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M.J. de Graaf, R. Severens, M.J.F. van de Sande, M.C.M. van de Sanden and D.C. Schram

Eindhoven University of Technology, Department of Physics, P.O. Box 513, 5600 MB Eindhoven, The Netherlands

H.J.M. Meijers and H. Kars

Rijksdienst voor Oudheidkundig Bodemonderzoek, Kerkstraat 1, 3811 CV Amersfoort, The Netherlands

For the development of H^+ and H^0 beams a new method has been developed based on the expansion of a cascade arc plasma. A partial aim was to develop an intense beam of atomic hydrogen. The result was a 100 A equivalent hydrogen atom beam with an energy conversion efficiency of typically 30–40%. The resulting hydrogen plasma has also been used to do preliminary experiments on restoration treatment of archeological artefacts according to the method of Vepřek et al. The present high density atom rich plasma beam with < 0.4 eV energies and temperatures proved to be effective in the treatment of these artefacts. The treated artefacts showed good erosion resistance compared to untreated as well as conventionally treated samples, notwithstanding the short treatment time of 20 minutes at a temperature of 400°C and the provisional character of the trial experiment.

For several applications in chemical and electronic industries the development of hydrogen atom beams of high intensity is very important. In other applications hydrogen ion beams are used. High beam intensities require a large production rate. Such a large production rate is achieved in employing thermal plasmas as has been shown in the work of Kroesen et al. [1] and Beulens et al. [2] for plasma deposition purposes. In this work a new high intensity particle source has been developed. The same principle was applied to achieve a high intensity hydrogen atom beam. The plasma source is a flowing thermal plasma in a cascade arc, with (three) cathodes at the upstream side, a stack of electrically floating copper cascade plates and an anode nozzle. The latter serves also as the connection to the vacuum-pumped treatment chamber. In fig. 1 the source is sketched. For diagnostic purposes the pressure and voltage drop at each plate can be monitored, as well as the heat transfer from the plasma to the cascade plates walls. As also the input energy is known from the voltage drops and the current (in the presented experiments 35–50 A) the efficiency of the energy conversion to the outflowing plasma can be obtained. As this energy is mainly carried in the chemical dissociation of the hydrogen gas, also the dissociation degree can be determined. In fig. 2 the dissociation degree is given as a function of the current for 3, 6 and 9 slm H_2 flow and for a three plate arc. It is evident that the dissociation degree is high; with the quoted flow rates the emanent hydrogen atom flux can increase to values over 100 A equivalent ($\sim 10^{21}$ H^0/s). This is shown in fig. 3 in which the H^0 atom flow is expressed in slm (1 slm is equivalent to 4.2×10^{21} H^0 atoms/s or 26 A equivalent). The ion density in argon is between 5–10%; in hydrogen it is not measured as yet but can be estimated to be between 1–5%. Through the anode nozzle the partially ionized gas expands in the pumped vacuum vessel. Depending on gas composition and ambient pressure the plasma first expands supersonically, then shocks after some cm and for the major part it expands subsonically. At pressure above 10 Pa the ionization is readily lost by a sequence of charge exchange and dissociative recombination

$$H^+ + H_2^{v \geq 4} \rightarrow H_2^+ + H, \quad (1)$$

where the molecular ion can be lost by

$$H_2^+ + e \rightarrow H + H, \quad (2)$$

or by the reactions

$$H_2^+ + H_2 \rightarrow H_3^+ + H, \quad (3)$$

$$H_3^+ + e \rightarrow H_2 + H, \quad (4)$$

$$H_3^+ + e \rightarrow H + H + H. \quad (5)$$

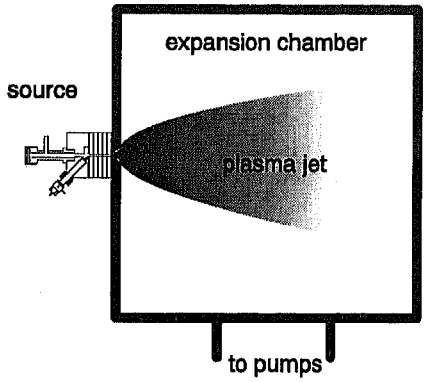

Fig. 1. The experimental setup: cascade arc thermal plasma source expanding into a treatment vessel.

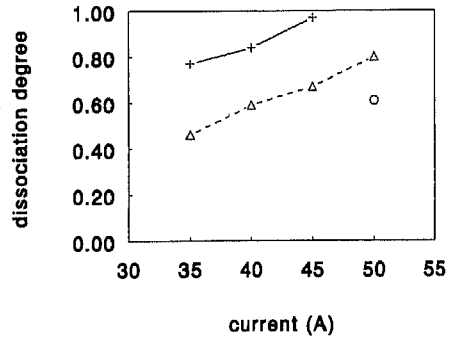

Fig. 2. Dissociation degree in the source versus current. The +, △ and ○ stand for a gas flow of 3, 6 and 9 slm, respectively.

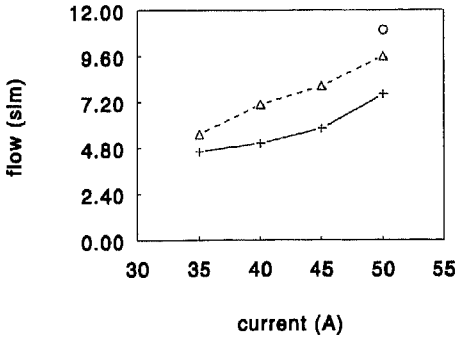

Fig. 3. Atomic hydrogen flow emanating from the source versus current. The +, △ and ○ stand for a gas flow of 3, 6 and 9 slm, respectively.

