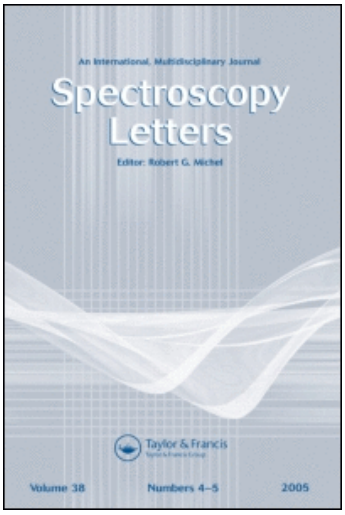

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Influence of Spectral Line Deconvolution on Measurement of Excitation Temperature in a Wall-Stabilized Thermal Plasma by Optical Emission Spectroscopy

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ABSTRACT

The measurement of temperature in a wall-stabilized thermal plasma is done by optical emission spectroscopy. The absolute intensity line method, relative intensity line method, and plot of the Boltzmann function are used. Another method, based on the measurement of electron density and calculations of concentrations, is also used. The influence of spectral line deconvolution on temperature profiles is studied. The results obtained by the four methods are compared. The influence of others parameters, such as the accuracy of spectroscopic constants, and the influence of the theoritical calculations of concentrations are also discussed. These

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methods employ Ar I, C I and O I lines to measure the temperature of an $Ar-CO_2$ mixture plasma produced in a wall-stabilized arc.

Key Words: Emission spectroscopy; Spectral line deconvolution; Temperature measurement; Temperature accuracy; Thermal plasma.

I. INTRODUCTION

Thermal plasmas are applied widely to many applications, such as the decomposition and synthesis of materials,[1] hydrogenation of silicon materials,[2] and plasma spraying.[3] In several arc applications, the arc energy transfer toward the anode is used for the treatment of metallic material used as the anode: welding, cutting,[4] or waste treatment.[5] Plasma parameters are not always well known, and calculated parameters must be checked experimentally. An important parameter of the plasma is its temperature T. The studied plasmas are characterized by a temperature ranging from less than 1 eV (11,600 K) to several electronvolts. At these temperatures, laser induced fluorescence is not an appropriate technique. Optical emission spectroscopy can be used to measure the plasma temperature.[6] We use, for an $Ar-CO_2$ thermal plasma, the absolute intensity line, the relative intensity line methods, and the plot of the Boltzmann function, on atomic lines of argon, carbon, and oxygen. Temperature is also deduced from the measurement of electron density and calculations of concentrations. This method is called the electron density method. All these methods need an accurate determination of the intensity of the spectral lines. Due to the different species of the plasma, it is often necessary to deconvoluate the spectral lines.

The aim of this work is to study the effect of line deconvolution on the temperature profiles. The results obtained by the four methods are compared. The effects of the accuracy of the spectroscopic constants chosen in the four cases are also discussed. We can note that the absolute intensity and electron density methods use calculations which presuppose the local thermal equilibrium (LTE). On the other hand, the relative intensity method and the plot of the Boltzmann function do not use these calculations and so do not require the hypothesis of LTE. For Boltzmann plot, the temperature accuracy depends on the slope accuracy, which is discussed. For the absolute intensity method, the influence of the theoretical calculation of concentrations used to determine the temperature is evaluated. The influence of the parameters on the relative intensity line method are also evaluated. The results, obtained on a pure argon and an $Ar-CO_2$ mixture plasma, are presented after a deconvolution known as Abel's inversion[7] due to the cylindrical symmetry of the arc.

II. EXPERIMENT

The experimental device has been described in a previous paper.[8] It is shown in Fig. 1. The wall-stabilized arc is produced in a modified Maecker chamber, which has been optimized.[9] It is an 85-mm-long arc chamber, consisting of a stack of six water-cooled copper plates, separated by thick insulating Celoron or Bakelite spacers as presented in Fig. 2. The copper plates have a 4 or 6 mm central hole diameter. A tight chamber is required to avoid pollution by the surrounding air. In the center of the chamber, 5-mm-thick quartz plate plane surfaces allow the observation of the radiation emitted radially by the arc and a disc on the top of the 20-mm-diameter chamber allows us to observe in the direction of the arc axis. The quartz has an optical bandwidth from IR to UV. Three flow rates allow to control the composition and velocity of the gas injected in the chamber. Two of them are used to inject the same quantity of pure argon near each tungsten electrode to protect them. The other one controls the injection of the gas studied, for example, CO_2 gas. The gas is ejected out of the chamber by eight holes between the center and the top of the chamber. The discharge current is adjusted with a DC generator from 30 to 80 A. The light emitted is observed perpendicularly to the arc column and focused on the entrance slit of a 1.5-m Czerny-Turner monochromator equipped with a 2400 lines/mm grating coupled with a CCD (512 × 512 pixels). The record of the different lines needs few minutes. A test of time reliability has been done. After switching the arc, first the intensities of the different spectral lines decrease with time. A 45-min working

