CALCULATION OF POPULATION DENSITIES OF EXCITED CI AND CII LEVELS IN HELIUM AND ARGON PLASMAS CONTAINING SMALL ADMIXTURES OF CO_2 — IMPORTANCE TO ANALYSIS OF RADIATION EMITTED FROM PLASMAS OF AXIAL SYMMETRY

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Population densities of excited CI and CII levels are calculated in helium and argon plasmas containing small admixtures of CO_2. Calculations are performed for a total pressure of 1 atmosphere, in a temperature interval from 8000 K to 40000 K, assuming local thermal equilibrium (LTE) and partial local thermal equilibrium (pLTE) conditions. Normal temperatures are obtained for selected excited CI and CII levels. The results are applied to a helium plasma with traces of CO_2, of cylindrical symmetry with presumed radial temperature distribution. Effective intensities of CI and CII spectral lines, corresponding to side-on radiance measurements along the cylinder diameter are evaluated. On hand of these effective line intensities and applying the Boltzmann plot method effective temperatures are evaluated and compared with the presumed temperature distribution.

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1. Introduction

Studies of processes occurring in plasmas require to evaluate its most important parameters: the temperature and densities of various components. In basic research as well as in technological applications helium and argon plasmas are often applied. Frequently, for diagnostic purposes, small amounts of additional matter (elements, molecules) are introduced to the working gas in order to obtain the corresponding spectra and — on that basis — to evaluate the required plasma parameters (e.g. [1–4]). It is also well known that introduction of traces of elements into rare gas plasmas (especially in helium plasmas), leads to a very efficient population of excited levels of the admixtures, which facilitates spectral line intensity measurements for these elements [5–9]. Often it is assumed that these small admixtures do not influence the plasma parameters noticeably (e.g. [10, 11]).

In many cases plasmas generated in various devices reveal axial or cylindrical symmetries — the plasma parameters, at a given cross section, depend only on the distance from the axis of the plasma. Measurements of lateral (side-on observation) distributions of radiation allow us to determine (after the Abel inversion) the radial distributions of emission coefficients of the plasma for bound-bound transitions and for transitions contributing to the continuum.

Such conditions are found in various arc discharges (including the wall-stabilized arc), in pinch discharges, in plasmas generated as a result of absorption of laser radiation in gases (so-called optical discharges) or plasmas arising at the surface of solid materials, where the laser beam is absorbed, and in many other devices.

Sometimes the determination of lateral distribution of radiation is difficult, sometimes even impossible, as in the case, when the plasma produced in a wall-stabilized arc is observed through a small hole in one of a stabilizing section [5–8]. In such cases, one is compelled to interpret the "averaged" radiation of the plasma and evaluate some "mean" plasma parameters. This procedure is entirely justified in many technological applications, e.g. in the case of plasmas appearing at metal sheets during cutting processes by the use of focused laser beams [12].

The aim of this work is to study the population densities of excited CI and CII levels in helium and argon plasmas with small amounts of CO_2. The results obtained for a plasma at atmospheric pressure are applied to a cylindrical plasma column with presumed radial temperature profile, simulating typical conditions for wall-stabilized arc discharges. In this way "effective" temperatures of the plasma are evaluated and compared with the presumed temperature distribution.

2. Calculations of parameters of plasma

Calculations of the plasma parameters were performed assuming that (i) the total pressure of the plasma amounts 1 atm, (ii) the plasma consists of electrons, neutral, singly and doubly ionized atoms of the noble gas (helium or argon) as well as of carbon and oxygen, (iii) the densities of oxygen and carbon in the plasma always correspond to the stoichiometrical ratio of CO_2, i.e. $(N_{\mathrm{O}} + N_{\mathrm{O}^+} + N_{\mathrm{O}^{++}}) = 2\,(N_{\mathrm{C}} + N_{\mathrm{C}^+} + N_{\mathrm{C}^{++}})$.

