Investigation on the Departures from Thermodynamic Equilibrium in a DC Plasma Jet

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Abstract

The distributions of the temperature and the electron density in a standard DC plasma spraying torch jet are measured at a power level of 16 kW using emission spectroscopy. The local values of the temperature were determined by three distinct spectroscopic methods: the 727.2 ńm argon neutral line, the stark-effect broadening of the H_β line of hydrogen and the Boltzmann method using 11 neutral argon lines. Results show progressively increasing deviations from LTE with decreasing pressure, increasing radial position and increasing axial distance away from the torch exit. Good agreement (±8%) is obtained between temperatures derived from the Boltzmann plot and those from the enthalpy probe measurements for electron densities greater than 1.3×10^{16} cm^{-3}.

Introduction

DC torches for plasma spraying and powder treatment perform an important role in plasma technology. Operation in a controlled atmosphere chamber changes the nature of the plasma - the jet containing visible shock waves is elongated, the gas velocity is increased, physical phenomena such as diffusion, chemically frozen flow and cold gas entrainment lead to deviations from LTE, especially when emission spectroscopy techniques are used for temperature assessment.

This paper is mainly concerned with the comparison of temperature profiles obtained from the spectroscopy measurements with those obtained from the calorimetric probe measurements. The enthalpy probe method is a reliable and meaningful diagnostic tool for investigating plasma properties in plasma jets since it measures the average temperature of the hot plasma gas and entrained cold gas. Incropera [1] has reported that emission spectroscopic methods are more accurate than enthalpy probe techniques for temperatures higher than 10, 000 K. Below this temperature, Pfender et al. [2] and Chen et al. [3] have shown that emission

spectroscopy data on the ArI 430.0 nm spectral line systematically indicate higher temperatures than enthalpy probes. On the other hand, Czernichowski [4] has demonstrated that the population relative to higher level excited states corresponds to the neutral species temperature as measured by Pitot tube experiments. The comparison of the different spectroscopic methods for temperature determination against the probe method will provide a set of limits for which emission spectroscopy may be reliably used. In this work, operating conditions were chosen such that LTE does not exist. Spectroscopic measurements were performed far away from the nozzle exit so as to be operating in the laminar-turbulent regime [3]. For experimental comparison purposes, this investigation was limited to a set of conditions under which enthalpy probe measurements had been carried out previously in our laboratory [5].

Experimental set-up

The main components of the experimental set-up are shown in Fig. 1. The DC plasma torch was fixed on the top apperture of a water-cooled reduced pressure controlled expansion chamber, equipped with quartz window for spectroscopic observations of the jet and a sealed scanning mechanism that could be installed at two axial levels for the enthalpy probe movement in the radial direction. The calorimetric probe and the spectroscopic assembly used to determine the local temperature profiles in the plasma jet has been described by Rahmane [5] and Sabsabi [6] respectively.

The DC plasma torch used was a 5 mm id. water cooled copper anode fitted with a thoriated tungsten cathode. The plasma gas mixture flow, which was essentially argon (<1% H_2), was set at 50 lpm. The plasma current was fixed at 400 A with a resulting arc voltage drop of 40 V. The chamber operating pressures was alternately set at 53 and 200 torr. Data were recorded at axial distances of 35, 45 and 55 mm downstream of the anode exit.

Temperature and electron density measurement

Local values of the temperature were determined by the three spectroscopic methods. A standard tungsten filament lamp was used to calibrate the absolute intensity of radiation measurement. The intensity of each line was obtained after subtraction of the adjacent continuum from the total signal. Local values of the emission coefficients were obtained by the Abel inversion of the measured radial intensity profiles.

In the first method, the temperature determination was made using the 727.29 nm atom spectral line. Calculations were based on the assumption of LTE. The temperature of the Ar-H_2 plasma was determined by means of the Saha-Boltzmann equation, the neutrality law and Dalton's law in conjunction with the measured local spectral emissivities.

The second method involved the use of the Boltzmann diagram. The following spectral lines, in nm, were selected: 727.29, 738.39, 750.38, 810.36, 842.46, 605.94, 675.28, 687.13, 549.59, 555.92 and 430.01.

The third method for determining the temperature, which is based on the assumption of local thermodynamic equilibrium, corresponds to the electron density

measured by the Stark broadening of the H_β line. The experimental procedure and analysis for determining the electron density has been described elsewhere [6].

