
DETERMINING OF THE ELECTRONIC EXCITATION TEMPERATURE IN ATMOSPHERIC PRESSURE PLASMAS BY USING EMISSION SPECTRA OF METALS WITH COMPLEX LOW-RESOLVED MULTIPLY STRUCTURE¹**V.YA. CHERNYAK,¹ I.V. PRYSIAZHNEVYCH,¹ OK.V. SOLOMENKO,¹ S.V. OLZEWSKI,¹ JA. DIATCZYK,² H. STRYCZEWSKA,² N.V. BELENOK,³ I. SHYHT¹**¹Taras Shevchenko National University of Kyiv, Radio Physics Faculty
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Plasma parameters of a gliding arc in the argon flow are investigated by optical emission spectroscopy. It is shown that there are a lot of overlapped low-resolved multiplet lines, which correspond to the emission of electrode material, in registered spectra of the investigated plasma. To process the obtained spectra, the simulation of main stainless steel components (Fe, Cr, Ni) and their alloy (stainless steel) is made with regard for the instrumental function of a spectrometer in a wide interval of electronic excitation temperatures $T_e^* = 2000-20000$ K. Spectral intervals for the adequate comparison of experimental data with results of simulation are suggested. It is found that the ratio of concentrations of Fe and Cr atoms in the investigated plasma correlates with the content of these elements in the used electrode material (Fe~70%, Cr~20%).

1. Introduction

The basic property of the plasma of arc discharges (gliding arc discharges (GA) [1], and a transverse arc [2], for example) is the contribution of a material of electrodes to its emission spectrum, since the electrodes contact the volume, where plasma is created. This can lead to prob-

lems in the identification of such emission spectra, especially when transition metals (Fe, Ni, Cr, etc.) and their alloys (stainless steel, etc.) are chosen as an electrode material. Spectra of such metals have complex multiplet structures which can be resolved by spectrometers with high resolution (tens of angstroms). Such devices are very expensive and inconvenient for the diagnostics of technological objects under manufacturing conditions. In addition, a lot of molecular bands are observed in a spectrum in the case where a molecular plasma-forming gas (air, for example) is used, which complicates the decoding of emission spectra.

Therefore, one of the available ways to determine plasma parameters (excitation temperatures) in the case where low-resolution spectrometers (with the instrumental function width at FWHM ≥ 0.5 nm) is to compare the experimental spectra of metals with the simulated ones, by taking the instrumental function of a used spectrometer into account. A similar approach with the simulation of molecular bands was used for the diagnostics of atmospheric pressure discharges [3]. In the present work, we simulate the emission of main stainless steel components (Fe, Cr, Ni) and choose the spectral intervals for the adequate comparison of experimental data

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Fig. 1. Gliding arc

with calculated ones. The results of spectroscopic researches of a gliding arc discharge with stainless steel electrodes in the air and argon flows are represented.

2. Experimental Setup

Experiments were done in a three-electrode GA plasma reactor of the vertical type (Figure 1) with knife-shaped electrodes made from the acid-resistant steel

0H18N9 (DIN X5CrNi810). The GA discharge worked in the quasicontinuous mode powered by a three-phase AC/DC/AC supply [4]. The output voltage reached several hundred volts, the discharge current was ten amperes, and the discharge power did not exceed 1–1.5 kW. As a working gas, argon at the atmospheric pressure was used. The gas flow rate G varied from 0 to 4 m³/h (flow velocity ~ 10 m/s).

The diagnostics of a GA plasma was made by means of optical emission spectroscopy (OES) with using a portable high-speed CCD-based OMA Solar SL40-3648 in the spectral range 210–1100 nm. The OES measurements were conducted at several cross-sections along the GA, by using a focusing optical fiber. The OES analysis has shown that the GA emission contains the emissions of components of the plasma-forming gases: ArI (912.3, 811.5, 801.5, 794.8, 763.5 nm), OI (777.2, 844.6, 926.6 nm), elements evaporated from the electrodes: FeI (516.7, 438.3, 404.6, 386.0, 374.6, 373.5, 372.0, 358.1, 344.1 nm), CrI (520.6, 429.0, 427.5, 425.4, 359.3, 357.9 nm), NiI (361.9, 352.4, 351.5, 349.3, 341.5 nm) and others.

