Proceedings

THERMIONIC ENERGY CONVERSION

Specialist Conference

October 11-12 1989

Eindhoven University of Technology, Netherlands
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THERMIONIC ENERGY CONVERSION

Specialist Conference
Eindhoven, The Netherlands
October 11-12, 1989

Edited by
L.R. Wolff, W.B. Veltkamp, J.M.W.M. Schoonen
and H.A.M. Hendriksen

Eindhoven University of Technology
Eindhoven
1990

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Thermionica

Thermionische energieomzetting / hoge-temperatuur-materialen / elektrisch vermogen in de ruimte.
Preface

The 1989 Thermionic Energy Conversion Specialist Conference was preceded by a short course on thermionics\(^1\). It was organized at Eindhoven University of Technology in the Netherlands October 9-12, 1989.

With this conference a series of international specialist conferences on thermionics was revived. Incidentally the last conference of the previous series of specialist conferences was also organized at the Eindhoven University in 1975.

In the period prior to 1975, almost the entire thermionic R&D effort was aimed at nuclear thermionics for space applications. After the energy crises the thermionic research effort in the Western world was reduced to a much lower level. Since then thermionic research in the USA and in Western Europe was directed almost exclusively towards terrestrial applications. Substantial progress was made in the development of combustion heated thermionic converters. Extensive system studies were carried out on various cogeneration systems like e.g. thermionic combustors.

In 1984, just when the demonstration phase had been reached in the USA, the Department of Energy of the USA decided to discontinue funding of terrestrial thermionics R&D. At the same time the USA interest in space applications of thermionics augmented. This resulted in a period of revival of space nuclear thermionic research in the USA.

Meanwhile, in Western Europe, the R&D effort in thermionics has seen a steady growth. Research groups working both on fundamentals and on combustion heated thermionic converters are presently active in Sweden, Italy and the Netherlands.

In Japan, thermionic R&D, aimed both at fundamentals and terrestrial thermionic applications has received steady support over the years. At present thermionic R&D is carried out at three locations in Japan by a University, a research institute and a major representative of Japan's heavy industry.

The most consistent and powerful research program on thermionics, however, is without doubt the program of the USSR. At a number of research institutes and universities, significant groups of soviet scientists and technicians have been engaged in thermionic R&D ever since the fifties. The Soviets have successfully developed and ground tested in core thermionic nuclear reactors. Some of these reactors were incorporated in satellites and subsequently launched into space.

Recently the Soviets also took a renewed interest in thermionic topping cycles for central station power plants. Furthermore there is a renewed European interest in space nuclear thermionics, which present really the only practical solution to large power requirements in space.

In our opinion these facts and the general consensus that thermionic scientists need to join forces in their research effort, marked the beginning of a new era of world wide cooperation in thermionic R&D.

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\(^1\)The course material of the Thermionic Short Course is available in a separate textbook
At the Eindhoven conference, attended by over 70 scientists from the USA, the USSR, Sweden, the UK, France, Italy, and The Netherlands, 30 papers in total were presented. Due to travel restrictions in the USSR, some of the soviet authors were unable to present their papers at the conference. However, since these papers contain valuable and interesting scientific information, the editors decided to include these papers in these proceedings, be it in a separate section.

At the end of the conference an international ad hoc committee of thermionic scientists made a proclamation, stating the desirability of international thermionic cooperation in some specified fields of general interest. An international thermionic liaison group will be established to further this cause. This declaration is also included in these proceedings.

At this place the editors would like to thank all authors for their high quality contributions, which, in their opinion, render these proceedings a valuable source of information on thermionics.

Eindhoven, November 1989

Lodewijk Wolff,
Bart Veltkamp,
Toon Schoonen,
Huib Hendriksen
Editors.
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We were very pleased and proud to be part of the first truly international conference in thermionic energy conversion in fourteen years. Truly this has been a landmark event. Recent achievements in this area have been nothing short of remarkable, especially given the successful flight tests of two thermionic reactor units based on the Topaz design, successful development of terrestrial thermionic converters and verification of thermionic fuel element components. We look forward to further developments in the future.

We respectfully suggest three major approaches for advancement on which the international thermionic community may wish to concentrate its efforts.

1. There are several specific applications which offer a unique opportunity as rallying points for the international technical community. Foremost among these is the contribution which thermionic energy conversion can offer towards a manned Mars mission. Because of the immense distances involved and the complex logistical support required, it is scarcely conceivable that this mission could take place without substantial sources of electrical and propulsive energy. Thermionic reactors offer a decisive opportunity for space development because of their superior electrical performance, compactness, static operation and reliability. An international focus on such a project can only be a stabilizing force for peaceful international relations. Another application of great importance is the development of efficient terrestrial combustion-heated thermionic converters for increased efficiency of commercial power plants, for co-generation applications and for autonomous power sources for remote power stations. Still other near term beneficial applications of thermionic power include its use in thermionic nuclear reactor power systems for international air traffic control satellites; for advanced television broadcast satellites and for advanced communications.

2. Concomitant with the potential benefits of thermionic space nuclear power is the responsibility for addressing the needs of the public for nuclear safety of these devices. Especially in the Soviet Union, vast resources have been expended to ensure that space nuclear power plants will operate without threat to the public. An international data base on space nuclear safety should be compiled and made available to the public to allow responsible, educated decisions to be made. Experience has shown that unnecessary restrictions on the release of technical data have contributed to public mistrust in this area. Conversely, increased public awareness and education on the great advantages and low risk space nuclear systems should lead to support for their use.
3. **Advancing the state of the art** of thermionic conversion entails several key technologies such as: diode physics (surface physics, plasma physics, materials research, techniques for barrier index reduction, use of additives, impurity effects and control); thermionic fuel element development; advanced converter development (including low temperature converters and diodes with very low spacing). These projects can be undertaken by individual countries, but can also form the basis for international research and cooperation.

To accomplish such ambitious goals, it will be necessary to provide an international mechanism for formulating proposals and carrying them to potential sponsors. Accordingly, it is our recommendation that an international thermionic coordinating group be established with the International Energy Agency in Paris. It is hoped that the result will be further cooperation, which may take the form of more international seminars and meetings, data exchanges, personnel exchanges and cooperative projects.
Thermionic Energy Conversion
Specialist Conference Eindhoven
October 11 and 12, 1989

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THERMIONIC ENERGY CONVERSION: REFLECTIONS AND PROJECTIONS

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ABSTRACT

The thermionic energy converter is a remarkably simple but effective heat engine using electrons directly as a working fluid for the production of electric power. Its history of development provides lessons that are useful in planning for stable and productive future international cooperation in this field. If international cooperation is to be more than periodic exchanges of information on independent programs, a mutually acceptable goal must be defined with joint projects as a focus. Several dilemmas arise in seeking to define such a goal. One is whether to emphasize engineering development based on the current state-of-the-art technology ("gadgets") or to emphasize research on revolutionary new technology ("breakthroughs"). Because of the tendency for program officials to choose a low risk approach with familiar technology, the "gadget" approach usually is emphasized. Substantial advances have been made in thermionic converter "breakthrough" technologies, but most current activity in the field is related to incremental advances in 1970-vintage "gadget" technology for applications that are internationally sensitive. It is proposed that international cooperation should seek a balanced and stable program with initial emphasis on exploiting basic "breakthrough" opportunities, concurrent with definition of a "gadget" goal that is compatible with the outcome of crucial and rapidly changing international factors. Subsequent joint research and development, based on new technology and international compatibility, thereby can avoid obsolescence and disruptions such as have occurred in the past.

I. INTRODUCTION

The purpose of this conference, in addition to the exchange of technical information, is to explore the possibility for reinitiation of international cooperation in this field. What is the purpose of such joint work? What are its objectives and how should it be accomplished technically? What is realistic politically in view of the rapidly changing nature of the field and relations between its primary participants?

There is not time available for me to review in detail the many scientific and engineering activities that have been proposed for potential cooperation. Furthermore, my impressions of the relative merit and priority of these activities are still changing since there are few fixed points in the rapidly evolving areas of application and the support for them. I will try, therefore, to address generic considerations that have dominated this field in the past, and that should be recognized in planning the future.
One troubling generic aspect of the field is that, because of the displacement of the electron tube by solid state electronics, relatively few people are trained in gaseous electronics. As a result there is a tendency to identify thermionic converters as esoteric devices having little in common with the more familiar heat engines. From a thermodynamic viewpoint, however, there is a great deal of similarity that permits recognition of the basic limitations on the present technology and of the potential for substantial improvement.

As can be seen in Fig. 1, the thermionic energy conversion cycle is similar to a modified Rankine (steam engine) cycle that uses electrons directly as the sole working fluid. The emitter is the "electron boiler" and the collector is the "electron condenser" which develop an electrical pressure (potential) difference to produce electrical work rather than a vapor pressure difference to produce mechanical work. The work functions of the electrodes are the heats of vaporization of the electrons. The near isentropic heating of condensed electrons in the emitter lead AB ("feedwater heating") and the isothermal vaporization of the electrons BC are similar to equivalent processes in the Rankine cycle. In the thermionic cycle, however, the hot electrons condense irreversibly (EF) into the low temperature collector, causing the efficiency of the ideal thermionic cycle to be near 60% of the Carnot thermodynamic efficiency compared with about 80% for the Rankine cycle. In the ignited mode converter that has been the basis for practical application, this ideal thermionic cycle efficiency is about halved to 30% of Carnot efficiency because about half of the electric power generated is dissipated internally to heat the electrons to the high temperature required for maintenance of the plasma. Advanced converters with more efficient cold plasmas can approach the ideal thermionic cycle efficiency. By comparison, the best thermoelectric (solid state) converters give only about 15% of Carnot efficiency, with an upper limit variously estimated at 20-30%. Present and projected thermodynamic efficiencies for thermionic conversion therefore are about double those for thermoelectric conversion.

II. HISTORICAL LESSONS

Past and ongoing thermionic programs are summarized briefly in Fig. 2. The first significant generation of thermionic power was demonstrated in the USSR by Marchuk in 1956 using the unignited cesium diode, followed in 1957 by demonstration in the US by Wilson of the more practical ignited diode, and in 1959 by in-core operation of the unignited diode by Grover, et al, apparently all acting independently. By 1965 sufficient exploratory experimentation and understanding had occurred to recognize the outstanding advantages of the in-core thermionic reactor using ignited converters, and engineering development programs were initiated in the US, USSR and Western Europe.

Rather than dwell on the evident details, it is instructive to notice some salient differences in the programs. First, whereas the US chose to develop the technology of the thermionic fuel element (TFE) beyond the 1965 level prior to constructing a test reactor, the USSR chose immediately to construct full scale test reactors based initially on the 1965 TFE technology.
Fig. 1 Basic thermionic energy conversion process.
### US Thermionic Program

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<th>Basic Physics</th>
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<td>Elementary Cesium Diode Exploration</td>
<td>Solar, Radioisotope &amp; Nuclear Reactor Exploration</td>
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<td>1965</td>
<td>Ignited Cesium Diode Consolidation</td>
<td>Thermionic Nuclear Fuel Element (TFE) Development</td>
</tr>
<tr>
<td>1973</td>
<td>Advanced Mode Exploration</td>
<td>Fossil-Fueled System Exploration</td>
</tr>
<tr>
<td>1975</td>
<td>Very High Temp. Exploration</td>
<td>TFE Technology Recovery</td>
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</table>

### USSR Thermionic Program (Estimated)

<table>
<thead>
<tr>
<th>Year</th>
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<td>1956</td>
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<td>1962</td>
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<td>1965</td>
<td>Unignited Diode Consolidation</td>
<td>Ignited Diode Consolidation</td>
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<td>1970</td>
<td>High Power TFE Development</td>
<td>Topaz Flight Test (Topaz Deployment)</td>
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<td>1975</td>
<td>Topaz Reactor Experiments</td>
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<td>1978</td>
<td>High Power Reactor Development</td>
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<td>1988</td>
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<td>Ignited Cesium Diode Consolidation</td>
<td>Thermionic Nuclear Fuel Element (TFE) Development</td>
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<td>1973</td>
<td>Ignited Mode 1 Cermet Emitter</td>
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<td>1975</td>
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<td>Ignited Mode 3 Cermet Emitter</td>
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<td>1985</td>
<td>Ignited Mode 4 Cermet Emitter</td>
<td></td>
</tr>
<tr>
<td>1990</td>
<td>Ignited Mode 5 Cermet Emitter</td>
<td></td>
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</table>

1- W. Germany, France, Euratom  
2- W. Germany, Netherlands  
3- W. Germany  
4- Sweden  
5- Netherlands

Fig. 2 Summary of thermionic energy conversion programs.
Second, whereas the USSR basic and engineering programs have been continuous and coherent since 1965, the US program has been characterized by gaps and a 10-year hiatus in thermionic reactor development during which combustion-heated (fossil-fueled) converters and advanced converter technology for electric power plants were explored. Third, basic work in the US has been performed primarily only in direct support of engineering programs and has suffered the disruptions accompanying the changes in those programs, whereas no such linkage is apparent in the USSR programs.

The approach to thermionic reactor development by three independent groups in Western Europe was TFE-oriented in the period 1965-1973 as in the US. The subsequent fossil-fueled work there was initially for auxiliary automotive power systems, however, and combustion-heated diode development here at Eindhoven has kept this technology alive after it was totally abandoned by the US.

It is important to recognize that Fig. 2 does not reflect the relative magnitude of manpower and facilities in the various activities. Typically, basic converter work has been less than 10% of the total activity. While the total thermionic work in the US and in Western Europe were comparable in the period 1965-1973, the magnitude of the Western European work has been much smaller than that in the US since then. Of overriding significance, however, is that the total thermionic activity in the USSR has been and at present is at least an order of magnitude greater than the total elsewhere in the world.

The continuity, coherence, magnitude and focus of the USSR work for the past 25 years has produced most of the scientific basis of thermionic energy conversion and the only operational fully-engineered thermionic systems. Yet the very lack of these characteristics in the US and Western European programs have caused their technology base to be more diverse, more innovative and more directed toward problem solving, particularly relative to obtaining long converter lifetime.

Accordingly, the past history of the field suggests that programmatic considerations have more to do with its progress and successes than scientific and technical considerations. There may be an opportunity now, through international cooperation, to combine the focus and long term commitment of the USSR thermionic program with the diversity and innovation of US and Western European programs in a more stable and productive joint program than could be achieved separately.

Two attempts were made previously for international cooperation in thermionics. The first (1965-1973), conducted under the auspices of the International Atomic Energy Agency and the Nuclear Energy Agency of the OECD, was dissolved when all participants but the USSR terminated their nuclear thermionic programs. The second (1976-1979), between the US and USSR, was terminated by unrelated political differences between the two countries. The two lessons to be learned here therefore are: 1) international cooperation should not focus so exclusively on one application that cooperation crashes if that application falls temporarily from favor; and 2) international
cooperation should include more than two countries so that the cooperation does not crash when temporary political differences arise between two countries.

III. COOPERATIVE PROGRAM PROSPECTS

As summarized in Fig. 3, the first decision that must be made is to chose between international cooperation and just continuing completely independent programs as in the past decade. If international cooperation is chosen because of the perceived substantial advantages just discussed, a choice then must be made between a fragmented cooperative approach and a goal-oriented joint program. In the former approach, which characterized the previous attempts at cooperation, the countries independently defined and pursued their separate goals, but met periodically in international meetings or exchanges of delegations to describe their progress and define mutual problem areas and solutions. If this approach is chosen, there is only need for a liaison committee to be formed that arranges and conducts the meetings and exchanges.

If instead a goal-oriented joint program is chosen as the basis for international cooperation, a mutually acceptable goal must be defined, which is substantially more difficult than just appointing a liaison committee. Several dilemmas immediately arise:

1. If the goal is important commercially or militarily, no country will want to share it. vs
   If the goal is unimportant, no country will want to pursue it.

2. If the goal is a specific application, cooperation crashes with either success or failure (see #1). vs
   If the goal is non-specific (no practical linkage), then the US at least loses interest.

3. Is the goal a "breakthrough" or a "gadget"?

IV. "BREAKTHROUGH" VS "GADGET" DILEMMA

In his book Science and Government, C.P. Snow deals with the fundamental competition for government funding between the advocates of "gadgets" (ingenious use of current state-of-the-art technology) and the advocates of "breakthroughs" (revolutionary new technology that renders obsolete the current state-of-the-art). Now the terms "gadget" and "breakthrough" are just code words for the different types of technology, and in no way are meant to depreciate or glorify either type. Similarly, there are all shades of semantic grey between the black and white extremes of "gadgets" and "breakthroughs", but all these semantic qualifications are unnecessary to define the essential dilemma. Snow describes in detail how the "breakthrough"
Fig. 3 Thermionic conversion program prospects.

THERMIONIC
"GADGET" TECHNOLOGY

OXIDE NUCLEAR FUELS
INSULATORS
TRILAYERS
CESIUM RESERVOIRS
FISSION PRODUCT VENTING
HEAT PIPES
RADIATORS
FUEL CLADDING
HOT SHELL
SYSTEM DESIGNS
COMPONENT LIFE TESTS

THERMIONIC
"BREAKTHROUGH" TECHNOLOGY

ADVANCED ELECTRODES
Cs/O VAPOR \( \rightarrow \) HIGH \( \theta_0 \)
DISPENSERS \( \rightarrow \) LOW \( \theta_c \)
COLD PLASMA
TRIODES
PULSED DIODE
MOLECULAR EXCITATION \( \rightarrow \) LOW \( V_d \)
OUTPUT CONDITIONING
INDUCTIVE COUPLING
RF GENERATION
VERY HIGH TEMPERATURE
CERMET NUCLEAR FUELS
HIGH HEAT & CURRENT DENSITY

fig. 4 "Gadget" vs "breakthrough" issue in thermionics.
technology of radar prevailed (through subversion via the afternoon tea) over "gadget"-oriented high-level official opposition early in World War II and probably saved Britain. "Gadgets" prevailed in Germany when the war-winning "breakthrough" technologies of ballistic missiles, jet aircraft and nuclear weapons were at hand. Clearly, though, neither side could have totally neglected the conventional "gadgets" of war in order to pursue "breakthroughs". A "gadget" vs "breakthrough" balance, therefore, is the vital issue.

To achieve this balance, it is essential first that both sides of this issue be understood, by the "breakthrough" and "gadget" advocates and by the government officials (or corporate managers) that make the choice between them. "Gadgets" usually win in those choices, resulting in grossly unbalanced programs. "Gadget" technology is "real": it is familiar to the officials and their advisors, and clearly available for current needs; "breakthrough" technology is esoteric, with a low probability of early success. Without understanding or confidence in the magnitude of what might be gained by taking the risk, the choice is easy -- "gadgets". The over-emphasis on "breakthroughs" in this presentation is an attempt to balance the unbalanced appeal and constituency of "gadgets". It is not an attempt to depreciate the primary importance of "gadgets".

Fig. 4 lists illustrative "gadget" and "breakthrough" technologies for thermionics. Advances in the "breakthrough" technologies might greatly relieve if not eliminate the dominant "gadget" problems. Yet "breakthrough" activities typically receive less than 10% of the funding in the US, and are the first to be dropped in the inevitable budget adjustments when the current "gadget" application either fails or succeeds. Lack of success with the meager resources provided is a predictable and convenient justification for dropping "breakthrough" research.

By way of further illustration of this dilemma let me share with you some quotations from officials who have sponsored my work:

* "The converter is good enough to compete with other systems." This is true, but underlying this statement is the assumption that the converter (circa 1965) is immutable by definition, and that there will be no advances in the competing systems.

* "You tell me more than I want to know." This is true, but he needs to know more to avoid obsolescence of the technology he funds.

* "You propose things that can't be done." This is true to the extent that they can't be done if there is insufficient funding to do them. British officials blocked radar because "it can't be done," i.e., it required a thousand-fold more powerful microwave generator than had ever been demonstrated or even conceived. Via the afternoon tea, scientists arranged enough research to discover the strapped cavity magnetron, an unexpected super-efficient and super-powerful microwave generator that only recently is fully understood.
V. BALANCE THROUGH COMMUNICATION

The only real way to achieve a "gadget" vs "breakthrough" balance is to provide a programmatic context for communication between the two sides of this issue. Fig. 5 shows a programmatic context that has been effective for coupling basic thermionic converter technology with engineering development. It consists of an iterative "basic loop" in which a detailed physical/analytical "fundamental model" is formulated from first principles that is consistent with data from basic thermionic converter experiments. Because the rigorous complexity of the fundamental model prevents it from being understood or useful to the "gadget" advocates, a much simpler "working model" is formulated that is consistent with the fundamental model in the practical regime. This working model provides direct intuitive insight into the dominant physical processes to focus basic converter experimentation and innovation onto current practical objectives.

The basic loop meshes with an iterative "engineering loop" in which the working model is used for system design innovation and evaluation, and for guidance and interpretation of engineering tests. Through this meshed two-loop process, therefore, the two sides can mutually recognize and focus upon the practical significance of a basic "breakthrough" for the current "gadget". Furthermore, this is precisely what the program manager needs to know in order to achieve a balanced program. Without the engineering loop as a directing focus, the basic loop tends to degenerate into irrelevant academic projects. Without the basic loop, the engineering loop freezes into obsolescence.

The basic principles of converter operation can be understood to at least the same degree as the internal combustion engine is generally understood, since the converter is substantially less complex and can be described much easier with quantitative precision. As shown in Fig. 6, the electrical output of a thermionic converter is characterized by the back-voltage ("barrier index") $V_B$, which is analogous to the back-pressure in a mechanical engine. It has three components, the arc drop (plasma energy loss) $V_d$, the current attenuation voltage $V_a$, and the collector work function (Surface energy loss) $\phi_c$. For a given output current density $J$, the output power density and efficiency of the converter increase linearly as $V_B$ is decreased.

Rigorous description of the ignited (hot plasma) mode of conventional converter operation is complex because the plasma must both transport the electrons across the interelectrode gap and heat the electrons to a high enough temperature to maintain itself. However, because the plasma is thin (only a few mean-free-paths in thickness) and has very low electrical resistance, an adequate description of converter performance for practical purposes can be obtained without integrating the complex set of differential equations that describe the detailed state of the plasma. Enforcement of energy and current continuity at the plasma boundaries yields a set of relatively simple algebraic equations describing converter performance and providing insight into the dominant physical processes and basic properties that determine $V_B$. These equations, expanded somewhat to cover a wider range
Fig. 5 Balanced & coherent advanced research & development.

Fig. 6 Basic thermionic converter electrical output characteristics.
of conditions, have been incorporated by John McVey into a computer program TECMDL that is widely used in the US for system design analysis and test data interpretation. He also has developed more rigorous analytical models and programs, based on numerical integration of the plasma differential equations, that serve as the "fundamental model" in Fig. 5 [1, 2]. The basic physics of electrode properties in cesium vapor similarly is readily understood and is included in the working and fundamental models.

VI. "BREAKTHROUGH" PROSPECTS

The analytical models provide insight and projections for substantial, even revolutionary, improvements in converter performance and application versatility. As can be seen in Fig. 7, these improvements are conveniently divided into two basic approaches. One approach is to decrease the internal barrier to electron flow in the converter, $V_b$, by advanced converter technology (ACT). The improvement can be through a manyfold increase in power density and doubling of efficiency at present emitter temperatures, or through maintaining present performance levels at much lower emitter temperatures. The other approach is to utilize the much higher levels of performance attainable with emitters at very high temperatures (VHT) using the present basic converter technology ($V_B = 2.1$ eV; i.e. $\phi_C = 1.6$ eV, $V_d = 0.4$ eV and $V_a = 0.1$ eV).

Fig. 8 compares conventional ignited mode converter operation with an advanced mode of operation. The ignited converter employs a high-density, highly-collisional hot plasma that is maintained by an internal potential drop $V_d$ that is subtracted from the output voltage. The working model of ignited operation tells us that the arc drop $V_d$ arises almost entirely from the energy lost by heating the plasma electrons above the emitter temperature, and that the prospects for circumventing this inherent limitation in ignited operation are not good. The advanced mode converter, however, employs a low density, quasi-collisionless cold plasma at about the same temperature as the emitter, such that its arc drop $V_d$ is nearly zero. Its plasma is maintained by an efficient externally powered ionization process.

Other aspects of advanced mode operation are even more important practically than the reduction of the arc drop. Advanced mode converters operate at more than an order of magnitude lower cesium pressure, which is a fundamental requirement for using much lower collector work functions $\phi_C$ at low collector temperatures, and which permits use of large electrode spacings. Furthermore, since its plasma is maintained by an external power source, the advanced mode converter can act as its own output power conditioner, e.g. as a switch for production of alternating current. Fig. 8 includes a schematic illustration of the TRICE (Thermionic Reactor with Inductive Coupled Elements) concept, in which the output of uninsulated fueled emitters in a nuclear reactor core are coupled by electromagnetic induction to the high voltage external load circuit [3].
Fig. 7 Options for thermionic converter performance improvement:
ACT: Advanced Converter Technology approach.
VHT: Very High Temperature approach.
Fig. 8 Comparison of conventional ignited mode thermionic converter with an advanced mode of converter operation and output coupling.
Fig. 9 illustrates the family of advanced mode converters, in which the plasma is maintained by electron or ion injection, and on which significant research already has been performed. In each case the plasma is maintained by a power supply \( P \) driven by the external circuit. The potential drop across \( P \) is the "equivalent arc drop" \( V_{eq} \) that is to be compared with the plasma-maintaining arc drop \( V_d \) in the ignited diode.

In addition to the challenge of obtaining \( V_{eq} \ll V_d \), advanced converter research must deal with basic plasma processes that have little effect on conventional ignited converter operation. Fig. 10 illustrates the interaction of the plasma with the self-generated magnetic field [4]. As shown, the converter output current \( I \) is strongly modulated by the magnetic field \( B \) in the interelectrode gap for converters with long electrode length \( L \) and long electron mean-free path \( \lambda \) at sufficiently high output current density \( J_0 \). Also, the Lorenz pondermotive force on the plasma by the magnetic field produces an axial pressure drop \( \Delta p \) that can approach the total plasma pressure in advanced converters. Furthermore, Coulomb scattering and associated local collective space charge effects can be the dominant current-limiting processes in advanced converters. All these effects can be viewed as obstacles that must be avoided in practical converter designs; conversely, they can be seen as strongly coupled feedback interactions between the output current and the plasma that might be the basis for innovation of a new class of thermionic RF power generators, analogous to the strapped cavity magnetron "breakthrough" in C.P. Snow's history of radar.

All advanced-mode converters require high emission currents at very low cesium pressure, which are contradictory requirements that have inhibited their practical use. A true "breakthrough" is the discovery that the use of cesium-oxygen vapor in the converter instead of cesium vapor alone allows removal of this contradiction. As shown in Fig. 11, the presence of only a very small partial pressure of an oxygen-bearing species allows the emission current to be increased by orders of magnitude into the practical regime at low cesium pressures because the oxygen increases cesium adsorption on the emitter [5]. Furthermore, the presence of oxygen also results in very low work functions \( \phi \), at low collector temperatures (and low cesium pressures) as shown in Fig. 12. Although the basic properties of electrodes in cesium vapor are well understood and can be represented by analytical models, the properties of electrodes in cesium-oxygen vapor and the technology for maintenance and control of the cesium-oxygen vapor need substantial further research to achieve a similar level of understanding for practical application.

Relative to the VHT "breakthrough" approach defined in Fig. 7, Fig. 13 shows the performance projected by the analytical models for converter operation at very high temperatures [6]. The two modes of conventional cesium diode converter operation are shown: the ignited (volume ionization) mode and the unignited (surface ionization) mode. Although ignited mode performance is superior below about 2500 K, the performance in both modes is comparable at higher temperatures. Refractory metal emitters with adsorbed cesium can be used for ignited operation, but unignited operation requires emitters with a
Fig. 9 Types of advanced mode (cold plasma) thermionic converters.

Fig. 10 Effects of self-generated magnetic field on cold plasma thermionic converters.

Thermionic Energy Conversion
Fig. 11 Emission enhancement in cesium/oxygen vapor.

Fig. 12 Minimum work function at optimum cesium pressure for Cs₂O pressure of 2 x 10⁻⁴ torr.
Fig. 13 Estimated thermionic converter performance at very high temperatures.

Fig. 14 Comparison of conventional and very high temperature converter performance.
bare work function near the optimum value. Experimental data are shown for unignited converters using zirconium carbide emitters (optimum near 2600 K), obtained by Maskevitch and co-workers who pioneered the VHT approach [7]. The order of magnitude increase in power density and doubled efficiency potentially available by the VHT approach are illustrated in Fig. 14.

Whereas advances in basic converter physics are required for "breakthroughs" by the ACT approach, advances in materials and innovations in converter and reactor configuration are required to utilize the "breakthrough" thermionic performance presently available at very high temperatures. Tungsten/UO₂ cermet nuclear fuels were developed for nuclear rocket application and operated stably for hours at 2800 K, but little is known about their lifetime at lower temperatures and higher fuel burnup. The fuel lifetime may be adequate even at 2800 K for short duration military space applications, however, and adequate at lower temperatures for the duration of an electrically propelled freight mission to Mars. Carbide-based fuels were operated at similar temperatures and times, but their vaporization rate is much higher than that of tungsten, and their vaporization products appear to be intolerable on high temperature collectors. A major challenge is to find new thermionic fuel element and reactor design innovations that accommodate the very large heat fluxes and output currents that characterize VHT operation.

VII. PROSPECTIVE APPLICATIONS

Thermionic reactor space power systems are a developed reality and probably will continue to be decisively superior to other systems because of the inherent advantages of in-core operation illustrated in Fig. 15. By far the largest investment and current activity in thermionic conversion has been in this application, especially in the USSR, and the USSR has proposed joint tests in their extensive thermionic reactor facilities as a focus for international cooperation. The choice of this application as a focus might be unfortunate, however, if the growing opposition worldwide to space nuclear systems (Fig. 16) leads to their being outlawed [8]. Deep space (e.g. Mars) missions might not be included in the prohibition. Accordingly, technologies addressing safety issues and the exceptional performance required for deep space missions should be emphasized if space nuclear power is a major justification for the joint research.

Except for the combustion heated ignited thermionic converter work being performed here at Eindhoven and the more limited work at Mitsui in Japan reported for the first time at this conference, there apparently is no other such work being performed at present elsewhere in the world. However, I am aware that there is interest in this application in both the US and USSR.

Substantial research and technology development for various fossil fueled thermionic applications were conducted in the 1975-1983 period. Fig. 17 shows examples of the types of terrestrial fossil fueled thermionic systems.
Fig. 15 Inherent advantages of in-core thermionic reactors.

MULTIPLE MODULAR REDUNDANCY

EXCEPTIONALLY HIGH CYCLE TEMPERATURES WITH CONVENTIONAL COOLANT TEMPERATURES

Fig. 16 Prospects for continuity of US space reactor programs.

**New York Times**

MONDAY, SEPTEMBER 11, 1989

ENERGY DEPT. AIMS TO STOP SPENDING ON SPACE REACTOR

BUDGET IS A MAIN FACTOR

But NASA and Pentagon Say Move Could Hurt Defense and Exploration Plans

By WILLIAM J. BROAD

The Energy Department wants to stop spending money on the Constellation space station program to develop a nuclear reactor for use in outer space, a Federal official said last week. The agency, which provides most of the money for the program, faced rising budget pressure, and the reactor program faces opposition in Congress and among groups who say they fear that an accident with an existing reactor would result in radioactive debris raining down on the Earth.
Fig. 17 Fossil fueled terrestrial thermionic systems.

(a) Topping of utility steam power plant.
(b) Topping of utility gas turbine generator.
(c) High temperature industrial process cogeneration system.
that have been evaluated in detail: thermionic topping of electric utility steam power plants and gas turbine power installations, and thermionic cogeneration systems for provision of high temperature heat and autonomous electrical power for industrial processes. Detailed system studies showed that fuel costs (efficiency) and capital costs (material expense, power density and lifetime) were marginally competitive with those of alternative systems for $V_o \approx 1.8$ eV, but clearly competitive for $V_o \leq 1.6$ eV. Other applications studied were thermionic bottoming of MHD Power plants and portable auxiliary electric power units.

VIII. CONCLUSIONS

Returning in summary to the decision tree in Fig. 3, an immediate solution to the central dilemmas posed in defining a specific goal for international cooperation is not apparent. This is primarily because of the uncertainties associated with the following crucial international factors that presently are in a state of rapid change:

1. Public acceptance of space nuclear power.
2. Domination of space nuclear power by military applications.
3. Commitment to joint interplanetary missions.
4. Absence of terrestrial fossil fueled thermionic R&D programs in most countries.

Because substantial changes in any of these factors could greatly affect the viability of an associated specific application goal, it is concluded on the one hand that commitment to such a goal should be postponed for 2-3 years while these factors are being resolved.

On the other hand, it is concluded that generic research in potential converter "breakthrough" areas is substantially independent of these factors, i.e. that it has no immediate implications of international or public policy, of great commercial or military sensitivity, or of relevance to the existence or non-existence of space or terrestrial application programs in the participating countries. However, as an engineer I believe that a cooperative program based solely on such research, without focus or practical justification, would be grossly unbalanced toward "breakthroughs", resulting in fragmented academic projects of little enduring interest to the sponsoring governments.

It is proposed, therefore, that international cooperation proceed in two phases, as summarized in Fig. 18. The goal of the First Phase would be achievement of selected "breakthroughs" in advanced converter performance, with joint experimental demonstration of a specific "breakthrough" as a focus. In support of the First Phase, preliminary engineering studies would be performed on a variety of space nuclear and terrestrial systems to define the best specific application and system concept for the Second Phase goal, consistent with the outcome of the crucial international factors and with the extent to which advanced converter technology is evolved in the First Phase.
<table>
<thead>
<tr>
<th>1ST PHASE (3-5 YRS)</th>
<th>2ND PHASE (3-5 YRS)</th>
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<tr>
<td><strong>GOAL</strong></td>
<td><strong>SPACE</strong> low power (communication)</td>
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<tr>
<td>GENERIC ADVANCED</td>
<td><strong>NUCLEAR</strong> high power (electric propulsion)</td>
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<tr>
<td>PERFORMANCE DEVELOPMENT</td>
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<td>- ADVANCED ELECTRODES</td>
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<td>- ADVANCED OPERATING &amp; OUTPUT MODES</td>
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<tr>
<td><strong>FOCUS</strong></td>
<td><strong>SPECIFIC DEMONSTRATION OF ADVANCED ENGINEERING COMPONENT WITH BASIC LOOP IN SUPPORT</strong></td>
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![Fig. 18 Proposed joint international program.](image-url)
The goal of the Second Phase would be engineering development of crucial component technology for the selected application and system concept, with joint experimental demonstration of the system or a major module as a focus. In support of the Second Phase, basic work would focus on the base technology of the selected concept, assist in resolution of problem areas and provide diagnostic interpretation of the engineering test data.

Three to five years would be required in the First Phase for adequate progress in the advanced technology, for evolution of the crucial international factors, and for development of sufficient confidence in international cooperation and Second Phase goal definition to justify commitment of the resources required for the Second Phase by the participating countries. A similar period would be required for the Second Phase, which might partially overlap the First Phase.

In summary, it is concluded that:

* New basic technology is available for
  - a second generation of high performance thermionic reactors.
  - a first generation of economically viable terrestrial thermionic power systems.

* International cooperation can provide the required diverse resources and stability by
  - defining goals common to interest of all countries but not dependent on one.
  - providing a focus that balances and integrates "gadget" and "breakthrough" forces.

Above all, the dismal cycle of slogging through dozens of peripheral problems for incremental improvements in 1970-vintage technology, until terminated by changes in international relations and national interests, should be avoided. The revolutionary advances potentially achievable in this field are too exciting and life is too short.
REFERENCES


Thermionic Energy Conversion
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EXPERIMENTAL RESEARCHES OF STATIONARY AND OSCILLATING

REGIMES OF KNUDSEN PLASMA DIODE

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The rather complete theory of the processes taking place in Knudsen plasma diode was developed up to date [1-3].

But some conclusions of the theory have not been confirmed experimentally.

The experimental research results of field distribution in the near-electrode layer in Knudsen plasma diode stationary regime, threshold of oscillations onset and field distribution in separate phases of current oscillations are given in this work.

Measurements were carried out with plane-parallel geometry diode which was structurally integrated with the electron-beam device for plasma probing by a planar electron beam which gave the opportunity to measure electric-field distribution in the interelectrode gap [4-5].

The experimental device scheme is given (see fig. 1).

1. STATIONARY REGIME

The potential change in the near emitter layer in Knudsen diode is Debye distance [1]

\[ \lambda = \frac{0.32(KTe)^{1/4}}{e^{1/2} m^{1/4} T_e^{1/2}} \]

In these conditions restricted resolution of the device (5.10 cm) enables to carry out only qualitative research of the near-emitter layer. In contrast the potential drop in the near-collector layer is determined by applied voltage and layer width forms significant part of the interelectrode gap, which gives the opportunity to carry out precise measurements. The experimental conditions were as follows: polycrystal tantalum emitter with work function \( \Phi_E = 4.7 \) eV; interelectrode gap \( d = 0.3 - 0.4 \) cm; cesium vapour pressure \( p \approx 10^{-5} \) Pa; \( T_e \approx 2000 \) K.

The ion current density \( J_i \) according to
The Saha-Langmuir formula with given $A_d$, $T_e$ and $p$ gives compensation coefficient $f = \frac{2}{3} \sqrt{\frac{3e}{m_e}}$ in the region $5 < \frac{T_e}{T_i} < 10$. Experimental definition of $f$ requires the measurement of electron density distribution, received by field curves differentiation. The near-emitter plasma layer with characteristic positive potential for $\frac{T_e}{T_i} < 1$

Fig. 1. Experimental device scheme:
1 - plasma diode; 2 - monitoring electrode; 3 - anode; 4 - electron gun cathode; 5 - quadrupol lens; 6 - diaphragm; 7 - deviating plates; 8 - luminescent screen; 9, 11 - rectifiers; 12 - power unit; 13 - generator; 14, 15 - power amplifiers; 16 - H.F. transformer; 17 - oscillograph; 18 - resistor.

and ion saturation current from the emitter. Electron thermoemission from the collector prevents measurement to the ion current from the emitter and the definition of arises some difficulties in such a way. However, the charge density definition in near-electrode layer enables to evaluate $\frac{f}{f}$ and plasma density.

Fig. 2 represents:

- a) volt-ampere characteristic (VAC) of plasma diode,
- b) field distribution curves,
- c) potential distribution curves received by field curves integration,
- d) charge density distribution, are seen on curves not far from the emitter surface with a corresponding barrier, which accelerates the emitted electrons and limits ion flow in plasma. At the collector positive potential in the gap the monotonous regime of potential distribution is formed (curve 1), which is characterized by the emitter positive jump, by the constant quantity in the plasma region and by the collector positive jump. At the negative collector potential the double ion-electron layer is formed at the emitter and this layer forms non-monotonous potential distribution. The plasma region follows the double layer and then passes into the negative charged near-collector layer (curves 2-4). In the presented figure the field distribution has linear character in a considerable part of the near-collector layer, this coincides with constant charge density and the parabolic potential distribution.

Fig. 3 represents the dependence of the

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dimensionless width $c$ of the near-collector layer on the dimensionless collector potential $\Phi_c = \frac{V_c}{kT_e}$.

Fig. 3.

This dependence is described well by the simple relation:

$$\frac{1}{2} \frac{\Phi_c}{c} = \Phi_c - 1 \quad (1)$$

Such field distribution character is observed only at low emission current from the collector $J_c$ when $J_c / J_c \gg 1$. In this case the contribution of the electrons, emitted from the collector to the space charge in the near-collector layer is insignificant. With the collector emission current increase the deviation from the mentioned dependence in the field distribution at the collector surface is observed.

2. CURRENT OSCILLATION THRESHOLD

During theoretical description of experimentally observed oscillations in plasma diode in Knudsen regime [3, 7, 9] the conclusion was drawn about the Pierce character of the instability. However, more or less complete experimental data, confirming Pierce relation justice are absent.

At Pierce instability the current density threshold value above which instability arises

$$J_o = \frac{\pi}{4} \frac{m^2}{e^2} \left(2 + \Phi_E\right) \frac{1}{kT_e}$$

where $u$ - beam velocity, $\ell$ - plasma region length [6].

For comparison of experimental results of the threshold current measurements with formula (2) one can notice that the plasma region length is $\ell \approx d - l_n$ (narrow near-emitter layer can be ignored). When $\Phi_c$ values are large, as it happened during the experiments, according to formula (1)

$$\ell = \frac{2\Phi_c}{\sqrt{2}} \frac{1}{kT_e}$$

the mean velocity is

$$v = \frac{2\Phi_c}{\sqrt{2}} \frac{1}{kT_e}$$

where $\Phi_E$ is dimensionless value of the emitter potential jump. Substituting these expressions in formula (2) we'll receive:

$$J_o = \frac{\pi}{4} \left[2kT_e(2 + \Phi_E)\right]^3$$

Knudsen diode stationary regime theory permits to calculate the emitter potential jump $\Phi_E$ precisely enough. The value of this jump at $\Phi_c \gg 1$ doesn't depend on $\Phi$ and is determined by the equation [1]

$$\chi = \frac{1}{2} \epsilon^2 \Phi_E \left(1 - \epsilon_\gamma \sqrt{\Phi_E}\right)$$

The measurements for testing Pierce condition in Knudsen plasma diode were carried out as follows: at given values of the interelectrode gap $d = 0.45$ cm, caesium vapour pressure $p$,
and applied voltage $V_c$ current value changed by increase of emitter temperature $T_e$. Dependence of current value $J$ on emitter temperature $T_e$ is recorded on the XY-recorder. When the current value threshold is achieved, field oscillations are seen on the electron-beam set screen.

Fig. 4.a. Current threshold density dependence on emitter temperature. Point numbers correspond to collector voltage:

1 - 0.25 V; 2 - 2 V; 3 - 4.8 V; 4 - 7 V; 5 - 10 V; 6 - 15 V; 7 - 20 V; 8 - 25 V; 9 - 28 V; 10 - 35 V; 11 - 40 V

By presenting the results of measurements given in fig. 4a as a threshold current dependence on a collector potential (fig. 4b) it allows to use them for testing Pierce condition satisfaction. For this purpose $J_e$ dependences on $Q_c$ are plotted by means of formula (3) for different values of $\gamma$ in the same figure. When carrying out calculations one can consider that the emitter temperature is equal to the mean value 2100 for all these experiments, that results in the error within ±5%. When $J$ changes within 1.5-15, that corresponds to the extremes obtained during $J_e$ calculation by means of Saha-Langmuir formula, the curves $J_e(Q_c)$ are in the narrow shaded region (fig. 4b) within which all the experimental points are.

3. FIELD DISTRIBUTION IN CURRENT OSCILLATION REGIME

When current threshold values $J > J_0$, the potential oscillations with small amplitude are observed in the limited region of the gap, occupied by neutral plasma. So the potential in the whole gap remains positive and potential oscillations don't result in the current oscillations in the circuit. When current increases the potential oscillation amplitude increases and current oscillations appear.

When the condition $J > J_0$ is reached the current oscillations become regular. Current variable component is then a significant part of the emitter current emission (50% or more).
Some papers are devoted to the theoretical research of oscillating processes in Knudsen diode. Their detailed description is given in [7, 7]. It is established, that current oscillations present the sequence of several processes, among which Pierce instability acts as a starting mechanism at the beginning of each current oscillation period [9]. The detailed research of the instability starting stage led to the conclusion that Pierce instability is nonperiodic and lies in the fact that initial potential distribution with a jump at the collector for time \( \frac{d\psi}{d\eta} \) turns into distribution with a jump at the emitter [7]. However, the received potential distribution is not self-consistent with ion density decrease and as a result the current limiting emitter potential barrier appears. Further the region with low ion concentration is transferred to the collector with the rate of ion movement and current remains limited. When this region reaches the collector the barrier disappears and whole emission current passes through the diode. The potential distribution returns to the initial state. The cycle is repeated.

For the experimental confirmation of the conclusion [7, 7] the field distribution measurement in the gap is necessary. The results of such research are presented in this part of the article. The description of the measurement method is given in [5].

Fig. 5 represents one of our experimental current oscillation curves and corresponding field and potential distribution (solid and dotted lines respectively) in separate phases of these current oscillations. The figure corresponds to the following experimental conditions:

\[ \lambda = 2 \times 10^{-2} \text{ cm}, \quad d = 0.3 \text{ cm}, \quad \frac{eU}{K^*T} = 40, \quad 5 \cdot 10^{-7} < \theta. \]

When analysing these curves we shall take as initial position 12 with a potential barrier for electrons near the collector. Field distribution 12 is unstable and for time \( 1 \times 10^{-7} \) s transforms into distribution 1 and then into distribution 2, in which the main voltage drop is concentrated in the near-emitter region and the current equals to the emitter emission current. Such a transition takes place due to Pierce instability [3, 7] which by the increasing nonperiodic process leads the system to the stable state 2. According to Pierce condition it means that \( J_{e1} \cdot J_{e2} \). It may be realized either by current limiting or by threshold value increase. Since during transition from state 12 to state 2 the beam current increases the current threshold value must increase more rapidly.

That is confirmed by observation of \( U_{e2} \) emitter potential jump dependence on value \( J_{e1} \).
At the given collector potential $V_c$, when $J$ increases the value $V_c$ increases too and may achieve values exceeding $V_c$.

Thus, Pierce instability in the Knudsen diode is revealed in current threshold value change $J_0$ while increase of beam energy by near-emitter jump. As a result the system becomes stable with respect to electron disturbances.

Subsequent process development corresponds to up-to-date theory. State 2 is stable with respect to electron disturbances but the obtained potential distribution isn’t consistent with ion distribution function change in time. Since ions are generated on the emitter surface and high emitter potential jump restricts ions entrance from the emitter to plasma and also draws off them from plasma to the emitter, ion density in plasma decreases.

As a result the near-emitter layer width increases gradually (positions 3-5). At the same time the double ion-electronic layer appears at the emitter where negative charge density increases in time and potential in the whole gap is positive. With developing of this process the increasing of negative charge density leads to formation of the potential barrier restricting the current passing through the diode (positions 6, 7). The time of formation of the double ion-electronic field with the potential barrier at the emitter is defined by the rate of ion loss from interelectrode gap plasma and is $0.8 \cdot 10^{-6}$ s for the given case.

On creating the potential barrier at the emitter the neutral plasma region behind the double ion-electron layer is formed, changing into electron-ion layer near collector in which the main potential drop applied to electrodes is concentrated (position 8). This layer represents the transition region from dense plasma near the emitter to less dense one near the collector.

The dense plasma region expands to the collector with time (positions 9-11). The rate of plasma boundary displacement is equal to $1.3 \cdot 10^{-6}$ cm $\cdot$ s $\equiv 3.5 \sqrt{E_i} \cdot m_i^{-1}$, which agrees satisfactorily with [3]. With plasma expansion its potential is increased, hence, the potential barrier decreases and the current increases (position 11). Within current increase the critical value is reached and instability develops (position 12-1). The transition time from (pos. 11) to (pos. 2) $\sim 10^{-7}$ s and then the cycle repeats. The received sequence of potential distribution change according to current oscillation phases agrees with the results of numerical solutions in [10].

It should be pointed out that potential distribution with the virtual cathode is unstable to the positive potential disturbance. This
instability is expressed in applying to the diode electrodes at given phase of current oscillations rectangular (\\) - shaped positive pulse with duration several times larger than a current oscillation period, the fore-front pulse leads to the transition from the given phase to the phase with the near-emitter jump of the potential and maximum current for 1.10^-7 s. When applying negative pulse the field distributions in given phase tunes in a very short time (1.10^-7 s) to the acting potential. The pulse rear front representing potential positive jump relatively to pulse plateau again leads to the transition to the state with the near-emitter jump of the potential and maximum current. Such influence of the current pulse was marked in [9] too. It permits to register the initial phase of current oscillations, thus giving an opportunity to synchronize the oscillating process with the probing electron beam. This method of synchronization was used in the work.

REFERENCES

Kurchatov Institute of Atomic Energy, Moscow, USSR.

NOTATION

\[ U \] - useful voltage
\[ j \] - total current density
\[ j_{e,c} \] - density of electron current coming into plasma from emitter, collector.
\[ j_{e,o,co} \] - density of emission current from emitter, collector.
\[ J_e = j_e / j, J_c = j_c / j, J_{co} = j_{co} / j \]
\[ \xi_{e,c} \] - emitter, collector work function.
\[ \xi_{e,c} \] - potential barrier value with respect to emitter, collector Fermi level.
\[ k \] - Boltzmann constant.
\[ e \] - electron charge.
\[ T_{e,o,co} \] - emitter, collector temperatures.
\[ T_{ee,ec} \] - temperature of plasma electrons near emitter, collector.
\[ T_c = T_{co} / T_{ec}, \Phi_c = e \kappa_{e,c} / k T_{ec} \]
\[ \Phi_c \] - potential jump value near collector.
\[ \eta_c = e \Phi_c / k T_{ec} \]
\[ j_{i,e,ic} \] - ion current density to emitter, collector.
\[ eU_i^{ef} \] - energy returned on electrodes with ions and plasma radiations per one ion.
\[ r \] - fraction of electrons coming from a collector to plasma and coming back to the collector without change in energy.
\[ V_d \] - voltage drop in plasma.
\[ P_{cs} \] - caesium vapors pressure.
\[ d \] - interelectrode gap value.

INTRODUCTION

Real processes of charges transfer and volume ionization in an interelectrode gap are associated with...
electric energy losses and result in the decrease of net power and efficiency of a real thermionic converter in comparison with an ideal one in which the electron current limiting caused by the volume charge is absent.

Main components of electric power losses in the arc mode are following:
- the decrease of net current due to reflection of part of electrons from a plasma and their return to an emitter;
- the voltage drop in an interelectrode gap, needed to sustain a volume ionization;
- the dissipation of electron energy on a collector.

The last type of losses is inevitable for an ideal converter as well. It is related both to the conversion of portion of electrons potential energy being equal to the collector work function to a thermal one and to the removal of electrons' kinetic energy. In the arc mode the voltage drop in the interelectrode gap and electrons' energy dissipation on a collector are interconnected, since the both kinds of losses depend on the character of potential distribution and the function of electrons velocity distribution near the collector.

Regardless a large body of material obtained both in theoretical considerations of processes in plasma and accumulated in the course of experimental investigations, the processes in the collector area and problems of an influence of collector's work function and emission on energy parameters have not gained an unambiguous interpretation so far. It is this problem that should be considered primarily to predict limit parameters for the converter in the arc mode operation.

**EFFECT OF AN EMISSION FROM THE COLLECTOR ON A VOLTAGE DROP IN THE INTERELECTRODE GAP**

From the energy equation

\[ U = \gamma_e \frac{\gamma_e}{\gamma_c} \frac{2K}{2e} [T_e (T_{ee} - T_{eo}) - T_{ee} + T_{ec} \frac{1 - 0.5 \exp(-\gamma_c) + T_{ee} (1 - T_c) (1 - p)}{1 - 0.5 \exp(-\gamma_c) (1 + 0.75 \gamma_c)}] - U_i \frac{R_i + j}{j} \]

and potential balance in the interelectrode gap:
\[ U = \kappa e f - \kappa c - U_d \]  

(2)

It follows that the sum of two last terms in equation (1) equals to the voltage drop in the interelectrode gap. The last term in brackets determines a removal of electrons kinetic energy on the collector at the Maxwell velocity distribution with allowance for an angular asymmetry of distribution function. The multiplier \((1-r)\) takes into account that not all electrons arrived from the collector at plasma have time to change their kinetic energy in consequence of interaction with plasma electrons. A part of them, as a result of scattering on atoms without change in energy, will return to the collector. A probability of this process is high due to a low plasma concentration and low value of retarding potential jump near the collector.

In the absence of emission from the collector the potential jump formed near its surface is determined by the energy delivered to the near collector area due to the electron gas heat conductivity; and it provides a removal of this part of energy through a collisionless region, where the heat conductivity mechanism is absent. In this case about a half of this energy being equal to \( \Delta \kappa_e J \) is converted in a jump into a potential one. The jump value over the range of \( \tau_c = 0.3 - 0.6 \) is well approximated by the expression [1]:

\[ \eta_c = 0.11 + 0.275 \tau_c \]  

(3)

With an occurrence of emission from the collector a part of energy delivered due to electron gas heat conductivity is consumed for heating collector's "cold" electrons arriving at plasma. As a result, the jump value decreases. Fig.1 shows the dependence of \( \dot{J}_{co}(\tau_c) \) at vanishing of the collector potential jump for two values of reflection coefficient: \( r = 0 \) (curve 1) and \( r = 0.5 \) (curve 2). With a further growth of emission from the collector the jump changes its sign and begins to limit the emission. The energy balance equation (1) in this case will be of the form:
\[ U = \kappa_{\text{ef}} - K_{\text{f}} - \frac{2K}{e} \left[ J_{\text{e}} (T_{\text{ee}} - T_{\text{eo}}) - T_{\text{ee}} + T_{\text{ec}} \left[ 1 + 2J_{\text{c}} (1 - T_{\text{c}}) (1 - r) \right] \right] - \frac{J_{\text{e}} + J_{\text{f}}}{J_{\text{c}}} \]  

(4)

The collector effective work function \( \kappa_{\text{ef}} \) including the potential jump limiting emission, is related with a current coming from the collector into a plasma \( J_{\text{c}} \) by Richardson equation.

If the collector work function only is changed, then from the analysis of equations (1) and (2) one can obtain the value of dimensionless current from the collector into plasma \( J_{\text{c}}^{\text{opt}} \) at which a useful voltage should be maximum at a fixed passing current. In this case one should bear in mind that if the collector work function only is changed, the plasma parameters remain invariable [1,2].

\[ J_{\text{c}}^{\text{opt}} = \frac{T_{\text{c}}}{\gamma (1 - T_{\text{c}}) (1 - r)} \]  

(5)

Fig.1 shows \( J_{\text{c}}^{\text{opt}}(T_{\text{c}}) \) dependences for two values of reflector coefficient: \( r=0 \) (curve 3) and \( r=0.5 \) (curve 4). Thus, if there is a possibility to change independently the collector work function only, as fast as it decreases the useful voltage will increase until the emission current reach the value given by the expression (5). A further behavior of the useful voltage will depend on the value of potential jump limiting the emission. If this value will be change in such a way, that the value of current coming from the collector into plasma will remain unchanged at the level of \( J_{\text{c}}^{\text{opt}} \) the decrease in the collector work function will not result in the change of the useful voltage. Otherwise, irrespective of whether the current from the collector into plasma will increase or decrease in comparison with the \( J_{\text{c}}^{\text{opt}} \) value, the useful voltage will drop after attaining the maximum value.

The analysis of changes of experimental volt-ampere characteristics with the rise of the collector temperature in the region of its high emission [2] makes it possible to draw an indirect conclusion that while changing the collector work function only the useful voltage after reaching the maximum should remain unchanged due to the value kept constant. In this case, from the comparison of dependences \( J_{\text{c}}(T_{\text{e}}) \) at \( n_{\text{e}} = 0 \) and
it follows that the value $J_{c}^{opt}(T_c)$ should be retained approximately at the level of collector work function at which the potential jump vanishes. From the constancy of $J_c$ it follows that with the increase in density of passing current the collector effective work function must decrease. Incidentally, this fact explains the growth of slope of volt-ampere characteristics at a high emission from the collector.

Under real conditions of a caesium converter the collector work function at a constant pressure of caesium vapours changes only at the cost of variation in the collector temperature. In these conditions the optimum value of $J_c$ at which the useful voltage is maximum, will be different from expression (5) and will depend on a concrete dependence of $\lambda_{c}(T_c)$.

