Use of thermochemical modelling for the analysis of energy extraction in a gas-core fission reactor

Citation for published version (APA):

DOI:
10.1016/0022-3115(89)90435-2

Document status and date:
Published: 01/01/1989

Document Version:
Publisher’s PDF, also known as Version of Record (includes final page, issue and volume numbers)

Please check the document version of this publication:
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USE OF THERMOCHEMICAL MODELLING FOR THE ANALYSIS OF ENERGY EXTRACTION IN A GAS-CORE FISSION REACTOR

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The chemical behaviour (dissociation and ionization processes) of the U–C–F–e system is analyzed in the temperature range of 2000 to 10000 K and for pressures of 1 and 100 bar. Increasing the amount of fluorine in the system, decreases the temperature at which UF₄ condenses on the wall because of formation of UF₅. At higher temperature UF₅ dissociates. At 10000 K the degree of ionization of uranium atoms is ~ 94 and 3% for pressures of 1 and 100 bar, respectively. A numerical equation of state, accounting for these dissociation and ionization processes is developed; it is given by

$$PV^{-a(P,T)} = b(P,T).$$

The influence of the dissociative heat on the total specific heat values of the mixture is investigated. For temperatures of ~ 2800 K (1 bar) and ~ 3800 K (100 bars), where UF₅ and CF₄ dissociate, the specific heat increases by a factor of five.

1. Introduction

The principles of operation of a gas-core fission reactor (GCFR) using uranium fluoride as a fuel have been discussed previously [1,2]. The gaseous mixture of uranium and carbon fluorides is in equilibrium with a graphite wall at a temperature of 2000 K and a pressure of about 2.5 MPa. In the bulk of the reactor the temperature is about 10000 K. Such a GCFR based on an energy production of 25 MWth (28 kg U, 30% enriched) can be incorporated in a fuel cycle in which the heavy actinides are recirculated and the fission products are extracted continuously. Corrosion problems in the fuel lines can be minimized by suitable adjustment of pressure, temperature and amount of fluorine [3]. Various possibilities of operation of gas-core fission reactors have been investigated previously in the USA [4] as well as in the USSR [5]. The main motivation was the applicability of a nuclear plasma reactor for propulsion in space.

In the USA the work was concentrated on:
- evaluation of a few types of UF₆ fueled reactors [6] for 2500 MWth energy production and gas temperatures of 1200 K, very much resembling the HTGRs;
- two types of pulsed reactors [7], fueled with UF₆ + He gas (90% ²³⁵U and most He gas).

They calculated the output of a mechanical piston engine (1 Hz) and of a GCFR steered by rotating neutron absorber discs. Both machines should stay below 2000 K at pressures of about 5 MPa to avoid dissociation of UF₆. The estimated output was about 5 MW electric. The work done by this group was primarily neutronics and reactor physics. Hardly any chemistry was done. Consideration of a gas-core fission reactor working at a high temperature of about 10000 K, automatically involves the possibility of energy extraction by means of magneto-inductive conversion of heat into electricity. This requires a high electron concentration. Consequently we investigated the degree of ionization of our U–C–F mixture. We also considered the possibility of increasing the electron concentration by means of addition of cesium. However, because of the high amount of free fluorine present in the mixture, F⁻ ions are formed and the electron concentration is hardly increased up to 6000 K.

In order to manipulate the gaseous mixture by means of a magnetic piston, as given in fig. 1, an equation of state and $c_p/c_v$ values, corresponding to the dissociating and ionizing UF₆–CF₄ mixture is required. A general equation of state for the pressure range of 1 to 100 bars and $c_p/c_v$ values (as derived from $P–V$ relations) was given previously [8]. In the present work we developed the equation of state for narrower pressure ranges, so that $c_p – c_v$ values can be calculated directly from it. All of the calculations were done on the basis of equilibrium compositions calculated with the computer pro-
2. Results of equilibrium calculations

As stated in the introduction the increase in the electron concentrations by means of addition of cesium is retarded by the free fluorine present in the mixture. Therefore we investigated the possibilities of operation with various amounts of fluorine in the gaseous mixture.