3. Methodology

It is known that the intensity of an emission spectral line can be defined by the expression [5]

$$I = C \frac{g_1}{g_2} \frac{f_{12}}{\lambda^3} N_2 \ell, \quad (1)$$

where C – numerical constant, g_1 – statistical weight of the lower level; g_2 – statistical weight of the upper level; N_2 – density of atoms in the upper excited electronic state; f_{12} – oscillator strength; λ – wavelength; and ℓ – thickness of the homogeneous transparent plasma layer.

In the case of the Boltzmann population distribution of excited electronic states of these metals, we have

$$N_2 = \frac{g_2}{g_0} N_0 \exp(-E_2/T_e^*), \quad (2)$$

where g_0 and N_0 – statistical weight and the density of atoms in the ground state; E_2 – energy of the upper level; and T_e^* – temperature of the population of excited electronic levels (electronic excitation temperature).

In the case of the plasma heterogeneity and the constancy of T_e^* along the view axis, the intensity of an observable emission line of the i -atomic component (Fe, Cr) can be described by the expression

$$I^i = C \frac{g_1}{g_0} \frac{f_{12}}{\lambda^3} e^{-\frac{E_2}{T_e^*}} \int_0^L N_0^i d\ell. \quad (3)$$

The calculated spectra were represented as a sum of all emission lines of a certain component (FeI, CrI, NiI, etc.) with wavelengths referenced in NIST database [6]. The emission spectra of main components (Fe, Cr, Ni) of stainless steel electrodes were simulated in the assumption of a local thermodynamic equilibrium (LTE) and the Boltzmann population distribution of excited electronic states.

Since the overlapping of different multiplet components occurs, we need to compare not only certain lines, but spectral regions in the calculated and experimental spectra.

By analogy with the Ornstein method (method of measuring the excitation temperature by relative intensities of two lines) [7], we will choose pairs of lines which belong to one certain component. In determining T_e^* with appropriate accuracy, the pair of lines with the highest difference between the energies of upper excited electronic states ($\Delta E_2 \geq T_e^*$) should be used. This means that there is a considerable difference in the behavior of intensities of these lines depending on the electronic excitation temperature. In addition, it is better

Spectroscopy constants from Database NIST [6]

Fe I (384-386 nm)				Fe I (435-440 nm)			
λ , Å	E_1 , eV	E_2 , eV	$g_1 f_1 2 \exp(-E_2/T_e^*)$ $T_e^* = 1 \text{ eV}$	λ , Å	E_1 , eV	E_2 , eV	$g_1 f_1 2 \exp(-E_2/T_e^*)$ $T_e^* = 1 \text{ eV}$
3840.437	0.99011	4.217584	0.00459707	4351.544	2.990352	5.838753	8.1258E-05
3841.048	1.607895	4.834856	0.006755658	4352.734	2.222712	5.070333	0.000339138
3843.256	3.046869	6.271975	0.001376713	4358.501	2.948793	5.792647	7.04527E-05
3846.8	3.251482	6.473617	0.001620814	4367.578	2.990352	5.828296	0.000156278
3849.969	1.011055	4.230538	0.001959153	4369.771	3.046869	5.883389	0.000526427
3856.372	0.051569	3.265706	0.001974533	4383.544	1.484864	4.312471	0.021226228
3859.212	2.404074	5.615847	0.000757063	4387.893	3.071339	5.896144	9.07509E-05
3859.911	0	3.21119	0.007872273	4388.406	3.602526	6.427001	0.0004302
3865.523	1.011055	4.217584	0.00153383	4390.951	3.017623	5.84046	9.76922E-05
3867.215	3.01725	6.222376	0.000754119	4401.29	3.602526	6.418733	0.000194056
				4401.442	2.831591	5.6477	7.93264E-05
				4404.75	1.557357	4.371352	0.00910922
				4407.708	2.175945	4.988051	8.11454E-05
				4408.414	2.197866	5.009522	0.000126808

not to use an intensive resonant line, since it can be re-absorbed.