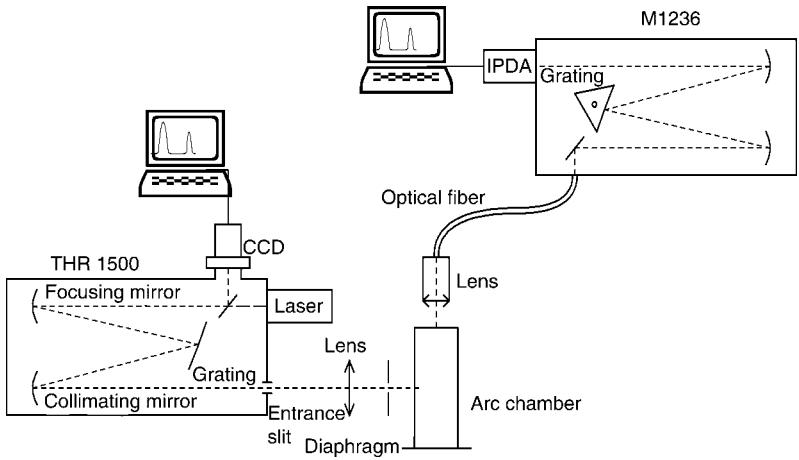

Figure 1. Schematic diagram of the experimental setup.

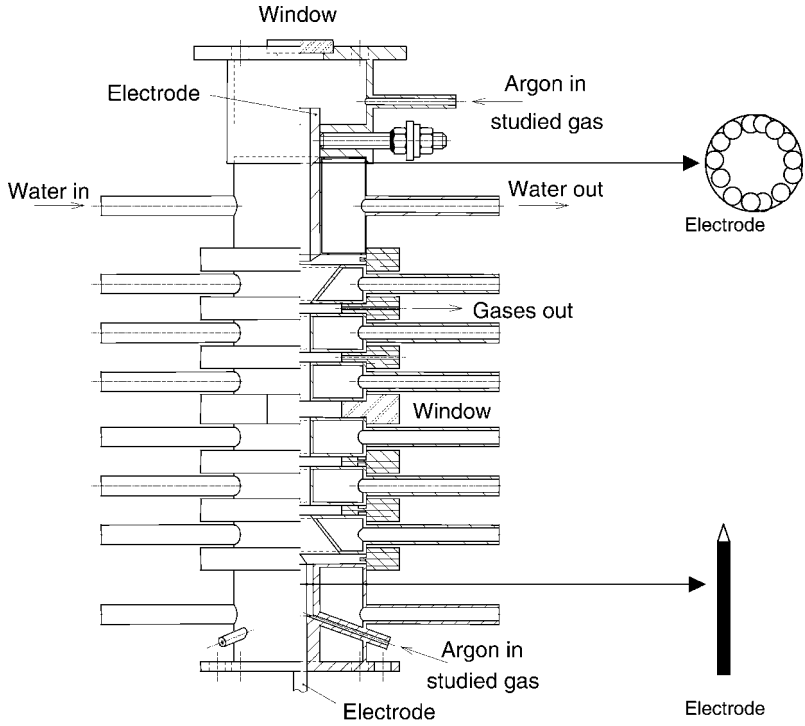

Figure 2. Sectional view of the plasma chamber.

duration is necessary to obtain constant intensities for the different spectral lines in all the wavelength range. The voltage evolution is similar. All the measurements are performed after a 60-min working duration which assured a very good stability of atomic lines.

III. TEMPERATURE MEASUREMENT PRINCIPLES

Four temperature measurement methods based on optical emission spectroscopy are used: the absolute and relative intensity line methods, the plot of the Boltzmann function, and the electron density method. The principles of these methods have been described already.[10–12] These methods make it necessary to know the coefficient of local intensity. In an arc discharge with a cylindrical symmetry, the coefficient of local intensity is deduced by using the inverse transform known as Abel's transform. This Abel's inversion used for the four methods has been explained in detail in a previous paper.[8]

The program has been tested with test functions used by several authors.[13,14] From these test functions, the mean discrepancy due to the calculation method can be estimated at 0.4%.

Without self-absorption, under LTE, the temperature can be deduced from the absolute intensity of atomic lines:

$$T = \frac{E_m}{k \ln((hc/4\pi)(1/\lambda_{mn})(g_m A_{mn}/I)(n(T)/Q(T)))} \tag{1}$$

where I is the intensity of the atomic line, $n(T)$ the atom density, ν_{nm} the frequency, A_{mn} the transition probability, g_m the statistical weight, E_m the excitation energy, and $Q(T)$ is the partition function of emitted level.

Using the same notation as for the absolute intensity method, the temperature can also be deduced from the relative intensity of two atomic lines I_{mn} and I_{pq}, respectively, from electronic level m to electronic level n and from electronic level p to electronic level q:

$$T = \frac{E_m - E_p}{k \ln((I_{pq}/I_{mn})(g_m A_{mn} \lambda_{pq}/q_p A_{pq} \lambda_{mn}))} \tag{2}$$

The accuracy is better when the difference between the energies of the two upper levels of the transitions is large. Unfortunately, in some plasmas (like argon plasma at a temperature lower than 15,000 K), the excited levels observed do not allow a good accuracy.

