THE DISTRIBUTION OF ATOMIC HYDROGEN IN AN ATMOSPHERIC-PRESSURE FREE-BURNING ARC DISCHARGE

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ABSTRACT

Relative radial concentration profiles of atomic hydrogen in an atmospheric-pressure argon-hydrogen free-burning 200 A arc discharge were measured using laser-induced fluorescence by 2-photon excitation of the ground state of atomic hydrogen. Radial profiles are presented for three axial locations. A comparison between the measured profiles and profiles calculated using a diffusion model that considers demixing processes is made, indicating that local chemical equilibrium is not maintained at larger radii in the arc.

INTRODUCTION

A complete understanding of diffusion in mixed-gas plasmas requires experimentally-determined species-concentration profiles in order to validate diffusion models. Hydrogen is one of the most common reactive gases added to plasmas. Because of its low mass and atomic radius, atomic hydrogen is expected to have a high diffusion rate. Also, the net effect of collisions (demixing due to collision-frictional forces) is to concentrate the lighter species in the high temperature region of the plasma. This has been observed in argon-hydrogen arcs [1,2]. Verification of diffusion models might therefore best be accomplished by comparing predicted ground state atomic hydrogen density profiles with measured profiles. We report in this paper relative concentration profiles of atomic hydrogen in an argon-hydrogen atmospheric-pressure free-burning arc discharge determined by 2-photon laser excitation of the ground state. The measured profiles are compared with profiles predicted by a diffusion model based on the Chapman-Enskog method.

THEORY OF ARC COMPOSITION

The simplest description of the composition of an arc containing more than one chemical element is that the relative concentration of each of the chemical elements is constant everywhere in the arc, and that the concentrations of the species containing these elements are those calculated assuming local chemical equilibrium (LCE). Diffusion can lead to significant variations from this description. The relevant diffusion-driven processes can be conveniently divided into two types, according to whether they lead to departures from LCE. We will use the term demixing [3,4] to describe those diffusion processes that lead to spatial variations in the relative concentrations of the chemical elements within the arc, while not causing departures from LCE. Those processes that lead to departures from LCE occur when the diffusion rate of a given species is large compared to the rate of ionization, dissociation, recombination or other chemical reactions involving that species.

A detailed discussion of the calculation of diffusion and demixing can be found elsewhere [5]. Here we briefly summarize the methods used. The effect of demixing on the density distribution of atomic hydrogen is calculated using the method described in Ref. [4]. Using the combined diffusion coefficients formulation [4], an expression that relates the gradient of the hydrogen mole fraction (i.e., the sum of the mole fractions of all hydrogen species) to the temperature gradient is derived. This expression is integrated in the radial direction to give the hydrogen mole fraction as a function of radius. The atomic hydrogen density is calculated from the hydrogen mole fraction assuming the existence of LCE. To estimate the extent of deviations from LCE in the arc fringes due to the rapid diffusion of atomic hydrogen, we calculate an average diffusion distance. This is defined as the product of the diffusion velocity and the recombination time of atomic hydrogen. The diffusion velocity is calculated from multicomponent diffusion coefficients found using the Chapman-Enskog method. The recombination time is given by $\tau = -1/2k[H][M]$, where [H] is the molecular concentration of atomic hydrogen, and k is the rate coefficient for the three-body recombination reaction $H + H + M \rightarrow H_2 + M$, where M denotes any third body.

EXPERIMENT

There are several techniques available for multiphoton excitation of the ground state of atomic hydrogen. These techniques are discussed in detail elsewhere [6]. For this work, we used two 205 nm excitation of the ground state to the n = 3 level. The subsequent fluorescence transition monitored was the n = 3 to n = 2 (H_α) transition at 656.3 nm.

The 205 nm laser wavelength was generated by frequency-tripling the 615 nm output of a neodymium-doped yttrium aluminum garnet (Nd:YAG) pumped pulsed-dye laser operated with sulfarhodamine dye. The fluorescence signal was collected and focused onto the entrance slit of a 1-m focal-length monochromator with a bandwidth of about 0.08 nm and detected with a photomultiplier tube (PMT). The PMT signal was integrated using a gated dual-channel boxcar averager averaging over 30 shots of the laser. More experimental details are presented elsewhere [5]. A schematic of the experimental set up is given in Fig. 6. All data were taken on a free-burning transferred argon-hydrogen arc at atmospheric pressure operating over a water-cooled copper anode. The nominal hydrogen concentration of the gas mixture studied was 5% by mole fraction in argon. The cathode-to-anode gap was 5 mm. The arc current was 200 A and the total gas flow was about 8 l $\min^{-1}$.

RESULTS AND DISCUSSION

Figures 2 and 3 compare the measured and calculated radial profiles of the atomic hydrogen density at 1 mm and 2.5 mm below the cathode, respectively. Profiles calculated both including (curve A) and neglecting (curve B) the effects of demixing are given for both positions. The calculations were based on the temperature profiles measured using frequency-integrated laser scattering for a 200 A argon arc [5]. The measured and calculated positions of the peak in the atomic hydrogen density differ slightly at both positions. The discrepancies probably arise from two effects. First, there are likely to be small differences between the axial position used for measurements of hydrogen density profiles and for the temperature measurements, because the measurements were performed on a different apparatus, making it difficult to ensure that the thermal expansion of the cathode was compensated for equally in both measurements. Secondly, the measured temperature distribution of a pure argon arc was used in calculating the atomic hydrogen distribution. This is expected to lead to errors. The addition of hydrogen to an argon arc will increase its thermal conductivity, and is expected to narrow the arc column, thus decreasing the temperature in the fringe regions of the arc. Taking this second effect into account in the calculation of the atomic hydrogen density profiles would decrease the radius of the peaks, improving the agreement between the experimental and calculated results. Simulations using the code described by Lowke, Kovitya, and Schmidt [7] show that this effect is larger further from the cathode. Figure 4 is the measured relative atomic hydrogen radial profile 4 mm below the cathode. Gas temperature data was not possible to obtain this close to the anode because of excessive stray laser light, so calculated profiles are not presented.

