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TWO APPROACHES IN DETERMINATION OF COPPER VAPOURS CONTENT IN THERMAL PLASMA BY OPTICAL DIAGNOSTICS

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This work presents a comparative study of two diagnostic approaches for determining copper vapour admixtures in the thermal plasma of an electric arc discharge. The emission of plasma of arc discharge between copper electrodes was analysed using optical emission spectroscopy (OES) under discharge currents of 3.5 and 30 A. Two sets of plasma parameters were used as inputs for equilibrium composition calculations: (1) radial distributions of excitation temperature and copper atom number density, and (2) radial distributions of excitation temperature and electron density. The excitation temperature was determined using the Boltzmann plot technique, while electron density was obtained from spectral line broadening using Fabry-Pérot interferometer and by applying the Saha equation. The consistency of the calculated plasma compositions using both input sets demonstrates the applicability of indirect diagnostic methods for evaluating the erosion resistance of electrode materials.

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INTRODUCTION

The presence of the metal vapour admixtures, caused by electrodes erosion, in the plasma of arc discharges plays a crucial role in setting its parameters such as temperature and electron density. For technological applications that use such discharges between melting electrodes (both single and multicomponent), these plasma parameters are primarily an indicator of their energy efficiency.

Another example of great importance role of material admixtures is the problem of plasma interaction with surface of walls in fusion reactor. This is one of the reasons for the intensification of the research and development of so-called plasma-facing materials [1], which are exposed to the plasma within which nuclear fusion occurs. That is why the issue of developing and improving methods for monitoring the presence and determining the content of electrode material vapours in the investigated plasma is relevant [2]. Traditionally, optical spectroscopy methods are used for this purpose [3]. However, this is a complex task that involves, first of all, clarification of the thermodynamic state of the plasma [4] for the optimal choice of methods for determining its parameters [5]. In addition, the use of verified spectroscopic data is crucial for the use of the above-mentioned diagnostic methods [6, 7].

The aim of this work is to analyse and apply various approaches to determine the content of metal vapours in the plasma of electric arc discharges by optical diagnostics.

1. EXPERIMENTAL SETUP

A free-burning arc was ignited in an air atmosphere between the flat surfaces of uncooled vertically oriented rod copper electrodes (diameter of 6 mm) at a current of 3.5 and 30 A. A discharge gap was 8 mm. Optical scheme of the experimental setup is shown in Fig. 1.

This scheme makes it possible to measure the spatial distribution of the observed intensity in a selected cross-section of the arc discharge. In this work, the central cross-section of the positive column of the arc discharge (at a distance of 4 mm from the edge of the electrodes) was investigated. The arc column is located in the focal plane of the F1 lens, which forms the parallel beam of the plasma radiation. The F2 lens is formed an image of the arc discharge on the entrance slit of the monochromator MDR-12. Since the entrance slit is vertically oriented the Dove prism is used to rotate the image by 90°. Specially developed program interface was used to control the experiment, recording and data treatment.