The results of analysis of equation (1) assuming that plasma parameters are invariable with changes only in collector's temperature and emission, are shown in Figs. 2 and 3. Fig. 2 gives dependences of optimum value of $J_{c}^{opt}$ on $T_c$ at $r=0$ for different functions of $\lambda_{c}(T_c)$ corresponding to real collectors. For a comparison the dependence $J_{c}^{opt}(T_c)$ is given (curve 5), when a work function changes independently of the collector temperature ($d\Phi_{c}/dT_c \to \infty$), as well as the dependence $J_{c}(T_c)$ (curve 6) corresponding to vanishing the potential jump. In fig. 3 analogous dependences are presented for the case $r=0.5$. From Figs. 2 and 3 is seen that the converter maximum useful voltage with real collector is realized at collector temperatures when an emission from it is still low enough and near the collector there exists the potential jump retarding plasma electrons.

If to take into consideration that the real collector is non-uniform in a work function and near its different spots the potential jump can have a different sign even when emission is absent, then, as it was shown in paper [1], the optimum value of $J_{c}^{opt}$ will be even lower. The corresponding region in fig. 2 is cross-hatched. The non-uniformity in the work function increases an realized work function of the collector approximately by 0.05-0.08 eV, which is due to a higher removal of energy from plasma to the non-uniform collector. The comparison of experimental results on polycrystalline and monocristalline collectors indicates that
the useful voltage in the last case is about 0.05-0.10 V higher, which conforms to the difference in the realized work function.

It is necessary to note that in an ideal converter the optimum value $J_{cc}^{opt} = J_{cc} / J^*$ is equal to expression

$$\frac{T_{cc} / T_{ce}}{1 - T_{cc} / T_{ce}}.$$ 

Thus for the collector work function which can be realized in ideal mode, is order 0.05-0.10 eV less than in the arc mode.

**OPTIMIZATION OF THE ARC MODE.**

The voltage drop in discharge characterizes energy consumptions to sustain an arc and equals to the difference in the maximum value of electrons' potential energy near by the emitter and the collector. The value $U_d$ is determined, firstly, by inelastic losses of electrons' energy at an excitation and ionization of caesium atoms - the last term in expression (1),(4); secondly, by the excess of electrons' kinetic energy removal from the plasma to electrodes over its arrival with emission electrons - the term in brackets.

On the section of characteristic with an excessive emission from the emitter, where $J_e$ and temperature of electrons are determined only by the value $P_{cs}, d$ [2] the voltage drop at fixed values of $T_{ce}, T_{cc}, J$ also depends on $P_{cs}, d$ only. Fig.4 shows $U_d(P_{cs}, d)$ dependences for various $T_{ce}$ at an absence of emission from the collector (a) and at its optimum value given by the expression (5)(b). The dotted line points to the dependence, when the data for electrons temperature are used from the paper [2], obtained on the assumption that $r=0.5$.

Fig.5 depicts dependences $U_d^{ef}$, where in addition to $U_d$, the losses related to the decrease of current due to a reflection of a part of electrons from plasma to the emitter are included in $U_d^{ef}$:

$$U_d^{ef} = U_d + \frac{e}{k} T_{cc} \ln J_e$$  \hspace{1cm} (6)

The optimum value of $P_{cs}, d$ increases slightly with the growth of emitter temperature. At a high emission from the collector it is somewhat higher than at a low one, due to the decrease of losses for heating the collector "cold".
electrons at a lower temperature of plasma electrons. The optimum $P_{c}$ value for $U_d$ is lower than for $U_d$ because of the decrease in back current from the plasma to the emitter.

As it was mentioned above, the optimum conditions on the collector at its given temperature are realized at the work function not exceeding the value corresponding to an optimum current from the collector to the plasma. With the growth of $P_{c}$ and appropriate drop of electrons temperature the value $J_{c}^{opt}$ increases and $h_c^{ef}$ decreases. An ion current to the emitter and, correspondingly, losses by inelastic processes increase with the growth of $P_{c}$ due to the increase in plasma concentration near by the emitter [2]. As a result, the specific power and efficiency of the converter at fixed values of $T_{eo}$, $T_{co}$, $j$ take maximum values at the definite value of $P_{c}$. Fig.6 illustrates dependences of specific power (solid lines) and efficiency (dotted lines) on $P_{c}$ for various densities of current at $T_{eo} = 2000$ K and $T_{co} = 800$ K under the condition that the collector work function is not higher than the value providing the optimum emission, and the emitter work function provides corresponding currents in the regime of excessive emission. From fig.6 is seen that $(P_{c} dB)$ is of the order of 1.0 torr.mm. With the decrease in $T_{eo}$ the optimum $P_{c}$ value is reduced and at $T_{eo} = 1600$ K it is of the order of 0.2 - 0.4 torr.mm. However, changes in both the specific power and in efficiency over the range of $P_{c} = 0.2 - 1.0$ torr.mm are negligible. The marked deterioration of parameters takes place only starting from $P_{c} > 1.2$ torr.mm.

Ultimate parameters depending on $T_{co}$ at $T_{eo} = 900$ K are shown in fig.7, and depending on $T_{co}$ at $T_{eo} = 2000$ K - in fig.8. Fig.9 presents dependences of minimum values of the barrier index $U_B = \frac{K_{o}}{e} \ln \frac{q_j \rho_{c}}{e^2} T_{co}^2 \cdot U$ characterizing total losses, including those thermodynamically unavoidable, for various densities of current at some temperatures of the collector. The highest experimental results of the arc regime are marked by points in the same place. From fig.9 is seen that at high $T_{co} = 1000 - 1100$ K, the best experimental data correspond to ultimate but at lower $T_{co}$ (especially at $T$ 800 K) the ultimate parameters are still far from reached.
The agreement of calculated ultimate parameters with the best experimental data at high collector temperatures evidences the validity of the procedure. The significant discrepancy between them observed at low temperatures of the collector is associated with the fact that materials providing a low work function in the given conditions are absent so far.

To reach limiting parameters at P_{cs} d ≅ 1.0 torr.mm the collector work function, depending on density of a current, should not exceed the value pointed out fig.10. The decrease of the required \( \phi_c \) with the growth of current density is stipulated, according to expression (5), by the increase in optimum value of current from the collector into the plasma

\[ j_c = j_c^{opt}. \]

With the descrease of \( P_{cs} \), d up to 0.2 torr.mm the value of the needed work function of the collector increases approximately by 0.1 eV at \( T_{ce} = 1100 \) K and about by 0.05 eV at \( T_{ce} = 800 \) K relative to the growth of \( T_{ce} \) and appropriate decrease of \( j_c^{opt} \).

The data of fig.10 were obtained on the assumption of complete energy relaxatation of emission electrons in plasma (\( r = 0 \)). At \( r = 0.5 \) the required work function of the collector should be approximately 0.05 eV less as compared with the data of fig.10.

The minimum value of the work function of present-day collectors is of the order of 1.4-1.5 eV. At working pressures of caesium vapors, determined by adsorption property of modern emitters, it is realized at comparativelly high, of the order of 1000-1100 K, temperatures of the collector. To reach ultimate parameters at \( T < 900 \) K the collector with \( \phi_c \leq 1.25 - 1.35 \) eV is necessary and with the condition that these values were realiazed at \( T_{ce} / T_{cs} \leq 1.4 - 1.5 \).

From fig.9 is see that the deviation of experimental parameters from ultimate increases with the rise of \( T_{ce} \). This is caused by a non-conformity of emitters to the requirements from the view-point of emission capacity at comparatively low \( P_{cs} \), determined by the optimum value \( P_{cs} \), d \( \leq 1.00 \) torr.mm at which plasma losses are minimum.

The emitter work function at a given \( P_{cs} \) depends on its
vacuum work function. In fig.11 there are dependences of the minimum vacuum work function $\psi_e$ on $T_{co}$ for two values of a gap and a number of $j$ values, which is necessary for obtaining an appropriate emission current $j_{co}$ at $P_{Co}$ \(d = 1.0 \) torr mm. If at low values of $j$ and $T_{co}$ a polycrystalline tungsten with $\psi_e = 4.5 \text{ eV}$ meets the above-mentioned requirements, then for high values of $j$ and $T_{co}$ the needed value of $\psi_e$ comes up to $5.3 \text{ eV}$, what can be met by the tungsten monocrystal with crystallographic orientation (110), iridium, platinum and oxygen-containing surfaces of metals. With reduction in the gap the requirements for the emitter vacuum work function are weakened but because of the increase of $P_{Co}$ the requirements on the parameters $T_{co} / T_{Co}$ at which the low work function of the collector should be realized become more stringent.

The low, of the order of 0.2 torr mm, values of $P_{Co} \cdot d$ do not lead so far to a marked growth of plasma losses but weaken requirements for the collector work function if in this case the value $P_{Co}$ decreases. But from the view-point of the emitter, this results in the necessity to have even higher vacuum work function as compared with the data in fig.11.

The contrary character of requirements on $P_{Co}$ from the view-point of emitter and collector gives rise to the growth of deviation of the reached parameters from ultimate ones with the decrease of $T_{co}$ and increase of $T_{Co}$.

REFERENCES


Fig. 1
Thermionic Energy Conversion

Figure 2

\[ \frac{d\phi}{d\theta} = \frac{2}{2}\phi - \frac{2}{2}\phi \]

\[ \phi = \frac{2}{2}\phi \]

\[ \frac{d\phi}{d\theta} = -2(2-1) \]

\[ \phi \]
$T_{co}=800\, K, \, J=3\, \% cm^2, \, J_{co}=0.$

Fig. 4a

$\eta_c \approx \frac{\kappa T_c}{\varepsilon} \ln \frac{4.12 \cdot T_{co}(T_{co}-T_0)(1-\chi)}{J}$

Fig. 4b

Thermionic Energy Conversion
\[ T_{co} = 800 \text{ K}, \quad j = 3 \text{ mA/cm}^2, \quad j_{co} = 0 \]

**Fig. 5a**

\[ U_{d}^{u}, V \]

**Fig. 5b**

\[ U_{d}^{e}, V \]

\[ T_{co} = 800 \text{ K}, \quad j = 3 \text{ mA/cm}^2 \]

\[ \chi \approx \frac{\kappa T_{e}}{e} \ln \frac{4 \cdot 120 T_{co}(T_{ec} - T_{co})(1 - r)}{j} \]

**Fig. 5b**

\[ \chi \approx \frac{\kappa T_{e}}{e} \ln \frac{4 \cdot 120 T_{co}(T_{ec} - T_{co})(1 - r)}{j} \]

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Fig. 6

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Fig. 7

Fig. 8
1 - D. Lieb, B. Gunther and al., Eindhoven, 1975 (W-W+WO3)
2 - G. Stax, M. Saunders, D. Lieb, 15th IECEC (Mo-Mo+O)
3 - D.B. Goodale, C. Lee, D. Lieb, P.F. Ottenger, 14th IECEC (W-W+WO3)
4 - E.A. Smith, Huffman, 19th IECEC, 1984 (W-Mo+O)
5 - F. Rufeh, D. Lieb, Topical Report NTE 4086-3-30, 1989 (W-Mo)
6 - B. Gunther, D. Lieb and al., Summary Report NTE 4188-6-74, 1974 (W-W+WO3)
7 - Результаты автора (W-Mo)

Fig. 9c

9 - G.L. Hatch, L. Manote, E.J. Britt, 14th IECEC, 1979 (Re-Mo)
10 - V.C. Wilson, J. Lawrence, TCSC, 1967, Palo Alto (W-NB)
11 - D. Lieb, F. Rufeh, TCSC, Miami, 1980 (W-NB)

Fig. 9d
Fig. 10

Fig. 11
1. Introduction

In our continuing experimental program to understand the basic processes in Thermionic Energy Converters we use mainly molecular beam and mass spectrometric methods. From these studies we have recently reported on the observation of negative ions of cesium, Cs\(^-\) [1]. In other experiments with cesium plasmas and cesium covered surfaces at high temperatures we have identified both highly excited (Rydberg state) atoms Cs\(^*\) and doubly excited cesium atoms Cs\(^{2*}\), which can carry considerable excitation energies [2,3].

We also continue our studies of real thermionic diodes of the open type we have used previously [4]. From the present study, we report now on the I-V curves when the conditions are chosen to give a large fraction of excited Cs atoms in the plasma.

2. Results

Fig. 1 shows a typical I-V curve taken under conditions where mass spectrometric sampling measurements show that field ionizable Rydberg states of Cs are common in the interelectrode space. Such highly excited states are easily and rapidly field ionized in even very weak fields. For example, a principal quantum number of 40 gives a diameter of the atom of 0.17 \(\mu\)m and an electron binding energy of 9 meV. This gives [5] a limiting ionizing field strength of 12 V/mm, which is easily reached in the converter.

A large number of experiments have been done, where the curve shape has been recorded under Rydberg conditions. The point where the Rydberg field ionization current appears should in the first approximation correspond to zero field strength in the converter. However, there must exist a certain low field strength before the ionization takes place, corresponding to a total voltage over the
converter space of $\Delta V$, as depicted in Fig. 2A. The point of zero field strength is, at least in principle, the point where the arc drop is also equal to zero, and then

$$V_{\text{out}} = \Phi_E - \Phi_C + \Delta V$$

is valid, where $\Phi_E$ is the emitter work function and $\Phi_C$ the collector work function. In Fig. 2B, the ordinary situation with an arc drop is shown. Since in that case

$$V'_{\text{out}} = \Phi_E - V_B$$

is true, where $V_B$ is the so-called barrier index, the relation

$$V_B = (V_{\text{out}} - V'_{\text{out}}) + \Phi_C - \Delta V$$

can be derived. In Fig. 3, the results for $V_B$ and $(V_{\text{out}} - V'_{\text{out}})$ are plotted. It could be assumed that $\Delta V$ is between 0.1 and 0.4 V. An extrapolation to zero then gives a common value of 1.0-1.3 V for the collector work function, and an arc drop $(V_{\text{out}} - V'_{\text{out}} - \Delta V)$ greater than 0.6-0.9 V in all cases. Of course, this treatment assumes that the work functions are unchanged on both the emitter and the collector at the two points depicted in Fig. 2.

3. Discussion

It is shown in many cases, that the arc drop, which includes several types of losses in the plasma and at the electrode surfaces, is the main cause of the non-ideal behaviour of thermionic converters. The size of the arc drop in the present cases is large, but the conditions in the converter have been chosen to demonstrate the existence of Rydberg states in the plasma. The simple analysis presented is only approximate, but points to the fact that the arc drop may be a large factor in the barrier index $V_B$. This agrees with the measurements of very low work functions of converter surfaces at realistic surface temperatures [6,7]. Thus, the future development of thermionic converters will depend on means of bringing the arc drop down to very low values.

As shown in Fig. 1, the linear variation of the Rydberg current with applied voltage indicates an almost metallic current transport through the plasma. The corresponding resistance is approximately 1 Ohm. This metallic current transport indicates an entirely different transport process with other loss processes than the ordinary electron transport through the plasma. This could mean new possibilities to maintain a current with low losses in thermionic converters.

4. Conclusions

The existence of a large field ionization current due to weak field ionization of very highly excited Rydberg states of Cs is demonstrated in a thermionic converter plasma. By an approximate theory, it is shown that Rydberg ionization can help to give separate values of the two dominating factors in the commonly used parameter, the barrier index. This means in the present case that arc drops...
greater than 0.6-0.9 V are found, similar in size to the collector work function which is 1.0-1.3 V. Finally, the possibility to use a more or less "metallic" mode of current transport through the converter is discussed.

Acknowledgements

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References


Figures

Fig. 1. Two oscilloscope pictures of converter I-V curves. Vertical scale 12.5 Acm⁻²/div, and horizontal scale 2 V/div. Emitter temperature was approximately 1500 K.
Fig. 2. Potential energy diagrams for electrons in the converter. A: at the onset of field ionization, and B: at the point where the current goes to zero in the first quadrant.

Fig. 3. Barrier index for a large number of runs, plotted against a modified arc drop, \( (V_{\text{out}} - V'_{\text{out}}) \), defined in Fig. 2. One line is a linear regression to all the data points, the other one is a line with the 45° slope predicted in the text.
Excited states of Cs in a thermionic energy converter

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1. Introduction

In previous publications from our group we have reported on the existence of highly excited states of Cs and Cs\textsuperscript{-} in the flux from a thermionic energy converter plasma and also from hot metal surfaces [1]. A more detailed mass spectrometric investigation has now been carried out to identify the excited states of Cs and other species in the plasma, and their relation to the converter characteristics. The characteristics of singly excited atoms in high Rydberg states are quite well known [2]. Such states can be formed both by atoms and molecules [3,4], and they are easily field ionized even at quite low field strengths [5]. Not very much is however known about doubly excited states or their reactions. In the present experiments, they originate from a thermal plasma. Exiting the two outermost electrons gives an energy up to 28.9 eV, which means that doubly excited atoms are energetic enough to ionize most atoms and molecules. They may be quite long-lived due to correlation effects between the two excited electrons, and we estimate their time-of-flight in the present experiments to be at least 200 μs.

2. Experimental

The experiments were performed with our "open" thermionic converter [6]. The cesium is brought into the interelectrode space through many small laser machined holes in the collector. Between the electrodes a voltage is usually applied, about 5 V AC at a frequency of 50 Hz, to ignite the plasma. In the chamber surrounding the converter a pressure of 10\textsuperscript{-5} mbar is maintained with diffusion pumps. The flux leaving the plasma is sampled in molecular beam form into a quadrupole mass spectrometer in a separately pumped chamber at 10\textsuperscript{-6} mbar. In front of the mass spectrometer, a shielded grid at a variable voltage is mounted in the present experiments. An extractor slit is located between the ion source and the quadrupole. In Fig. 1, the apparatus used is shown schematically.
3. Results

In the mass spectra, exemplified in Figs. 2 and 3, several mass peaks are observed without any electron emission in the source, i.e. with field ionization at field strengths up to 600 V/cm. This indicates that a large part of the flux is in high Rydberg states, which are known to field ionize under such conditions. The large peaks of Cs$^{2+}$ often found indicate the existence of doubly excited atoms Cs$^{2*}$. In the flux from the converter, the states Cs$^*$, Cs$^{2*}$ and probably H$_2^*$ are identified by field ionization. Gas inlets into the converter is shown to give rise to other field ionizable excited states, like N$_2^*$. The doubly excited states Cs$^{2*}$ react rapidly with gases like CO$_2$, O$_2$, N$_2$ in the ion source of the mass spectrometer, giving rise to singly and sometimes doubly charged molecular ions. This is shown in Fig. 3 for the case of CO$_2$. Due to its small excitation energy, Cs$^*$ does not give reactions like this.

4. Discussion

Both singly and doubly excited states of Cs are detected in the flux from the plasma. Also other excited states with large n can be observed directly from the plasma, e.g. of N$_2$ and H$_2$. The field strength necessary to field ionize a singly excited Rydberg state is proportional to quantum number n as 1/n$^4$. At a field strength of 100 V/cm, all atoms with n>42 are field ionized, and at 600 V/cm, with n>27.

One interesting aspect is the large reactivity of the doubly excited states of Cs. The reaction processes we have observed are due to electron excitation energy transfer from doubly excited Cs$^{2*}$ atoms, giving highly excited Rydberg states of atoms and simple molecules, which are easily field ionized. From the size of the energy transfer cross sections, between 4000 and 8000 $\AA^2$, we calculate principal quantum numbers approximately equal to or larger than 10 for the doubly excited states of Cs.

This large reactivity may be an important factor to consider in the construction of thermionic converters, since it may severely limit the number of materials which will survive in a thermionic converter environment. As described by reports from our group, excited alkali atoms may also be responsible for the strong catalytic promotor effects of alkali atoms in ordinary heterogeneous catalysis [7]. This indicates that surface corrosion effects will be severe if impurities (other than rare gases) are allowed to enter the converter space. Further, the facile formation of excited states in the plasma, which have large cross sections for electron collisions, makes it necessary to preserve the cleanliness of the converter space for long times.

Acknowledgements

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References


Figures

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Fig. 1. A schematic of the experimental arrangement.
Fig. 2. A typical field ionization mass spectrum with large mass peaks at m/z 133 and 66.5.

Fig. 3. A field ionization mass spectrum with CO$_2$ admitted into the ion source.
1.0 INTRODUCTION

Two new computer models have been developed for converter diagnostics. The first is a model of the unignited (diffusion) mode which is used for inferring the emitter temperature in in-core experiments.* The second is a model of plasma decay in a mixed cesium-argon pulse-ionization diode.** This model is intended to investigate the mechanisms leading to long decay times and high output power in such converters. Comparisons with experimental data have shown very good agreement for both models.

The unignited mode model was designed to aid the inference of emitter temperature using the electron-rich unignited mode. This mode is extremely useful for diagnostics because the form of the I-V characteristic depends more strongly on the emitter temperature than on other converter parameters. Comparisons of calculated results with data from both planar and cylindrical converters show extremely good agreement. The program has been used in conjunction with a computer-aided data acquisition system to guide the start-up and long term diagnostics of fueled cells in test reactors.

The model of the pulsed diode is used to explore the factors which affect the decay phase duration and output power. Comparison with data from a variable-spacing, planar pulsed-ionization diode gives very good agreement.

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2.0 BASIC PHYSICAL MODEL DESCRIPTION

In the case where charged particles make several collisions in crossing the interelectrode space, the plasma can be described by a set of transport equations [1]. These equations assume that there are enough collisions in the interelectrode space and the electric field is so small that the inertia of the charged particles can be neglected. They are given by:

\[ J_e = -\mu_e \left[ \frac{an}{e \partial x} + enE + (1+k_e)nk_e \frac{\partial T_e}{\partial x} \right], \]  

(1)

\[ J_i = -\mu_i \left[ \frac{an}{i \partial x} - enE + (1+k_i)nk_i \frac{\partial T_i}{\partial x} \right], \]  

(2)

\[ q_e = \left[ \frac{5}{2} + k_e \frac{T_e}{e} + \psi \right] J_e - \kappa T_e \frac{\partial T_e}{\partial x}, \]  

(3)

\[ E = \frac{\partial \psi}{\partial x}, \]  

(4)

\[ \frac{\partial J_e}{\partial x} = \frac{\partial J_i}{\partial x} = e(S_{an} - an^3 - \frac{an}{\partial t}), \]  

(5)

and

\[ \frac{\partial q_e}{\partial x} = (\psi - V_i \frac{\partial J_i}{\partial x} - \frac{3}{2} \frac{\partial}{\partial t}(nkT_e) - eV_i \frac{\partial n}{\partial t} - \Delta E_{rad}. \]  

(6)

Where:

- \( J_{e,i} \) is the electron (ion) current,
- \( \mu_{e,i} \) is the electron (ion) mobility,
- \( k \) is Boltzmann's constant,
\( T_{e,i} \) is the electron (ion) temperature,

\( n \) is the plasma density,

\( E \) is the electric field,

\( k_{e,i}^T \) is the electron (ion) thermal diffusivity ratio,

\( q_e \) is the electron heat flux,

\( \psi \) is the electron potential,

\( \kappa_e \) is the electron thermal conductivity,

\( S \) is the ionization coefficient,

\( n_a \) is the density of neutral cesium atoms,

\( \alpha \) is the three-body recombination coefficient,

\( V_i \) is the cesium ionization potential,

\( \Delta E_{\text{rad}} \) is the electron energy loss by radiation.

Boundary conditions for the transport equations come from an analysis of the electrostatic sheaths adjacent to the electrodes [2]. In this analysis, the sheaths are assumed to be of negligible thickness. The ions near the electrodes are assumed to be in thermal equilibrium with the electrodes. Non-monotonic sheaths and the Schottky effect, important in the ignited mode, have been neglected in this treatment.

At the emitter, for a positive (electron-retaining) sheath:
\[ J_e = J_R - (J_{re} - J_e/2)e^{\frac{-eV_E}{kT_e}}, \]
\[ J_i = J_i e^{\frac{eV_E}{kT_e}} - (J_{ri} - J_i/2), \]
\[ q_e = J_R(2kT_e + V_E) - (J_R - J_e)(2kT_e + V_E). \]

For a negative (ion-retaining sheath):

\[ J_e = J_R e^{\frac{eV_E}{kT_e}} - J_{re} - J_e/2, \]
\[ J_i = J_i e^{\frac{eV_E}{kT_e}} - (J_{ri} - J_i/2)e^{\frac{eV_E}{kT_e}}, \]
\[ q_e = J_{re}(2kT_e - 2kT_e) + J_e(kT_e + kT_e). \]

At the collector, for a positive collector sheath:

\[ J_e = (J_{re} + J_e/2)e^{\frac{-eV_C}{kT_e}} - J_C, \]
\[ J_i = J_{ri} + J_i/2, \]
\[ q_e = J_e(2kT_e + V_C + \psi_C) + J_C(2kT_e - 2kT_C). \]

For a negative collector sheath:

\[ J_e = J_{re} + J_e/2 - J_C e^{\frac{eV_C}{kT_C}}, \]
\[ J_i = (J_{ri} + J_i/2)e^{\frac{eV_C}{kT_C}}, \]
\[ q_e = J_{re}(2kT_e - 2kT_C) + J_C(kT_e + kT_C + \psi_C). \]
The random electron (ion) current is calculated by:

\[ J_{re,i} = \frac{e\bar{v}_{e,i}}{4}, \]

where

\[ \bar{v}_{e,i} = \left( \frac{8kT_{e,i}}{\pi M_{e,i}} \right)^{1/2}. \]

The symbols used here are:

- \( V_{E,C} \) is the emitter (collector) sheath height,
- \( T_{E,C} \) is the emitter (collector) temperature,
- \( \psi_C \) is the electron potential at the collector sheath-plasma interface,
- \( J_R \) is the Richardson emission current density from the emitter,
- \( J_{iE} \) is the Saha-Langmuir ion current density from the emitter,
- \( J_C \) is the electron current density from the collector,
- \( M_{e,i} \) is the electron (ion) mass,
- \( \bar{v}_{e,i} \) is the average thermal velocity.

3.0 **UNIGNITED MODE MODEL**

The unignited mode model was developed for the purpose of accurately inferring the emitter temperature of in-core thermionic converters by fitting the unignited mode portion of the I-V curve with calculated results. The
unignited mode has several advantages for emitter temperature diagnostics. Under typical conditions for TFE tests the unignited mode is in the "electron-rich" regime, where the I-V curve is insensitive to parameters other than the emitter temperature, interelectrode spacing, cesium pressure, and collector work function. In addition, the unignited mode saturation current or ignition current is insensitive to collector work function but is very sensitive to emitter temperature.

In the unignited mode model the time derivatives in Eqs. 5 and 6 are neglected in order to give the steady state case. Numerical solutions are obtained for the transport equations and boundary conditions. The method of "shooting" to a fitting point in the center of the interelectrode space is use [3]. In this method, some of the unknown values at the electrodes are guessed in order to provide initial conditions for the transport equations. These are \( T_e(0), V_E, T_e(d), V_C, \) and \( \psi_C \). An efficient numerical integration routine is used to integrate the transport equations from the electrodes to the center, where the degree of mismatch in \( T_e, n, \psi, J_e, \) and \( q_e \) is evaluated. The unknown values are adjusted so as to make the mismatch small. The method depends on a good starting guess for the unknown values in order to converge. This is provided by an approximate unignited mode model. In calculating a current density-voltage curve the values at the previously calculated point are used.

The cesiated emitter work function is calculated from the emitter temperature cesium pressure and bare work function using the Rasor-Warner formulation [4]. The cesiated collector work function is calculated from an analytical fit to measurements on planar thermionic converters with the same electrode materials as the H-series cells.

It has proven to be important to retain volume ionization, recombination and radiation terms in the formulation in order to accurately match I-V curves at high emitter temperatures. These terms are normally neglected in descriptions of the unignited mode. For emitter temperatures above about
1700 K there is no distinct saturation region in the J-V curve. Inclusion of ionization, recombination, and radiation in the model enable the ignition portion of the I-V characteristic to be modeled accurately. This is a beneficial feature when fitting a calculated curve to data at high emitter temperatures.

4.0 UNIGNITED MODE MODEL RESULTS AND COMPARISON WITH DATA

A comparison between calculated and experimental volt-ampere characteristics in the electron-rich, unignited mode is shown in Fig. 1. The experimental curve was obtained from the PD-6 variable-spacing planar converter. Excellent agreement is obtained between calculated and experimental results. The ionization/recombination terms included in the model enable the ignition behavior of the I-V curve to be accurately calculated in both the forward and reverse directions. Additional comparisons with experimental data have shown that the model remains accurate over a wide range of collector temperatures and cesium pressures.

The effect of emitter temperature over a range from 1600-1800 K is shown in Fig. 2. Again, a high degree of accuracy is obtained.

A match to experimental data from an in-core cylindrical converter is shown in Fig. 3. Here the emitter temperature is used as a fitting parameter. A very good match is obtained at an emitter temperature of 1745 K.

The computer model has been incorporated into a system for real-time display and analysis of experimental I-V characteristics. It has been used to guide the start-up and continuing diagnostics of two in-core converters.
Fig. 1  Comparison between Unignited Mode model calculation and PD-6 data
Fig. 2 Match between Calculated Results and PD-6 Emitter Temperature Family
Fig. 3  Calculated fit to 1H1 in-core cell data

- $T_E = 1745 \, K$
- $T_C = 1115 \, K$
- $T_R = 601 \, K$
- $D = 10 \, \text{mils}$

Calculated = Solid Curves
5.0 PULSED-IONIZATION DECAY MODE MODEL DESCRIPTION

The physical model is of a partially ionized vapor of cesium and inert gas, with cesium as the sole ionized species. For this model Eqs. 1 through 6 are combined into two equations: the ambipolar diffusion equation which describes the change in the plasma density, and the energy equation which describes the change in the electron temperature. The formulation is similar to a previous description by V. A. Zherebtsov [5].

The ambipolar diffusion and energy equations form a set of two second-order non-linear differential equations with non-linear boundary conditions. As such, they must be solved numerically. It was required that the numerical method minimize complexity and provide for a high degree of modularity in program construction. Previous experience indicated that simple finite-difference methods suffered from instability and cumbersome algebra. The method of lines was selected to solve this problem. In this method, all spatial derivatives in the equations are replaced by finite difference approximations while the derivatives in time are left alone. This results in a system of coupled ordinary differential equations with initial conditions.

Due to the inclusion of the energy equation in the formulation, the problem of multiple time scales is introduced. The characteristic time for electron temperature is on the order of 10 nanoseconds, while the characteristic time for the plasma density is on the order of 10 microseconds. If the time step for the numerical integration is larger than 10 nanoseconds, even after the electron temperature has reached equilibrium, the solution becomes unstable. Such systems of equations are referred to as stiff systems. Fortunately, there exist efficient algorithms to solve stiff equations. Such methods require that the Jacobian matrix of the problem be provided. Due to the complexity of the expressions for the time derivative, numerical differences are used to estimate the partial derivatives which form this matrix.
The initial condition for the plasma density and electron temperature is set arbitrarily. Results from the model confirm that after an initial transient period of about 1 microsecond the behavior of the plasma is insensitive to the initial condition, provided that the density and temperature start out higher than a certain level. For most of the calculations done to date spatially independent values of $10^{14}$ cm$^{-3}$ for the density and 2500 K for the electron temperature were used.

Two versions of the program were developed to treat constant current density or constant voltage decay. Constant voltage requires more computational work than constant current due to the need to iterate to find the value of current density and the much more complex structure of the Jacobian matrix.

The mobilities and thermal diffusivity ratios for ions and electrons, as well as the electron thermal conductivity, must take into account coulomb scattering and scattering by cesium and inert gas neutrals. It was found in development of the model that inclusion of the energy dependence of electron-neutral scattering was important to achieving a good match between calculation and data, particularly at low current densities. Energy dependent values were used for the electron-cesium [6] and electron-argon [7] collision cross-sections. The method of expansion in Sonine polynomials [8] was used to calculate the electron mobility, thermal conductivity, and thermal diffusivity ratio. This gives a set of simultaneous linear equations in which the coefficients are computed by a numerical integration. These equations are then solved to yield the required electron transport coefficients. Values for the cesium-cesium ion and argon-cesium ion scattering were taken from Houston [9] and Bogdanov, et. al. [10], respectively. Values for cesium ionization and recombination coefficients were taken from results by Lawless [11].
6.0 PULSED-IONIZATION MODEL RESULTS AND COMPARISON WITH DATA

A comparison between a calculated and an experimental decay envelope family is shown in Fig. 4. Decay envelopes result from measuring the decay of voltage at constant current at various current values. The experimental results were taken from a variable spacing planar diode with a platinum (111) single crystal emitter. Measured values were used for the cesiated emitter and collector work functions.

The small diameter of the electrodes in the experimental diode results in plasma decay by radial ion loss becoming important under some circumstances. Full two-dimensional treatments of the transport equations required excessive amounts of computer time. Accordingly, radial loss is treated in an approximate way. The term \( n/r \) is subtracted from the right-hand side of Eq. 5, where

\[
r = \frac{1}{D_a} \left( \frac{R}{2.404} \right)^2.
\]

Where \( D_a \) is the ambipolar diffusion coefficient and 2.404 is the first zero of the \( J_0 \) Bessel function. A similar term can be used in Eq. 6 to account for radial loss of electron energy.

Fig. 5 indicates the expected effect of radial ion loss in the experimental geometry. The dashed curves in Fig. 5 are the same as the calculated curves in Fig. 4 and correspond to an electrode radius of 0.55 cm. The solid curves correspond to an essentially infinite radius. As can be seen, the finite radius of the experimental electrodes caused a slight decrease in decay time.

The curves in Fig. 5 are compared with an ideal diode characteristic. The results show the voltage loss due to the relatively high value of plasma resistance. This is due to the low value of electron temperature (about equal to the emitter temperature) and the large interelectrode gap (1 mm). The
**Fig. 4**  Comparison between a Calculated Decay Family and Experimental Data

*Experimental Value Approximate
Solid Lines - Experimental
Dashed Lines - Calculated

- $T_E = 1500 \text{ K}$
- $T_C = 800 \text{ K}$
- $T_{CS} = 475 \text{ K}$
- $d = 1 \text{ mm}$
- $P_{Ar} = 5 \text{ torr}$
- $R = 0.55 \text{ cm}$
- $\Phi_E = 2.05 \text{ eV}$
- $\Phi_C = 1.52 \text{ eV}$
- Constant Current Decay
Fig. 5  Effect of Radial Ion Loss
resistance can be reduced by reducing the interelectrode gap. Operation of pulsed diodes at high current densities requires that methods be perfected of increasing decay time in order to compensate for the decay time reduction resulting from the smaller gap.

Fig. 6 indicates the expected change in the decay family if the decay had been at constant voltage instead of at constant current. At a voltage of 0.6 volts and a decay duration of 80 microseconds the combination of radial loss and constant current decay causes the current density to be 50% lower than would have been possible with no radial loss and constant voltage decay. This indicates the utility of the computer model in estimating effects of non-idealities in the experimental set-up.

Figs. 7-10 examine the details of the plasma parameters during decay. The decay is calculated at a constant 0.6 volts for the conditions used in Fig. 6. The time dependence of current density is shown in Fig. 7. The calculation indicates the reduction in the rate of decay at low current densities as a potential well for ions forms. This effect is shown in the motive diagram in Fig. 8, which indicates the growth of ion-retaining sheaths at the electrodes.

Fig. 9 shows the time and spatial dependence of the plasma density. The density falls almost two orders of magnitude from its initial value to a level of about $10^{12}$ cm$^{-3}$ at 80 microseconds. After 20 microseconds the slope of the density at the emitter is nearly flat, indicating that there is very little plasma loss there. This is due to the rise of a large ion-retaining emitter sheath. As an ion-retaining sheath is formed at the collector, the slope at that side is reduced as well.

Fig. 10 shows the time and spatial dependence of the electron temperature. At 1 microsecond the average temperature is close to the emitter temperature, which gives rise to a high plasma resistivity as previously discussed. As time progresses, the electron temperature near the collector rises due to ohmic heating. This is similar to the electron temperature in the unignited mode.
Fig. 6  Comparison of I-V curves with constant current and constant voltage decay
All Conditions as in Fig. 6
Constant Voltage Decay
V = 0.6 Volts

Fig. 7 Time dependence of current
All Conditions as in Fig. 6
Constant Voltage Decay

Fig. 8 Time evolution of motive diagram
All Conditions as in Figure 6
Constant Voltage Decay
\( V = 0.6 \) Volts

Fig. 9 Time evolution of plasma density
Fig. 10  Time evolution of electron temperature

All Conditions as in Fig. 6
Constant Voltage Decay
$V = 0.6$ Volts
Figs. 11 and 12 examine the possibility of improved performance by reducing the cesiated electrode work functions, something which may result from improved control of cesium-oxide vapor sources. The decay is constant current, with no radial loss for these cases. In Fig. 11 decay families are compared for collector work functions of 1.52 and 1.30 eV. The effect is very nearly to simply add the difference in output voltage to each decay envelope. In Fig. 12 the emitter work function has been reduced in order to increase its emission current density from 35 to 70 amperes/cm$^2$. As seen, this has the effect of a very large increase in decay time, particularly at the larger current densities. For a working current density of 6 A/cm$^2$ and an emission current density of 35 A/cm$^2$, the voltage decays to zero in about 25 microseconds. The same case with 70 A/cm$^2$ emission takes about 60 microseconds to decay to zero.

7.0 DISCUSSION, CONCLUSIONS, AND FUTURE WORK

The unignited mode computer model provides the ability to fit unignited mode I-V characteristics with calculated curves and has proved to be a very useful and accurate way to determine emitter temperatures in in-core thermionic experiments. Future efforts will apply this technique to experiments with series-connected strings of thermionic cells, all with different emitter temperatures.

The decay phase computer model has been shown to accurately describe experimental data in the regime of interest. It has indicated the quantitative effect non-idealities in the experiment have on the results. It has indicated that performance improvements can be realized by further reduction in collector and emitter work functions.
Fig. 11  Effect of reducing collector work function
Fig. 12 Effect of reducing emitter work function

Current Density (A/cm²)

Electrode Voltage

$T_E = 1500 \text{ K}$
$T_C = 800 \text{ K}$
$P_{CS} = 0.08 \text{ torr}$
$P_{Ar} = 5 \text{ torr}$
$d = 1 \text{ mm}$
$\phi = 1.52 \text{ eV}$
$R = 10 \text{ cm}$

Constant Current Decay
Solid Lines - $\phi_E = 2.05 \text{ eV}$
Dashed Lines - $\phi_E = 1.96 \text{ eV}$
REFERENCES


2. Ibid, pp. 428-430.


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THERMIONIC PROPERTIES OF NIOBIUM-OXYGEN MONOCRYSTALS USED AS CAESIUM THERMIONIC CONVERTER COLLECTOR

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It is known that caesium and oxygen double layers on niobium can have low work function (1.1-1.3 ev). These layers are usually formed by consequent adsorption on niobium surface, first of oxygen then of caesium. At temperatures higher than 800-900 K these layers are unstable due to volume diffusion of oxygen.

In works by R.Pantel, M.Bujor, J.Bardolle [1] it is noted that the surface structures are formed at segregation of oxygen solved in metal. This effect can be used to form stable at elevated temperatures oxygen-caesium layers on thermionic converter collector. Polycrystalline compositions of niobium oxygen have high brittleness. In the difference from polycrystals niobium monocrystals containing 0.05% of oxygen have satisfactory plastic properties and known treatment methods can be applied to them: grinding, electrolytic polishing, cutting, electron-beam welding, so they can be used as thermionic energy converter (TEC) collector material.

The investigation of adsorption-emissive properties of (110), (111) planes of niobium-oxygen monocrystals, produced by diffusion saturation of niobium with oxygen was held in the present work. Work function of the said planes was measured in vacuum and barium. The reciprocal tungsten emitter and niobium-oxygen collector influence on their emissive properties during TEC work was also investigated.

The experimental samples were made of niobium polycrystal or monocrystal rods obtained by electron-beam zone melting. Samples with diameter 20 mm and height 4-5 mm and height 4-5 mm were made from monocrystal rods with (110) and (111) axis orientation. The deformed layer was removed by grinding and electrolytic polishing, then samples were saturated with oxygen. Oxygen concentration was measured by neutron activation method. With oxygen concentration increase samples microhardness varied in a linear mode from 0.7-0.9 GPa for initial samples to 3.2-3.4 GPa for oxygen 0.5 mass %. The oxygen in niobium was in way of solid
solution. The microhardness measurements along the sample cross section showed their uniform saturation with oxygen. X-ray and microscopic investigations did not show significant structural variation or second phase precipitation after their saturation with oxygen.

The scheme of experimental device is shown at fig. 1. Six samples were simultaneously mounted in a mobil and during measurements could take two places: opposite Faraday cylinder during adsorption properties measurements - or opposite tungsten cathode when these samples were used as diode anode. Gas release from collector and mass transfer of tungsten from emitter to collector were simulated in a diode.

Caesium spraying was help from thermostate and barium spraying directly from ampoule. The residue gases pressure during the measurements did not exceed 5.10⁻⁶ Pa.

The samples thermovacuum preparation (TVP) was made by step annealing at 900 K - 80 hours, 1300 - 40 hours, 1600 K - 20 hr., 2000 K - 1 hr. Each annealing regime ended with surface adsorption properties measurements by measuring thermionic current at $T = 450-500$ K in caesium flow of constant intensity.

During TVP S-shaped curves of emission current from samples in caesium constant flow shifted with reverse temperature ($1/T$). All this shows that samples surface adsorption increases with gas release. For samples with oxygen concentration up to 0.2% curves shift took place after every annealing step, for samples with higher oxygen concentration after $T = 1300$ K, 40 hr annealing S-curves stabilisation took place (see fig. 2). It was the evidence that samples surface at 1300 K already cleans from impurities like carbon, sulphur.

Work function values $\frac{U}{T/T_R}$ - is caesium reservoir temperature obtained by processing S-curved measured on TVP-treated samples are shown on fig. 3. With oxygen con-

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Fig. 1. The scheme of the experiment

1 - sample; 2 - yoke; 3 - electron heater; 4 - Faraday cylinder; 5 - caesium reservoir; 6 - steam pipe-line; 7 - effusive hole; 8 - water cooling screens; 9 - barium ampoule; 10 - immobil electrode
Fig. 2. S-curves of the sample (111) Nb–0.25% O measured in the TVP process. Caesium atoms flow 1.10⁻⁶ at/cm². s

The annealing regimes: 900 K, 800 h; o 1300 K, 40 h; △ 1600 K, 18 h; Δ 1900 K, 1 h.

Concentration increase minimal work function decreases from 1.50–1.55 eV to 1.35–1.40 eV. The $U/T_{TR}$ curves slope to the left of the minimum is also decreases. In the similar adsorption conditions minimum for (110) samples can be seen at temperatures of 60 K higher then for (111) samples.

S-curves measurements held at different values of caesium flow showed that adsorption heat of caesium

Fig. 3. $U/T_{TR}$ dependences of the monocristals (111) Nb–0, (111) NbO

a) o (111) Nb; △ (110) Nb–0, 2%O
   △ (110) Nb–0,35%O △ (110) Nb–0,5%O
b) o (111) Nb; X (111) Nb – 0,2%O
   △ (111) Nb–0,25%O △ (111) Nb – 0,5%O

Initial close packing surface structure with loose (110) NbO, (220) NbO and (111) NbO structures with corresponding work function decrease. This very process can be used to
for oxygen-containing samples is decreasing with the approach to minimal work function value (see fig. 4). This can explain the slope decrease in the left part of fig. 3.

The (110) samples work function in vacuum decreases with oxygen concentration growth from 4,65 to 4,4eV and (111) samples work function increases from 4,1 to 4,4 eV (see table 1).

Barium adsorption decreases work function of (110) Nb – 0,35% O sample to 1,6 eV and (111) Nb – 0,5% O sample to 1,7 eV. (110) samples work function versus temperature dependence curves have plateaux 1,7 eV; 2,4 eV; 2,7 eV values in barium flow.

Barium layers thermal desorption in vacuum showed existence of stable states with the same work function values corresponding temperatures 750 K, 1050 K, 1480 K. The same behavior was seen at barium desorption from (111) samples.

The results achieved can be described satisfactory with the help of existing data on caesium and oxygen adsorption on niobium monocrystal surface. For example, in works [1-3] at oxygen adsorption on Nb (110) plane the consequent change of initial explain Nb (110) samples work function decrease with oxygen concentration growth.

Caesium, adsorption investigations in niobium oxidation phases [4] showed that Cs-O on Nb (111) structures is in depolarized states thus providing higher values of minimal work function. In our case, with surface segregation of the solved oxygen, (110) samples gave lower work function values in barium and caesium flows than (111) samples. These very assumptions about adsorption character can explain the aperiodic behavior of (T) curves at barium adsorption and caesium adsorption heat decrease in the minimal work function region.

The further investigations of Nb-O composition as TEC collector were aimed to bring an understanding on releasing from collector during TVF components influence on tungsten emitter surface work function and tungsten emitter surface mass-transfer on collector emission properties. Monocrystalline tungsten samples (112) W mounted into mobil yoke (fig. 1) and non-moving sample was (110-Nb – 0,35% O) monocrystal.

At a constant emitter temperature ~ 1700 K collector temperature increase to 1000 – 1100 K led to emitter work function growth from 4,7 to 5,2 eV, but after few hours of diode work at this collector temperature, emitter work function dropped to 4,7 eV. The further collector temperature increase to
These experiments showed that gaseous components being evaporated from Nb-O collector surface at temperature lower than 1400 K, the existence of the said components causes the emitter work function increase. With their pump of from interelectrode gap the emitter reduces its properties. The components being evaporated at higher temperatures are probably niobium oxides, with hydrogen existence reduces on emitter hot surface, leaving, niobium layer with corresponding work function 4,3 eV and 4,9 eV for (112) and (110) planes.

In TEC interelectrode gap with tungsten emitter and Nb-O monocrystal collector cyclic transport reactions can take place. The hydrogen existing in caesium vapour phase falling on collector surface containing oxygen forms water vapours and after that caesium oxyhydrates. Caesium oxyhydrate by diffusion reaches emitter, were it is being decomposed with fluid tungsten oxides formation. The letters are being condensed on collector emission properties.

To simulate this process (110) Nb - 0,3 % and (111) Nb - 0,3% O samples were intralled into mobil yoke, non-moving sample was (110) W. Niobium samples were annealed over Faraday cylinder during 40 hours at 1300 K, after that S-curves of their surface were measured in caesium flow. After that niobium samples were placed for one hour over (110) W emitter having temperature 1950 K.

During this operation the oxygen was filled into experimental device up to the pressures 10 -1 Pa, providing about 50 tun-
gsten monolayers evaporation from emitter. Niobium samples were returned to Faraday cylinder and once again S-curves were measured. The experimental results are shown in Fig. 5 in a way of (T/Tₐ) plots.

![Graph](image)

**Fig. 5.** \( (T/Tₐ) \) dependences of the collectors (111) and (1100 Nb - 0.3% O before \( \Delta \) and after \( 0 \) tungsten condensation

As one can see from the Fig. 5 samples emission properties coincide after tungsten condensation. Their minimal work function appears to be the same as initial for (110) N - 0.3% O plane and this value is 1.4 eV.

At authors suggestion tests of TEC with plane and cylindrical electrodes shape with emitter made of tungsten monocrystal and collector made of Nb - (0.2 - 0.3)% O monocrystal were held in Sukhumi Physico-Technical Institute. Emitter vacuum work function values increase was observed together with collectors work function decrease in caesium vapour. In cylindrical device after 400 hours of operation emitter made of (111) W average vacuum work function increased from 4.9 eV to 5.2-5.3 eV, and (111) Nb - 0.3% O collector in caesium vapour decreased from 1.6 to 1.5 eV. Auxiliary collector heating at \( T = 1200 \) K during two hours led to work function decrease to 1.35 eV. During further tests emission properties of electrodes remained constant.

Investigations of collector surface chemical composition held after this tests showed existence of tungsten oxides layers with thickness no more than 0.05-0.1 m.

As a result of the processes described TEC reached stable parameters, presented in table 2, which did not change during all 1240 hours of tests.

Electric output parameters of the same TECs, with the only difference being collector made of molybdenum are presented in the same table 2, these results were obtained by us earlier. As it can be seen from the comparison, using collector made of Nb - (0.2-0.3%) O gives a significal increase in characteristic TEC power.

**REFERENCES**

1. R.Pantel, M.Bujor, J.Bardoll. Continuons


ФИЗИКА ТВЕРДОГО ТЕЛА, 1975, N 12, c.3562.


ФИЗИКА ТВЕРДОГО ТЕЛА, 1961, N 5.

4. E.V.Klemenko, Mutual adsorbtion on the face (110).

УКР. ФИЗИЧ. ЖУРНАЛ, 1982, N 7, c. 1087 - 1093.

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### Table 1

**Characteristics of the researched samples niobium-oxygen**

<table>
<thead>
<tr>
<th>Orientation</th>
<th>Oxygen content in the sample</th>
<th>$U_0$</th>
<th>$U_{\text{min at Cs}}$</th>
<th>$U_{\text{min at Ba adsorption}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>of monocry-</td>
<td>mass %</td>
<td>T=1800 K, eV</td>
<td>vacuum</td>
<td>adsorption, eV</td>
</tr>
<tr>
<td>stal sample</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(110)</td>
<td>0.02</td>
<td>4.65</td>
<td>1.5</td>
<td>2.0</td>
</tr>
<tr>
<td>initial</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0.20</td>
<td>4.60</td>
<td>1.4</td>
<td>1.7</td>
<td></td>
</tr>
<tr>
<td>0.35</td>
<td>4.50</td>
<td>1.4</td>
<td>1.6</td>
<td></td>
</tr>
<tr>
<td>0.50</td>
<td>4.40</td>
<td>1.35</td>
<td>-</td>
<td></td>
</tr>
<tr>
<td>(111)</td>
<td>0.02</td>
<td>4.10</td>
<td>1.5</td>
<td>2.0</td>
</tr>
<tr>
<td>initial</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0.20</td>
<td>4.25</td>
<td>1.46</td>
<td>-</td>
<td></td>
</tr>
<tr>
<td>0.25</td>
<td>4.30</td>
<td>1.45</td>
<td>-</td>
<td></td>
</tr>
<tr>
<td>0.50</td>
<td>4.40</td>
<td>1.4</td>
<td>1.7</td>
<td></td>
</tr>
</tbody>
</table>

### Table 2

The results of experimental converter tests $T = 1870 K$, $T = 870 K$, $d = 0.4$ mm

<table>
<thead>
<tr>
<th>Electrodes</th>
<th>$W$, W/cm²</th>
<th>$V$, V</th>
<th>$P$, Pa</th>
<th>$V$</th>
<th>Geometry</th>
</tr>
</thead>
<tbody>
<tr>
<td>W-(110) Nb - 0.3% O</td>
<td>13.4</td>
<td>0.7</td>
<td>345</td>
<td>0.9</td>
<td>flat-parallel</td>
</tr>
</tbody>
</table>
| (110)W-(111)Nb-0.2%O | 11.2 | 0.5 | 440 | 0.82 | -"-
| 111 W - 111 Nb - 0.3% O | 11.6 | 0.53 | ... | 0.8 | cylindrical |
| (110)W - Mo | 7.3 | 0.45 | 720 | 0.66 | flat-parallel |
| 111 W - Mo | 6.6 | 0.46 | 280 | 0.67 | cylindrical |
SURFACE PROPERTIES OF EMITTER ENVELOPES WITH MONOCRystal TUNGSTEN COATING AND THEIR IMPACT ON THERMIONIC CONVERTER PERFORMANCE


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ABSTRACT

The paper presents the results of surface properties research into monocrystallic [111]-axis oriented molybdenum emitter envelopes with tungsten coating and the study of emissive emitter non-uniformity impact on thermionic converter (TEC) performance.

The results of the surface property analysis include the detailed analysis of element composition temperature evolution, crystallographic orientation and electron work function (Ψ) by the methods of Auger electron spectroscopy (AES), high resolution total current spectroscopy (HRTCS) and contact potential difference (CPD).

The tungsten coating has been found to be monocrystalline, as to the type of crystallographic orientation, it is polyhedral azimuthally alternating direction [110] and [112] on the surface and the corresponding vacuum work function Ψ₀ variation from 5.35 eV to 4.7 eV.

It has been revealed, that the emissive non-uniformity of the emitter envelope associated with the polyhedrality of tungsten coating can cause the increase of barrier index (V₀) by -0.2 eV.

INTRODUCTION

The research of TEC electrode surface properties impact on the output electric characteristics is a required stage of work when validating the choice of electrode materials. In developing of the reactor-converter "TOPAZ" [1,2] we carried out complex analyses of TEC laboratory models with a monocrystal emitter envelope with tungsten coating and a niobium-based alloy collector.

Tungsten was coated by the chemical vapor deposition (CVD) from WF₆ [3] on the monocrystallic molybdenum envelope with crystallographic axial orientation [111]. Upon coating tungsten the emitter envelope (EE) was not subjected to any treatment. The diameter of EE was 10 mm. The value of TEC spacing envelope (SE) at a room temperature of electrodes (d) was 0.4 mm, the effecting working area of electrodes was 10 cm².

It is evident that the choice of technological process of EE can greatly affect the emission-adsorption properties of TFE emitters, since it involves the corresponding changes in the element composition, chemical state of surface and near-surface layer, crystallographic surface orientation and work function.

This work dealt with the solution of the problem of establishing interrelation between the composition, crystallographic surface orientation, distribution of Ψ₀ over the EE surface and the corresponding TEC characteristics variation.

RESEARCH METHODS, TECHNOLOGY, EQUIPMENT

The experiments were conducted in two stages. At the initial stage the evolution of element composition, crystallographic surface structure and the distribution of work function (Ψ₀) over the EE surface were tested in the device for surface analysis. The value of the effective EE work function (Ψ₀) was calculated by the data on Ψ₀ distribution measurements.

At the second stage the variation of emission-adsorption characteristics of electrodes and J-V characteristics of TEC in the range of emitter temperature (Tₑ) variation 1200-2000 K, the collector temp-
perature (\(T_p\)) 723–1273 K, cesium reservoir temperature (\(T_{Cs}\)) 470–620 K was studied on the laboratory rig with electric heating. All in all 7 experiment runs were conducted with different laboratory TEC, the time of characteristics measurements in each run was 100–200 hours. The \(V_B\) value variation and the analysis of EE emission non-uniformity impact on the value of minimum barrier index \((V_B\) \textit{min}) obtained similar to [4] were found by the measurement data experimentally and by calculation.

The detailed research on the surface and near-surface EE region characteristics was made through a complex surface analysis by the AES, HRTCS and CPD methods in the analytical device, whose design diagram is shown in fig.1.

Fig.1 - Design diagram of an analytical device and its main units for the analysis of EE surface.

(a) - general view: 1 - analytical chamber; 2 - X,Y,Z,\(\theta\) - manipulator of specimens; 3 - AES; 4 - sublimation pump; 5 - Ti - evaporator; 6 - preliminary pumping valve; 7 - sputter-ion pump; 8 - vacuum transducer; 9 - gas bleeding-in system; 10 - HRTCS; 11 - monopolar mass-spectrometer transducer.

(b) - cross-section: 1 - AES collector; 2 - AES; 3 - gas bleeding-in system; 4 - HRTCS; 5 - Cs source; 6 - viewing port; 7 - 6-position holder - manipulator; 8 - electron gun; 9,10 - analyzing electrodes.

The detailed description of the surface analysis methods employed and the corresponding technology and devices are given in Ref. [5]. A salient feature as opposed to [5] was allocation of specimens under study (fig.1,c) - annular EE specimen-witness fragments (point 2) tested in TEC on a special holder. The holder design provided for an opportunity to rotate the specimens about the axis to detect azimuthal dependences \(f(\phi)\), a composition, structure and work function about a fixed analyzing focus on the surface. The geometrical resolution on the surface in the AES-mode was 20 \(\mu\text{m}\), in the HRTCS and CPD-mode - 0.1 mm. The measurement error \(\Phi_0\) was \(\pm 0.02\) eV. The AES sensitivity for carbon was as good as \(10^{-2}\) of monolayer. In addition, the holder design allowed the specimens to be heated by electron bombardment (point 3) up to \(T = 2200\) K and the temperature to be measured using tungsten-rhenium thermocouples TR 5/20 (point 4). The pressure of residual gases in the analytical chamber in the course of measurements was as low as \(10^{-8}\) Pa.