The description of the set of equations, which has to be formed, in order to evaluate the required parameters of the plasma, can be found elsewhere [13, 14]. The set was completed by two relations for determining the lowering of the ionization energy, which has to be applied in the Saha equations and for evaluation of the pertaining partition functions [15, 16]. The numerical values of the partition functions were obtained from Tables of Drawin and Felenbok [17].

The calculations were performed by (a) applying the LTE-model and (b) applying a simplified pLTE-model. Within the pLTE case we assumed that only the ground states of the neutral atoms are overpopulated. The overpopulation factors a were defined according to [13] as follows:

$$\frac{N_m}{N_1} = \frac{1}{a}\frac{g_m}{g_1}\exp\left(-\frac{E_m}{kT}\right), \qquad \frac{N^+}{N^0}N_{\mathrm{e}} = \frac{1}{a}S(T, \Delta\chi), \tag{1}$$

where: N_m, g_m, E_m are the population, statistical weight and excitation energy of the selected level m, respectively; N_1 is the density of the atoms in the ground level; N^+, N^0, N_e are the ion, atom and electron densities, respectively; $\Delta\chi$ is the lowering of the ionization energy; $S(T, \Delta\chi)$ is the Saha function; kT is the temperature in energy units. The a-values were chosen somewhat arbitrarily: $a_{He} = 100$, $a_{Ar} = 10$, $a_O = 10$ and $a_C = 2$, scaling with the third power of the maximum energy gap in the corresponding atomic energy level system. In the same way scales the criterion for the electron density value, which is indispensable for establishing LTE in the plasma [18, 19]. The results obtained within collisional-radiative models, developed for helium [20, 21] and argon plasmas [22, 23] indicate that our arbitrary choice of the set of a-values is reasonable.

3. Results and discussion

3.1. Discussion of the population densities of CI and CII levels

Population densities, defined as $\rho_m = N_m/g_m$, of various excited CI and CII levels, in helium and argon plasmas, containing small amounts of CO_2, were calculated in the temperature range from 8000 K to 40000 K. In Fig. 1, the population densities of the selected CI term ($3p\,^3D$) with an excitation energy of 8.64 eV, are shown as a function of temperature for 3 different admixtures of CO_2, corresponding to the fraction of 0.01%, 0.1% and 1% of carbon in the helium (a) and in the argon (b) plasma. In both figures, the population density functions (excitation functions) obtained for a plasma produced from decomposition of CO_2 at atmospheric pressure are presented for comparison (33% carbon content). In the same figure the temperature dependences of the resulting electron density for the plasmas with 0.1% carbon content are shown.

As can be seen from Fig. 1a, the excitation functions of CI levels in the argon plasma show a typical course in both the LTE approximation as well as in the case of the simplified pLTE model. On the contrary, the excitation functions of the CI levels in the helium plasma (see Fig. 1a) exhibit two distinct maxima, the low and the high temperature maximum. These temperatures are called the normal temperatures [24]. Similar results have been obtained by Diermeier and Krempl [25] for excited atomic levels of Na, Al, Fe and Zn in the case of an argon plasma at atmospheric pressure with small admixtures of always one of these elements. For details about the causes and processes, which lead to the appearance of these two maxima we refer to the last reference.

The excitation functions for all CII levels reveal typical temperature dependences, with maxima depending mainly on the excitation energy of the respective level and much less on the kind of the main plasma component (argon or helium) as well as on the applied approximation (LTE or pLTE). The normal temperatures for levels with $E_m \geq 16$ eV exceed the value of 25000 K and are very close to the normal temperatures obtained for the CO_2 plasma.