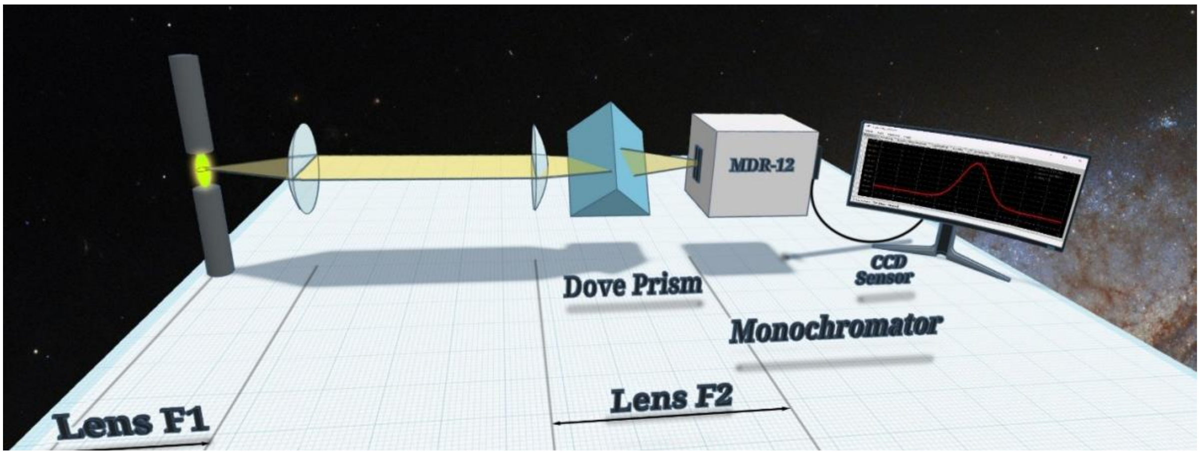

Fig. 1. Optical scheme of the experimental setup

2. EXPERIMENTAL TECHNIQUES

Two approaches for determining the plasma composition were investigated in this work. The plasma composition, in turn, serves to determine the spatial distribution of the aforementioned metal vapour admixtures [6]. The first approach is performed using the experimentally determined radial distributions of the plasma temperature *T(r)* and the number density of metal atoms (in this work copper) $N_{Cu}(r)$. The second approach is to utilize the radial distribution of the electron density $N_e(r)$ instead of the number density of metal atoms.

The method of Boltzmann plots based on radial distributions of absolute emission intensities of spectral lines of atoms of electrode origin is used for realisation of the first approach. This method allows one to determine both the plasma temperature (in case of realisation of local thermodynamic equilibrium) and the number density of radiating particles (in particular, metals). The absolute intensities themselves can be obtained from the absolute values of the spatial distribution of the observed intensity of spectral lines by the inverse Abel transform [8]. In turn, the spatial distributions of the observed intensity (radiance) of spectral lines can be obtained experimentally, having previously calibrated the optical scheme in absolute values. The calibration algorithm and the method of Boltzmann plots are described in details [9]. Typical obtained radial distribution of the absolute intensity of Cu I 515.3 nm spectral line at arc current of 3.5 A is shown in Fig. 2.

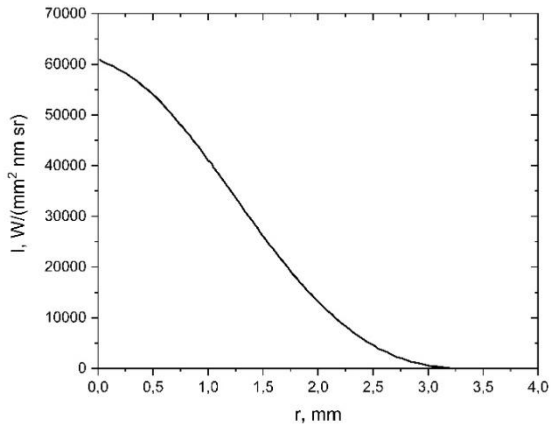

Fig. 2. Typical radial distribution of the absolute intensity of Cu I 515.3 nm spectral line at arc current of 3.5 A

Spectral lines Cu I 465.1, 510.5, 515.3, 521.8, 570.0, and 578.2 nm were selected for the following utilization in Boltzmann plot technique. The spectroscopic data (in particular the product of the statistical weight and the oscillator strength $g_i f_{ik}$) for the six Cu I lines of interest mentioned above are taken from the previous work [6]. The obtained radial distributions of temperature and copper number density with its confidence intervals, obtained by Boltzmann plot technique at arc currents of 3.5 and 30 A, are shown in Figs. 3 and 4, respectively.

For the second approach radial distribution of the electron density should be determined. The most common approach for determining the electron density is a direct calculation from the width of the spectral line contours, broadened due to the Stark effect [10].

