ESTIMATION OF ATOMIC DENSITY DISTRIBUTION IN DC ARC PLASMA USING SELF-ABSORBED EMISSION LINES

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Self-absorbed spectral lines emitted by plasma were used for the determination of copper content in a free burning DC arc discharge. At the first step, the plasma temperature radial distribution was obtained by Boltzmann plot method using non-absorbed spectral lines. The distributions of spectral line contours in different spatial points of plasma emission were registered. At the next step, the numerical model was developed to describe the contours of Cu I spectral lines in the case of optically thick plasma. The copper content was obtained by fitting of the experimentally registered line profiles and numerically modelled profiles, taking into account the plasma self-absorption.

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INTRODUCTION

Optical emission spectroscopy (OES) techniques are appreciably complicated in the case of plasma inhomogeneity and self-absorption of spectral lines in its volume. These properties can significantly change intensity distribution and profiles of spectral lines in comparison with "optically thin" plasmas. It results in a considerable accuracy decrease of the conventional optical diagnostic techniques when plasma absorption is neglected.

The problem of emission absorption in plasma and its influence on spectral line profiles is widely known. Many theories were proposed earlier [1-3] to describe such absorption influence. These theories were used extensively and experimentally verified [4-6]. However, model assumptions strictly limit a scope of theirs usage. For example, assumption of a constant halfwidth and shift of emissivity profile could not be applied since plasma is usually inhomogeneous and the line profile parameters are strongly depended on plasma properties. Bartels factors *M* and *Y*, introduced in [1], which connect maximal spectral intensity on self-reversed profile $I(\lambda)_{\max}$ and black body emission intensity $B(T_{max}, \lambda)$ corresponding to maximal temperature of plasma source, do not depend on the line broadening parameters. Moreover, Bartels theory is not applicable to the resonance lines. This is a serious restriction because the resonance lines are significantly more affected by absorption than non-resonance ones. Later in [7] authors proposed the calculation of a factor *M* for the resonance lines of sodium and aluminum. This factor depends strongly on a plasma temperature and ionization degree. Nevertheless, a model does not take into account a line asymmetry.

An absorption factor *p* of the Cowan–Dieke model [2], which depends on the number of absorbing atoms along the line of sight, does not reflect a line self-absorption rate. It is also defined only for unshifted line center. Fishman [8] proposed further model based on Cowan–Dieke results [2]. It takes into account profile halfwidth and shift dependence on plasma inhomogeneity. However, the vast number of model parameters complicates its usage. Similar model with many parameters was proposed in [9, 10]. The factors of dip depth and peak separation of a self-reversed line profile were obtained which describe plasma inhomogeneity and absorption. The results were obtained only for neutral mercury lines and it is difficult to use the model for other element spectral lines.

Nowadays numerical modeling allows to avoid the inverse problem solving and to get a direct dependence of a spectral line profile on plasma parameters. Moreover, local thermodynamic equilibrium (LTE) assumption, which is valid for the most of plasma sources under atmospheric pressure, makes possible to reduce a model parameters number.

This paper presents a numerical modelling of self-reversed copper atom line profiles with the aim to obtain a copper density distribution in plasma. Experimentally obtained copper atom line profiles and the plasma temperature in electric arc discharge between copper electrodes were used as initial data in this modelling.

1. EXPERIMENT

A scheme of experimental setup is shown in Fig. 1. A free burning arc was ignited between two non-cooled copper electrodes (E1, E2) in the ambient air atmosphere. To register a distribution of spectral line contours in different spatial points of plasma emission simultaneously, an optical system was used [11]. It includes lenses O1, O2, the Fabry-Perot interferometer (FPI) and the MDR-12 monochromator, equipped by CCD linear image sensor. Dove prism rotates the arc image on MDR-12 entrance slit. The apparatus halfwidth is determined by the free spectral interval of the FPI and was estimated as $\lambda^2/14d$, where $d = 0.3\,mm$ is a distance between interferometer mirrors. Thus, the apparatus halfwidth varied from 0.038 to 0.086 nm in the investigated spectral range of 400…600 nm. The arc DC current was of 3.5 A, on which a current pulses up to 100 A of 30 ms duration were applied. A 7 ms delay between pulse front and emission registration was set to provide study in a quasi-stationary mode of arc discharge. The exposure time was less than 1 ms.