On the basis of the requirements represented above, the following low-resolved groups of lines were selected for the simulation: (I) Fe I $\lambda = 438.4 \text{ nm}$ and $\lambda = 386.1 \text{ nm}$; (II) Cr I $\lambda = 475.6 \text{ nm}$ and $\lambda = 520.7 \text{ nm}$. There are a lot of spectral lines (multiplets) within chosen intervals of wavelengths: nearly 21 lines in Fe I (384–386 nm) and nearly 34 lines in Fe I (435–440 nm). The most intensive lines are represented in Table.

To determine T_e^* by comparing experimental spectra with the simulated one, it is necessary to choose spectral groups of lines (look like wide bands) which mainly belong to the one component and don't overlap with others. For that purpose, the simulation of emission spectra of main stainless steel components (Fe, Cr, Ni) was made at atmospheric pressure and a fixed temperature with regard for spectral constants [6] and the instrumental function of a real spectrometer, whose shape is close to Gaussian. Equal initial concentrations of these components were accepted during the calculations. Results of the simulation are represented in Fig. 2.

From Fig. 2, it can be seen that, in the selected spectral interval 384–388 nm, the Fe I emission makes the main contribution into the total emission spectrum within this narrow spectral region. So, it is possible to neglect the Ni contribution, since its relative concentration in the electrode material is much smaller than that of Fe. For the same reason, we can ignore its contribution in the interval of 474–478 nm as well (see Fig. 2,c) and can assert that the Cr emission is dominant in the chosen spectral interval. As is seen from Fig. 2,b, the

emission of two components (Fe and Cr) is observed in the spectral interval 436–440 nm. While the character of the Cr emission is close to monotonic, we can neglect it like the almost constant background during the measurement of the peak height of the chosen spectral group of lines (at $\lambda = 438.4 \text{ nm}$).

These groups of lines were chosen in such way that one of them is very sensitive to changes in the temperature and another one is not. The result of calculations of the Fe I emission in the chosen spectral intervals for various electronic excitation temperatures is represented in Fig. 3.

From Fig. 3, we can see that the contour of the group of lines (436–440 nm) and the position of its maximum change with a variation of the electronic excitation temperature. This can be related to a redistribution of the intensities of multiplet components with a change in the temperature (see Table). At low temperatures ($T_e^* \leq 5000 \text{ K}$), the Fe line at $\lambda = 437.6 \text{ nm}$ ($E_1 = 0 \text{ eV}$, $E_2 = 2.83 \text{ eV}$) makes the main contribution to the total emission spectrum within this spectral interval. Thus, Fig. 3 demonstrates that one of the group of lines (436–440 nm) depends on the temperature and another group of lines (384–388 nm) almost doesn't.

To decrease the inaccuracy related to the presence of the background, we suggest to use the relative maxima and minima in certain spectral intervals for each group of lines (see Fig. 4).

The curves of the ratios of maximum intensities (I_1, I_2) in the chosen spectral regions as functions of the electronic excitation temperature for Fe and Cr components are shown in Fig. 5.