Results and discussion

Temperature profiles determined from the excited atom density of the 727.29 nm ArI line, Tex, are shown in Fig. 2. The measured electron densities on the axis at 35 mm downstream of the nozzle exit are very similar ($\pm$ 6%), yet, the values of Tex at 200 torr is much larger compared to the 400 torr operation. The latter condition is the lower pressure limit for LTE to exist on the axis where significant deviation from LTE starts to appear for points furthest from the nozzle, beyond 25 mm [6]. At 35 mm and 400 torr, the electron density profile measured from the H_β lines is lower than that deduced from LTE considerations. For example, the value of Ne,lte at r=0 is smaller than $Ne_{H\beta}$ by 23 % and this difference increases as the distance from the axis increases. This discrepancy is larger than the experimental uncertainty which is estimated to be $\pm$ 15% for the $Ne_{H\beta}$ determination. The comparative approach, which as been used to establish LTE [7], asserts that LTE conditions do not hold for all the data points measured by emission spectroscopy in this work .

Temperatures determined from the various methods are compared in Fig. 3 for the 400 torr operation at 35 mm downstream. The temperature deduced for LTE conditions at $Ne_{H\beta}$ is Tne, the excited temperature for the 727.29 nm ArI line is represented by Tex, the temperature obtained from a Boltzmann plot is Tb and the temperature obtained using the enthalpy probe is given by Tp. Relatively good agreement between the values determined by the four methods is observed for various locations on the axis. There, the average deviation in the temperature measurement is only $\pm$ 3.5%. As seen above, this is misleading as far as LTE establishment is concerned since all temperature profiles obtained by different methods are systematically lower than the Tne profiles. On the axis, the probe temperature, which represents local gas characteristics, is lower than Tne by 6.5 %. This difference, which is of the same   order of experimental error in Tp determination, becomes more important as the radial distance increases.

Fig. 4 compares the different temperatures along the plasma axis at 400 torr. As expected, the difference between Tne and Tb is more pronounced at 45 mm downstream. This situation is further accentuated at lower pressure operations. Also noted on Fig. 3 and 4 is the proximity of Tb and Tp. The Boltzmann temperature is obtained in terms of the excited atom density Nm/gm associated with upper energy level of several neutral Ar I lines. The variation of the population of all the excited states of ArI used in this work in terms of the upper excited energy levels show that we have Boltzmann distribution and a state of equilibrium between the different excited states. This method is less constraining since it does not require the assumption of LTE. This partial equilibrium permits determination of the electron temperature. Considering that the uncertainty in the determination of Tb is $\pm$ 600 K, the agreement between the two temperatures extends to r=2mm. Beyond r>2 mm, deviation of Tp from pLTE values only cannot explain the observed differences. Knowledge is lacking on collisional-radiative processes, ambipolar diffusion effects and cold gas

entrainment. There is, however, earlier research where the comparison is made between plasma spectral diagnostics, using the pseudo-equilibrium approach, and the determination of the neutral species temperature with a Pitot tube, extended to much lower temperatures [4]. The comparative agreement reported by the authors probably rests on the choice of spectral lines used and the nature of the argon plasma produced in their laboratory.

An overall picture of the extent of departures from LTE is shown in Fig. 5a and 5b where the temperatures determined by different methods are compared as a function of the measured electron density at 400 torr and 200 torr respectively. Since the Stark broadening method is not based on a state of equilibrium, it can therefore provide an indication of the non-equilibrium effects. As seen in Fig. 5a, the plasma is close to equilibrium conditions on the axis at 400 torr only where the difference between the gas temperature, T_p, and the electron temperature, T_b, is 650 K. The measured electron density of 1.76×10^{16} cm^3 is close to the generally recognized lower limit of 2×10^{16} cm^{-3} for a plasma to approach a complete state of LTE [8,9]. If the uncertainty limits of the measurements are taken into account, relatively good agreement between the electron temperature, as given by a Boltzmann plot, and the neutral particle temperature as given by the enthalpy probe, extends down to an electron density of about 1.3×10^{16} cm^{-3} for both pressure operations and axial positions.

Conclusions

The application of emission spectroscopy for plasma diagnostics must be used with care in plasma regions where the entrainment of the surrounding gas plays an important role. The selection of an appropriate set of upper level energy populations of the neutral argon atom yields an electron temperature closer to the neutral gas temperature as provided by an enthalpy probe than would the LTE assumption. Since enthalpy probe is generally recognized as a reliable tool for measuring temperatures between 2,000 K and 10,000 K, it may be employed as a basis for comparing the temperature determined by the different spectroscopic methods and for establishing limits of applicability of spectroscopic methods. The validity range in which the gas temperature and the electron temperature agree, within the experimental error, show an electron density greater than 1.3×10^{16} cm^{-3}.