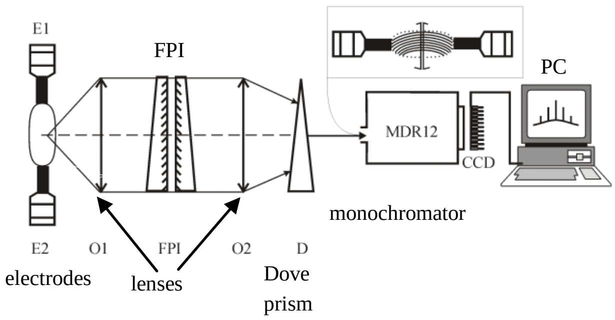

Fig. 1. Experimental setup

The emission of plasma was studied in the middle cross section of the discharge gap (see Fig. 1) at arc current 100 A. As an example, the experimental interferogram is shown in Fig. 2. One can see that each registered line contour is composed of several overlapping peaks, corresponding to different distances of the line of sight from discharge axis. Using multi-peak fitting procedures, one could obtain a distribution of relative line intensity as well as a distribution of observed line halfwidth along the plasma cross section.

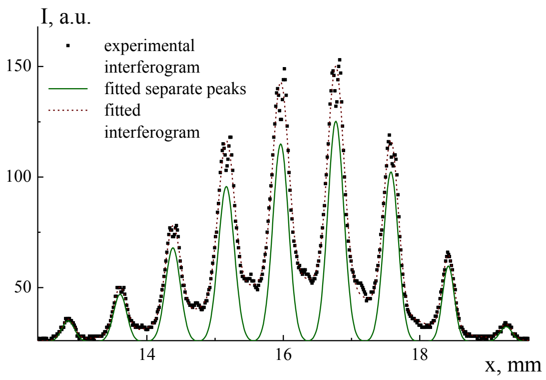

Fig. 2. Experimental interferogram of Cu I 510.5 nm – line with symbols and fitted – solid lines

To obtain local halfwidths of spectral line, a deconvolution procedure was used. Though the apparatus function of Fabri-Perot interferometer is described by Airy function conventionally, for simplification we assumed that apparatus function is Gauss.

2. MODELING

The numerical model was developed to describe the contour of spectral lines in the case of optically thick plasma [12]. The plasma pressure of 1 bar was considered, while an air/copper ratio distribution was varied during fitting of calculated and experimentally measured line halfwidth [13, 14]. The numerical model includes solutions of the Saha-Boltzmann equation set and the radiation transfer equation. Due to a high temperature in plasma, the plasma composition was simplified as a mixture of N, O and Cu atoms, theirs ions and electrons. More detailing calculation of the plasma composition taking into account N_2, O_2 molecules and other gases like Ar, CO_2 was carried out. Fig. 3 shows that the discrepancy between simplified mixture and detailed one is the most prominent for the nitrogen atom concentration at the temperatures less than 7500 K. Nevertheless, the difference for copper atom and ion concentrations as well as for electron density is insignificant.

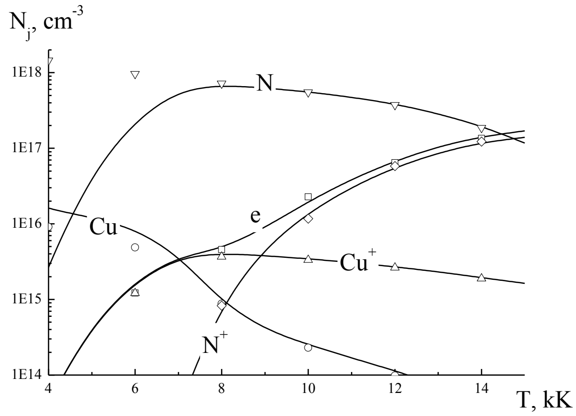

Fig. 3. Detailed (lines) and simplified (dots) plasma air-Cu (1 vol.%) compositions

Local profiles of spectral lines were calculated in assumption of the quadratic Stark effect. Stark parameters: width $w(T)$ and shift $d(T)$ were taken from [15]. It is supposed the local profile is a Lorentz one, which halfwidth γ_e and shift δ_e are proportional to the electron density N_e (equation (1)). Parameters of the simulated copper density distribution were adjusted to make the calculated halfwidth of the line profiles be fitted to the experimentally measured ones [13, 14]

$$\gamma_e = w(T) \cdot N_e \cdot 10^{-16},\ \delta_e = d(T) \cdot N_e \cdot 10^{-16}. \qquad (1)$$

Integrated over the line of sight intensity $I(\lambda)$ was calculated by solving the radiation transfer equation (2) for a set of wavelength values within line profile $\phi(\lambda)$. The wavelength step is chosen to describe this profile with sufficient accuracy for line parameters determination (halfwidth, peak separation, profile dip).