Three further discrepancies are apparent between the measured and calculated profiles. First, the measured hydrogen density decreases radially towards the arc center to almost zero within 2.5 mm of the peak. This agrees qualitatively with similar measurements made on a free-burning arc discharge at a pressure of 0.5 atm [8]. However, the calculated density is still approximately 50% of its maximum value at this position. Secondly, the measured profile decreases much less rapidly than the calculated profile from the peak towards the arc edge. Thirdly, the measured profiles demonstrate small secondary peaks at radii greater than 10 mm; these are not reflected in the calculated profiles. The reason for these peaks is presently not understood.

The first discrepancy can probably be explained in terms of Stark broadening of the H_α signal that is detected. One cannot be certain if the lack of a fluorescence signal in the center of the arc is because there is not a detectable amount of atomic H present or because only a small portion of the of Stark broadening signal overlaps with the bandwidth of the monochromator. It is therefore meaningless to compare the measured profile with the predicted profile in this region. It was verified that the linewidth of the peak fluorescence signal was less than 0.05 nm at radial positions greater than 5 mm.

The second discrepancy between the measured and calculated atomic hydrogen density profiles may be attributed to the large diffusion rate of atomic hydrogen relative to the rate of recombination to form molecular hydrogen. The average diffusion distance of atomic hydrogen at radii is calculated to be some millimeters near the peak in the atomic hydrogen density, increasing rapidly with radius. Since the direction of diffusion is radially outward, we expect the atomic hydrogen density at large radii to be greater than the calculated densities shown in Figs. 2 and 3, which we derived assuming LCE. It should be noted that the diffusion distance was calculated assuming an LCE composition. The increase in atomic hydrogen density to levels higher than predicted under LCE leads to decreases in both the diffusion velocity and the recombination time of atomic hydrogen, hence limiting the diffusion distance and ensuring that the atomic hydrogen remains confined within the arc.

It is further worth noting that the rapid diffusion of atomic hydrogen can not move the position of the peak in atomic hydrogen density to a larger radius than that calculated assuming the existence of LCE. This is because the position of the peak in n_H is determined by the equilibrium chemistry, occurring at that temperature at which recombination of atomic hydrogen is in equilibrium with the dissociation of molecular hydrogen. At larger radii, no matter how large the diffusion flux of atomic hydrogen, the dominance of recombination reactions will cause a decrease in n_H. In fact, a large diffusion flux of atomic hydrogen can, counterintuitively, lead to the peak in n_H being shifted to a smaller radius, since an increase in n_H increases the temperature at which recombination and dissociation reactions are in equilibrium. This effect is, however, relatively small, since the equilibrium temperature only increases from 3940 K to 4390 K when the mole fraction of atomic hydrogen is increased from 0.05 to 0.50.

CONCLUSIONS

Diffusion processes in an atmospheric-pressure argon-hydrogen arc discharge were studied using 2-photon laser-induced fluorescence to determine radial relative concentration profiles of the ground state of atomic hydrogen. The measured profiles were compared to profiles calculated assuming the existence of LCE, but taking into account demixing. The measured atomic hydrogen density profiles of were found to decrease less rapidly with increasing radius at temperatures below the recombination temperature of atomic hydrogen than predicted by calculations. The discrepancy was explained by demonstrating that the diffusion rate of atomic hydrogen is larger than its recombination rate in this region. It may be concluded that LCE is not maintained in the low temperature region of the arc due to the rapid diffusion of atomic hydrogen.

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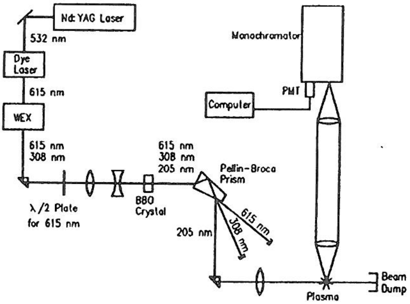

Fig. 1. Experimental schematic.

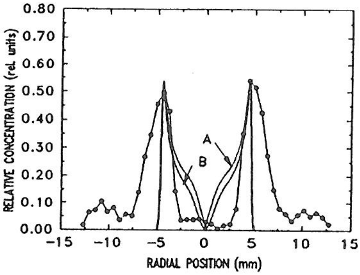

Fig. 2. Relative H concentration as a function of radius 1 mm below the cathode.

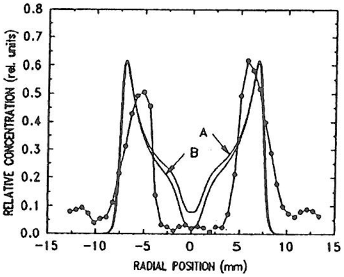

Fig. 3. Relative H concentration as a function of radius 2.5 mm below the cathode.

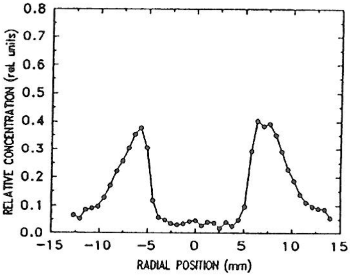

Fig. 4. Relative H concentration as a function of radius 4 mm below the cathode.