Application of line-intensity-ratio method for measurement of electron temperature of radio-frequency plasma of argon in magnetic field inside the plasma separator

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Abstract. The buffer plasma in plasma separator of substances must not lead to an additional ionization of ions to be separated, therefore it is needed to monitor its electron temperature. For this purpose an emission line-ratio method was used. In the paper optical diagnostic of argon rf-plasma \((f \approx 5 \text{ MHz})\) in magnetic field (130 and 650 G) was carried out. Argon pressure was of 5 mTorr. Corona model was used for the description of emission processes in plasma. Radial distribution of electron temperature was obtained using measured intensities of 763.5 and 811.5 nm argon emission lines. For data reprocessing the excitation from the ground state and metastable levels was taken into account. The obtained results are in agreement with data, measured from a double probe.

1. Introduction

Reprocessing of a spent nuclear fuel (SNF) is one of the priority tasks for modern atomic energy for the most purpose of nuclear fuel cycle closure in view of a nature resources limitation. One of the alternatives to chemical reprocessing methods is a plasma separation technology [1]. This technology implies SNF conversion in a low-temperature plasma flow that spatially separates in presence of electromagnetic field into two gropes (actinides and fission products) in terms of charge to mass ratio. Required electrical potential shape is produced in magnetized buffer plasma that also increase productivity of the method. During the implementation of the SNF plasma separation method it is necessary to avoid an additional ionization of flow ions by electrons of buffer plasma. Therefore a task of buffer plasma parameters diagnostics takes place. For this purpose a line intensity ratio method was applied for argon rf-plasma electron temperature \((T_e)\) measurements. Argon spectral lines to be observed (763.5 and 811.5 nm) were chosen form the assumption of low values of \(T_e\) (1–10 eV) realizing in the experiment. It should be noted that line intensity ratio method is widely used for argon plasma diagnostics [2,3], in particular in the area of dusty plasma [4].
2. Used theory of line intensity ratio method

For description of radiation and excitation processes of atoms in studied plasma a corona model was used [5]. Area of its application lays at low pressures of 0.1–1 Pa and low ionization degrees \( \sim 10^{-6} \text{–} 10^{-4} \) [6], that are close to the parameters of measuring rf-plasma of plasma separator \((P_{\text{Ar}} \approx 0.5 \text{ Pa, } \alpha \approx 0.01–1\% )\) [7]. Within the framework of corona model an elementary two-level system is usually considered, where the number of excited atoms is low in comparison with ground state (g.s.) population. The population of top level forms only by collisions with electrons and de-excitation happens only as a result of spontaneous radiation. The balance between these two processes defines intensity of spectral line. In used model spectral line intensity as a function of electron energy \( E \) is [2]:

\[
I_{ij} \sim n_0 n_e \int_0^\infty \sigma_{ij}^{\text{Opt}}(E) f(E) \sqrt{2E/m_e} \, dB,
\]

where indices \( i \) and \( j \) indicate ground and excited level correspondingly, \( \sigma_{ij}^{\text{Opt}} \) is an optical cross-section, \( f(E) \) is the electron energy distribution function, \( n_0 \) is a density of atoms in g.s., \( n_e \) is an electron density, \( m_e \) is the electron mass.

It was considered that plasma electrons in our rf-discharge have Maxwellian distribution, that confirms by estimation of ratio of power transferred to electrons from rf-field to power of thermalization. In this case relation (1) becomes

\[
I_{ij} \sim n_0 n_e \int_0^\infty \sigma_{ij}^{\text{Opt}}(E) e^{-E/(kT_e)} \sqrt{2E/m_e} \, dB,
\]

where \( k \) is the Boltzmann constant. From formula (2), one can see that the relation of two lines intensities depends only on electron temperature, and it allows determining of \( T_e \) by experimental measurement of \( I_{ij}/I_{ab} \).

The key point during an adaptation of this method to specific experimental conditions is a choice of spectral lines for the observation. It is defined first of all by expected range of electron temperature, and it allows determining of \( T_e \) by experimental measurement of \( I_{ij}/I_{ab} \).

In figure 1 argon \( 2p_0 \) excitation cross-sections [8] from g.s. (blue) and from 1\( s_5 \) metastable level (red) and also Maxwellian distribution for \( T_e = 3 \text{ eV} \) are presented. It is clearly visible that for only g.s. \( \rightarrow 2p_0 \) transition in (2) a significant part of plasma electrons will be considered within the integral if they have equilibrium temperature close to the threshold energy. Otherwise the noticeable amount of electrons will not contribute in population of excited level and will not be accounted at calculation of \( T_e \). The usage of levels which population is formed by excitation from metastable (for example 1\( s_5 \)) provides a significantly bigger intersection of excitation cross-section and electron distribution function, that will impact on accuracy of \( T_e \) determination and will reduce influence of \( f(E) \) deviation form Maxwellian distribution on its tail. Therefore in line-ratio method for diagnostics of the system with \( T_e \) of 1–10 eV we include in consideration the third level—metastable, and the excitation was accounted from it and from g.s. In this extended corona model the formula (2) for intensities ratio becomes [9]

\[
\frac{I_{ij}}{I_{ab}} = \frac{n_0 \int_0^\infty \sigma_{ij}^{\text{g.s.}}(E) e^{-E/(kT_e)} \sqrt{2E/m_e} \, dB + n_m \int_0^\infty \sigma_{ij}^{\text{m}}(E) e^{-E/(kT_e)} \sqrt{2E/m_e} \, dB}{n_0 \int_0^\infty \sigma_{ab}^{\text{g.s.}}(E) e^{-E/(kT_e)} \sqrt{2E/m_e} \, dB + n_m \int_0^\infty \sigma_{ab}^{\text{m}}(E) e^{-E/(kT_e)} \sqrt{2E/m_e} \, dB},
\]

where indices \( m \) and g.s. mean metastable and the ground state, \( n_m \) is the density of atoms in metastable level.
Taking into account the above reasoning we chose argon lines 763.5 (2p<sub>6</sub> → 1s<sub>5</sub>) and 811.5 nm (2p<sub>9</sub> → 1s<sub>5</sub>) for observation. A population on these levels forms by excitation from g.s. and metastable level (1s<sub>5</sub>) approximately equally [10].