$$I(\lambda) = \int_0^\infty \varepsilon(\lambda, y) ch\left(\int_0^y \kappa(\lambda, y')dy'\right) dy \cdot \exp(\tau_\lambda), \qquad (2)$$

where emission coefficient ε_λ is:

$$\varepsilon_\lambda = \frac{1}{4\pi} A_{mn} \frac{hc}{\lambda_0} N_m \phi(\lambda), \qquad (3)$$

and line profile $\phi(\lambda)$ is described as follows:

$$\phi(\lambda) = (\gamma_e / \pi)[\gamma_e^2 + (\lambda - \lambda_0 - \delta_e)^2]^{-1}, \qquad (4)$$

absorption coefficient κ_λ is defined as:

$$\kappa_\lambda = \frac{\pi e^2 \lambda_0^2}{mc^2} f_{nm} N_n \left[1 - \exp\left(-\frac{hc}{\lambda_0 kT}\right)\right] \phi(\lambda), \qquad (5)$$

and optical depth $\tau(\lambda)$ is:

$$\tau(\lambda) = \int_0^\infty \kappa(\lambda, y) dy. \qquad (6)$$

It must be noted, that most informative parameter is a line halfwidth. Peak separation and profile asymmetry is also valuable information which allow to estimate plasma inhomogeneity. Unfortunately, the latter two parameters could not be measured with acceptable accuracy because the convolution of FPI apparatus function with intensity line profile significantly blurs the profile parameters. Line halfwidth of self-reversed lines depends mostly on plasma optical depth $\tau(\lambda)$ and these line profiles data allow to estimate the neutral copper density.

3. RESULTS AND DISCUSSIONS

Experimental investigations of the spectral lines contours showed that Cu I 427.5, 510.5, 515.3, 521.8 and 578.2 nm lines are self-absorbed for mentioned above discharge mode. The self-absorption rate R is defined as

$$R = 1 - I_1 / I_0 , \qquad (7)$$

where I_1 is an integral line intensity:

$$I_1 = \int_0^{\infty} I(\lambda) d\lambda . \qquad (8)$$

I_0 is integral line intensity in the hypothetical case of absorption absence:

$$I_0 = 2\int_0^{\infty} d\lambda \int_0^{y^*} \varepsilon(\lambda, y) dy . \qquad (9)$$

Table shows the R values for some spectral lines of copper atom. They depend on many factors like the Stark parameters, radiation transition probabilities and density distributions of absorbing particles. It is well known [12], that the ratio between Stark parameters significantly effects on self-absorption. Lines with lower *d/w* ratio have the maximal R values. For example, Cu I 510.5 nm has *d/w* ratio less than 0.1 and its R value is the largest one among considered spectral lines Table.

Self-absorption rate of Cu I lines(discharge axis in the middle cross-section)

λ, nm	E_{up}, eV	f_{ik}	g_{up}	g_{down}	I_0/I_1	R, %
427.5	7.74	0.126	8	6	2.02	50.4
448.0	6.55	0.009	2	2	1.00	0.4
510.5	3.81	0.0052	4	6	3.14	68.2

Such behavior of 427.5 and 510.5 nm spectral lines is caused by significant optical density of plasma, and obviously, these lines cannot be used in traditional optical spectroscopy techniques. The self-absorption of other spectral lines, for example, 448.0, 465.1 nm, can be neglected, therefore they were used in plasma temperature and electron density determination.

The FWHM radial distributions were calculated on the base of experimentally obtained plasma temperature [16] (Fig. 4) and atomic data [17]. To obtain density distribution, calculated FWHM distributions for lines with high R were fitted to maximal coincidence with experimental ones using the least-squares method. Due to the uncertainties of radiative transition and broadening constants, it is rather complicate task to fit the data for several lines simultaneously. An example of the best fit for Cu I 427.5, 448.0 and 510.5 nm lines is shown in Fig. 5.

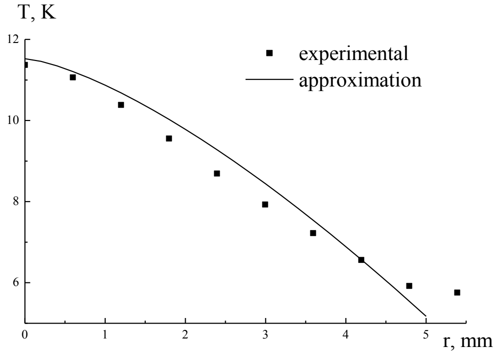

Fig. 4. Radial plasma temperature distribution of the discharge at arc current 100 A