The plot of the Boltzmann function does not require the calculation of the densities of the different species vs. temperature. Relation (1) can be written:

$$\ln\left(\frac{\lambda I}{g_m A_m}\right) = -\frac{E_m}{kT} + \ln\left(\frac{4\pi Q(T)}{hcn(T)}\right) \tag{3}$$

Assuming a Boltzmann distribution, the temperature T can be deduced from the slope of the plot of the Boltzmann function. All the measurements have been done with oxygen lines except for pure argon plasma.

The electron density method allows the calculation of the temperature and its corresponding atom and ion concentrations from electron density measurements. The electron density measurement is based on the Stark effect broadening. The measurements have been done with an H_α hydrogen line ($\lambda_H = 656.26$ nm) and an argon line ($\lambda_{Ar} = 430.01$ nm) for pure argon plasma. The results are similar. Accuracy is better for H_α line. In $Ar-CO_2$ plasma mixtures, the Ar line is disrupted by other lines. Therefore, the H_α hydrogen line profile is used. Broadenings of spectral lines can be due, for thermal plasmas, to apparatus function, Doppler effect, and Stark effect. Apparatus function and Doppler effect are gaussian profiles. The Stark effect is reduced to a lorenztian profile if we only take the electron broadening

into account and neglect the effect of ion broadening.[15] In the static ion approximation, the effect of ion broadening depends on the distribution of the electric microfield.[16] In this model, the motions of the perturbing plasma ions during the radiative process are ignored. On the other hand, the dynamic ion model includes the effects of ionic motions in the line shapes.[17] This effect results from the fact that the free ions of the plasma, which perturb the radiator, have enough time to move during the radiative process. Moreover, as the radiator is charged, the interactions between them and the other plasma charges are correlated to the variations of the radiator velocities.[18] These two models lead to a non-analytical profile. In dense plasma, Doppler and instrumental profiles are negligible and the Stark broadening can be fitted by a lorentian shape.[19] In our experimental conditions, a good fit of the experimental profiles are obtained reducing the Stark effect to a lorentian profile. The convolution of the gaussian and the lorenztian profile is a Voigt profile. In order to distinguish the different causes of broadening, a numerical deconvolution is done and the Stark effect broadening is isolated. The width at half maximum is compared with those calculated by Smith et al.[20] for different temperatures and electron densities, so the electron density is deduced. The knowledge of n_e allows us to obtain the excitation temperature. The absolute intensity method and the electron density method need the calculations of the different species of the plasma vs. temperature assuming LTE. Figure 3 presents the composition for an $Ar-CO_2$ mixture (volume percentage: 90% Ar-10% CO_2) for temperature ranging from 4000 to 14,000 K. The accuracy of the electron density method, which depends on the determination of the width at half maximum, the Smith et al. calculations, and the concentration calculations can be evaluated at about 10%.

IV. RESULTS AND DISCUSSION

All the measurements are performed at atmospheric pressure. The oxygen, argon, and carbon lines used are listed in Table 1 with their energy level transitions, statistical weights, and transition probabilities.[21,22]

An absolute calibration on intensity is made using a calibrated tungsten ribbon lamp. The influence on the temperature accuracy of the absolute calibration accuracy is weak and can be neglected. For all spectral lines, the continuum is subtracted from the measured signal. A weak difference on the intensity of some spectral lines can generate a non-negligible accuracy on the temperature. In pure argon, overlapping between spectral lines is less important. Although the consequence of the deconvolution is less important, it is necessary to exactly determinate the area of each spectral lines. A deconvolution of the profile of each line is done. After this deconvolution for each

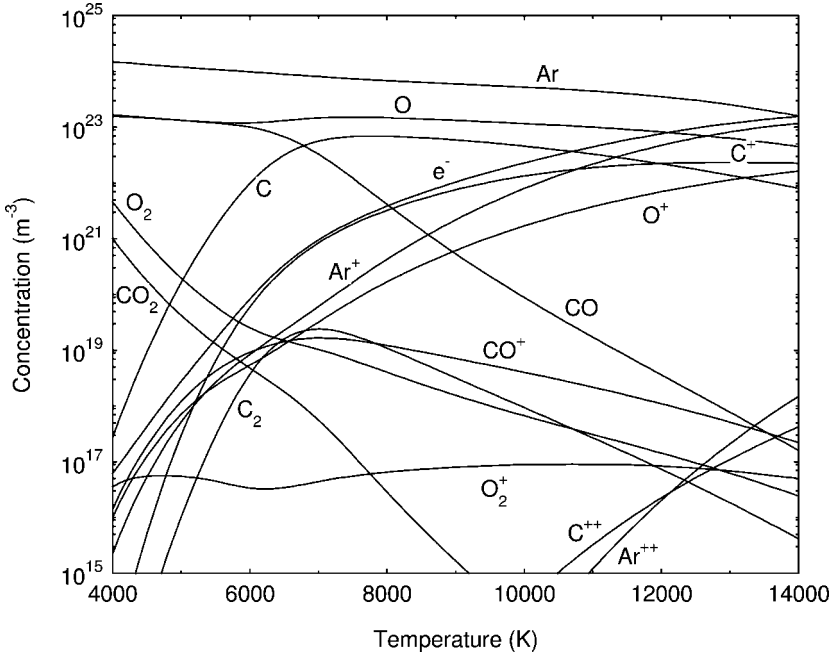

Figure 3. Plasma composition vs. temperature in an Ar–CO_2 mixture: 90 vol.% Ar and 10 vol.% CO_2.

