Modeling And Measurements Of The Demixing Effect In Arc Plasmas Containing Hydrogen

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Abstract. In this work are presented the results of the measurements of the plasma parameters in the wall-stabilized arc working in several mixtures of argon, hydrogen and sometimes helium, showing the lateral and longitudinal demixing effect. Lateral demixing in Ar+H_2 is compared with the theoretical calculations of the plasma temperatures and molar ratios for Ar+H_2 mixture, and its agreement with even the very simplified model of diffusion in arc plasma. The experiment shows the considerable lateral variations in the hydrogen percentage in the plasmas containing helium and, as well, the visible variations in electron density in such case.

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INTRODUCTION

The wall-stabilized arc is a widely used source of the atomic and ionic radiation [1-2]. This type of plasma is very popular in the spectroscopic research, e. g. for measurements of atomic and ionic structure data, such as spectral line strengths or Stark shifts and widths. The arc plasma sources used also for many other industrial and scientific applications – especially as the plasma generators for various plasmatrons and jets.

Hydrogen is a very popular tool for plasma diagnostics – very small admixtures of this element can be found in most gases and the lines of the Balmer series, exhibiting strong Stark broadening, are for years widely used for determining the electron density.

Problems with behavior of admixtures in an arc plasma, exhibiting strong gradients of plasma parameters, is rather complex. In the case of hydrogen, especially longitudinal demixing can be very important for plasma diagnostics, essential for determination of atomic and ionic parameters. In the case of end-on measurements, or unresolved side-on measurements, the hydrogen lines are assumed to originate predominantly from the same plasma volume as the studied component, which can be untrue, especially in the helium-based plasmas , which are very popular due to their high temperature and low electron density.

SIMPLIFIED MODEL OF DEMIXING

Diffusion in a gas can be described by equations for the mass flux J_i, relative to the mass-average velocity, of each of the species *i* present.

Murphy [3] has shown that in a mixture of two homonuclear gases that do not react with each other the treatment of diffusion can be greatly simplified if local chemical equilibrium is assumed. In this case, instead of considering the diffusion of individual species separately, one can consider the diffusion of gases.

It is possible to derive an expression for the mass flux of a gas, defined as the sum of the mass fluxes of all the species that make up the gas.

In the equilibrium situation, in which the mass fluxes of the gases have vanished, the equilibrium mole fraction gradients of the two gases are calculated by setting all fluxes to 0, giving.

$$\nabla \overline{x_B} = -\nabla \overline{x_A} = \frac{\rho}{n^2} \frac{\left(\overline{D_{AB}^{T1}} + \overline{D_A^{T}}\right)\nabla \ln T}{\overline{m_A}\,\overline{m_B}\,\overline{D_{AB}^{x}}} \tag{1}$$

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where $\overline{m_Y}$ is the average mass of the heavy species of the particular gas Y and $\overline{x_Y}$ is the sum of the mole fractions of all species of this gas. $\overline{D_Y^X}$ are the combined diffusion coefficients of both mixed gases.

To estimate the demixing effect in a plasma, the plasma parameters have also to be taken into account. In the general model, several conservation equations have to be taken into account. Simplification is possible if the following conditions are fulfilled:

- velocity of the plasma flow is negligible
- plasma is in stationary state
- temperature gradient is perpendicular to the arc current
- radiation losses are negligible.

Only the energy conservation equation is then important, which for two components plasma, taking into account demixing, takes the form:

$$\frac{\bar{j}^2}{\sigma} + \nabla\cdot\left(\frac{\kappa}{c_p}\nabla h\right) - \nabla\cdot\left[\left(\overline{h_A} - \overline{h_B}\right)\left(\frac{\kappa}{c_p}\nabla\overline{Y_A}\right)\right] = 0 \qquad (2)$$

where $\overline{h_Y}$ is the enthalpy of one of the gases, and $\overline{Y_Y}$ is the mass fraction of the gas Y.

Both of the above mentioned equations can be still simplified, if the cylindrical symmetry is assured, so the three dimensional gradients can be reduced to the radial derivatives.

This model cannot predict any longitudinal (axial) demixing, as it does not take into account the diffusion due to electric forces, and one of the assumption is the cylindrical symmetry of the source. Te calculations based on this model can, theretofore, describe only the lateral demixing and its dependence on plasma composition.

EXPERIMENT

The experiment was performed using the cascade arc consisting of eleven copper segments of 6 mm width, separated by 1.5 mm thick teflon rings). The discharge channel diameter was 4 mm. The thoriated tungsten electrodes were attached to the boundary segments. The mixture of argon and helium was inserted by 4 mm diameter inlets alongside the whole plasma channel. The arc current was 40 A.