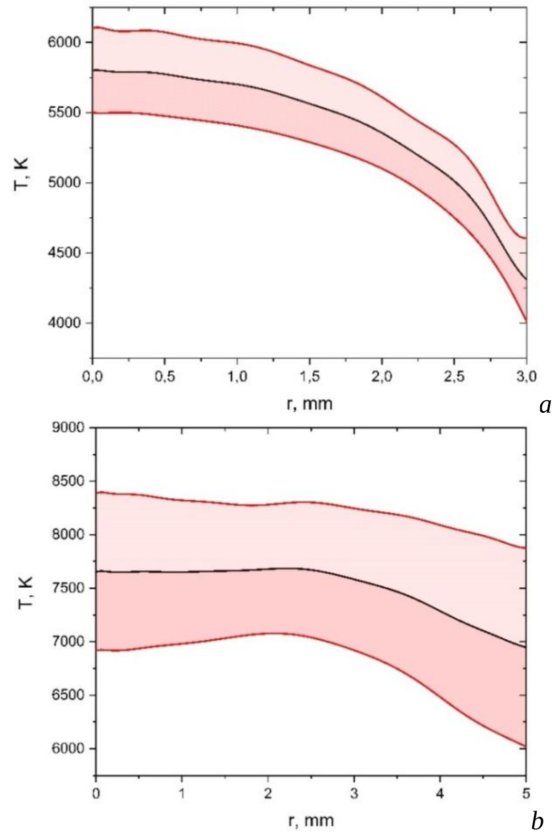

Fig. 3. Radial distributions of temperature obtained by Boltzmann plot technique at arc current of (a) 3.5 A and (b) 30 A

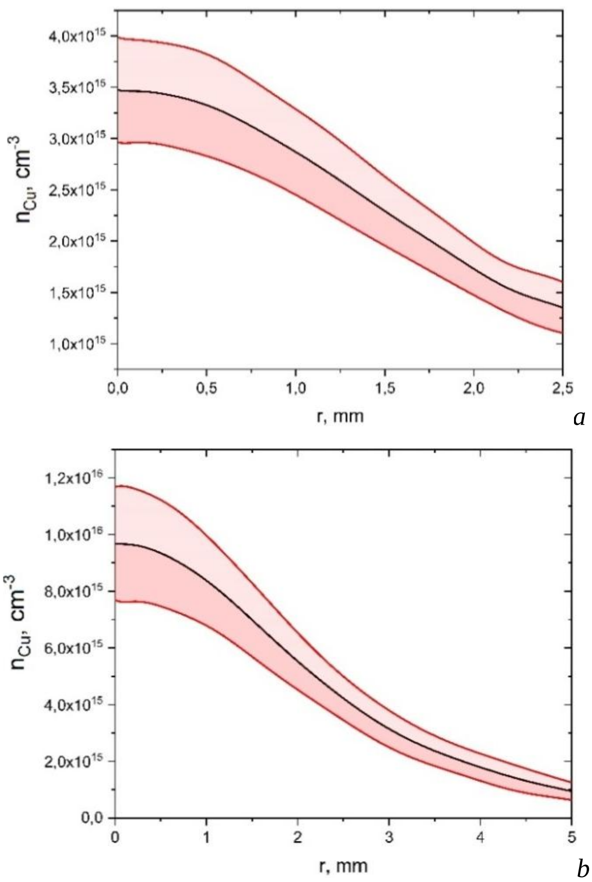

Fig. 4. Radial distributions of copper number density obtained by Boltzmann plot technique at arc current of (a) 3.5 A and (b) 30 A

This approach requires the use of a high-resolution spectral device for detailed observation of the shape of the contours of spectral lines, especially for the emission of low-current electric arc discharges as model plasma sources. A Fabry-Pérot interferometer (FPI) is used to achieve high resolution in measuring the broadening of spectral lines at different points in the arc discharge plasma. This device is configured in an optical scheme (Fig. 5) to sample plasma emission at specific angles [11]. Consequently, by measuring the width of the corresponding interference maximum from the obtained interferogram, the width of the spectral line at a specific spatial point of the plasma object can be determined. The specially developed program interface is also used to process the obtained interferograms (typical example is shown in Fig. 6).