line, the spectrum is recalculated and compared with the original spectrum in order to confirm the deconvolution. Figure 4 shows this deconvolution for some oxygen lines. The influence of this deconvolution on the accuracy of the temperature is studied for each method.

First, the influence of the accuracy of the different experimental points on the slope of the plot of the Boltzmann function and on the temperature as well is presented. The temperature is calculated using 13 oxygen lines. Table 2 presents the temperature at the center of the arc column ($r = 0$) and the corresponding correlative coefficient of the slope of the Boltzmann plot (R). In order to simulate the sensitivity in the calculation of the original spectrum, the temperature is calculated for different values of the areas of different oxygen lines. The corrections of the line areas are optimized values that represent the relative difference between the original spectrum and the calculated spectrum. The temperature T_{nc} is obtained without any correction of the line area and R_{nc} is the corresponding correlative coefficient. T_i are the different temperatures obtained with different values of O lines areas and R_i the corresponding correlative coefficients. T_1 is obtained with a correction of 5% on the area of the O line ($\lambda = 665.3834$ nm);

Table 1. Wavelength, energy, statistical weight, transition probability, transition probability accuracy for oxygen, argon, and carbon atomic lines.

	Wavelength (nm)	Energy of upper level (eV)	Statistical weight	Transition probability ($10^8\,\mathrm{sec}^{-1}$)	Transition probability accuracy (%)
O I	394.7295	12.2861984	7	0.00326	25
O I	394.7481	12.2860501	5	0.00325	25
O I	436.8258	12.3588694	3	0.0066	25
O I	645.5977	12.6608615	5	0.0331	10
O I	665.3834	16.2348592	1	0.6	10
O I	747.1404	15.7821574	3	0.0114	25
O I	747.3241	15.7817496	5	0.102	10
O I	747.6439	15.7810401	7	0.408	10
O I	747.7234	15.7821574	3	0.17	10
O I	747.9074	15.7817496	5	0.306	10
O I	748.0671	15.7821574	3	0.226	10
O I	777.1944	10.7409353	7	0.34	10
O I	777.4166	10.7404795	5	0.34	10
O I	777.5388	10.7402289	3	0.34	10
O I	788.6273	15.9437434	5	0.37	10
O I	795.0803	14.0996633	7	0.331	10
O I	795.2159	14.1003765	5	0.313	10
Ar I	415.8591	14.5289187	5	0.014	10
Ar I	430.0101	14.5060729	5	0.00377	10
Ar I	451.0734	14.575954	1	0.0118	10
Ar I	470.2317	14.464001	3	0.00109	10
Ar I	565.0704	15.1005492	1	0.032	10
Ar I	696.543	13.3278619	3	0.0639	10
Ar I	727.2935	13.3278619	3	0.0183	10
Ar I	738.398	13.3022323	5	0.0847	10
Ar I	750.3868	13.4798917	1	0.445	10
Ar I	801.4785	13.0948773	5	0.0928	10
C I	477.174	10.085373	5	0.012	50
C I	505.217	10.138166	5	0.017	50
C I	538.04	9.988524	5	0.016	50

T_2 with a correction of 10% on the same line; T_3 with a correction of 5.8% on the oxygen triplet lines ($\lambda = 777.5388\,\mathrm{nm}$, $\lambda = 777.4166\,\mathrm{nm}$, $\lambda = 777.1944\,\mathrm{nm}$); T_4 without taking into account the O line $\lambda = 436.86\,\mathrm{nm}$ which was far from the plot. Raskovic et al.[23] have estimated the error of the slope of the Boltzmann plot from 2% to 5.2% in the temperature determination of an argon stabilized U-shaped DC arc burning.