The profile analysis of specimen element composition was made at a room temperature on the Auger spectrometer PHI-590, the quantitative processing of the Auger-spectra was carried out by the method of element sensitivity coefficients using the data of Ref. [6].

Fig.2 shows the design diagram of the device and the laboratory TEC applied for the converters testing. The preparation of TEC and vacuum-cesium supply lines to the experiments was basically traditional including thermovacuum preparation (TPV) with residual gas mass composition monitoring in the course of gassing, distillation and Cs supply to the service reservoir (point 11 in fig.2) etc. A salient feature of these devices is the implementation of SE pumping and supply lines with relatively large clear openings and pumping rate and the stringent requirements to the accuracy of TEC measurement and maintaining. The former was achieved due to implementation of both-way pumping of TEC SE, shortening of pumping line, (point 6 in fig.2) and their connection to two magnetic discharge pumps with the total pumping speed \(~350\) 1/sec using a heated valve 50 mm in bore.
Fig. 2 - Design diagram of laboratory TEC and system vacuum - cesium supply lines.
1 - EE; 2 - collector; 3 - thermocouples; 4 - Y-shaped tungsten heater of the emitter; 5, 7 - system of heating and cooling of the collector unit; 6 - TEC pumping system; 7/8, 9, 10 - heated valves 50, 10, 10, 10 (mm) in bore; 11 - Cs-reservoir; 12 - Os distillation system; 10 - monopolar mass-spectrometer transducer; 14, 15 - protective vacuum chamber and its pumping system.

The latter was achieved due to the location of 7 tungsten-rhenium thermocouples TR 5/20 over the length of the emitter working part (point 3 and 1 in fig.2), the utilization of emitter heater providing the EE heating by irradiation up to $T_E \approx 2000$ K with the temperature drop over the length as low as 50 degrees. The temperature of electrodes and Os reservoir was kept at a fixed level using a temperature regulator with the deviation $T$ as low as for $T_E$, $T_C \approx \pm 15^\circ$, $T_{CS} \approx \pm 0.5^\circ$, the absolute measurement error $T$ did not exceed 1%. When determining the value $T_E$ the experimental profiles $T$ were averaged by a parabolic dependence. The system of J-V characteristics registration incorporated an electronic device and a scanning type data-logging system of temperature sensors, current, voltage transducers etc., with the processing and recording on the "ROBOTRON-1715" - type personal computer. The rectangular measuring pulses with a width 1 ms and on-off time ratio over 50 were made use of to reduce the thermal effect.

RESULTS AND DISCUSSION

Figs. 3-5 present the most characteristic results of the research into surface properties and EE near-surface area by the data of a number of experiments.

Fig. 3 shows a stable distribution over the EE surface of crystallographic orientation and $\varphi_0$ (fig. 3,b) established following the specimen heating at $T=2000$ K.

Fig. 3 - Variation of crystallographic orientation and EE surface work function.

- typical view of delay curves: 1 - W(110) standard; 2 - W(112) standard; 3 - EE region with (110) orientation; 4 - EE region with (112) orientation.
- azimuthal distribution of work function and crystallographic orientation over the EE surface: 1 - pure surface; 2 - with 0 impurity.
- design dependence of EE effective work function on the portion of surface with (110) orientation regions: • - by data of 6.
The data of HRTCS and CPD were referred to the crystallographic orientation of surface and EE work function with a use of primary standards with the known performance of W(110) and W(112) monocrystals' specimens with the values of work function $\varphi_{110} = 5.35 \pm 0.03$ eV and $\varphi_{112} = 4.70 \pm 0.05$ eV (fig. 3,a), which is in agreement with the data of Ref. [7]. Fig. 3,c shows the results of $\varphi_0$ calculation by the model discussed below as a function of (110) face surface fraction on the EE ($S_{110}$).

The result of calculation $\varphi_0 = 4.9$ eV (a black dot on the curve, fig. 3,c) is in agreement with the experimental data of $\varphi_0$ distribution on the EE (fig. 3,b, curve 1). When measuring the distribution $\varphi_0(\varphi)$ for various basic series of EE tested in this work the specific sizes of sections with the maximum $\varphi_0$ ((110)-face) in the azimuthal direction were measured from 0.5 to 1.2 mm. It agrees with the measurement of $S_{110}$ from 10 to $\approx 20\%$ and $\varphi_0$ from 4.9 to 5 eV.

The analysis of the results given in fig. 3 demonstrates that the EE tungsten coating is monocrystal, polyhedral by the crystallographic surface orientation, with the sections alternating every 60° azimuthally, which are oriented in the direction [112] and [110], and in the whole agree with the crystallographic orientation of Mo [111] monocrystal backing-envelope surface. The variation of $\varphi_0$ from 4.7 to 5.3 eV over the EE surface in the azimuthal direction is consistent with the corresponding change of surface crystallographic orientation from [112] to [110]. In a number of cases an oxygen impurity with a degree of coating $\delta = 0.1$ was noted, which can insignificantly shift up the dependence $\varphi_0(\varphi)$ towards higher values (curve 2 in fig. 3,b), which agrees with the data of Ref. [5].

Figs. 4 and 5 a,b show certain salient features of tungsten coating of EE applied, which are related to the availability of C and O background impurities introduced into the near-surface region of coatings in the course of CVD settling of tungsten. According to the data of profile Auger analysis (fig. 5, a,b) a content of O in the near-surface region at a depth more than 4 mm is on a level $5 \cdot 10^{-1} - 10^{-1}$ mass%.

Fig. 4 - EE Auger-spectra segments.
- "self-cleaning" when heated.
- behaviour of C in various EE regions.
- behaviour of O in various EE regions.

Fig. 5 - Data of EE profile Auger analysis (a,b) and dependence of emitter work function on T_e/T_{Cs}.
- by data of [13],
--calc by form. (5).
The distribution of O and C by the EE azimuth is non-uniform, they are accumulated largely on "ribble" surface regions corresponding to direction [121] (fig.4, b and c). Oxygen is responsible for the process, which we called EE "self-cleaning" from the impurities in particular from C, which normally goes into a "carbide" chemical state and cannot be removed by heating for molten monocrystals W and Mo being heated in vacuum [5,8]. In our case O entered in the course of W settling, when heated, diffuse on the surface, interact with C and transforms it to CO monoxide, which is readily resorbed from the surface at T ~ 1200 K. The process of EE "self-cleaning" was especially checked by model contamination of C in the form of saturated carbide by acetone vapour decomposition on a heated backing like in [15]. Heating the artificially carbidized specimen of EE up to T = 2100 K resulted in fast reduction (= 5 min) of carbon peak in the Auger spectrum (fig.4, a) and the corresponding growth of O peak like in [5] up to the initial value. Dynamics of C at diverse stages of "self-cleaning" by heating the EE artificially carbidized specimen is shown in fig.4, a. It is shown, that the surface has cleaned to the initial state, i.e. "self-cleaning" from the C impurity entered on the surface by way of its oxidation by O contained in the coating. Multiple heating of EE up to T = 2100 K following the "self-cleaning" did not result in build-up of C. The "self-cleaning" process can be assumed to be determined both by O diffusion on the surface dependent on T and C ingress rate with the source of hydrocarbons in SE TEC available. The lifetime of this O source will depend on its consumption rate (leaving the surface) and the total inventory in the W - coating layer.

The EE "self-cleaning" in the experiments on J-V characteristics of TEC considered below seems to have taken place as early as at the TFV stage. We attribute to it a stability of EE emission - adsorption characteristics observed in all measurement runs.

As an example fig.6, a and b (curve 1) show the most typical results from 7 experiment runs with TEC at the conditions studied with specified T_E, T_C, d and T_C - VAR. The value of effective vacuum work function calculated by Richardson-Deshman's formula as a function of current density in the knee of J-V characteristics (\( \psi_0 \)) of arc modes taking into account a correction for electron-cumium atom scattering and averaged over the results of measurements of all the runs was \( \psi_0^e = 4.90 \pm 0.06 \) eV.

Averaging over the same runs for the value of minimum collector work function (\( \psi_C \)) obtained with the use of back emission method and \( \psi_B \) gave the following values:
\[
\psi_C \min = 1.48 \pm 0.03 \text{ eV},
\]
\[
\psi_B \min = 2.23 \pm 0.04 \text{ eV}.
\]

The confidence limits of deviations for these values were determined with a probability 95%.

The analysis of the experimental data obtained during TEC tests by the data of integral measurements of characteristics demonstrates a good correlation of \( \psi_0^e \) with the design value (fig.3, c) by the data of local measurements \( \psi_0(\alpha) \) (fig.3, b). However, a comparison of experimental values \( \psi_B \) with the design ones provided a difference of the order of 0.2 eV (fig.6, c, curve 1 and 2). The design value \( \psi_B \)
\( V_B \) was calculated like in [4] by the formula
\[
V_B^{\text{calc}} = \gamma_C + \Delta V_C + \Delta V_{\text{transport}} \quad (1),
\]
where the term \( \Delta V_C \) takes into account a reduction of output voltage due to emission of electrons from the collector and was determined according to recommendation [9] by the formula
\[
\Delta V_C^t = \frac{kT_e}{e} \ln \left[ 1 + \frac{\dot{E}_C}{\dot{j}_C} \left( \frac{T_e - T_C}{T_e} - \frac{1}{\rho_{ts}} \right) \right] \quad (2).
\]

Here and below \( \dot{j}_C \) is the collector electron emission current density, \( \dot{j} \) the transmission current density, \( k \) the Boltzmann constant, \( e \) the electron charge, \( \rho_{ts} \) the Cs vapour pressure, dimensionality \( \rho_{ts} \) - [mm mercury column x mm], \( \rho_{cs} \) - pressure in accordance with recommendation [10] was calculated by the formula
\[
\rho_{cs}^T = -\frac{0.4053 \cdot 30}{0.0945282} T_e + 9.92534 \quad (3).
\]

The value \( \Delta V_{\text{transport}} \) corresponding to transport losses of voltage in SE was considered constant and was selected following the recommendations of Ref. [11]. It is evident that the limiting value \( V_B \) (fig.6, b, curve 3) can be found as
\[
V_B^{\text{lim}} = \lim_{\varphi_C \to 0} V_B^{\text{calc}}. \quad (4)
\]

Fig.6 shows that at \( T_C/T_{cs} \approx 2 \) \( V_B^{\text{calc}} \approx V_B^{\text{lim}} \), i.e. the TBC output voltage in this range of ratios \( T_C/T_{cs} \) is defined not by \( \varphi_C \), but it is only limited by electron emission of the collector. It is evident that the output voltage with the ratio \( T_C/T_{cs} = 1.9 \) is basically defined by \( \varphi_C \).

The comparison of values \( V_B \) and \( V_B^{\text{calc}} \) shows that their difference remains approximately constant, and in different experimental runs it was \( 0.15 \sim 0.25 \text{ eV} \).

The difference of \( V_B \) value determined in experiment from \( V_B^{\text{calc}} \) indicates a significant shortage of output electric characteristics and efficiency of TEC with polyhedral EE. The attempts to explain this barrier index value exceeding the expected one, that we know from the literature, on the basis of the \( V_B^{\text{calc}} \) design estimates were based on the interpretation of processes associated with the collector impact, in particular, with a feasible impact of electron reflection from the collector, with flecks with reference to \( \varphi_C \), an availability of virtual collector was also suggested. However, it had no conclusive explanation supported by the experiments with TEC. In our case, proceeding from the particular attention to purity and monitoring of experimental conditions, the rise in \( T_B \) measurement accuracy, the analysis of EE surface detailed characteristics, we attribute a fairly considerable difference of \( V_B \) from \( V_B^{\text{calc}} \) to the effect of polyhedral EE emission structure. With this aim in view let the design estimate \( V_B \) be quoted taking into account the experimental results (fig.3,b) and certain our semi-empirical representations given below.

\( \varphi_E \) of the emitter contained in Cs vapours is known to be normally determined either with the so-called "Razor's curves" [12] or by the formula of empirical technique [13]. We believe, that the following formula is more convenient to calculate the emitter emission current density value:
\[
J_e = \exp \left[ \frac{\varphi_e}{k} - \frac{T_e}{250} + \frac{T_e - 15.85}{a_0} \right], \quad A/cm^2 \quad (5).
\]

The comparison of \( \varphi_E \) calculated by formula (5) and by the data of Ref. [13] given in fig.5,c shows an agreement of their field of application for \( 4.3 \text{ eV} < \varphi_E < 6 \text{ eV} \) and \( 2.3 \text{ eV} < \varphi_E < 3.3 \text{ eV} \). Formula (5) is more convenient for the qualitative analysis of the phenomena occurring in TEC.

Let the polyhedral EE be considered, where the regions with high (\( \varphi_1 \)) and low (\( \varphi_2 \)) vacuum work function alternate azimuthally. The portion of EE surface with \( \varphi_1 \approx S_1 \), and with \( \varphi_2 \approx (1 - S_1) \). According to the experimental measurements shown in fig.3,b for the EE with the diameter 10 mm, the width of regions with \( \varphi_1 \) and \( \varphi_2 \approx 1 \text{ mm} \), which is essentially more than \( d \). This enables us to employ the characteristics additivity principle when constructing the J-V characteristics of TEC.

If \( \frac{J_1 - J_2}{J_1 \cdot J_2} \ll 1 \) (here \( J_1 \) and \( J_2 \) are current densities of the corresponding EE regions), then we can confine ourselves only to \( J_1 \) when considering the total emission current. Note, that this ratio is \( \ll 1/30 \) for our operating conditions.
For the segment of J-V characteristic with a hindered arc mode in the general case we can write
\[ j = a \cdot e^{-\frac{eV}{kT_e}} \]  
(6),
where \( a = \text{const} \), \( V \) is a current value of output voltage. In our case, taking into account only \( j \), with \( S \), we have
\[ j = S \cdot e^{-\frac{eV}{kT_e}} \]  
(7).

Equating (6) to (7) the value of output voltage losses is obtained (or the rise of \( V_B \)) due to EE polyhedral emission structure:
\[ \Delta V_B = V - V_1 = -\frac{k}{e} \ln S_4 T_e \]  
(8).

To calculate \( \psi_E \) of polyhedral EE let us make use of the additivity principle:
\[ j = j_1 S_4 + j_2 (1 - S_4) \]  
(9).

Substituting formula (5) into (9) we shall have
\[ \psi_E = \frac{\psi_0}{S_4} \cdot \psi_0^{S_4} + (1 - S_4) V_{\alpha} \]  
(10),
then
\[ \psi_E = \psi_E^0 + 0.2 \ln (S_4 + \varepsilon) \]  
(11),
where the correction is \( \varepsilon = (1 - S_4) e^{-\frac{\psi_0}{S_4}} = \frac{1}{32} \).

It is significant only at low values of \( S_4 \). For our EE with \( \psi_E^0 = 4.90 \pm 0.06 \) eV taking into account the variation of \( \psi_0 (\varepsilon) \) (by the data of fig.3,b) between the values from \( \psi_0 = 4.7 \) eV to \( \psi_{112} = 5.3 \) eV we shall have \( S_4 = 0.08 \pm 0.03 \), which is in good agreement with the results of local measurements \( \psi_0 \), \( S_110 = 10\% \) (fig.3,c).

In conclusion let the value \( V_B \) be estimated taking into account the additional voltage losses due to polyhedral emission structure of EE. Let the value \( \psi_C \) be equal to the averaged minimum value \( \psi_C \) of \( \psi_0 \) for the real operating conditions of TEC (\( T_E = 1800 \) K, \( T_0 = 900 \) K) will be \( \psi_C = 1.48 \) eV. The rise of \( \Delta V_B \) according to (8) for the real operating conditions of TEC (\( T_E = 1800 \) K, \( T_0 = 900 \) K) will be \( \Delta V_B = 0.3 \) eV.

Then the value \( V_B = \psi_C + \Delta V_B + 0.5 + \Delta V_B = 1.48 + 0.03 + 0.5 + 0.3 = 2.31 \) eV, which is in a good agreement with that obtained experimentally in TEC with the polyhedral EE \( V_B = 2.23 \pm 0.04 \) eV. Hence, a conclusion can be drawn that the above calculation evaluates properly both the \( V_B \) growth value and the distribution of emission regions areas on the EE.

Therefore, proceeding from the results of TEC characteristics analysis conducted, we can conclude, that the reason of relatively low efficiency of polyhedral EE TEC is an emission ability non-uniform structure of the effective area. In this context, it can be assumed that with the increase of \( S_{110} \) share up to the value \( \approx 50\% \), the rise in TEC output voltage by \( 0.15 - 0.2 \) eV can be expected.

CONCLUSIONS

The tungsten coating built-up by a gaseous-phase method on a monocrystal EE made of Mo[111] is monocrystalline. By its crystallographic orientation of emission surface it is polyhedral with the areas in directions [112] and [110] alternating azimuthally every 60°, and with the corresponding variation of \( \psi_0 \) from 4.7 to 5.3 eV.

A typical size of the region with the maximum work function in the azimuthal direction varies within the range 0.5-1.2 mm for diverse EE. The effective vacuum work function variation corresponding to it by the data of \( \psi_0 \) local measurements can be within the range \( \psi_E = 4.9 - 5 \) eV.

A property of tungsten coating "self-cleaning" from carbon has been revealed due to its oxidation by oxygen diffusing from the near-surface coating region to the surface, when the EE is heated. Concentration of oxygen entering the surface in the course of gaseous-phase tungsten settling varies in the near-surface region in the range \( 10^{-5} - 10^{-1} \) mass %.

According to the data of measurements of EE emission-adsorption characteristics as part of TEC with a niobium-based alloy collector \( \psi_E = 4.90 \pm 0.06 \) eV, \( V_B \) min = 2.23 \pm 0.04 eV.

The emissive non-uniformity of EE polyhedral tungsten coating results in the increase of \( V_B \) (reduction of TEC output voltage) by \( \approx 0.2 \) eV.
REFERENCES


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Abstract

Effective work function ($\phi_e$) is one of the important considerations in the selection of the electrode materials for high temperature thermionic energy converters in space power applications. The effective work functions of W-Re-0.35HfC with various Re contents were measured in the temperature range of 1700-2500 K. The results indicated that the effective work function increased with increasing temperature and Re content. The increase in the effective work function with Re content can be explained by the increase in the potential barrier at the metal-vacuum boundary, associated with the volume effect--decrease in the lattice constant. Conversely, adding HfC lowers the effective work function at temperatures below 2100 K. Properties associated with high temperature mechanical stability such as tensile and creep strength are also important considerations for the selection of electrode materials for thermionic energy conversion. Tensile properties of W-3.6Re-0.4HfC were evaluated in the temperature range of 1950 to 2980 K. The tensile strength of a W-3.6Re-0.4HfC alloy is much greater than that of pure W or W-3.6Re alloy above 0.5 $T_m$. The excellent high-temperature strength of W-3.6Re-0.4HfC is due to the strengthening effect of HfC particles. The strengthening effect of HfC particles decreases with increasing temperature. Above 2700 K, the contribution of HfC particles to tensile strength is very small. The decrease in the strengthening effect of HfC particles above 2700 K is considered to be caused by the coarsening of HfC particles. The fracture mode of W-3.6Re-0.4HfC above 0.5 $T_m$ is ductile dimple tearing. The tendency for ductile tearing increases with temperature. Extensive grain boundary sliding takes place above 2450 K and this is responsible for the rapid increase in tensile elongation of W-3.6Re-0.4HfC above 2450 K. Step-load and step-temperature creep tests were performed in the temperature range of 1955-2783 K and in the stress level of 10 to 70 MPa. The steady-state creep rate of W-4Re-0.33HfC alloy is a function of stress to the 5.3 power and is not sensitive to the temperature from 1955 to 2783 K. The W-4Re-0.33HfC alloy is more creep resistant than pure W, W-5Re, W-26Re, and W-23.4Re-0.27HfC alloys. The activation energy for creep deformation is about 105.5Kcal/mole at temperatures from 1955 to 2190 K. It is not sensitive to the applied stress from 10 to 70 MPa. Grain-boundary sliding initiated the wedge-shaped cracks and caused the rupture of the creep specimen. The creep deformation mechanism is most likely governed by dislocation core diffusion.
INTRODUCTION

Generation of multi-megawatts in space demands nearly ultimate alloys for high-temperature nuclear reactors and attendant energy converters. Such ultra-alloys derive from tungsten (W) incorporating rhenium (Re) to reduce W creep, recrystallization and embrittlement[1-6]. The work function of a metal surface characterizes thermionic emission[7]. It represents the difference between the electron motive far outside the free surface and the internal Fermi energy. This emission property is important in determining the efficiency of thermionic energy conversion. The useful life of the thermionic energy converter (TEC) relates strongly to the selection of emitter and collector materials and to their operating-temperature regimes. These factors are important because TEC has been chosen as one of the candidates for space nuclear power systems. W, Re alloys are promising electrode materials for high-temperature TEC.

In such space applications W provides nearly ultimate resistance to fracture, creep, recrystallization, vaporization and melting. However poor room-temperature ductility, inadequate oxidation resistance at elevated temperatures and difficulties associated with joining or welding often limit the use of this refractory metal. There was a significant effort at NASA Lewis Research Center during the Apollo era to develop W-base alloys with improved low-temperature fabricability and high-temperature strength [7-17].

For space-power systems W,Re alloys are considered to be effective candidates as electrode materials in TEC because of their high bare work functions as well as high-temperature mechanical stability. In general high bare work functions result in low cesiated work functions to produce necessary electron emission for efficient TEC. There has been an attempt to develop metal-matrix composites reinforced with W fibers for structural application in advanced aero propulsion systems.

The previous studies showed that moderate addition of Re can improve low-temperature ductility and high-temperature strength of W[8,9,17,18]. In order to further increase the high-temperature strength of W and W-Re alloys, various carbides have been added[11,12,13]. It was found that the presence of HfC particles dramatically increased the high-temperature strength of W and W-Re alloys[10,12,13].

The need of developing high-power systems for space satellites has renewed the interest in developing suitable refractory metals for these applications[19]. In the earlier studies of the effects of Hf and C concentrations on high-temperature mechanical properties of W and W-4%Re alloys, Rubenstein[11] and Klopp and Witzke[12] concluded that the optimum HfC contents for maximum strengthening at elevated temperatures were in the range from 0.35 to 0.5 mol.%. In response to such technology requirements, this study presents $\phi_e$ values for 5-20% Re in W with 0.35% HfC and determines the thermionic effects of hafnium carbide(HfC) and Re
variation in W alloys. It also presents the high temperature (> 0.4 T_m, melting temperature in Kelvin) tensile and creep properties of W-Re-HfC alloys. The strengthening effect of HfC and possible deformation mechanisms were also discussed.

EXPERIMENTAL PROCEDURES

Sample Preparations for Electron-Emission Study

The W-Re-HfC samples were prepared by powder metallurgical techniques. After sintering and arc melting the samples underwent electron-microprobe analyses to ensure homogeneity in composition. Then these discs were ground and sliced using a diamond wafering blade to a common 9.31 mm diameter and 2.72 mm thickness. Hohlraums each of 0.7 mm diameter and 7 mm depth were machined by electric discharge. Next the samples were polished to obtain smooth surfaces. Microstructures were examined and photographed for future comparisons with the results obtained from the thermionic emission microscope. Finally the samples were annealed and outgassed at 2500 K and a pressure less than 1.3x10^-5 Pa for an hour before testing.

Materials for mechanical testing

The starting materials were two 25.4 mm diameter W-Re-HfC rods provided by NASA Lewis Research Center. Both rods were processed by an arc-melting technique. One of the rods was received as arc-melted and the other was slightly swaged. The chemical compositions of both rods were determined by microprobe analysis. The as arc-melted rod was analyzed as W-3.6%Re-0.4mol.%HfC and the swaged rod was analyzed as W-4%Re-0.33mol.%HfC. Plate-type specimens were machined by electrical discharge machining. The gauge lengths of the specimens for tensile and creep tests were 8 mm and 12.7 mm, respectively. All specimens were first mechanically polished and then chemically polished with a 10% NaOH solution.

Effective Work Function Measurements

Electron work-function studies were performed in a thermionic-emission microscope. The thermionic emission microscope used for measuring the effective work function of individual grains. A counterwound W filament heated the sample by electron bombardment. Electrons emitted from the heated sample passed through a series of objective immersion electrostatic lenses and then a drift tube before striking a luminescent phosphor screen. A small aperture at the center of the phosphor screen allowed some of the electrons to reach the Faraday collector. The electron current to the Faraday collector was recorded with a Keithley 642 electrometer. This collector-current value was monitored by a computer and used to calculate the effective work function using
the Richardson-Dushman equation.

Temperature steps of 100 K were used for tests on at least five grains selected to represent different areas of the emitting surface. The sample could be moved and controlled by a three-dimensional linear traveling mechanism. Photos of the grains were taken for identification and comparison purposes.

Mechanical tests

Tensile tests were performed in an ultra-high vacuum test chamber mounted on an Instron machine. In order to provide a uniform recrystallized microstructure, all tensile specimens were initially annealed at 2450 K for 30 min. in a vacuum better than 1.3 x 10^{-5} Pa prior to testing. All creep tests were performed in a custom-made creep testing system. Creep specimens were degassed at 1273 K for 12 hours in a vacuum of 1.3 x 10^{-4} Pa followed by recrystallization annealing at 2438 K for 1.5 hours and then at the test temperature for 30 min. prior to creep testing. Specimens were heated with self-resistant heating by passing a electric current through the specimen. Temperature was measured by an optical pyrometer calibrated with a ribbon filament lamp prior to testing. The maximum uncertainty of the measured temperature was within 10 K in the whole temperature range employed in this study.

Tensile tests were conducted in a vacuum better than 1.3 x 10^{-5} Pa and at the strain rate of 10^{-3} per second. Both side and fracture surfaces of the tested specimens were examined with a JXA-840 scanning electron microscope(SEM). Step-load and step-temperature creep tests were performed at pressures below 1.3 x 10^{-4} Pa. The load was applied to the specimen through a bellows. Strain was measured from the movement of the loading rod with a linear variable differential transducer. Step-load creep tests were conducted at temperatures range from 1955 to 2783 K and at stresses range from 10 to 70 MPa. The step-temperature creep test was performed at a constant stress of 42.3 MPa and the temperature was increased sequentially from 2013 to 2190 K. The post-test specimens were examined by an optical microscope and a SEM.
EXPERIMENTAL RESULTS

Effective Work Function

Figures 1 and 2 are typical emission micrographs of the W-5Re-0.35HfC surface at 2000 K before and 2300 K after vacuum annealing.

Figure 3 presents the change of $\phi_e$ of W-5Re-0.35HfC with time at 2200 K. During a period of 180 minutes the effective work function decreased by less than 0.08 eV. Thus after stabilization at high temperatures, time had very little effect on $\phi_e$ for the W-Re-HfC alloys investigated in this program.

Figure 4 shows the influence of temperature on the effective work function of W-Re,HfC alloys: raising the temperature from 1700 K to 2500 K raised $\phi_e$ for these ultralloys by 0.96 eV for 5% Re and by 1.12 eV for 20% Re.

Fig. 1 Emission micrograph of the W-5Re-0.35HfC surface at 2000 K before vacuum annealing.

Fig. 2 Emission micrograph of the W-5Re-0.35HfC surface at 2300 K after vacuum annealing.
Fig. 3  Time dependence of the effective work function for W-5Re-0.35HfC at 2200 K.

Fig. 4  Temperature dependence of the effective work function of various W-Re-0.35HfC alloys
Figure 5 illustrates the effect of Re content on $\phi_e$ for the W-Re,HfC alloys of this study: $\phi_e$ increased by 0.40 eV at 1700 K and by 0.45 eV at 2500 K as the Re concentration rose from 5% to 20%. Therefore within the experimental ranges for these ultralloys the effective work function had a weaker dependence on Re content than on temperature.

![Graph showing the effect of Re concentration on the effective work function of W-Re-0.35 HfC at various temperatures.](image)

Fig. 5 Effect of Re concentration on the effective work function of W-Re-0.35 HfC at various temperatures.

**Tensile Properties**

Fig. 6 shows the effect of test temperature on the ultimate tensile strength of W-3.6Re-0.4HfC. The ultimate tensile strength of W-3.6Re-0.4HfC is significantly higher than that of pure W or W-3.6Re up to 2700 K. The comparison of the results with those of pure W or W-3.6Re illustrates the dramatic effect of 0.4 mol.% HfC on the tensile strength up to 2700 K. The tensile strength of W-3.6Re-0.4HfC decreases rapidly above 2700 K and becomes similar to that of pure W or W-3.6Re at 2980 K.

The effect of temperature on the 0.2% offset yield strength of W-3.6Re-0.4HfC above 0.5 Tm (1842 K) is shown in Fig. 7. The results of arc-melted pure W [10] and W-3.6Re[8] are also shown in the same figure for comparison. It can be clearly seen that the yield strength of W-3.6Re-0.4HfC is much greater than those of pure W and W-3.6Re up to 2700 K.
Fig. 6 Effect of temperature on the ultimate tensile strength of W-3.6Re-0.4HfC alloy.

Fig. 7 Effect of temperature on the yield strength of W-3.6Re-0.4HfC alloy.
In Fig. 8, the percent elongation of W-3.6Re-0.4HfC is plotted as a function of temperature. The percent elongation increases slowly with increasing temperature up to 2450 K and then rapidly increases above 2450 K. All specimens exhibited a typical ductile dimple fracture in the experimental temperature regime. Figures 9 (a) and (b) show the SEM micrographs of the fracture surfaces of the specimens deformed at 1950 K and 2450 K, respectively. Although both specimens show a similar fracture mode, it can be seen that there is a tendency of ductile tearing increases with rising temperature.

![Fig. 8 Effect of temperature on the tensile elongation of W-3.6Re-0.4HfC alloy.](image)

![Fig. 9 SEM fractographs of W-3.6Re-0.4HfC deformed at (a) 1950 K (b) 2450 K.](image)
Creep Properties

Fig. 10 shows the typical strain-time creep curves of W-4.0Re-0.33HfC obtained from the step-load creep tests at 1955, 2190 and 2873 K. Upon initial loading, these three curves all show a primary creep region with decreasing creep rate followed by a secondary creep region with a constant creep rate. In the secondary creep region, there is a balance between the strain-hardening rate and the recovery rate. An increase in the applied stress shifts this balance and the primary creep takes place again. Eventually tertiary creep occurs with necking in the gauge section followed by creep rupture (the creep test at 1955 K was terminated before rupture took place).

Fig. 11 shows a power-law relationship between the steady-state creep rate of W-4.0Re-0.33HfC and the applied stress at various temperatures. The stress exponent $n$ for creep in the power law creep rate equation was determined by measuring the slope of each straight line and was found to be in the range of 5.0 to 5.4 in the temperature range of 1955 to 2783 K.

The creep strength of W-4Re-0.33HfC alloy at a creep rate of $10^{-6}$/sec is compared with pure W [20], W-5Re [18], W-26Re [14], and W-23.4Re-0.27HfC alloys [15] in Fig. 12. This figure clearly shows the excellent creep strength of W-4.0Re-0.33HfC. For example, the comparison of the data shows that W-4.0Re-0.33HfC is about three times stronger than W-5Re at 1973 K. At 2200 K, the W-4Re-0.33HfC alloy is about 2 times stronger than W-5Re, W-26Re, or W-23.4Re-0.27Hf-C.

![Fig. 10 Strain-time creep curves of the W-4Re-0.33HfC alloy in the step-load tests at various temperatures.](image-url)
Fig. 11 Power-law relationship between steady-state creep rate and stress at various temperatures for W-4Re-0.33HfC.

Fig. 12 Comparison of the creep strength at a creep rate of $10^{-6}$/sec for various materials.
The fracture of all creep specimens was initiated by intergranular cracking. Fig. 13 shows a typical micrograph of the side surface of the creep specimen after test. Although this particular picture was taken from the creep specimen tested at 2190 K, the creep specimens tested at other temperatures showed similar features. The picture shows typical wedge-shape intergranular cracks initiated by grain boundary sliding. Therefore, it can be concluded that grain-boundary sliding caused the rupture of the creep specimen.

![200µm](image)

Fig. 13 Wedge-shape intergranular cracks observed on the 2190 K creep-tested sample.

DISCUSSIONS

Effective Work Function

Five grains as shown in Figure 1 were selected to represent different emitting surfaces. Comparing Figure 1 and Figure 2 reveals that grain sizes of the alloys were considerably larger after vacuum annealing than before. Throughout the subsequent experimental period grain sizes of the alloys changed very little. Thus it can be assumed that the grain size was fairly stable.

As shown in Figure 3 time had very little effect on the effective work function. In a previous study the effective work functions of the W,Ta alloys remained unaffected by heating the samples for 50 to 70 hours at 1900 K to 2100 K and were only slightly changed by subsequent heating for 15 hours at 2300 K[22]. Stabilities of effective work functions of the W,Ta alloys indicated at least up to 2300 K, the surface layers were not depleted of Ta. Effective work functions of W,Mo alloys were also unaffected by heating at 1900 K to 2000 K for 50 to 70 hours. In other studies a W,25Re filament was observed for more than three months at room temperature in a vacuum in the 10^{-9} torr range and for about one month at 2000 K. During these observations the effective work
function remained constant. And no change in the electron emission characteristics were detected [23].

The effective work function of W-Re-0.35HfC increased monotonically with both temperature and Re concentration within the present experimental regime. In other words, there were no minimum or inflection points. This behavior was not unexpected because no major physical change such as a phase transition occurred on the surface. Also it was consistent with Husmann that the effective work function underwent a continuous nonlinear growth with increasing amounts of Re alloyed in W [23].

Dyubua et al. discovered a reduction in the effective work function of W with HfC dissolved [24]. They explained this effect by the reduction in the potential barrier at the metal-vacuum boundary associated with a purely volume effect—an increase in the lattice constant. This influence depended on small amounts of Hf which may be available in ultralloys containing HfC even though the free Hf is an excellent getter. Gorov et al. observed a similar effect in the W,Hf alloys [25]: They attributed this effect to the change in the kinetic and potential energies of the free-electron gas due to an alteration in the conduction-electron density and the excess charges of Hf. The excess charge appears because the number of valence electrons for Hf is two less than that of W. In contrast a monotonic increase of work function can be explained by another volume effect decreasing the lattice parameter as Re content increases [26,27]. Also the extra charge of Re raises the superficial electron-potential barrier and causes an effective work function increase as Re concentration grows. However the extremely dilute amount of HfC could be responsible for the lowering of the effective work function of W,Re alloys in comparison with values from previous work [28-29]. In addition rising temperatures often push the effective work functions upward monotonically: at higher temperatures such as above 2100 K, the effective work function approaches that of the simple W-Re system. The effect of HfC diminishes due to the desorption of the HfC monolayer from the W-Re surface. For the binary alloy with an effective work function smaller than either of its components this temperature coefficient was positive [22]. The ultralloys under study followed the same trend despite the addition of HfC.

**Mechanical Properties**

The excellent high-temperature strength of W-3.6Re-0.4HfC, as shown in Fig. 6 & 7, is probably due to the solid-solution strengthening effect of Re and precipitation strengthening of HfC. However, the results of Klopp et al. indicated that the addition of 3.6 at.% Re to pure W only slightly improved the yield or ultimate tensile strength above 0.5 Tm. Therefore, it can be concluded that the superior strength of W-3.6Re-0.4HfC above 0.5 Tm is due to the addition of 0.4 mol% HfC. The finely dispersed HfC particles probably act as effective barriers to dislocation motion and improve the high-temperature strength of W-3.6Re up to 2700 K. The
rapid decrease in high-temperature strength above 2700 K is most likely caused by the coarsening of HfC particles. Klopp and Witzke[12] also reported that coarsening of HfC particles took place above 2273 K.

The comparison of Figs. 6 and 7 shows that the difference between the yield strength and the ultimate tensile strength of W-3.6Re-0.4HfC decreases with increasing temperature. The difference becomes very small above 2700 K. This is most likely caused by a decrease in work hardening rate. The work hardening behavior of a material can be expressed by the strain-hardening exponent n in the equation $\sigma = K\varepsilon^n$, where $\sigma$ and $\varepsilon$ indicate true stress and true strain, respectively. The constant K is known as the strength coefficient of a material. The strain-hardening exponent n is a function of material, microstructure, temperature and strain rate. A large value of n indicates a high work hardening capability and therefore a high resistance of a material to further plastic deformation. Fig. 14 shows the effect of temperature on the strain-hardening exponent of W-3.6Re-0.4HfC. Unfortunately, the strain-hardening exponent of arc-melted pure W or W-3.6Re is not available in the literatures. In Fig. 14, the result obtained from sintered and recrystallized W-3Re[30] is plotted for comparison. The strain-hardening exponent of W-3.6Re-0.4HfC decreases rapidly from 0.48 to 0.17 as the temperature increases from 1950 to 2450 K and then decrease slowly to 0.15 at 2980 K. The comparison of data indicates that the strain-hardening exponent of W-3.6Re-0.4HfC is much higher than that of W-3Re up to 2450 K and both alloys show a similar value of n above 2473 K. The formation of Orowan dislocation loops around finely distributed HfC particles is considered to be responsible for the high strain-hardening exponent of W-3.6Re-0.4HfC up to 2450 K.

![Figure 14: Effect of temperature on the strain-hardening exponent of W-3.6Re-0.4HfC.](image)

Fig. 14 Effect of temperature on the strain hardening exponent of W-3.6Re-0.4HfC.
As can be seen in Fig. 9 (a) and (b), the fracture mode of W-3.6Re-0.4HfC alloy above 0.5 \( T_m \) is ductile dimple tearing. The tendency for ductile tearing increases with increasing temperature. This tendency was also confirmed from the fractographs of the specimens deformed at other temperatures. The examination of the side surfaces of the deformed specimens revealed that extensive grain boundary sliding occurred in the specimens tested at 2700 and 2980 K. Therefore, the rapid increase in tensile elongation above 2450 K is considered to be caused by the grain boundary sliding. It is noteworthy that the rapid increase in tensile elongation takes place at the temperature where HfC particles begin to lose their effectiveness in strengthening this alloy. It appears that coarsening of HfC particles also decreases the effectiveness of HfC particles in hindering grain boundary sliding.

The present study clearly shows that W-Re-HfC alloys have not only exceptional high-temperature tensile strength but excellent resistance to high-temperature creep as shown in Fig. 12. The creep strength of W-4.0Re-0.33HfC appears to be better than any high-temperature alloys ever developed. The creep mechanism is often deduced from the activation energy for creep. The activation energy can be determined from the slope of a line by plotting the steady-state creep rate at a constant applied stress as a function of the reciprocal of the test temperature, as shown in Fig. 15. In the present study, the activation energy for creep of W-4.0Re-0.33HfC is calculated to be about 105 Kcal/mole in the temperature range of 1955 to 2190 K. It appears that the activation energy remains constant within the stress range of 20 to 50 MPa.

![Figure 15](image-url) Temperature dependence of the steady-state creep rate for W-4.0Re-0.33HfC.

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<th>( \sigma ) (MPa)</th>
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The activation energy for creep deformation usually corresponds to an activation energy for bulk diffusion at temperatures above \(0.5 \, T_m\). The activation energies for bulk diffusion of Re in W and self-diffusion in W were reported to be 163 and 153 Kcal/mole [31], respectively. The activation energy for creep of W-4.0Re-0.33HfC is found to be 105 Kcal/mole in the present study. Therefore, the creep deformation of this alloy is not controlled by bulk diffusion. The activation energies for grain-boundary diffusion and for dislocation core diffusion were reported to be 92 and 90 Kcal/mole, respectively [32]. Since the steady-state creep rate and the applied stress has a power-law relationship for this material, creep is not governed by grain-boundary diffusion, which indicates a linear relationship between creep rate and stress [33]. Therefore, at temperatures from 1955 to 2190 K the creep deformation of the W-4Re-0.33HfC alloy is most likely controlled by dislocation core diffusion.

CONCLUSIONS

The effective work functions of W-Re-0.35HfC with various Re contents were measured in the temperature range of 1700-2500 K. The tensile properties of an arc-melted W-3.6Re-0.4HfC alloy were examined in the temperature regime of 1950-2980 K. The creep behavior of the W-4Re-0.33HfC alloy was investigated by step-load and step-temperature creep tests. The following conclusions can be drawn from the above experimental results:

1. The effective work function increased with increasing temperature and Re content. The increase in the effective work function with Re content can be explained by the increase in the potential barrier at the metal-vacuum boundary, associated with the volume effect--decrease in the lattice constant. Conversely adding of HfC lowers the effective work function at temperatures below 2100 K.

2. The tensile strength of a W-3.6Re-0.4HfC alloy is much greater than that of pure W or W-3.6Re alloy above 0.5 Tm. The excellent high-temperature strength of W-3.6Re-0.4HfC is due to the strengthening effect of HfC particles.

3. The strengthening effect of HfC particles decreases with increasing temperature. Above 2700 K, the contribution of HfC particles to tensile strength is very small. The decrease in the strengthening effect of HfC particles above 2700 K is considered to be caused by coarsening of HfC particles.

4. The fracture mode of W-3.6Re-0.4HfC above 0.5 Tm is ductile dimple tearing. The tendency for ductile tearing increases with increasing temperature. Extensive grain boundary sliding takes place above 2450 K and results in a rapid increase in tensile elongation of W-3.6Re-0.4HfC above 2450 K.
(5) The steady-state creep rate of W-4Re-0.33HfC alloy is a function of stress to the 5.3 power, and is not sensitive to the temperature from 1955 to 2783 K.

(6) The W-4Re-0.33HfC alloy is more creep resistant than pure W, W-5Re, W-26Re, and W-23.4Re-0.27HfC alloys.

(7) The activation energy for creep deformation is about 105.5Kcal/mole at temperatures from 1955 to 2190 K, and is not sensitive to the applied stress from 10 to 70 MPa.

(8) The creep deformation mechanism is most likely governed by dislocation core diffusion.

(9) Grain-boundary sliding initiated the wedge-shaped cracks and caused the rupture of the creep specimen.

ACKNOWLEDGMENTS

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REFERENCES


THERMIonic WORK FUNCTION OF POLYCRYSTALLINE OSMIUM, SINGLE CRYSTAL (110)
IRIDIUM, AND SINGLE CRYSTAL (-1012) RUTHENIUM

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ABSTRACT

The effective work function of osmium, (110) iridium, and (-1012) ruthenium were measured as part of an ongoing effort at Arizona State University to characterize the emission behavior of promising emitter materials. For polycrystalline osmium the work function measured 4.68 eV at 1800 K and increased to 5.21 eV at 2600 K. The (110) single crystal iridium work function also increased with temperature from 5.40 eV at 2000 K to 5.55 eV at 2500 K. The increase for iridium was linear whereas for osmium it was not. No temperature dependence was observed for the (-1012) single crystal ruthenium. It had an average value of 4.90 eV over the temperature range of 1700 K to 2200 K.

The osmium emission remained transient for several hours after switching to a new temperature. The time and temperature dependencies exhibited by osmium could have been related to the pressure, but such a relationship was not confirmed. The increase in the iridium work function was definitely not a function of residual gas pressure.

INTRODUCTION

During the past several years, the thermionics laboratory at Arizona State University (ASU) has studied the work function behavior of candidate emitter materials, mostly alloys. Dilute solutions of rhenium in tungsten were examined [1-6]; recently studies of the thermionic emission from alloys of osmium in tungsten were conducted [7]. The characterization of promising alloys is continuing at ASU and presently consideration is being given to alloys of iridium in tungsten. To better understand the observed thermionic emission from such alloys it was felt that additional analysis of some less-common metals was warranted.

For example, thermionic emission from polycrystalline osmium was studied by Wilson [8] and independently by Houston [9]. The work function reported by Houston was considerably higher than that by Wilson. A literature search revealed no temperature dependent studies for ruthenium. The present interest in tungsten, iridium alloys justified an evaluation of emission.
properties of iridium. This paper presents findings for osmium, ruthenium, and iridium.

SAMPLE PREPARATION

An arc melted button of polycrystalline osmium was obtained from Dr. Edmund Storms (research scientist at Los Alamos National Laboratories). The button was fabricated by pressing and sintering high purity osmium powder (99.97 percent) prior to arc melting. A cylindrically shaped disk 9.2 mm in diameter and 2.5 mm thick was ground from the button. The top surface was polished to a mirror finish using common metallographic techniques. A 0.05 \( \mu \)m alumina slurry was used in the final lapping stage. A black body hole 0.7 mm in diameter and 7 mm deep was machined radially into the disk by electrical discharge machining (EDM). The sample was annealed at 2500 K for several days before measurements were taken.

A single crystal of iridium approximately 8 mm in diameter and 50 mm in length was purchased from Aremco Products, Inc. (23 Snowden Avenue, Ossining, New York). It was oriented with (110) planes perpendicular to its lengthwise axis. A slice 2.5 mm thick was cut from the single crystal rod using a diamond wafering blade. The (110) surface was polished the same as the osmium sample and a black body hole was machined by EDM. It was annealed at 2400 K for 24 hours prior to testing.

A polycrystalline rod of ruthenium was also purchased from Aremco Products, Inc. It was prepared in the same fashion as the iridium crystal. However during the annealing step a large grain developed at the center of the sample face surrounded by four or five small grains at the edges. A Laue pattern of the large grain showed it to be oriented with planes of the form \([-1012]\) parallel to the surface.

EXPERIMENTAL PROCEDURE

The work function was obtained from high-temperature electron emission measurements with a guard-ringed vacuum emission vehicle (VEV). The VEV [10] is depicted in Figure 1 and a simplified electrical schematic is shown in Figure 2. The collector was 3.5 mm in diameter with a 0.14 mm gap between collector and guard ring. The guard ring had a diameter several times that
of the emitter area. The effective collector area was taken as the emission area plus one half of the area of the gap.

The well-known Richardson, Dushman equation [11] was used to calculate the effective work function,

\[ J_o = AT^2 \exp\left(-\Phi_E/kT\right) \]  

(1)

where \( J_o \) = zero field saturation current density (A/cm\(^2\)),
\( A \) = constant (120.4 A/cm\(^2\)K\(^2\)),
\( T \) = temperature (K),
\( \Phi_E \) = effective work function (eV),
\( k \) = Boltzmann constant (0.861x10\(^{-4}\) eV/K).

To obtain \( J_o \) from the measured current density, \( J_s \), the Schottky equation [12] was used,

\[ \ln J_s = \ln J_o + 4.403 \left(E^{0.5}/T\right) \]  

(2)

where \( J_s \) = electron saturation current density (A/cm\(^2\))
\( E \) = applied field (V/cm)

Equation (2) is valid for low to medium strength fields. To determine the precise range of validity data are gathered as a function of applied field at a constant temperature. The linear portion of a Schottky plot \( (\ln J_s \text{ versus } E^{0.5}) \) represents the valid range. The zero field current density, \( J_o \), is obtained by extrapolating the linear section to zero field. An example Schottky plot taken from the ruthenium sample is shown in Figure 4.

Once it had been determined that the applied field was well within the linear range then Equations (1) and (2) were combined to yield

\[ \Phi_E = kT \ln\left(\frac{AT^2}{J_s}\right) + 3.79 \times 10^{-4} E^{0.5} \]  

(3)
From this equation the effective work function was computed. The immediate benefit of Equation (3) was that data were acquired as a function of time with the field held constant. The work function was then calculated as rapidly as desired facilitating time dependent studies.

The total pressure was continuously monitored with a Bayard-Alpert type ionization gauge. In addition, a mass spectrometer was used to measure residual gas partial pressures during the iridium tests. To evaluate the effect of residual gases various gases were introduced into the VEV through a high-vacuum leak valve (Figure 4).

Samples were heated by means of electron bombardment by a tungsten filament. The temperature was measured by focusing a micro-optical disappearing filament pyrometer on the black body hole. The pyrometer was calibrated routinely to an NBS certified tungsten strip lamp.

A polycrystalline sample of molybdenum was tested as a standard to check the accuracy of the VEV. The molybdenum data are plotted in Figure 5. They agreed closely with reported values [13-15].

Ultra high vacuum (UHV) was maintained by following a rigorous bake-out procedure. After each sample change the entire station was baked at 500 K for 24 hours pumping continuously with a turbomolecular pump. Ion pumps were then switched on and the sample heated to its highest expected operating temperature while still baking at 500 K. The sample was held at high temperature for 12 to 24 hours before isolating the turbomolecular pump from the VEV. The final pressure obtained while operating depended markedly on the sample and the temperature. At room temperature, pressures in the $10^{-10}$ torr range were achieved.

RESULTS

The averaged work function values are listed in Table 1. At each temperature many measurements were made. It was estimated that the results were accurate to ±0.04 eV. However, the precision was higher; measurements were reproducible to ±0.02 eV. Most of the error and uncertainty was attributed to temperature measurements.
TABLE 1. Averaged Vacuum Work Functions, \( \phi_E \) (\( \pm 0.04 \) eV).

<table>
<thead>
<tr>
<th>Temperature (K)</th>
<th>Osmium (eV)</th>
<th>Iridium(110) (eV)</th>
<th>Ruthenium(11012) (eV)</th>
<th>Molybdenum (eV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1700</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1800</td>
<td>4.68</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1900</td>
<td>4.69</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2000</td>
<td>4.71</td>
<td>5.40</td>
<td></td>
<td></td>
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<tr>
<td>2100</td>
<td>4.74</td>
<td>5.43</td>
<td></td>
<td></td>
</tr>
<tr>
<td>2200</td>
<td>4.83</td>
<td>5.48</td>
<td></td>
<td></td>
</tr>
<tr>
<td>2300</td>
<td>4.93</td>
<td>5.51</td>
<td></td>
<td></td>
</tr>
<tr>
<td>2400</td>
<td>5.07</td>
<td>5.52</td>
<td></td>
<td></td>
</tr>
<tr>
<td>2500</td>
<td>5.12</td>
<td>5.55</td>
<td></td>
<td></td>
</tr>
<tr>
<td>2600</td>
<td>5.21</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

**Osmium**

The osmium work function results from this study are plotted in Figure 6 together with the results of Wilson and of Houston. Figure 7 shows the work function data in a graph with the total pressure. An increasing pressure with increasing temperatures at very high temperatures was unavoidable. Wilson nor Houston reported residual gas pressures to use for comparison.

The thermionic emission varied with time as well as temperature. Depending on the temperature, up to ten hours, were required for the emission to reach a steady state after switching to a new test temperature. Figure 8 shows this time anomaly at three different temperatures. The pressure during each of these runs was virtually constant and well in the \( 10^{-9} \) torr range.

Osmium was heavily etched from high-temperature annealing. A photo-
A micrograph of the thermally etched micro-structure is included as Figure 9. X-ray diffraction studies were inconclusive in determining if a high temperature osmium allotrope exists.

Iridium

A comparison of the single crystal data with the total pressure is shown in Figure 10. Polycrystalline iridium has a reported work function of 5.3 eV [16]. This value was confirmed at ASU [17]; the polycrystalline data of [18] are plotted in Figure 11 together with the single crystal (110) Iridium data and the polycrystalline data of Wilson [8].

Different gases (oxygen, nitrogen, carbon dioxide, and argon) were introduced into the VEV while capturing iridium data. The partial pressure of each was increased independently from approximately $1 \times 10^{-10}$ torr to $1 \times 10^{-7}$ torr. The temperature was varied from 1900 K to 2400 K. The work function was not affected by an increase in any of the added gases. Thus it was concluded that the work function increased linearly with temperature as illustrated in Figure 12. At just above 2500 K the iridium crystal partially melted where it joined the tantalum holder. Apparently it had reacted with the tantalum to form a low melting point alloy. The osmium sample was similarly destroyed and for subsequent tests a tungsten sample holder was employed.

Ruthenium

As mentioned, a single grain encompassed most of the emitting surface of the ruthenium sample. The grain was oriented with (-1012) planes parallel to the emitting surface and it was larger than the effective collector area. Thus the data obtained represent the (-1012) single crystal planes. Data are plotted in Figure 13; In Figure 14 the single crystal data is compared with data for polycrystalline ruthenium [17]. No temperature dependence on the work function was observed, and from Figure 13 it would appear that the residual gas pressure was not a factor.
DISCUSSION

From a consideration of Figure 6 it is clear that the conflicting published results for osmium were here resolved. However it is still uncertain if the work function varied as a result of an increase in temperature or an increase in pressure. The time dependent results (Figure 8) would tend towards a temperature dependency since the work function was seen to vary while pressure was held constant. Still, delayed adsorption/desorption reactions at different temperatures could have been responsible. Figure 7 certainly suggests a pressure relationship. Unfortunately the osmium sample was destroyed (reacted with its tantalum holder) before the effect of leaking known gases into the vacuum chamber could be assessed.

The non-linear behavior observed for the work function of osmium displays the advantage in reporting the effective work function as it varies with temperature as opposed to the "temperature independent" Richardson work function,

$$\phi_R = \phi_E - \alpha$$

Where $\phi_R =$ Richardson work function,
$\alpha =$ temperature coefficient.

When $\phi_R$ is substituted into the Richardson-Dushman equation then the constant A is no longer necessarily equal to 120.4 A/cm²K². Such a substitution is valid only if the effective work function, $\phi_E$, varies linearly with temperature. Had Houston chosen to report $\phi_E$ instead of $\phi_R$ he likely would have observed a temperature dependence similar to that reported here. The time dependencies seen for osmium would also make difficult an accurate calculation of $\phi_R$ using the Schottky method.

The (110) planes of a tungsten crystal exhibit a work function much higher than the polycrystalline value of 4.52 [18-19]. Therefore, data for the (110) planes of the already high work function material, iridium, were of interest. The values obtained are probably representative of the highest
vacuum work function of any metal. The very refractory properties of iridium combined with its resistance to corrosion and high work function should make it an attractive candidate for future emitter studies.

The inert properties of iridium no doubt account for the stable work function regardless of residual gas pressure. The general agreement of iridium work function studies in the literature is certainly indicative of this pressure independent data. The iridium data and the molybdenum data reported herein lends excellent credibility to measurements made in the VEV at ASU.

The ruthenium data are useful in that they illustrate the importance of considering grain size and orientation in electron emission work. It is also interesting to contrast the three group VIIIA metals; ruthenium, osmium and iridium, especially ruthenium and osmium which have similar valence shell electrons. Surprisingly each displays unique temperature trends (Figure 15). Ruthenium (-1012) showed no temperature dependence with respect to the work function, iridium (110) increased linearly, and osmium increased non-linearly and was time dependent.

CONCLUSION

Conflicting vacuum work function data for osmium were explained. Osmium has a low work function of 4.68 eV at 1800 K which increases rapidly to 5.21 eV at 2600 K. The possibility that residual gases were responsible for the increase was not confirmed but could not be ruled out altogether. An extremely high work function for iridium (110) was measured. It increased linearly with temperature registering 5.40 eV at 2000 K to 5.55 eV at 2500 K. Residual gases were not responsible for the increase. The work function of (-1012) Ruthenium remained constant at approximately 4.90 eV from 1700 K to 2200 K.

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The Main Elements of the Vacuum Emission Vehicle (VEV) at ASU.

Simplified Electrical Diagram of the VEV.

Figure 1

Figure 2
Figure 3  Experimental Configuration for Leaking a Known Gas into the VEV.
Figure 4  A Typical Schottky Plot, Data are from a Sample of Ruthenium.

Figure 5  The Effective Work Function of Polycrystalline Molybdenum and the Total Pressure During Data Acquisition.
Figure 6  The Effective Work Function of Polycrystalline Osmium from 1800 K to 2600 K from This Work and Data from Two Earlier Reports.