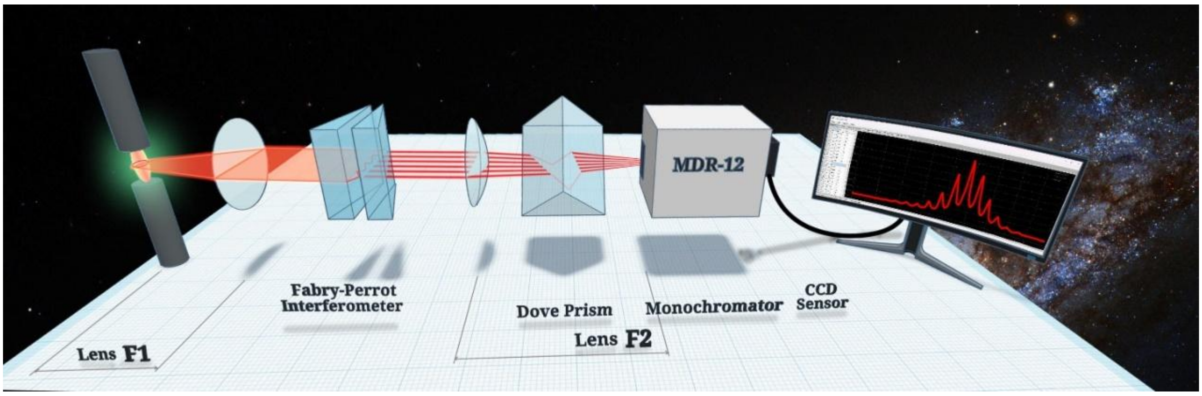

Fig. 5. Experimental setup for recording interferograms of spectral lines

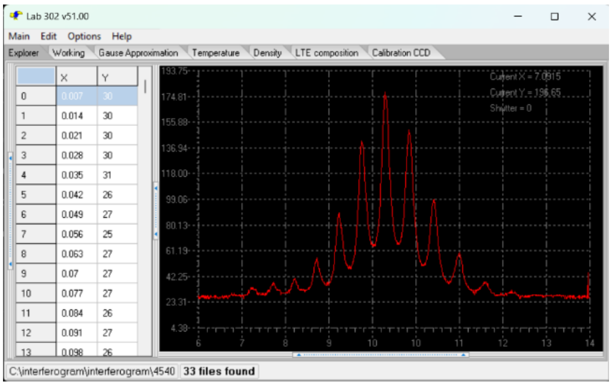

Fig. 6. Example of the obtained interferogram of the Cu I 515.3 nm spectral line obtained at current of 30 A

It should be noted that for the second approach (which involves electron density) the use of Boltzmann plot technique is not obligatory. In such a case, the method of relative intensities can be used. Since the optical scheme based on a monochromator makes it possible to register just one spatial distribution of spectral line radiance per single act of registration, reducing the number of used spectral lines to two can significantly accelerate the assessment of the erosion resistance of the materials under study. In this case, the method of relative intensities may be preferable when the time of investigation is important. Therefore, the temperature can be determined by the relative intensity method by eq. (1):

$$T = \frac{E_2 - E_1}{ln\left(\frac{\varepsilon_1}{\varepsilon_2}\right) - ln\left(\frac{A_1 g_1 \lambda_2}{A_2 g_2 \lambda_1}\right)}, \quad (1)$$

where E is the energy of the upper energy level of atom; ε is the emission intensity; A is the Einstein coefficient for spontaneous emission; g is the statistical weight of the upper energy level; λ is the wavelength at the centre of spectral line. Indices 1 and 2 correspond to the number of the spectral line used in the calculation. Thus, the radial distribution of temperature can be obtained by selection of two spectral lines. However, the choice of the line pair also affects the calculated temperature. In fact, the method of relative intensities is a special case of the Boltzmann plot method (using only two spectral lines). Fig. 7 shows the Boltzmann plot with linear approximation for different pairs of spectral lines, namely 510.5/515.3, 510.5/465.1 and 515.3/465.1. In particular, it is shown how the slope of the approximating line, which corresponds to the change in temperature [9], changes depending on the selected pairs of lines: $tg(\alpha_i) = \frac{1}{k_B T_i}$.

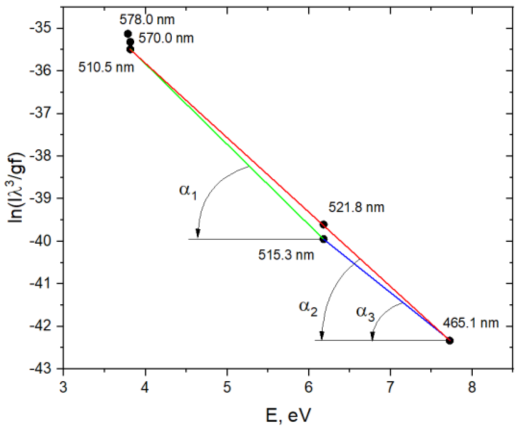

Fig. 7. Boltzmann plot on the axis of the arc (r = 0 mm) with linear approximations along two lines

Fig. 8 shows the radial temperature distributions obtained by the Boltzmann plot technique and the method of relative intensities for the pairs of lines 510.5/515.3, 510.5/465.1, and 515.3/465.1 at arc current 3.5 A. Thus, one can conclude that the most preferable pair of spectral lines for determining the temperature by the method of relative intensities is the pair 510.5/515.3, since it is closest to the temperature determined by the Boltzmann plot technique (at least at the arc axis) by both value and distribution behaviour.