The construction of the arc is cylindrically symmetric. The gas inlets are located in the side window, and the outlets are between each of the arc segments along the arc column. Also the arrangement of both electrodes at the end of the discharge channel is identical. The weak and symmetrical gas flow through the discharge volume assures, that the distribution of the gas mixture in the absence of the arc current is uniform. Thus, the departures from the uniformity (axial and radial gradients of the parameters) in the plasma column can be explained only as a result of the cooling originating from the arc walls and the demixing effects in plasma.

The light emitted from the plasma channel was registered in side-on geometry. The teflon spacers were slitted to allow the observation of light between nine inside cascade arc segments – from second to tenth. The plasma was imaged by applying the spherical mirror ($f = 730$ mm) onto the entrance slit of the PGS-2 spectrometer. The dispersed light was registrated by an Optical Multichannel Analyzer (OMA). The CCD matrix of the OMA was divided vertically to form 256 tracks, for measuring the spatial distribution of the emitted light. From the measured light intensity (integrated over the line of sight), assuming the axial symmetry, after Abel inversion the radial distribution of emission coefficients of atomic and ionic spectral lines were obtained. The width of one track corresponded to 0.026 mm distance across the arc channel. The observed plasma diameter was larger than the plasma channel (4 mm) due to the plasma expansion in the region between the copper segments.

To register the light emitted from the spaces between different arc segments, the arc was translated perpendicularly to the optical axis of the spectrometer. In this way, a three-dimensional map of the emission coefficients of different atomic and ionic lines have been determined.

Three experiments were performed with different plasma compositions: argon plasma, with only a few percent of hydrogen, the “intermediate” mixture, where the gas was still mostly argon, but with an important addition of hydrogen (about 20% molar percentage), and the plasma mixture with the same argon to hydrogen ratio as before, but containing about 50% helium.

The plasma diagnostics was performed using the emission coefficients of argon (693.8, 696.5 and 703.0 nm) and hydrogen (H_α and H_β) lines and the HWHM of the hydrogen lines (both H_α and H_β).

RESULTS AND DISCUSSION

Longitudinal Demixing

Results of the study of the longitudinal demixing of hydrogen in argon and argon-helium mixture are shown in figs. 1 and 2. For easier comparison, the results are averaged in the ring of the 0.1 mm radius around the arc axis. The results show, that for the mixture of argon and hydrogen only the axial uniformity is not bad, showing the greater discrepancies only for the vicinity of the cathode (the minimum in electron density in the middle of the arc may be an artifact). The situation in the mixture of argon, hydrogen and helium is much more complicated, and the resulting lineshape registered in the end-on geometry needs not to be in agreement with any of the local values, especially as the strong hydrogen radiation is mostly from the region of high electron density, which can suggest the overestimation of the overall on-axis electron density.

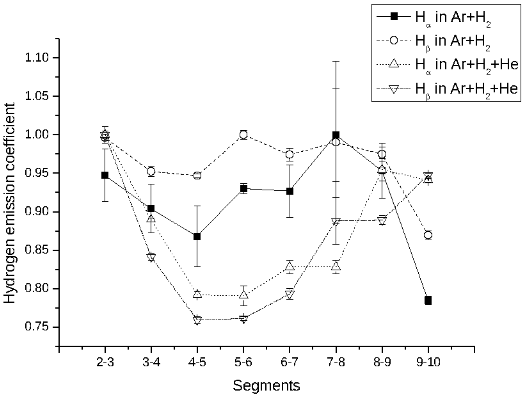

FIGURE 1. Values of the hydrogen lines emission coefficient, normalized to their maximal values.

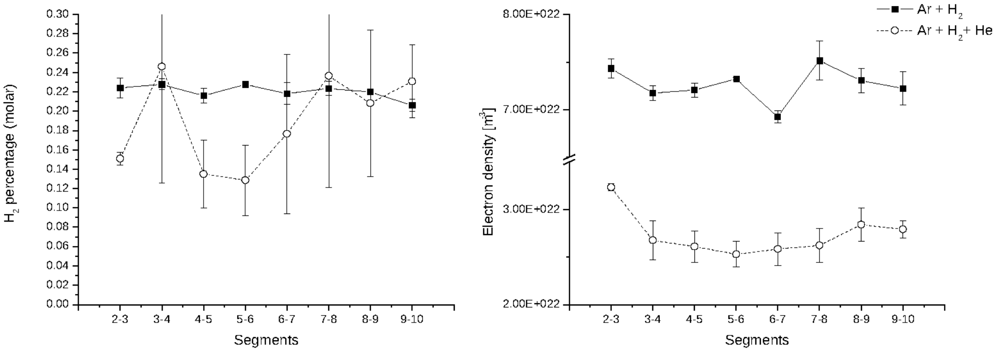

FIGURE 2. Plasma parameters plotted along the arc axis for two gas mixtures.

