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Axial variation of electron number density in thermal plasma spray jets

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Abstract. The electron number density has been measured in a plasma spray torch using Stark broadening of H_β and Ar-I (430 nm) line. A small amount of hydrogen (1% by volume in argon gas) was introduced to study the H_β line profile. Axial variation of electron number density has been determined up to a distance of 20 mm from the nozzle exit point of spray torch. The plasma torch was operated at 5 and 10 kW power level and flow of argon was kept at 25 liters per minute. Using the measured excitation temperature data under same experimental conditions, the electron number density has also been calculated using Saha equation. Comparison of electron number densities measured from Stark broadening with those derived from excitation temperature measurements under the assumption of local thermodynamic equilibrium (LTE) in thermal plasma jets indicate about the deviation from LTE in thermal plasma jets. The electron number density measurement using Stark broadening of Ar-I (430 nm) line will be particularly useful when only argon gas is used in thermal plasma jets.

PACS. 52.70.Kz Optical (ultraviolet, visible, infrared) measurements – 52.77.-j Plasma applications – 52.25.-b Plasma properties

1 Introduction

The plasma spray jet temperature, velocity and electron number density are important parameters for determining the heating and acceleration of spray powder and for determination of thermo-physical properties of such plasmas. Although temperature and velocity data for plasma spray torches are available in literature [1,2], limited studies have been made for the electron number density determination. The electron density is the key parameter for establishing the ionization equilibrium and for energy transfer in thermal plasma jets.

The electron density profiles using Stark broadening of H_β line have been measured in Ar/He plasma jet [3] by adding a small amount of hydrogen. The stark broadening technique can be used even if plasma is not in local thermodynamic equilibrium (LTE) state. Moreover, sophisticated theoretical calculations for Stark broadening of H_β line in plasma [4–6] are available and using Vidal, Cooper and Smith (VCS) tables [5] electron density can be determined with an uncertainty of $\pm 5\%$. It has been shown that electron densities derived from Stark broadening are substantially higher than the LTE predictions and these higher values may be attributed to the chemically frozen flow situations caused by the high axial jet velocities and to non LTE effects caused by radial diffusion.

The Stark broadening of seven Ar I transitions (826.5 nm, 794.8 nm, 750.3 nm, 720.7 nm, 714.7 nm, 703.0 nm and 696.5 nm) have been determined in a direct current argon plasma jet [7] and used for the determination of electron density and electron impact parameter. An experimental investigation of the conditions for local thermal equilibrium to exist in an atmospheric pressure argon plasma jet has been made and it has been shown that LTE does not exist below an electron density of $6\times10^{22}/m^3$ [8]. Effect of pressure variation on temperature and electron density measurement has been studied in Ar–H_2 (1%) thermal plasma jet [9] at power input of 4.5 kW. The pressure was varied in the range 0.3–1 bar. Strong discrepancies exist between measured electron density using Stark broadening of H_β line and those from argon continuum measurement based on the LTE assumptions.

Electron temperature and electron density profiles of an atmospheric pressure argon plasma jet [10] have been measured from the analysis of the electron feature of Thomson scattered laser light by the plasma. Measured centerline values of the electron temperature are in excess of 20 000 K at the torch exit and these measurements suggest that significant deviation from LTE persists throughout the plasma jet. Comparative measurement on thermal plasma jet characteristics in atmospheric and low pressure plasma spraying have been made [11] and electron density in plasma jets was measured using a Langmuir probe rotating across the plasma jets.

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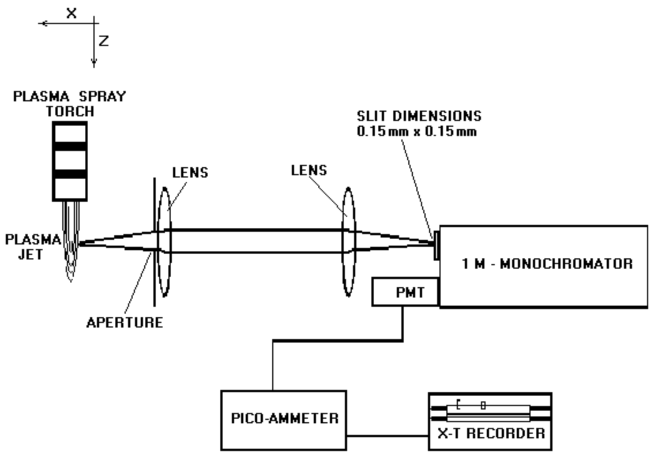

Fig. 1. Schematic of the experimental set-up for line profile measurement.