In addition, it is important to note that electron density determination using FPI is optimal for the 30 A arc mode, but is not feasible at 3.5 A arc discharge current.

This is due to the fact that when using FPI in the 3.5 A mode, large losses of emission intensity are observed. In this regard, another approach to calculating the electron density at a discharge current of 3.5 A is realised.

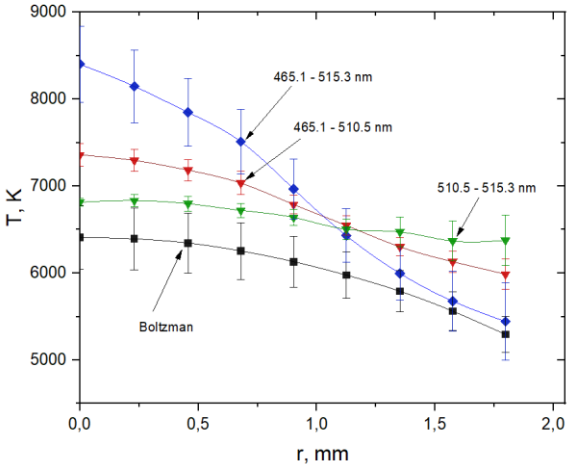

Fig. 8. Radial distributions of temperature obtained by the Boltzmann plot technique and the method of relative intensities at arc current of 3.5 A

Having experimentally determined the radial distribution of emission intensity of the Cu I 465.1 nm spectral line [9] at the arc current of 3.5 and 30 A, it is possible to find their ratio at different currents according to eq. (2):

$$\frac{\varepsilon_1}{\varepsilon_2} = \frac{A_1 g_1 \cdot n_1 \cdot \lambda_2 \cdot \Sigma_2}{A_2 g_2 \cdot n_2 \cdot \lambda_1 \cdot \Sigma_1} \cdot \frac{e^{-\frac{E_1}{kT_1}}}{e^{-\frac{E_2}{kT_2}}}. \qquad (2)$$

In eq. (2), the notations E, ε, A, g, T and λ are the same as in equation (1); n is the number density of radiating atoms (copper in this case); Σ is the partition function of atoms; k is the Boltzmann constant. Indices 1 and 2 correspond to the arc mode, namely arc current 3.5 and 30 A, respectively. Since the intensity ratio of the same spectral line is used, then $A_1 g_1 = A_2 g_2$, $E_1 = E_2$, and $\lambda_1 = \lambda_2$. Considering that electrons in the plasma appear mainly as a consequence of ionisation of metal atoms, then the Saha equation for ionisation of copper atoms can be written by eq. (3), as follows:

$$\frac{n_e^2}{n} = \frac{2\Sigma^+}{\Sigma} \cdot \left(\frac{2\pi m T}{h^2}\right)^{\frac{3}{2}} e^{-\frac{E_i}{kT}}, \qquad (3)$$

where n_e is the number density; Σ^+ is the partition function of ions; m is the mass of electron; h is the Planck constant; E_i is the ionisation energy of copper atom. Considering that the upper level energy of the Cu I 465.1 nm spectral line (7.737 eV) is approximately equal to the ionisation energy of copper (7.727 eV) and substituting eq. (3) into eq. (2), one can obtain eq. (4) for the ratio of electron density in an arc discharge at current of 3.5 and 30 A:

$$n_{e1} = n_{e2} \cdot \sqrt{\frac{\Sigma_1^+}{\Sigma_2^+} \cdot \frac{\varepsilon_1}{\varepsilon_2} \cdot \left(\frac{T_1}{T_2}\right)^{\frac{3}{2}}}. \qquad (4)$$

Radial distributions of electron density obtained using FPI at arc current of 30 A and using eq. (4) at arc current of 3.5 A shown in Fig. 9,*a*,*b*, respectively.