Figure 7  The Effective Work Function of Polycrystalline Osmium and the Total Pressure During Data Acquisition.
The Effective Work Function of Polycrystalline Osmium as a Function of Time. Data were Acquired at 2300 K after Increasing from 2200 K, at 2200 K after Increasing from 2100 K, and at 2100 K after Decreasing from 2300 K.

Optical Photomicrograph of Thermally Etched Polycrystalline Osmium, 100X.
Figure 10  The Effective Work Function of Single Crystal (110) Iridium and the Total Pressure During Data Acquisition.

Figure 11  The Effective Work Function of Single Crystal (110) Iridium Compared to Polycrystalline Iridium.
\[ \phi_E = 4.81 + 2.99 \times 10^{-4} T \]

Figure 12 Linear Temperature Relation of (110) Iridium Work Function Data.

Figure 13 The Effective Work Function of Single Crystal (-1012) Ruthenium and the Total Pressure During Data Acquisition.
Figure 14  The Effective Work Function of Single Crystal (-1012) Ruthenium Compared to Polycrystalline Ruthenium.

Figure 15  Comparison of the Effective Work functions of Ruthenium (-1012), Iridium (110), and Polycrystalline Osmium.
REFERENCES


1.0 INTRODUCTION

Desirable improvements in the performance or capability of nuclear thermionic converters can take several forms:

a. Higher power density without penalty to output voltage or conversion efficiency, resulting in more compact reactors.

b. Efficiency of conversion to limit the size and weight of the waste heat radiator.

c. Widening of the interelectrode gap without penalty to either the conversion efficiency or electrical power density, of importance to the in-core reactor concepts where fission-induced fuel swelling can cause closure of the gap.

d. High saturation currents combined with low cesium pressure in the interelectrode gap, a critical necessity for advanced converters and induction coupling [1] and/or direct AC output.

As shown both theoretically and experimentally, one approach to achieving some of these performance improvements is simply to increase the emitter temperature. However, this enhances the rates of mass transport in both the reactor heat source and the converter, normally resulting in eventual adverse performance changes and penalizing the system lifetime capabilities.

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Another well-known and experimentally confirmed method for performance improvement, which could be incorporated with only small design consequence to the reactor concepts, is to introduce small amounts of oxygen into the converter. The benefits of oxygen addition have been demonstrated experimentally in several laboratory converters (out-of-pile electrically heated) [2].

The oxygen additive work done to date is, however, only indicative of the potential offered by this mechanism. This past work has been limited in its quantitative content, the objectives having been to demonstrate that the effect does indeed exist, is stable and reproducible, and that the addition can be accomplished in prototypic converters in a relatively simple way by oxygenating the collector. However, this approach makes the oxygen-releasing process dependent on the collector temperature, which is a critical system parameter in space thermionic reactors. The precise control of the amount of oxygen released, therefore, becomes difficult.

If the full potential and understanding of oxygenated converters is ever to be realized, an essential first step is to devise a means of obtaining and maintaining a known oxygen content in the converter vapor. The objective of the present research is to examine the possibility of using an oxygen ion carrying solid electrolytic cell to that effect.

2.0 THE SOLID ELECTROLYTE APPROACH

To control the oxygen content, or "activity", in a thermionic converter, it has been suggested to use a solid electrolytic cell capable of carrying oxygen ions [3]. The operating principle is illustrated in Fig. 1. Under thermodynamic equilibrium and assuming that, for the particular range of temperature and oxygen activity, the electrical conductivity is exclusively ionic, the cell open circuit voltage is given by Nernst's law [4]:

\[ E_0 = \frac{RT}{4F} \ln \left( \frac{a_1}{a_2} \right) = 2.16 \times 10^{-5} T \ln \left( \frac{a_1}{a_2} \right), \text{ in } V/K^{-1} \]  

(1)
Fig. 1 Operating principle of oxygen control in a thermionic converter, by means of solid electrolytic cell capable of carrying oxygen ions
where $T$ is the cell temperature, $a_1$ and $a_2$ the oxygen activities at the anode and cathode respectively. If, for instance, $a_1$ is known, $a_2$ can be immediately derived from Eq. 1. When a voltage is externally superimposed onto $E_0$, then a net ionic current, in this case carried by $O^{2-}$, flows through the cell, the ultimate result being the transport of $O_2$ molecules from one side to the other. N. Rasor has expanded on the original suggestion with the concept of a Collector Oxygen Electrolytic Dispenser (COED), in which the oxygen ion electrolytic cell is added to the collector of a thermionic converter: this concept is shown schematically in Fig. 2a and its possible integration into the multilayer collector structure of a "flashlight"-type fuel element for a thermionic nuclear reactor is depicted in Fig. 2b.

The COED structure consists of a solid oxygen-source counterelectrode layer, an oxygen-ion-conducting solid electrolyte layer, and an oxygen-permeable electrode layer that also serves as the collector of the thermionic converter. The oxygen-source layer consists of a mixture of a metal and its oxide which has a thermodynamically-fixed oxygen activity. The solid electrolyte is a metal oxide having essentially pure oxygen ion conductivity. The collector layer is a metal that is compatible with cesium vapor and in which oxygen is highly soluble and mobile.

In the thermionic converter application, Fig. 2b, the COED cell is envisioned to be integrally combined with the converter collector-insulator-cladding tri-layer structure, which is required in contemporary thermionic fuel elements to isolate the collector electrically from the liquid metal coolant. It is further anticipated that the additional solid-electrolyte and oxygen-source layers can be incorporated by the same or similar methods as those now used to form the tri-layer structure, not substantially increasing the total thickness. The possible inclusion of an outer oxygen barrier, region 10 of Fig. 2b, is also indicated, but may prove unnecessary.

A preliminary experiment, demonstrating the feasibility of using an oxygen-ion-conducting solid electrolytic cell to introduce oxygen in a cesiated diode, was recently performed at RAI as part of the TRICE program [3]. In this experiment the introduced oxygen formed cesium oxides in the diode, providing a source of oxygen-bearing species which enhanced the electron emission of a tungsten filament.
Fig. 2a Basic thermionic energy converter with COED cell

Fig. 2b Integration of COED cell into multilayer collector structure of "flashlight"-type fuel element for thermionic reactor
The COED concept is much more ambitious, due to the integration of the oxygen source and of the thermionic converter collector in the same structure. But this integration also seems like a very logical development since the operating temperatures of both the solid electrolyte and the collector are quite similar, e.g. 900-1100 K.

At the same time, the COED concept offers substantial improvements over the previous ways of dispensing oxygen at the emitter surface:

-- the rate of oxygen dispensation is not solely dependent on the collector temperature, as in the oxide collector approach.

-- the rate of oxygen dispensation is controllable by an outside voltage, hence opening the possibility of feedback control.

-- oxygen transport is theoretically reversible.

-- the oxygen activity inside the converter can be measured.

Thus, the implementation of the COED concept offers the unique opportunity of controlling and measuring the amount of oxygen released in the converter, and thereby should provide an unequalled approach to determining quantitatively the effects of that oxygen release on the overall converter performance. The practical use of the solid electrolyte cell approach is, however, contingent on two major issues: the first one is the adaptation of this technology to the requirements imposed by the nuclear thermionic converter; the second one is the ability of having a reversible electrolytic cell in the converter environment, i.e. the ability of adding oxygen to, as well as removing it from the converter inner atmosphere. It is likely that at the beginning of oxygen introduction into the converter, oxygen will be consumed until thermochemical equilibrium is reached between the various species. Removing oxygen from the converter will become necessary if there is an optimum oxygen activity or if oxygen is released inside the converter through other processes. One such process could be the diffusion of oxygen from UO₂-based fuels through the emitter cladding.
This paper examines the problems associated with the cell integration into the converter structure:

-- selection of suitable solid electrolytes;
-- verification of materials' compatibility;
-- ability to achieve a leakfree joint between metal and solid electrolyte.

3.0 INTEGRATION OF A SOLID ELECTROLYTE CELL INTO A THERMIONIC CONVERTER

3.1 Selection of Suitable Solid Electrolytes

Oxygen-carrying solid electrolytes are based on oxides of Group IV elements [5]. A quick search revealed that, of those, only calcia-stabilized zirconia (CaSZ) and yttria-doped thoria (YDT) were available off-the-shelf in the tubular shape envisioned in the design of COED converters for nuclear power applications.

3.1.1 Selection criteria

The integration of the electrolytic cell in the oxygen-permeable collector structure imposes new requirements on the electrolyte material:

a. It must be compatible with the cesium vapor and possibly also with cesium oxide vapors. At this stage in the design, it is not yet clear whether cesium oxides are actually formed on the porous collector surface or readily dissociate.

b. A leaktight joint between the electrolyte and the converter envelope must be formed.

c. For in-core thermionics, the electrolyte must be stable in the neutron flux. However, for the design of a laboratory converter, this constraint may be ignored.
d. The electrolyte should have reasonable resistance to thermal shocks, and reasonable mechanical strength.

3.1.2 Cesium compatibility test

To our knowledge, only one article in the scientific literature deals with the compatibility of YDT and yttria-stabilized zirconia (YSZ) and liquid cesium at -1000 K [6]. In this severe environment, after about 50 hours YSZ showed some penetration, while YDT showed some sign of attack only by a liquid Cs₂O₂-CsO₂ phase. YSZ "crumbled" after 7 h at 800 K in a similar liquid mixture, but the process leading to this attack was not identified.

A cesium compatibility was therefore performed. An all-nickel vessel, fired in wet hydrogen to remove carbon, was loaded with samples cut from YDT and CaSZ tubes purchased respectively from Cerac* and Alfa**. The final configuration was one sample of each material in contact with the liquid cesium and with the vapor above it. The vessel was heated for 230 h at 980 K (cesium vapor pressure around 1 atm).

A comparison of the weight of the respective samples before and after this first test revealed that ZrO₂ samples had gained about 1% in weight, while the weight decrease for the ThO₂ samples was less than 0.03%. The Cs-exposed ZrO₂ samples crumbled when squeezed with plastic tweezers, but the ThO₂ samples remained mechanically sound. The obvious conclusion is that ZrO₂ had been severely attacked. This was confirmed by ESCA (x-ray photoelectron spectroscopy) analyses performed after ion-etching about 0.1 μm to eliminate surface contaminants (ESCA probes only about 100 Å deep). ESCA spectra for Cs-exposed ZrO₂ and ThO₂ are compared in Fig. 3, clearly showing the presence of Cs in the bulk of the ZrO₂ sample, about 3% in atomic concentration, but undetectable (less than 0.5%) in ThO₂.

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* ThO₂ 15% wt Y₂O₃, Cerac, Milwaukee, WI 53201
** ZrO₂ 5% wt CaO, Alfa, Danvers, MASS 01293
Fig. 3  ESCA spectra for cesium-exposed ZrO$_2$ and ThO$_2$ samples
It is suspected that the presence of SiO$_2$ as an impurity in ZrO$_2$ is the cause of the severe attack by cesium due to the formation of silicates. SiO$_2$ is added by manufacturers to strengthen mechanically ZrO$_2$. In our case, the amount of SiO$_2$ was about 0.5% in weight.

Since admittedly the conditions of this first test were much more severe than those expected in a thermionic converter, where liquid cesium is not in direct contact with the solid electrolyte and the cesium pressure only of the order of a few Torrs, a second test was run on four ZrO$_2$ samples exposed to cesium vapor at 3 Torr. One sample was from Alfa (as before), and the three remaining from Zircoa (SiO$_2$ less than 0.25% w). The test ran for 240 hours. Weight measurements showed an increase of 0.1% on a Zircoa disk, but none on the other samples. All samples had remained mechanically strong.

A third test in which ZrO$_2$ samples of various SiO$_2$ contents were held at 900 K in 19 Torr of cesium for 420 hours, revealed however that a Zircoa sample became very brittle, although the weight increase was again only 0.1%. The results of this final test are being analyzed.

3.1.3 Comparison of thoria and zirconia electrolytes

The machining, brazing and operation at collector temperatures of the two electrolytes under consideration require reasonable mechanical properties and resistance to thermal cycling, the latter being expressed as a composite of thermal and mechanical properties.

Table I allows to compare these properties, the cesium compatibility and the resistance to neutron irradiation. A subjective scale was used with rankings 1 through 3 in order of decreasing quality.
TABLE I

Comparison of thoria (YOT) and zirconia (YSZ)

<table>
<thead>
<tr>
<th>ELECTROLYTE</th>
<th>YOT</th>
<th>YSZ</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cesium compatibility</td>
<td>1</td>
<td>2</td>
</tr>
<tr>
<td>Resistance to thermal shock</td>
<td>3</td>
<td>1</td>
</tr>
<tr>
<td>Thermal conductivity</td>
<td>2</td>
<td>3</td>
</tr>
<tr>
<td>Resistance to neutron irradiation</td>
<td>3</td>
<td>1</td>
</tr>
</tbody>
</table>

(Rankings 1-3 in order of decreasing quality)

Thus, for the experimental converters to be built to test the COED concept, YOT seems a better choice. For the eventual design of in-core converters with oxygen control, the importance of the long-term effect of cesium exposure on zirconia will have to be assessed against the neutron fertility of thoria.

3.2 Testing of Metal-Solid Electrolyte Seals

Since the electrolyte is to be integrated in the collector structure, leakfree seals between the electrolyte and the converter envelope have to be developed. YOT and CaSZ ceramic materials having thermal expansion coefficients similar to that of alumina, kovar was chosen as the metal. It also offers some resistance to oxidation, which can occur on the high oxygen activity side. Two earlier seal designs, with YOT tubes copper-brazed to kovar, tested during the TRICE program [3, 7] are shown in Fig. 4.

The seal design in Fig. 4a had the thoria in compression, leading to mechanical stress and cracking of the thoria itself. The design in Fig. 4b, a butt-seal with backup thoria ring, alleviated this problem, but operation at high oxygen activity ($10^{-2}$ Torr) at 900 K resulted in oxidation of the copper braze.
Fig. 4a  First design of a YDT cell

Fig. 4b  Second design of a YDT cell
The two main problems for this type of seal were, therefore:
-- resistance to thermal cycling,
-- resistance to oxidation.

3.2.1 Brazed kovar-electrolyte samples

CaSZ and YOT tubes, about 1.2 cm OD and 0.9 cm ID, were cut to form cylinders, 1.2 cm long, and rings, 0.2 cm thick. Kovar foil, 0.13 mm thick, was cut in disks matching the tubes OD’s. CaSZ were metalized with a W-Mn paint, which involved treatment at 1200°C in wet H₂-N₂. YOT were metalized with a Mo-Mn paint, which involved treatment at 1140°C in the same atmosphere. Copper-brazed samples were assembled by INTA for thermal cycling and oxidation resistance tests. Such samples are shown in Fig. 5.

A special corrosion-resistant plating was used involving alternate electroplating of nickel and rhodium layers. The tests of resistance to oxidation and thermal cycling are more important for YDT, as failures of such brazed joints occurred frequently during the TRICE project, and since the issue of the cesium compatibility of ZrO₂ has not been resolved yet.

3.2.2 Thermal cycling tests on Kovar-Thoria joints

The brazed samples, helium leaktight, were cycled in vacuum from 25°C to 700°C and back to room temperature with a temperature rise (or decline) of ±4°C/min, and a soak at 700°C of 30 minutes. After each cycle, the sample was leak-checked. All four Kovar-Thoria joints were found leaktight after three cycles.

3.2.3 Oxidation resistance tests on Kovar-Thoria joints

The same samples were then cycled in 1 x 10⁻³ Torr of oxygen at about 620°C, staying at that temperature for three hours. All four Kovar-Thoria joints were found leaktight, incidently completing a total of six temperature cycles. Some oxygen uptake by the thoria occurred, shown by the light yellow discoloration (Thoria heated in vacuum is bluish-white).
Fig. 5  Brazed Kovar-Zirconia (left) and Kovar-Thoria (right) joints
4.0 PRELIMINARY CONCLUSIONS AND FUTURE WORK

Since the new Kovar-Thoria brazed joints performed so well, and given the excellent resistance of thoria to cesium corrosion, thoria is obviously the best choice for the electrolyte for laboratory converters. The relationship between silica content, mechanical properties and cesium corrosion of Zirconia is being investigated in more detail to determine the suitability of Zirconia as an electrolyte material.

Future work will first concentrate on the issue of the reversibility of the electrolytic cell in a converter environment. The concern here is that the oxygen activity inside the converter might be so low, due to the oxygen affinity with cesium, as to limit the removal rate to unacceptable levels. Future work will then address the question of finding a correlation between the oxygen activity inside the converter, as determined by the electrolytic cell open-circuit voltage and the emitter work function.
REFERENCES


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MODEL OF MASS TRANSFER BETWEEN ELECTRODES
IN A THERMIonic CONVERTER
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Abstract
Chemical mass transfer of a material under nonisothermal conditions through multicomponent gas phase containing caesium vapour is considered. The system of nonlinear equations is obtained on the base of up-to-data model notions of adsorption and lateral interaction between adatoms on the surface of solids. It allows to calculate surface concentrations of the elements which the gas phase contains and to describe mass transfer processes between two surfaces of electrodes. Different kinds of dependence of impact molecules sticking probability and adatoms desorption energies from a surface on the degree of gas phase components coating are considered. The influence of oxygen in gas phase on mass transfer of electrode materials is estimated. These developed notions are applied to the analysis of molybdenum and tungsten electrodes mass transfer in a thermionic converter.

When describing chemical mass transfer between two surfaces under nonisothermal conditions, one must take into consideration chemical reactions and mass transfer through a gaseous medium. One can distinguish three steady-state conditions: 1) diffusive, when mass transfer rate is determined by diffusive process in a gaseous medium; 2) kinetic, when adsorption processes on surfaces are principal and 3) intermediate (diffusive-kinetic). The diffusive conditions are studied well enough \(1\) and are used at relatively high pressures, when free path length of gas particles is considerably less than the surface spacing. In other cases adsorption and desorption processes play an important part and a gaseous medium acts as a diffusive barrier reducing mass transfer rate in comparison with the kinetic conditions when molecules overcome the surface spacing without any collisions. The latter two conditions are used in a caesium thermionic converter. For their description a gaseous medium and solids interaction behaviour is necessary to know.

The mass balance condition during steady-state mass transfer between two surfaces having the same areas results in the following atom-molecule flows relation:
where \( r_i \) is the number of \( i \)-atoms in \( j \)-molecule; \( S_j^{(0)} \) is \( j \)-molecule sticking probability on \( r \)-surface; \( \lambda_j \) is mass transfer ratio for \( j \)-molecules which depends on the mean molecule rate, the surface spacing and diffusion coefficients of considered molecules in a gaseous medium; under the kinetic conditions \( \lambda_j = 1/2 \).

Set of equations (1) contains \( S_j \) and \( \lambda_j \) which are functions \( \Theta_i \) of surface coverage with the components of gaseous medium. The number of the unknown \( \Theta_i^{(0)} \) and \( \Theta_i^{(1)} \) is twice larger than the number of equations. The additional equations making the solution of the set of equations simple can express the amount of each element on the electrode surfaces

\[
N_i = A \left( n_i^{(1)} + n_i^{(0)} \right)
\]

or the mean partial pressures of compounds formed from the considered elements

\[
\begin{align*}
\bar{p} & = \frac{2}{i} \frac{S_j^{(0)}}{1 - \lambda_j} \frac{M_i}{R} \left( \frac{2\pi m_i k_B T}{h^2} \right)^{1/2}, \\
\bar{p} & = \frac{2}{i} \frac{S_j^{(1)}}{1 - \lambda_j} \frac{M_i}{R} \left( \frac{2\pi m_i k_B T}{h^2} \right)^{1/2},
\end{align*}
\]

where \( A \) is area of the electrodes, \( T \) is surface temperature, \( \Theta_i \) is number of the adsorption centres on the surface and \( M_j \) —mass of \( j \)-molecule.

When considering interaction of a gaseous medium with a surface, one can obtain \( S(\Theta_i) \) and \( \lambda(\Theta_i) \) dependances. Let it be a metal surface. At low pressures and high temperatures one can suppose that the interaction is described by set of balance equations each of which can be written as:

\[
\sum_j f_{ij} \lambda_j^{(1)} \cdot \lambda_j^{(0)} \cdot \Theta_i^{(1)}
\]

where \( f_{ij} \) is \( j \)-molecule flow flying on the surface.

The sticking probability \( S_j \) of \( j \)-molecules is function of adsorption \( P_n \) and desorption \( P_{n+} \) probabilities from different kinds of centres and probabilities of finding these centres \( P_n/3-6/ \). When adsorbing \( AB \) molecule, the sticking probability is equal to:

\[
S_{AB} = \frac{P_n P_{n+} (\Theta_A, \Theta_B)}{(P_n + P_{n+}) (\Theta_A, \Theta_B) + \sum_n P_n P_{n+} (\Theta_A, \Theta_B)}
\]

Adsorption and desorption probabilities must take into account the adatom migration along the surface and in general depend on the surface temperature. The equations of type(4) allow generalizing to more complex molecules and presence of a greater number of adatom kinds on the surface but result in complicated relations. When processing experimental data, it is more convenient to use proximate but simpler relations.

The right-hand sides of equation (3) contain the partial flows of atoms and molecules desorbing from the surface. If one consider desorption in terms of the theory of absolute reaction rates /7/, one can write for the desorption rate:
where \( k \) is Boltzmann's constant, \( h \) is Planck's constant, \( n_i \) is concentration of \( j \)-particles on the surface and \( j_i \) is desorption energy of \( j \)-particles.

Let the particles be a two-dimensional gas. If at the desorption the activated complex is in equilibrium with the particles forming it, different equilibriums will be reached between the atoms and the molecules on the surface. In the case of a two-component gaseous medium, for example, consisting of oxygen and hydrogen the equilibriums can be written as:

\[
XH(s) + yO(g) + 2H(g) = H_2 + O_3 \quad (8)
\]

If it is assumed that the sum of the particle states on the surface differs from the sum of the states of the corresponding particle in a gas phase by the loss of a translational degree of freedom and the energy change by the value of desorption activation energy \(/7,8/\), one can obtain a relation between the molecule flow and the flows of the atoms, forming this molecule:

\[
J_{M_j} = \frac{kT}{h} n_j e^{-\frac{J_j}{kT}},
\]

\[
(5)
\]

where \( k \) is Boltzmann's constant, \( h \) is Planck's constant, \( n_j \) is concentration of \( j \)-particles on the surface and \( J_j \) is desorption energy of \( j \)-particles.

Let us consider the atom desorption from metal surface. The atom desorption rate is determined by equation (6) too. Its dependence on the surface concentration can be explained in terms of \( n_i \) and \( j_i \). For the first quantity the dependence is rather simple \( n_i = n_x \cdot \theta_i \) but the atom desorption energy dependence requires special consideration.

The atom activation energy from a metal surface is usually obtained from thermal desorption experiments. Numerous experimental data /9/ show considerable spread in desorption heat values, for example, when desorbing oxygen from a molybdenum surface, \( j_o \sim 360-650 \) KJ/mol and \( j_o \sim 370-640 \) KJ/mol, respectively. Such spread in data is associated with the effect of many factors on the atom activation energy /9-12/; lateral interactions between adatoms, surface non-uniformity, etc.

Statistical adsorption models /11-16/ don't usually concretize the interaction nature and only take into account a pair-
wice interaction between the nearest neighbours. Among the known models only the surface electron gas model /13-14/ allows to take into consideration the mutual effect of different kinds of adatoms on the desorption energy.

According to the model assumptions the desorption energy can be written as:

\[ J_i = J_0 + \sum_j J_{0j} \theta_i, \]  

(9)

where

\[ J_{0j} = -\frac{k^2}{4\pi m^*} \eta_j \theta_j \]  

and

\[ \eta_j \]  

is an effective charge of 1-atom and \( m^* \) is an effective mass of an electron of the surface electron gas. While derivating relations (9) and (10), it was assumed that:

1) the adsorption activation energy equals zero;
2) was calculated according to the data on electron work function, polarizability and radii of particles /15/;
3) the linear interpolation of the experimental dependences of the desorption activation energies on coverage was used for estimating \( m^* \)/9,16/.

The values obtained as a result of estimating the quantities in equations (9) and (10) are given in table I for a number of materials. Thus, the surface electron gas model gives an opportunity of calculating the desorption energies, taking into account the adatom interaction and obtaining \( J_i(\theta_i) \) - dependence in a multicomponent gaseous medium.

The taken assumptions, when considering the adsorption and desorption, can be tested on the basis of the available experimental data on tungsten evaporation in oxygen and water vapor. The calculated results of tungsten interaction with oxygen, carried out by a numerical method in equations (3) and (7) and the experimental data on the tungsten evaporation rates/19-30/ are given in fig. 1.

The oxygen sticking probability was taken as:

\[ S_0 = \frac{0.56(1-\theta)}{(1-\theta_s)^2 + 0.59\theta_s (1-\theta_s) + 0.36\theta_s^2} \]  

(11)

and the equilibrium constants are taken from /31/.

The constants for calculating the desorption processes are given in table I.

<table>
<thead>
<tr>
<th>Refractory metal</th>
<th>( i )</th>
<th>( \eta_i )</th>
<th>( \theta_i )</th>
<th>( J_{0i} )</th>
<th>( J_{0i} )</th>
<th>( J_{0i} )</th>
<th>( J_{0i} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>O H</td>
<td>0.10</td>
<td>75432</td>
<td>-36313</td>
<td>20659</td>
<td>7266</td>
<td></td>
<td></td>
</tr>
<tr>
<td>W</td>
<td>1.65</td>
<td>33711</td>
<td>12771</td>
<td>-6866</td>
<td>-5209</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Mo H</td>
<td>1.65</td>
<td>35708</td>
<td>6525</td>
<td>-5587</td>
<td>-3191</td>
<td></td>
<td></td>
</tr>
<tr>
<td>W-Cs H</td>
<td>1.05</td>
<td>16950</td>
<td>11240</td>
<td>-4086</td>
<td>-6565</td>
<td></td>
<td></td>
</tr>
<tr>
<td>W-Cs H</td>
<td>1.42</td>
<td>5580</td>
<td>-24840</td>
<td>9311</td>
<td>13350</td>
<td></td>
<td></td>
</tr>
<tr>
<td>W-Cs H</td>
<td>0.82</td>
<td>74500</td>
<td>-17410</td>
<td>9621</td>
<td>11270</td>
<td></td>
<td></td>
</tr>
<tr>
<td>W-Cs H</td>
<td>0.82</td>
<td>34780</td>
<td>9621</td>
<td>-4781</td>
<td>-5695</td>
<td></td>
<td></td>
</tr>
<tr>
<td>W-Cs H</td>
<td>0.41</td>
<td>34815</td>
<td>22240</td>
<td>-11740</td>
<td>-14510</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

The experimental data on tungsten evaporation rates in the mixture of water vapor and hydrogen /33/ were used for estimating \( S_{0i} \) because of scantiness of
information on the water sticking probability on refractory metals /32/.

Let sticking probability of H₂O-molecules be written as:

\[ S = \frac{1 - \Theta_a - \Theta_b}{A + B \Theta_a + C \Theta_b} \]  \hspace{1cm} (12)

A=2.16; B=6.31 and C=0 were obtained by a least-squares technique.

\[ M_{12W}(t) \text{cm}^{-2}s^{-1} \]

Fig. 1. The tungsten evaporation rate versus the oxygen pressure at different temperature

- experiment /19-29/- calculation /30/ the present work.

Taking into account the partial filling of the surface with oxygen, H₂-sticking probability was given as:

\[ S_a = 0.051(1 - \Theta_a - \Theta_b) \]  \hspace{1cm} (13)

The experimental and calculated data on tungsten evaporation rate in H₂O+H₂-mixture are given in fig 2. The same operations were carried out for Mo-O-H-system. The calculated results of molybdenum evaporation rates in oxygen and in water vapor and the known experimental data /34/ are shown in fig 3 and 4. The calculated and experimental data are in satisfactory agreement.

If the \( M_a(\Theta_a) \) and \( S_a(\Theta_a) \) dependences of different gas j-particles are known, we can calculate the interaction of the multicomponent gaseous medium with metallic surfaces as in caesium thermionic converter.

Let us limit ourselves to considering a gaseous medium consisting of three elements Cs₂O.H. It should be noted that in Cs₂O-H-system under thermionic converter conditions according to the thermodynamic estimations the oxygen is contained mainly as caesium hydroxide CsOH. Therefore, we shall consider Cs+CsOH-mixture where refractory metals evaporate. The calculated results are given in fig 5 and 6. The data in fig 5

\[ M_{12W} \text{cm}^{-2}s^{-1} \]

Fig. 2. The tungsten evaporation rate versus the temperature in H₂O+H₂-mixture

1. \( P_{\text{H}_2} = 1.33 \cdot 10^4 \text{ Pa}, P_{\text{O}_2} = 1.33 \cdot 10^7 \text{ Pa}; \)
2. \( P_{\text{H}_2} = 1.33 \cdot 10^2 \text{ Pa}, P_{\text{O}_2} = 1.33 \text{ Pa}; \)
Fig. 3 The molybdenum evaporation rate versus the temperature and oxygen pressure

\[ \log P_{O_2} \text{(Pa)}: -0.18(1); -0.58(2); -0.88(3); -1.18(4); -1.58(5); -1.88(6). \]

Fig. 4 The molybdenum evaporation rate in \( H_2O \)-vapor versus the temperature

\[ M_{Mo}, \text{cm}^2 \text{s}^{-1} \]

is in Cs and CsOH vapors mixture. While considering particles desorbing from a surface, it is necessary to take into account ions besides atoms and molecules \( /34/ \) as follows from fig. 5. The tungsten evaporation rates in different gaseous media: oxygen, water vapors and caesium hydroxide are compared in fig. 6.

Fig. 5 The tungsten evaporation rate at different pressures of CsOH and Cs versus the temperature

Fig. 6. The tungsten evaporation rate in oxygen, water vapor and caesium hydroxide versus the temperature

When describing the mass transfer processes in the interelectrode gap, the availability of steady-state conditions of mass transfer between the electrodes is supposed, and model of refractory metal interaction with

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multicomponent gaseous media is used, that has an effect on the set of equations (1)-(2). In this case the mass flow through the interelectrode gap from each electrode surface is sum of the flows desorbing from the surface and reflected by the metal surface of the particles. The sticking probabilities and the desorbing flows are functions of coverage with the elements forming the compounds of the gaseous medium of the thermionic converter interelectrode gap.

Therefore, as a result of solving the set of equations, when calculating both the multicomponent gaseous medium interactions and the mass transfer through the intersurface gap, the values of coverage with the metal surface elements are obtained as partial results. Changing in oxygen and hydrogen coverages with the temperature, when evaporating tungsten in the water vapor and hydrogen mixture, is shown in fig.7. From the abovementioned dependences it follows that the surface oxygen concentration is much higher than the surface hydrogen one and it is the surface oxygen that is responsible for increasing the tungsten evaporating rate. This conclusion can be continued in the following way: the availability of oxygen compounds in a gaseous medium can cause an increase in the refractory metal (W and Mo) evaporation rate and in the interelectrode mass transfer, in particular, in mass transfer from an emitter to a collector.

The described model can be used for estimating the processes proceeding in a thermionic converter, when being prepared for operation and used under operating conditions.

Using the data on the partial pressures and partial rates one can obtain the initial data for equation (2b), and having solved the set of equations (1) and (2) one can estimate the effect of the thermal preparation conditions on the mass transfer of the electrode materials. The availability of the results on the electrode material mass transfer values under operating conditions allows to estimate the possible concentrations of the impurities (oxygen and hydrogen) on the electrode surfaces of a
caesium thermionic converter and to connect them with the electrode emission characteristic variation and with the generated electric power variation.

Conclusions
1. The model of the multicomponent gaseous medium interaction with the refractory metals (molybdenum and tungsten) was developed.

2. The interelectrode mass transfer model, allowing to calculate the oxygen and hydrogen amounts on the electrode surfaces in a caesium thermionic converter and the electrode material mass transfer, was developed.

REFERENCES


2. Термоэmissionное преобразование энергии: сборник статей, Пер. с англ., Атомиздат, т.1, 1964, Ричардсон А. с.144, Таэр Д., с.151.


7. Глестом С.Лейдлер К.Эририн Г. Теория абсолютных скоростей реакции. М.: Изд. ИД., 1948, с.583.


17. Тосбик Д.К. Поверхность, 1962, N9, с.15 N10, с.45.


31. Термодинамические свойства индивидуальных веществ.
PLASMA SPRAYED TUNGSTEN AS EMITTER MATERIAL

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ABSTRACT
The electron emission of etched and unetched plasma sprayed tungsten was measured in the unignited ion rich mode. Work functions were derived from the measured saturation current densities. In the ignited mode, I-V characteristics were measured. Using an unetched and an etched plasma sprayed tungsten emitter, barrier indexes of 2.35 and 2.05 eV respectively were found. At 1400°C, in the case of an unetched plasma sprayed emitter, a power density of 1.5 W/cm² was found while for an etched plasma sprayed emitter a power density of 4.5 W/cm² was measured.

1. INTRODUCTION
In order to facilitate commercial application of combustion heated thermionic energy converters, higher efficiencies and lower material costs are required. For the realization of these goals we suggested the utilization of plasma sprayed tungsten as an emitter material [1] instead of tungsten produced by Chemical Vapor Deposition. The plasma spraying process is presented schematically in Fig.1. The tungsten powder is melted in a plasma torch and sprayed on a substratum. A hot shell for a combustion heated converter is produced by plasma spraying tungsten on a brass spindle with a diameter of 5 cm. According to the supplier (H.C. Starck, Berlin) the tungsten purity is better than 99 w%, and the C content is less than 0.05 w%. The spraying process is performed in the air. Tungsten powder with a grain size of 5-20 μm is injected into the plasma flame. The plasma sprayed layer is about 0.5 mm thick. After the spraying the tungsten is reduced in hydrogen at a temperature of 1500 °C. Next, the tungsten is sintered in vacuum (10⁻⁵ mbar) at a temperature of 2000 °C.
2. THE RESEARCH DIODE

In order to measure the current-voltage (I-V) characteristics a plasma sprayed tungsten disk was produced and joined to a molybdenum emitter substructure of a research diode by vacuum brazing. Mo-42 at%Ru was used as filler material. The plasma sprayed emitter faced a molybdenum collector. The emitter and collector diameters were 1.3 cm. The various flanges are joined by copper gaskets. The emitter was heated by an electron gun (W-6%Re filament, 0.5mm in diameter). A sketch of the diode is presented in Fig.2. Collector and cesium reservoir temperature were measured and controlled automatically using Pt/Pt-10%Rh thermocouples. The emitter temperature was measured with a Pt-6%Rh/Pt-30%Rh thermocouple. In order to avoid the effect of the electrical resistance of the leads, a four point measuring method was used [2]. The I-V characteristics were obtained using a Tektronix 577 D2 curve tracer with sense connection. The diode was out gassed using an AEI triode ion pump (120 l/min). The ultimate temperature was 1600°C. The ultimate pressure was \(10^{-8}\) mbar. After testing the plasma sprayed tungsten emitter in the research diode, this emitter was electrolytically etched [3] and tested once more. The electrolytically etching was performed at a voltage of 2V in a 3% NaOH aqueous solution.
3. ELECTRON EMISSION OF UNETCHED PLASMA SPRAYED TUNGSTEN IN THE UNIGNITED ION RICH MODE

In the unignited mode various I-V characteristics were measured at a low inter electrode distance [4](see Fig.3.). The emitter and collector temperature were kept constant. Various cesium reservoir temperatures were employed resulting in ion rich conditions. Using the Richardson equation the work function ($\Phi$) can be calculated from the saturation current ($I_s$):

$$\Phi = kT_0 \ln\left(120T_o^2/I_s\right)$$

where $T_e$ is the emitter temperature in K and $k$ is the Boltzmann constant. These work functions are presented as a function of the reduced temperature, i.e. the emitter temperature divided by the cesium reservoir temperature (see Fig.4.).
Fig. 3. Electron emission in the ion rich unignited mode using a plasma sprayed tungsten emitter. The emitter and collector temperatures were 1200°C respectively 350°C. Various cesium reservoir temperatures were used: 150, 160, 170 and 180°C. The inter electrode distance was 0.02 mm.

Fig. 4. Work functions calculated from the saturation current density in the ion rich unignited mode. Measurements at various cesium reservoir temperatures were performed at emitter temperatures of 1200, 1300, 1400 and 1500°C. Literature values [5] are indicated (o) too.
4. THE UNETCHED PLASMA SPRAYED TUNGSTEN EMITTER IN THE IGNITED MODE

In the ignited mode, various I-V characteristics were obtained at fixed emitter and collector temperatures and fixed inter electrode distance [4]. Only the cesium reservoir temperature was varied. It was kept at a specified temperature, which was raised 10 degrees in every subsequent experiment (see Fig. 5.). The Boltzmann line is indicated. From the voltage difference between the Boltzmann line and the measured I-V characteristic, the barrier index is deduced. At the conditions presented in Fig. 5 the barrier index is 2.40 eV.

The power density of the converter can be calculated from the voltage current characteristics. In Fig. 6. the power density measured at three temperatures is presented. A power density of 1.5 W/cm\(^2\) was found at 1400 °C. This is a power density one would expect for a poly crystalline tungsten emitter.

Fig. 5. I-V characteristics in the ignited mode using a plasma sprayed tungsten emitter and a molybdenum collector. The emitter temperature and collector temperature were 1400 °C and 635 °C respectively. The cesium reservoir temperatures ranged from 250 to 300 °C (step 10 °C). The inter electrode distance was 0.3 mm. The dashed line is the Boltzmann line (V+2.30V).
Fig. 6. Power density of the converter with plasma sprayed tungsten emitter and molybdenum collector at emitter temperatures of 1300, 1400 and 1500°C.

5. ELECTRON EMISSION OF THE ETCHED PLASMA SPRAYED TUNGSTEN EMITTER IN THE UNIGNITED ION RICH MODE.

In order to expose the (110) tungsten crystal planes at the surface, the plasma sprayed tungsten emitter was electrolytically etched. The work function of the etched plasma sprayed tungsten emitter was calculated from the measured I-V characteristics in the ion rich unignited mode (see Fig.7.). It was found [4] that the work function of the tungsten emitter changed less than 0.1 eV upon etching (see Figs.8. and 5.).
Fig. 7. Electron emission of etched plasma sprayed tungsten in the ion rich unignited mode. The emitter and collector temperature were 1300°C respectively and 370°C. Various cesium reservoir temperatures were used: 180, 190, 200, 210 and 220°C. The inter electrode distance was 0.05 mm.

Fig. 8. Work functions calculated from the saturation current density of the etched plasma sprayed tungsten emitter in the ion rich unignited mode. Measurements at various cesium reservoir temperatures were performed at emitter temperatures of 1200, 1300 and 1400°C.
5. THE ETCHED PLASMA SPRAYED TUNGSTEN EMITTER IN THE IGNITED MODE

I-V characteristics were measured in the ignited mode at various condition [4] (see Fig.9). Power densities are reproduced in Fig.10. It is seen that the power density is much higher using an etched plasma sprayed emitter (at $T_e=1400^\circ C$, $P=4.5W/cm^2$ as compared to $1.5 W/cm^2$ for unetched plasma sprayed tungsten).

Fig.9. I-V characteristics in the ignited mode using an etched plasma sprayed tungsten emitter and a molybdenum collector. The emitter temperature and collector temperature were $1400^\circ C$ respectively $630^\circ C$. The cesium reservoir temperature ranged from 260 to $320^\circ C$ (step $10^\circ C$). The inter electrode distance was $0.4mm$. The dashed line is the Boltzmann line ($v+2.00V$).
Fig. 10. The power density of the converter with the etched plasma sprayed tungsten emitter and molybdenum collector. The emitter and collector temperatures were 1400°C and 630°C respectively. The cesium reservoir temperature was optimal. Various inter electrode distances were used.

6. DISCUSSION AND CONCLUSION

Comparing Fig. 5 and 9 the effect of the etching of the emitter can be deduced. It is seen that at the same cesium reservoir temperature, the etched emitter results in a higher current density and in a lower barrier index. Which explains the much higher power density of the diode with the etched emitter. The higher current of the etched emitter is expected, because a lower cesiated emitter work function was measured too. The measured higher current density in the ignited mode corresponds with the 0.1eV lower emitter work function.
However, the bare work function of a (110) plane: $\Phi_{(110)} = 5.3$ eV and that of a poly crystalline surface plane $\Phi_{pol} = 4.5$ eV differ much more, i.e $\Delta\Phi = 0.8$ eV. It is generally accepted that the difference of the the cesiated work function equals the difference of the bare work functions [5]. It is concluded that the (110) tungsten planes developed during etching disappear during operation at high temperature (emitter temperature up to 1500$^\circ$C). However, using a etched tungsten emitter, a stable and surprisingly low barrier index is found.

For the etched and unetched emitter, the characteristics were calculated [4] using the model of N.Rasor [6]. The experimentally determined emitter work functions were used in the calculation. The value of the collector work function was kept the same in both cases. It resulted from the calculation that the current density is higher using the lower emitter work function (in the etched case, 0.1 eV lower). But the barrier index is the same in both cases. We conclude from this that the work function of the collector has changed in the experiments using the etched emitter. As a result of the evaporation of material from the etched emitter the composition of the collector surface changes.

The molybdenum collector surface was thoroughly investigated with X-ray diffraction, Scanning Electron Microscopy and AUGER. Using various acceleration voltages in the Scanning Electron Microscope the composition and the layer thickness of the outer layer can be determined. Using X-ray diffraction, carbides were detected on the collector surface. The combined analyses indicated [4] that a tungsten carbide layer ($W_2C$, about 0.02 $\mu$m thick) was condensed on to the molybdenum collector.

The above measurements indicate that plasma sprayed tungsten, after reduction and sintering, is a suitable emitter material. Etching the plasma sprayed tungsten emitter resulted in a high power density of 4.5 W/cm$^2$ at an emitter temperature of 1400$^\circ$C.

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REFERENCES


TRANSPARENT THERMIONIC CONVERTERS
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Summary

A novel thermionic converter concept is described in which the collector is transparent in that it absorbs a minimum amount of thermal radiation from the emitter, transmitting the greater part. Such a configuration can be accomplished by use of sapphire as the structure of the collector with a coating of metal tens of Angstroms thick serving as the electrode surface.

One application of this concept is a high efficiency thermionic converter. The transparent collector would be incorporated into a converter of otherwise conventional design, such as a cesium diode of planar or cylindrical configuration with emitter heated by any means (solar, combustion or nuclear fission heat). Outside of the transparent collector would be an ideally adiabatic reflecting surface. This reflecting surface would operate at the same temperature as the emitter, and so the total thermal radiation from the emitter would be zero. Since thermal radiation is parasitic to the thermionic power conversion process, elimination or reduction of radiation enhances converter efficiency directly.

High Efficiency Thermionic Converter

Efficiency of a thermionic converter is defined by the equation

$$\eta = \frac{V J}{q_e + q_r + q_c + Q_s/A}$$

where the terms in the numerator are the converter electric power output and those in the denominator the converter thermal power input. $V$ is the converter output voltage and $J$ is the interelectrode current density. The thermal power components are

$q_e$ energy flux associated with the interelectrode current, called also emitter electron cooling

$q_r$ thermal radiation flux from emitter
$q_c$ thermal conduction flux through cesium vapor from emitter

$Q_s/A$ the net heat from the power source that bypasses the interelectrode space, mainly by way of the emitter lead and other structure, per unit electrode area.

Thermionic converter designers make an effort to minimize $Q_s$, and years of development effort in thermionics have focused on raising the $VJ$ product. The concept of a transparent converter aims to minimize $q_r$.

A converter design is proposed as illustrated in Figure 1. The collector is composed of a structure of sapphire coated with tens of Angstroms of a metal, which serves as the electrode. Outboard of the collector would be a vacuum with multi-foil type insulation. The coated sapphire collector would either reflect or transmit the greater part of thermal radiation originating from the emitter, and the surface of the multi-foil insulation would reflect thermal radiation back through the transparent collector to the emitter. As a result, the surface of the multi-foil insulation would operate at near the emitter temperature with a minimal net radiative heat transfer off of the emitter, thus minimizing $q_r$.

The collector would run at a temperature determined by the electron cooling and cesium conduction which impinge upon it, the Joule heating generated within and the thermal conductance of periodic penetrations of the sapphire which connect it to the collector base. Representative temperatures are shown in Figure 1.

Sapphire is the suggested material for the transparent collector. The spectral distribution of the emitter radiation, whatever the emitter material might be, can be expected to follow the Planck distribution for black body radiation. As can be seen from Figure 2, for a temperature of 1800 K the distribution peaks around 1.6 $\mu$m falling between 0.7 and 5.0 $\mu$m. Compare this distribution to the spectral transmittance of sapphire in Figure 3, where it is a maximum over the same range. Note also that the spectral reflectance of sapphire is minimal in this wavelength band.

The metallic collector coating must be compatible with long-term thermionic performance and highly electrically conductive. Its thickness is
optimized balancing transparency with lateral electrical conductance. (Thermal conduction is accomplished through the sapphire and the metal.) For the purpose of initial feasibility studies, copper, nickel or platinum are considered candidate materials for the coating. Transmittance through the coating is modeled according to electromagnetic theory (Ref. 2).

\[ T = e^{-d/d_0(\lambda, T)} \]

where

\[ d_0(\lambda, T) = (\lambda c/\sigma(T))^{1/2} \]

and \( \lambda \) is the wavelength of radiation and \( \sigma(T) \) is the temperature dependent electrical conductivity. A 20\( ^\circ \)A layer of copper has a calculated transmittance of 97%.

Because of the thin collector electrode, the transparent thermionic converter must operate at low current density compared to "conventional" high pressure cesium diode type thermionic converters. High conversion efficiency can be obtained, however, at low current density. A standard reference for thermionic converters is Thermo Electron Planar Converter #46 (Ref. 3), which was tested over a range of emitter temperatures and with interelectrode spacing between 0.03 and 1.0 mm providing electrode output curves for optimum cesium pressure. Emitter was (110) oriented tungsten and collector was niobium. Figure 5 shows the hypothetical optimum efficiency of converter #46 in ignited mode operation at representative constant electrode temperatures with assumed varying values of \( q_r \) ranging from the estimated value for the electrode pair to one tenth of that value. Clearly the maximum efficiency rises, and it occurs at lower current densities.

Even higher efficiencies can be obtained operating a transparent converter in the unignited mode or in the quasi-vacuum mode. Very close electrode spacings are required, however.

Figures 6 and 7 presents typical results of calculated performance of transparent converters. Calculations use properties of the sapphire collector structure with copper collector electrode coating. Radiative heat exchange through the collector is modeled as one-dimensional. The variable \( L \) is the
relative pitch between current leads on the collector. The configuration modeled is shown in Figure 8.

Figure 9 is a summary comparison of calculated maximum (limiting case) converter efficiencies for transparent converters in two modes of operation with performance based upon actual performance of three experimental devices. Shown in the figure are the theoretical maximum lead efficiencies (in whatever operating mode) for varying values of barrier index, $V_b$. Plotted at points A and B are the calculated efficiencies for the best results of experimental devices Thermo Electron #46 and SAVTEC-15 (Ref. 4) as they were built. Each, respectively, operated the ignited and unignited mode. Shown at points A' and B' are the values of their efficiency if they were transparent converters and operated in the same mode. Point A" is the resulting best efficiency of Converter #46 in the unignited mode.

Dramatic increases in converter efficiency appear to be possible with the transparent collector concept, and in particular when combined with the SAVTEC converter configuration, a conversion efficiency near 30% might be expected.

Acknowledgement

Contributions to the development and evaluation of the concept described in this paper have been made by Ned Rasor of Rasor Associates, Inc. and Dan Allen of General Atomics.

References


Figure 1 - Transparent Collector

Figure 2 - The Planck distribution law, spectral radiance of blackbody radiation as a function of temperature and wavelength.
Figure 3 - Analyzed normal spectral transmittance of aluminum oxide (Sapphire). (Ref. 1)

Figure 4 - Angular spectral reflectance of aluminum oxide (Sapphire). (Ref. 1)
Figure 5 - Transparent C46 Converter (ignited mode)

Figure 6 - Comparison of Converter 46 as normal converter and transparent converter.
Figure 7 - Comparison of SAVTEC-15 as normal converter and transparent converter.

Figure 8 - Collector
Figure 9 - Summary of Efficiency Improvements
TECHNIQUES FOR THE STUDY OF DIFFUSION IN ADVANCED TEC ELECTRODES

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ABSTRACT

The next generation of thermionic fuel element (TFE) converters being developed in the United States may incorporate high creep strength emitters to minimize fuel swelling problems. The anticipated design for such an emitter incorporates a substrate of a high creep strength material like W-1% HfC, which is formed into the emitter shape and coated with CVD W.

A potential problem with this emitter design is the stability of the surface properties of the CVD W. These properties are extremely sensitive to the presence of adsorbed or surface segregated species, even in quantities less than one atomic layer. Thus, the possibility of Hf, C or impurities from the substrate diffusing to the emitter surface and altering its emission properties is a serious concern.

We discuss means for monitoring the diffusion of species through CVD W layers, for determining diffusion rates and for measuring the effects of these species upon the emitter surface properties. Techniques to be used include thermionic projection microscopy to measure the emission uniformity of the emitter surface and to monitor its change with time, and surface sensitive analytical techniques including Auger electron spectroscopy and secondary ion
mass spectrometry to study diffusion parameters.

We also plan to study Cs adsorption onto CVD W surfaces with adsorbed diffusion products, in order to measure the effects of these species on cesiated emitter surface properties. This is part of a larger effort to investigate coadsorption of Cs and fission reaction products onto electrode surfaces.

INTRODUCTION

The next generation of thermionic fuel element (TFE) converters being developed in the United States will probably incorporate high creep strength emitters to minimize fuel swelling problems. One possible design for such an emitter incorporates a substrate of a high creep strength material like W-1% HfC, which is formed into the emitter shape and coated with CVD W. This design provides the high-temperature mechanical strength necessary for long converter lifetime. The CVD W layer provides the proper emitter work function during operation in Cs vapor, and the surface of this layer must have stable properties over the lifetime of the device.

A potential problem with this emitter design is the stability of the surface properties of the CVD W. These properties are extremely sensitive to the presence of adsorbed impurities, even in quantities less than one atomic layer. Thus, the possibility of Hf, C or impurities from the substrate diffusing to the emitter surface and altering its emission properties is a serious concern. In particular, any such contamination which modifies the intrinsic work function of the CVD W or its ability to adsorb Cs can have a detrimental effect upon the power output of the converter.

Another potential problem with TFE converters operated for long times is diffusion of fission products into the interelectrode space of the TEC diode.
Whether such diffusion will occur in advanced, composite emitter systems has not yet been determined, but it certainly seems possible when 5-10 year operating lifetimes are considered. If this diffusion does occur, these fission products may have significant effects upon TEC operating characteristics.

We are initiating a two-pronged program of study of these potential diffusion effects. First, we will perform measurements to determine whether significant diffusion can be expected during the anticipated TEC device lifetime, using accelerated testing methods and appropriate modeling. Second, and in parallel with these measurements, we will study the effects of potential diffusing species upon the surface properties of TFE electrodes. In this paper, we discuss the experimental techniques to be used in these studies.

EXPERIMENTAL TECHNIQUES

Diffusion from the bulk to the surface of a solid specimen commonly produces a phenomenon known as surface segregation, that is, an enrichment of the diffusing species in the surface layer. In the case of TEC electrodes, any surface segregation of diffusing species may be detrimental to device operation. It is therefore clear that the most direct method of tracking diffusion-induced effects is to use surface-sensitive analytical techniques. In particular, changes in the surface chemical composition and electronic properties need to be studied.

1) Work Function

The work function may be measured most directly using some form of retarding potential technique (1). We will use the field emission retarding potential method (FERP) which yields the absolute work function of the surface...
directly (2). FERP provides the most accurate way to measure the true work function. This method works best on flat single-crystal specimens at least 2 mm in diameter. It may also be used on practical, polycrystalline surfaces, where it measures an arithmetic average of the work functions of the various planes exposed at the surface. FERP is most suitable for the study of surfaces at room temperature, and is difficult to implement for heated samples. The FERP technique may also be used to determine the low-energy electron reflection characteristics of a surface. In the study of diffusion in TEC electrodes, FERP will be used primarily to determine changes in work function of controlled surfaces during adsorption of impurities, both with and without the presence of adsorbed Cs (simulation of the effects of diffusants at surfaces).

A second method for the measurement of work functions and changes in work functions is the thermionic projection microscope (TPM). This instrument allows the distribution of electron emission intensities to be measured from the pattern on a fluorescent screen. Several talks given at this conference discuss results of measurement made using various embodiments of the TPM (3-4). Our particular experimental arrangement is discussed in detail elsewhere (5), but is described briefly here for completeness.

Figure 1 shows schematically the implementation of our TEC. The electrons emitted from the heated specimen are accelerated and projected onto a phosphor screen which is coated with a thin layer of aluminum to attenuate the photons emitted from the hot specimen. The pattern on the screen is recorded with a vidicon tube and frame grabber which provides a digitized position versus intensity map to a personal computer. The image is coded with a color scale for visual display, and can also be plotted in pseudo-three-
Figure 1. Schematic drawing of the TPH experimental tube.
dimensional and gray-scale forms (Figs. 2 and 3). The digitized data is processed with background subtraction and median averaging routines and is corrected for distortion induced by projection of a hemisphere onto a flat screen. Then, the emitted current density \( J \) from any region of the specimen may be calculated. The temperature \( T \) of the specimen is measured pyrometrically, so the equation

\[
J = AT^2 \exp(\phi_e/kT)
\]  

(1)

may be used to calculate the effective work function \( \phi_e \) assuming the pre-exponential factor \( A \) is 120 A·cm\(^{-2}\)·K\(^{-2}\). Using the TPM we will be able to determine the emission uniformity of the CVD emitter surface, monitor its change with time and do accelerated testing by increasing the specimen temperature.

2) Surface Composition

Although it is essential to know what effect diffused surface impurities have upon the work function of the emitter surface, it is equally important to determine the nature of these impurities. With the surface sensitive analytical techniques of fixed-beam and scanning Auger electron spectroscopy (AES) and secondary ion mass spectrometry (SIMS) we will be able to determine precisely which species have diffused to the emitting surface and where the diffusion has occurred (for example, along grain boundaries). AES is particularly useful for quantitative measurement of species which segregate to the surface in quantities of at least a few percent of the surface layer. Surface segregation is common behavior for many bulk impurities in metals, and is driven by surface energy considerations.

SIMS is an especially sensitive technique, and it provides an inherent depth profiling capability. Thus, we will be able to probe the distribution of diffusing species within the CVD W layer of a high creep strength emitter.
Figure 2. Pseudo-three-dimensional image of the thermionic emission pattern from the hemispherical end of a $<110>$-oriented W rod.
Figure 3. Gray-scale image of the thermionic emission pattern from the hemispherical end of a <110>-oriented W rod. The electron emission profile shown at the bottom indicates relative emission intensities along the line bisecting the pattern.
Our particular SIMS uses liquid metal ion source technology, developed in our laboratory, to provide a sub-micron lateral resolution capability. Thus, SIMS analysis of diffusion at grain boundaries within the CVD W layer is practical with this instrument. The information provided by these surface compositional analysis techniques will allow us to determine the diffusion constants of the diffusing species in the CVD W layer.

3) Cs Adsorption

A final set of experiments will involve Cs adsorption onto CVD W surfaces with adsorbed diffusion products. Of particular importance will be the determination of work function versus coverage of Cs on the contaminated CVD W surface, which will be determined by the FERP method already discussed. Furthermore we will use thermal desorption mass spectrometry (TDS) to determine the binding energies of Cs on these surfaces. We are presently using an elemental Cs source and the FERP, AES and TDS techniques to study coadsorption of various species and Cs onto collector surfaces, and the investigation of Cs on emitter surfaces with adsorbed diffusants is a logical and compatible extension of that work.