Taking into consideration: (i) the general criteria for establishing LTE in plasmas, (ii) the specific results of collisional-radiative models for helium and argon plasmas, and (iii) the electron density dependences on temperature (shown in Fig. 1) one can conclude that our pLTE results comply much better in the low

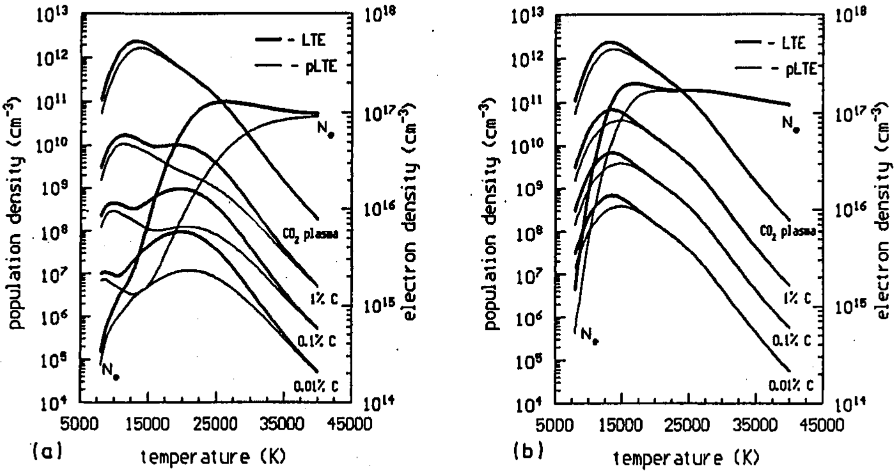

Fig. 1. Population densities of the CI term $3p\,^3D$ with an excitation energy of 8.64 eV as a function of temperature in a helium (a) and in an argon (b) plasma at atmospheric pressure, for 3 different admixtures of CO_2, and for a plasma produced from decomposition of CO_2. The courses of electron density correspond to a 0.1% carbon content in the helium (a) and argon (b) plasma. The thick lines represent the LTE results, the thin lines the pLTE results.

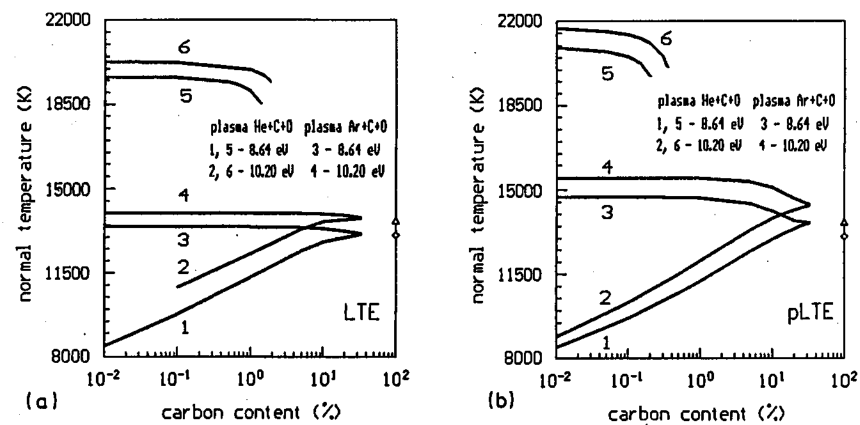

Fig. 2. Normal temperatures of 2 selected CI levels evaluated in the case of helium and argon plasmas with CO_2 admixture as a function of carbon content in the plasma. The results for 33% carbon content correspond to the case of a plasma produced from decomposition of CO_2 (without helium or argon admixture). The symbols corresponding to 100% carbon content represent results for a pure carbon plasma (LTE) at atmospheric pressure: ◇ for $E_m = 8.64\,\mathrm{eV}$, Δ for $E_m = 10.20\,\mathrm{eV}$. The curves 5 and 6 show the second (higher) normal temperature appearing in the helium plasma. At higher carbon concentrations the second maximum vanishes, see also Fig. 1a. In part (a) and (b) the results obtained within the LTE and pLTE approximation are shown, respectively.

temperature range ($T \leq 22000$ K for He, and $T \leq 11500$ K for Ar plasma), while the LTE results comply in the high temperature range ($T \geq 22000$ K for He and $T \geq 11500$ K for Ar-plasma).

In Fig. 2 the evaluated normal temperatures for two selected levels are shown as a function of the carbon content in the helium and in the argon plasma. In the part (a) of the figure the results obtained applying the LTE model, while in the part (b) applying the pLTE approximation, are presented. In the case of helium plasmas also the second (higher) normal temperatures for CI levels, obtained at very low carbon admixtures are presented. (At higher carbon concentrations the second maximum in the excitation function vanishes, see also Fig. 1a.)

