CHARACTERISTICS OF A SPRAY PLASMA

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ABSTRACT

The measured evolution of the electron density and the plasma temperature
are compared with the results of a model based on the conservation laws.
It is concluded that ambipolar diffusion is the governing process, that
the system must be close to LTE and that also in the expanding plasma jet
currents can not be excluded.

1. INTRODUCTION

In plasma spraying techniques a plasma gun is used to provide the hot
plasma flow to heat and to accelerate spray particles to be deposited on
substrates. In order to model these processes it is important to know the
temperature field of the plasma. It appears that the heat transport from
the plasma to the particle depends primarily on the plasma temperature.
Also the viscosity which plays a dominant role in the acceleration
process depends on the plasma temperature.

In most treatments the plasma is assumed to be close to equilibrium, which
gives a definite relation between electron temperature and density at
given pressure. In the active plasma the neutral ground state must be
slightly overpopulated as this is required to provide the finite particle
source. In the expanding plasma beyond the anode this is not required any-
more and the plasma may be close to equilibrium or even slightly recombin-
ing, i.e. the neutral ground state may be underpopulated. It appears that
very accurate temperature measurements are needed (better than 5%) to
allow conclusions at this point.

A third item which demands attention is the possible difference between
heavy particle (ion and atom) temperature and the electron temperature.
In this paper we will compare the measured evolution of $n_e$ and $T_e$ with
the results of a model based on mass- and energy-balances and arrive at
several conclusions concerning the matters discussed above.

2. MEASUREMENTS

In the literature [1-2] several detailed measurements of $n_e$ and $T_e$ have
been reported on. We will use for illustration (fig.1) the $r$, $z$
dependences of $n_e$ and $T_e$ as reported in [1].

The electron density has been measured by $H_\alpha$-broadening, using Griems
relation between density and broadening. The influence of Abel inversion
has been evaluated by Abel inversion of lateral profiles at various
settings. This influence amounted to corrections of the order of 10%. The electron temperature has been determined in two ways. First, absolute values at the axis are obtained from the ratio of Ar II 480.6 nm line to the continuum. This method has a precision of better than 5%. It appears that these values differ only slightly from the $T_e$ LTE ($n_e$) at the values of $n_e$ at a pressure of 1 bar (see section 3). As the difference is small, most points in fig. 1.b are derived from $n_e$ under the assumption of LTE.

Fig. 1: Spatially resolved electron density (a) and electron temperature (b).

$\phi$ anode = 6 mm, $I$ = 450 A, 3% H$_2$.

3. THE EVOLUTION OF THE ELECTRON DENSITY AND THE MASS BALANCE

The mass balance for ions, $i$, and neutrals, $a$, read for stationary argon plasmas (not in LTE):

$$\nabla \cdot n_{i-a} = \nabla \cdot n_{a-a} = K_1 n_{a1}^S \delta b_1 n_e - n_e^2 k_{rad} A_{rad} = \text{net source}$$

in which $K_1$ is the total excitation and ionization rate and $n_{a1}^S$ is the Saha density of the neutral ground state; defined as:

$$n_{a1}^S = n_e^2 \frac{g_1 \cdot \frac{2\pi m_e kT_e^{-3/2}}{\hbar^2}} \exp \left[ \frac{E_i^+}{kT_e} \right] = n_e^2 S_1(T_e).$$

For ionization, only the out of equilibrium portion of the neutral ground state is effective; this is indicated by the parameter $\delta b_1$ which indicates the deviation of equilibrium:

$$\delta b_1 = \frac{n_{a1}^S - n_{a1}}{n_{a1}}.$$

The second term of the RHS of eq. (1) describes the radiative recombination to the ground state with rate coefficient $k_{rad}$; the expression is corrected for the effect of trapping by the escape factor $A_{rad}$. Though the latter is dependent on the actual plasma geometry we will use for simplicity the value by Hermann for cylindrical arcs [3].

If we assume LTE i.e. only the atom ground state is out of equilibrium, then the quantities $T_e$, $n_e$, $\delta b_1$ and the total pressure, $p$, are inter-
related. In fig. 2 the product $K_1 S_1$ and the electron density at LTE have been plotted as functions of the temperature. At this point it is useful to make a distinction between convection and diffusion. If we introduce the convective velocity, $\mathbf{w}$, as

$$ \mathbf{w} = \frac{\mathbf{n}_i \mathbf{v}_i + \mathbf{n}_a \mathbf{v}_a}{n_i + n_a} $$

then it follows from eq. (1) that the convective heavy particle flow is divergence-free:

$$ \nabla \cdot (n_i \mathbf{w}_i) = 0. \quad (4) $$

The small difference between the total ion drift velocity, $\mathbf{w}_i$, and the mass flow velocity, $\mathbf{w}$, can be interpreted as the diffusion velocity:

$$ \nabla \cdot \mathbf{n}_i \mathbf{v}_i + \nabla \cdot \mathbf{n}_i \mathbf{w} = \text{net source} \quad (5) $$

where $D_A$ is the ambipolar diffusion coefficient:

$$ D_A = \frac{5.5 \times 10^{18}}{n_a + n_i} T_e^{0.64} $$

So, the ion mass balance reads:

$$ -\nabla \cdot D_A \nabla n_i + \nabla \cdot n_i = \text{net source} \quad (6) $$

The second term of the l.h.s. can be rewritten as:

$$ \nabla \cdot \mathbf{n}_i \mathbf{v}_i = \nabla \cdot \left[ \frac{n_i}{n_a + n_i} \right] \mathbf{n} \left( n_a + n_i \right) \quad (7) $$