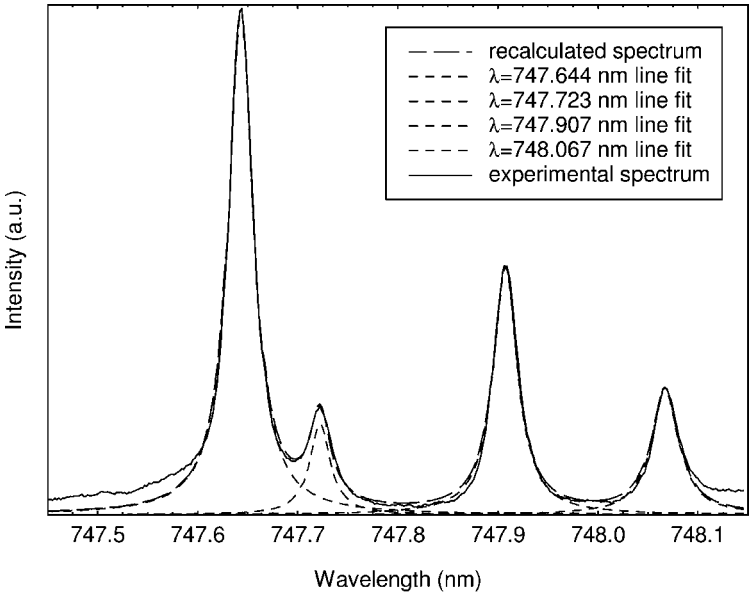

Figure 4. Profiles of oxygen lines ($\lambda = 747.644$ nm, $\lambda = 747.723$ nm, $\lambda = 747.907$ nm, $\lambda = 748.067$ nm): emitted lines (full curve), deconvoluted lines (dashed curve), and recalculated spectrum using deconvoluted lines (chain curve). 90 vol.% Ar–10 vol.% CO_2 mixture, plasma diameter $\phi = 4$ mm, $I = 30$ A.

Figure 5 presents the Boltzmann plot for the non-corrected areas and for a correction of 5.8% on the oxygen triplet lines. We can note the very weak influence of the middle points on the slope. Conversely, the extreme points have a great influence on the slope, particularly the oxygen triplet lines at $\lambda = 777$ nm. A 5.8% difference on the oxygen triplet line areas (first abscissa

Table 2. Temperatures and correlative coefficients of the slope of the Boltzmann plot on the axis of the plasma column, 90 vol.% Ar − 10 vol.% CO_2 mixture, plasma diameter $\phi = 4$ mm, I = 30 A.

T_{nc} (K)	R_{nc}	T_1 (K)	R_1	T_2 (K)	R_2	T_3 (K)	R_3	T_4 (K)	R_4
107,40	0.9927	10,233	0.9928	9,875	0.9917	9,567	0.9967	10,775	0.9982

Note: T_{nc}: non-corrected area; T_1: correction of 5% of the O lines area $\lambda = 665.6834$ nm; T_2: correction of 10% of the same O line; T_3: correction of 5.8% of the O triplet line areas; T_4: without the O line $\lambda = 436.826$ nm; R: correlative coefficient of the Boltzmann plot slope; the indexes have the same meaning as for T.

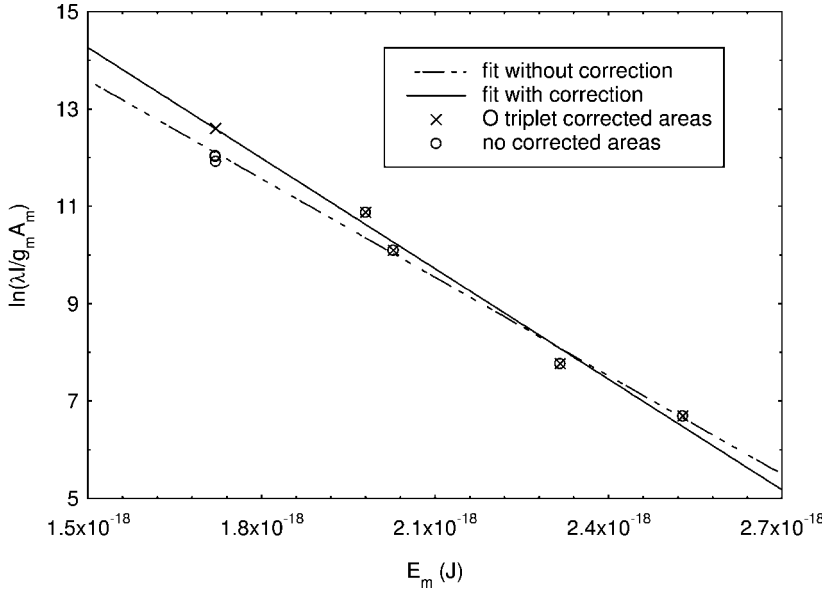

Figure 5. Boltzmann plot with oxygen lines for temperature measurements in a 90 vol.% Ar–10 vol.% CO_2 mixture, plasma diameter $\phi = 4$ mm, $I = 30$ A.

points) can involve an 11% difference on the temperature. The great influence on the slope of some points is pointed out by Se Youn Moon et al.[24] The measured excitation temperature of Ar I lines, in an atmospheric microwave-induced plasma, is obtained using a Boltzmann plot without taking into account some spectral lines.

In Table 3, we compare the temperature vs. radial co-ordinate of the plasma column obtained without correction of the line areas (T_{nc}) and with a correction (T_c) on the areas of the oxygen triplet lines ($\lambda = 777$ nm). All the other line areas are similar for the two calculations. The correlative coefficients R show very good linear relationship between $\ln(\lambda I/g_m A_m)$ and E_m. The difference between the two temperature ranges from 10.4% to 15.5%. With regard to R, the correlation is better with the correction of the line areas whatever the radial position is.