References
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Figures
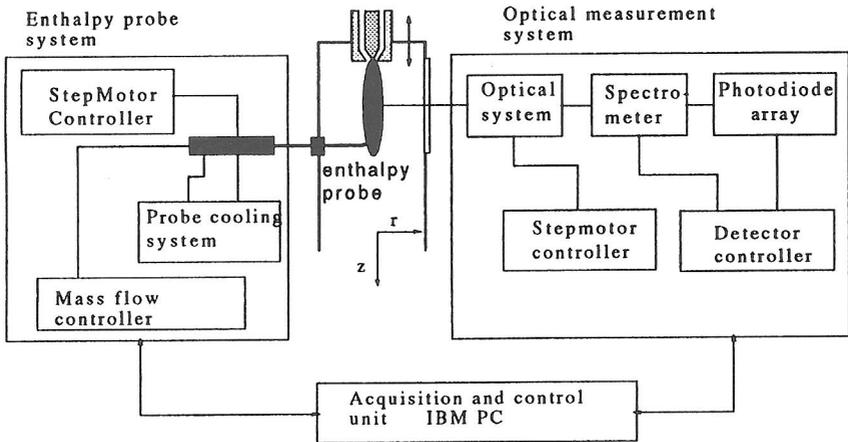
Fig. 1   Schematic diagram of the measurement system
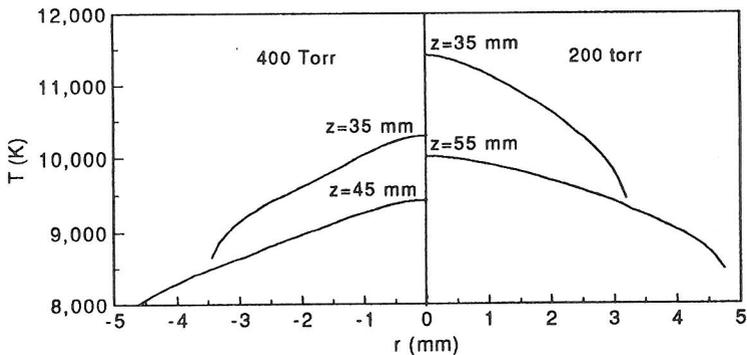
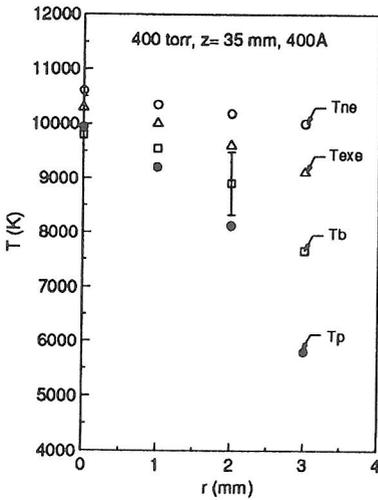Fig. 2   Radial temperature profiles determined from the 727.29 nm neutral argon line

Fig.3 Comparison of radial temperature profiles determined from different methods 35 mm downstream of the plasma jet nozzle exit at 400 torr
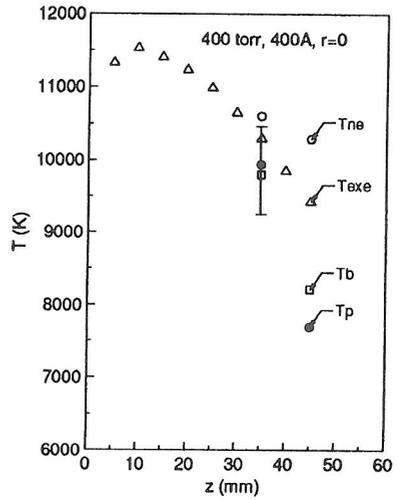
Fig.4 Comparison of axial temperature profiles determined from different diagnostic methods on the plasma jet axis at 400 torr
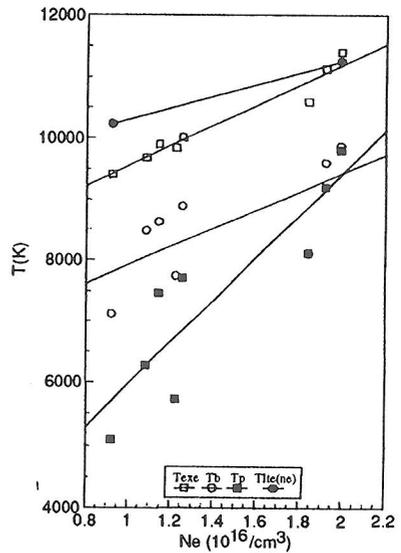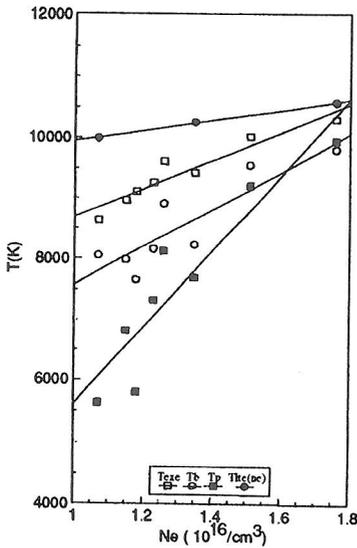

Fig. 5 Comparison of the different temperatures as a function of the measured electron density (left 400 torr, right 200 torr).