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The role of vibrationally excited molecules in a recombining hydrogen plasma

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1 Experimental

The experiments presented in this paper are performed on a plasma apparatus consisting of a cascade arc expanding into a vacuum chamber. This type of setup is researched as a source for a:C-H deposition [1], a:Si-H deposition [2], and for hydrogen radical and ion production [3]. Main characteristics of the cascade arc are a high pressure (0.1 to 1 bar) and a high power density, up to 50 GWatt m^{-3} . Due to the high density mainly atomic ions will be produced. The plasma generated in the arc can expand into the vacuum chamber through a nozzle in the anode plate. The vacuum chamber is operated at a pressure of 0.1 to 20 mbar. Neutral density in this section is typically $5 \cdot 10^{21} m^{-3}$, the plasma temperature (gas and electron temperature are not too different) is in argon 3000 K and in hydrogen 1500 to 2500 K.

The measurements are performed with a Langmuir double probe system. The probe system was moveable over an area from 20 to 30 cm downstream the nozzle and 5 cm perpendicular to the plasma axis. Thus radial electron density scans were made at three axial positions. By integrating these radial electron densities over the plasma cross section, a 1-dimensional measure $n(x)$ is obtained. The plasma velocity over the measured area is around 600 m/sec and varies only slightly [4], so this measure is directly proportional to the electron density. A time scale of the recombination processes can be determined: 20 to 30 cm at a velocity of 600 m/s means a time of flight in the order of 0.3 msec.

2 Results and discussion

Figure 2 shows radial profiles at 20, 25 and 30 cm for a pure argon plasma, a 10 difference between the three is the absolute level of the density. The pure argon case has a maximum of approximately $3 \cdot 10^{19} m^{-3}$, whereas in the hydrogen containing plasmas a three respectively four orders of magnitude smaller value is found. A second important observation is that, although the absolute densities in the presence of hydrogen are far smaller, still recombination goes much faster than in hydrogen. This becomes more evident if the evolution of the electron flux along the plasma axis is determined as described above (Figure 2). The anomalous fast recombination in hydrogen can not be explained with two- or three particle recombination. Molecular reactions are fast enough to give the observed ionization loss; however, the cascade arc source is known to contain only atomic ions. Therefore a channel is needed to convert atomic ions to molecular ions. This can happen by the reaction



The vibrational excitation of the hydrogen molecule is vital: otherwise the reaction is endothermic and rate coefficients are very small. Niedner et al [[5]] have shown that this charge transfer is a two step mechanism: a vibrational excitation to $v \geq 4$ followed by a resonant, exothermic charge transfer. In a plasma, a fraction of the molecules will already be in a vibrational state $v \geq 4$. Cross sections for the second step, the exothermic charge transfer, are not available. To address the potential contribution of the CT channel to the recombination, we assumed a typical charge transfer cross section of $10^{-18} m^2$. Now a 'time scale analysis' of recombination processes can be made. Figure 2 shows the evolution of the electron density, starting from $10^{20} m^{-3}$ as a result of three particle recombination and of the proposed charge transfer channel. It shows that at the time scale of 0.3 msec in argon three particle recombination can lead to the observed densities, whereas in hydrogen the molecular channel is needed.

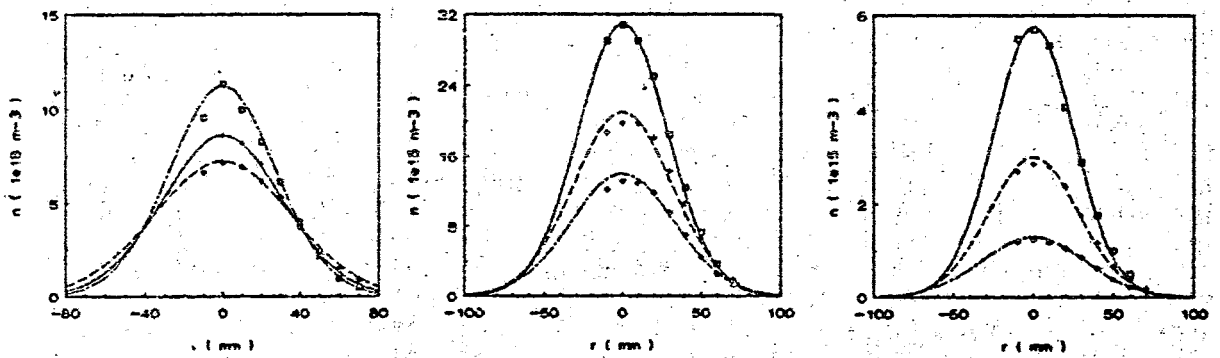
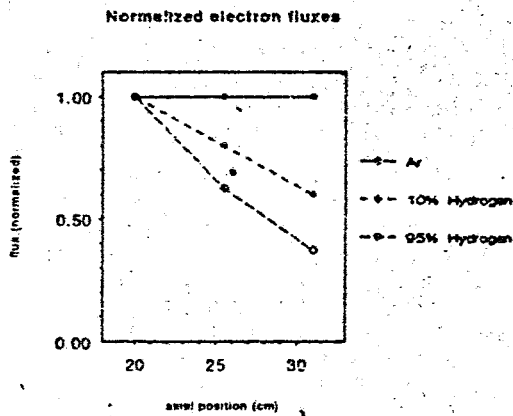


Figure 1: Measured radial density profiles for argon, 10% and 95% hydrogen.



Recombination in hydrogen plasma

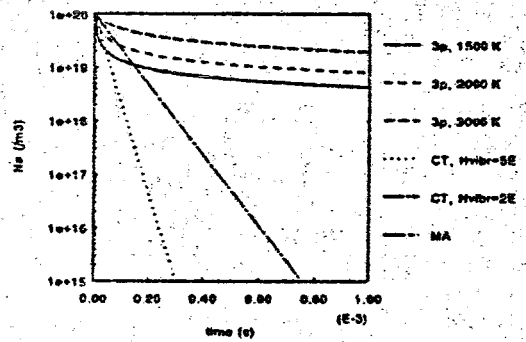


Figure 2: Ion flux at 3 positions along the plasma Figure 3: Recombination processes starting from 10^{20} m^{-3} .

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