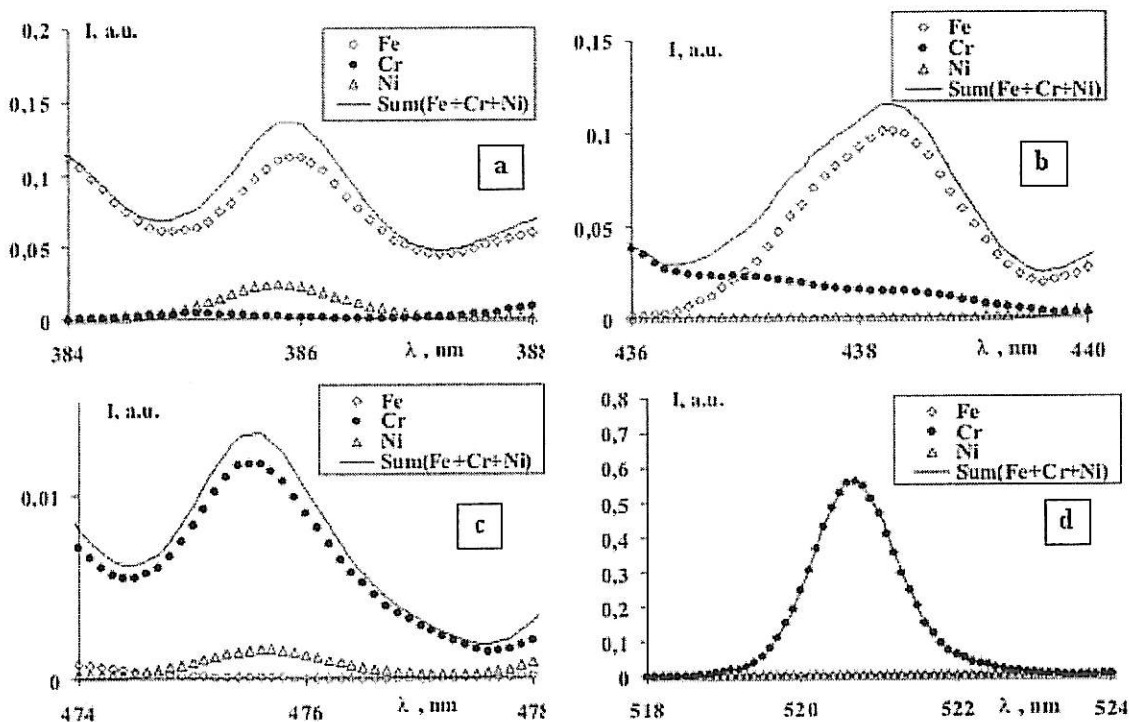


Fig. 2. Results of a simulation of the emission spectra of the main steel components and their sum in the chosen spectral intervals in the case of equal initial concentrations at a fixed temperature and a fixed pressure. All spectra were normalized on the intensity of the Fe line at 248.8 nm

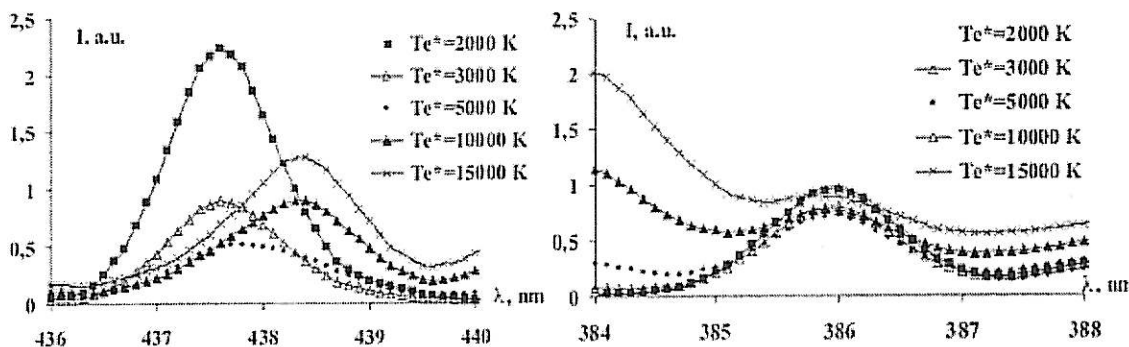


Fig. 3. Calculated Fe I emission in the spectral intervals 384–388 nm and 436–440 nm at various electronic excitation temperatures T_e^*