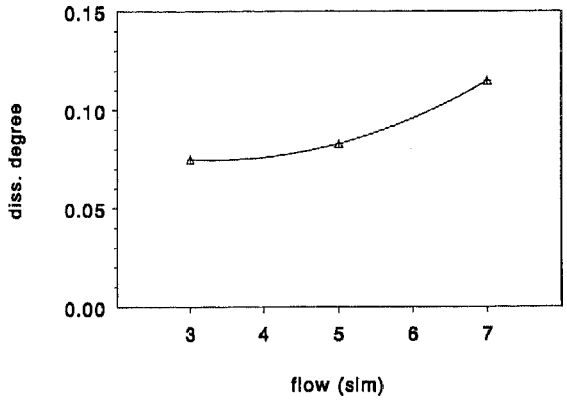

Fig. 4. Dissociation degree in the treatment vessel 25 cm downstream the source.

Apparently, for ion source application low pressure operation combined with magnetic confinement is essential. In this paper we will focus on the use as a neutral particle source and concentrate on operation at high pressure (> 10 Pa).

The dissociation degree in the treatment vessel has been measured with RF excited active actinometry. *Active* actinometry is needed since the essential electronic excitation is absent because of the low electron temperature (between 0.15 and 0.4 eV). Without the RF excitation all light emitted by the plasma results from dissociative reactions as discussed above, which usually end in excited fragments. Hence passive line-emission spectroscopy can only be used to monitor the recombination reactions described above. In the active actinometry method the electrons are locally heated by an RF probe exciter with typical powers of 10 W. As a result H Balmer lines and the H_2 Fulcher rovibrational band are excited and can be spectroscopically observed. After calibration line ratios can be used to give the dissociation degree in the plasma. In fig. 4 the result is shown. Two striking differences are apparent: first the dissociation degree is much smaller than in the emanating source plasma, second the dissociation degree increases rather than decreases with increasing flow. These results point to the influence of wall association and of recirculation. Actually, this influence is to be expected since the dwelling time, typical 0.1 s, is much longer than the transit time through the machine, less than 1 ms. Also wall association,

$$H + H(\text{absorped}) \xrightarrow{\text{wall}} H_2(\text{desorped}), \qquad (6)$$

is known to be effective and to result in rovibrational excited hydrogen molecules. Thus it also forms a source

for the $H_2^{v\geq 4}$ molecules which play an essential rule in the destruction of the ion fraction as described in reaction (1). Still the incident H atom flux is very large compared to more classical plasma physics techniques.

Therefore it was investigated whether a considerable reduction of the passivation treatment time of archeological artefacts could be achieved. The artefacts chosen for this study are selected from material of Roman age and derive from sandy soils with strong oxiding preservation conditions. Similar finds were treated earlier with the conventional method, i.e. washing in a natriumhydroxide/natriumsulfite solution, followed by an impregnation with epoxy resin. It turned out, however, that the process of corrosion of these heavily oxided objects could not be stopped by using this method. To investigate the plasma jet method a trial experiment was set up in which samples were treated according to the method earlier described by Vepřek, Patscheider and Elmer [3–5]. In the trial experiments a treatment time of 20 minutes was chosen. During the experiments the temperature was monitored through a NaCl window by an IR pyrometer. The temperature was not homogeneous over the objects, but never exceeded 400°C. After the treatment it showed that the outer rim of corrosion products, which in fact for the greater part is composed of quartz sand and clay minerals, was very easy to remove and easy to separate from the so-called patina underlayer underneath that has to be saved. Another main result was that surface details were kept better than using the conventional treatment of grinding. The artefacts discolour black during the treatment; however, from esthetical point of view of the conservator this is not considered a disadvantage. After the hydrogen treatment two methods of conservation have been experimentally applied: impregnation with molten microcristalline wax at a temperature of around 100°C and impregnation with epoxy resin (Araldite AY103-AH956) at around 80°C. The surface of the objects became porous as a result of the hydrogen plasma treatment, which was facilitating for the impregnation. The treated objects were stored under conditions similar to those generally present in Dutch museums and store rooms, with – varying with the seasons – a relative humidity ranging from 40 to more than 60%. Inspection of the artefacts after a period of one year showed no significant corrosion, contrary to similar objects treated with the conventional method. Best results were obtained with the Araldite impregnation after plasma treatment. Resuming, the results are encouraging: despite the short treatment time, 20 minutes whereas in litterature 2 hours is common, and the provisional character of the experiments the typical improvements disclosed in earlier publications were observed: the cleaning of the artefacts was strongly facilitated and the treated objects show no corrosion contrary to pieces treated with the conventional methods. One aspect of the present method (apart from the high flux) could also be important in this respect: the low ion and neutral energies (< 0.4 eV). In classical plasma methods a self-bias is created because of the difference in diffusion behaviour of the light electrons and the heavy ions. This self bias potential accelerates the ions incident on the substrate. In the present method the few ions which remain obtain only sub eV energies and the major part of the flux consists of low energy atoms. This leads to a very gentle, but intense particle flux. This feature is the consequence of using a *passive* plasma for treatment and thus of the separation of plasma production and plasma treatment.

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