CONCLUSIONS

The research discussed here addresses a potentially serious problem with composite TFE emitters intended for long-term, high-temperature applications. The experimental techniques applicable to this research are all well-established, and do not represent any dramatic departure from the tradition of surface analysis. Nevertheless, these studies are expected to provide important insight into the viability of new types of emitters for use in advanced TFE designs.
REFERENCES


MEASUREMENT OF THE EMISSION CHARACTERISTICS OF THE TEC ELECTRODES BY MEANS OF A THERMIOTIC EMISSION MICROSCOPE.

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The operation experience of many years shown that a thermonic emission microscope (TEM) is a reliable tool for investigating the emission properties of electrode materials for thermonic energy converters (TECs) and offers some incontestable advantages as compared with other methods, such as diode measurements [1]. The main hopes of recent years for the further improvement of TECs have been set on the development of collectors having low work functions and high-efficiency emitters ensuring a required current density in cesium vapor. It seems to be very promising, therefore, to apply a TEM to studying the surface emission properties of the TEC emitter and collector materials in cesium vapor under conditions adequate to those of the TEC normal operation.

Cesium can be fed to the surface under study in the TEM only by a directed beam because adsorption of cesium from the gas phase on the specimen surface is impossible under conditions required for the normal TEM operation [2]. In consequence, it has been chosen a scheme of cesium injection through a capillary channel in the body of the first electrode of the TEM immersion lens [3].

The experimental device represents a modified TEM version [4] the block diagram of which is given in Fig. 1. The exceptional feature of the TEM in comparison with the averaging methods of analyzing the surfaces of the TEC materials consists in the possibility of measuring the emission current from a small local area (10 μm) of the analyzed surface, which makes it possible to take the spectra of distribution of electron work function over the surface. The possibility of observation on a
fluorescent screen the studied surface in emitted electrons in combination with determining the spectra of distribution work function on surface, enables the local and integrated emission properties to be compared under conditions of one experiment. A working vacuum of 10^-3 torr in the TEM column is attained with an oil-free pumping means. Moreover, there is a possibility of checking during the measurements the spectrum of residual gases and the dynamics of its change by a radio-frequency mass-spectrometer. A silver and a palladium thermodiffusion leakers built into the TEM vacuum system to introduce oxygen and hydrogen, respectively, as well as a special system for introducing other gases by a HV gas dosing valve make it possible to vary the composition and the partial pressures of gaseous components in any direction required in the experiment.

A cesium molecular gun built into the first electrode of the TEM immersion lens produces an orientated molecular flux of cesium atoms directed into the surface under study. A narrow directed beam of cesium atoms is formed by a 10-mm-long channel 0.5 mm in diameter. The slope of the channel axis is 14°. The design of the cesium injector assembly is shown in fig.2. The cesium source represent a heated ampoule containing a cesium-graphite compound. The temperatures of the cesium source and the cesium channel are registered by chromel-alumel thermocouples. The temperature of the specimen is measured by a tungsten-rhenium thermocouple and by infrared pyrometer in the low-temperature (below 1000 K) range and by a precision optical pyrometer at high temperatures (1000-2800 K). The specimens, the TEM optics, the cesium injector and the system of thermocouples are at high potentials. This is the main difficulty in developing the cesium system for the TEM operation conditions. This problem was solved at the stage of development of the cesium molecular gun and its introduction into the TEM. To avoid condensation of cesium on the inner surface of the channel, the temperature of the channel walls is maintained sufficiently high by a special heater. To prevent the spreading of cesium in the TEM pumping and spectrometric channels the immersion lens and cesium injector assemblies and so on.
The flow of cesium atoms through the outlet channel of the injector is molecular. From the general ideas of the kinetic theory of gases one can easily derive for ideal gases the formula for the channel conductivity.

\[
\frac{1}{F} = \left( \frac{2.394}{d^3} + \frac{3.184}{d^2} \right) \left( \frac{M}{73.144 \cdot 10^6} \right)^{1/2}
\]

where \( F \), \( d \) and \( l \) are the conductivity, diameter and length of the channel, respectively; \( M \) is the molecular weight; \( T \) is the temperature of the channel wall. Thus, the channel conductivity is determined practically by the design features of the injector and equal to 1.287 cm\(^5\)/s for the design described here.

The reactions of high-temperature transformations in the cesium-graphite compound is a physical basis for the operation of the cesium injector of such a type. These reactions can proceed by the following way with formation of cesium [4]:

\[
\begin{align*}
5 \text{ C}_2\text{Cs} & \rightleftharpoons 4 \text{ C}_{10}\text{Cs} + \text{Cs} \\
12 \text{ C}_{10}\text{Cs} & \rightleftharpoons 5 \text{ C}_{24}\text{Cs} + 7 \text{Cs} \\
3 \text{ C}_{24}\text{Cs} & \rightleftharpoons 2 \text{ C}_{36}\text{Cs} + \text{Cs}
\end{align*}
\]

The conditions for saturation of graphite with cesium were chosen taking into account formation of the \( \text{C}_2\text{Cs} \) phase.

Knowing the injector channel conductivity \( F \), it is possible to determine the density of cesium atom flux, \( N_t \), through the channel in the form \( N_t = F \cdot n \), where \( n \) is the density of cesium atoms in the channel obtained from the relationship \( n = 9.656 \cdot 10^{18} \text{P/T} \cdot \text{cm}^2 \). Here \( P \) is the pressure of saturated cesium vapor in torr at a reservoir temperature \( T \). The value of \( P \) for \( \text{C}_2\text{Cs} \) was determined from sorption isosteres [4]. With the slope of the channel axis relative to the studied surface \( \alpha = 14^\circ \), taken into account, the expression for the density of cesium atom flux reaching the specimen surface can be written in the form \( N = \)
4.62.10 P/T at. Cs/(cm².s). The dependences N(T) shown in fig.3 were obtained from the family of curves P(T). However, it should be noted that real values of N can be somewhat lower than the calculated ones because of the possible scattering of the beam. A mathematical model for the scattering of the beam of cesium atoms at the injector outlet was developed to refine the physics of the interaction of the beam and the specimen surface. The results of the mathematical modelling are presented in fig.4 where R and ψ are the planar coordinates in the plane of the specimen. Taking into account that the linear dimensions of the specimen surface under study are 6x8 mm² it is possible to draw the following conclusion: the whole area of the specimen surface is exposed essentially to the uniform flux Cs atoms, i.e. there is no anisotropy of irradiation. From this point of view it is possible to ignore the surface diffusion of Cs atoms and to consider only the process of steady-state equilibrium between the incident beam and the desorption flux from the surface when determining the coverage of the surface with the cesium layer at a fixed temperature of the substrate.

The local work function is determined by the total current method from the Richardson equation. The density of current from the specimen is calculated as:

\[ j(x, y) = J(X, Y)/S_0/M^2, \]

where \( J(X, Y) \) is the current to the Faraday cylinder measured by an electrometric amplifier; \( S_0 \) is the area of the calibrated orifice in the plane of the screen; \( M \) is the magnification of the immersion lens. The relative error in calculating the work function from the total current does not exceed 1%.

After being placed into the working chamber of the experimental device, the specimen investigated were subjected to the standard thermovacuum pretreatment to remove the traces of the carboniferous components from their surfaces. The local vacuum work function measured after this pretreatment in some points of the surface, for example, on the surface of
a W(110) single crystal varied in the range of 5.15 to 5.35 eV. The corresponding temperature dependences of the statistical mean values of work function for W(110), W_{el} (110) Mo(110) and W_{el} (110) single crystals measured by the total current method are shown in Fig. 5 [5].

In order to obtain an emission pattern in the low temperature range (650-1000 K) which is essentially closer to the temperature regime of the TEC collectors the measurements were made under conditions when the beam of cesium atoms affects the surface of a Mo single crystal. The typical emission pattern of the Mo(110) surface on the TEM screen in the high-temperature (T = 1900 K, a) and low-temperature (T = 900 K, b, c) ranges obtained in the action of the cesium beam on the surface is given in Fig. 6. One can see the lines on the surface drawn specially in order to determine the magnification of the TEM.

The dependence of the logarithm of the emission current density on reciprocal temperature obtained for four fixed temperatures of the cesium - graphite compound are shown in Fig. 7. The densities of cesium atom fluxes on the investigated surface in the 5-shaped characteristics varied from 10 to 10^{17} at. Cs/(cm^2 s). The values of the local work functions were calculated from the obtained values of the current density and the corresponding temperatures of the specimen T. It is convenient to present the temperature dependences of work function in the form of the Rassor curves \( e^0 = f(T/T_R) \) [6], where \( T_R \) is the temperature of a fictitious reservoir with liquid cesium which ensures the same flux of cesium atoms as the cesium-graphite compound at the temperatures noted above. Figure 8 shows the emission dependences \( e^0 = f \left( T/T_R \right) \) for the specimens of the electrodes from W(110), Mo(110), LaB_6 and polycrystalline molybdenum. As can be seen from the figure, the obtained curves characterize the emission properties both in the collector and emitter ranges of temperatures. Thus, the use of a TEM with a cesium molecular gun makes it possible to obtain the basic characteristics of the TEC electrodes in all temperature ranges used in a TEC.

The dynamics of the emission properties of various collector at different stages of the TEC operation was studied in detail by means of a TEM [7]. It was shown that the
emission-adsorption properties of the collectors varied essentially at the initial stages of the TEC operation and then were stabilized in time. The TEM allows also the modelling of some processes proceeding on the electrodes in the operating TEC and the comparison of the obtained properties of the electrodes with those for a real TEC.

REFERENCES


3. V. A. Koryukin, V. P. Obresumov, P. D. Chilingarishvili.
A cesium injector for a thermionic emission microscope.

Sources of working medium for thermionic energy converters.
Energoatomizdat, 1986.


7. V. A. Koryukin, V. P. Obresumov, V. I. Vybyvats.
Figure Captions

Fig. 1. The block diagram of the thermionic emission microscope:
1 - vacuum chamber; 2- cathode assembly; 3- specimens;
4- specimen transport system; 5- electrostatic system of immersion lens;
6- ampoule with cesium-graphite compound; 7- cooled panel;
8- luminescent screen; 9- guard electrode; 10- Faraday cylinder;
11- thermal current measuring unit; 12- total and partial pressure measuring system; 13- gas inlet system; 14-
pumping system; 15- unit of high-voltage sources.

Fig. 2. The immersion lens assembly with a built-in cesium injector:
1,3,4 - electrodes of immersion lens; 2- channel of cesium injector; 5- cesium source.

Fig. 3. The temperature dependence of the densities of cesium molecular flux for the given design of the cesium source.

Fig. 4. The calculated results on the uniformity of the incident flux of cesium atoms on the specimen surface.

Fig. 5. The temperature dependences of work function for W(110), W(110), W(112) and Nb(110) single crystal (curves 1,2,3 and 4, respectively).

Fig. 6. The emission patterns of the surface of a Mo(110) single crystal on the TEM screen.

Fig. 7. The dependences of the emission current density on the Mo(110) reciprocal temperature for different temperatures of the cesium source.

Fig. 8. The temperature dependences of work function in the Rasor coordinates:
1- polycrystalline Mo; 2- Mo(110); 3- W(110); 4- LaB₆.
Fig. 3

Fig. 4

Thermionic Energy Conversion
Fig. 5

Fig. 6
Thermionic Energy Conversion
Plasma in the Knudsen TEC mode is essentially nonequilibrium. The distribution function of charged particles differs strongly from the Maxwellian one. Under these conditions an instability sets in and high amplitude oscillations develop. To describe the nonlinear nonstationary processes the special methods are required. Calculations of the nonstationary processes are also necessary in the case when voltage is applied to the collector or auxiliary electrode, i.e. at the initial stage of the Knudsen discharge development.

Before studying the nonstationary processes let us consider some particularities of the Knudsen TEC mode. The Knudsen number under the TEC conditions is defined as follows

\[ \text{Kn} = \frac{\lambda}{d} \]  

Here \( \lambda \) - mean free path, \( d \) - value of interelectrode gap. In the Knudsen mode (\( \text{Kn} > 1 \)) electrons pass across the gap without collisions. In this case power losses due to current passing turn out to be minimum and the TEC efficiency is most near to the maximum attainable thermodynamic value of the efficiency given by Carnot. The compensation of space charge may be realized for instance by means of the essential decrease of the gap (to \( \approx 1 \mu \text{m} \)). TEC with the full compensation of space charge is the ideal TEC. But such decreasing of gap is difficult to realize in practice. Therefore ions are used to neutralize the electron charge. As a rule the cesium is chosen as an ionizable addition. For providing conditions most near to those of ideal TEC it is advisable to produce ions by means of the surface ionization. The discharge occurring in the arc mode leads to the inevitable voltage losses and the TEC efficiency decreases.

The important parameter of the Knudsen mode TEC with the surface ionization is the degree of neutralization

\[ \gamma = \frac{n^+_i(0)}{n^+_e(0)} \]  

\[ (2) \]
Here $n^+_i(0)$ and $n^+_e(0)$ - densities of ions and electrons with positive velocities measured directly at the emitter surface. This parameter depends on the properties of the emitter surface. Next important parameter is the ratio $d/\lambda_D$ (here $\lambda_D$ - the Debye length). As a rule $d/\lambda_D \gg 1$, i.e. plasma is present in the gap. Under Knudsen conditions it is impossible to rise essentially the cesium vapor pressure. In this case as experiments have shown work function in the presence of cesium vapors is lowered insignificantly and the use of cesium vapors doesn't permit to obtain the optimum emitter.

The idea of using additional component with high absorption energy in the Knudsen mode proved to be fruitful. Barium vapor appears the most suitable for this purpose. Due to relatively high ionization potential Ba is actually not ionized on surface but it effectively reduces the emitter work function. One can vary emitter work function in a wide range without changing other TEC parameters by means of controlling the Ba pressure. It is especially important for the physical investigations. The TEC with Cs-Ba filling was first studied by Psarouthakis [1]. However in this paper overstatement results have been presented, which are probably due to mistakes in the temperature measurements. Further systematic investigations of Cs-Ba TEC were carried out in [2], where the attention was paid mainly to the dependence of saturation current on the Ba vapor pressure (Figure 1a). At fixed values of emitter temperature and Cs vapor pressure this dependence has a rather sharp maximum. Existence of the maximum is associated with the competition of the two processes which take place during the lowering of emitter work function: increasing of the electron emission and decreasing of the ion emission. At the maximum point the parameter $\gamma$ proves to be near unity and to the right from this point the regime turns out to be undercompensated. While in the overcompensated regime the saturation current coincides with the emission current of emitter, in the undercompensated regime it is restricted by a noncompensated electron space charge. First calculations of potential distributions in the undercompensated Knudsen regime were carried out using assumption of full filling of potential well by ions [3]. It follows from these calculations that in undercompensated regime the saturation current doesn't depend on $\gamma$, i.e. the dependence $j$ on the $P_{Ba}$ should be like a curve in Figure 2 denoted by dashed line. Essential difference of experimental results
from the calculated ones is due to a low degree of filling of potential well by ions.

Results of solving of self-consistent system of kinetic equations and the Poisson equation in assumption that ion capture is absent point to the existence of a virtual cathode near emitter (Figure 3). Height of virtual cathode increases with decreasing $\gamma$ [4]. Because of the strong growth of potential barrier retarding the electrons decrease of $\gamma$ results in reducing of the current (Figure 2b). Such calculations well correlate with experiment.

An actual emitter surface is always inhomogeneous in terms of work function. The emitter is covered with patches having the different work functions. Since the potential wells in which the capture of ions may take place are located near surfaces the patches with a high work function may be the effective ion absorbers. Even if these patches occupy a small part of surface the ion absorption by the patches may be significant since ions oscillate and approach the surface many times.

Dependence $J(P_{Ba})$ has been investigated for the different materials of substrata [5]. Figure 3 shows these dependences for polycrystalline W, monocrystalline $W_{110}$ and Ta at the same cesium pressure and emitter temperature. In all these cases the curves are qualitatively similar. The only effect of the emitter material is to change the value of the $P_{Ba}$ at which the maximum current is reached. Among these materials the polycrystalline tungsten has the maximum adsorption energy for the barium atoms. In this case the maximum current is attained when the barium pressure is the lowest. This result is in a good agreement with the experimental data [6,7]. For example, in accordance with [6] the specific energy of barium adsorption for monocrystalline $W_{110}$ is 2.3eV/atom, for $W_{111}$ - 4.6eV/atom and for $W_{100}$ - 5.2eV/atom. Thus, the (110)-face with the maximum work function has the lowest barium adsorption energy as compared with another faces. It follows from the Figure 3 that the use of barium practically solves the problem of the emitter material selection. Optimization of the emitter work function and obtaining of the maximum current value are ensured with any substratum. The value of this current is universal under the fixed other conditions. It was shown that this value is

$$J_0 = 1.57 \cdot J_{eq}$$  \hspace{1cm} (3)
Here $J_{eq}$-equilibrium current density in a closed isothermal cavity at the fixed temperature of the walls ($T=T_E$) and Cs pressure.

Table 1 shows the values of the $J_{eq}$, $J_o$ and $\Phi_E^O$ for a fixed cesium flux to emitter surface $P_{Cs}=10^{18}$ cm$^{-2}$ sec$^{-1}$ at different emitter temperatures. The magnitude $\Phi_E^O$ is the optimum value of emitter work function. The decrease $\Phi_E^O$ with growing $P_o$ results in the maximum saturation current increase according to the law

$$J_o \sim (P_{Cs})^{1/2}, \quad J_o \sim (P_{Cs})^{1/2}$$

Figure 4 shows the theoretical and experimental dependences $J_o(P_{Cs})$ at $d=0.8$ mm and various $T_E$. The good agreement between the theory and the experimental data is shown in a wide range of the cesium pressure from $5 \times 10^{-3}$ to $2 \times 10^{-2}$ Torr. It may be seen that at very low pressures experimental points are above the theoretical curves. It is due to the thermomolecular pressure effect occurring when cesium is introduced through a nonisothermal tube. This effect is observed at very low pressure. The decrease of the experimental value at increasing the cesium pressure above $2 \times 10^{-2}$ Torr is due to scattering of electrons by cesium atoms. Here we see again the competition of two processes: the increase of $J_o$ with growing $P_{Cs}$, and scattering of electrons. Condition Kn $\rightarrow 1$ for cesium corresponds to

$$P_{Cs} d = 2 \times 10^{-2} \text{Torr} \cdot \text{mm} \quad (5)$$

If the gap decreases, a value of current density may be increased by the law

$$J_o \sim d^{-1/2} \quad (6)$$

The maximum power of TEC under Knudsen conditions, $W_o$, was calculated for various $T_E$ and $T_C$ [5]. Figure 5 shows...
$W_o$ as a function of $T_E$ for various $\varphi_C$ ($\varphi_C$ is connected unambiguously with $T_c$). The experimental data are also presented here. They well agree with theoretical curves if we suggest that $e\varphi_C^o$ is within a range $2.2-2.3\text{eV}$. It should be noted that the experimental data were obtained at very high collector temperatures (1300-1500K). The possibility of obtaining a high specific power at a high collector temperature is based on the fact of Knudsen TEC efficiency being close to the Carnot one.

Now attention should be paid to the typical features of TEC having a developed or multicavity surface emitter. The geometric surface development implies the use of surface slots having rectangular cross-section with walls perpendicular to the emitter surface. The preliminary experiments involving such surfaces showed that the surface development results in the noticeable increase of TEC power. However, many effects observed were not explained. In [8] the transverse magnetic field was used to analyze the anisotropy of the velocity distribution function of electrons emitted by a multicavity emitter. In our experiment anisotropy was observed; electron velocities in a plane parallel to the cavity walls proved to be higher. A model was suggested which explained the formation of the electron distribution function within the cavity as a result of electron scattering on non-one-dimensional potential structures in the vicinity of patches on the wall surface.

For obtaining most reliable data on differences between parameters of TEC having the smooth and multicavity emitters a special experiment was carried out (see [9]) in which operating conditions were the same for both emitters. The main error in results of Cs-Ba TEC study appears in measurements of the emitter temperature. It is due to changing transparence of the sapphire windows acted upon by barium. There exist additional errors of measurements of the multicavity emitter temperature associated with the technology of its fabrication. Therefore in the experiment considered the emitter was made in the form of a tablet and one half of an operating surface was a multicavity emitter while another half was a smooth emitter. Against each of them two isolated collectors were placed (Figure 6a).

Figure 6b shows the dependencies of current and power gain coefficients on $P_{Ba}$. It is seen that the main gain was obtained in the overcompensated mode; it amounted to 5. As a consequence, the $W(P_{Ba})$ dependence for the multicavity emitter has a maximum broader than that
for case of the smooth emitter. The comparison of maximal power values on the \( W(P_{Ba}) \) dependences indicates that the gain attained in the case of the multibavity emitter application is as high as 40%.

We have always observed current oscillations of large amplitude in the Knudsen TEO with a smooth emitter as well as with a multicavity emitter. Oscillations exist mainly in a region of current saturation (Figure 7). Their frequency is about 1 MHz at \( d \approx 1 \text{mm} \).

When one investigates theoretically the nonlinear time-dependent processes in a bounded collisionless plasma at each time \( t \) electrons and the electric field may be assumed to adjust themselves "instantaneously" to the ion distribution at that time. The kinetic equation for ions moving in the time-varying electric field and a stationary kinetic equation for electrons together with the Poisson equation for the known ion distribution are solved in turn.

The computation of the ion velocity distribution function is reduced to calculating of some particle trajectories. The main feature of the computational method is as follows: each trajectory at a point of \((x,t)\)-plane is calculated by integrating the equation of motion for a particle backward in time starting at this point and ending at the time the trajectory intersects the emitter. This method guarantees the high accuracy of the distribution function calculations [10].

The calculation of the electron and potential distributions for a fixed ion distribution reduces to solving a second-order ordinary differential equation with boundary conditions \( \Phi(0)=0, \Phi(d)=\Phi_C \). This nonlinear boundary-value problem has as a rule a nonunique solution. The number of possible solutions, as well as their stability, may be analyzed by means of the \( \eta,\varepsilon \)-diagram [11]. To construct the diagram the electric field strength near the emitter is varied and the \( E(0) \)-dependence of the potential value at the collector \( \Phi(d) \) calculated each time is plotted. The diagram intersections with \( \Phi(d)=\Phi_C \) line give all the boundary-value problem solutions. The plasma state for which the diagram slope at the point of intersection is negative is stable relative to small electron perturbations. In the opposite case it is unstable.

The calculations have shown that the nonstationary process in plasma involves the alternation of two stages: the slow (ion) and fast (electron) ones [8]. The fast electron transitions develop from the Pierce-type instability. They play the prevalent role in the nonlinear structure formation during the oscillation processes. Figure 8 gives
An example of a time-dependent process in the Knudsen TEC with a relatively small gap value of $d/\lambda_D = 8$ which in the end transits into a stable stationary state. It has been supposed that the ion flux from emitter is turned on instantly at a time $t = 0$. The ion transit time across $\lambda_D$ is chosen as a unit of time. Under conditions considered nonstationary process is aperiodical and involves only one jump. Note that the stage into which the transition occurred was prepared by the plasma beforehand (see the dash line in Figure 8b).

Figure 9 shows the process of nonlinear oscillations in a diode with a larger gap: $d/\lambda_D = 16$ [12]. One observes here several electron jumps, a moving virtual cathode which limits the electron current passing across a diode and strong electron fields accelerating ions towards the electrodes.

The ion intersection with the moving nonlinear potential structure leads to the formation of the strongly accelerated ion beams in energy space (Figure 10). As a consequence, the ion kinetic energy is localized in space (Figure 11a). Note that the ion density curve in the gap has no such sharp humps (Figure 11b).

The nonlinear part of the fast stage (an evolution of the Pierce-type instability) has been studied in Ref. [13]. The electron transit time across $\lambda_D$ is chosen as a unit of time (in cesium plasma it is about $1/500$ of the ion transit time). It has been shown that the process develops aperiodically and ends in a new stage which differs strongly from the initial one (Figure 12).

The next problem is investigating of transient processes existing at the initial stage of the Knudsen TEC discharge. Such processes occur under conditions when the voltage $\Phi_i$ exceeding the ionization potential $\Phi_i$ is applied between the electrodes. If the surface ionization is negligible the initial electron current corresponds to the well-known Langmuir's 3/2 power law: $j_{3/2} = (1/9\pi)(2e/m_e)^{1/2}\Phi_i^{3/2}/d^2$. Current rise at the initial discharge stage is due to the compensation of the electron space charge by ions which are generated in an interelectrode spacing as a result of direct impact ionization of atoms by electrons. Appearance of the ions leads to expansion of the ionization region and to sagging of the potential in this region. In its turn it causes the further increase in ion generation (Figure 13). In order to describe quantitatively this process it is necessary to solve a system of kinetic equations for charged particles distribution functions and the Poisson equation for...
the potential of self-consistent electrical field in the spacing taking into account ion generation. Effective numerical - analytical methods of solving such nonlinear system developed make it possible to calculate all diode states available and to define a critical values of current (\( J_{cr} \)) and voltage (\( \Phi_{cr} \)) corresponding to the discharge ignition.

For the calculation of transient processes at the initial stage of discharge (14) as well as for investigation of the surface ionization TEC oscillations an adiabatic approach was used. Poisson equation involved in this approach is

\[
\frac{d^2\eta}{dt^2} = \frac{4}{9} I \cdot \eta^{3/2} / \eta^{1/2} - N_1(\xi, \tau), \quad \frac{dn}{d\xi} \bigg|_{\xi=0} = 0 \quad (7)
\]

Here \( \eta \) and \( \xi \) - dimensionless potential and distance from the cathode, \( I=J_e / J_{3/2} \) - relative electron current, and \( N_1(\xi, \tau) \) - ion charge density at a point \( \xi \) at a time \( \tau \) (the unit of the potential is \( \Phi_1 \), distance -\( d \), time - \( t_o = d/(2\phi_1/m_1)^{1/2} \), charge density - \( n_{1o} = \phi_1/(4\pi\epsilon_0 d^2) \). The current and potential distributions are found from this equation if the ion density is known. The main problem is to find \( N_1 \). For numerical solving of the kinetic equation for ions phase space \( \xi, v \) is separated in a sufficiently large number of small cells \( \Delta\xi, \Delta v \) and the distribution function is represented by densities \( F_{pj} \) of the ions and their mean velocities \( V_{pj} \) in the cells. In order to calculate the evolution of the ions from different cells during a time step the particle number conservation law is used. Such conservative method ensures the high accuracy and stability of the numerical scheme.

An important parameter which defines the character of the ionization process development is

\[
b = (m_1/m_e)^{1/2} \sigma_0 n_a d
\]

Here \( n_a \) - atom density, \( \sigma_0 \) - typical ionization cross section. At the fixed value of the spacing \( d \) parameter \( b \) is proportional to atom pressure \( P_a \).

The results of calculation for the fixed \( b \) and different anode voltages near the ionization threshold are represented in Figure 14. It is seen that at \( \Phi_a < \Phi_{cr} \) during the time \( t \approx t_o \) a steady state with
the monotonic potential distribution is established which corresponds to the preignition I-V curve point at a given anode voltage (Figure 14a, curves 1-2). On the contrary, if $\Phi_a > \Phi_{cr}$ the steady state does not exist and the continuous growth of the current (Figure 14a, curves 3-6) and expanding of the ionization region (Figure 14b, solid curves) are observed, time necessary to attain zero field strength at the anode being inversely proportional to the value of overvoltage $\Phi_a - \Phi_{cr}$. It is seen also that the retardation of the current growth at $t > t_o$ is connected with the beginning of ion neutralization at the cathode.

Dependence of an ionization cross section on electron energy in the wide range of energies is nonmonotonic. For Cs it was measured in [15] and is represented in Figure 16b. As it is seen from calculations [16] such behavior of the cross section results in the presence of the critical pressure $P^o_a$ below which the Knudsen discharge does not develop (Figure 15, curves 1-4). At $P_a > P^o_a$ there are two branches of I-V curves (Figure 15, curves 6-10) in the region of monotonic potential distributions and, respectively, two values of the discharge ignition potential (Figure 16a). For the case of Cs filling and $d=1\text{mm}$ the value of $P^o_a$ turns out to be $3 \cdot 10^{-3}\text{Torr}$ and corresponding value of the ignition potential $\Phi_{cr}^o$ is equal to $40\text{V} (\approx 10\Phi_i)$. As $P_a$ increases a voltage range in which the developed discharge with nonmonotonic potential distribution exists is expanded.

Both phenomena - the Pierce-type instability in the Knudsen TEC mode and the existence of high-voltage low-current branch on the preignition I-V curve are of great importance: they open new ways for controlling the current in systems with Knudsen bounded plasma.
REFERENCES

Figure 1. Saturation current vs. $P_{Ba}$ and $\gamma$.

a: Experimental results, $d = 0.8\text{mm}$, $T_E = 2100\text{K}$, $T_C = 1100\text{K}$;
b: Calculated results.

Figure 2. Profiles of a virtual cathode at various $\gamma$.

1 - $\gamma = 0.001$, 2 - 0.002, 3 - 0.004
4 - 0.005, 5 - 0.01, 6 - 0.02, 7 - 0.04, 8 - 0.05, 9 - 0.1, 10 - 0.2. $\lambda_D$ - Debye length, calculated for plasma parameters at a virtual cathode top, $\Phi$ - potential, measured from the emitter potential.

Figure 3. Saturation current density vs. barium vapor pressure for various emitter materials;

d=0.1mm, $P_{Cs} = 10^{-3}\text{Torr}$,
- $T_E = 2100\text{K}$, o - 2000K.
Figure 4. Maximum saturation current density $j_0$ versus $P_{CS}$ for various $T_E$.

Figure 5. Maximum specific power versus $T_E$ for two Cs flux values. Solid curves calculated for $d=0.1$ mm, $F_a=10^{19}$ cm$^{-2}$s$^{-1}$, dashed - for $d=1$ mm, $F_a=10^{18}$ cm$^{-2}$s$^{-1}$.

Figure 6. a: Schema of electrode arrangement in the experiment with smooth and developed emitter surfaces. b: Current density (1, 2), specific power (3, 4), current gain (5), power gain (6); °-smooth, and x - multicavity emitter. $d=0.5$ mm, $P_{CS}=2 \times 10^{-2}$ Torr, $T_E=1928$ K, $T_C=1263$ K.
Figure 7. a - I-V curve; 
b - temporal oscillogram of the current at the short-circuit point for \( d = 1 \text{mm} \), \( P_{\text{Cs}} = 5 \times 10^{-4} \text{Torr} \), and \( T = 2060 \text{K} \).

Figure 8. Evolution of the \( \eta, \varepsilon \)-diagram (a), of the electric field strength at the emitter (b), and of the potential distribution in the gap (c) for a diode with \( \gamma = 2 \), \( \eta_{\text{C}} = 5 \), \( \delta = d / \lambda_{\text{D}} = 8 \); \( \zeta = x / \lambda_{\text{D}} \), \( \tau = t / (\lambda_{\text{D}} / (kT_{\text{E}} / m_{\text{f}})^{1/2}) \), 
\( \lambda_{\text{D}}^2 = kT_{\text{E}} / (4\pi e^2 N_e(0)) \). 
In (b), dashed and dotted lines indicate stable and unstable states, respectively.

Figure 9. The convection current (a), the electric field strength near the emitter (b), and the potential distribution in the gap during an oscillation period (c) vs time for a Knudsen diode with \( \gamma = 2 \), \( \eta_{\text{C}} = 5 \), \( \delta = 16 \). Electron jumps are indicated by little circles. In (c), the dashed curves give the potential distribution after the respective electron jumps.
Figure 11. Change in the distributions of the ion kinetic energy (a) and the ion density (b) along the gap during an oscillation period for a diode with \( \gamma = 2, \eta_C = 5, d/\lambda_D = 16 \).

Figure 12. \( \eta, \varepsilon \)-diagram at the time of tangency (a), potential distribution at different time moments (b), and time dependence of the potential perturbation maximum \( \Delta \eta_m \) (c); \( \gamma = 2, \eta_C = 1.73, d/\lambda_D = 11.523, \tau = t/(\lambda_D/(kT_F/m_e)^{1/2}) \). In (b), solid curves define initial (0) and final (\( \infty \)) states, and dashed curves are the potential distributions at \( \tau = 50 \) and \( \tau = 60 \).
Figure 13. Schematic pictures of interelectrode gap (a) and potential distributions (b). 1) Initial distribution without ionization (coincides with vacuum one); 2) Distribution in the presence of ions. Ion generation region is shaded.

Figure 14. a: Time dependence of the relative current for $\beta=6.3$ and various values of $\eta_a$: 1) $\eta_a=1.5$; 2) 1.67; 3) 1.70; 4) 1.72; 5) 1.8; 6) 2.0; $\eta_{cr}=1.69$. b: Distributions of the potential (solid curves) and of the ion density (dashed curves) corresponding to points I-IV of the curve 4 in Figure 2a. (for Cs at $d=1\text{mm}$ $n_{\text{fc}}=2\times10^{-8}\text{cm}^{-3}$ and $t_{\text{e}}=0.4\ \mu\text{s}$).

Figure 15. $I$ vs $\eta_a$ for Cs: 1) $\beta=1$; 2) 1.4; 3) 1.54; 4) 1.55; 5) 1.56; 6) 1.6; 7) 1.7; 8) 1.8; 9) 2.5; 10) 10.6. Dashed curve: zero differential resistance. Dot-dashed curve: zero differential conductivity. Dotted curve: points in which zero field is attained at the anode.

Figure 16 a: $\eta_{cr}$ (potential of the points with zero differential resistance) vs $\beta$. b: Ionization cross section $\sigma$ for Cs vs energy $W$. $\sigma_{\max}=10^{-15}\text{cm}^2$. 

Thermionic Energy Conversion
DEVELOPMENT OF A DIAMOND INSULATOR FOR
THERMIONIC APPLICATIONS

by

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Development of a Diamond Insulator for Thermionic Applications

by

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Abstract

Diamond (D) and Diamond Like Carbon (DLC) films and thick sections are being considered for several applications, one of which is as electrical insulators for the converter of the in-core thermionic generator. While it is conjectured that properties of D and DLC should make it ideally suited for this, there is dearth of solid experimental data under planned thermionics operating conditions.

The purpose of our work is to fill this need for data. In specific terms, our goals are to:

(i) Optimize several methods of D & DLC depositions,

(ii) Characterize the thermomechanical, optical and electrical properties at room and elevated temperatures, and

(iii) Determine the effects of fast neutron and gamma irradiation on the structure and properties of these films.

D and DLC films using a variety of H$_2$/CH$_4$ gas mixtures, have been deposited on silicon and niobium substrates, using four different deposition techniques. These include i) RF-coupled plasma enhanced CVD method; ii) RF-induced plasma enhanced CVD method; iii) laser assisted CVD process and iv) micro-wave assisted CVD method. The resultant films have been examined using optical and scanning electron microscopy and Raman spectroscopy. The results with respect
to the two techniques involving RF assisted plasma are very encouraging. The laser based and the bare wire ionization techniques have not been fully explored at this time.

Very good to excellent diamond coatings have been achieved through RF based methods with the substrate temperature being about 950°C and H₂/CH₄ (volume) ratio being about 50 at a flow rate of approximately 100 cc/min. With lower substrate temperatures (T < 600°C) diamond like carbon coatings are produced. Raman spectra and SEM and optical microscopy confirm above results.

A high vacuum, high temperature electrical resistivity measuring apparatus capable of going up to 1200°C, was designed and fabricated specifically to measure the highly insulating D and DLC films. The resistivity measurements are in progress and initial results indicated that in the case of pure diamond coating, the resistivity of 5 μm thick coating is in excess of 10⁵ Ω cm at temperatures of up to 600°C.

Plans are being made to irradiate D and DLC films in the EBR-II reactor to study the behavior of these insulating materials under the simultaneous exposure to neutron, gamma fluxes and high temperatures.

1.0 Introduction

Solution of the sheath insulation problem in the thermionic converter is central to the effective utilization of thermionics for high temperature energy conversion. A typical in-core thermionic fuel-element is shown schematically in Figure 1. The insulator is located between the inner and the outer sheaths.

In so far as the insulator is concerned, the properties of most interest are thermal conductivity, electrical resistivity and structural stability. The higher the electrical resistivity and the thermal conductivity, the better the overall performance of the fuel-element. In addition, for space based applications, the specific values of these properties (the thermal conductivity and electrical resistivity per unit mass) are of extreme importance. The adhesion and structural strength are also of importance.
insulator has to withstand a certain minimum number of thermal cycles, caused by the start-up, shut-down and power-ramp operations.

The basic design criteria for the insulator for a nuclear power reactor in the range of 0.5 to 5.0 MW(e) are as follows.

1. Operating temperature range - 900 to 1300 K
2. Applied voltage in the range of 15-100 V
3. Minimum thermal conductance -- 1 W/cm-K
4. Maximum fast neutron fluence of $4 \times 10^{22} \text{n/cm}^2 (E > 0.1 \text{MeV})$
5. Operating lifetime of about 7 years

Additional requirements are thermal compatibility with the niobium substrate and compatibility with cesium gas.

The development of a high temperature electrical insulator of a high thermal conductivity which can successfully operate without mechanical or electrical degradation in the hostile environment of high temperature and high neutron fluences has plagued the thermionic program for a number of years. Most of the previous insulator work has concentrated upon conventional ceramics technology involving hot-pressed oxides. However, the oxides are unsatisfactory because of the problem of ionic conduction, large irradiation caused swelling, poor ceramic-metal (Nb) bond joint and inherent structural brittleness.

Diamond coatings, on the other hand, have the potential for satisfying all the needs as the insulator for the thermionic generator. The thermal conductivity of diamond is the highest of all known materials -- for example, it is about 5 times that of copper at room temperature. Theoretical electrical resistivity is also, the highest of all material and is about equal to $10^{70}$ ohm-cm. Because of the impurities commonly present in the natural diamonds the experimentally measured maximum electrical resistivity is only
about $10^{17}$ ohm-cm. Nevertheless, the value of $10^{17}$ ohm-cm is several orders of magnitude greater than the best of the experimentally measured resistivities of all oxides and ceramics, including sapphire. All of the structural strength parameters of diamond are also very favorable.

In this paper we describe our recent diamond deposition experiments and the various results obtained from the deposited films. In particular, we have developed independent deposition techniques and a special high temperature and high vacuum resistivity measurement apparatus capable of reaching up to 1200°C. Finally, we present a brief discussion of the current state of the theoretical understandings on thermal stability and irradiation effects as a prelude to our planned future work.

2.0 Experimental

2.1 General Process Description

There are various practical methods of producing diamond coatings. While an exact, detailed theory based on first principles for diamond deposition has not been developed by anyone as yet, the diamond deposition has been successfully attained through, in general, by variations of the CVD technique. A generalized CVD process is summarized through the following steps:

a) A mixture of an inert gas or hydrogen together with a hydrocarbon gas is dissociated through rf or other types of ionizing mechanism -- at an absolute pressure value of less than 100 torr. The basic thermodynamic equations are:

$$CH_4 + Ar \xrightarrow{\Delta H} C^+ + 2H_2 + Ar + ne^-$$

$$CH_4 + H_2 \xrightarrow{\Delta H} C^+ + 3H_2 + ne^-$$

The energy of dissociation, $\Delta H$ is provided by and through the high temperature, or by the rf, or by the laser or by direct ionization. In
some instances, it has been noted that the substrate itself acts as a catalyst in the initial stages of dissociation process.

b) The dissociated gas is usually mixed with a stream of hot hydrogen gas. Typical value of the ratio of hydrogen to the hydrocarbon gas is in excess of 90.

c) A suitably heated target surface is exposed to the flux of dissociated gas.

d) The carbon from the dissociated gas deposits on the surface with diamond or graphite structure.

e) The hot hydrogen gas molecules and hydrogen ions (present from the dissociation process) combine with carbon in the graphitic form, forming hydrocarbon gaseous molecules -- leaving behind a clean diamond structure.

Good diamond/diamond-like carbon films can be formed by controlling the deposition and removal rate of graphitic carbon formed simultaneously to the formation of the diamond structure. Thus, the rate of graphite etching should equal or exceed the graphite deposition rate.

In addition, the surface condition has to be just right for the deposited diamond to adhere on to the surface. If the stresses induced by differential thermal expansion exceed the allowable shear stresses at the interface, upon cooling or repeated thermal cycling, the diamond coating may spall-off. Therefore, in some instances, a suitable thermal expansion matching-layer may be required to be deposited on the substrate of interest prior to diamond film deposition.

2.2 Deposition Methods:

Since the nucleation and growth of the diamond lattice is dependent upon the plasma details, we have chosen to deposit D/DLC films using five independent techniques, as described later in this section, to determine,
which of these would produce the best insulator for our intended use. Additionally, since some of the properties are known to be dependent upon the manufacturing technique, (i.e., plasma chemistry) we have decided to integrate manufacturing methodology into our overall goal of producing effective insulator material for the thermionic fuel element.

We have successfully deposited diamond and diamond like carbon films via LCVD, plasma-coupled CVD and micro-wave CVD techniques. In addition, two more apparatuses have been constructed for depositing D/DLC coatings. These include direct ionization CVD and plasma-induction CVD methods. Figures 2 through 6 schematically show the design drawings of these apparatuses. We are able to vary all of the chemical concentrations for each of these techniques.

The principal features of the LCVD or the photoionization method (Figure 2) consists of i) the excimer laser beam, ii) the rotating (source) graphite target, iii) the substrate holder with independent temperature controller, iv) the inlet stream of mixture of H₂: Ar: O₂ gases, v) independent substrate bias controller (0-2 KV), vi) quartz chamber construction, vii) variable substrate-to-target distance adjuster, viii) high vacuum system and ix) the MKS brand gas flow (mass) controller (not shown in Figure 2).

In the photoionization method (Fig. 2) a highly intense laser beam hits a rotating graphite target and creates an ionized plume of carbon atoms. These ionized carbon atoms are attracted on to a hot substrate kept at a negative potential (0-2 KV). The ionized carbon atoms nucleate into graphite and diamond structure (graphite to diamond ratio of between 1000 to 10,000). A continuous stream of ionized hydrogen atoms is allowed to impinge upon the substrate. The ionized hydrogen attacks preferentially the graphitic-carbon, leaving intact the diamond nuclei. Thus, if the process is continued, the diamond nuclei continue to grow and form the diamond film. The actual apparatus, in the as-built state is shown in the photograph of Figure 7.

The laser parameters and other details of the deposition runs were as follows:

excimer laser (wavelength = 308 nm)
Pulse energy 300 mJ

f = 125 Hz or 250 Hz

Pulse width = 20 ns

Beam spot size = 0.5 mm x 1.5 mm

Pulse fluence = 40 J/cm²

The RF coupled sputter deposition process is schematically shown in Fig. 3. The basic underlying principle is somewhat different from the photoionization process. The RF coupled sputter deposition process converts carbon from a hydrocarbon gas, such as methane (CH₄) into diamond and graphite structures. Only a minor fraction of the deposited film originates from the sputtering of the graphite cathode. Figure 8 shows the actual apparatus while Fig. 9 shows the RF-glow discharge around the substrate during one of the deposition runs. The RF power generator and controller are the Advance Energy/U.S. Gun Model #RFX-600 and #8TX-600, respectively.

The RF induced deposition technique is somewhat similar to the RF coupled sputter deposition method except that there is essentially no sputtering taking place. Figure 4(a) shows a schematic sketch of the RF induced deposition apparatus, while Figure 4(b) shows the actual apparatus. No deposition runs have been made on the apparatus as yet. The RF power generator is the LEPEL Model #T-5-3-DF-J-S.

The direct ionization method is schematically shown in Figure 5. This is the simplest of all methods. It consists of essentially a vacuum system and a hot wire to ionize the source gas and hydrogen. The drawbacks of this method are: short life span of the hot wire, leading to frequent changes and low ionization efficiency of the hot wire.

Finally, three independent runs were made using microwave assisted CVD apparatus (ASTEX model HPMS system (Figure 6)). Two of the runs were made for
a single crystal silicon substrate of (100) orientation and the third one was made for a polycrystalline niobium substrate. These three runs were made at -500°C, -800°C and -950°C (on niobium) respectively.

2.3 Apparatus for Resistivity Measurement

A high vacuum-high temperature resistivity apparatus is schematically shown in Figure 10. The specimen holder and the electrical connection details are schematically shown in Figure 11 while the actual, as-built apparatus is shown in Figure 12. A clean high vacuum is provided (final stage) by a cryopump. The furnace is capable of continuous and stable use of up to 1200°C.

2.4 Substrate Preparation

Except for one deposition run, all runs have been made using (100) single crystal silicon substrates. The one exception was with a polycrystalline niobium substrate.

The silicon substrates were further conditioned by one or more of the following operations:

1) electro-mechanically polished (mirror polish-rms < 0.01 μ)

2) chemically etched using 49% conc. HF acid

3) mechanically scratched using 3 or 6 or 9 μ diamond polishing powder.

The niobium substrate was mechanically polished (final stage) by using 6 μ diamond powder.
3.0 Results

3.1 RF Couple CVD Method

The results from the RF coupled technique are very interesting. The coatings were extremely hard (scratch test on MOH scale > 8.0) and adherent. The effect of the scratches on the nucleation and growth phenomenon is quite vividly shown in the optical photomicrograph of Figure 13. Figure 13a was taken from the transition region between electropolished and 9 µ deep scratched region. The electropolished region (left 1/3 of Figure 13a) is essentially free of any deposition. The scratched region (right 2/3 region, Figure 13a) has good coverage. Figure 13b (at 520x) shows the coating on the fully scratched up region of the substrate. Some of the specimens have been examined via a scanning electron microscope (Model JEOL 400). Figure 14, 15 and 16 are from a typical deposit as developed on the chemically etched surface. Figure 14 is a low magnification (150 x) photomicrograph. The dark band about 3/4" wide running from top to bottom is the scratch from the scratch test -- which was performed to determine the hardness of the coating. The coating is (clearly) seen to be (i) rough and (ii) made up of many "hills" and small "domes." Figure 15 is a higher magnification (10,000 x) photomicrograph taken from a region near the scratch mark in Figure 14. It shows that (i) the coating itself is about 1.0 µ thick and (ii) that the surface contains many small "islands." The "islands" are more clearly shown in Figure 16 (at 40,000x). These photomicrographs indicate that the diamond/diamond like carbon film develops via the classical nucleation and growth phenomenon.

Finally, a few Raman spectra were obtained on some of the specimens. A typical Raman spectrum is shown in Figure 17. Both the diamond and the graphite peaks are present, indicating that these coatings are not made of pure diamond lattice and hence, are of the DLC type. We believe that had the substrate temperature been 1000°C, instead of 700°C, the coatings would have been of pure diamond lattice.
3.2 Microwave Based - CVD Method

3.2.1 Silicon Single Crystal (100) Substrate

There were two runs made for silicon single crystal substrates. During the first run, the specimen was at too low a temperature (~500°C) and it resulted in a sooty deposit. The other sample was deposited at 800°C and it developed a diamond like carbon coating of about 10 μm average thickness, as shown in the photomicrographs of Figures 18 and 19. However, the coating does not appear to be of pure diamond -- there are graphite like platlets distributed through the thickness, as indicated at locations marked "A" in Figure 19.

With regard to the niobium substrate, the deposition run was made at 950°C. The resulting deposit appears to be made of pure diamond crystallites, as shown in the Figures 20 and 21.

3.3 Resistivity Measurements

Resistivity measurements were made on the two micro-wave CVD samples -- DLC on silicon (Spec. #86) and D on niobium (Spec. #89). The results are very encouraging and shown in the graph of Figure 22. As seen in Figure 22, the resistivity of the diamond film is about one to two orders of magnitude greater than that of the diamond like carbon coating.

4.0 Discussion

4.1 D and DLC Deposits

From our preliminary results, it appears that it is possible to deposit D and DLC films using the CVD type methods. Additional deposition runs will elucidate the detailed conditions necessary for developing either D or DLC films. However, from our preliminary data, it appears that for substrate temperature of less than 800°C, pure diamond film does not result. Instead, a mixture of diamond and graphite composite film results. For substrate temperature of 950°C, pure diamond lattice results. Thus, the key parameter,
in addition to the plasma chemistry needs, is the substrate temperature during deposition. It is conjectured that at low ($T \leq 800^\circ C$) substrate temperatures, not all of the graphite is etched away by the hydrogen ions.

4.2 Thermal and Irradiation Stability

Two major issues relating to the use of diamond or diamond films in a reactor are the stability of the diamond form with respect to temperature as well as stability with respect to neutron irradiation. Although very little data is available for diamond films, a great deal of data exists for natural diamond. It is expected that diamond films will be comparable to natural diamond, although this statement needs to be verified through additional future work.

Several investigators have indicated that diamond in its natural form will anneal radiation damage. An excellent example of this is provided by Woods [1] for natural diamond irradiated with energetic neutrons and electrons. Wood's data indicated that essentially all of the damage was annealed out between 600°C and 800°C.

Vance [2] performed experiments with fast neutron damage of natural diamond at 373 K. Irradiations of $2.4 \times 10^{19}$, $2.5 \times 10^{20}$, $4.8 \times 10^{20}$ and $1.63 \times 10^{21}$ nvt were performed. Fast neutron fluence is expected to be especially damaging because of knock-on collisions caused by the scattered nucleus. Thermal reactors are expected to be much less damaging than fast reactors. Transmutation of the carbon nucleus is very rare (the thermal cross section is only .004 bn).

Most of the samples experience some density decrease upon irradiation, indicating that damage had occurred and some lattice expansion had taken place. Radiation swelling of diamond results as localized vacancies in the lattice migrate and eventually coagulate, forming macroscopic voids and pores. X-ray crystallographic analysis showed that at $2.6 \times 10^{20}$ nvt, diffuse scattering occurred, indicating that the lattice had become more disordered. Samples were annealed for one hour periods at temperatures ranging from 923 K to 1923K (some samples were annealed several times at increasing
The results indicated that for samples with volumetric swelling of less than 12%, annealing was effective in restoring the samples to their original size and single-crystal orientation, although some diffuse x-ray scattering was noted. Above 12%, the samples reverted to a semi-amorphous state.

Irradiations by Primak et al. [3] produced similar results. Lattice distortion was observed in many of the samples which were irradiated at up to $10^{21} \text{nvt}$. Successive anneals at temperatures ranging from 350 K to 1270 K were successful in reducing the lattice constant to about the original value.

Qualitatively, the significance of these results to the proposed use of diamond films in a reactor environment is to show that defects in the radiation-damage diamond lattice can be annealed out with temperature. Wood [1] estimated that vacancy annealing temperature is about 500°C for diamond lattice. Because the data by Vance were taken at 373 K, self-annealing was retarded and could not be as effective as it would be at thermionic trilayer operating temperatures conditions, it can be anticipated that many of the localized vacancies which are formed will be annealed out of the lattice before they have a chance to coalesce.

Thermal reactors of the Topaz type would experience fluences within the limits of some of the data. Unfortunately, the aforementioned researchers did not study the effect of radiation damage upon the electrical resistivity. It is presumed that the electrical resistivity would remain high after irradiation and annealing, but this needs to be verified experimentally. It is also presumed that the electrical characteristics of diamond films would be similar to those of natural diamond. This ignores the possible effects of impurity dopants, effects along grain boundaries and so forth. The similarities between natural and diamond film samples need to be verified experimentally under the conditions of high temperature and neutron fluence.
Evans [4] and Davies and Evans [5] have presented considerable data and analysis concerning the behavior of natural diamond at high temperatures. At temperatures above - 1500 K, graphitization of the surfaces occurs. Graphitization is a nucleation process and for the most part does not occur internally except at defect sites, cracks and so on.

Graphitization can be described by the following Arrhenius type of equation,

\[ \frac{dx}{dt} = C \exp \left( -\frac{E_Q}{RT} \right) \]

where \(\frac{dx}{dt}\) is the rate of graphitization in units of microns per unit time, \(E_Q\) is the activation energy (experimentally determined to be 730 +/- 50 kJ per mol, \(R\) is the gas constant and \(T\) is the temperature. Unfortunately, Davies and Evans do not indicate that value of \(C\). However, \(C\) can be calculated knowing that the .02 g test samples (density = 3.51 g/cm\(^3\)) were about 68% graphitized at 2273 K in ten minutes, corresponding roughly to a linear graphitization rate of 0.35 cm per 10 minutes, or .035 mm/min. Thus, the value of \(C\) would be about 2 x 10\(^{-15}\) mm/min. For \(T = 1000K\), and \(t = 7\) years, then the depth of graphitization is only 5.7 x 10\(^{-17}\) mm, a meaningless amount. Thus, if graphitization can be adequately explained as an activation energy process, it would not affect diamond insulators for thermionic applications. Of course, the rate of attack is affected by chemical reactants such as oxygen. It remains to be verified that the diamond will be chemically inert with respect to the refractory metal surfaces in the trilayer thermionic insulator.

Qualitatively, then, it is our expectation that damage produced by irradiation in diamond crystals will be annealed by temperature resulting in a very radiation-hard material. Based on our understanding of the effects of the thermal and radiation environment, diamond and diamond films should be extremely stable. It is not expected that temperature and radiation will combine in some synergistic manner to weaken the lattice. If this prediction can be confirmed experimentally, a major technological problem with the thermionic generators will have been solved.
5.0 Future Work Plan

Plans are to proceed along the general objectives outlined earlier, subject to the availability of adequate resources. Specifically, we shall perform the following experiments and/or theoretical calculations and modeling:

a) Fully characterize D and DLC films on several substrates using standard metallography methods and specially developed resistivity measuring apparatus. Data shall be obtained in the temperature range of room temperature to 1200°C.

b) Perform irradiation experiments, using the EBR-II fast reactor facilities. Irradiations shall be performed at several temperatures including the design normal and design basis accident maximum temperatures.

c) Compare data obtained from the irradiated specimens with the unirradiated specimens,

d) Develop theoretical models and understanding of the mechanisms responsible for the measured irradiation effects, and,

e) Modify current apparatuses to facilitate deposition on cylindrical surfaces.
6.0 Reference


Figure 1. Typical In-Core Thermionic Fuel-Element
Figure 2. Schematic of LCVD Apparatus
MEGAHERTZ SYSTEM STRONGLY ASYMMETRIC
(SMALLPOWERED ELECTRODE LARGE GROUNDED ANODE)

PYROMETER
OPTICAL EMISSIONS IN 8-12 μm
SPECTRAL RANGE (NOT AFFECTED
BY PLASMA)

BARATRON
VACUUM
GAUGE

ZnSe
WINDOW

SUBSTRATE

H₂O
COOLED ELECTRODE

FORE
PUMP

DIF
PUMP

FLOW METER

GAS MIXING
CHAMBER

CH₄

MASs FLOW
METERS

H₂

BIRD #43
RF-POWERMETER
IMPEDANCE MATCH

RF MATCHING
UNIT π TYPE

2.2 mH
COIL

M Ω
DC
VOLTMETER

MEASURE
NEGATIVE
DC SELF-BIAS

CHoke

2.2 mH
COIL

13.6
MHz

400-1800 V

50 Ω
OUTPUT

Figure 3. Schematic of the RF Coupled CVD Apparatus
Figure 4a. Schematic of RF-Induced CVD Apparatus
Figure 4b. As-Built RF-Induced CVD Apparatus
Figure 5. Schematic of Direct Ionization CVD Apparatus
Figure 6. Micro-Wave Based CVD Apparatus: Models ASTEX Model #HPMS
Figure 7. As-Built LCVD Apparatus
Figure 8. RF-Coupled CVD Apparatus

(a) Overview

(b) Chamber Detail
Figure 9. View of a Specimen During a Deposition Run: RF Coupled CVD Method
HIGH TEMPERATURE INSULATOR
ELECTRICAL RESISTIVITY MEASUREMENT STATION

Figure 10. Schematic of the Resistivity Measuring Apparatus
Figure 11. Schematic of the Specimen Holder (See Figure 10)
Figure 12. Details of the Specimen Holder (Figure 11) For Resistivity Measurement
Typical photomicrograph showing the effect of the scratches in the development of Diamond/Diamond Like Carbon coatings. Figure 13(a) is from the boundary between scratched and unscratched region. The left 1/3 portion of Figure 13(a) is, essentially, devoid of any coating and it was also in the electropolished condition. The right 2/3 portion shows good development of the coating. Figure 13(b) shows the coating at higher magnification in the fully scratched region (with 9 μ deep scratches).
Scanning electron micrographs of Diamond Like Carbon coatings deposited via RF-coupled technique. The dark band in Figure 14 is the scratch from the scratch hardness test. The coating thickness is measured to be about 1 μm in thickness (Figure 15).
Figure 16.