From Fig. 5,a, it is seen that the calibration curve for the measurement of T_e^* has a minimum nearly at $T_e^* = 5000$ K. Therefore, we have two branches: the “low-temperature” branch, $T_e^* < 5000$ K, and the “high-temperature” one, $T_e^* > 5000$ K. The nonmonotonic character of the curve (Fig. 5,a) results from the redistribution of the intensities of multiplet components with

a variation of the temperature. In this spectral region, there are the overlapping different multiplets with different energies of the upper states E_2 (see Table). By The simulation indicates that, to specify a branch we should use during the measurement of T_e^* , it is necessary to estimate the ratio $I_{386.1}/I_{382.3} = C$. It was found that, at temperatures $T_e^* < 5000$ K, $C > 1$ (the left “low-

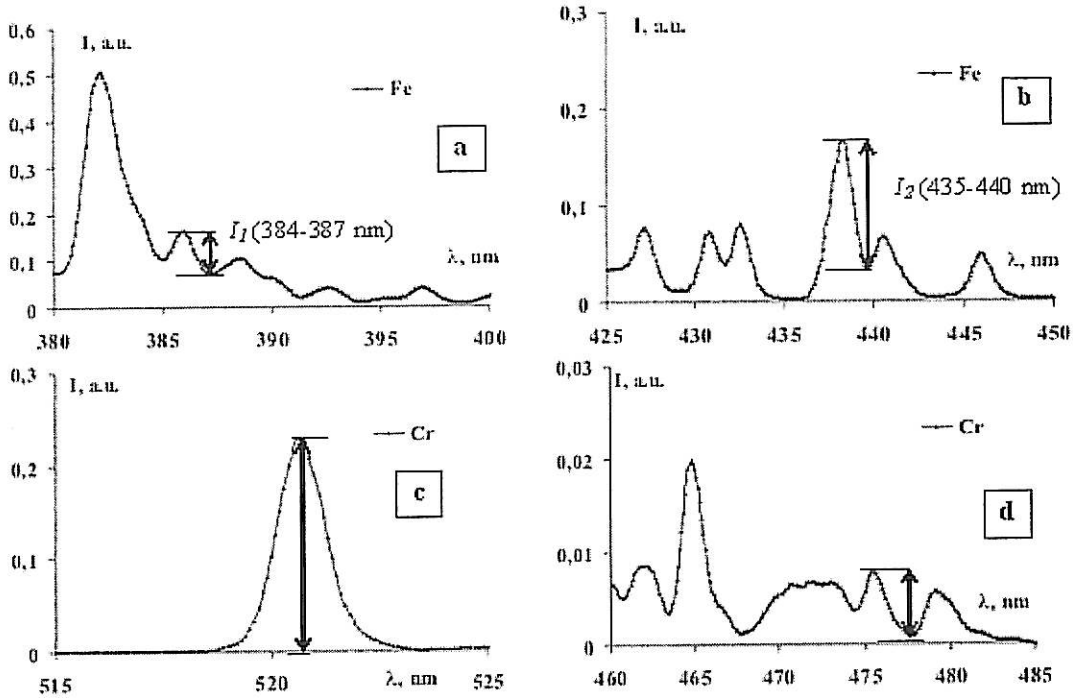


Fig. 4. Emission spectra calculated at $T_e^* = 8500$ K for: a, b - Fe I; c, d - Cr I