2.1. Fluorine potential

We define as fluorine potential the number of moles of fluorine, divided by the number of moles of actinides present in the gaseous mixture. This definition should not be confused with the usually used definition for “oxygen potential” \( G_{O_2} = -RT \ln p_{O_2} \). If the hold-up consists of \( p \) moles of \( U \) and \( q \) moles of \( F \), the fluorine potential \( FP \) will be given by the relation

\[
FP = \frac{q}{p}.
\]

However, inside the reactor, an equilibrium is established between the gaseous mixture and the graphite wall. Various uranium and carbon fluorine compounds are formed. This equilibrium composition is temperature and pressure dependent. Moreover, it depends on the fluorine potential of the gaseous mixture, as will be discussed below.

2.2. Equilibrium compositions

2.2.1. Boundary gas layer

Equilibrium compositions in the boundary gas layer were published previously [3]. In all of these calculations we considered that the graphite wall is in equilibrium with the gaseous mixture which it contains. We found that for low fluorine potentials, i.e. \( FP = 4.0 \) to
4.2 The gaseous mixture contains a mixture of lower valent \(C_mF_n\) compounds \((m = 1, 2; n \leq 4)\). However, above a fluorine potential of about 4.6 the main component is \(CF_4\); small amounts of \(CF_2\) and \(C_2F_2\) are present too. In fig. 2 we give the main components present in the equilibrium compositions as a function of the fluorine potential.

Clearly, condensation of \(UF_4\) is dependent on the fluorine potential of the gaseous mixture. We define as "condensation point" the temperature at which condensation of the corresponding species begins. In order to find this temperature we calculated equilibrium compositions in the temperature range of 2050–1850 K, at intervals of 5 K. In fig. 3 we give the condensation temperature of \(UF_4\) as a function of the fluorine potentials. This condensation temperature is an important factor in establishing the wall temperature of the reactor.

2.2.2. Bulk gas mixture

In fig. 4 we give the equilibrium compositions of the gaseous mixture \(U-C-F-e\) inside the reactor, in the temperature range of 2000 to 10000 K, for pressures of 1 and 100 bars, as they will be used for the development of the equation of state. The calculations were performed with the computer program "SOLGASMIX". We included 49 gaseous species, which are given in table 1. In all of our calculations we considered a constant molar overall mean ratio \(F : U : C = 4.568 : 0.800 : 0.202\). This specific ratio corresponds with chemical equilibrium with a graphite wall at 2000 K, 25 bars, which are the assumed operating conditions of our reactor.

From these equilibrium calculations we find the total number of moles resulting from the dissociation and ionization of one mole of initial gas mixture, with the composition given above. They are given in fig. 5. It can be seen that the total number of moles increases especially:

- for 1 bar: between 2400 and 3000 K, where the dissociation of \(UF_4\) and \(CF_4\) takes place, and between 4000 and 6000 K, where \(UF_4\) and lower valent uranium and carbon fluorides dissociate;
- for 100 bars: between 3000 and 4400 K, and between 5500 and 8500 K, because of the same reasons.

As shown in fig. 4a these dissociation processes result in a large amount of monoatomic fluorine gas.

2.3. Degree of ionization

Denoting by \((UF_n)_{\text{init}}\) the number of uranium fluorine species \(n\) which are initially present in the system and by

\[
\sum_{n=0}^{5} (UF_n(T))^+,
\]

the number of positive uranium fluorine molecular-ions which are present in the system at a temperature \(T\), we define the degree of ionization as the fraction of positive ions formed.

\[
\xi = \sum_{n=0}^{5} \frac{(UF_n(T))^+}{(UF_n)_{\text{init}}}. \tag{1}
\]

In table 2 we give values of \(\xi\) for pressures of 1 and 100 bars for temperatures of 4000 to 10000 K.
Fig. 4. (a) Equilibrium compositions of the gaseous system U–C–F–e ions. Major components. Number of moles available: F : U : C are 4.568 : 0.800 : 0.202. —— P = 1 bar; ———— P = 100 bar. (b) Equilibrium compositions of the gaseous system U–C–F–e ions. Main minor components. P = 10 bar. (c) Equilibrium compositions of the gaseous system U–C–F–e ions. Main minor components. P = 100 bar.
Fig. 5. Total number of moles as a function of temperature. Parameter: pressure in bar. Number of molecules available: \( F:U:C = 4.568:0.800:0.202 \). Corresponding electron concentrations are given in table 3. From fig. 4b and 4c it can be seen that up to 8000 K (1 bar) and up to temperatures above 10000 K (100 bar) the concentration of the free electrons is lower than the concentration of the positive ions. This is due to the formation of \( F^- \) ions. For simplicity we did not present the \( F^- \) concentration in fig. 4.