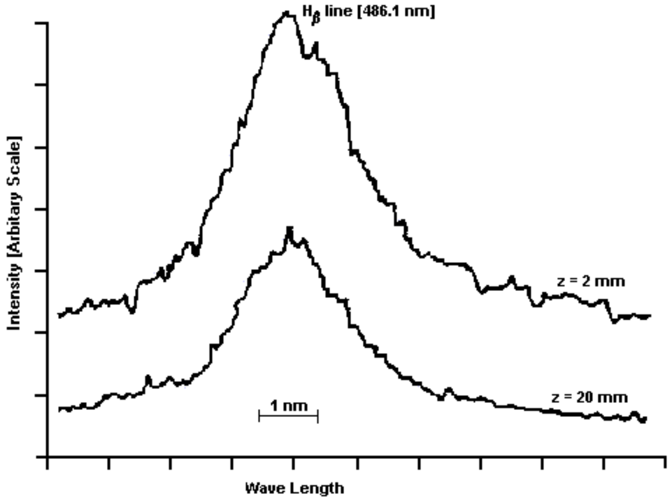

Fig. 2. Stark broadened profile of H_β line at axial distance 2 mm and 20 mm from torch nozzle exit at 5 kW torch power.

The effective line profile emitted by plasma jet can be approximated by a Voigt function. The experimental line shape should be properly deconvoluted accounting for Doppler and instrumental profile to obtain the Lorentzian half width of the transition. In this paper, Stark broadening of H_β line and Ar I transition (430 nm) has been used for the determination of electron number density. The electron density values obtained here are line of sight (LOS) values. Although, it is well-known that typical plasma spray jet exhibit fluctuations, the measured electron density values in this paper are time averaged and line of sight averaged values. An axial density profile up to a distance of 20 mm from the nozzle exit has been obtained. These values are compared with the previously reported values for direct current argon plasma jet using various techniques. There exists a large discrepancy between electron density determined by Saha equation using measured spectroscopic temperature values and Stark broadening techniques.

2 Experimental set-up and measurement

The plasma torch used in these experiments consists of a water-cooled thoriated tungsten cathode and a water-cooled copper anode nozzle of 8 mm internal diameter. The arc is vortex stabilized and it operates at atmospheric pressure. The torch has been operated at 5 kW (23 V, 220 A) and 10 kW (23 V, 450 A) power levels with argon flow rate of 25 l/mn. To measure the spectral line profile of H_β line, hydrogen gas with a flow rate of 250 cc/mn was introduced along with argon gas.

A schematic diagram of the experimental set up for measuring the line profiles is shown in Figure 1. A 1-m Monochromator (Monospek-1000) along with an EMI-9658B photo multiplier tube, a picoammeter, and an X-t recorder was used for measuring the intensity profile of Ar I line (430 nm) and H_β line (486.1 nm). The monochromator has a reciprocal linear dispersion of 0.82 nm/mm. The speed of monochromator for recording the intensity profile of Ar I line and H_β line was kept at 1 nm/min and 10 nm/min respectively.

The torch is mounted on a screw driven X-Y-Z table. The z-axis had its positive direction down wards in the direction of cathode anode axis as shown in Figure 1 and $z = 0$ is the nozzle exit point. The plasma flame is observed in a direction parallel to x-axis. For a fixed z-position, the X and Y-screws were moved and point of maximum intensity was fixed to obtain the intensity along the given line of sight chord from the plasma jet. The excitation temperature along the line of sight has been measured under the same experimental conditions of plasma jet using atomic Boltzmann plot method and central axis temperature using modified atomic Boltzmann plot method [12]. Spectral measurements of line profile of Ar-I line and H_β line have been made at 2 mm increments from an axial position 2 mm (from nozzle exit point) to a distance of 20 mm. At the same location, forward and backward scan of line profile of H_β or Ar-I line were recorded and average of these scans were taken in order to minimize the effect of intensity fluctuations. Using these line profiles and properly deconvoluting them, the electron density along the line of sight (LOS) in thermal plasma jets can be obtained using the Stark broadening techniques. Beyond the distance of 20 mm from the nozzle exit, the jet is still visible but its intensity drops drastically. For our experimental conditions, the argon jet is discharging into surrounding air and visible flame length is approximately 28 mm.