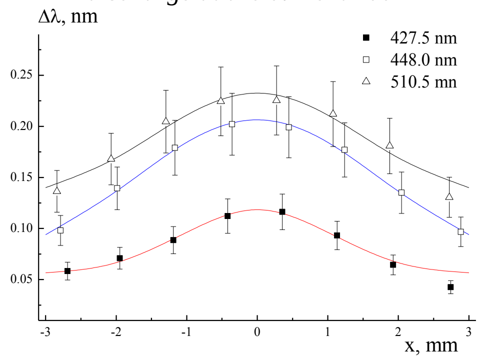

Fig. 5. Line FWHW distribution for Cu I: symbols – experimental data; lines – calculation

The results of fitted FWHM allowed to obtain the radial distribution of copper atom density in the discharge. Using plasma temperature (see Fig. 4) the content of copper X_{Cu} in discharge plasma was calculated as the sum of neutrals and single ionization ions densities. The ions of higher degree of ionization can be neglected because the maximum temperature was near 1 eV.

The radial distribution of copper content in plasma is shown in Fig. 6. The copper density rises to discharge edge from approximately 0.03% in the arc center to 1% at periphery. The radial distribution is limited by 3 mm narrow emission zone. However, proposed method has an appreciable advantage: it does not requires an external light source to scan plasma. It could be critical for remote plasma sources investigations.

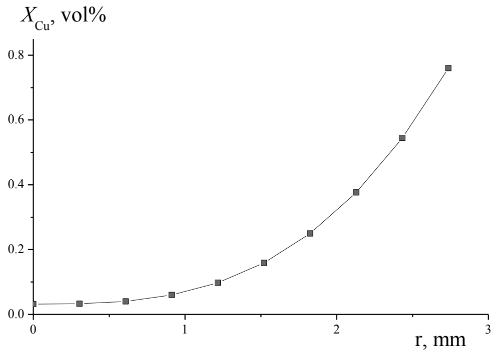

Fig. 6. Radial distribution of copper content

CONCLUSIONS

The technique of atomic density distribution estimation in dc arc plasma using self-absorbed emission lines is developed. Self-absorbed Cu I lines were used for the determination of copper content in a free burning electric arc at current 100 A. Copper content in the discharge was numerically obtained by modeling of Cu I spectral lines contours in the case of optically thick plasma. Experimentally measured plasma temperature and distributions of spectral line contours in different spatial points of the discharge were used as input data in simulation.

It was shown that the copper content at discharge axis was less than 0.1% and increased to the arc periphery up to 1%.

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ОЦЕНКА РАСПРЕДЕЛЕНИЯ ПЛОТНОСТИ АТОМОВ В ПЛАЗМЕ ДУГИ ПОСТОЯННОГО ТОКА С ИСПОЛЬЗОВАНИЕМ САМОПОГЛОЩЕННЫХ СПЕКТРАЛЬНЫХ ЛИНИЙ

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Содержание меди в свободно горящем дуговом разряде постоянного тока определяли с помощью самопоглощенных спектральных линий излучения плазмы. На первом этапе определяли радиальное распределение температуры плазмы с использованием не самопоглощённых спектральных линий методом диаграмм Больцмана. Регистрировали распределения контуров спектральных линий в различных пространственных точках излучения плазмы. На следующем этапе разработали числовую модель для описания контуров спектральных линий Cu I для оптически толстой плазмы. Содержание меди получали путем подгонки экспериментально зарегистрированных и численно моделируемых профилей линий с учетом самопоглощения в плазме.

ОЦІНКА РОЗПОДІЛУ ГУСТИНИ АТОМІВ У ПЛАЗМІ ДУГИ ПОСТІЙНОГО СТРУМУ З ВИКОРИСТАННЯМ САМОПОГЛИНЕНИХ СПЕКТРАЛЬНИХ ЛІНІЙ

В.Ф. Борецький, К.Ю. Кацалап, Є.О. Єршов-Павлов, Л.К. Станчітц, А.М. Веклич

Вміст міді у вільно існуючому дуговому розряді постійного струму визначали за допомогою самопоглинених спектральних ліній випромінювання плазми. На першому етапі визначали радіальний розподіл температури плазми з використанням не самопоглинених спектральних ліній методом діаграм Больцмана. Реєстрували розподіли контурів спектральних ліній в різних просторових точках випромінювання плазми. На наступному етапі розробили числову модель для опису контурів спектральних ліній Cu I для оптично товстої плазми. Вміст міді отримували шляхом підгонки експериментально зареєстрованих і чисельно модельованих профілів ліній із урахуванням самопоглинання в плазмі.