### Table 2

<table>
<thead>
<tr>
<th>Temperature (K)</th>
<th>Pressure 1 bar</th>
<th>Pressure 100 bar</th>
</tr>
</thead>
<tbody>
<tr>
<td>4000</td>
<td>9.08 x 10^{-4}</td>
<td>7.26 x 10^{-3}</td>
</tr>
<tr>
<td>5000</td>
<td>4.59 x 10^{-2}</td>
<td>9.61 x 10^{-4}</td>
</tr>
<tr>
<td>6000</td>
<td>0.275</td>
<td>0.012</td>
</tr>
<tr>
<td>7000</td>
<td>0.485</td>
<td>0.059</td>
</tr>
<tr>
<td>8000</td>
<td>0.728</td>
<td>0.143</td>
</tr>
<tr>
<td>9000</td>
<td>0.876</td>
<td>0.235</td>
</tr>
<tr>
<td>10000</td>
<td>0.943</td>
<td>0.334</td>
</tr>
</tbody>
</table>

### 2.4. Addition of cesium

In order to increase the electron concentration, we tried to add some cesium, to see if this electron-donor could contribute to the density of the free electrons. Equilibrium compositions were calculated by adding to the U–C–F system the following additional species: Cs(s, l), CsF(g), CsF(s, l), CsF(g), CsF, CsFu, CsF. However, up to 6000 K the free fluorine atoms bind practically all the electrons to form \( F^- \) ions. Decreasing the fluorine potential from 5.71 to 4.00 increases the electron concentration by about 20%. Above 6000 K the negative ions are thermally destroyed. Consequently, addition of a few percent of cesium has only a minor effect on the electron concentration in the boundary layer (see Discussion). This is shown in table 4. From a technological point of view it is unlikely that higher concentrations of Cs will be used; it is better avoid it completely.

### Table 3

<table>
<thead>
<tr>
<th>Temperature (K)</th>
<th>Pressure 1 bar</th>
<th>Pressure 100 bar</th>
</tr>
</thead>
<tbody>
<tr>
<td>4000</td>
<td>0.014</td>
<td>0.29 x 10^{-3}</td>
</tr>
<tr>
<td>5000</td>
<td>1.01</td>
<td>0.16</td>
</tr>
<tr>
<td>6000</td>
<td>3.84</td>
<td>7.61</td>
</tr>
<tr>
<td>7000</td>
<td>6.72</td>
<td>55.82</td>
</tr>
<tr>
<td>8000</td>
<td>8.56</td>
<td>145.6</td>
</tr>
<tr>
<td>9000</td>
<td>9.01</td>
<td>230.8</td>
</tr>
<tr>
<td>10000</td>
<td>8.76</td>
<td>308.4</td>
</tr>
</tbody>
</table>

### Table 4

<table>
<thead>
<tr>
<th>Temperature (K)</th>
<th>Species</th>
<th>Cs concentration 1 mol%</th>
<th>Cs concentration 10 mol%</th>
</tr>
</thead>
<tbody>
<tr>
<td>2500</td>
<td>( \Sigma UF_n^+ )</td>
<td>0</td>
<td>&lt; 0.0001</td>
</tr>
<tr>
<td></td>
<td>Cs(^+)</td>
<td>0.029</td>
<td>0.09</td>
</tr>
<tr>
<td></td>
<td>F(^-)</td>
<td>0.029</td>
<td>0.09</td>
</tr>
<tr>
<td></td>
<td>( \bar{\varepsilon} )</td>
<td>&lt; 0.001</td>
<td>0.0002</td>
</tr>
<tr>
<td>6000</td>
<td>( \Sigma UF_n^+ )</td>
<td>13.1</td>
<td>6</td>
</tr>
<tr>
<td></td>
<td>Cs(^+)</td>
<td>5.5</td>
<td>37</td>
</tr>
<tr>
<td></td>
<td>F(^-)</td>
<td>10.0</td>
<td>24</td>
</tr>
<tr>
<td></td>
<td>( \bar{\varepsilon} )</td>
<td>8.6</td>
<td>19</td>
</tr>
</tbody>
</table>
3. A numerical equation of state