Details of Figure 15. The individual islands (of diamond) are measured to be about 50 to 100 nm in size.
Figure 17.

A typical Raman spectrum from a DLC coating. Diamond peak at -1332 cm$^{-1}$, carbon peak at -1600 cm$^{-1}$. 
Figure 18. Low Mag Showing Coating Cross Section: Thickness \( \approx 10 \ \mu m \)

Figure 19. High Mag Showing Mixture of Diamond and Graphite Plates ("A")

DLC on Silicon: Micro-Wave CVD: Substrate Temperature 800°C
Figure 20. Low mag (1,000 x)

Figure 21. High mag (4,000 x)

Diamond Film on Niobium: Substrate Temperature 950°C.
Figure 22. Resistivity Measurement Data From DLC and D films: Micro-Wave CVD Method
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THERMIONIC SPACE NUCLEAR POWER SYSTEMS
WITH LONG LIFE: KEY ISSUES AND PERSPECTIVES
OF DEVELOPMENT

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ABSTRACT

The main factors affecting lifetime of thermionic fuel element (TFE) are:
- deformation of the fueled emitters under swelling fuel down to short circuit of thermionic cells;
- the qualitative and quantitative composition of impurity gases in the interelectrode spacing;
- the electrical resistance and electrical strength of the TFE insulation.

The constructional and technological measures for a severe decrease in the mass transfer intensity in interelectrode spacing are described. Cesium vapor supply systems of space nuclear power system (SNPS) "TOPAZ" based on a single usage of cesium vapors are presented; the data on gaseous impurities are given. Cesium vapor supply options are presented schematically and an assessment of their effectiveness from viewpoint of extraction of
impurity gases from interelectrode spacing is given. The electrical insulation resistance and strength requirements in the thermionic reactors are presented. Practical realization of described measures may provide converter efficiency in thermionic reactors of no less than 10% during a three-five-year lifetime, if the range of emitter heat flux is 35-40 W/cm². With such TFE parameters and at coolant temperatures 600°C and 750°C for thermal and fast thermionic reactors respectively mass and dimensional performances of thermionic SNPS will be satisfactory.

INTRODUCTION

Thermionic reactor power systems may be used for providing electric power in space from several kilowatts to megawatt levels. At power levels from 20 kWe and more thermionic SNPS will have an inherent advantage over solar power systems taking into account mass, operation and economic performances in the near term. The basic requirements for the thermionic SNPS are:

- a three-five-year lifetime at initial stage with increasing up to seven-ten years;

- power range of 15-20 kWe to several hundred kilowatts for long duration space applications, and the increasing to megawatt level for advanced space missions of the 21st century (for example, Mars mission);

- compatibility of mass and dimensional performances with possibilities of advanced launchers;

- competitive ability in reliability, safety and cost in comparison with another types of SNPS and solar power systems.
THE DEFORMATION OF FUELED EMITTERS

The lifetime will be primarily defined by a TFE and the long lifetime requirements set limits on the electrical parameters, fuel rod and emitter temperatures.

It is not possible, in the present state of the art, to connect a TFE lifetime with power density and its other parameters. Nevertheless, the certain quantitative conditions may be formulated which are necessary for the realization of long lifetime.

One of the main factors limiting lifetime of TFE is the deformation of the fueled emitters under swelling fuel up to short circuit of thermionic cells. The deformation of the emitters with the uranium dioxide is determined by the following factors [1-4]:
- swelling of uranium dioxide with columnar structure of grains, because for SNPS with power levels beyond 20 kWe fuel temperature exceeds 1700 C and structure rebuilding from equiaxed into columnar structure of grains will be accomplished in several thousand hours;
- creep resistance of emitter envelope;
- creep resistance of fuel rod;
- enough free porosity inside fuel rod;
- gas venting out of fueled emitter.

At burn up of 2...2.5% which is characteristic for SNPS with power levels of up to 300 kWe the porosity of fueled emitters may be as great as 0.7...0.8. Emitter design must fulfill requirements that the creep resistance of emitter envelope to be essentially higher than the creep resistance of fuel at their working temperatures. In this case swelling fuel may be redistributed into in-
inner porosity. The use of refractory tungsten alloys containing Re, Hf, Ta, Nb and so on for the emitter envelope is very promising; as this takes place, the emitter envelope is covered by a fine emissive layer of monocrystalline tungsten (a few tens of micrometer in thickness).

These measures might be effective only when in use a gas venting system from the fueled emitters, since its failure results in short circuit of a thermionic cell at times not more than several thousand hours because of envelope deformation under inner gas pressure at any possible values of envelope thickness. For reliability of gas venting system, e.g. with capillary head, it is essential that the capillary head temperature in isothermal cavity, formed by revaporised fuel surface, should be not lower than the temperature of this surface [1]. This places certain requirements upon the capillary head positioning and upon tube heat resistance from capillary outlet up to the point of its connection with the end of a thermionic cell. Another solutions of gas venting systems at the same conditions of their reliability are possible.

Emitter diameter change of a thermionic cell under swelling fuel may be approximately described using the linear law with time

\[
\frac{dA}{de} = a (q_v, T_e) \cdot (t - t_o),
\]

where \(d_{ei}\) - initial emitter diameter,
\(T_e\) - emitter temperature,
\(\overline{q_v}\) - average volume heat power density,
\(t_o\) - time of changes in structure, consisting small part of SNPS lifetime.
Considering, that for the compensation of degradation processes SNPS thermal power will be increasing within some boundaries approximately linearly over time, emitter diameter change can then be described using equations:

\[ \frac{\Delta d_e}{d_{ei}} = \chi(t), \]  

\[ \chi(t) = \alpha(\bar{q}_v, T_e)(t-t_o) + \beta (t-t_o)^2, \]  

where \( \alpha \) - accidental magnitude,
\( \beta \) - determinative magnitude.

At instant of time \( t \) magnitude \( \chi(t) \) must not exceed a limiting value \( \chi_{lim} \), from beginning with which thermionic cells may be regarded to be completely short circuiting. Thus:

\[ \alpha(\bar{q}_v, T_e) \leq \frac{\chi_{lim} - \beta (t-t_o)^2}{t-t_o} \]  

If we take that \( \alpha(\bar{q}_v, T_e) \) is distributed by normal law with regard to the values of geometry sizes and strength properties, then

\[ B_{ep} \left[ \chi(t) \leq \chi_{lim} \right] = \frac{1}{\sqrt{2\pi} \sigma} \int_{-\infty}^{\kappa} \exp \left[ -\frac{(a-\bar{a})^2}{2 \sigma^2} \right] da = \Phi(\kappa-\bar{a}) = p(t) \]  

where \( \bar{a} \) - mathematical expectation,
\( \sigma \) - dispersion.
Thermionic cell failure probability as a result of short circuit may be written as

$$Q(t) = 1 - P(t)$$  \hspace{1cm} (6)

For the degradation of thermionic reactor electrical performance we need to divide all the thermionic cells into some groups according to heat power density within the limits of $[q_v, q_v + \Delta q_v]$. Let us assume distribution characteristics $(\bar{a}, \sigma')$ in every group to be the same. Then medium number of the thermionic cells which are short circuited at instant of time $t$ in group $i$, containing $n_i$ thermionic cells, will be

$$\bar{n} = n_i Q_i(t).$$  \hspace{1cm} (7)

The total number of thermionic cells which are short circuited in all groups at instant of time may be written as

$$\bar{n} = \sum_{i=1}^{m} n_i Q_i(t),$$  \hspace{1cm} (8)

where $m$ - number of chosen thermionic cell groups.

The medium number of the thermionic cells which are short circuited discharged per one TFE:

$$\bar{x}(t) = \frac{1}{n_{TFE}} \sum_{i=1}^{m} n_i Q_i(t),$$  \hspace{1cm} (9)

where $n_{TFE}$ - number of TFEs in a core of thermionic reactor.

As a condition of TFE characteristic stability we may propose
inequalities

\[ x(t) \leq a, \quad t \leq T_e, \quad a \ll 1 \]  \hspace{1cm} (10)

At given value \( a \) there can be assessed requirements of the magnitudes \( a \) and \( \sigma \) in order to get these inequalities.

THE INFLUENCE OF GAS IMPURITIES

For the lifetime stability of TFE characteristics composition of gas impurities in interelectrode spacing is very important. These gas impurities are responsible for mass transfer processes in interelectrode gap which may lead to the change of radiative and emission-adsorption properties of the electrodes, to additional current losses and destruction of construction elements in fueled emitter. Basically, dioxide fuel is the source of oxygen. When the fuel and other TFE elements content carbon impurities it should be possible the presence of carbonic oxides (primarily, monoxide) in the interelectrode spacing. Carbonic oxides were registered during the ground tests of SNPS "TOPAZ" while taking samples from the cesium cavities pumping out system of the thermionic reactor; monoxide content exceeded dioxide content by nearly one order of magnitude. Flowrate of carbon monoxide reached 4...5×10^\((-5)\) Watts at temperature 300 C after 3000 hours of the ground tests. In this case carbon monoxide pressure in the TFE interelectrode spacing could be estimated as high as 10 Pa.

The interaction of carbonic oxides with cesium vapor may lead to condensed phases formation, for example, \( Cs_2CO_3, CsOH \) and carbon at the collector temperatures when oxides pressures exceeding eq-
ulilibrium over condensed phases. Pressure evaluation of carbon monoxide interelectrode spacing of SNPS "TOPAZ" does not exclude condensation of carbon compound on the collectors.

In thermal thermionic reactor hydrogen from zirconium hydride moderator penetrates to TFE interelectrode spacing. During the ground tests of SNPS "TOPAZ" in which measures for long-term retention of hydrogen was not incorporated, appearance of hydrogen in the interelectrode spacing was detected at 250-300 hours after the beginning of the tests. After 3000 hours of tests the hydrogen flows reached $5 \ldots 7 \times 10^{-4}$ Watts at the temperature 300 K; in the total gas flow hydrogen fraction exceeded 90 %. In this case partial hydrogen pressure in TFE interelectrode spacing could exceed 65 Pa. The negative influence of such hydrogen pressures in the gap on TFE electrical characteristics is connected with the increases of heat conductivity and plasma losses because of the interaction of cesium atoms and electrons with molecules and hydrogen atoms.

It should be noted that the effect of these factors is quite sufficient to explain efficiency degradation ratio on order of 20 % for 4400 hours lifetime, that was observed during SNPS "TOPAZ" tests.

The presence of hydrogen in gap along with carbonic oxides may lead to hydrocarbon formation at the collector temperatures and its subsequent decomposition on the emitter surface. It may result in "carbidization" of the emitter surfaces with formation of compounds (e.g. $W_C$ and $W_2C$) and reduction of its bare work function.
THE MEASURES FOR DECREASE MASS TRANSFER PROCESSES IN TFE GAP

In order to decrease the mass transfer processes in interelectrode spacing the several constructional and technological measures must be performed, which include [1, 5-7]:

- the use of uranium dioxide with oxygen content close to stoechiometric;
- the use of high temperature degassing from all components of TFE (at the temperatures not exceeding operating) reducing gas impurity concentration in it;
- the use of special passages for removing fission and impurity gases which will restrain their penetration into the interelectrode gap;
- the use of special protective covers for decrease hydrogen leakage (from the hydride cavity of thermionic reactor) and prevent hydrogen penetrating into interelectrode gap;
- the use of cesium vapor supply system with sufficiently high flow rates (up to several tens grammes per hour).

The activities performed in the last item are summarized below. Cesium vapor supply system with single cesium usage is used in the thermionic SNPS "TOPAZ". Cesium vapor from the reservoir that is maintained at given temperature level by means of electric heater, passes through the inlet commutation chamber, through all connected hydraulically in parallel to interelectrode gaps of TFEs, through the outlet commutation chamber further the throttle, limiting cesium vapor flow rate in the system and reach pyrolytic graphite cesium trap. With the fixed amount of liquid cesium in the reservoir the cesium vapor flowrate unambiguously determines the lifetime of SNPS. The trap capacity is
designed for absorption of the whole amount of cesium. The non-condensed gases are removed from the trap into the outer space.

The application of such cesium supply system is essentially restricted by following factors:

-the volume and the mass of the reservoir increase proportional to lifetime SNPS at fixed vapor flowrate;

-the cesium vapor flowrate is in principle restricted (a few tens of grammes per day), due to interelectrode gap hydraulic resistance and increasing of electric losses when cesium vapor pressure drop in TFE reach value of about 130 Pa.

To cancel the first of this restrictions it is efficient to employ multiple-usage cesium vapor supply systems in which the flowrate doesn't practically connect with lifetime of SNPS and on the amount of liquid cesium. It is such systems that are most perspective for longlife SNPS.

The cesium vapor generator in such systems may use the principle of heat pipe with zones of evaporation and condensation, from latter of these removal of noncondensed gases may be provided.

The determining value for distribution of impurity gases partial pressures is a quantity

$$\Theta = \frac{u(\bar{\rho}) \cdot \Delta \rho \cdot L}{\Delta \cdot \pi \cdot d_e \cdot \bar{\rho} \cdot D},$$

where $\bar{\rho}=(\rho_1+\rho_2)/2$ - mean cesium vapor pressure in gap;

$$\Delta \rho = \rho_1 - \rho_2;$$

$\rho_1,\rho_2$ - pressures in the inlet and outlet commutation chambers, respectively;

$L$ - TFE length;
$d_e$ - diameter of the fueled emitter;
$\Delta$ - size of interelectrode gap;
$D$ - diffusion coefficient of impurity gases in cesium vapor (distinctive for different gases);

$u(\bar{P})$ - gas-dynamic conductivity of interelectrode gap.

Since $u(\bar{P}) = \Delta^{3/4} \bar{P}^{1/4}$, where $\bar{d} = (d_e + d_c)/2$, and $d_c$ - is the collector diameter of TFE, then the quantity $\Theta$ does not depend on $L$ and very slightly depend on $d_e$.

One-dimensional equation for the distribution of impurity gases pressures in the interelectrode spacing $P_g$, considering those gases as being "freezed-in" into the cesium vapor flow and the gases generation rate is constant at the gap length, can be written as

$$
\frac{d^2}{dx^2} \frac{P_g}{\bar{P}} - \frac{\Theta}{L} \frac{d}{dx} \frac{P_g}{\bar{P}} = \frac{Q_g}{\Delta \cdot \iota \cdot d_e \cdot P \cdot D \cdot L},
$$

where $Q_g$ - the amount of total gases generation in TFE.

Using the solution of this equation we can obtain the next expression for the mean pressure of impurity gases in the interelectrode gap:

$$
\bar{P}_g = \bar{P} \frac{Q_g}{Q_{cs}} \left[ \frac{1}{\Theta} (1 - M) - M + \frac{1}{2} \right] + \rho_0 \cdot M,
$$

$$
M = \frac{1}{\Theta} \left[ 1 - \exp(-\Theta) \right],
$$

where $Q_{cs}$ - cesium vapor flowrate in a single TFE;
$\rho_0$ - the pressure of the impurity gases in the outlet commutation chamber of thermionic reactor, which
is defined from the equation:

$$
\rho_0 = \rho_0 \frac{n_{TFE} Q_p}{Q_\Sigma},
$$

(14)

where $Q_\Sigma$ - total cesium vapor flowrate in this chamber.

It seems interesting to compare the cesium vapor supply system of the SNPS "TOPAZ" with that of the one-side diffusion scheme for removal of the impurity gases from interelectrode spacing. For this scheme the mean pressure of impurity gases in gap is described by expression:

$$
\bar{p}_g = \bar{p}_c s \left( \frac{n_{TFE} Q_p}{Q_\Sigma} + \frac{1}{3} \frac{Q_p L}{\Delta \mathfrak{m} \cdot d_e \cdot \rho \cdot D} \cdot \frac{T_{ig}}{T_t} \cdot \xi_{em} \right),
$$

(15)

where $T_{ig}$ and $T_t$ - temperatures of the impurity gases in the gap and in the throttle, respectively ($T_{ig} = (T_e + T_c)/2$);

$\xi_{em}$ - is the coefficient, considering the influence of electromagnetic forces upon the distribution of the cesium vapor pressure in the gap.

At $\Delta p_{em}/p_{cs}^{in} \leq 0.1$, the value $\xi_{em} \approx 1$, $p_{cs}^{in}$ - cesium vapor pressure in the inlet of TFE, $\Delta p_{em}$ - cesium vapor pressure drop in gap, due to electromagnetic forces. In this scheme it is assumed that the cesium vapor is pumped through the common for all the TFE cesium chamber.

We shall introduce the ratio of mean impurity gas pressures in both considered cesium systems:
\[
\alpha = \frac{I}{e} \left( 1 - \frac{M}{T_{FE}} \right) + \frac{I}{2} \left[ 1 + \frac{3}{2} \frac{n_{TFE}}{T_{FE}} \frac{T_{i}}{T_{t}} \frac{T_{i}}{T_{t}} \right],
\]

From the condition that the cesium vapor pressure drop in inter-electrode gap does not exceed 120 Pa we may evaluate the limiting values of \( \Theta \). For \( \Delta = (0.4 - 0.5) \times 10^{-3} \) m, \( T_{ig} = 1400 \) K, and \( T_{t} = 850 \) K this value for technological gases, with the molecular mass near 28, is of about 1 and of about 0.6 for hydrogen. At the above values of \( \Theta \), we obtain that \( \alpha \approx 0.7 \) at \( \Theta = 0.6 \) and \( \alpha = 0.6 \) at \( \Theta = 1.0 \).

We consider these values to be the limiting estimates of the advantages of the cesium vapor supply of SNPS "TOPAZ" in comparison with the one-side diffusion scheme, having similar cesium vapor flowrates. It must be noted that at the fueled emitter swelling the value of \( \Theta \) decreases proportionally to the square of the gap size.

Application of systems with multiple usage of cesium vapor makes it possible to improve essentially its effectiveness as far as the removal of the impurity gases from interelectrode gap. Thus, for instance, in the one-side diffusion scheme at

\[
Q_{z} \rightarrow \infty, \quad \bar{P}_{g} \rightarrow \frac{1}{3} \rho_{ar} \frac{T_{i}}{T_{t}} \frac{Q_{g} L}{\Delta \pi d_{g} \bar{P} d}
\]

The evident condition of of reasonable cesium vapor flowrate increase is the following:
A still higher effectiveness may be reached in the two-side diffusion scheme, where the impurity gases are removed from both ends of interelectrode gap into two cesium cavities either by means of diffusion only, or by means of diffusion with the provided pumping of cesium vapor through the gap with limited flowrate; pumping-over of cesium vapor through each chamber is accomplished at a higher flowrate.

At $\theta \ll 1$ and $\xi_{em} \approx 1$, within use of such cesium vapor supply system, the impurity gas mean pressure is defined by the correlation

$$p = \bar{p}_{cr} \left( \frac{\nu_{eff} Q_g}{Q_x} + \frac{1}{12} \frac{Q_g L}{\Delta \Phi \cdot d_e \cdot \bar{p} \cdot D} \cdot \frac{T_i g}{T_t} \right).$$

(19)

Hence at $Q_x \to \infty$, $p = \frac{1}{12} \bar{p}_{cr} \frac{T_i g}{T_t} \frac{Q_g L}{\Delta \Phi \cdot d_e \cdot \bar{p} \cdot D}$.

Reasonable cesium vapor flowrate in this system is:

$$Q_x \geq \frac{12 \cdot \Delta \Phi \cdot d_e \cdot \bar{p} \cdot D \cdot T_t}{L \cdot T_{ig}}.$$  

(20)

The increase of value $\theta$ to the limiting levels in conformity with the above mentioned estimates will lead to a not more than 25 % decrease of impurity gas mean pressure in gap as compared to the case, when $\theta \ll 1$.

**REQUIREMENTS TO ELECTRICAL INSULATION RESISTANCE AND STRENGTH**

To provide the thermionic reactor performance, the insulation
resistance and its electrical strength are of great significance. To restrict the current leakage through the insulation to not more than \( \beta \) - the set part of thermionic reactor total current \( I \), the insulation resistance is to satisfy the condition:

\[
R_{in} \geq \frac{U_{TR}}{\beta I},
\]

(21)

where \( U_{TR} \) - thermionic reactor voltage.

From the condition of electric strength breakdown voltage of insulation \( U_{bd} \geq \gamma U_{TR} \), where \( \gamma \) - is the electric strength margin factor. It is possible to connect the middle point of thermionic reactor electric circuit by means of finite or zero resistance with "mass" then the demands on \( U_{TR} \) may be reduced up to two times.

THERMIONIC REACTOR PERFORMANCES

Theoretical and experimental investigations demonstrate that with use of effective emitter and collector materials, providing vacuum work function of about 5 eV and effective emissivity of electrodes \( E_{ef} < 0.2 \), beginning of life TFE efficiency and mean emitter surface heat density from 25 to 40 W/cm² can reach 12 % at optimal collector temperatures and cesium vapor pressure. For long lifetime stability of such characteristics (taking into account its permissible degradation 20 %) it is essential to realize the above mentioned conditions in the design of TFE and thermionic reactor.

Considering above mentioned parameters of TFE in thermal thermionic reactor with maximum core volume of up to 0.08 m³, the electric power on reactor terminals exceeding 100 kW can be
achieved, and the thermionic reactor specific mass, considering its main service subsystems (cesium system, control drives, etc.) will amount to 10-12 kg/kW. The SNPS radiator surface at efficiency of not less than 10 % and of maximum coolant temperature at thermionic reactor outlet not more than 600 C will not exceed 60 m**2.

As for the fast thermionic reactor with minimal critical dimensions on base uranium dioxide fuel, having reactor core volume of about 0.1 m**3, the total emission surface of up to 9 m**2 and using sodium-potassium eutectic alloy as coolant, the mean mass characteristics will be to 5-6 kg/kW. The SNPS radiator surface at electric, power thermionic reactor about 300 kWt and maximum coolant temperature of up to 750 C will be to 75-80 m**2. At the above mentioned characteristics, the SNPS having thermal and fast thermionic reactor at electrical power of up to several hundred kilowatts, will be fully compatible with the possibilities of space launchers.

CONCLUSIONS

On the base of available theoretical and experimental data it is possible to formulate the requirements to TFE and thermionic reactor of long lifetime thermionic SNPS. At optimal parameters of TFE emitter and collector units and realization of the design and technological measures aimed at providing the TFE electric characteristics stability, during the lifetime of about several years and at mean emitter heat densities of 25-40 W/cm**2, conversion efficiency not less than 10 % may be achieved. At such TFE parameters SNPS with thermal and fast thermionic reactor will
have satisfactory mass-dimensional characteristics fully compatible with the possibilities of space launch vehicles.

Acknowledgements

This work was conducted at the SIU "Krasnaya Zvezda" (Moscow, USSR) under funding of SIU.

References

7. Gryaznov G.M., Zhabotinsky E.E., Zrodnikov A.V., Serbin V.I.,
SAVTEC, a self-adjusting versatile thermionic converter, has potential application to advanced incore-thermionic power systems. A good lead efficiency, typically 10% or more, is achievable with SAVTEC at relatively low emitter temperatures, e.g. 1300 K to 1400 K. This contrasts to the emitter temperature of 1700 K to 1800 K for the normal ignited mode thermionic converter. The lower operating temperature of SAVTEC allows the fuel cladding to be at a lower temperature where it has greater creep strength. By making the cladding strong relative to the fuel, this reduces the fuel swelling problem. Lower temperatures may also permit the tungsten inside the reactor to be replaced by niobium for better neutronics. Also, SAVTEC can operate efficiently at lower power densities than conventional thermionics. The combination of lower temperatures and lower power densities than conventional thermionics permits larger incore fuel elements to be used in fast reactors. This can lead to reducing driver rod requirements in fast reactors.

The feasibility of the SAVTEC concept of close-spaced thermionic converters was demonstrated five years ago. Additional work is needed to demonstrate their manufacturability, reliability, and lifetime. SAVTECs have a system reliability advantage because they can be tested and replaced as necessary before launch. In the radiatively coupled TFE design, the SAVTEC TFE can be tested out of core similarly to single-cell TFES. With additional manufacturing experience, improved quality control procedures, and improved designs, we believe that manufacturing yield can greatly improved. Several design improvements have been suggested for improved manufacturability and reliability. These include: (1) the use of a pedestal with improved stiffness, (2) careful surface polishing before assembly, (3) improved lead attachment, and (4) the choice of materials which prevent cold-welding in space. A manufacturing procedure is discussed which attempts to automate production while maintaining careful quality control.
II. Introduction

It has long been recognized that near ideal thermionic converter performance could be achieved if the inter-electrode gap could be reduced to a few microns. SAVTEC (Self Adjusting Versatile Thermionic Converter) technology has achieved these spacings. In the SAVTEC concept, the inter-electrode spacing is controlled using thermal expansion. This approach is potentially important in both solar and nuclear power applications. The principal issues to be resolved are reliability and lifetime. In particular, SAVTEC power density, efficiency, and operating temperatures appear to be well suited for in-core nuclear reactor concepts.

A schematic of a research SAVTEC converter is shown in Figure 1. The emitter lead also functions as a structural support for the emitter. Thermal expansion of this lead causes the emitter to separate from collector by a few microns. In this design, the emitter is radiatively heated. For higher total power, many SAVTEC converters can be combined on a single substrate as shown in Figure 2.

This paper will review demonstrated SAVTEC performance and address issues of nuclear SAVTEC design and SAVTEC manufacturing. Some concepts for nuclear thermionic fuel elements (TFEs) that use SAVTEC will be discussed in Section IV. SAVTEC design options for improved reliability will be presented in Section V along with the outline of a possible manufacturing procedure.

III. SAVTEC Performance

SAVTEC converters operate in the quasi-vacuum mode of thermionic conversion. By maintaining the inter-electrode gap as small as a few microns, space charge effects can be minimized. Thus, the voltage loss associated with the plasma in ignited mode converters is eliminated. The resulting efficiencies may be very high, approaching that of the vacuum ideal converter.

A series of SAVTEC experiments were performed at Rasor Associates several years ago. Measured lead powers are shown in Figure 3. At low emitter temperatures, and hence low current densities, there is good agreement with vacuum diode theory. At higher emitter temperatures, the behavior is described by unignited theory.

A comparison of efficiency versus emitter temperature for various types of thermionic converters is shown in Figure 4. Two theoretical lines are shown for barrier voltages of 1.6 and 2.0 V. A SAVTEC converter, denoted S15, was operated at 1750K and achieved an efficiency of 18%. This is much higher than the efficiency of conventional ignited mode thermionic converter as represented by the TFE design. Other SAVTECs are shown operating at lower temperatures. Comparison with the Vb=2.0 V line, shows that performance was substantially better than that of an ignited-mode converter.
SAVTEC THERMIONIC CONVERTER

Platinum or Mo/Ru Braze
Emitter
Blackbody Hole
Emitter Lead
Collector
Ceramic Insulator
Emitter Lead Braze
Emitter Base
Ceramic Insulator
Retaining Lip

(-) Output Voltage Thermocouple
(+) Output Voltage

FIGURE 1.
DIAGRAM SHOWING COMPONENTS OF SAVTEC POWER CONVERTERS

Heat Wall

Emitter (typ)

Collector (typ)

Ceramic

Diode Gap

Radiation

0.5 cm

0.5 mil

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Figure 3. Measured maximum output from a Savtec diode.
SAVTEC DATA FROM THE EXPERIMENT AND THEORY CURVES

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**FIGURE 4.**
Some SAVTEC experimental results are summarized in Table 1. The rows marked SAVTEC refer to tests on individual converters. The rows marked Hotshell refer to SAVTECs operated in an array inside a combustion heated hotshell. The hotshell arrangement is illustrated in Figures 5 and 6. Not all the SAVTECs installed in this hotshell operated reliably. Sample results are shown in Figure 7. The SAVTECs which successfully opened in that test are marked with X. The others did not open in that test. For two of these, the problem appeared to be inadequate collector cooling which reduced the desired differential thermal expansion. To further complicate interpretation of the results, the combustion provided uneven heating and the SAVTEC converters which were installed were designed for operation at a uniform temperature. This may explain why those SAVTECs towards the outside of the hotshell, where it was cooler, were more likely to fail to open.

IV. In-Core SAVTEC Reactor Concept

SAVTEC has several potential advantages for in-core nuclear reactor operation:

1. A good lead efficiency, typically 10% or more, is achievable with SAVTEC at relatively low emitter temperatures, e.g. 1300 K to 1400 K. This contrasts to the emitter temperature of 1700 K to 1800 K for the normal ignited mode thermionic converter.

2. The lower operating temperature of SAVTEC allows the fuel cladding to be at a lower temperature where it has greater creep strength. By making the cladding strong relative to the fuel, this reduces the fuel swelling problem.

3. The lower temperatures may permit the tungsten inside the reactor to be replaced by niobium for better neutronics.

4. SAVTEC can operate efficiently at lower power densities than conventional thermionics. The combination of lower temperatures and lower power densities than conventional thermionics permits larger incore fuel elements to be used in fast reactors. This can lead to reducing driver rod requirements in fast reactors.

5. SAVTEC converters can be tested out of the core.

6. The fuel body can be removed leaving the SAVTECs in place.

A conceptual arrangement for SAVTECs within a thermionic fuel element (TFE) are illustrated in Figure 8. The walls of the TFE are lined with SAVTECs which are heated radiatively by the uranium fuel. As shown in Figure 8, the fuel is hexagonal. Fuel swelling may cause distortion in this hexagon. In this case, it may be better to state with a circular fuel body as shown in Figure 9. Note that swelling of the fuel affects SAVTEC performance only if it physically touches and damages the SAVTEC. Unlike conventional TFEs, the fuel cladding is not the emitter. The SAVTEC design is shown in more detail in Figure 10. The SAVTECs may be connected in series along the length of a TFE as shown in Figure 11.
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<tr>
<td>HOTSHELL B</td>
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<td>960</td>
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<tr>
<td>SVT-1B</td>
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<td>800</td>
<td>6.5</td>
<td>1.56</td>
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</table>

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**TABLE 1**
Cross-Section of Converter Array and Cesium Envelope

Schematic of Converter Array Prior to Installation of Emitters

FIGURE 5.
SAVTEC ARRAY

SAVTEC Array Assembly Prior to Installation in Hot Shell

Completed Converter Prior to Testing

FIGURE 6.
19-CELL FLAME-HEATED SAVTEC EXPERIMENT

Test 4

A S B C S
D E E F G S
H I J K L
M N O P
Q R S S

X Open
X Shorting Due To High Collector Temperature
S Unopened

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THERMIONIC FUEL ELEMENT
With Hexagonal Fuel Body

Emitters
Collectors
Fuel Cladding (Tungsten)
Central Void
UO₂

FIGURE 8.
FIGURE 9.

THERMIONIC FUEL ELEMENT
With Round Fuel Body

Emitters
Collectors
Fuel Cladding (Tungsten)
Central Void
UO₂
Outer Sheath
FIGURE 10.
FIGURE 11.

THERMIONIC FUEL ELEMENT

Collector
Emitter Base
Emitters
Voltage of a Series of SAVTEC Cells

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092589-30
Unlike conventional flashlight TFE designs, the SAVTEC converters are not in physical contact with the fuel or its cladding. Furthermore, when out of the core, the fuel cladding can be resistively heated to test the SAVTECs. The fuel rod may even be removed if desired. This permits SAVTEC TFEs to be constructed with greater reliability.

V. SAVTEC Manufacturing Issues

While SAVTECs have been built and tested in a research environment, reliable manufacturing of them has not been demonstrated. 60-90% of the early SAVTECs worked at least once. Further, 40-60% appeared to work reliably. At 40-60%, the yield is better than many space PV cells. An alternate design with possibly greater reliability is illustrated in Figure 12. In this design, the emitter is supported by a ceramic pedestal. For greater mechanical stability, the pedestal has a much larger outer diameter than the emitter support shown in Figure 1. Thermal expansion of this pedestal is responsible for lifting the emitter off the collector under operation.

If the converter must be cycled on and off, there is the possibility of cold welding between the emitter and collector. This can potentially be prevented by coating the collector with a metal such as copper. Because the molybdenum and copper are not miscible, they cannot cold-weld.

In a practical application, it is likely that many thousands of SAVTEC converters would be required. Some method for automated manufacturing would be required. For the SAVTEC design in Figure 12, an approach for automating the final assembly and brazing is shown in Figure 13. After the emitter and collector pieces have been lapped for surface accuracy, they would be placed in a holder as illustrated. First, the holder would then be placed on a support and the metallized ceramic pedestal would be inserted. The emitter-pedestal braze would then be made. Secondly, the holder would be turned over and the collector-pedestal braze would be made. During this braze, the emitter and collector would be in light contact. The assembled SAVTEC would then be removed from the holder.

The accuracies required in SAVTEC assembly are similar to those in the electronics and optics industries. SAVTEC manufacturing can benefit by using technology they have developed such as lapping and clean environments.

A strong quality control process would be key to successful manufacturing. This would occur in all steps from material procurement to assembly of a reactor. To achieve repeatable results, careful control of the brazing process is particularly important. Also, as shown in Figure 14, testing should be carried out both before and after the TFE is assembled.
IMPROVED SAVTEC DESIGN FOR MORE RELIABLE MANUFACTURING

FIGURE 12.
FIGURE 13.

1) Emitter - Pedestal Brazing
Bottom Rest Positions
the Sapphire Support Tube

2) Turn-over and Braze Collector Base - Pedestal
Emitter and Collector are in
Contact (Shorted) During Braze

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SAVTEC QUALITY CONTROL PROCESS

Fabrication
- Materials Procurement
- Cutting
- Drilling
- Capping
- Brazing

Testing and Thermal Cycling
Burn-in

Reject

Assembly of TFE

Out-of-Core
Testing

Rejects

Assembly of Reactor

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FIGURE 14.
VI. CONCLUSION

Only a very SAVTEC experiments have been performed so the data is limited. Results to date indicate high efficiency and good agreement with theory. Applications have been identified for solar, nuclear, and flame-heated systems. Of the few that have been fabricated, a 40-60% manufacturing yield is indicated. Many of the manufacturing issues are similar to those in the semiconductor and optics industries. By applying techniques borrowed from these industries, a greatly improved yield is expected.
Abstract

The dominant failure mechanism of an in-core thermionic converter is considered to be emitter deformation, which results from $\text{UO}_2$ fuel swelling, and can lead to shorting to the collector and hence to a loss of power output. The performance of each converter in the core of a SP-100 design thermionic reactor was evaluated relative to this failure mechanism in conjunction with the modified LIFE-4 fuel-clad model and experimental converter performance data. The results of the analysis showed that, even in the worst case scenario, initial shorting will start after about four years and that the power system will last an additional two years. Thermionic energy conversion is thus a potential candidate for future space missions.

Introduction

Thermionic energy conversion is a potential candidate for space nuclear power sources for future space missions. A thermionic nuclear reactor power system was designed several years ago for the first phase of SP-100 [1]. The reactor core consists of a close packed array of 176 thermionic fuel elements, TFEs, housed within a cylindrical vessel. Each TFE contains six thermionic converters, with $\text{UO}_2$ fuel inside, connected in series and housed within a cylindrical outer sheath. Control of the nuclear reaction is provided by cylindrical control drums which surround the vessel and by seven in-core shut-down rods. The reactor is cooled by a pumped liquid metal loop and associated heat pipe radiator.

One of the requirements of such a space nuclear power source is the ability to provide power continuously over a long time period, such as 7 years or more. It is thus necessary to determine the possible failure modes and model them so that the lifetime of this type of thermionic reactor can be predicted. The main failure mode that will be considered here is emitter deformation which, as a result of $\text{UO}_2$ fuel swelling, can lead to shorting to the collector and hence lead to a loss of output power. This is considered to be the dominant degradation mechanism. Open circuits are possible, though not likely, but have also been considered. In order to make life predictions a model was developed which combined converter performance data and fuel-clad deformation rates to determine when and where the initial interelectrode shorts would occur and as a
result when the power system would start to fail.

Performance Model

The system model that was developed included a detailed analysis of the projected performance of each converter in the reactor core. The thermionic converter that was assumed for the design is a cylindrical converter with a CVD <110>-W emitter and a niobium collector as depicted in Figure 1. The thermionic converter performance was based on a phenomenological analysis which calculates the current density versus voltage as a function of emitter and collector temperatures, cesium pressure, emitter and collector work functions and the interelectrode spacing [2]. The converter performance model was calibrated to planar converter 46 (tested by Thermo Electron in 1970 [3]) which underwent extensive performance mapping over a wide range of emitter and collector temperatures, cesium pressures and interelectrode spacings. The performance data for a planar converter was used because insufficient data was available for a cylindrical one. However, the difference between them is expected to be small based on the small amount of data that is available. Both planar converter 46 and the cylindrical converter used in the reactor design used a CVD <110>-W emitter and a niobium collector.

The inputs which are required for the thermionic reactor system performance model include the reactor, coolant loop and radiator geometries, the average emitter temperature, coolant loop temperature drop, cesium reservoir temperatures, and the relative power density profile for the reactor. The reactor core was divided into five radial rings and 6 layers, each of which has the potential for a different power density and hence different emitter and collector temperatures. Each emitter temperature is calculated by requiring that the average temperature corresponds to the input value and that the resultant thermal power density profile is attained. The thermal power is calculated as the sum of electron cooling, cesium conduction, thermal radiation and conduction, and resistive heating and the total depends on the emitter and collector temperatures. The collector temperature is calculated by the thermal power rejected from each converter, and the fluid temperature at that particular location within the core. The fluid loop temperature is determined by the total heat to be rejected and the effective radiator area.

The reactor core was designed to try and flatten both the radial and axial power profiles. The core was designed with five radial zones, with the fuel in each zone having a different central hole diameter in the UO₂ pellets. Preferentially removing fuel from the central rings by means of larger central holes, flattens the radial heat generation rate. The core also has six different axial layers, one for each converter within the TFE. Each converter has a different fuel pellet length and thus a different emitter and collector length. The fuel pellet lengths have been shortened in the center of the core and lengthened at the top and bottom of the core in order to flatten the axial heat generation rate. A
constant fuel enrichment has been maintained throughout the core. This effort to flatten the fission power distribution has led to a relatively uniform emitter temperature distribution throughout the core.

The cesium pressure within the interelectrode gap is another crucial parameter relative to the converter performance. Each TFE has to have a single cesium pressure within it, because all of the cells are interconnected. However it is possible to vary the cesium pressure depending on which radial zone the TFE is in. The code thus allows for five different cesium pressures, one for each radial zone. These pressures were optimized to give the best converter performance.

The electrical network for the thermionic converters is also an input. The cells are connected in a series parallel network. The circuit which was considered consisted of four parallel strings of 264 converters connected in series. All cells in series must have the same current, and those in parallel must have a common voltage. This means that most of the cells will not be operating at the optimum point on the I-V curve, depending on the electrical network. The principal degradation mechanisms that were considered in the model are those which either cause a short circuit or an open circuit in the network. In addition, radiator degradation which results from the failure of heat pipes due to the impact of meteoroids and debris was considered.

The primary emphasis on determining the performance of a thermionic reactor as a function of life was emitter swelling, which could ultimately lead to the emitter touching the collector and causing a short circuit. Emitter clad swelling depends on the choice of fuel and clad, what their temperatures are, and what the fuel power density and void fraction and burn-up is. The existing LIFE-4 fuel-clad model was modified after an extensive review of the data on high temperature UO$_2$ fuel clad tests and measured rates of fission gas release [4]. The factors which were identified as most strongly affecting distortion rates are the emitter temperature, the tungsten emitter thickness, and the fuel power density and void fraction. These factors were mapped over their full possible design and operating ranges (with three void fractions and three power densities being considered) so that the emitter distortion rates could be predicted as accurately as possible.

The system model estimates the emitter distortion as a function of time. The interelectrode gap was assumed to be 20 mils (0.5 mm) at the beginning of life and to decrease over time. Both theory and converter test data, indicate that the converter performance will improve with decreasing gap to a maximum at between 5 and 10 mils (0.25mm) and finally dropping off rapidly for a gap of less than 5 mils (.13mm). The converter performance model calculates the performance as a function of the interelectrode gap. However, the emitter does not distort uniformly. The shape of the emitter distortion that was used in the model was based on that
experimentally measured in TFE 6F3 [5]. The code thus calculates two interelectrode gaps, one which is the minimum at the point of maximum distortion and is considered for shorting, and a second which considers the overall emitter distortion and is the effective gap which is used for performance calculations. As a result of the emitter swelling, performance of the thermionic reactor will initially increase with time, thus allowing a reduction in emitter temperatures while maintaining a constant net electrical power output of 100 kilowatts.

Results

The results of the analysis indicates that a beginning of life average emitter temperature of from 1745 to 1767 K is required in order to provide a net electrical power of 100 kilowatts to a user at a distance of 25 meters from the reactor. The range of temperatures is due to uncertainties in heat transfer calculations and mass optimization. The reactor is operated in a manner to provide a constant 100 kilowatts of electric power at a constant voltage of 100 volts throughout the lifetime of the mission. Running the model with the input reactor relative power density profile, presented in Figure 2, resulted in the emitter temperature profiles at the beginning of life and after 60,000 hours as indicated in the Figure.

The results of the model showed that the first interelectrode shorts (considered to be when the interelectrode gap is equal to or less than 3 mils (0.076 mm)) occurs in those converters with the highest emitter temperatures, smallest fuel voids, and highest power densities after 70,000 to 80,000 hours (8 to 9 years) of operation. As a worst case, an emitter deformation rate 23% greater than that predicted by the modified LIFE-4 code was used. This was based on the extreme discrepancy between predicted and actually measured distortion rates in the 6F3 test. In this worst case, initial shorting would occur after 40,000 to 50,000 hours (4.6 to 5.7 years) of operation. The effect of one and two open circuits per year was, also, determined. The effect of two open circuits per year reduced the time at which the first short circuit would occur, in the worst case, to about 32,000 to 42,000 hours. These results are summarized in Figure 3.

The actual life of the power system is not set by the first short (which produces a small reduction in power) but rather by the time at which the reactor coolant temperature exceeds the Curie point temperature in the electromagnetic coolant pumps. This will occur only after the loss of about 30% of the converters in the reactor. The model predicts that such a loss will occur about 20,000 hours after the first short circuit. Hence, the objective of meeting a 7 year (60,000 hours) life with the continuous generation of 100 kilowatts of electric power from a thermionic reactor is feasible.
Summary

A model was developed to predict the lifetime of an SP-100 design thermionic reactor, required to provide 100 kilowatts of electrical power. The system model, which included a converter performance model, a modified fuel-clad model, and a model for the heat removal loop and heat pipe radiator, considered emitter deformation to be the main degradation mechanism. In addition, heat pipe failures caused by the impacts of meteoroids and debris and potential open circuits in the thermionic converter network were considered. The results indicate that under nominal assumptions the first short circuit, caused by emitter distortion, would occur after 8 to 9 years of operation. The results also indicate that, in the worst case scenario, the first short circuits would occur after about 32,000 to 40,000 hours (about 3.5 to 4 years) and that the power system would fail about 2.3 years after the first shorts. Thus, a long-life (at least 7 years) in-core thermionic nuclear space power system is feasible.

Acknowledgements

We would like to thank Dr. G. Fitzpatrick and G. Stapfer for the many helpful discussions. The work described in this paper was carried out at the Jet Propulsion Laboratory, California Institute of Technology, under contract with the National Aeronautics and Space Administration.

References


4. Baars, R., Private Communication

Figure 1. Thermionic Cell
RING

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BOL AVERAGE $T_E = 1767$ K

INPUT REACTOR RELATIVE POWER DENSITY PROFILE

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RESULTANT REACTOR TEMPERATURE PROFILE

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AFTER 60,000 HOURS: AVERAGE $T_E = 1726$ K

RESULTANT REACTOR TEMPERATURE PROFILE

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Figure 2. Reactor Temperature and Power Profile
Figure 3. Emitter Temperature versus Time to First Short Including Open Circuits
INTRODUCTION

The recent success of the in-core thermionic reactor concept, as demonstrated by space-based testing of two 10 kW(e) Topaz reactor units, provides a point of departure for further research and development. The single-cell thermionic fuel element (TFE) used in these reactors offers many operational advantages in contrast to multicell TFEs including: testability outside the reactor core using resistive heating; a relatively small number of manufacturing operations required to build a TFE; simplicity of providing paths for supply of cesium as well as exhaust of fission gas, etc.

However, the significant drawbacks include 30-50% resistive losses due to the high currents and long electrical transmission path; and a corresponding lack of scalability beyond the ten or twenty kilowatt level. Accordingly, the efficiency of single cell TFES is limited to five or six percent, or roughly half of what is attainable in multicell TFES. Thus, above the ten kilowatt level, multicell TFES represent the component of choice, based on today's technology. However, the advantages of simplicity of manufacture and non-nuclear testing are lost.

It is worthwhile to inquire whether the use of advanced technology might be able to extend the usefulness of the single cell diode to higher power levels while retaining the important benefits of simplicity, testability and long life with stable operation.
DESIGN RATIONALE

To this end, there are some broad technical requirements which must be satisfied to achieve a workable design. To begin with, the desire for a small system mass leads to a design of a zirconium hydride-moderated reactor core, which creates excellent nuclear fuel economy. However, the maximum tolerable burnup, together with the energy conversion efficiency of the system places a constraint on the minimum amount of nuclear fuel required for a given energy density. The current state of the art in TFE design is directed towards burnup levels of 4.1% average burnup or less. Thus, a 30 kW(e) system operating for seven years at ten percent efficiency requires an excess amount of fuel equal to

\[ m_{\text{burnup}} = F m_a t/A \]

where \( F \) is the fission rate = \( 9.38 \times 10^{15} \) f/sec for a 300 kW(t) reactor; \( m_a \) is the atomic mass of the fuel = 235 grams, and \( A \) is Avogadro's constant = \( 6.02 \times 10^{23} \) atoms/mol. Thus,

\[ m_{\text{burnup}} = 808 \text{ grams}. \]

The total amount of fuel required at beginning of life is then

\[ m_o = 808 \text{ g} / .041 = 19.7 \text{ kg of uranium, or 22.4 kg of UO}_2. \]

which happens to be close to the minimum critical mass of a hydride moderated thermionic reactor. If the conversion efficiency is only 5%, the fuel requirement is 39.4 kg. Therefore, the critical loading and ultimately the system mass is substantially penalized if an energy conversion efficiency of about ten percent or higher is not realized.

For thermal neutron spectrum reactors, it is neutronically inefficient to have a fuel pin diameter greater than about 1-1.5 cm, because of the flux depression in the center of the element (that is, the center of the fuel is not very accessible to thermal neutrons because they will be strongly absorbed in the first few millimeters of the fuel). Thus the minimum practical emitter area will be achieved with a fuel diameter of about 1.5 cm. Assuming an 85% theoretical density fuel, the number of elements required is equal to
where \( p \) is the density of UO\(_2\), \( M_0 \) is the mass of UO\(_2\), \( d_1 \) is the fuel pellet diameter, \( d_v \) is the diameter of a fission vent tube and \( l \) is the active length of the fuel element. Using a UO\(_2\) fuel density of 9.21 g/cm\(^2\), \( d_v \) equal to 0.3 cm, and an active fuel element length of 35 cm,

\[
n_{\text{min}} = 41 \text{ TFEs},
\]
corresponding to an electrode area equal to

\[
A = n\pi l(d_2)
\]

\[
= 8100 \text{ cm}^2
\]

where \( d_2 \) is the emitter diameter (assumed equal to 0.3 cm greater than the fuel pellet diameter for this case). Thus the maximum power density on the emitter electrode will be no higher than 3.5 W(e)/cm\(^2\). This is significant because it permits the researcher to place an upper bound on the power density of interest for application to this system.

A lower value is obtained by selecting the smaller diameter fuel. For this case, then \( n_{\text{max}} = 114 \), \( A = 15,000 \text{ cm}^2 \) and \( P'' = 2.0 \text{ W(e)/cm}^2 \). Thus a reasonable thermionic operating point can be selected within the range of 2.0 to 3.5 W(e)/cm\(^2\). Higher power density can only be considered in the event that higher thermionic efficiencies can be reached, or if shorter lifetimes are acceptable.

Between the lower and upper bound, the tradeoffs will involve the reactor core size (which will tend to increase as metal is added to each cell) and the thermionic lead efficiency (which will decrease if metal is taken away). Currently, a point design is baselined at an emitter diameter of 1.4 cm and a fuel pellet diameter of 1.1 cm. Figure 1 and Table 1 describe the baseline design.

Because the thermionic converter is inherently a low voltage, high current device, Joule losses are very significant in the power density range required. For example, consider a single-cell TFE with a emitter resistivity \( \rho \). The effective resistance is calculated from the differential version of
VENT TUBE 0.3 CM
FUEL PELLET 1.1 CM
EMITTER 1.5 CM
PLASMA GAP 1.6 CM
COLLECTOR 1.8 CM
INSULATOR 1.81 CM
OUTER SHEATH 1.90 CM

Figure 1. Thermionic Cell Cross-Section

<table>
<thead>
<tr>
<th>TABLE 1. TFE PARAMETERS</th>
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<tr>
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<tr>
<td>EMISSION AREA, CM²</td>
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<tr>
<td>EMITTER TEMPERATURE, K</td>
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<tr>
<td>COLLECTOR TEMPERATURE, K</td>
</tr>
<tr>
<td>CESIUM EFFECTIVE TEMP, K</td>
</tr>
<tr>
<td>CURRENT DENSITY, A/cm²</td>
</tr>
<tr>
<td>ELECTRODE VOLTAGE</td>
</tr>
<tr>
<td>LEAD VOLTAGE</td>
</tr>
<tr>
<td>OUTPUT POWER DENSITY, W/cm²</td>
</tr>
<tr>
<td>EFFICIENCY</td>
</tr>
<tr>
<td>EMITTER EFFECTIVE BARE WORK FUNCTION, eV</td>
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<tr>
<td>CESIATED SURFACE WORK FUNCTION, eV</td>
</tr>
<tr>
<td>COLLECTOR CESIATED WORK FUNCTION, eV</td>
</tr>
<tr>
<td>NUMBER OF TFEs</td>
</tr>
</tbody>
</table>
Ohm's law,
\[ \int_{0}^{V} dV = \int_{0}^{1} \left[ \frac{4}{4\pi(d_2^2 - d_1^2)} \right] J''\pi d_0 dl \]
\[ = J''2\pi d_2 \left( \frac{2\rho_1}{p(d_2^2 - d_1^2)} \right) \]
\[ = J''AR_{\text{eff}} \]
\[ = IR_{\text{eff}} \]

from which \( R_{\text{eff}} = \frac{2\rho_1}{p(d_2^2 - d_1^2)} \). This neglects the variation of thermionic performance along the length of the element, which is adequate for the coarse analysis used here. The total effective electrode resistance, \( R_e \), for the case in which power is extracted from both ends of the TFE, is simply one-half the sum of the collector and emitter resistance.

Defining a load resistance \( R_L \) and electrode resistance \( R_e \), the Kirchhoff equation is
\[ V = J''A(R_L + R_e) \]

Since the power consumed by the load is
\[ P = (J''A)^2 R_L \]
then
\[ R_L = \frac{P}{(J''A)^2} \]

Thus, making the substitution \( I = J''A \),
\[ V = I(\frac{P}{I^2} + R_e) \]
or
\[ I^2 - \left( V/R_e \right)I + P/R_e = 0 \, . \]

The quadratic equation in \( I \) yields two solutions. The solution for higher efficiency and smaller current is the practical solution, or

\[ I = \left( \frac{1}{2R_e} \right) \left[ V - \left( V^2 - 4R_eP \right)^{1/2} \right] \, . \]

This equation also produces a constraint for the minimum value of \( V \) or

\[ V_{\text{min}} = \left( 4R_eP \right)^{1/2} \, . \]

Physically, this means that for \( V < V_{\text{min}} \), it is impossible to produce the required load current. An increase in thermal power will result in more power being dissipated as internal losses.

Substituting the numbers for the actual case at 1.5 cm emitter with \( n = 79 \), and \( P = 190 \) W(e) per half cell, the effective emitter resistance for the case in which power is extracted from both ends is

\[ R_{\text{em}} = \frac{\rho l}{\pi [d_2^2 - d_1^2]} \, . \]

\[ R_{\text{em}} = 50 \times 10^{-6} \, \text{\Omega cm} \times 35 \, \text{cm} / \pi (1.5^2 - 1.1^2) \]

\[ = 0.54 \, \text{m\Omega} \, . \]

Similarly, the collector effective resistance is 0.38 m\( \Omega \). Adding an assumed lead resistance of 0.5 m\( \Omega \) yields

\[ R_c = 1.41 \, \text{m\Omega} \, , \]

\[ V_{\text{min}} = \left( 4 \times 1.41 \times 10^{-3} \, \text{m\Omega} \times 190 \, \text{W} \right)^{1/2} \, , \]

\[ = 1.04 \, \text{v} \, . \]
Thus if $V$ is not at least 1.04 volts, then it will be impossible to produce the required current, no matter what the theoretical I-V characteristic of the diode may be. In other words, the electrode and lead resistance imposes a limit on the slope of the load line on an I-V curve. Note that 50% of the output power is wasted at this voltage, so achieving a reasonable efficiency of about 10% will require that the electrode voltage be about 1.2 volts or higher, as shown in Figure 2.

Alternatives are to increase the thickness of the electrodes (already undesirable because of nuclear criticality considerations), shorten the active length (also undesirable for the same reason), increase the number of TFEs or to use improved technology to develop higher output voltage capability. The last alternative provides the technical challenge to thermionic community.

An obvious method to improve the output voltage of the thermionic converter is to seek higher temperatures, which will raise the effective surface work function of the emitter, while providing adequate electron emission potential.

The state of the art thermionic emitter uses chemical-vapor-deposited (CVD) tungsten. The life limiting feature of the emitter is creep strength. This can be augmented by utilizing tungsten alloys which exhibit superior
strength characteristics. Of particular interest are tungsten alloys such as W-HfC, W-Re-HfC and perhaps W-ThO₂ which can be used to fabricate a reinforcement matrix prior to depositing CVD material. The presence of small amounts of alloying agents is believed to be sufficient to prevent slipping along grain boundaries of tungsten, which is presumed to be the main mechanism of creep deformation of emitters. Such alloys justify optimism for long lifetime even at temperatures of 2000 K or even higher.

The high temperature emitter can be coupled advantageously with oxygenated electrode surfaces to produce reasonable current output at high output voltage. An oxygenated tungsten surface can be expected to achieve an effective bare work function of about 5.2 eV, corresponding to a cesiated work function of about 3.1 eV. It is believed that the oxygen will be the controlling factor in determining the work function; the unexpected presence of hafnium or carbon at the surface can probably be tolerated to an extent. The use of oxygenated electrode surfaces requires that a means be found to regulate the activity of oxygen in the electrode space. Initially, it was thought that reduction of the cesiated collector work function might also be desirable as a means of boosting the output voltage. However, electron back-emission from the collector is described by the Richardson-Dushman equation as

\[ J^\text{back} = 120T^2e^{-\phi/kT} \text{ (A/cm}^2\text{) } , \]

where \( \phi \) is the work function, \( k \) is the Boltzmann constant = 8.617 \( \times 10^{-5} \) eV/K. For \( \phi = 1.5 \) eV and \( T = 880 \) K, the \( J^\text{back} = .238 \) A/cm\(^2\). For \( \phi = 1.3 \) eV, \( J^\text{back} = 3.3 \) A/cm\(^2\), which is higher than the forward emission. Thus there is not much point in seeking lower collector work functions unless the total current density is much higher (which, as pointed out earlier, is not practical in a single-cell TFE), or if the collector temperature is lower than 800 K (resulting in a heavy radiator).