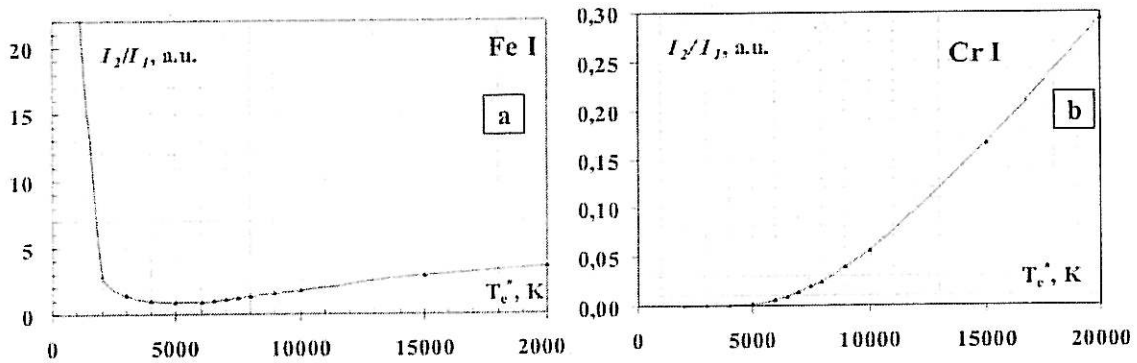


Fig. 5. Curve for determining the electronic excitation temperature: for Fe (a) and Cr (b)

temperature" branch should be used). At $T_e^* > 5000$ K, $C < 1$ (then the right branch of the curve in Fig. 5, a should be used).

4. Results

The emission spectra of GA plasma in the Ar flow $G = 4$ m³/h at the discharge current $I_d = 15$ A are measured. The electronic excitation temperatures $T_e^*(Fe) \approx T_e^*(Cr)$

$= 7500$ K, $T_e^*(Ar) = 7000$ K, and $T_e^*(O) = 6000$ K are determined.

To estimate the relative concentrations of radiating species (main stainless steel components are Fe and Cr) in the ground state under the assumption of their identical distribution along the view axis, we applied the method proposed in work [8]:

$$\frac{[N_{Cr}]}{[N_{Fe}]} \approx \frac{I_{exp Cr} I_{cal Fe}}{I_{exp Fe} I_{cal Cr}} \quad (4)$$

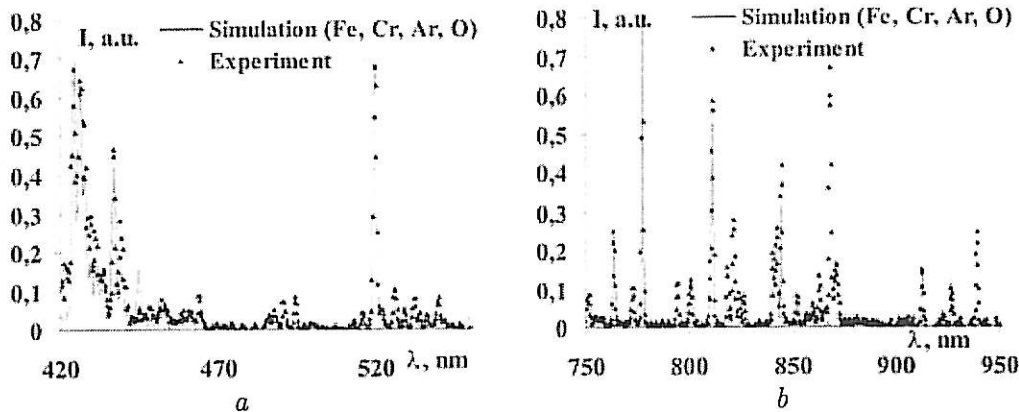


Fig. 6. Emission spectra of GA plasma in the argon flow – in the range: *a* – 420–550 nm, *b* – 750–950 nm. The best agreement between experimental and simulated spectra was obtained at $T_e^*(\text{Fe, Cr, Ni}) = 7500$ K, $T_e^*(\text{Ar}) = 7000$ K, and $T_e^*(\text{O}) = 6000$ K

Here, I_{exp} and I_{cal} – peak heights of the chosen spectral groups of lines in experimental and calculated spectra of the corresponding components (Fe or Cr).