The total number of moles in fig. 5 reflects the degree of dissociation and ionization at various temperatures and pressures. This total number of moles \( n(P, T) \) implies a volume \( V \), to be calculated by

\[
P V = n(P, T)RT.
\]

In fig. 6 we present a \( P-V \) diagram as calculated with "SOLGASMIX" for one mole of initial gas mixture with an overall molar ratio of \( F:U:C = 4.568:0.800:0.202 \). In order to get a mathematical expression for the equation of state, we investigated the expression:

\[
\log P = a \log V + b
\]

or

\[
P V^{-a(T)} = 10^{b(T)}.
\]

The coefficients \( a \) and \( b \) are temperature dependent. In order to find the numerical values of these coefficients for every temperature, we have introduced numerical values of \( P \) and \( V \) (from fig. 6) into a "least square fit" computer program [9]. In a previous work [8] we used the \( P \) and \( V \) data in 20 pressure steps in the range of 1 to 100 bars. In that case the equation of state reproduced the input data from fig. 6 within 7%. However, for the calculation of specific heat values (which will be discussed below) more accurate values of the coefficients \( a(T) \) and \( b(T) \) were required. We calculated \( a \) and \( b \) in the temperature range of 2000 to 10000 K, applying small temperature intervals of 200 K. For every temperature we considered pressures of 1, 20 and 100 bars, with pressure ranges of 10% (i.e. 0.9–1.1, 18–22, 90–110 bars), each range divided into 8 small steps. In this case reproducibility of the \( P-V \) data was within 0.5%. Values of the coefficients \( a \) and \( b \) for three pressures are given in fig. 7a and 7b.

From a point of view of physical interpretation the coefficient "\( a \)" corresponds to the increase in pressure due to compression. For an ideal gas \( a = -1 \). Because of recombination processes of dissociated molecules at higher pressures the increase in pressure is less: \( |a| < 1 \) as can be seen from fig. 7a. With increasing pressure the extreme values of "\( a \)" shift to higher temperatures, corresponding to the dissociation processes given in fig. 4 and 5. The coefficient "\( b \)" corresponds to the extrapolation of the straight line given by eq. (3). Actually, it is the logarithm of the pressure which would be exerted by the mixture if it expands from a volume \( V(P, T) \) to a volume of 1 m\(^3\) at the temperature \( T \). Considering eq. (2) and the slope and intercept of the straight line given by eq. (3), values of \( a(P, T) \) and \( b(P, T) \) can be calculated according to the equations

\[
a = \frac{\log(P_1/P_2)}{\log(n(P_1, T)/P_2/n(P_2, T)P_1)},
\]

\[
b = \{\log P_2 \log n(P_1, T) - \log P_1 \log n(P_2, T) \}
+ (\log RT)(\log P_2/P_1) \}
\{\log[n(P_1, T) /n(P_2, T)] + \log P_2/P_1 \}^{-1},
\]

where \( P_1 \) and \( P_2 \) are two close marges of \( P \) (i.e. 0.90\( P \) and 1.1\( P \)). They are chosen arbitrarily and depend on the accuracy and the range of applicability required: the values of \( n(P_1, T) \) and \( n(P_2, T) \) can be found from two corresponding equilibrium calculations.
For constant "n" the value of \( b \) reduces to
\[
b = \log nRT.
\]

4. Specific heats of the system U-C-F-

We denote by:
- \( c_p \) = the molar specific heat of the gaseous mixture at a pressure \( P \) and temperature \( T \);
- \( c_v \) = the molar specific heat of the gaseous mixture at a volume \( V \) and temperature \( T \).

In order to calculate the ratio \( c_p/c_v \) we have to:
1. calculate the mean values of \( c_p \) for the gaseous mixture, corresponding to the various equilibrium compositions;
2. calculate the difference \( c_p - c_v \).