Another benefit of oxygenation is that it permits a low cesium density in the interelectrode space and hence permits a wider electrode spacing. This is important because there is some chance that the emitter may bow slightly in response to thermal stress (of course, this should not happen with a perfect design, but not everyone is as good as the Soviets).

Oxygen regulation is best accomplished with an electrochemical approach in which the cell voltage controls the activity of oxygen.
Figure 3 represents the $J''$-$V$ characteristics with varying emitter work function and cesium pressure. Under the conditions described, it is predicted that the electrode output voltage will be about 1.2-1.3 volts before electrode/lead losses. Thermionic efficiency is projected to be about 10-12%.

Because of the 50-100 °C temperature variation expected along the length of the TFE due to the difference between coolant inlet and outlet temperatures, it is likely that present converter physics models will need to be extended to two or even three dimensions. Therefore, a semi-three-dimensional code is presently being developed for use to describe single-cell long diodes.

Additional manufacturing considerations must be taken into account for the long diode. For example, the multicell emitter is allowed to expand lengthwise, the emitter being fixed only at one end. Another means must be used to allow the single cell emitter to expand. Either some space must be left in the middle of the element, or some compliance must be designed into the lead area.

The use of advanced insulator materials is desirable to enhance thermal conduction from the collector to the coolant, radiation stability, temperature stability and physical robustness. Diamond films are an
excellent candidate technology, assuming that they can be shown to retain their excellent electrical resistivity characteristics under conditions of reactor operation. Diamond films would also be much thinner (about 10-50 microns versus the present 500 microns for alumina), partially compensating for the increased thickness of the thermionic electrodes required for single cell operation. In addition, the ability of diamond to act as a hydrogen barrier could be extremely important in avoiding corrosion problems in the interelectrode space, particularly since the presence of oxygen is already planned (note that the single cell design allows the entire TFE to be protected by a solid sheath, which is not generally the case for the multicell design). Applying a uniform diamond coating to a large area surface (~100 cm$^2$) represents a technical challenge and most diamond film efforts to date have concentrated on producing high quality films over only a few square centimeters at most. Accomodating the thermal expansion coefficient mismatch between the metallic collector and the diamond, which has a very low thermal expansion coefficient, represents another technical concern. Thinness of the diamond is one factor in its favor.

From a system standpoint, it is desirable to operate with as high a heat rejection temperature as practically achievable, in order to minimize radiator mass and area. Currently, the reactor temperature is thought to be limited by two factors: the use of stainless steel liquid metal containment, and the containment of hydrogen. Liquid metal fast breeder reactor technology provides an excellent data base on stainless steel operating at 920 K peak temperature and below. Thus a thermionic reactor with a reactor outlet of 920 K and a reactor inlet of some 820 to 870 K represents an upper bound on achievable temperatures. Similarly, the migration of hydrogen requires that a suitable coating be used to prevent leakage. Current state of the art technology based on the SNAP program is sufficient for peak temperatures of about 850 K and no higher. Extrapolation to 920 K based on improved containment technology is probably achievable in the near term. It is also possible that diamond films may be of value in increasing the temperature capability of the containment, but there are currently no ongoing programs to investigate this promising concept. Ultimately, the heat rejection temperature may be extended to 1100 K through the use of superalloy or even refractory metal concepts. However, the extreme difficulty and prohibitive cost associated with operating a high vacuum reactor test facility for refractory metal systems will preclude their use for many years.

The desire for reliability and redundancy leads to an increased
emphasis on modular concepts. This is probably the only way to ensure against threats such as debris collisions, for example, which can vaporize entire sections of spacecraft radiators. Thus it is desirable for heat removal to be accomplished with a modular system such as heat pipes or capillary pumped loops. These techniques offer the additional advantage of reduced temperature drop along the length of the TFE. Programs to adapt and develop modular cooling techniques specifically for zirconium hydride thermionic reactor cores are currently in an exploratory stage only. Thus, initial tests may be accomplished using a single liquid metal forced convection loop, although in the far term modular heat removal may be mandatory.

SUMMARY

Technical advances are required to extend the single cell TFE to the few tens of kilowatts regime. Of prime importance is realizing a high cell output voltage, which has resulted in the need for higher emitter temperatures, oxygenated electrode surfaces and material improvements such as the use of advanced insulator materials. These projected advances appear attainable, and research is underway to prove it.
In-Core Thermionic Space Reactor Design Integration

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I. Introduction

The design of any space reactor system encompasses the integration of a number of distinct, yet dependant subsystems. The major systems considered in a space nuclear power system (SNPS) include the reactor, or core, which produces heat, a nuclear shield, which protects any payload from the harmful nuclear radiations generated by the reactor/core, a power conversion system, which converts the thermal energy from the core into electricity, a heat removal system which transports the fission energy out of the core, and a heat rejection system which delivers the excess, or waste, heat to the ultimate heat sink—the deep recesses of space. This paper discusses some of the primary design variables and design constraints associated with the above subsystems, and also discusses the results of calculations concerning the nuclear design and safety evaluation of three reactor core systems.

The overall objectives in the design of a SNPS will vary depending upon the specific mission that the power system will be performing. For example, a SNPS powering a North Atlantic commercial radar system will place a high premium on long lifetime, reliability, and flexibility, while a SNPS for a Mars rover vehicle would require low mass, short lifetime and high reliability, or a system for an interplanetary probe may require highly reliable intermittent operation during a planetary fly-by. In general, however, the overall system design objectives can be placed into one of three categories: minimum mass, long lifetime, and high reliability.

In a design optimized to obtain the minimum mass, it is often not sufficient to simply minimize the mass of each system sub-component without regard to the mass of the entire system. Since all of the systems are inter-related, it is important to consider the effects of design changes in any of
the sub-systems on the overall mass of the system. Changes in one system may have large consequences in others. For example, reducing the outlet temperature of the coolant coming from the reactor will reduce the requirements on the materials in the reactor, but may also end up reducing the effective heat rejection temperature in the radiators and result in an increase in the required radiator area and the radiator mass. Also related to the overall system mass are the constraints posed by the launch vehicle. All launch vehicles have restrictions on the amount of payload mass and volume which can be raised into earth orbit.

Long lifetime of a SNPS is dependent upon two basic variables. The first is the provision of sufficient nuclear fuel to ensure that the specific mission can be completed, and the second is the ability of the system designer to protect the SNPS from the harsh environment of space. Providing enough fuel is a relatively simple task as nuclear analysis computer codes have reached a mature state of development and can be counted on to determine the amount of fissile material required. Protecting the SNPS from the elements of space requires detailed knowledge of the various threats to the system. Some of these are the naturally occurring micrometeoroids and an assortment of space debris ranging in mass from micrograms to many kilograms, cosmic radiation, atomic oxygen in low earth orbit, and control of the thermal environment of the SNPS.

A SNPS is required to be highly reliable as it is designed to provide power for the successful completion of the designated mission. Such a requirement becomes dependent upon the inherent safety, redundancy, and diversity placed in the design. This may also require simple control systems, artificial intelligence, and other features to ensure that the SNPS is able to
provide the necessary amounts of power for the intended lifetime of the system. The SNPS may also be designed to be repaired, or be able to recover from a mishap and continue to provide power. Such a scenario is difficult to envision for an unmanned earth orbiting satellite; however, on advanced missions to the lunar or martian surfaces it is entirely believable to consider the repair and recover of the operation of an SNPS.

II. Design Variables and Constraints

Within each of the above systems there exist a vast number of variables which can be adjusted and on which the SNPS can be optimized. Similarly there are many more constraints which effectively limit the range over which these variables can be used. Many of these variables can also be found to be dependent upon additional parameters; thus a great deal of juggling is often necessary before a final design is achieved.

In the design of the reactor core many choices are possible on all levels of the design. Table 1 lists some of the available parameters which affect the design. The primary choices, fuel type, enrichment, cladding type, and coolant type, set the major characteristics of the reactor and determine many of the other system choices. Once chosen, the available parameter space over which changes can be made can become rather limited. For instance, once a decision has been made to utilize a thermal reactor, cladding and coolant choices become restricted to those with small thermal neutron cross sections, and arrangements must be made to ensure the availability and the retention of sufficient hydrogen for moderation.

In the case of the core design, some of the significant design constraints can be seen in Table 2. Safety in the design of any nuclear system is of paramount importance during launch and operation.
Table 1. Reactor Core Design Variables

<table>
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<th>Category</th>
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<tr>
<td>FAST OR THERMAL ENRICHMENT</td>
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<td>FUEL TYPE</td>
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<td>CLADDING TYPE</td>
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<td>COOLANT TYPE</td>
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<td>CONTROL TECHNOLOGY</td>
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<td>PIN / CELL ARRANGEMENT</td>
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<td>PLATE DIMENSIONS</td>
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<td>PLATE CLADDING</td>
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<td>SOLID CORE ARRANGEMENT</td>
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<td>FISSION PRODUCT RETENTION</td>
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<tr>
<td>POWER LEVEL</td>
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<td>POWER DENSITY</td>
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<tr>
<td>POWER SHAPING AND DISTRIBUTION</td>
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<tr>
<td>FUEL TEMPERATURES</td>
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<tr>
<td>COOLANT TEMPERATURES - INLET AND OUTLET</td>
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<tr>
<td>COOLANT FLOW RATES</td>
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<tr>
<td>HEAT TRANSFER AND HEAT FLUXES</td>
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<td>FUEL BURNUP</td>
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</table>
Table 2. Reactor Core Design Constraints

<table>
<thead>
<tr>
<th>Category</th>
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<tr>
<td>SAFETY (LAUNCH AND OPERATIONAL)</td>
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<td>CRITICAL HEAT FLUXES</td>
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<tr>
<td>FUEL TEMPERATURE LIMITS</td>
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<td>FUEL CLADDING LIMITS</td>
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<tr>
<td>CRITICALITY</td>
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<td>LIFETIME</td>
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<tr>
<td>POWER DISTRIBUTION DURING BURNUP</td>
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<tr>
<td>STRUCTURAL INTEGRITY</td>
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<tr>
<td>MATERIALS COMPATIBILITY</td>
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</table>
Temperature limits on the materials used (fuel, cladding, and coolant—in the form of critical heat flux) also provide the primary constraints, and all are related to the probability that the reactor will be able to reach its design performance level.

The shield is required to attenuate the reactor radiation to meet any payload radiation exposure requirements. Available design variables include the choice of neutron and gamma shielding materials and their arrangement. Since neutrons are emitted with high energy from the core, it is necessary to choose shielding materials which will first thermalize, or reduce the neutron energy, and then absorb the neutrons. In the choice and arrangement of the gamma shielding materials it is important to consider both the primary (those emitted directly from the core) and the secondary (those caused by neutron absorptions in the shield) gammas. Computer calculations must be made to determine the appropriate thicknesses and arrangements to optimize the shield design. In many cases, the minimization of the shield mass is the primary design driver. The design must also meet the constraints posed by the need to remove any heat generated in the shield to avoid any materials temperature limits within the bounds of the compatibility of the shield materials.

The heat removal system is mainly responsible for insuring the integrity of the SNPS during power operation and during any operational transients. Since a reactor generates a considerable amount of heat once it is placed in operation, particular attention must be placed on the design of this system. Table 3 lists some of the important design variables and constraints for the heat removal system. Care must be taken in the design to provide sufficient redundancy and diversity in the heat removal system if possible to ensure that the SNPS can achieve its design goals. Design choices include the use of
Table 3. Heat Removal Design Variables and Constraints

VARIABLES
- REDUNDANCY AND DIVERSITY
- SINGLE PHASE VS. TWO PHASE
- PUMPED LOOP VS. HEAT PIPE OR COMBINATION
- INTEGRATION WITH REACTOR AND HEAT REJECTION SYSTEMS
- WORKING FLUID TEMPERATURES
- MATERIALS PROPERTIES
  - DENSITIES
  - STRENGTH
  - DUCTILITY

CONSTRAINTS
- PUMPING CAPACITY
- MATERIAL COMPATIBILITY
- COMPATIBILITY WITH HEAT REJECTION SYSTEM
- PROTECTION FROM ELEMENTS
  - MICROMETEOROIDS AND DEBRIS
  - COSMIC RADIATION
  - ATOMIC OXYGEN
  - THERMAL CONTROL
- ENTRAINED GAS REMOVAL
single- or two-phase flow, working fluid core inlet and outlet temperatures
all limited by the constraints of materials compatibility, environmental
protection, and available pumping capacity.

Similar choices are necessary in the heat rejection system. Any design
configuration must remain compatible (temperatures and materials) with all
other parts of the system. The choices of materials which can be protected
from the space environment, provide sufficient surface emissivity to enhance
the rejection of heat to space, and are compatible with the working fluid are
critical. Since higher radiator temperatures mean smaller required surface
areas, it may be important to use as high temperature materials as possible to
reduce radiator mass. Also, since the radiator provides the largest surface
area component in the SNPS, multiple heat rejection circuits are needed to
reduce the possibility of system failure due to punctures from micrometeoroids
and other space debris.

Lastly, the choices involved in the design of the power conversion
system require attention. In general, the primary choices and variables
include the choice of the power conversion technology, its overall efficiency,
its location, and the working fluid temperatures needed to make it possible.
In specific, the use of thermionic energy conversion allows the choice of
location either in the core or out of the core. Other authors at this
conference will discuss and fully describe many of the thermionic energy
conversion design variables and constraints. Some of these include emitter,
collector, and cesium reservoir temperatures, gap spacings, and output power
densities. The reader is referred to the numerous other papers for further
details.
III. In-core Thermionic Reactor Core Design Study

Nuclear feasibility and criticality safety assessments of a number of reactor concepts were determined based upon Monte Carlo three-dimensional calculations of the effective multiplication factor, $k_{\text{eff}}$, using the MCNP neutron transport code, version 3A (1). All calculations were performed on the OSU Nuclear Engineering Department’s Apollo Domain Series DN3000 engineering work station. First order criticality results were obtained for the proposed reactor concepts utilizing homogeneous, three-dimensional models of each reactor. Greater detail for this preliminary design study was not warranted as it would have greatly increased the required computational time. The cross-section set utilized was the ENDF/B-IV data set supplied by the Radiation Shielding Information Center, Oak Ridge National Laboratory, Oak Ridge, Tennessee with the MCNP code (2).

The cases considered each represent a specific scenario. These include startup and operational life capability, launch pad and ascent shutdown capability, water immersion criticality and safety, both for normal launch configuration with all shutdown subsystems in place and a post-impact launch abort configuration with all exterior control and shutdown systems removed, and a sand burial scenario for the postulated post-impact launch abort configuration. Further details of the configurations are contained in reference 3.

III.A. Maximum Reactivity

In this configuration, the maximum operating reactivity was determined in order to evaluate the initial criticality of each reactor concept. All control rods were fully withdrawn and all reflectors were positioned in such a way as to provide the maximum amount of neutron reflection. Any fixed poisons
were assumed to remain in the core and the objective was to estimate the maximum amount of excess reactivity available for normal startup.

The target values of $k_{\text{eff}}$ for these cases was required to fall between 1.05 and 1.09. These limits were chosen to allow for statistical variances in the calculational techniques, cross section inaccuracies and temperature effects on startup, and to ensure sufficient reactivity margins to provide for reactor operation for a seven year period due to burnup. In all cases examined, the statistical variance of the results was found to be less than 2% which was accomplished using a combination of variance reduction techniques and running sufficient histories.

III.B. Launch Configuration

In the launch configuration, all movable poisons were placed in such a manner that a subcritical assembly was maintained prior to and during launch. Control rods were fully inserted into the core and any movable reflectors used for control were removed and stored in their launch positions. These cases were designed to test the amount of shutdown margin available to the reactor during the fabrication of the concept and the safety of the concept after it is loaded into the launch vehicle. They also give some measure of the capability to shutdown the reactor system after initial criticality in space should a problem develop. Ideally, the values for $k_{\text{eff}}$ for these cases should be as low as possible; however, a value of less than 0.9 was more than acceptable from an initial feasibility standpoint.
III.C. Water Immersion

For water immersion cases, an accident was simulated in which the reactor system was dropped into water. Such an accident could occur during a launch which failed to place the reactor into orbit or during transportation of a completed reactor system to the launch site. In these cases it was assumed that the launch configuration described above was maintained, no water was allowed to enter the reactor system, and the entire reactor system was placed at the center of a 5 meter radius sphere of water where the water acted as an additional reflector and external neutron moderator. No neutron moderation, other than from designed core materials, was included within the reactor system. Also, it was assumed that no physical damage to the reactor core occurred and that there was no redistribution of core or reflector materials (i.e., no compaction). For water immersion accident scenarios, an acceptable upper limit for $k_{\text{eff}}$ was chosen to be 0.95.

III.D. Water Flooding

The water flooding cases modelled the water immersion accident with no allowance made for active shutdown systems external to the core. All movable components exterior to the core were assumed to have been removed on impact, including movable reflectors. It was further assumed that the core itself and fixed reflector sections would remain intact on impact. Water was allowed to fill any and all of the voids within the reactor system, including coolant flow channels inside the core, and the void spaces in the thermionics and heat pipes. It was assumed that all coolant volume fractions in concepts which utilize liquid coolant were replaced with water and that any core heat pipes were filled with water. The resulting configuration was then submerged at the center of a 5 meter radius sphere of water as in the water immersion cases.
No allowances for the compaction of the reactor core and reflectors were made in this scenario. Acceptable levels of subcriticality could be assumed for such cases if $k_{\text{eff}}$ was found to be less than 0.95. Again, this included a margin to allow for statistical and data accuracy, but did not leave very much margin in the cases where compaction of the core was possible.

III.E. Sand Burial

Sand burial cases simulated the flooded reactor buried in water-saturated sand. All movable components exterior to the core were again assumed to be removed by impact, and water filled all voids, coolant channels, etc. No sand was assumed to seep into the reactor and no compaction was assumed to occur. The porosity of the sand was assumed to be 50% -- i.e., 50% by volume sand, and 50% by volume water. Additionally, in an attempt to more fully approach actual conditions, two types of sands were modeled: 100% silicon dioxide and 100% calcium carbonate, as some Florida beaches are made up almost exclusively of crushed coral. Acceptable levels of subcriticality could again be assumed if $k_{\text{eff}}$ was found to be less than 0.95.

IV. In-core Thermionic Reactor Systems Analysis

Three small, low-power in-core thermionic reactor designs are discussed here. The following assumptions were made:

a) All reactor configurations were without any reactivity control, approximating a cold clean startup for the determination of the maximum amount of positive reactivity.

b) The overall core height/diameter ratio was assumed to be maintained at 1.

c) Cylindrical geometry.

d) 7 cm reflector thickness on top, bottom and sides.
e) Each reactor region was treated as a homogeneous mixture of the appropriate materials.

f) 95% enriched uranium was used in the fuel.

g) Reactivity control was accomplished by louvered radial reflectors. To model the shutdown scenarios, it was assumed that the radial reflector density was 10% of the value used in the maximum reactivity cases.

IV.A. Solid Core Reactor Configuration

This design used a solid core and heat removal was solely through conduction. The nominal power for this reactor system was 1-10 kW. The fuel region was uranium dioxide (60 volume percent) in a tungsten matrix (40 volume percent), with a uranium enrichment of 95 percent. The core was surrounded by a 7 cm thick reflector of BeO. The thermionics were assumed to be 40 volume percent tungsten, 40 volume percent niobium and 20 volume percent void. Waste heat removal was accomplished by heat pipes using potassium as the working fluid. They were modeled as 20 volume percent potassium, 20 volume percent tungsten and 60 volume percent void. An additional 23 cm of BeO was placed on top of the reactor as a shield. Reactivity control was accomplished by a louvered radial reflector. Positive shutdown in the launch configuration was by use of a boron carbide control rod (with fuel follower) through the center of the core. Figure 1 schematically shows the core material arrangement for the solid core reactor configuration.

The initial feasibility results for the solid core reactor were quite encouraging as seen in Table 4. A minimum critical radius of 13.9 cm was achieved and the analysis of the additional configurations yield reasonable
Figure 1. Solid Core (UO₂) with External Reflector.
Table 4. Results: Solid Core (UO$_2$) with External Reflector

MAXIMUM REACTIVITY

<table>
<thead>
<tr>
<th>Value</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.06</td>
<td>R = 13.9 CM</td>
</tr>
<tr>
<td>1.03</td>
<td>R = 13.4 CM</td>
</tr>
</tbody>
</table>

LAUNCH CONFIGURATION

<table>
<thead>
<tr>
<th>Value</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.842</td>
<td>1.5 CM RADIUS SHUTDOWN ROD + THIN REFLECTOR</td>
</tr>
</tbody>
</table>

WATER IMMERSION

<table>
<thead>
<tr>
<th>Value</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.905</td>
<td>1.5 CM RADIUS SHUTDOWN ROD + THIN REFLECTOR</td>
</tr>
</tbody>
</table>

WATER FLOODING

<table>
<thead>
<tr>
<th>Value</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.926</td>
<td>1.5 CM RADIUS SHUTDOWN ROD, NO REFLECTOR</td>
</tr>
</tbody>
</table>

SAND BURIAL

<table>
<thead>
<tr>
<th>Value</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.935</td>
<td>1.5 CM RADIUS SHUTDOWN ROD, NO REFLECTOR, SiO$_2$</td>
</tr>
<tr>
<td>0.932</td>
<td>1.5 CM RADIUS SHUTDOWN ROD, NO REFLECTOR, CaCO$_3$</td>
</tr>
</tbody>
</table>
results. However, such a reactor configuration is probably unreasonable since uranium oxide has very low thermally conductivity and thus will reach very high temperatures without actively cooling the core interior.

IV.B. Uranium Nitride Core (SP-100)

In general, the geometry of this case was identical to that of case A above. The difference was in the composition of the fuel region: the core consisted of uranium nitride fuel pins (with a rhenium liner and niobium-1 percent zirconium clad) in a hexagonal array. These fuel pins were identical to those planned for the SP-100 GES program, however, due to the unavailability of rhenium cross sections, tungsten was substituted for this analysis. The pitch-to-diameter ratio was 1.1 and the interstitial areas were filled with static liquid lithium. The lithium was used to conduct the heat generated in the fuel rods to the outside of the reactor vessel where it was converted to electricity. Heat pipes were again used to reject the waste heat to space. The fuel was 95% enriched and the nominal power for this reactor was 10-30 kWe. The dimensions of the fuel pins used were:

- fuel pellet diameter 1.021 cm
- liner thickness 0.013 cm
- gap 0.006 cm
- clad thickness 0.038 cm
- overall fuel pin diameter 1.135 cm

The results for the uranium nitride core are shown in Table 5. The system easily meets the first three objectives (maximum reactivity, launch configuration, water immersion); however, it had trouble meeting the water flooding criteria. This was a consequence of replacing the static lithium bath with water. Also the concept has significant excess reactivity in the
Table 5. Uranium Nitride Core (SP-100) with External Reflector

MAXIMUM REACTIVITY

<table>
<thead>
<tr>
<th>Reactivity</th>
<th>Configuration</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.975</td>
<td>R = 9.35 CM</td>
</tr>
<tr>
<td>1.050</td>
<td>R = 10.35 CM</td>
</tr>
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</table>

LAUNCH CONFIGURATION

<table>
<thead>
<tr>
<th>Reactivity</th>
<th>Configuration</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.809</td>
<td>1 PIN/QUARTER SHUTDOWN ROD + THIN REFLECTOR</td>
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</tbody>
</table>

WATER IMMERSION

<table>
<thead>
<tr>
<th>Reactivity</th>
<th>Configuration</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.878</td>
<td>1 PIN/QUARTER SHUTDOWN ROD + THIN REFLECTOR</td>
</tr>
</tbody>
</table>

WATER FLOODING

<table>
<thead>
<tr>
<th>Reactivity</th>
<th>Configuration</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.994</td>
<td>1 PIN/QUARTER SHUTDOWN ROD, NO REFLECTOR</td>
</tr>
</tbody>
</table>

SAND BURIAL

<table>
<thead>
<tr>
<th>Reactivity</th>
<th>Configuration</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.027</td>
<td>1 PIN/QUARTER SHUTDOWN ROD, NO REFLECTOR, SiO₂</td>
</tr>
<tr>
<td>1.007</td>
<td>1 PIN/QUARTER SHUTDOWN ROD, NO REFLECTOR, CaCO₃</td>
</tr>
</tbody>
</table>
sand burial cases, even exceeding the water flooding cases. This problem was
due to the tungsten in the core which has the effect of shifting the neutron
spectrum and increasing the positive reactivity.

IV.C. Uranium Nitride Core (SP-100) with Large Pin

This case is identical to case B except that the fuel pellet diameter
was increased by 20% to 1.2252 cm, giving an overall fuel pin diameter of
1.3392 cm. Nominal power was 10-40 kWe.

Table 6 contains the criticality results for the large pin SP-100
system. As with the smaller fuel pins, the large fuel pin system easily
satisfied the maximum reactivity, launch configuration and water immersion
objectives. It exceeded the desired water flooding reactivity, however, again
as a result of water replacing lithium in the fuel region. Also, it is seen
that the sand burial cases appear to be less of a problem for the larger fuel
pin design than for the nominal SP-100 design.
Table 6. Results: Uranium Nitride Core (SP-100, with large pin) with External Reflector

<table>
<thead>
<tr>
<th>Condition</th>
<th>Reactivity</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>MAXIMUM REACTIVITY</td>
<td>1.050</td>
<td>R = 10.4 CM</td>
</tr>
<tr>
<td>LAUNCH CONFIGURATION</td>
<td>0.835</td>
<td>1 PIN/QUARTER SHUTDOWN ROD + THIN REFLECTOR</td>
</tr>
<tr>
<td>WATER IMMERSION</td>
<td>0.887</td>
<td>1 PIN/QUARTER SHUTDOWN ROD + THIN REFLECTOR</td>
</tr>
<tr>
<td>WATER FLOODING</td>
<td>1.000</td>
<td>1 PIN/QUARTER SHUTDOWN ROD, NO REFLECTOR</td>
</tr>
<tr>
<td>SAND BURIAL</td>
<td>1.027</td>
<td>1 PIN/QUARTER SHUTDOWN ROD, NO REFLECTOR, SiO₂</td>
</tr>
<tr>
<td></td>
<td>1.012</td>
<td>1 PIN/QUARTER SHUTDOWN ROD, NO REFLECTOR, CaCO₃</td>
</tr>
</tbody>
</table>

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V. Acknowledgement

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References


STATUS REPORT ON THERMIONIC ENERGY CONVERSION RESEARCH IN JAPAN

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*Umezono, Tsukuba-city, Ibaraki 305 JAPAN

ABSTRACT

Electrotechnical Laboratory (ETL) has conducted a research on a lanthanum hexa-boride (LaB6) thermionic converter. The converter is a plane-parallel type, both emitter and collector electrodes are made of polycrystalline LaB6 disks, active surface area is 0.64 cm², inter-electrode spacing is 1.1 mm. LaB6 collector workfunction ~c in cesium (Cs) vapor was measured from the reverse saturation current Jrev in un-ignited mode V-I curves. Although ~c depends on the emitter and/or collector temperature, the minimum ~c is 1.35V when Tc=720K, Tc/TR=1.53, here Tc and Tn are collector and Cs reservoir temperature. Because the value of ~c minimum =1.35V is far less than the value of refractory metal collector materials, the LaB6 thermionic converter has possibility to greatly improve power generation efficiency.

Mitsui Engineering & Shipbuilding Co., LTD (MESCO) has been conducting a combustion heated thermionic converter R & D. A tori-spherical CVD converter has been developed and power generation tested. The converter is made of a hot shell-emitter structure which consists of silicon carbide-graphite-tungsten three layer structure. The diameter of the hot shell is 60 mm, the length 90 mm. The collector is made of nickel and air cooled. The active surface area of the converter is 20 cm². The converter is installed in a kerosene-fired furnace and heated up to 1600°C for power generation test. The maximum performance of the converter at emitter temperature 1474°C was; output power 82.5W, output voltage 0.38V, output current 217A. The converter successfully demonstrated to be able to endure temperatures up to 1600°C in combustion gas atmospheres.

Research Institute for Scientific Measurements, Tohoku University (RISM) has been studying thermionic emission materials of more than 30 kinds, such as metals, carbides, oxides, and borides. Hafnium carbide (workfunction=3.31V), zirconium carbide (workfunction=3.56V) are considered to be promising emitter electrode materials from the point of view of materials evaporation loss.

1. Research on LaB6 thermionic converter(1), (2)

LaB6 has a workfunction value of 2.7~2.8V without cesium (Cs) adsorption which is suited for an emitter electrode of a thermionic converter. Therefore, Cs pressure could be determined mainly to minimize collector workfunction (~c) and also to reduce interelectrode plasma loss. It would be possible that a LaB6 thermionic converter could establish higher efficiency than refractory metal electrodes converters. This paper reports on the experimental results of output current characteristics of a LaB6 thermionic converter developed in ETL, and the LaB6 emitter workfunction (~e) and collector workfunction (~c) measurements in un-ignited mode volt-ampere curves (V-I curves).

1.1 Method of workfunction measurement(3)

Thermionic electron emission saturation current density Jₑ from a uniform surface of an electrode is given by Richardson equation, (eq.1);

$$J_e = A T^2 \exp\left(-\frac{e \phi}{k T}\right) = 120 T^2 \exp\left(-\frac{e \phi}{k T}\right)$$

Here Jₑ: electron saturation current density (A/cm²), A: Richardson constant 120 (A/(cm²·K²)), k: Boltzmann constant 1.380622·10⁻²³ (joule/K), T: temperature (K), e: electrical charge of electron 1.6021917·10⁻¹⁹ (coulomb), ~φ: workfunction of an electrode (V).

The ion saturation current density Jᵢ is given by the relation;

$$I_i = e P_g \cdot 10^{-4} / \left[ (2 \pi M_g k T_g)^{1/2} \cdot \left(1+2 \exp\left(V_i - \phi/k T\right)\right) \right]$$

Here Iᵢ: ion saturation current density from an electrode (A/cm²), P_g: Cs pressure (N/m²), M_g: Cs mass 2.206935·10⁻²⁵ (Kg), Tᵢ: Cs gas temperature (K), Tₑ: emitter temperature, Vi: Cs ionization voltage 3.89V. The ion-richness ratio β is defined by the relation;

Thermionic Energy Conversion
\[ \beta = \frac{n_i}{n_e} = \left( \frac{m_i}{m_e} \right)^{1/2} \cdot \frac{J_s}{J_s} = 492.2 \cdot \frac{J_s}{J_s} \] -----(3)

Here \( n_i \): density of ions (particles/m\(^3\))
\( n_e \): density of electrons (particles/m\(^3\))
\( m_i \): the mass of Cs ion, \( m_i = m_g \)

The workfunction \( \phi \) is expressed from eq. (1);

\[ \phi = \left( \frac{kT}{e} \right) \ln \left( \frac{A T^2}{J_s} \right) \] -----(4)

Because Cs gas of the pressure \( 1.33 \times 10^5 \sim 1.33 \times 10^3 \) Pa is filled in a thermionic converter, we must pay close attention to some points when \( \phi \) is measured by eq. (4).

(1) effect of sheath

The workfunction \( \phi \) should be measured on condition of \( \beta \geq 1 \), otherwise electron sheath is formed near the surface of the electrode and the measured apparent workfunction might take higher value of the order of electron sheath voltage.

(2) effect of electron scattering

In order to avoid the decrease of electron current flowing from an emitter to a collector by scattering collision between electrons and neutral Cs atoms, the condition of eq. (5) must be satisfied;

\[ \lambda \cdot e^{-a} > d \] -----(5)

Here \( \lambda \cdot e^{-a} \): mean free path for electron-neutral Cs atom elastic collision.

The mean free path \( \lambda \cdot e^{-a} \) is described by eq. (6), using the cross-section for electron-neutral Cs atom elastic collision \( e^{-a} \).

\[ \lambda \cdot e^{-a} = 1 / (n \cdot \lambda \cdot e^{-a}) \] -----(6)

Here \( \lambda \cdot e^{-a} \): mean free path for electron-Cs atom elastic collision (cm), \( n \): Cs gas density of the thermionic converter (particles/cm\(^3\)), \( e^{-a} \): scattering cross-section for electron-neutral Cs atom elastic collision (cm\(^2\)).

\( e^{-a} \) values are given (4);

\[ e^{-a} = 300 \times 10^{-16} \text{(cm}^2\) in case of \( T_e = 1000 \sim 2000 \text{K}\)
\[ e^{-a} = 400 \times 10^{-16} \text{(cm}^2\) in case of \( T_e = 2000 \sim 4000 \text{K}\) \] -----(7)

Here \( T_e \): electron temperature.

The Cs gas density \( n \) is given by eq. (8) from ideal gas equation of state;

\[ n = \frac{P_g}{(RT)} \text{(mol/m}^3\)}
\[ = \frac{P_g}{N_A \cdot 10^{-5} / (RT)} \text{(particles/cm}^3\)}
\[ = 7.243 \times 10^{16} \cdot \frac{P_g}{T} \text{(particles/cm}^3\) \] -----(8)

Here \( R \): gas constant for ideal gas equation of state \( R = 8.31434 \text{(joule/(mol\cdot K))}\)
\( T \): Cs gas temperature \( T \geq T_e \), \( T_e \): Cs reservoir temperature, \( N_A \): Avogadro's number \( N_A = 6.02252 \times 10^{23} \text{(particles/mol)}\)

Taking \( \sigma \cdot e^{-a} = 400 \times 10^{-16} \text{(cm}^2\) from eq. (7) and substituting \( \sigma \cdot e^{-a} \) and eq. (8) to eq. (6), the mean free path \( \lambda \cdot e^{-a} \) is given by eq. (9);

\[ \lambda \cdot e^{-a} = 3.451 \times 10^{-4} \cdot \frac{T}{P_g} \text{(cm)} \] -----(9)

The experimental conditions should be chosen so that the value of \( \lambda \cdot e^{-a} \) given by eq. (9) satisfies the condition of eq. (5).

The method of determining workfunction from the output current characteristics is as follows.

As shown in Fig. 1, output current characteristics in un-ignited mode operation are divided into three regions, forward saturation current \( J_{for} \), reverse saturation current \( J_{rev} \), and the Boltzmann line region in between \( J_{for} \) and \( J_{rev} \).

![Fig. 1 Schematic of output current characteristics of a thermionic converter](image)

In Fig. 1, the positive direction of abscissa means that the collector electric potential is negative with respect to the emitter, i.e., retarding potential against electrons flowing from the emitter to the collector. If the conditions of \( \beta \geq 1 \) and \( \lambda \cdot e^{-a} > d \) are satisfied and the emitter ion saturation current density \( I_{es} \) is negligibly smaller than the collector electron saturation current density \( I_{cs} \),

\[ J_{for} \sim J_{es} = J_k \text{ (J_k: emitter electron saturation current density)} \] -----(10)

\[ J_{rev} = I_{es} + J_{cs} \sim J_{cs} \text{ (J_{cs}: collector electron} \)
saturation current density) \[ \text{--------(11)} \]

Emitter and Collector workfunctions can be obtained by substituting \( J_{ES} \) and \( J_{CS} \) respectively to eq.(4).

1.2 Experimental converter and method of experiment

The experimental converter is shown in Fig. 2.

![Schematic diagram of LaB₆ thermionic converter](image)

**Fig.2 (a) Schematic diagram of LaB₆ thermionic converter**

![Photo of LaB₆ converter](image)

**Fig.2 (b) Photo of LaB₆ converter**

The converter is plane parallel type, the emitter is a poly-crystalline LaB₆ disk 14.8mm diameter, 2mm thickness. The LaB₆ disk was brazed to the tantalum emitter base, the brazing material: Ti 95w%, Ni 5w% fine powder, brazing temperature: 1650°C, brazing time: 30min. under a certain pressure.

The collector is also made of a poly-crystalline LaB₆ disk 9mm diameter, 7mm thickness. The interelectrode spacing \( d \) is 1.1mm at room temperature. The converter is evacuated by 8 lit./sec ion pump and titanium sublimation pump. The converter is mounted inside a stainless-steel bell jar of which outer diameter is 250mm.

The output current characteristics were measured by applying 50Hz AC small voltage to the converter, and sampling output voltage and current signals for a period of one cycle (20msec) and recording them by digital memory. The emitter was heated by electron bombardment up to 1800K.

1.3 Emitter workfunction

In Fig. 3, an example of the output current characteristics in un-ignited mode operation of the LaB₆ thermionic converter is shown. \( J_{for} \) and \( J_{rev} \) are distinctly shown in Fig. 3.

![Output current characteristics in un-ignited mode of LaB₆ converter](image)

**Fig.3 Output current characteristics in un-ignited mode of LaB₆ converter**

Emitter workfunction \( \phi \) was calculated by eq.(4), supposing that forward saturation current \( J_{for} \) in un-ignited mode output current characteristics equals to emitter electron saturation current density \( J_{ES} \). In calculation, the data which satisfy conditions of \( \beta \geq 1 \) and \( d/\lambda \epsilon \alpha < 1 \) were chosen and plotted. Truly, because of the lack of data which strictly satisfy these two conditions, the conditions were relaxed to \( \beta > 0.2 \) and \( d/\lambda \epsilon \alpha < 10 \) and data were plotted.

First before Cs is distilled into the converter, electrodes were sputter-cleared by argon ions to remove surface contamination. The sputtering conditions were argon pressure 1.33*10⁻³Pa, applied AC voltage 175V, sputtering current 1mA, and sputtering time 2.5hours. After the sputtering, \( \phi \) in vaccum was 3.4V. Although this value was still higher than \( \phi \) of 2.7~2.8V of atomically clean surface of LaB₆, Cs was distilled at this time. The results of the measurement of emitter workfunction \( \phi \) in Cs vapor is shown in Fig. 4.

\( \phi \) shows a tendency of slow and linear change with respect to \( Te/T \) and corresponds well to presumption by Rasor & Warner theory on workfunction of Cs adsorbed metal surfaces(3).
The workfunction of the LaB$_6$ collector was derived from the measurement of the reverse saturation current density $J_{rev}$ in un-ignited mode output current characteristics, assuming that $J_{rev}$ equals electron saturation current density from the collector $J_{cs}$. In calculation, it was confirmed that the emitter ion saturation current density $I_{ES}$ was negligibly smaller than $J_{cs}$. The results were shown in Fig.5.

In Fig.5 in the case of $Te > 1400K$, $\phi_c$ shows a tendency of linear and upward change as to $Tc/TR$. On the other hand, in the case of $Te < 1352K$, $\phi_c$ takes a minimum value of 1.35V at $Tc/Ta=1.53$, and the $\phi_c$ curve shifts downward about 0.24V from the curve of $Te > 1400K$.

So, the results of the experiment showed that the collector workfunction $\phi_c$ differs depending on collector temperature. In the experiments, $Tc=850K$ in the case of $Te=1400-1420K$, and $Tc=720K$ in the case of $Te=1337-1352K$. The minimum value of $\phi_c$ in the experiment was 1.35V. The reason why $\phi_c$ depends on $Tc$ is not clear.

1.5 Output current characteristics in ignited mode

The output current characteristics in ignited mode operation are shown in Fig.6. In Fig.6, the maximum power was obtained at output voltage 0.5V, output current density 0.79A/cm$^2$, output power density 0.39W/cm$^2$, $Te=1746K$, $Tc=1018K$, $Tc=454K$, $d=1.1$mm nominal. The barrier index $V_b$ was $V_a=2.5V$, and was not better than refractory metal converters. The reasons are (1) collector temperature was too high to use lower $\phi_c$ because of insufficient size of collector cooling fin, (2) interelectrode spacing was not optimized.

It is the subject for a future study to demonstrate low workfunction of LaB$_6$ collector in ignited mode operation. The LaB$_6$ emitter allows its operation with more than an order of magnitude lower Cs pressure than refractory metals electrodes. So, the LaB$_6$ converter with auxiliary emitter, so called plasmatron...
type triode, filled with both Cs and inert gas, is expected to improve efficiency. In the plasmatron, the Cs pressure is mainly controlled to minimize collector workfunction, and positive ions are generated by electron impact to inert gas in an ignited auxiliary discharge between LaB₆ emitter and auxiliary electrode. The direct ionization process is considered to be highly efficient to produce ions for neutralization of negative space charge in the converter than spontaneous ion generation process in the elementary ignited diode, because of no appreciable increase of average electron temperature.

2. Combustion heated thermionic converter development

Mitsui Engineering & Shipbuilding Co., Ltd (MESCO) is conducting research and development on thermionic energy conversion to utilize unused high temperature heat energy such as a boiler, a gas-turbine, an industry furnace etc. Test fabrication of combustion heated thermionic converters and their power generation test results are reported in this paper.

2.1 Thermionic converter

A photograph of a thermionic converter used in the power generation test is shown in Fig. 7 and the specification is shown in Table 1.

### Fig. 7 Photograph of a combustion heated thermionic converter

#### Table 1. Specification

<table>
<thead>
<tr>
<th>Hot shell</th>
<th>Torispherical, Dia. = 60mm, Length=90mm, outer surface is SiC coated as high temperature oxidation protection layer</th>
</tr>
</thead>
<tbody>
<tr>
<td>Collector</td>
<td>High purity Ni</td>
</tr>
<tr>
<td>Spacing</td>
<td>0.9mm at room temperature, 0.3-0.4mm at operation</td>
</tr>
</tbody>
</table>

The hot shell has a composite structure, i.e., silicon carbide (SiC) is coated on the outer surface of the graphite as a barrier to combustion gas atmosphere, and tungsten (W) is coated on the inner surface as an emitter electrode. A collector is made of nickel (Ni), ceramic seal made of high purity alumina.

2.2 Power generation test

A photograph of a furnace for power generation tests is shown in Fig. 8, and the formation of the furnace is shown in Fig. 9.

### Fig. 8 Photograph of a power generation test furnace

The converter is mounted on the furnace with a coupling structure and heated by kerosine combustion gas. The collector electrode and the coupling structure are cooled by dry and clean air. The surface temperature of the converter is measured by optical pyrometer through an observation window on the opposite side of the furnace wall. The collector temperature Tc and the Cs reservoir temperature Ts are measured by chromel-alumel thermocouples. The power generation characteristics (the volt-ampere curves) of the converter are measured by AC voltage sweeping method which simulates an external electrical load change.

2.3 Result of power generation test

Power generation characteristics of the thermionic converter depends largely on emitter temperature Te, collector temperature Tc, Cs reservoir temperature Ta, and interelectrode spacing d. In the tests, power generation characteristics were measured under the various combination of Ts (truly, surface temperature Ts
of the converter, $T_c$, and $T_k$. The conditions are as follows:

$$T_s = 1300 \sim 1480^\circ C$$ (supposing emissivity = 1),
$$T_c = 400 \sim 650^\circ C$$, $T_k = 240 \sim 290^\circ C$.

The output current characteristics and output electrical power are shown in Fig. 10 and Fig. 11 when $T_c$ and $T_k$ are held constant and $T_s$ is changed.

Similarly, the V-I curves when $T_s$, $T_k$ are held constant and $T_c$ is changed is shown in Fig. 12. And the V-I curves when $T_s$, $T_c$ are held constant and $T_k$ is changed is shown in Fig. 13.

These test results have demonstrated that the test converters have enough endurance to high temperature, oxidation atmosphere of around 1600°C combustion gas. The output power of the converter increases as $T_s$, $T_c$ increase, and there exists optimum $T_k$ reservoir temperature $T_s$ for each electrode temperature. In the tests, the maximum output power at the terminal of the converter is 82.5W, output voltage 0.38V, output current 217A, output power density about 4W, at $T_k = 1474^\circ C$, optimum $T_k$ of about 270°C ($P_{cs} = 1\text{torr}$).
3. Emission materials research

Research Institute for Scientific Measurements, Tohoku University (RISM) has been studying thermionic emission materials of more than 30 kinds, such as metals, carbides, oxides, and borides. Hafnium carbide ($\phi = 3.31$V), zirconium carbide ($\phi = 3.56$V) are considered to be promising emitter electrode materials from the point of view of materials evaporation loss. Besides, carbides and borides of rare earth atoms are expected to have workfunction values of about 3.0V at low evaporation loss in vacuum, and systematic researches on these materials will be needed.

References:
(2) N.Kwai, et al; "Research on lanthanum borides" Researches report of national institute for reseach in inorganic materials No.17 (1978)
(4) J.M.Houston; "Cross section values to use in analyzing the cesium thermionic converter" Thermionic conversion specialist conf. (Oct. 1964)
RADIATIVELY HEATED THERMIONIC CONVERTERS
FOR NUCLEAR SPACE POWER SYSTEMS

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Loral Electro-Optical Systems (LEOS)
Pasadena, California

ABSTRACT

This paper presents results of a two-year effort of a five-year program to develop a radiatively heated thermionic converter capable of being coupled to a Romashka type nuclear reactor. Converter design parameters as influenced by the nuclear reactor system consideration will be delineated. Program successes and problems encountered will be described.

INTRODUCTION

This development program was initiated in response to the need for spaceborne power systems in the 5 to 40 kW range. Numerous studies indicate that the reactor thermionic power system is a system of choice when compactness, lightweight, reliability, and survivability are of the primary consideration.

One of the promising system concepts is based on a Romashka type reactor coupled to a thermionic power conversion system. For this concept (depicted in figure 1) the nuclear heat is conducted from the uranium dicarbide core to the graphite trays which are radiatively coupled to a large number of thermionic converters (via the emitter shoe structure), connected in series and parallel combination, providing redundancy and acceptable dc voltage for efficient and further power conditioning. Within the thermionic converter, heat is converted into electricity, and the waste heat is removed from the collector backside by a high temperature heat pipe to a radiator.

High Carnot and system efficiencies are obtained because the thermionic emitter typically operates between 1800°K to 2000°K while the collector/radiator operates between 900°K to 1000°K. The high temperature radiator, when compared to other heat-engine or photovoltaic systems, offers a lightweight and compact system. The significantly low observable cross section and high degree of maneuverability are important factors in survivability against the anti-satellite attack or other forms of threats.

In terms of the reactor thermionic systems, the Romashka concept offers one important advantage: the elimination of the liquid metal coolant loop which is replete with problems associated with the electromagnetic pump, material compatibility, corrosion, and the extraneous power consumption. Another important advantage is the avoidance of the converter interelectrode distortion caused by the nuclear fuel swelling, which normally occurs in the thermionic in-pile concept.

Other advantageous factors include:

- Ready adaptability of planar thermionic diode whose technology is more mature than the other type.
- Less stringent requirement on thermal insulator than the in-pile design.

Work sponsored by Aero Propulsion Laboratory, Air Force Wright Aeronautical Laboratories, Aeronautical System Division, United States Air Force, Wright-Patterson, Ohio 45433-6563.
Figure 1. Thermionic Romashka Nuclear Power System
Out-of-core testing ease with electrical heat source that closely simulates the real reactor conditions of radiative couplings, save the nuclear environments.

This paper presents results of a two-year effort of conceptual design, tradeoff studies, device design, and fabrication and testing of the Preliminary Converters which have a goal of achieving a device efficiency of at least 10%. The remaining tasks are concerned firstly with the improvement of converter performance with the advanced converter materials being investigated by Loral and other laboratories and secondly with the integration and operational aspect of a number of converters (a minimum of four) connected in series.

### SYSTEM TRADEOFF

In 1967, Rasor et al. proposed a Thermionic Romashka concept, having as a baseline system capable of 5 kW electrical power output. Twenty years later G A Technologies proposed a similar concept (STAR-C) with a 6 kW(e) baseline. For this study LEOS considers a system of 9.6 kW(e) size. Results of our tradeoff study are summarized in Table 1, in comparison with the previous studies, which include the pertinent system parameters such as the dimensions and temperatures of the reactor core, along with the converter temperatures. Table 2 summarizes the reactor thermal parameters used in the studies.

The GA and LEOS studies indicate the trend toward operating the emitter at a moderate temperature (between 1800° and 1900°K) while extracting greater power density from the reactor to generate more electrical power. The rationale for the moderate emitter temperature is that the reactor core radiating area dictates the size of the emitter shoe and hence a large active emitter area.

### TABLE 1

| Conceptual Romashka Thermionic Power System (UC₂-Graphite Reactor) |
|------------------|-----------------|-----------------|
| Reactor Power (kW) | 35              | 44.4           | 69.2           |
| Electrical Output (kW) | 5              | 6              | 9.6            |
| System Efficiency (%) | 14.2           | 13.5           | 13.9           |
| Core CL Temperature (°K) | 2173           | 2293           | 2270           |
| Core Surface Temperature (°K) | 2240(1)       | 2011(1)        | 1995(1)        |
| Emitter Temperature(°K) | 1973           | 1963           | 1850           |
| Collector Temperature (°K) | 1000           | 1016           | 990            |
| Carnot Efficiency (%) | 49.3           | 45.5           | 46.5           |
| Core Dimensions   |                 |                |                |
| o Diameter (cm)   | 27.9            | 25             | 25.5           |
| o Length (cm)     | 40.0            | 32             | 50             |

**NOTES:**
(1) Assume net emissivity exchange 0.8 (e₁ = e₂ = 0.9)

### TABLE 2

<table>
<thead>
<tr>
<th>Reactor Thermal Parameters</th>
</tr>
</thead>
<tbody>
<tr>
<td>Core Diameter (cm)</td>
</tr>
<tr>
<td>Core Length (cm)</td>
</tr>
<tr>
<td>Thermal Power (kW)</td>
</tr>
<tr>
<td>Power Density (kW/houre)</td>
</tr>
<tr>
<td>Radiant Flux (W/cm²)</td>
</tr>
</tbody>
</table>

By keeping the overall system temperature moderate, problems of material evaporation, structural integrity, and performance degradation of the reactor and power conversion system will be alleviated. At temperature between 2100°K to 2300°K the evaporation of UC₂ and graphite (in comparison with tungsten) will be as indicated in Table 3.

### TABLE 3

<table>
<thead>
<tr>
<th>Evaporation of Reactor Materials (mm/10 years)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Materials</td>
</tr>
<tr>
<td>UC₂</td>
</tr>
<tr>
<td>Graphite</td>
</tr>
<tr>
<td>Tungsten</td>
</tr>
</tbody>
</table>
Clearly the UC2 fuel disc and the graphite fuel tray should be encased in a low vapor pressure coating. During the study phase LEOS investigated several refractory carbides for desirable properties such as low vapor pressure, low fast neutron cross section and high emissivity. At the operating temperature between 2000°K and 2300°K, niobium and hafnium carbides have comparable vapor pressure (Figure 2). Their fast neutron cross sections are also comparable. Hafnium carbide however has a much higher emissivity, being in the range of 0.8 to 0.9 as opposed to 0.45 of niobium carbide (figure 3). LEOS has performed emittance measurement of plasma-sprayed HfC on tantalum substrates. Our preliminary results indicate the emittance to be 0.9 to 0.95 in the 900°K to 1200°K temperature. Measurements of this material with different substrates and at higher temperatures are planned.

We have not performed an extensive configuration study to evaluate the optimal configuration in terms of the series-parallel option and the associated problem of mechanical integration. However, some realism must be factored-in in the converter design. Since the emitter shoe planform area is 4 x 4 cm and the core (radiating) length is 50 cm, it is convenient to consider the surrounding thermionic cells (circumferentially) as consisting of four sectors. Each sector will then consist of 60 cells in series and parallel connection as shown in figure 4. Each sector will produce 2.4 kW at 300A and 8V output feeding into a dc/dc converter. Although the dc/dc converter output was shown to be 120V, the output voltage can be any value depending on the user's need and on the spacecraft integration scheme.

**THERMIonic CONVERTER DESIGN**

For this work the thermionic converter will be a planar type similar to the SET (Solar Energy Thermionic) converter developed by LEOS for Jet Propulsion Laboratory in the 1960's.

The goals which the thermionic converter designer is striving for are (1) high power density, (2) high conversion efficiency, and (3) high output voltage.
TOTAL ELECTRICAL OUTPUT
TOTAL THERMIONIC CELLS
FOUR (4) SECTORS, EACH CONSISTING OF
SECTOR VOLTAGE (12S X 5P)
SECTOR CURRENT

<table>
<thead>
<tr>
<th>Description</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>TOTAL ELECTRICAL OUTPUT</td>
<td>9.5 Kw</td>
</tr>
<tr>
<td>TOTAL THERMIONIC CELLS</td>
<td>240</td>
</tr>
<tr>
<td>FOUR (4) SECTORS, EACH</td>
<td>60</td>
</tr>
<tr>
<td>CONSISTING OF</td>
<td>CELLS</td>
</tr>
<tr>
<td>SECTOR VOLTAGE (12S X 5P)</td>
<td>8 V</td>
</tr>
<tr>
<td>SECTOR CURRENT</td>
<td>300A</td>
</tr>
</tbody>
</table>

Figure 4.

Often high output voltages are specified at the sacrifice of power density to alleviate the power conditioning requirements. There are four converter parameters which strongly influence the above-mentioned objectives, namely:

- The emitter material and operating temperature
- The collector material and operating temperature
- The cesium vapor pressure (hence, the cesium reservoir temperature)
- The interelectrode spacing.

A typical range of operating temperatures is 1800 to 2000 K for the emitter and 900 K to 1000 K for the collector. In the order of increasing temperature, the converters are 10 to 18 percent efficient, with the power density varying from 10 to 15 watts/cm², and output voltages from 0.5 to 0.8 volt. With future improvements, thermionic converters operating in the 1800 to 2000 K range may be conservatively estimated as 15-25 percent efficient, with the power density of 20-40 watts/cm² and output voltage approaching 1.0 volt.

Essentially, the improvement will be realized by lowering the collector work function through new material or a specially prepared collector surface or by inclusion of additive to the cesium vapor. Low collector work function contribute directly to an increase in the output voltage and power density. Past experience has indicated that a reduction in collector work function from 1.6 volts to 1.4 volts leads directly to a 0.2 volt increase in the output voltage at a constant current.

The effects of additives on the converter performance are not well understood and must await until more reproducible experimental data are obtained. In order to provide rational for device optimization, various converter parameters and effects will be discussed.

Pressure-Spacing Effect. A thermionic converter operating in the arc mode is essentially a gas discharge device. That is to say, there will be variations of power output with spacing. The power output density has been found to be maximum when the product of cesium vapor pressure and electrode spacing is approximately 17 mil-torr. Figure 5 shows the interelectrode spacing effect on the converter performance. Table 4 substantiates the optimum pressure-distance to be about 17 mil-torr for a rhenium converter (4).