The result of superimposing the experimental spectrum with the simulated one is shown in Fig. 6. A good agreement between the experimental and simulated spectra shows that the ratio of concentrations of Fe and Cr atoms in the plasma-forming gas correlates with the content of these elements in the used electrode material, steel 0H18N9 (Fe~70%, Cr~20%).

5. Conclusions

The technique of determining the electronic excitation temperature in the plasma of arc discharges with stainless steel electrodes is suggested. It is found that T_e^* of metal atoms (Fe, Cr) is slightly higher than T_e^* of blowing gas atoms (O). This correlates with plasma parameters of a transverse arc in air and Ar gas flows [2]. It is shown that the ratio of concentrations of Fe and Cr atoms in the plasma-forming gas correlates with the content of these elements in the used electrode material, stainless steel.

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ВИЗНАЧЕННЯ ТЕМПЕРАТУРИ ЗАСЕЛЕННЯ ЕЛЕКТРОННИХ ЗБУДЖЕНИХ РІВНІВ АТОМІВ У ПЛАЗМІ АТМОСФЕРНОГО ТИСКУ З ВИКОРИСТАННЯМ ЕМІСІЙНИХ СПЕКТРІВ МЕТАЛІВ ІЗ СКЛАДНОЮ СЛАБКО РОЗДІЛЕНОЮ МУЛЬТИПЛЕТНОЮ СТРУКТУРОЮ

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Резюме

Методом оптичної емісійної спектроскопії досліджено параметри плазми ковзаючої дуги в потоці аргону. Показано, що зареєстровані спектри дослідженої плазми містять багато перекладаних слабко розділених мультиплетних ліній, які відповідають емісії матеріалу електродів. Щоб обробити отримані спектри, було проведено моделювання основних компонентів сталі (Fe, Cr, Ni) а також їхніх сплавів (нержавіюча сталь) із врахуванням апаратної функції використаного спектрометра в широкому діапазоні температур заселення електронних рівнів $T_e^* = 2000$ –20000 K. Запропоновано спектральні інтервали для проведення адекватного порівняння експериментальних

даних з результатами моделювання. Виявлено, що відношення концентрацій атомів Fe та Cr у дослідженій плазмі корелює з вмістом цих елементів в матеріалі використаних електродів (Fe~70%, Cr~20%).

ОПРЕДЕЛЕНИЕ ТЕМПЕРАТУРЫ ЗАСЕЛЕНИЯ
ЭЛЕКТРОННЫХ ВОЗБУЖДЕННЫХ УРОВНЕЙ
АТОМОВ В ПЛАЗМЕ АТМОСФЕРНОГО
ДАВЛЕНИЯ С ПОМОЩЬЮ ИСПОЛЬЗОВАНИЯ
ЭМИССИОННЫХ СПЕКТРОВ МЕТАЛЛОВ
СО СЛОЖНОЙ СЛАБО РАЗДЕЛЕННОЙ
МУЛЬТИПЛЕТНОЙ СТРУКТУРОЙ

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Резюме

Методом оптической эмиссионной спектроскопии исследованы параметры плазмы скользящей дуги в потоке аргона. Пока-

зано, что зарегистрированные спектры исследуемой плазмы содержат множество перекрывающихся слабо разрешенных мультиплетных линий, которые соответствуют эмиссии материала электродов. Чтобы обработать полученные спектры, проведено моделирование основных компонент стали (Fe, Cr, Ni) а также их сплавов (нержавеющая сталь) с учетом аппаратной функции используемого спектрометра в широком диапазоне температур заселения электронных уровней $T_e^* = 2000-20000$ К. Предложены спектральные интервалы для проведения адекватного сравнения экспериментальных данных с результатами моделирования. Обнаружено, что отношение концентраций атомов Fe и Cr в исследуемой плазме коррелирует с содержанием этих элементов в материале используемых электродов (Fe~70%, Cr~20%).