4.1. Specific heats

The specific heats of our dissociating gas mixture can be calculated from the equilibrium compositions and the individual molar enthalpies of each species \( i \), according to the relation
\[
c_p(P, T) = \sum_{i=1}^{N} n_i(P, T_2) H_i(T_2) - \sum_{i=1}^{N} n_i(P, T_1) H_i(T_1)
\]
\[
\frac{1}{[\bar{n}_{\text{tot}}(P)](T_2 - T_1)}
\]
(7)

where
- \( n_i \) = number of moles of species \( i \),
- \( H_i \) = molar enthalpy of species \( i \),
- \( \bar{n}_{\text{tot}}(P) = [n_{\text{tot}}(P, T_2) + n_{\text{tot}}(P, T_1)]/2. \)
Values obtained for pressures of 1, 25 and 100 bars are given in fig. 8. We give the total specific heat, as calculated by eq. (7), and the mean molar specific heat calculated from the mole fractions and the $c_i^l$ values of the individual species [i.e. $i = 1.49$ (see table 1)]. Clearly, the differences are due to the dissociation processes involved.

4.2. Calculation of $c_p - c_v$ values

Values of the difference $c_p - c_v$ can be calculated according to the relation [10]

$$c_p - c_v = \frac{1}{\eta(P, T)} TV\alpha^2/\beta,$$  \hspace{1cm} (8)

where $\alpha$ is the isobaric expansion coefficient, given by

$$\alpha = \frac{1}{V} \left( \frac{\partial V}{\partial T} \right)_P$$ \hspace{1cm} (9)

and $\beta$ is the isothermal compressibility coefficient given by

$$\beta = -\frac{1}{V} \left( \frac{\partial V}{\partial P} \right)_T.$$  \hspace{1cm} (10)

Considering eq. (4) for our system one obtains

$$\alpha = -a^{-2} (\ln P - b \ln 10) \frac{da}{dT} - a^{-1} \ln 10 \frac{db}{dT}$$ \hspace{1cm} (11)

and

$$\beta = -a^{-1} P^{-1}.$$  \hspace{1cm} (12)

The values of $da/dT$ and $db/dT$ are calculated for temperature intervals of 200 K at the mean pressure of 1 and 100 bar. In fig. 9 we give values of $c_p - c_v$ as calculated by eqs. (8), (11) and (12). For purposes of comparison we also give values calculated by the relationship:

$$c_p - c_v = -\frac{T(\partial V/\partial T)_P^2}{n(P, T)(\partial V/\partial P)_T}.$$ \hspace{1cm} (13)

5. Discussion and conclusions

5.1. Influence of dissociation and ionization on energy extraction

The concentration of the free electrons as obtained for temperatures above 6000–7000 K is suitable for the manipulation of the plasma by means of a magnetic piston as given in fig. 1 [11]. Nevertheless, the “cool” gas layer between the wall and the position where 6000 K is reached will act as a leak for the magnetic piston. There the electron concentration is too low. The size of this leak depends on the temperature profile in the reactor. Van Dam and Hoogenboom calculated the temperature profile neglecting the dissociation and ionization processes [12]. However, because of these processes the composition of the gas-mixture as a function of the distance to the wall change [2]. This causes a decrease in the temperature gradient near the wall, and a decrease of the temperature maximum in the bulk of the gas. Details will be published in the near future.
In order to analyze the thermodynamic performance of a pumped gas-core reactor, one has to calculate a \((P, V)\) loop integral, based on the first law of thermodynamics

\[
Q = \oint dU + \oint P \, dV, \tag{15}
\]

where: \(Q = Q_{\text{nuclear}} - Q_{\text{loss}}\). Because of the complexity of our equation of state (resulting from the dissociation processes) one has to calculate this integral by using values of "\(a\)" and "\(b\)" for short intervals of temperature and pressure, i.e. a step by step calculation has to be used. The values of \(c_P - c_v\) are given in ref. [8]. They are from \(\sim 1.1\) at 2000 K to \(\sim 1.4-1.5\) at temperatures above 8000 K, depending on the pressure considered.

**Appendix. Ionization due to nuclear reactions in a GCFR**

Schematized the physical situation is as follows:
- a nuclear-critical system with a stationary energy production of 50 MWth in a volume of 10 m\(^3\), with 20 m\(^2\) wall area;
- the mean working pressure be 65 bar, and the temperature of the bulk of the gas 10000 K;
- the top layer of the graphite wall is at 1800 K and with a temperature gradient in front of it of \(3 \times 10^6\) K/m.