As can be seen from Fig. 2, at low carbon admixtures, the normal temperatures for a given CI level depend strongly on the "working" gas, in which the carbon atoms are excited. The difference between the normal temperature for the CI level with $E_m = 8.64$ eV in helium and argon reaches even 5000 K. In the case of CII levels the difference does not exceed 100 K. Generally, in the case of the argon plasma the carbon content does not change essentially the normal temperatures for both the CI as well as CII levels. Only at larger carbon densities in the plasma, the normal temperatures are influenced. This is obvious, because argon then plays the role of admixture in the CO_2 plasma.

In the case of the helium plasma and the CI levels, the "first" normal temperature increases nearly linearly with the logarithm of the carbon content. Below carbon admixtures of 0.01% it is difficult to determine the normal temperatures (see the trend of the shape of the excitation functions in Fig. 1a). Our calculations show also that the normal temperatures only insignificantly depend on the applied equilibrium model, compared with the influence of the kind of the "wórking" gas.

3.2. Application of the results to the analysis of a plasma of cylindrical symmetry

The complex temperature dependence of the excitation functions of CI levels may be essential for the analysis of radiation emerging from plasmas with a temperature gradient along the direction of radiation emission. As can be seen from Fig. 2, in the case of helium plasmas, the first normal temperature strongly depends on the carbon content in the plasma. The temperature interval, in which the normal temperatures appear for the different plasma compositions, corresponds to conditions of typical arc discharges at atmospheric pressure. Therefore we decided to apply our results to the analysis of a plasma of cylindrical symmetry, simulating the side-on emission of CI and CII spectral lines from wall-stabilized arcs. Detailed analyses were performed for a cylindrical ($r = 2$ mm) helium plasma with an admixture of CO_2, which corresponds to a 0.2% content of carbon and meets the conditions of experiments [5, 8].

Calculations of the radial distribution of population densities were accomplished on the basis of a presumed radial temperature distribution, which is typical for a wall-stabilized arc at atmospheric pressure. Again two approximations were applied — the LTE and the simplified pLTE. Because for a plasma with temperature and electron density gradients one has to expect the occurrence of a demixing effect [26–29], two sets of calculations were performed assuming that (a) the concentration of the admixture increases with increasing radius, and (b) neglecting the demixing effect, i.e. assuming that the distribution of carbon and oxygen is uniform. It was assumed that the stoichiometrical relation of CO_2 holds at every point of the plasma volume.

For comparison similar calculations were performed also for an argon plasma, with a slightly changed radial temperature distribution and with the same amount

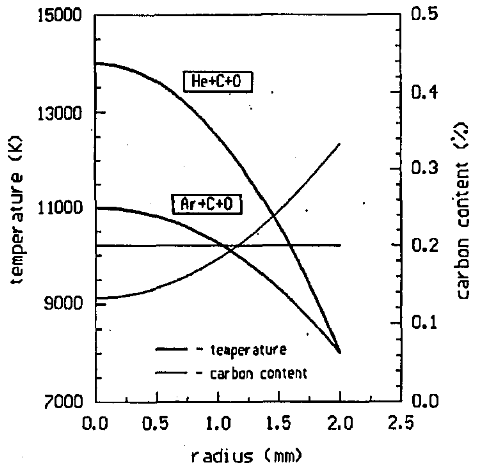

Fig. 3. Presumed radial distributions of the temperature and the carbon density, simulating the conditions of a wall-stabilized arc.