3 Results and analysis

The measured intensity profiles of H_β line and Ar-I line at 2 mm and 20 mm from the nozzle exit point are shown in Figures 2 and 3 for the operating torch power of 5 kW and 10 kW respectively. For the case in which there are two or more competing mechanisms such as instrumental

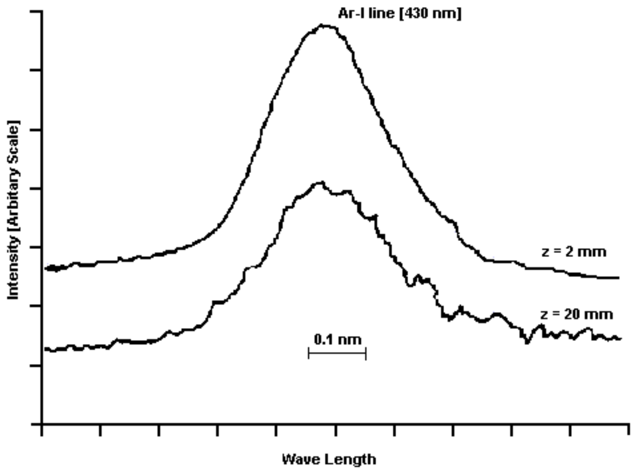

Fig. 3. Stark broadened profile of Ar-I line at axial distance 2 mm and 20 mm from torch nozzle exit at 10 kW torch power.

($\Delta\lambda_{1/2}^{\mathrm{inst}}$) and Doppler ($\Delta\lambda_{1/2}^{\mathrm{Doppler}}$) broadening represented by a Gaussian function their deconvolution is obtained by adding their half widths in the following way:

$$\Delta\lambda_{1/2}^{\mathrm{total\,G}} = [(\Delta\lambda_{1/2}^{\mathrm{inst}})^{1/2} + (\Delta\lambda_{1/2}^{\mathrm{Doppler}})^{1/2}]^{1/2} \quad (1)$$

where $\Delta\lambda_{1/2}^{\mathrm{total\,G}}$ is the total Gaussian width. On the other hand two Lorentzian half widths due to Stark and natural broadening are added linearly *i.e.*

$$\Delta\lambda_{1/2}^{\mathrm{total\,L}} = \Delta\lambda_{1/2}^{\mathrm{Stark}} + \Delta\lambda_{1/2}^{\mathrm{natural}}. \quad (2)$$

If the profiles of a line due to two different broadening mechanisms are $I_1(\Delta\lambda)$ and $I_2(\Delta\lambda)$, then the resultant profile is obtained by a convolution integral:

$$I(\Delta\lambda) = \int_{-\infty}^{+\infty} I_1(\Delta\lambda - \Delta\lambda')\, I_2(\Delta\lambda')\, \mathrm{d}(\Delta\lambda'). \quad (3)$$

The deconvolution procedure is described in detail in reference [13] and has been adopted in present studies to evaluate the half width due to Stark effect.

The linear Stark effect of the hydrogen lines, specially H_β line have received special attention [4–6] and here the full width at half maximum (FWHM) increases with $n_e^{2/3}$:

$$\Delta\lambda_{1/2} = 2.5 \times 10^{-9} \alpha_{1/2} n_e^{2/3} \quad (4)$$

where $\Delta\lambda_{1/2}$ is the FWHM in Å, n_e is the electron density in cm^{-3} and $\alpha_{1/2}$ is the shape factor. The values of $\alpha_{1/2}$ for H_β line have been taken from reference [5]. The axial variation of electron density using the H_β line Stark broadening for spray torch operating at power level of 10 kW and 5 kW is shown in Figures 4 and 5.