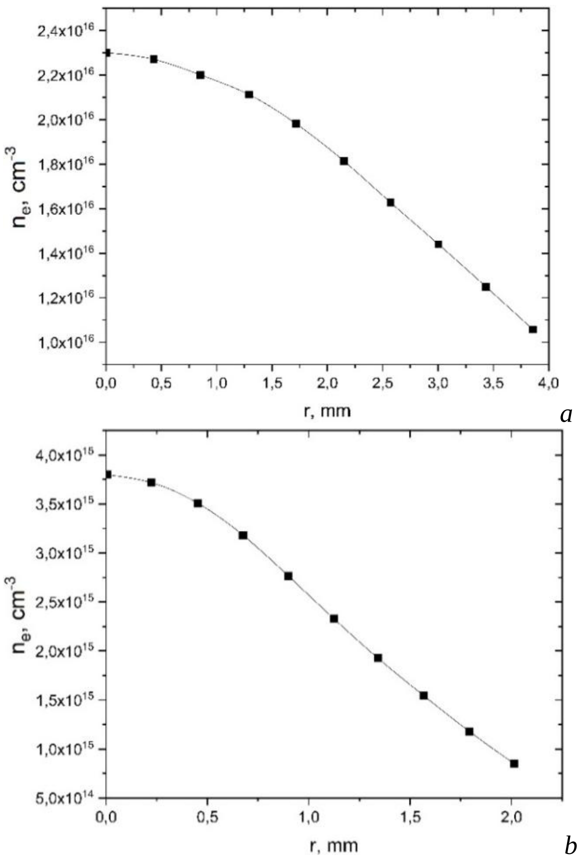

Fig. 9. Radial distribution of electron density in the arc discharge plasma at arc current of (a) 30 A and (b) 3.5 A

3. CALCULATION OF THE COMPOSITION OF THE ELECTRIC DISCHARGE PLASMA

Having determined the radial distributions of temperature and number density of copper atoms in the first case and the radial distribution of electron density in the second case, the composition of the plasma can be calculated using system of equations (5)-(9):

Saha equation for Cu, O, N atoms:

$$S_i(T, N_e) = \frac{N_e N_i^+}{N_i}; \; i \in Cu, O, N. \qquad (5)$$

Dissociation equations for O_2, N_2:

$$D(T) = \frac{N_i^2}{N_{i2}}; \; i \in O, N. \qquad (6)$$

The electroneutrality equation

$$N_e = \sum N_i^+; \; i \in Cu, O, N. \qquad (7)$$

The ideal gas equation:

$$B(T) = N_e + N_{O_2} + N_{N_2} + \sum N_i + \sum N_i^+; \; i \in Cu, O, N. \qquad (8)$$

The ratio of nitrogen to oxygen in the air:

$$\left[\frac{2N_{N_2} + N_N + N_{N^+}}{2N_{O_2} + N_O + N_{O^+}}\right] = 3.72. \qquad (9)$$

By solving this system of equations, it is possible to obtain the equilibrium plasma composition in two approaches. Radial distributions of equilibrium composition of plasma at arc current of 3.5 and 30 A obtained by eq. (5)-(9) with different input parameters are shown if Figs. 10 and 11, respectively.

One can see in Figs. 10 and 11, the radial distributions of n_e, n_{Cu}, and n_{Cu^+} coincide, which provides the possibility determining the erosion resistance of electrode materials in indirect way. This can be done by following two approaches: in the first approach, using temperature and electron density as input parameters, and in the second, using temperature and the number density of metal atoms.

Thus, the agreement between the two methods demonstrates the feasibility of estimating the erosion resistance of electrode materials through indirect calculations, based on the equilibrium plasma composition in both approaches.