As $n_i$ is much larger than $\frac{n_i}{n_a + n_i}$ the second term of eq. (7) can be neglected and $\nabla \cdot \mathbf{n}_i \mathbf{w}$ can be replaced by $\mathbf{w} \cdot \nabla \mathbf{n}_i$. This is in agreement with the experimental observation, that the variation of $\mathbf{w}_z$ is small. The resulting equation is then $(\mathbf{w}_z \gg \mathbf{w}_r, \mathbf{w}_i \gg \mathbf{w}_z) :

$$ -\frac{1}{r} \frac{\partial}{\partial r} D_A \frac{\partial n_i}{\partial r} + \mathbf{w}_z \frac{\partial n_i}{\partial z} = \delta_b \frac{K_1 S_1}{n_e} n_e - \frac{N_e}{n_e} \mathbf{k} \text{ rec \ rec} \quad (8) $$

In fig. 3 the plasma velocity, $\mathbf{w}_z$, is shown as measured by L.D.A. on small 3 μ size particles. A slight increase of $\mathbf{w}_z$ is observed over the first few cm. One can show from the total momentum balance that a pressure drop of 40% could explain this increase. It may also partly be due to finite drag. In this analysis we will assume that the plasma
velocity is 300 m/sec up to \( z = 30 \) cm. The source term still contains the non-equilibrium contribution. The measurements of Vaessen [1] indicate a near to LTE (\( \delta b_1 = 0 \)) or a slight underpopulation and thus negative value of \( \delta b_1 \). However, the accuracies required here (2% in \( T_e \), 10% in \( n_e \) and \( p \)) hardly allows conclusions at this point. So, we will assume that the system remains close to equilibrium. The results of the model calculations are shown in fig.4.

Fig.4 : Measured (*) and calculated (-----) radial density profiles at various distances from the anode : a) \( z = 2 \) mm, b) \( z = 6.5 \) mm, c) \( z = 11.5 \) mm, d) \( z = 16.5 \) mm, e) \( z = 21.5 \) mm, f) \( z = 26.5 \) mm

The overall agreement confirms that the ambipolar diffusion is the major effect. The radiative recombination is only a weak effect, as the contribution of inelastic processes provided \( |\delta b_1| < 0.1 \). So in a good approximation the particle source term (which is negative for recombining plasmas) can be ignored. As far as the data allow conclusions, there are indications that the total number of electrons (integrated over the convection) increases somewhat which would mean net production and a positive \( \delta b_1 \).

4. ENERGY BALANCE

The total energy balance can be obtained by adding the balances for electrons and heavy particles. The result is:

\[
H(T_e) \delta b_1 + n_e H(T_e) - \frac{5}{2} kT_e \Delta S + \frac{p}{kT_e} \nabla V - \mathbf{w} \cdot \mathbf{V}_p + \mathbf{V} \cdot q = j \mathbf{E}
\]

The function \( H(T_e) \) stands for:

\[
H(T_e) = k_1(T_e) S_1(T_e) (E_1^+ + \frac{5}{2} kT_e)
\]

where \( E_1^+ \) = ionization energy.
The term \(n_e G(T_e)\) describes the radiative losses, except for the radiative recombination and resonance radiation to the ground state which are accounted for in the inelastic first term.

The total heatflow \(q\) can be written as \(q = \kappa c T\) where \(\kappa\) is the total heat conductivity, which for temperatures above 10,000 K is mainly carried by electrons. Note that the radiative losses are treated separately and are not included in the definition of \(\kappa\).

Radiation, heat conduction and inelastic losses if \(\delta b_1 > 0\) lead to energy loss and hence to a decrease of the temperature. From the magnitude of the radiative term alone \(n_e^2 G(T_e)\) with \(G(T_e) \sim 6 \times 10^{-38}\) we can conclude that the temperature should decrease much faster than observed in fig.1.b.

At \(z = 0\) the cooling rate \(\frac{\partial T}{\partial z}\) would be about 20-30/m due to radiation- and expansion-cooling and heat conduction. This is not observed; so, if we assume no net current density in the plasma jet, the only possibility is that \(\delta b_1\) must be negative. In this way the plasma is heated during the expansion by recombination. This deviation has been estimated by Vaessen[1] to be between \(-\delta b_1 = 0.1-0.2\).

There is however another intriguing possibility: Joule dissipation. If we assume that a convective current loop is formed in the expanding plasma of about 10% of the main current between anode and cathode, then enough Joule dissipation occurs to cope with the heat losses. Such a current density of about \(2 \times 10^6 \text{ A/m}^2\) is equivalent to a difference between electron and ion drift velocities of only half the plasma velocity. This can certainly not be excluded. If this mechanism would be operative then \(|\delta b_1|\) can be close to zero even positive and the system would be close to LTE as assumed in the modelling of the density evolution.

CONCLUSIONS

The evolution of the electron density in an expanding plasma jet is shown to be mainly due to ambipolar diffusion. As the line integral of the density does not decrease significantly and even increases somewhat we must conclude that the system is still ionizing. The energy balance predicts a rather fast decrease of the temperature unless heat is dissipated by recombination \((\delta b_1 < 0)\) and/or Joule dissipation of small convective currents in the plasma jet. Since from the mass balance near equilibrium \((\delta b_1 > 0)\) is concluded, we suspect convective currents to exist in the spray plasma.


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