The determination of \( T_e \) using measured plasma radiation spectrum was performed with the help of calculated function \( I_{763.5}/I_{811.5} = f(T_e) \) based on (3) and reference data of cross-sections [11] and relative population \( n_m/n_0 \) [9] for argon pressure of 5 mTorr.

### 3. The experiment

Diagnosed argon plasma was generated in steel cylindrical chamber with a length of about 2 m and internal diameter of about 1 m [7]. Argon working pressure was set by a vacuum leak valve and was \( 5 \times 10^{-3} \) Torr, residual gas pressure was less than \( 5 \times 10^{-5} \) Torr. The discharge was initiated with the help of a lamp generator that applied ac voltage on frequency close to 5 MHz to a double-turn antenna. For the decrease of capacitive coupling between the antenna and the plasma and for more effective rf-power transfer into the plasma a Faraday shield was used [7]. The expansion of argon plasma along the system axis was provided by presence of external magnetic field which intensity on the center of the chamber can be varied from 0 to 650 G. Visually area of plasma glow had a diameter of near 30 cm.

Optical diagnostic of plasma radiation was carried out in a central plane that is perpendicular to the axis of system symmetry (figure 2). For the determination of \( T_e \) radial distribution in this plane with the help of achromatic objective a radiation from different chords was collected. The diameter of radiation collection area from a chord was about 1 cm. The accuracy of the distance between the center and a chord was not worse than 2 cm. The plasma radiation was registered by previously calibrated wide-range spectrometer SDH-IV, which spectral resolution was of 0.4 nm in a wavelength range of 450–900 nm. Signal exposure of spectrometer ranged from 30 to 500 ms. The results of optical measurements of \( T_e \) radial distribution were compared with data, obtained by double probe.
4. Results and discussion

During the experiments intensities of chosen spectral lines were determined along 9 different chords of the chamber. Next, registered distributions were approximated by a smooth curve and with the help of reverse Abel transform [12] lines intensity distributions along chamber radius were calculated. An example of approximation results and calculated line intensity radial distributions are presented in figure 3.

Optical diagnostics of the discharge was carried out at two intensities of magnetic field 130 and 650 G. In figure 4 there are results of optical (blue points) and probe (red points) $T_e$ measurements along the radius.

As can be seen in figure 4(a) at magnetic field of 130 G the results of $T_e$ measurements by optical method and double probe are coincide within the error. Before $r = 17$ cm $T_e$ is approximately constant of about 3.5 eV and on the periphery there is a light decrease to 2.5 eV. At high magnetic field of 650 G optical measurements give slightly higher results on 1–2 eV, figure 4(b). Both methods give a similar initial slope at $r = 0$ to 5 cm. Near $r = 10$ cm $T_e$ is stable but optical results give value of 5 eV and probe show 3 eV. The most significant difference is at $r > 15$ cm, obtained results show opposite trends of $T_e$. It should be noted that plasma density away from the axis decreases by more than order of magnitude compared to the center ($10^{10}$ against $3 \times 10^{11}$ cm$^{-3}$ by double probe data).

Observed difference in $T_e$ measurement at $B = 650$ G can be caused also by specificity of probe measurements at high magnetic fields. When moving from $B = 130$ to 650 G the value of ratio between the Larmor radius of the electron to the diameter of the probe ($d = 0.3$ mm) for $T_e = 3$ eV passes through the unit (changing from 2 down to 0.4). So, the collection conditions of charged particles by the probe are changing, and it can effect on measurement results [13]. Other purposes of the results mismatch at high magnetic field should be considered separately.

The main error sources of used line intensity ratio method should be listed also. We do not point optical method error in figure 4 because it is hard to assess, but we assume it is not exceed 30%. Used plasma model do not account excitation from resonance levels $1s_2$ and $1s_4$ because an effect of radiation trapping is low at low pressures and it give negligible contribution in population of high levels [14]. An excitation from metastable $1s_3$ was neglected because of significant lower cross-sections of $2p_6$ and $2p_9$ excitation than from $1s_5$. Major errors can
be connected with dependencies of $n_m/n_0$ and $\sigma_{ij}^{Opt}$ from pressure, which define the relation $I_{763.5}/I_{811.5}$ from $(T_e)$. So, the instability of pressure during the experiment can lead to mistakes. In general, to increase the accuracy of the line-ratio method it is possible to include in the consideration (if necessary) levels mentioned above, as well as to use the collisional-radiative model instead of the coronal model that is significant simplification.
5. Conclusion
Let us list the main obtained results. For determination of argon rf-plasma electron temperature the line intensity ratio method was used, where main plasma processes were described by corona model. Accounting low values of measuring $T_e$ two spectral lines were chosen for observation, they are 763.5 (2p$_6$ → 1s$_5$) and 811.5 nm (2p$_9$ → 1s$_5$). Based on registered radiation from different cords of cylindrical plasma volume the radial distribution of line intensities was obtained and $T_e$ distribution was calculated. The measured $T_e$ at low magnetic field of 130 G is in good agreement with results of probe measurements. At high magnetic field of 650 G average optical measurement results exceed probe one by 1 eV. In general applied in this work line intensity ratio method can be used during $T_e$ measurements in plasma separator.

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