We want to discuss radiative ionization in the bulk and near the wall. The bounding gas layer is assumed to have a mean gas temperature of 3500 K, with a size of a few millimeters.

- The mean particle density near the wall is \(15 \times 10^{19}\) cm\(^{-3}\) (22 mg/cm\(^3\)) and in the bulk \(5 \times 10^{19}\) cm\(^{-3}\) (4 mg/cm\(^3\)). Due to the temperature difference, dissociation and ionization, the density of U-nuclei is 25 times larger near the wall than in the bulk. Therefore also the energy production per cm\(^3\) in the bulk is 5 J/cm\(^3\) s but 125 J/cm\(^3\) s near the wall.

- The mean stopping power for fission products between 100 and 10 MeV, if passing through fluorine gas is about 37 MeV/mg cm\(^2\) [13]. This gives a track-length of 0.7 cm in the bulk and 0.1 cm near the wall. Each fission event produces 3.2 \(\times 10^{-11}\) J.

- If charged particles at high velocity pass through a gas the Coulombic (electronic) stopping process creates one ion pair per 30 eV energy loss [14]. Therefore each fission track has \(3 \times 10^6\) ion pairs. The production of ion pairs in the bulk is \(9 \times 10^{17}\) cm\(^{-3}\) s but near the wall \(200 \times 10^{17}\) cm\(^{-3}\) s.

- The loss of ion pairs is mainly due to recombination according to the chemical rate equation

\[
\frac{dn^+}{dt} = -n^+ \sigma_{\text{rec}} \bar{v}_{\text{ion}}.
\]
where \( n^+ \) and \( n^- \) are the ion densities \((n^+ = n^- = n_i)\), \( \sigma_{\text{rec}} \) is the cross section for recombination in \( \text{cm}^2/\text{ion} \), and \( \bar{v}_{\text{therm}} \) is the mean thermal speed of the fastest particle.

**Bulk losses**

Loss processes at 10000 K are due to recombination of free electrons with positive atomic ions. Such cross sections are always of the order of \( 10^{-17} \text{ cm}^2/\text{ion} \). The mean thermal speed of the electrons at 10000 K is \( 6 \times 10^7 \text{ cm/s} \).

**In the stationary state** we have

\[
9 \times 10^{17} = n_i^2 \times 10^{-17} \times 6 \times 10^7 \rightarrow n_i = 4 \times 10^{13} \text{ cm}^{-3},
\]

which corresponds to a degree of ionization due to radiation of \((n_i/n_{\text{neutral}}) \approx 10^{-6}\). As the thermal degree of ionization is about \( 10^{-1}\) this means that in the bulk of the GCFR, at 10000 K, radiative ionization can be neglected.

**Losses in the boundary gas-layer**

At 3500 K, 65 bar, the concentration of free electrons is negligible in a gas mixture of uranium- and carbon fluorides (electron negative gases). Recombinative loss processes take place between heavy molecular ions \((\sigma_{\text{rec}} \approx 10^{-15} \text{ cm}^2/\text{ion})\). The ion transport can be kinetic \((\bar{v}_{\text{therm}} = 10^5 \text{ cm/s})\), or diffusive \((\vec{v} = 10^{-1} \text{ cm/s})\). In the first case we find \( n_i = 4 \times 10^{14} \text{ cm}^{-3} \), and in the last case \( n_i = 4 \times 10^{17} \text{ cm}^{-3} \), which is an upper limit. In comparison with the thermal equilibrium ion concentration, which is of the order of \( 10^{15} \text{ cm}^{-3}\), diffusive recombination in the dense and "cool" wall layer shows a dominating influence of radiation (factor 100 more ions). But the importance for magneto hydrodynamic processes is negligible. The concentration of free electrons is negligible and therefore the electronic conductivity of the boundary gas layer is very low.

**Acknowledgements**

This work is part of the research program of the Stichting voor Fundamenteel Onderzoek der Materie (Foundation for the Fundamental Research on Matter) and was made possible by financial support from the Nederlandse Organisatie voor Wetenschappelijk Onderzoek (Netherlands Organization for the Advancement of Research) and the Stichting voor Technische Wetenschappen (Foundation for Technical Science).

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