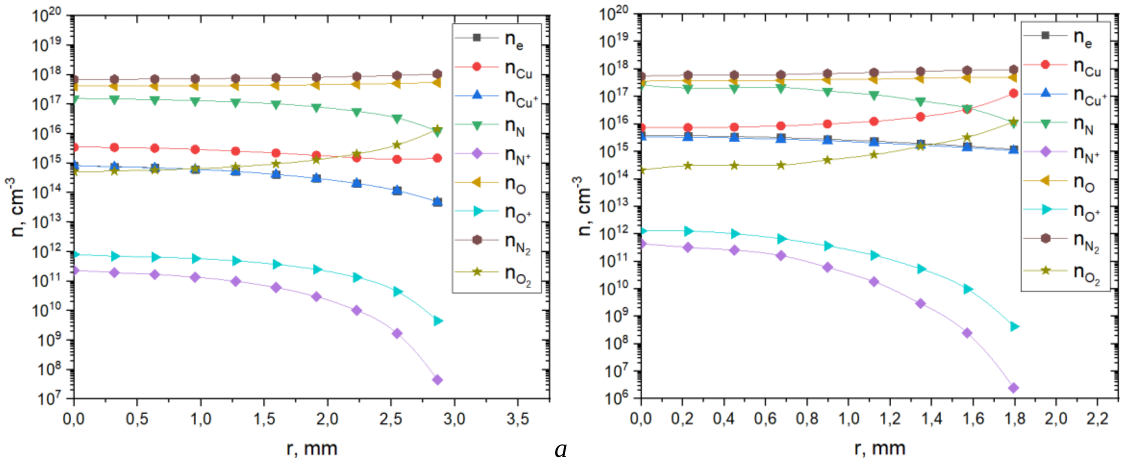

Fig. 10. Radial distribution of the equilibrium composition of arc discharge plasma at current of 3.5 A calculated (a) with temperature and copper number density and (b) with temperature and electron density as input parameters

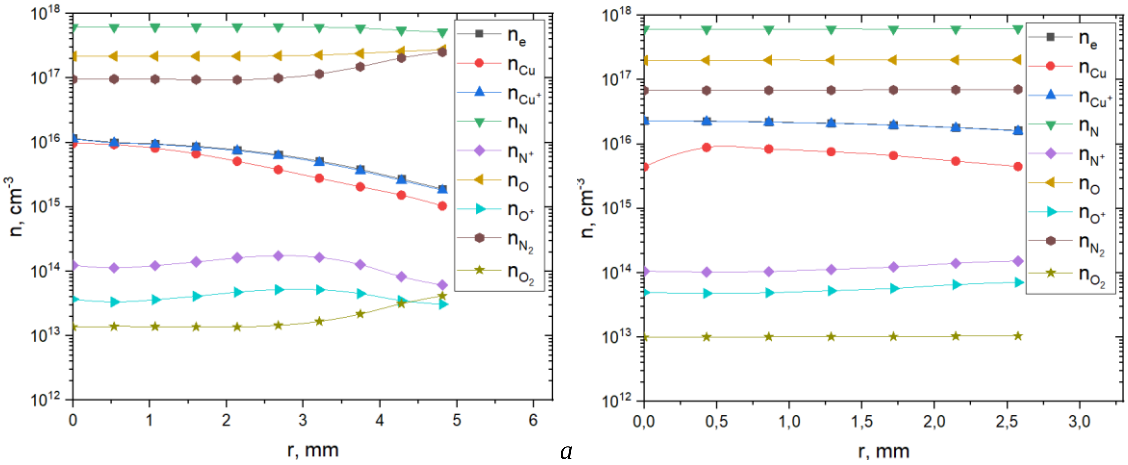

Fig. 11. Radial distribution of the equilibrium composition of arc discharge plasma at current of 30 A calculated (a) with temperature and copper number density and (b) with temperature and electron density as input parameters

CONCLUSIONS

Two complementary optical diagnostic approaches were applied to determine the equilibrium composition of copper-containing arc plasma: one based on excitation temperature and copper atom number density, and another on excitation temperature and electron density.

The excitation temperatures were successfully determined using both the Boltzmann plot technique and the relative intensity method. The pair of spectral lines 510.5/515.3 nm was found to be the most suitable for relative intensity diagnostics due to its close agreement with the full Boltzmann plot.

Electron density was obtained from Stark broadening of the Cu I 515.3 nm spectral line using Fabry-Pérot interferometer at arc current of 30 A, and indirectly for 3.5 A using intensity ratios and the Saha equation.

Comparison of the equilibrium plasma compositions obtained by both approaches revealed a good agreement in the calculated electron density, number densities of copper atoms and ions.