Lateral Demixing – Theory And Experiment

The results – temperature and molar percentage of hydrogen – obtained by resolving numerically equations 1 and 2 for the parameters of this experiment are shown in figure 3 and compared with experimental results. This figure shows, that addition of hydrogen to argon results in "squeezing" the hot region of the plasma, which rises a bit the plasma temperature (the difference starts to be visible, though, only for very high percentage of hydrogen). Radial dependence of the molar percentage of hydrogen shows the on-axis maximum and another maximum, on the fringe of the arc, which can be observed for overall hydrogen percentage exceeding 2-3% and show closer and closer to the plasma axis as the overall percentage of hydrogen rises.

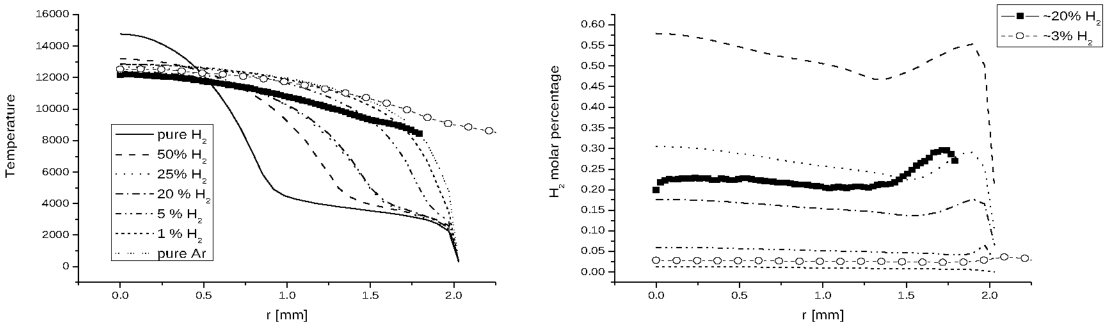

FIGURE 3. Temperature and molar percentage of hydrogen in hydrogen-argon mixtures – calculations and experimental values. Theoretical curves are presented as lines, experiments as points joined by lines.

The simplified model can in fact describe the situation deep in the plasma channel, not in the regions where the experiment is performed – i.e. between the arc segments. The real values of the temperature are a bit lower and the temperature distribution is definitely broader. Taking this into account we can see, that the LTE values of the experimental temperature are similar to theoretical curves, within the error bars (which in this case are around 10%). The plasma composition fits even better – the shape of the dependence of the hydrogen amount on the radius are very nicely reflected by experimental values. The experiment agrees with calculations as well in the feature, that for increasing amount of hydrogen its distribution is „squeezed", being definitely wider for small amount of hydrogen than for higher one.

CONCLUSIONS

Longitudinal demixing of hydrogen in argon is very slight – the intensity of hydrogen lines is notably different only in the vicinity of the cathode (the difference of the 7-8 segment gap is due to the experimental problems there). There is also a difference in the electron density in the middle of the arc, but it is pretty small (a few percent). Much bigger are the gradients in the mixture containing helium – the differences in electron density are up to 20%, and the hydrogen density can vary even more.

The radial demixing in Ar+H_2 mixture can be pretty accurately predicted using the simple one-dimensional model of two-gas demixing, in the kind developed by A. B. Murphy, and using his coefficients for detemining the plasma parameters. The differences between the experimental results and the model are mostly due to the simplification of the model from one side, and the problems with partially non-LTE atomic level excitation distribution in argon (the calculations of temperature assuming LTE distribution are significantly different than the temperature derived from Boltzmann diagram).

Data presented here show, that the transversal and longitudinal demixing of the hydrogen in the plasma is significant, especially in the case of the plasmas containing helium. This demixing can, as well, affect the plasma parameters, especially electron density (gradients of temperature due the changes in plasma composition are much less visible). This problem should be taken into account in the end-on measurements, where the longitudinal uniformity of the hydrogen emission is ordinarily assumed, and used to determine the electron density.

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