![Figure 5](image-url)
The optimum "pd" relationship was also found to hold across the realm of electrode material combinations, such as rhenium-rhenium, rhenium-molybdenum, tungsten-molybdenum and tungsten single crystal materials. The only major effect of changing electrode materials appears to revolve around the capacity of the emitter to absorb cesium and give an adequate emission density at reasonable pressure and reasonable spacings. For example, in order to achieve 15 to 20 watts/cm² at 0.70 volt from a Ta-Mo system, an interelectrode spacing of approximately one mil must be resorted to, with a cesium vapor pressure of approximately 20 torrs, whereas in the Re-Mo or Re-Re system, the same power density can be achieved at a spacing of 3 to 5 mils and a cesium pressure of approximately 5 torrs. These latter spacings, of course, are much more suitable for obtaining reproducible and reliable thermionic diodes.

\[ \text{TABLE 4} \]

**SUMMARY OF PRESSURE DISTANCE DATA TAKEN FROM INTERELECTRODE SPACING VS VOLTAGE OUTPUT CURVES OF RHENIUM-RHENIUM TEST VEHICLE**

<table>
<thead>
<tr>
<th>( T_{\text{EMITTER}} ) (K)</th>
<th>( T_{\text{CEASER}} ) (°C)</th>
<th>( P_{\text{CEASER}} ) (torr)</th>
<th>( d_{\text{EVT}} ) (mil)</th>
<th>( pd_{\text{targets}} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>1600</td>
<td>291</td>
<td>1.22</td>
<td>12.5</td>
<td>16.7</td>
</tr>
<tr>
<td>1700</td>
<td>291</td>
<td>1.43</td>
<td>11.0</td>
<td>15.7</td>
</tr>
<tr>
<td>1700</td>
<td>303</td>
<td>1.96</td>
<td>8.0</td>
<td>15.8</td>
</tr>
<tr>
<td>1800</td>
<td>310</td>
<td>2.35</td>
<td>7.1</td>
<td>16.8</td>
</tr>
<tr>
<td>1800</td>
<td>320</td>
<td>3.01</td>
<td>5.3</td>
<td>15.9</td>
</tr>
<tr>
<td>1800</td>
<td>331</td>
<td>4.02</td>
<td>3.9</td>
<td>15.7</td>
</tr>
<tr>
<td>1900</td>
<td>331</td>
<td>4.02</td>
<td>3.9</td>
<td>15.7</td>
</tr>
<tr>
<td>1900</td>
<td>331</td>
<td>4.02</td>
<td>3.9</td>
<td>15.7</td>
</tr>
<tr>
<td>2000</td>
<td>344</td>
<td>5.30</td>
<td>3.0</td>
<td>15.9</td>
</tr>
<tr>
<td>2008</td>
<td>350</td>
<td>6.06</td>
<td>2.7</td>
<td>16.3</td>
</tr>
</tbody>
</table>

**Emitter Materials.** There are two important aspects concerning the selection of emitter materials. First, it is desirable to have materials with the highest bare work function, since for a given emission density, a low cesium pressure will be required, with a resultant wider spacing allowable. Secondly, it is desirable to operate the converter at a highest temperature allowable, that is without an excessive evaporation of emitter materials which will plate out at the collector. Contamination of the collector surface results in an increase in collector work function and subsequent converter performance degradation.

**Effects of Cesium Pressure on Emitter and Collector.** The presence of cesium on the emitter or collector surface is to cause the effective work function to be lower than the work function of the bare surface with the resultant increase in emission density. The emission characteristic as a function of the electrode and cesium reservoir temperatures can be summarized by the Taylor-Langmuir S curves. Figure 6 shows the saturated emission from a polycrystalline rhenium surface. For the emitter operating a 2000K and cesium pressure of 4 torr, the saturated current density is approximately 40 amp/cm². The corresponding emitter work function is 2.72 volts. The collector work function appears to be converging at 1.45 volts for collector temperatures from 900 to 1100K.

**Low Work Function Collectors.** The Taylor-Langmuir data similar to figure 6 for other collector materials were obtained and their minimum cesiated work functions are empirically correlated with their corresponding bare work functions. Results shown in figure 7 clearly indicate that low work function collectors are obtained by starting with the highest bare work function materials.

**Figure 6**
Summary. Data presented in figures 5 to 7 inclusive point to the fact that at the current state of affairs, chemical vapor deposit rhenium is the best material for both emitter and collector. Its low collector work function will provide:

- High output voltage and power density (also efficiency)
- Less probability of collector contamination (Re-Re system)
- Relatively large spacing (3-5 mil) at 1800 to 2000°K

The CVD-Re system will be used as the baseline design for the "Preliminary Converter".

Selected Design Point

A variable parameter test vehicle of the rhenium-rhenium converter was fabricated and extensively characterized under the JPL-sponsored contract (4). The converter performance data pertinent to the present work are summarized in table 6.

<table>
<thead>
<tr>
<th>TABLE 6</th>
<th>EXPERIMENTAL DATA OF LEOS CONVERTER</th>
</tr>
</thead>
<tbody>
<tr>
<td>No. 1</td>
<td>No. 2</td>
</tr>
<tr>
<td>Emitter Temperature (°K)</td>
<td>1800</td>
</tr>
<tr>
<td>Collector Temperature (°K)</td>
<td>983</td>
</tr>
<tr>
<td>Cesium Reservoir (°K)</td>
<td>583</td>
</tr>
<tr>
<td>Output Voltage (V)</td>
<td>0.5</td>
</tr>
<tr>
<td>Spacing Range (mils)</td>
<td>6-8</td>
</tr>
<tr>
<td>Current (A)</td>
<td>38</td>
</tr>
<tr>
<td>Power (W)</td>
<td>19</td>
</tr>
<tr>
<td>Power Density (W/cm²)</td>
<td>9.5</td>
</tr>
</tbody>
</table>

As it was indicated in System Tradeoff, the reactor size consideration favors a large area emitter operating at a moderate temperature between 1800 and 1900°K and still meeting the program objective. The emitter temperature of 1850°K is therefore chosen as a baseline. Table 7 summarizes the converter other design parameters.

<table>
<thead>
<tr>
<th>TABLE 7</th>
<th>CONVERTER DESIGN PARAMETERS</th>
</tr>
</thead>
<tbody>
<tr>
<td>Emitter Temperature</td>
<td>1850°K</td>
</tr>
<tr>
<td>Collector Temperature</td>
<td>990°K</td>
</tr>
<tr>
<td>C, Reservoir Temperature</td>
<td>620°K</td>
</tr>
<tr>
<td>Spacing</td>
<td>4 mils</td>
</tr>
<tr>
<td>C, Pressure</td>
<td>4 torr</td>
</tr>
<tr>
<td>Output Voltage @ Max. Power Density of 10.5 W/cm²</td>
<td>0.57 V</td>
</tr>
<tr>
<td>@ Power Density of 8.9 W/cm² (Des.Pt.)</td>
<td>0.67 V</td>
</tr>
<tr>
<td>Emitter Area</td>
<td>4.67 cm²</td>
</tr>
<tr>
<td>Converter Output Per Cell</td>
<td>40 w (70 w max.)</td>
</tr>
<tr>
<td>Emitter Shoe Planform Area</td>
<td>4 x 4 cm²</td>
</tr>
</tbody>
</table>
Figure 8 illustrates the thermionic converter assembly. The salient features of this converter under development are:

- Emitter shoe made of w-5% Re selected on the basis of high creep strength.
- CVD rhenium for both the emitter and collector
- Niobium-sodium heat pipe for waste heat removal from the collector to the Nb-radiator
- High emissive coating of hafnium carbide ($e = 0.9$) will be applied to the emitter shoe and the radiator.

![Figure 8](image.png)

**PROGRESS**

The program milestones at the end of two years of design studies and development efforts are:

- Development of CVD rhenium collector integral to the niobium-sodium heat pipe which is brazed to the hafnium carbide coated radiator.
- Development of the CVD rhenium emitter which is integral to the emitter shoe made from tungsten-rhenium alloy. The shoe heat receiving surface is coated with hafnium carbide.
- Integration of the emitter and collector heat pipe into a complete converter.

- Development of a simulated heat source capable of radiatively transferring up to 400 W at 2400°K to the emitter shoe surface.

The overall summary of the progress is that the integral assembly of collector heat pipe/radiator has been successfully tested and it is ready to be integrated into a complete converter. For the emitter assembly, we have a schedule setback due to the problem of the CVD process which is being corrected. The simulated heat source has been successfully be tested with a mock-up emitter shoe. The heat source is capable of receiving 470 W electrical input and heating the emitter shoe up to 2000°K. The complete converter should be ready for test, in a radiatively heated mode, in the near future. Details of program achievements and some of the problems encountered will now be described.

Collector Heat Pipe/Radiator Assembly. The sodium heat pipe is made with niobium. For maximum reliability, the grooved capillary design is chosen over the wick screen mesh approach.

The cylindrical container section was made by the CVD process. Figure 9 shows the molybdenum mandrel which will be used to make the CVD niobium structure.

![Figure 9](image.png)
Figure 10 shows the CVD niobium structure in the as-received and after machining conditions. The evaporator end cap which serves also as the collector was made from molybdenum. (Originally it was to be made from niobium, however it was discovered that the CVD rhenium could not be reliably deposited on the niobium substrate.)

The collector heat pipe/radiator assembly was tested in a horizontal position to mitigate the gravity effect. The cylindrical section (except the emitter lead terminal) was heat shielded with six layers of molybdenum foils and alumina fiber sheets. The CVD rhenium collector face was heated by an EB gun. Figure 12 shows the temperature profile of the heat transport section which is relatively flat indicating the proper functioning of the heat pipe.

Figure 11 shows the assembly of the collector heat pipe complete with the finned radiator which has been coated with the high emissive coating of hafnium carbide.
Figure 13 shows the temperature profile of the radiator which indicates that the radiator design needs to be improved. One solution is to replace the fins with two small sodium heat pipes which are brazed to the condenser of the primary heat pipe and to the radiator plate. The two small heat pipes might be loaded with non-condensable inert gas. When properly designed such a radiator (and hence the collector) can have a fixed temperature (say 1000 ±50 K) but a two-to-one heat rejection capability. Such a flexibility is important for two reasons. First, at this stage of development the device conversion efficiency is not precisely known but the collector temperature should be held to within 50 K of the design point. Second, the radiator heat rejection capacity should accommodate the variability of the load demand, including an occasional overload condition.

Emitter Assembly The 4.7-cm\(^2\) circular emitter proper is an integral part of the square emitter shoe but to be coated with CVD rhenium. The substrate material is W-3.5\%Re. An earlier fabrication experiment was done to establish the CVD processing procedure, using a small circular substrate. Bonding of the CVD rhenium layer (25 mils) was tenacious. However when it came to coating the real part, bonding was found unsatisfactory. It appears that the fixture used was unsuitable for the geometry of the part. The CVD vendor is in the process of correcting the deficiency.

Simulated Heat Source. It is a requirement of this program that the converter be heated radiatively; i.e., not by an EB gun. The reason for this is because both the surfaces of the heat source and the emitter shoe are coated with high emissivity coating of hafnium carbide and they must be tested for durability at high operating temperatures. (From the reactor side, graphite itself has high emissivity but hafnium carbide is needed to suppress the high evaporation rate.)

Figure 14 shows the simulated heat source which is made from tungsten wire and coated with hafnium carbide. It is designed to operate at 2400 K with 400 W from a 60-Hz source. The shape of the heater is designed to cover the 4 cm x 4 cm planform area of the emitter shoe. The heater element is mounted in the insulated cavity made from molybdenum foils and alumina fiber sheets.
Preliminary tests with a mockup emitter shoe indicate that the emitter shoe reached the temperature 2000°K with the input of 470 W. Efforts are continuing in minimizing the heat loss with additional heat shields.

**Artist's Model** Figure 15 illustrates five full-scale converter models on the circumference of a 26-cm diameter circle. In the real reactor system the space between the heat pipes will be filled with beryllium to satisfy the neutronic requirement. The converter design is modular such that they can be integrated and tailored to meet the power system need from 5 to 40 kW.
REFERENCES


INTRODUCTION

For the past thirty years it has been generally recognized that the static nature and high heat rejection temperature of the thermionic conversion process were highly advantageous characteristics with regard to the use of this process in nuclear reactor space power supplies. There are two general ways of using thermionic conversion in a space reactor. One of these, termed the out-of-core method, involves transferring heat from a hot nuclear fuel mass to an array of thermionic converters either by radiation (Becker 1968) or by the use of high temperature heat pipes (Koenig 1979). The requirement for either very high fuel temperatures (for radiative heat transfer) or electrolytically stable electrical insulation and chemically stable heat pipe operation at very high temperatures (for heat pipe systems) has presented a major roadblock to the development of out-of-core systems.

The other general method of building a thermionic reactor, the in-core method, is to incorporate nuclear fuel into the emitters of cylindrical thermionic converters and actually form a critical reactor mass from an arrangement of such converters (Grover 1958, Ranken 1960). This approach has received the most attention over the past three decades, resulting in the recent demonstration by the USSR of a 10 kWe moderated thermionic reactor operating in orbit for almost one year (Ponomarev-Stepnoi 1989). In the US degradation-free performance has been obtained on thermionic fuel elements (TFEs) for similar periods of time (Gulf General Atomic Co. 1973). These units were designed as core elements of an unmoderated (fast neutron spectrum) reactor, but were tested in a thermal test reactor.

The above test results are very impressive, particularly the USSR test, which demonstrated orbital operation of a space reactor for a much longer period of time than had previously been achieved. However, the lifetime goals for space power supplies are generally in the range of 7 to 10 years. Furthermore, at high power levels (above 100-200 kWe) it is highly advantageous to use a fast neutron spectrum design for a thermionic reactor. This results in a particularly harsh environment for the TFE in terms of a combination of high fast neutron flux, high temperature, and high electrical fields. Demonstrated lifetimes under conditions equivalent to a high power fast reactor environment have been on the order of 5% of the goal lifetime.

It was to improve on this situation that the Thermionic Fuel Element Verification Program was initiated in the US.
The Thermionic Fuel Element Verification Program is a development and demonstration effort begun in 1986 to show that TFEs could be built that would have lifetimes exceeding 7 years in a fast neutron spectrum in-core thermionic reactor with a power output capability of 2 MWe. It was established as a TFE development program rather than a reactor program because achieving a long life TFE was considered to be the main technology challenge for this type of power system. Because the reactor coolant temperature is in the range of experience of terrestrial liquid metal cooled reactors, much of the technology already demonstrated for the terrestrial reactors can be applied to the thermionic reactor design provided a successful TFE design is demonstrated. A further justification of the TFE approach is that many design studies have been done for thermionic reactors in the past so that the needed parameters for TFE design are well known in the power range from 5 kWe to 5 MWe. It is also well known that a single TFE design can cover this entire power range with little modification of the basic construction.

Accepting that verification of seven year lifetime for a TFE is the essential element of establishing seven year lifetime capability for an in-core thermionic reactor still leaves the problem of how to demonstrate seven year TFE lifetime. Ideally, a development/demonstration program for a TFE suitable for high power thermionic reactors would involve a great amount of irradiation testing in fast neutron spectrum testing reactors such as the FTR or EBR-II. However, the long duration testing of TFEs in either of these two reactors is not practical unless the reactors can be operated continuously at about 10% of design power. This would mean that the test reactor would have to be dedicated to the thermionic program, a costly proposition. In addition, the length of a program to show 7 year lifetime would be very long.

An alternative, basically low budget, approach to TFE verification is to conduct a testing program that will demonstrate the capability of individual components of the TFE to withstand the temperature/irradiation environment and, where applicable, the electric field environment of a fast thermionic reactor for the required lifetimes. Because the lifetime goals are so long, it is necessary that accelerated tests be conducted in order to reduce the span of the development program. This is the approach being taken in the TFE Verification Program. The component testing includes irradiation testing of fueled emitters, sheath insulator assemblies, seal insulators, integral cesium reservoirs, intercell emitter alignment assemblies and intercell insulator coatings. These tests are supported and augmented by bench tests of electric field effects on insulator candidates and performance tests of individual thermionic converters. The test program results are fed into an analytical modeling effort with the goal of predicting component behavior as a function of total exposure as well as exposure rates (acceleration factors) so that the test results can be reliably extrapolated in time to the seven year requirement. Relatively long duration tests of partial length TFEs, conducted in a thermal spectrum reactor, serve.
to integrate the component test program and specifically evaluate such factors as fission product vent performance and fission product and fuel migration effects on TFE electrical performance. The program is planned to culminate in a two month test of a six cell TFE in a fast spectrum test reactor.

The two lifetime limiting phenomena of greatest concern in TFE design are emitter swelling caused by fission gas formation in the fuel and fast neutron damage to ceramic insulation materials. The nature of these problems can be seen by referring to Fig. 1, which is an illustration of part of a TFE. The figure shows that the emitter, a cylindrical tungsten cup containing UO₂ pellets, is separated from the collector by a narrow gap. Should its diameter increase by more than 3% to 4% it would touch the collector and electrically short the converter, thus causing loss of electrical output. Emitter swelling can be caused by fission gas formation in two ways. The fission gases that escape from the fuel can collect in the emitter cavity and exert a pressure that causes the emitter wall to creep slowly outward until contact is made with the collector. This effect can be alleviated by venting the released fission gas (approximately 90% of the total generated) from the emitter cavity. Venting is essential for achieving lifetimes of more than a few thousand hours, but does not completely eliminate fuel swelling. The fission gas that remains in the fuel causes it to push outward on the emitter cup wall. Swelling of the emitter caused by this mechanism can be reduced by increasing the strength of the emitter wall relative to the fuel, either by thickening the emitter wall, or strengthening it, or by weakening the UO₂ fuel.

The locations where ceramic is used in the TFE can also be seen in Fig. 1. The most critical use is the sheath insulator that isolates the collectors in the series string of converters from the outer metallic sheath. The latter is required to protect the ceramic sheath from the reactor coolant as well as to give structural integrity to the fuel element. Because the reject heat from the converter collector must flow radially outward to the reactor coolant, it is necessary, in order to minimize radial temperature drops, that the sheath insulation layer be thermally bonded to both the collectors and the outer metallic sheath. The insulation must also be thin for the same reason. However, it also must standoff at least half of the reactor output voltage and hence it can experience relatively high electrical fields which can cause electrolytic decomposition of some ceramic materials at the operating temperatures of the reactor coolant.

Another important use of insulation in the TFE is the insulator that isolates the emitter from the collector of each converter. This insulator is required to be a hermetic seal for TFE designs where the fission gases are vented in special channels so that they do not come into contact with the electrode surfaces or the cesium vapor in the interelectrode space. For designs where venting occurs through the cesium vapor space hermeticity is not a requirement and the intercell insulation task is relatively easily
accomplished. Insulation is also required for the emitter alignment structure that serves to restrict emitter vibration during rocket launch into orbit as well as provide positive alignment at the free end of the emitter. Still another insulation use, which is not shown in the figure, are ceramic coatings required in the intercell region to facilitate TFE assembly.

The problem of fast neutron damage to ceramic materials is primarily one of dimensional growth. If the ceramic swells and the metal to which it is bonded does not, then debonding will eventually occur. In the case of the sheath insulator this will cause large temperature drops leading to excessive collector temperature and consequent poor converter performance. For the seal insulators where hermeticity is a requirement, ceramic swelling can cause leakage. For the emitter alignment insulator and ceramic coatings, debonding caused by ceramic swelling can result in loss of function.

TESTING PROGRAM -- OUT-OF-CORE

While the main emphasis in the TFE Verification Program has been on irradiation testing to verify suitable emitter and ceramic component performance, a corollary program of laboratory testing of converters and components has also been established. Part of this support program is devoted to converter performance determination and improvement. Both planar and cylindrical geometry converters have been constructed and operated to study methods of achieving emitter oxygenation and to learn the effects of such oxygenation on converter performance. Long term effects of cesium graphite reservoirs on diode performance are also under study as are such practical concerns as the ability of converter performance codes to predict cylindrical diode performance from planar diode data and to predict the effect of nonuniform emitter swelling on converter performance.

Among the results of the out-of-core converter testing are that cylindrical diode performance can be predicted very well using planar diode data as can the effect on performance of a nonuniform emitter-collector gap. Emitter oxygenation allows the emitter-collector gap to be increased to as much as one millimeter with little degradation of performance at emitter temperatures in the vicinity of 1800 K.

Laboratory studies of seal insulators have been primarily concerned with fabrication methods and thermal cycling capability. Candidate specimens have been Al2O3, Al2O3-Nb cermet, Y2O3, Y2O3-Nb cermet, yttria alumina garnet (YAG), and YAG-Nb cermet brazed to Nb end pieces of various geometries. One of the major successes of this part of the program has been the development of leak tight seals of YAG brazed to Nb with high temperature braze material.
Because of the great expense of testing sheath insulators with an impressed voltage in the fast neutron spectrum reactor, laboratory testing of these components is especially important. An extensive series of tests is underway to determine the effectiveness of various types of insulator as a function of time, temperature, and electric field strength. The candidate specimens are the same as those for the seal insulators. Significant results to date are that alumina specimens maintain their electrical integrity very well under equivalent TFE electrical and temperature conditions and that the loss of resistance of yttria specimens with time at temperature is largely attributable to a carbon impurity. It has also been learned that YAG sheath insulator specimens maintain good resistance when tested in a high vacuum environment despite radial cracks in the ceramic layer. The testing of cracked specimens reflects the lack of success to date of fabricating crack free YAG sheath insulator specimens.

TESTING PROGRAM -- IN-CORE

Program Scope

The irradiation testing portion of the TFE Verification consists of an extensive series of tests in three separate test reactors. Some idea of the scope of this program can be gotten from Fig. 2 which shows the various irradiation tests and the schedule in terms of experiment design, experiment fabrication, capsule assembly, irradiation, and post irradiation examination (PIE) times. The identifying symbols for the tests appear in the column on the far right of the figure along with the acronyms identifying the test reactors. The latter include the Fast Test Reactor (FTR) at Richland, WA, the Experimental Breeder Reactor (EBR-II) at the Idaho National Engineering Laboratory in Idaho, and the TRIGA reactor at the General Atomics facility in San Diego, CA. The first two of these are fast neutron spectrum reactors while the last is a water moderated thermal spectrum test reactor.

The TFE Verification testing schedule was initially laid out as a four year effort. The fact that it now spans more than six years is a reflection of changes in funding availability. The FTR was selected as the test facility for insulating components and cesium reservoirs because of existing test vehicles easily adapted to testing at regulated temperatures in the desired range. The components in question are being tested in four batches, the last of which is a test of sheath insulator specimens subjected to an electric field while at operating temperature in a fast neutron flux. This last test is very costly and has, as a result been subjected to more schedule slippage than the others.

In the case of the UFAC series of fueled emitter irradiations the test reactor of choice was the EBR-II primarily because of shorter run times and testing flexibility which allow periodic neutron radiography and changing specimens in the irradiation test capsules when required. Two sizes of fueled emitters are being irradiated. One is prototypical for the 2 MWe fast
reactor design with real time thermal power density in the fuel, and the other is one half of prototypical diameter so that the power density can be increased by a factor of four without changing the inside to outside fuel temperature ratio relative to the real time testing case.

The other group of tests shown in Fig. 2 are TFEs that are largely prototypical except for having less than the full complement of converters. Most of these tests will be conducted in the TRIGA reactor. The initial tests in this series are being done with TFEs that have just one converter. Subsequent tests will have three and eventually six converters. The purpose of these tests is to demonstrate successful TFE fabrication, assembly, and operation, to investigate the electrical performance stability with integral cesium reservoirs, to study the effect of fission gases and vapors mixing with the cesium atmosphere, and to ascertain the effectiveness of the fission product trapping system in the emitter support stem and the operation of the fission gas venting channels. The culminating test in this series, and in the TFE Verification Program as a whole, will be the testing of a six cell TFE in a fast reactor test facility.

Insulator Seals

Candidate insulator seal materials are included in the UCA-1, 2, and 3 FTR test vehicles. UCA 1 and UCA 2 have been completed except for some of the PIE investigations. The fast neutron fluences to which they were exposed were:

- UCA-1: $2.5 \text{ to } 7.2 \times 10^{22} \text{ n/cm}^2$
- UCA-2: $2.5 \text{ to } 6.7 \times 10^{22} \text{ n/cm}^2$ ($E_n \geq 0.1 \text{ MeV}$)

The reference design fluence goal is $2.7 \times 10^{22} \text{ n/cm}^2$.

Fig. 3 shows the seal geometries that were included in the irradiation tests. Some of the major findings of this testing are that the trilayer seal specimens that were made with yttria and alumina/Nb cermet appeared to have withstood irradiation very well. This configuration has the advantage of the greatest potential strength of the ceramic to metal bond. Some helium leakage was observed for some of these specimens during PIE, but the rate was low enough to be acceptable for seven year reactor operation.

Taper geometry seal insulator specimens also show considerable promise. The single YAG specimen that has been irradiated to date in the taper configuration had a visual appearance unchanged from the pre-irradiation condition except for blackening of the ceramic. However, it was not leak tight. The taper geometry seals that were made with sapphire showed no visual evidence of cracks extending through the ceramic body, but there were short axially directed cracks in the thinnest part of the taper in the niobium skirts and some peeling away of the niobium from the ceramic in this region. The assemblies were not leak tight. Further investigation is
required to determine the cause of the loss of hermeticity of both types of taper seal specimens.

The butt seal specimens that were tested did not fare well in that they showed a propensity for cracking and for generally low post-test strength. This configuration has been dropped from the program.

Sheath Insulators

Several geometries of sheath insulator were tested. These are shown in Fig. 4. They differed primarily in the treatment of the ends where stresses caused by ceramic swelling reach peak values. Both the alumina and yttria specimens tended to bell out at the ends of the specimens and circumferential cracks were formed in the ceramic layer. If, as appears to be the case from visual observation and diameter measurements, the depth of these end cracks is on the order of a millimeter, then the sheath insulator would still be capable of performing its function of providing good electrical resistance while permitting radial heat flow with little temperature drop. In general, the amount of damage to alumina specimens was much less that anticipated based on earlier measurements at one fifth the present fluence levels. The end cracking appeared no more severe and the diameter increase was somewhat under 1%, which was only about 50% more than the increase found at the lower fluence levels.

An important finding of the sheath insulator irradiations was that tapering of the metal components at the ends of the specimens did not have the effect, predicted from finite element stress analysis, of relieving the end stresses and preventing the belling out type of distortion mentioned above. The wrap around geometry was tested only for the alumina/niobium cermet which is very resistant to the end distortion phenomenon in any case. No cracking was observed for this specimen. More surprising were the test results for the specimens with partial outer sheath cutaway and complete outer sheath cutaway (see Fig. 4). The former geometry appeared to reduce the amount of distortion observed for alumina and yttria specimens and the latter reduced it still further, in some cases completely eliminating it.

Tentative conclusions from the partial PIE results obtained on the sheath insulator specimens are that yttria with cutaway stress relief is a leading candidate if the out-of-core test results show it to retain adequate electrical resistivity for seven year equivalent lifetime. The alumina/niobium cermet looks very promising from both a mechanical stability and electrical resistivity standpoint.

Cesium Graphite Reservoirs

The major concern with cesium/graphite reservoirs is that fast neutron damage would change the properties of the graphite lattice into which the cesium atoms are intercalated so that the cesium pressure for a
given reservoir temperature would change radically. To study this effect it was necessary to measure the cesium vapor pressure versus temperature relationship of cesiated graphite specimens both before and after irradiation. Initial attempts to do this were unsuccessful because specimens of highly oriented graphite apparently increased in volume so much from neutron damage (possibly several hundred per cent) that they burst the container they were in and removed any possibility of post test measurements. However, successful measurements have been made on one specimen of graphite with randomly oriented fine crystal structure. The results of these measurements are shown in Fig. 5 where cesium vapor pressure is plotted as a function of reciprocal temperature for the before and after irradiation cases. The results indicate that the irradiation to a fluence value of $4.2 \times 10^{22}$ n/cm$^2$ caused less than a factor of two increase in cesium vapor pressure. This change is small enough to be tolerable in reactor design and hence is a very encouraging result relative to the possibility of incorporating the cesium/graphite type of integral reservoir into the TFE.

**Fueled Emitters**

The irradiation data on fueled emitters includes testing of nine tungsten clad $\text{UO}_2$ specimens that was initiated during the competitive phase of the SP-100 space reactor development program and continued under the umbrella of the TFE Verification Program. The emitter specimens were sized for a fast spectrum 100 kWe thermionic reactor design and were somewhat more than twice the diameter of the present reference design emitters. They were contained in three capsules irradiated in TRIGA. The status of these tests is given in Table 1.

| TABLE 1. Status of Fueled Emitters in TRIGA as of August, 1989 |
|---------------------------------|-----------------|---------------|---------------|
| Capsule 1 (Removed)            | 19,425          | 0.2           | 0.5           |
| Capsule 2                      | 35,000          | 0.3           | 0.6           |
| Capsule 3 (Removed)            | 24,000          | 0.3           | 0.6           |

Periodic neutron radiographs have been taken of the TRIGA test capsules to determine emitter distortion. From this it has been possible to study dimensional increase with time. The composite behavior of the emitters has been brought together into one representation shown in Fig. 6.
The curves indicate the closure of the emitter-collector gap with time and are based on experimental data to which approximate corrections for temperature variations and extrapolation to gap closure times have been made based on analytical models. It is apparent that gap closure time is very dependent on relative cladding thickness and also that seven year lifetime at an emitter temperature of 1800 K is achievable with emitters of this size.

As shown in Fig. 2, real time and accelerated testing are underway in EBR-II in test vehicles UFAC-2 and UFAC-1, respectively. The emitters in these two units, referred to as Batch 1, were placed in the reactor in May, 1987. Batch 2 will be put into the reactor in autumn of 1989 in test vehicle UFAC-3 and a reconstitution of UFAC-2. The status of these irradiations as of October, 1989 is shown in Table 2.

TABLE 2: Status of Fueled Emitter Irradiations in EBR-II as of October 1989

<table>
<thead>
<tr>
<th>Test Variables</th>
<th>Fast Fluence $10^{22}$ n/cm²</th>
<th>Burnup Atom %</th>
<th>Status</th>
</tr>
</thead>
<tbody>
<tr>
<td>Emitter thickness, Fuel volume</td>
<td>7</td>
<td>1.4</td>
<td>Irradiation continuing</td>
</tr>
<tr>
<td>Emitter thickness and temperature, Fuel volume</td>
<td>7</td>
<td>4</td>
<td>Beginning PIE</td>
</tr>
<tr>
<td>Fuel volume, Adv. emitter material, Adv. Fuel</td>
<td>0</td>
<td>0</td>
<td>Irradiation beginning</td>
</tr>
<tr>
<td>Adv. emitter material, Adv. fuel, Emitter temp.</td>
<td>0</td>
<td>0</td>
<td>Irradiation beginning</td>
</tr>
</tbody>
</table>

Three of the accelerated emitters and one of the real time emitters of Batch 1 were removed early because neutron radiography showed that the emitter support had fractured at the tungsten-to-tantalum bond, most likely...
during transport for radiography, because of a combination of void formation at the bond and an inadequate diffusion bond. Both problems were addressed in the Batch-2 fabrication. Initial PIE results show generally very good appearance of the fuel and cladding except for the fracture location and some minor chemical interaction between the UO₂ fuel and the WRe fuel support plates at the ends of the emitter. A typical dimensional change measurement result for an accelerated emitter is shown in Fig. 7.

**TFE Prototypes**

Testing of TFE prototypes began in September, 1988 with the installation of the one-cell unit, 1H1, into TRIGA. A schematic illustration of this unit is shown in Fig. 8. 1H3, a one cell unit with fission gases vented through the interelectrode gap, was installed into TRIGA in August, 1989. Both units are operating without incident. The former has accumulated one year of test time without performance degradation. Its performance is very close to that predicted from archival planar converter data as well as planar and cylindrical converter data generated for 110 oriented tungsten emitters in this program.

**CONCLUSIONS**

The TFE Verification Program is an experimental and analytical program to demonstrate the design, performance, and lifetime capability of thermionic fuel elements. The major emphasis in this program has been placed on the irradiation testing of components and prototypes under conditions as close as possible to the exacting conditions that TFEs will experience in a seven year lifetime 2 MWe power level in-core thermionic reactor. Test results to date have been very encouraging, particularly with regard to insulator options, fueled emitter behavior, and integral cesium reservoir stability. Some of these results are summarized in Table 3. It is expected that the verified design emerging from this program will be a basic building block for power systems with requirements from a few kilowatts to megawatts of electrical power.

**ACKNOWLEDGMENTS**

The TFE Verification Program is jointly sponsored by the Department of Energy and the Department of Defense (SDIO). The prime contractor for this work, charged with overall management of the technical program and production of test specimens, is General Atomics. Subcontractors assisting in carrying out the program are Thermoelectron Technology Corporation and Rasor Associates. The Westinghouse Hanford Company has played a major role in planning and managing the fast reactor irradiation testing and postirradiation examination in a highly effective manner. The Argonne National Laboratory has an important supportive role for irradiations conducted in the EBR-II. Technical oversight for the program has been furnished by the Los Alamos National Laboratory.
### TABLE 3: Summary of Particularly Significant Findings of the TFE Verification Program.

<table>
<thead>
<tr>
<th>ITEM</th>
<th>FINDINGS</th>
</tr>
</thead>
<tbody>
<tr>
<td>TFE</td>
<td>1H1 operated for more than 1 yr at predicted performance level. TFE 1H3 started operating in TRIGA in August 1989.</td>
</tr>
<tr>
<td>Insulator Seals</td>
<td>Trilayer seal configurations with yttria and alumina/niobium cermet have withstood neutron fluences up to $7 \times 10^{22}$, almost three times the 7 yr fluence for the 2 MWe reference design reactor. Observed leakage rates are acceptable for 7 yr reactor operation. YAG taper seals also appear promising.</td>
</tr>
<tr>
<td>Sheath Insulators</td>
<td>High neutron fluence irradiation results show that yttria and alumina/niobium sheath insulator assemblies with cutaway end design will remain crack free and well bonded for 7 yr reactor lifetime. Graded alumina dimensional increase was much less than expected based on earlier lower fluence tests. Cutaway end design eliminates end distortion.</td>
</tr>
<tr>
<td>Intercell Insulators</td>
<td>Alumina, yttria, and YAG coatings and yttria emitter-alignment insulators show no visual damage after exposure to high neutron fluence.</td>
</tr>
<tr>
<td>Cesium Graphite Reservoir</td>
<td>Cesiated POCO graphite irradiated to a fluence of $4 \times 10^{22}$ n/cm$^2$ maintained the cesium vapor pressure within a factor of two over the temperature range of interest.</td>
</tr>
<tr>
<td>Fueled Emitters</td>
<td>A seven year lifetime at an emitter temperature approaching 1800 K is achievable.</td>
</tr>
</tbody>
</table>

**REFERENCES**


Fig. 1 Section of thermionic fuel element
Fig. 2 Irradiation testing schedule for TFE Verification Program.
Fig. 3  Interelectrode insulator seal assemblies tested in FTR.

Fig. 4  Sheath insulator configurations tested in UCA-1 and UCA-2 irradiation vehicles.
Fig. 5  Dependence of cesium vapor pressure on inverse reservoir temperature before and after irradiation to $4 \times 10^{22}$ n/cm$^2$ ($E_n > 0.1$ MeV).

Fig. 6  Dependence of interelectrode gap closure time on cladding thickness to emitter diameter ratio.
Fig. 7  Comparison of measured and predicted radial emitter deformation versus irradiation time for accelerated test fueled emitter specimen.

Fig. 8  Schematic illustration of 1H1 prototypical TFE assembly undergoing irradiation in TRIGA.
INVESTIGATIONS OF EMISSIVE NON-UNIFORMITY OF A CYLINDRICAL THERMIonic
CONVERTER EMITTER

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The emitter surface of thermionic energy converter (TiC), created by means of
gas-phase tungsten deposition on a cylindrical surface of single-crystal molybdenum with axis orientation [III] is characterized by multifaced structure, repeating with definite periodicity on azimuthal angle. In this connection the necessity appears to characterize TiC emitter not by averaged work function value, but by its distribution on surface. Such a surface characteristic may occur to be effective for the control of emitter production techniques and also in the stability investigations of emissive properties of tungsten emitter surface to long-term temperature effect.

In present papers we give the descriptions of the device and the method of distribution registration of work function on cylindrical emitter surface and the results of the investigations of work function distribution on surface: 1) cylindrical single-crystal molybdenum with axis orientation [III], 2) tungsten surface, deposited by gas-phase method onto cylindrical surface of single-crystal molybdenum, and 3) the same sample after long-term temperature effect.

The diagram of the device for registration of work function distribution on cylindrical emitter surface is shown in Fig. 1.

The device is a thermionic microscope, in which the role of cathode is played by a cylindrical emitter, and an immersion objective is a transaxial electron-optical system. Focusing electrode 2 and electron accelerating anode 3 are cylindrical electrodes, placed coaxially to emitter, having a slot on the circle. Such an electron-optical system combines the properties of a cylindrical projector with thus of a cylindrical lens. Cylindrical projector provides with the image enlargement on the circle with the factor of \( M_z = R/\varphi \), where \( R \) is the circle radius, where a luminescent screen 4 and Faraday cylinder 5 are situated, \( r \) is the emitter radius. Cylindrical lens, looking as a circle slot, gives the enlargement along the forming surface with the factor \( M_o \). Slot dimensions, diameters of electrodes 2 and 3 and potentials on them are selected to provide with equation.

The emitter is heated by thermal radiation and electron bombardment from heater 6. The emitter temperature is registered by photoelectric multipliers 7. The emitter is supplied with rotation and displacement mechanisms and position sensors. In front of Faraday cylinder diaphragm 8 is placed, having the aperture of \( \varphi \) 2mm, determining the square of registered emitter surface (when the image enlargement factor is equal to 50, the linear dimension of registered surface is equal to 4.10^-5m). To avoid the dynatron effect in Faraday cylinder we use grid 9 with transparency of 0.9, having negative potential as relate to the cylinder.
The device works by the following way. To the emitter we apply accelerating voltage $-5 \text{ V}$, the anode is grounded, to the focusing electrode is applied regulated voltage, which is negative as relate to the emitter. To prevent the influence of the magnetic field current of the emitter heater on the emissive non-uniformity image, the latter is seeded by the rectified half-period current, with the frequency of 50Hz; synchronously with flowing current, to the focusing electrode is applied negative voltage, blocking an electron ray.

The electron ray, formed in such a way, gives an image on a luminescent screen and emissive current is registered by Faraday cylinder from a given emitter range. Emission current, coming to Faraday cylinder, through a current amplifier reaches the input "Y" of two-coordinate plotter, and the signal from the emitter position sensor reaches the input "X". During emitter rotation or displacement the emission current dependence upon the emitter position coordinate is written on the plotter. To judge the emissive emitter structure it is necessary to use a comparing analysis of an investigated sample with the model of an ideal single-crystal cylinder with the analogous structure.

In Fig.2 you may see the dependence plot of work function of a cylindrical ideal single-crystal with axial face orientation $[111]$ upon the azimuthal angle. On the plot the main crystallographic directions, received from stereographic crystal projection [1], have been compared with corresponding work function values [2]. The dots, corresponding to main face directions, are connected by conventional curves, because work functions of intermediate faces are known. The work function values have been taken according to the supposition, that recommended in [2] values are related to perfect faces, and possible measured values, which are less than given, will correspond to structure deviation from the perfection.

As in the formation of an epitaxial tungsten layer the sublayer structure plays the main part, that's why the emissive structure of single-crystal molibdenum with axial face orientation has been investigated [111]. In Fig.3 the emission current and work function dependence of this single-crystal upon the azimuthal angle is shown. The sample temperature is $T = 1730 \text{ K}$. From the comparison of the plots in Figures 2 and 3 it follows that for both the investigated sample and the ideal single-crystal we observe the periodicity of emissive properties with the period of 60°. As distinct from the ideal single-crystal we do not observe any structures separately in the investigated single-crystal, corresponding to faces $(12)$ and $(123)$. There is some divergence in absolute values of maximum and minimum work function. The emissive properties are quite uniform along the cylinder forming surface. In Fig.4 the emissive characteristics is shown via the azimuthal angle of epitaxial tungsten coating $(T=1990 \text{ K})$. As it is obvious from the plot there is no strong periodicity in the emissive structure. The maximum work function value is equal to $\varphi \approx 5 \text{ eV}$, and the minimum one

$$\varphi_{\text{min}} \approx 4,75 \text{ eV}.$$  

The surface with $4,75 \text{ eV}$ work function occupies a slight part of the circle. The main part of the surface corresponds to the work function not less than 4,9eV. These work function values are close to $\varphi$, corresponding to structure $(110)$, the divergence can be explained by some face $(110)$ disorientation. Work function distribution along the forming surface is non-uniform and has a chaotic character, (spread in work function is $\Delta \varphi \approx 0,2 \text{ eV}$).

In Fig.5 you see emissive characteristics of epitaxial tungsten coating, subjected to long-term temperature effect,
on the azimuthal angle. In contrast to the described above sample the given sample reveals quite exact periodical structure, which is characteristic of single-crystal sublayer. The maximum work function value is kept approximately in the range of 4.9 eV. Such a stability of coating work function is connected with the sublayer structure stability.

Work function distribution along the forming cylinder surface becomes evident. On the base of fulfilled investigations it is possible to come to the following conclusions:

1. The performed device and the worked out methodology provide with the possibility to investigate work function distribution along the cylindrical emitter surface;

2. Cylindrical single-crystals with axial face orientation [111] are characterized by multifaced surface with periodical structure (period 60°);

3. Epitaxial tungsten coatings take sublayer structure as a result of long-term temperature effect.

REFERENCES:

Fig. 1. Thermionic microscope diagram.

Fig. 2. The dependence of work function of an ideal cylindrical single-crystal W with axial face orientation [111] upon the azimuthal angle.
Fig. 3. The dependence of emission current and work function of a single-crystal Mo with axial orientation [111] upon the azimuthal angle.

Fig. 4. The dependence of work function of epitaxial tungsten coating upon the azimuthal angle (initial sample).

Fig. 5. The dependence of work function of epitaxial tungsten coating upon the azimuthal angle (after 10000 hours of heating).
Introduction

In recent time, the interest to cesium-barium thermionic devices appears in the connection with the working out of high temperature converting systems [1-2]. In the devices of this type at considerably low pressures (1-20Pa) barium vapours provide optimum emitter work function, and cesium ions compensate negative spatial electron charge. Simultaneously, optimum collector operating temperature enhances on ~ 400^oK and the power output of the device increases [2,3]. Cesium and barium vapour injection into the operating volume of the thermionic device is carried out from the working media block (WMB). That's why the creation of compact WMB with stable cesium and barium vapour injection into the thermionic device for extended periods of time is rather actual problem.

2. Cesium-Barium WMB Design

At the present some variants of cesium-barium working media blocks have been worked out, based on laminated system of cesiated graphite and two-component gas-controlled heat pipe (HP).

One of the first variants of WMB [4] consisted of two reservoirs placed in different "points" of operating volume of TiC with various temperature. To prevent barium condensation into cesium reservoir special throttling device has been used. It made TiC design more complex in the whole not excluding barium recondensation completely [6]. In the papers cesiated graphite C_C (n=10,24) has been used as a cesium vapour source with the optimum operating temperature close to the optimum operating temperature of barium reservoir.

Nevertheless, uncontrolled changing of partial cesium pressure took place as a result of barium atoms instillation into laminated structure of cesiated graphite [7]. To avoid this phenomenon in Ref. 8 integrated cesium and barium vapour source has been suggested, in which interaction of barium atoms with cesiated graphite was practically excluded.

Block-diagram of mentioned WMB is given in Fig.1a, WMB is a metallic reservoir, having two interconnected chambers. In
one chamber metallic barium is placed, in the other - cylindrical sample of cesiated pyrographite, symmetry axis of which coincides with the crystalline axis C direction. Active element produces along C-axis external pressure, preventing barium atoms instillation into cesiated graphite. Transfer Measure Device (TMD) allows us to control anisotropic changing of cesiated pyrographite geometric dimensions along C-axis and - according to the known dependence of cesiated pyrographite considerable expansion along C-axis upon cesium concentration in it 2 - not only to define phase compound $C_nC_{s'}$ but to control it by means of saturation and desorption.

In Fig.1b you may see WMB variant with preliminary barium distillation. Barium is in separate vertically-set reservoir, connected with lower WMB chamber. Barium distillation is achieved in argon atmosphere by the method of heat pipe (HP). Rectified barium is condensed into lower WMB chamber, and heat pipe vapour channel is vacuum-tightly cut off from WMB operating volume. The main shortcoming of cited above variants lies in the control difficulty of cesiated pyrographite stochiometric compound. It complicates the optimization process of output parameters of thermionic device on vapour partial pressure of barium. This task can be solved with the help of introduction into existing constructive design an additional cesiated pyrographite block. Block-diagram of such a WMB variant is shown in Fig.1c. Cesiated pyrographite blocks are washers, inner cavities of which are interconnected by means of a steampipe. Cesium vapour supply into operating volume of the thermionic device is achieved from outer surface of the main cesiated pyrographite block, serving as a core. Cesium addition feeding of inner core surface is done in account of cesium atoms, getting it from an auxiliary cesiated pyrographite block. This allows us to vary partial cesium pressure in the operating volume of the thermionic device at constant partial barium vapour.

In considered above cesium-barium WMB vapour pressure control of working media is achieved by a thermal way. Meanwhile, this method is not always acceptable for pressure control of two-component working media, because, at first, it demands heat WMB mode stabilization with a rather high accuracy, and at second, in some cases temperature WMB agreement with device units is complicated to some extent. These problems can be solved with the help of the electromagnetic method. According to the values 9 the use of ponderomotor effect in two-component filler TiC in some definite conditions may lead to the changing of mixture concentration along converter electrodes. It allows to control stochiometric WMB compound in interelectrode spacing of the device.

Principal circuit of cesium-barium WMB with electromagnetic WMB pressure control is shown in Fig.1d. It includes cesium-barium source [8] and electromagnetic plasma diode placed between cesium-barium source and thermionic converter. The main plasma diode units are made of heat-proof metals; emitter 10 and collector 9 of cylindrical form and also metallo-ceramic seals II. With the help of external power supply 12 on the account of constant current flow in emitter the magnetic field $B$ appears and power supply 13 in its turn ignites low-voltage flashing discharge which produces cesium-barium plasma. Under the influence of electromagnetic forces along the plasma diode length WMB pressure gradient $\Delta P$ is produced depending on the current direction $J_r$. Pressure gradient may be directed either to the WMB reservoir side or to the thermionic device side. In the first case $\Delta P$ is conventionally considered to be nega-

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tive and in the second case - positive.

Such WMB performance suggests cesium and barium vapour supply through inter-electrode spacing of a plasma diode and partial pressure control of WMB components with the help of values and direction changings of electrical current $I_{r}$, flowing through the diode.

One of the cesium-barium WMB variants is two-component cesium-barium heat pipe [10]. As a heat carrier in the heat pipe we use barium-high-temperature filler component and as a noncondensed gas we use saturated vapour of low-temperature component-cesium. The scheme of working medium of this type is shown in Fig.1d. Vapour tapping from given construction is created by means of vapour drawing off sleeve, placed in the region of "barium-vapour-cesium vapour" distribution and having the opportunity to transfer along axial TT axis.

The control of partial pressure of low-temperature component and high-temperature component vapours in a given construction is carried out in the following way. As you know, special distribution of heat carrier vapour and buffer gas takes place in gas controlled heat pipe (GCHP) with the formation of "vapour-gas" division boundary. The length of division boundary usually is comparable with the heat pipe diameter. Typical distribution of partial vapour and gas pressures along HR is shown in Fig.1. In a given figure it is evident, that partial pressure in the region of division boundary is changing monotonously- vapour pressure in the direction of "cold" end pipe decreases, and gas pressure increases. In the described source construction we use, as it was mentioned above, vapour of low-temperature filler component. Moreover naturally, the temperature of "cold" end HP must be supported higher than the condensation temperature of the low-temperature filler component.

Therefore, implementation of steam bleeding from different "points" of HP it is possible to get vapour mixture of required stchiometry. Pull pressure control may be carried out on account of vapour changing of low-temperature component. Control of partial pressure ratio is realized by two ways -by transmission of vapour drawing off device along HP, or by heat power control, supplying to the HP evaporator.

Lower threshold of HP pressure, as you know, reaches the value of 60 Pa, whereas optimum pressure of cesium-barium mixture in thermionic invertors does not exceed 10-20 Pa. That's why, you need to realize running supply scheme with vapour throttling at the application of cesium-barium sources based on HP.

3. Kinetic and some thermodynamic characteristics of cesium-barium WMBs

The kinetics of reaching of pre-set working mode of thermionic device and the rate of switching to some other mode are determined by working medium pressure in the operating volume [2]. That's why partial pressure for cesium-barium WMB based on luminated graphite structures and metallic barium & at the given reservoir temperature is defined by phase compound of cesiated graphite $C_{n}C_{s}(n=8, 10,24,36)$ [2,11], and barium partial pressure will be equal to the value of the saturated vapour pressure of metallic barium at the given temperature [12].

To substantiate the optimum working modes of cesium-barium WMB with electromagnetic pressure control working medium the calculation theoretic evaluations of the process of concentration changing of cesium-barium mixture in thermionic device.
have been carried out. It was supported, that working medium components have similar temperatures and pressure drop of barium atoms along plasma diode channel is absent (\( \Delta \rho_{Ba} = 0 \)). The density magnitude of radial current along the diode has been defined from the expression [13]:

\[
\Delta \rho = 6.4 \times 10^{-3} \left( \frac{\rho_{Ba}}{\rho_{Ba} + 0} \right) \rho_{Ba} x \rho_{Ba} L^2
\]

(1)

where \( \Delta \rho \) - cesium pressure drop, Pa; \( \rho_{Ba} \) - radial current density, A.cm\(^{-2} \); \( L \) - plasma diode length, cm. From (1) it follows, that at \( L = 25 \) it is:

\[
\rho_{Ba} = \frac{1}{2} \sqrt{\Delta \rho}
\]

(2)

The value of \( \Delta \rho \) has been estimated on the thermodynamic data of metallic barium [12] and cesiated graphite [21].

The calculations on definition of the \( J_r \) and \( \Delta \rho \) parameters have been done for two devices.

1. for thermionic energy converter at barium and cesium partial pressure values equal to \( P_{Ba} = 13 \) Pa (\( T_{Ba} = 1018 \) K) and \( P_{Cs} = 13 \) Pa [14];

2. for plasma inverter at \( P_{Ba} = 6.6 \times 10^{-2} \) Pa (\( T_{Ba} = 785 \) K) and \( P_{Cs} = 1.3 \) Pa [15].

The results obtained are shown in the table, where the phase compounds of cesium-graphite system \( C_n Cs \) (\( n = 8, 10, 24, 36 \)). Values \( J_r \) and \( \Delta \rho \) with the notation of the polarity mark are represented.

<table>
<thead>
<tr>
<th>( C_n Cs )</th>
<th>( J_r, A cm^{-2} )</th>
<th>( \Delta \rho, ) Pa</th>
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<tr>
<td>( C_8 Cs )</td>
<td>1.3</td>
<td>0.6</td>
</tr>
<tr>
<td>( C_{10} Cs )</td>
<td>1.8</td>
<td>0.5</td>
</tr>
<tr>
<td>( C_{24} Cs )</td>
<td>18.6</td>
<td>2.5</td>
</tr>
<tr>
<td>( C_{36} Cs )</td>
<td>57.0</td>
<td>8.5</td>
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</table>

The analysis of these results shows that the most acceptable compounds of cesium-graphite system, used as a cesium source in the cesium-barium WMB for the inverter and TEC modes are the compounds of \( C_8 Cs \) and \( C_{36} Cs \).

Some experimental investigations have been carried out with the use of single component cesium source to ground working modes of magnetic plasma diode as a regulating element. These investigations have shown the opportunity to control volt-ampere characteristics at the constant WMB temperature. In Fig. 2 you may see volt-ampere characteristics for various pressure gradients at \( T_{WMB} = const \) in the wide range of \( \Delta \rho \), the values of which are in the range of 0-3 hPa and are estimated according to the formula (2). It should be noted, that the process of control is realized practically inertialessly.

4. Experimental investigations of cesium-barium WMB in the TEC structure

Some above-cited cesium-barium WMB have been tested in the structure of model cylindrical TEC. Thermal treatment methods of TEC electrodes and vacuum system as a whole, as well as getting data of envelope J-V device were standard [2]. On the base of envelope of J-V characteristics the dependence of the ratio of the output TEC power to the maximum output converter power (\( W/W_{max} \)) upon the temperature of various WMB (version 1a) which for \( T_{Ba} = 2100 \) K and \( T_{Cs} = 1300 \) K is shown in Fig. 3. Curve 1 corresponds to cesium WMB, curve 2- cesium-barium WMB of \( C_8 Cs + Ba \) compound, curve 3- \( C_{24} Cs + Ba \) and curve 4- \( C_8 Cs + Ba \). As you see, for \( C_8 Cs + Ba \) the optimum working reservoir temperature is 800 K, for \( C_{24} Cs + Ba \) -950 K, for \( C_8 Cs + Ba \) -1150 K, and \( W/W_{max} \) considerably weakly depends upon the working WMB temperature.

In Fig. 4 you see the dependence of ratio of the maximum electrical output power of the converter with cesium-barium filling (\( W_{max} \)) to the maximum power of cesium TEC (\( W_{max} \)) upon the collector temperature at \( T_{Ba} = 2000 \) K for the WMB having an additional cesiated graphite.
block (version 1). Curve 1 corresponds to \( C_2C_3 - \beta_3 \), when the additional block doesn't work; curve 2 -to \( C_2C_3 - \beta_3 \) with the additional block of \( C_3C_3 \) compound at the temperature of 800 K ; curve 3 -\( C_2C_3 - \beta_3 \) with the additional block of \( C_3C_3 \) compound at the temperature of 720 K . The character of the represented curves shows that the regulation of partial cesium pressure in the operating TEC volume at the constant barium vapour-pressure allows us to increase the output power of the device. Moreover, the higher working collector temperature is the more essential power growth is.

5. Conclusions

1. Cesium-barium WMB have been worked out based on the laminated structures of cesiated graphite and metallic barium; having different ways of partial cesium and barium pressure control.

2. The construction of cesium-barium WMB based on two-component heat pipe has been suggested where the working medium vapour mixture supply was carried out from the range of "vapour-cesium - vapour barium" distribution.

3. Some cesium-barium WMB variants with thermal and electromagnetic control of vapour mixture of the working medium have been tested as a part of experimental thermionic devices.

REFERENCES


4. Psarouthakis J. Thermionic energy converters, filled vapour mixture// FPTEE and TE. 1987, N9, p.79.


FIGURE CAPTIONS

Fig. 1. Cesium-barium sources.
1 - frame; 2 - silphon; 3 - cesiated pyrographite; 4 - steamline;
5 - valve; 6 - distillator; 7 - cavity; 8 - thermionic energy converter;
9, 10 - diode collector and emitter; 11 - insulator; 12, 13 - power supplies;
14 - wick.

Fig. 2. Volt-ampere characteristics of cesium $TE_1$ for various pressure gradients at $T_{wa}=const$ ($TE_1 = 1770 \text{K};
\ T_C = 870 \text{K}$).
(a) $T_{wa}=510 \text{K}$. Current density via diode $S_r$, A cm$^{-2}$: 1- 0; 2- 2,8; 3- 4,8;
b) $T_{wa}=523 \text{K}$; $S_r$, A cm$^{-2}$; 1$^1$- 0; 2$^1$ - 2,5; 3$^1$ - 4,2.

Fig. 3. Dependences $W/W_{\text{max}}$ upon the temperature sources at $TE_1 = 2100 \text{K}$ and $T_C = 1300 \text{K}$: 1 - Cs; 2 - Cs$\cdot$Ba; 3 - Cs$\cdot$Cs$\cdot$Ba; 4 - Cs$\cdot$Cs$\cdot$Ba$\cdot$Ba.

Fig. 4. Dependences $W_{\text{max}}/W_{\text{max}}$ upon the collector temperatures at $TE_1 = 2000 \text{K}$.
1 - Cs$\cdot$Cs$\cdot$Ba$\cdot$Ba (without complementary block); 2 - with complementary block of Cs$\cdot$Cs$\cdot$Ba$\cdot$Ba compound at $T = 800 \text{K}$; 3 - with complementary block Cs$\cdot$Cs$\cdot$Ba$\cdot$Ba at $T = 720 \text{K}$.
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