Transport of H and H2 in an expanding hydrogen plasma

P.Vankan, S.Mazouffre, R.Engeln, and D.C.Schram
Department of Applied Physics, Eindhoven University of Technology
P.O. Box 513, 5600 MB Eindhoven, The Netherlands

Introduction
Hydrogen plasma expansions can be found in a wide variety of objects, ranging from large scale, e.g. solar flares, to very small scale, e.g. laser spots [1]. At an intermediate scale many hydrogen plasmas concern the future of energy production like fusion plasmas and expanding plasmas used for thin layer deposition, e.g. hydrogenated amorphous silicon layers (a-Si:H) for the next generation solar cells [2].
In all of these examples the particle transport is a main issue. For instance in a-Si:H deposition hydrogen radicals are essential for the growth and quality of the layers. The location where the radicals are produced is different from the reaction location and different from the deposition location. Therefore it is of importance to understand how the radicals are transported between the different sites. In the experiments reported here the transport of hydrogen atoms and molecules has been investigated by measuring the density, temperature, velocity and velocity distribution in a pure hydrogen expanding plasma jet as a function of the axial position.

Experimental arrangement
A cascaded arc [3] operated at a current of 55 A and a voltage of 150 V is used as a plasma source. The plasma is created from a hydrogen flow of 3.5 slm and expands freely into a vacuum chamber. The stagnation pressure inside the arc is 0.14 bar and the background pressure in the vessel is either 20 Pa or 100 Pa. The arc is mounted on a movable arm enabling spatially resolved measurements.
The hydrogen atoms have been probed using the two-photon absorption Laser induced fluorescence technique (TALIF). Using this technique the hydrogen atom densities, temperatures, velocities and velocity distributions have been determined. The details of the technique are published elsewhere in this proceedings [4].
The hydrogen molecule densities have been measured using Rayleigh scattering. Since the H atom densities are known the H2 densities can be determined from the Rayleigh signal. The scattering experiment has been performed using the 205 nm laser beam from the TALIF setup. Both techniques can be used simultaneously.

H2 expansion
In Figure 1 the H2 density has been plotted as a function of the axial position along the jet axis. In the supersonic part the density decreases until the stationary shock wave is reached. There the density increases because of forward flux conservation. In the background the density increases slightly, since the plasma flows at constant pressure. The H2 density profile is in agreement with the free jet flow theory. The plasma flow has

Figure 1: The axial density profile of molecular hydrogen measured by Rayleigh scattering.
been modeled using a hydrodynamic code [5]. The model shows a good qualitative agreement with the measured H₂ densities.

**H atom expansion**

In Figure 2 the H atom axial temperature and velocity profile have been plotted. The different regions in the expansion have been determined from these profiles. The supersonic region ranges until the temperature increases and the velocity decreases. At the end of the shock wave, where the Mach disk is located, the velocity becomes smaller than the sound speed and the temperature starts to decrease because of heat transfer to the wall.

The H atom density profile has been plotted in Figure 3. In the supersonic part the expected density decay can be seen. Contrary to molecular hydrogen the H atom density profile does not exhibit a density jump at 20 Pa, and at 100 Pa the density jump is far too small. This directly means that the H atom forward flux is not conserved, i.e. the hydrogen atoms are lost from the core of the expansion. This loss is not due to volume recombination since the densities and temperatures in our plasma are far too low for the 3-body recombination process to be efficient. The only other way the radicals can be lost is due to diffusion out of the plasma jet [6]. The hydrogen radicals have a very high probability to recombine on the vessel walls. Combined with the very long residence time in the background this leads to a very large chance that the radicals are lost on the walls. The H atom partial pressure in the background is therefore very low inducing large gradients between the core of the jet and its surroundings. Those gradients are responsible for the diffusion out of the plasma.

This effect is a very general effect. It applies to all radicals having high wall recombination probabilities. This has far-reaching consequences. The radicals are the reactive particles in the plasma, i.e. the particles needed for the chemical processes. Due to the outward diffusion most of the radicals are lost and therefore the chemical potential of the plasma is

**Figure 2:** H atom axial temperature and velocity as a function of the axial position along the jet centerline at 20 Pa and 100 Pa background pressure. The sound velocity at 20 Pa calculated from the temperature is depicted by a solid line.

**Figure 3:** The H atom density profile at 20 and 100 Pa background pressure.
reduced drastically. In the design of vessels used for radical expansions one has to take the loss mechanism into account. The wall recombination probability can for example be reduced by using materials at which radicals show a low association degree. If acceptable for the process, higher background pressures can be used to improve the radical confinement.

**Velocity Distribution Function**

Due to the large axial velocity of the light H-atoms (and thus the large Doppler shift) and the relatively high temperature in the plasma it is possible to determine the velocity distribution function (VDF) from the spectral profile. A Gaussian spectral profile means a Gaussian VDF, determining the flow as Maxwellian, i.e. one mean velocity and one mean temperature. The flow is then in LTE. In Figure 4 H-atom VDF’s have been plotted at a background pressure of 20 Pa at different axial positions along the expansion axis. The upper left plot is the VDF in the supersonic region. The VDF is Gaussian pointing to LTE for the H atoms in this region.

When moving along the expansion into the shock region (from z=10 to 50mm) we clearly see a departure from LTE. It can be explained by the essential property of shock waves: the transformation of a supersonic flow into a subsonic flow. During this process the supersonic VDF in front of the shock has to be adapted to the subsonic VDF behind the shock. The H atom VDF has been decomposed into two separate Gaussian VDFs, matching the conditions before and after the shock.

Moving through the shock we can see that the density of the supersonic component is decreased to zero and that the subsonic component appears and the density is increased until the complete flow is subsonic as depicted in the lower right hand side plot. The bi-modal approximation of the VDF in a shock wave has been studied theoretically by Mott-Smitt [5]
Conclusions
The transport of hydrogen atoms and molecules has been investigated using the two-photon absorption Laser induced fluorescence and Raleigh scattering techniques. The results show that radicals can easily escape the plasma jet under the right circumstances. This has to be taken into account when the chemical potential of the plasma is needed. In the shock wave the flow is not in LTE. This is a direct consequence of the transfer of a supersonic flow into a subsonic flow.

References
[4] see the contribution by S.Mazouffre, P.Vankan, R.Engeln, D.C.Schram in this proceedings.
[5] M.Playez; Von Karman Institute, Brussels