A formula for calculating the electron density using half width of H_β line has been proposed by Ovsyannikov [15]:

$$\log n_e = 1.452 \log \Delta\lambda_{1/2} + 16.017 \quad (5)$$

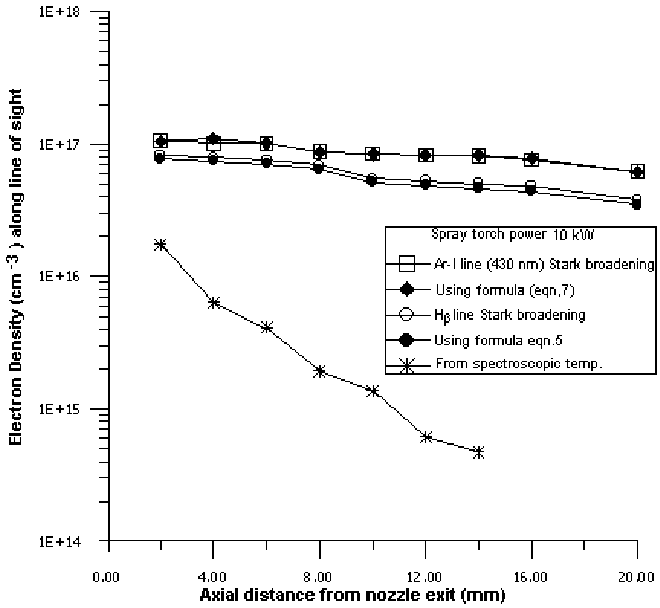

Fig. 4. Axial variation of electron density at 10 kW torch power.

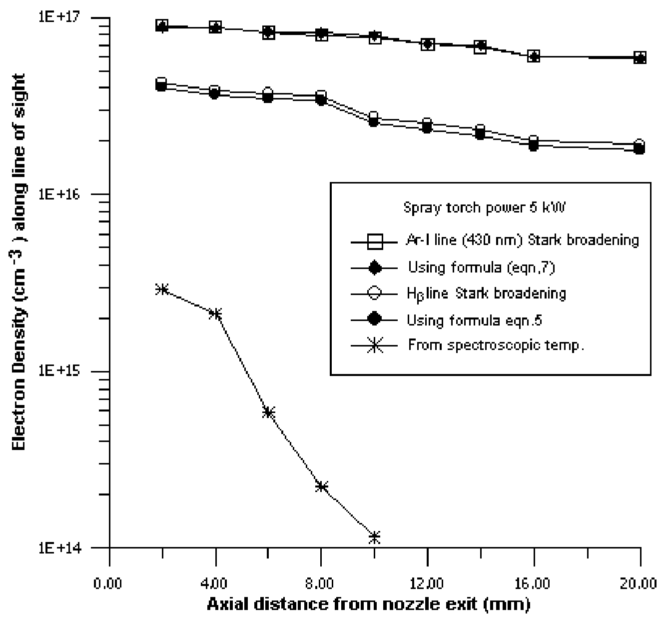

Fig. 5. Axial variation of electron density at 5 kW torch power.

where n_e is in cm^{-3} and $\Delta\lambda_{1/2}$ is in nm. It can be seen from Figures 4 and 5 that n_e values calculated using this formula agree well with the values obtained from experimentally observed Stark broadened H_β line profile.

The spectral lines of non-hydrogenic lines exhibit quadratic Stark effect and here the half width is proportional to the electron density [13,14]

$$\Delta\lambda_{1/2} = 2 \times 10^{-16} \omega n_e \times [1 + 1.75 \times 10^{-4} n_e^{1/4} \alpha \{1 - 0.068 n_e^{1/6} / T_e^{1/2}\}] \quad (6)$$

where ω is the electron impact parameter and α is the ion impact parameter. The values of ω and α for Ar-I line (430 nm) has been given by Griem [4]. Although, various Ar I lines have been studied for Stark broadening purpose, but still large discrepancies exists and the values of Stark parameters are known with an uncertainty of $\pm 15-25\%$. The values of n_e obtained using Stark broadening of Ar-I line (430 nm) are higher than that of using H_β line broadening as shown in Figures 4 and 5.

A curve fitted empirical formula for calculating the electron density n_e (cm^{-3}) using half width of Ar I line ($\Delta\lambda_{1/2}$ in nm) is given below;

$$\log n_e = 17.432 + 0.662 \log \Delta\lambda_{1/2}. \quad (7)$$

Under the assumption of LTE, the average excitation temperature measured using atomic Boltzmann plot method [12] can be taken same as the ionization temperature. If we use excitation temperature values measured from spectrometric technique and calculate n_e from Saha equation, the value of n_e varies from $1.76 \times 10^{16}/cm^3$ to $4.75 \times 10^{12}/cm^3$. These values are substantially lower than that determined from Stark broadening. It can also be seen that small change in temperature causes large change in electron density under LTE condition in thermal plasmas. The values calculated using Saha equation are substantially lower than that determined from Stark broadening. The discrepancies between electron densities derived from LTE condition and from Stark broadening increases with increasing distance from the nozzle exit and at low torch power.