Figure 6 presents the temperature vs. the radial co-ordinate of plasma column obtained without corrections of the oxygen line areas and with deconvoluated oxygen lines. On the axis of the column ($r = 0$), the accuracy calculated with the deconvoluated lines are 5% when it is 20% with the non-corrected lines.

Table 3. Temperature and correlative coefficient vs. radial co-ordinate of the plasma column, 90 vol.% Ar–10 vol.% CO_2 mixture, plasma diameter $\phi = 4$ mm, $I = 30$ A.

Radial co-ordinate (mm)	T_{nc} (K)	R_{nc}	T_c (K)	R_c
0	10,740	0.9927	9,567	0.9967
0.1	10,742	0.9927	9,455	0.9968
0.2	10,575	0.9931	9,472	0.9974
0.3	10,491	0.9937	9,355	0.9977
0.4	10,374	0.9937	9,143	0.9981
0.5	10,285	0.994	8,907	0.9983
0.6	10,166	0.994	8,913	0.9987
0.7	10,049	0.9938	8,696	0.999
0.8	9,896	0.9936	8,689	0.9992
0.9	9,721	0.9936	8,219	0.9992
0.10	9,537	0.993	8,155	0.9994
0.11	9,359	0.993	8,030	0.9995
0.12	9,188	0.9923	7,845	0.9993

Note: T_{nc}, from non-corrected areas of the oxygen triplet lines areas and T_c, after correction of the oxygen triplet lines areas.

Using the absolute intensity of atomic lines, the discrepancy due only to transition probability (Table 1) gives less than 1.5% for s_T/T (s_T is the standard deviation). When the temperature is deduced from Eq. (1), the logarithm term reduces the error due to the determination of the line area and weakens the deconvolution of the spectral line error on the temperature to 2%. Using the absolute intensity method, the temperature is deduced from the oxygen, carbon, and argon atoms concentration calculations vs. temperature (Fig. 3) under LTE assumption. Departures from LTE can exist, so the main cause of accuracy of the temperature is due to the accuracy of the concentration calculations.

The accuracy of the relative method depends on the accuracy of the spectroscopic constants, the line areas, and the difference of energy between the two upper levels of the transitions.[11] The difference of energy $\Delta E = |E_m - E_p|$ between the two upper levels of the transitions considered depends on the gases studied. Our experimental set up allows us to detect wavelengths ranging from 300 to 800 nm. In some plasmas, for our temperature range, upper atomic levels are not populated enough to be well detected and used for measurements. In our pure argon plasma, the different lines which can be detected have a ΔE lower than 2 eV. Due to this low value, the accuracy of this method is less satisfactory than the other ones. In Ar–CO_2 mixture, with a ΔE near 5.5 eV for O lines, we obtain a better degree

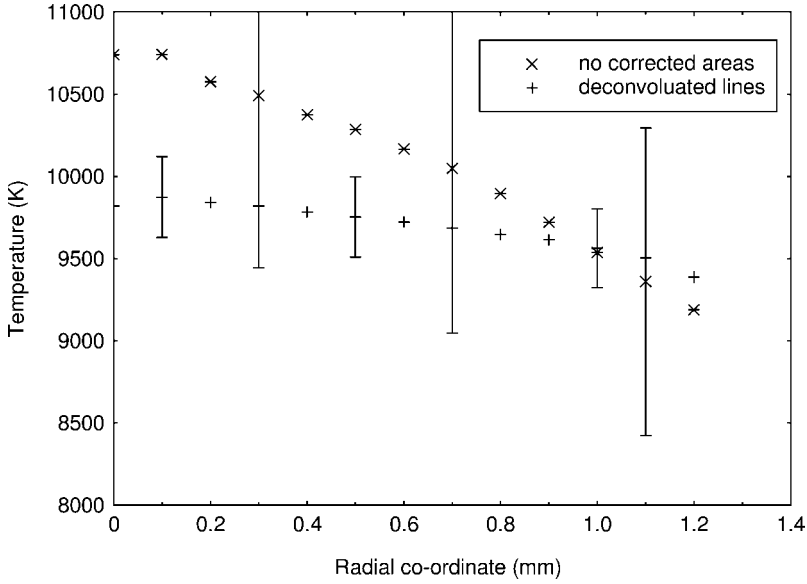

Figure 6. Excitation temperature vs. radial co-ordinate of the plasma deduced from non-corrected areas and deconvoluated oxygen lines in an $Ar-CO_2$ mixture: 90 vol.% Ar, 10 vol.% CO_2, $I = 50$ A.

of accuracy. Nevertheless, even with the deconvolution processes, the accuracies obtained with the relative intensity lines are worse than those obtained by the other methods.

The electron density method needs the deconvolution of the spectral lines in order to distinguish the different causes of broadening and to isolate the Stark effect broadening. It is impossible to use this method without deconvolution and so to calculate the contribution of the deconvolution of the spectral lines. The accuracy of the temperature depends on the calculations of concentrations with LTE assumption, the accuracies of the Smith et al. calculations,[20] the determination of the full width at half maximum of the spectral lines, and so the deconvolution of the spectral lines. The accuracy can be estimated to 10%.