This consistency confirms that both diagnostic strategies are valid for analysing arc plasma properties and can be effectively used for estimating the erosion behaviour of electrode materials under different discharge modes.

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REFERENCES

1. H. Sattar, H. Ran, W. Ding, M. Imran, M. Amir, H. Ding. An approach of stand-off measuring hardness of tungsten heavy alloys using LIBS // *Applied Physics B*. 2014, v. 126, p. 1-11. https://doi.org/10.1007/s00340-019-7355-0.
2. A.B. Murphy. The effects of metal vapour in arc welding // *Journal of Physics D: Applied Physics*. 2010, v. 43(43), 43401. https://doi.org/10.1088/0022-3727/43/43/434001.
3. A.M. Rahal, B. Rahhaoui, S. Vacquie. Copper vapour diffusion in a nitrogen arc chamber // *Journal of Physics D: Applied Physics*. 1984, v. 17(9), p. 1807-1822. doi: 10.1088/0022-3727/17/9/006.
4. V. Boretskij, A. Veklich, Y. Cressault, A. Gleizes, P. Teulet. Non-equilibrium plasma properties of electric arc discharge in air between copper electrodes // *Problems of Atomic Science and Technology*. 2012, № 6, p. 181-183.
5. I.L. Babich, V.F. Boretskij, A.N. Veklich. Shapes of Spectral Lines of Nonuniform Plasma of Electric Arc Discharge Between Copper Electrodes // *AIP Conf. Proc*. 2007, v. 938, p. 252-257. https://doi.org/10.1063/1.2800139.
6. L. Babich, V.F. Boretskij, A.N. Veklich, et al. Spectroscopic data and Stark Broadening of Cu I and Ag I spectral lines: selection and analysis // *Advances in Space Research*. 2014, v. 54, p. 1254-1263. https://doi.org/10.1016/j.asr.2013.10.034.
7. A.N. Veklich, A.V. Lebid, T.A. Tmenova. Spectroscopic Data of W I, Mo I and Cr I Spectral Lines: Selection and Analysis // *Journal of Astrophysics and Astronomy*. 2015, № 4, p. 589-604. https://doi.org/10.1007/s12036-015-9342-0.
8. K. Bockasten. Transformation of Observed Radiances into Radial Distribution of the Emission of a Plasma // *Journal of the Optical Society of America*. 1961, v. 51(9), p. 943-947.
9. A. Murmantsev. Investigation of spatial distribution of metal vapours admixtures in the plasma of an electric arc discharge // *Problems of Atomic Science and Technology*. 2023, № 4, p. 139-146.
10. R. Konjevic, N. Konjevic. Stark broadening and shift of neutral copper spectral lines // *Fizika*. 1986, v. 18(4), p. 327-335.
11. A.M. Veklich, V.Ye. Osidach. The determination of electron density in electric arc discharge plasma // *Bulletin of the University of Kyiv. Series “Physics and Mathematics”*. 2004, № 2, p. 428-435.

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ДВА ПІДХОДИ ДО ВИЗНАЧЕННЯ ВМІСТУ ПАРІВ МІДІ В ТЕРМІЧНІЙ ПЛАЗМІ ЗА ДОПОМОГОЮ ОПТИЧНОЇ ДІАГНОСТИКИ

А. Веклич, Є. Сорокун, І. Слісаренко, А. Мурманцев, В. Осідач

Представлено порівняльне дослідження двох діагностичних підходів до визначення домішок парів міді в термічній плазмі електричного дугового розряду. Випромінювання плазми дугового розряду між мідними електродами аналізували за допомогою оптичної емісійної спектроскопії (ОЕС) при силі струму розряду 3,5 та 30 А. Як вхідні параметри для розрахунків рівноважного складу використовували два набори параметрів плазми: (1) радіальні розподіли температури збудження та концентрації електронів і (2) радіальні розподіли температури збудження та концентрації атомів міді. Температуру збудження визначали за допомогою методу діаграм Больцмана, тоді як концентрацію електронів отримували з розширення спектральної лінії, визначеної з використанням інтерферометра Фабрі-Перо, та застосуванням рівняння Саха. Узгодженість розрахованих складів плазми з використанням обох наборів вхідних даних демонструє застосовність непрямих діагностичних методів для оцінки стійкості електродних матеріалів до ерозії.