The average electron density variation along axial direction in a spray torch may be given by an expression:

$$n_e(z) = n_e(0) \exp(-z/L) \quad (8)$$

where $n_e(0)$ is the average electron density measured along line of sight (LOS) at $z = 0$ and L is the visible flame length. Since $n_e(0)$ can not be measured due to experimental difficulties, it has been estimated by extrapolating the curve between $n_e(z)$ and z/L. The measured average electron number density (LOS) using H_β line profile for spray torch power 5 kW and 10 kW are plotted in Figure 6. By extrapolating this curve, the value of $n_e(0) = 4.79 \times 10^{16}/cm^3$ for 5 kW torch power and $9.39 \times 10^{16}/cm^3$ for 10 kW torch power.

It may be seen that electron densities derived from Stark broadening technique, Thomson scattering technique and rotating Langmuir probe technique are in the same range ($10^{17}/cm^3$ to $10^{16}/cm^3$) for atmospheric pressure plasma spray jets under typical operating parameters of plasma torches. High temperature electrons are generated in the discharge region of plasma torch. These electrons exit the torch with a bulk axial velocity of 1000 m/s. Owing to the high velocity, the traveling time of the plasma from nozzle exit point to axial position $z = 20$ mm down stream, is of the order of few microseconds, which is of the same order of magnitude as relaxation times. Therefore, a partially frozen flow kind of situation may occur and this finite rate of chemical reactions coupled

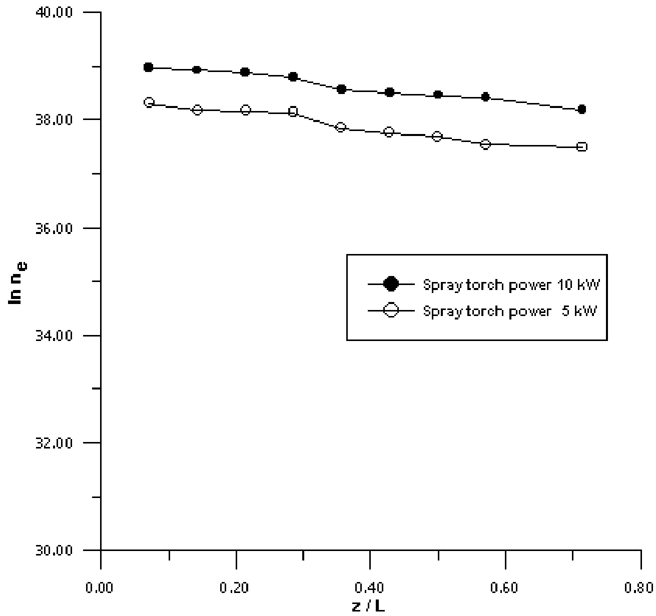

Fig. 6. Plot of $\ln n_e$ *vs.* z/l for plasma jet discharging in to air.

with fast macroscopic translation of plasma can maintain higher electron densities as measured data show using Stark broadening parameters.

4 Conclusion

The axial variation of electron number density (LOS) in thermal plasma jet has been measured using Stark broadened line profile of Ar-I transition (430 nm) and H_β (486.1 nm) transition. The experimental line profiles were deconvoluted to account for Doppler and instrumental broadening. The measured values of n_e from H_β Stark broadening and Ovassanikov's formula [15] agree with in the range of $\pm 7\%$. The measured values of n_e from Ar-I line Stark broadening give values on higher side. A curve fitted formula relating Ar-I line (430 nm) Stark broadening width and number density has been given. The use of Ar-I line to monitor electron density is important in processing application when addition of a small amount of hydrogen may be undesirable.

There is a large discrepancy between Stark broadened n_e values and those predicted by spectrometric temperature measurement. It can be seen from Figures 4 and 5 that plasma may be in LTE state only near the exit point and as one moves away in axial direction, deviation starts and will also depend on plasma torch power. However, it may be seen that electron number densities for spray torch measured using various diagnostic technique is in the range 10^{17} to $10^{16}/cm^3$. The higher electron densities obtained in plasma jet may be assigned to finite chemical reaction rate coupled with high exit velocity of plasmas.

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