Figure 7 presents the excitation temperature vs. the radial co-ordinate r of the plasma column for $I = 50$ A. In pure argon plasma, absolute intensity method and Boltzmann plot method use argon lines. In $Ar-CO_2$ mixture, argon, oxygen, and carbon lines are used; the temperatures obtained are similar. Oxygen atomic lines are used to construct Fig. 7. In the side of the discharge, the spectral lines are less intense; so the accuracy of the

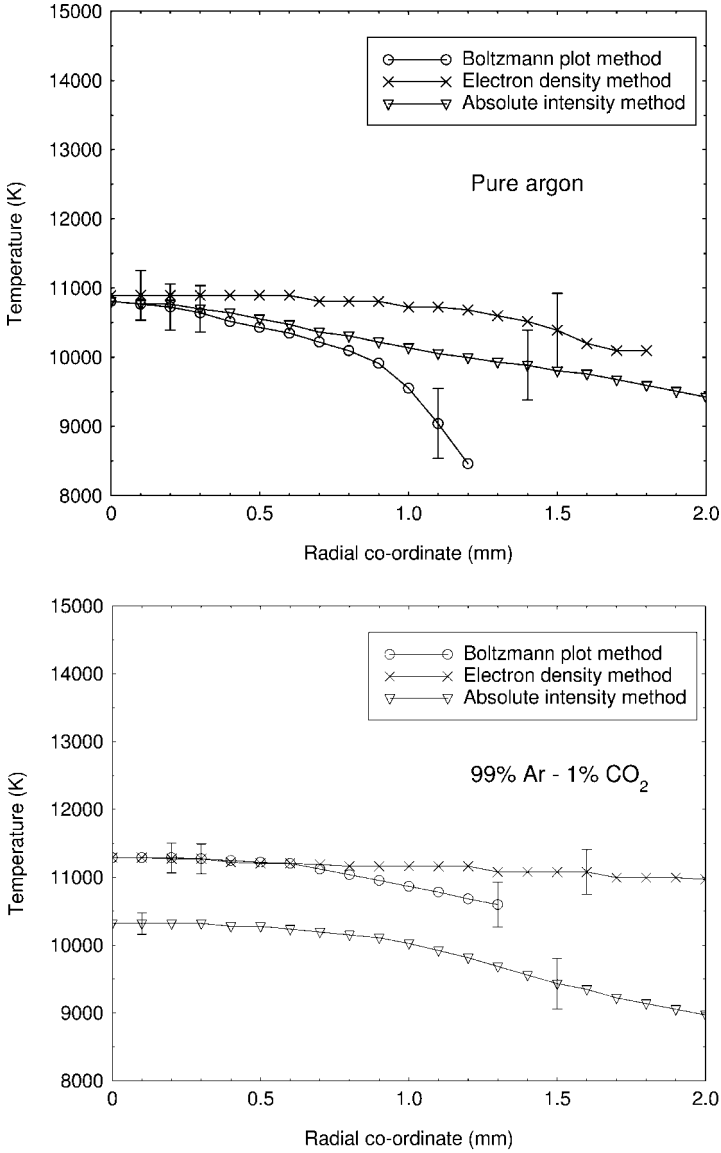

Figure 7. Excitation temperature vs. radial co-ordinate of the plasma, $I = 50$ A, plasma diameter $\phi = 4$ mm. Comparison of absolute intensity method, Boltzmann plot method and electron density method. (a) Pure argon plasma, (b) Ar–CO_2 mixture: 99 vol.% Ar, 1 vol.% CO_2, (c) Ar–CO_2 mixture: 95 vol.% Ar, 5 vol.% CO_2, (d) Ar–CO_2 mixture: 90 vol.% Ar, 10 vol.% CO_2.

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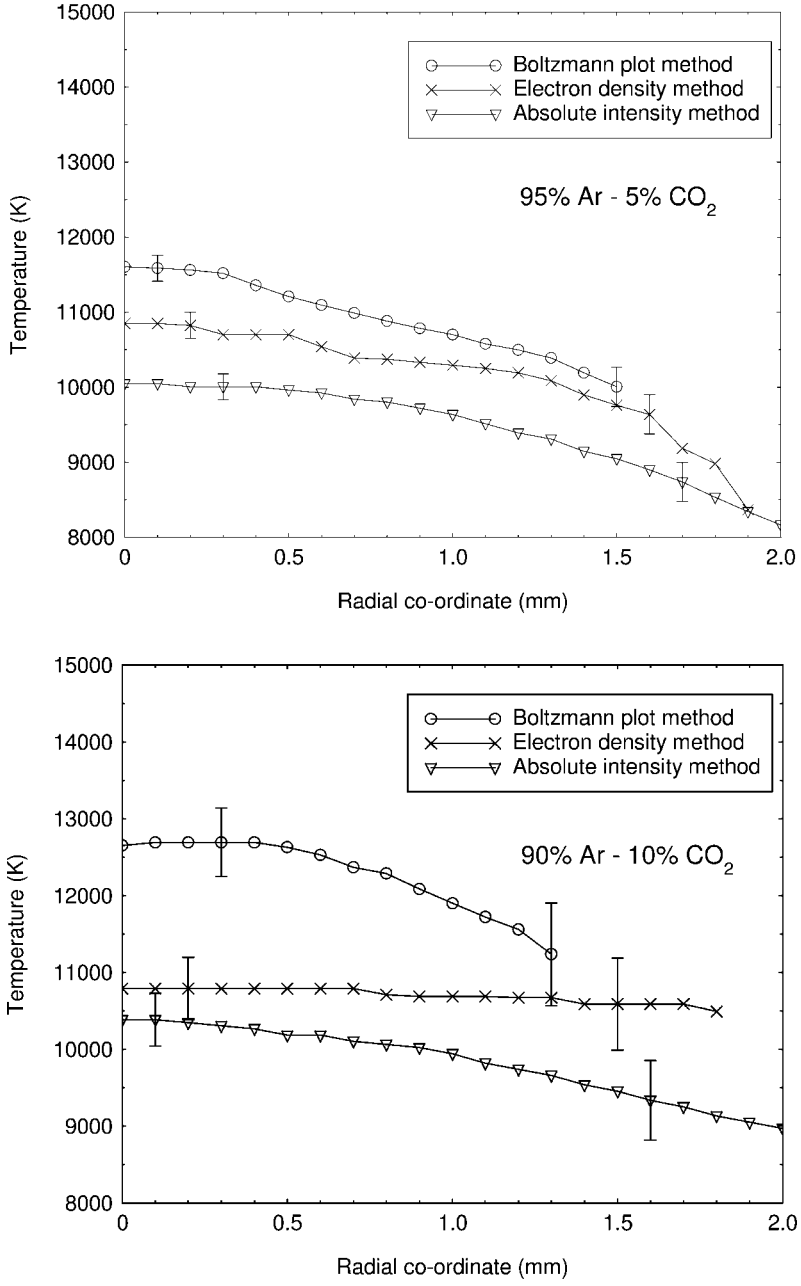

Figure 7. Continued.

temperature is lower. The temperature profiles, as determined using the absolute intensity and the electron density methods, cover a wider radial distance than does the Boltzmann method. In pure argon [Fig. 7(a)], the results of the three methods are similar in the center of the plasma column ($r \leq 1$ mm) where LTE exists. On the side of the column where departures from LTE can exist,[25] differences appear. Temperatures obtained with the electron density method and the absolute intensity method which assume LTE are close to one another. On the other hand, the Boltzmann plot method gives lower temperatures. In $Ar-CO_2$ mixtures [Fig. 7(b)–(d)], temperatures obtained with the absolute intensity method are always lower than those determined by the Boltzmann plot method. The electron density method gives middle values. In $Ar-CO_2$ mixtures, absolute intensity method uses oxygen and carbon atomic lines to calculate the temperature. In these plasmas, demixtion phenomena can appear and in the side of the discharge, concentrations of carbon and oxygen atoms are higher. The oxygen and carbon atomic lines recorded are more intense. So the oxygen and carbon concentrations measured are higher than those calculated with the hypothesis of homogeneity of the plasma. For temperature ranging from 8000 to 14,000 K, concentrations of carbon and oxygen atoms decrease when the temperature increases (Fig. 3). So temperature calculated using absolute intensity method is lower than temperature calculated using Boltzmann plot method.

V. CONCLUSIONS

The optical emission spectroscopy gives a non-perturbative and accurate measurement to determine such parameters as concentrations or temperatures. The effect of line deconvolution on the temperature profiles has been studied and the four methods and their accuracies have been compared. The relative intensity line method gives a lower degree of accuracy than the other ones due to the transitions observed in our plasma. Nevertheless, it can be used to get a rapid estimation of the temperature. The absolute intensity line method, the Boltzmann plot, and the electron density method allow us to obtain accurate temperatures subject to certain precautions: spectroscopic constants must be selected correctly and line areas must be correctly evaluated and spectral lines often require a deconvolution of their profile. Under these conditions, the Boltzmann plot, absolute intensity of atomic lines, and electronic method can be used for a temperature diagnosis of thermal arc plasmas. The results do not suggest which spectroscopic method to use to characterise a given plasma. In the case of pure argon plasma, the Boltzmann plot method seems to be the best. As a matter of fact, this method does not

require LTE, hypothesis which is not always verified on the sides of the column. But in mixtures, the results are more complex making more difficult, clear, and accurate choice. The absolute intensity method seems to undervalue the temperature due to the hypotheses attached to the method. The Boltzmann method does not require an LTE hypothesis but it is more sensitive to the overlapping of spectral lines. An accurate deconvolution of spectral lines is necessary; nevertheless the main differences and discrepancies between the methods are due to the assumptions as LTE.

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