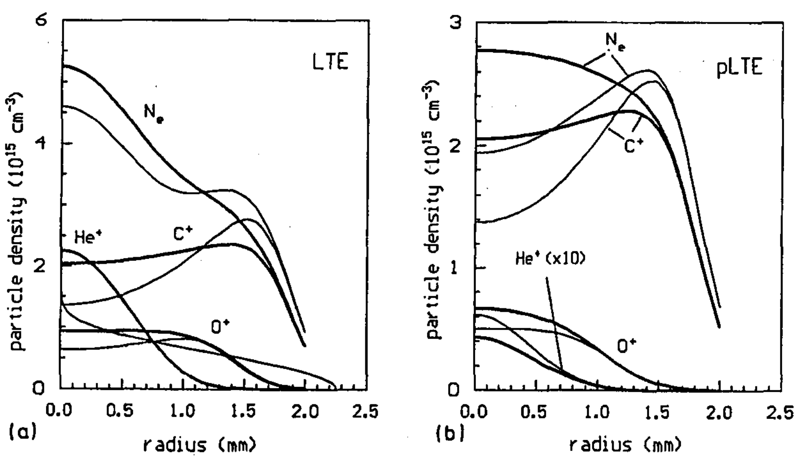

Fig. 4. Evaluated radial distributions of charged particles in the helium plasma with 0.2% admixture of CO_2. The thin lines represent results including the occurrence of the demixing effect, while the thick lines represent results neglecting the demixing effect. Part (a) shows the results obtained within the LTE approximation, while part (b) those within the pLTE approximation.

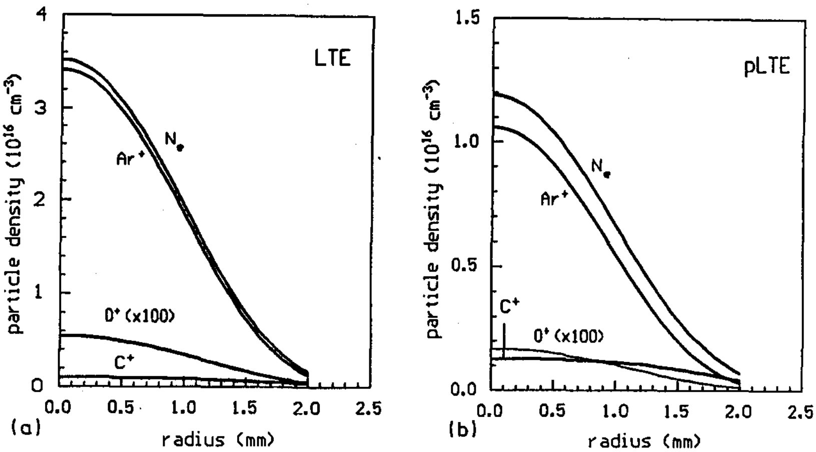

Fig. 5. Radial distributions of charged particles in the argon plasma with 0.2% admixture of CO_2. (a) LTE results, (b) pLTE results.

of CO_2 uniformly distributed in the plasma. (The demixing effect in the argon plasma is expected to be less pronounced, because the ionization energies of all three plasma components are similar.)

Figure 3 shows the postulated radial temperature distributions for the helium and argon plasmas and the assumed radial distributions of the carbon impurity. In Fig. 4 the evaluated radial distributions of the electron and ion densities in the case of the helium plasma are shown. In Fig. 5 the corresponding results for the argon plasma are presented.

As one can see from Fig. 4 the pLTE calculations predict rather flat N_e distributions in the central part of the arc ($r \leq 1$ mm) and strong gradients in the outer part of the plasma column ($r > 1.5$ mm). The assumption of a rather weak demixing effect (the concentration of carbon at the arc center is only 2.5 times smaller than at the arc boundary) leads even to a maximum electron density outside the arc axis. This is directly caused by the efficient ionization of carbon, yielding the main contribution to the electron density, despite the very low CO_2 admixture. In the case of the LTE approximation this effect is less pronounced but clearly evident, leading to a complex N_e radial distribution. In the case of the argon plasma, the main contribution to N_e is originating from the ionization of the main plasma component (argon) and therefore, as expected, no peculiarities in the distribution of the plasma components appear.

3.3. Evaluation of averaged plasma temperature equivalent to side-on line intensity measurements

As mentioned earlier, in some cases one is compelled to interpret the measured "averaged" spectra, i.e. the radiation which is integrated along the diameter of the plasma column, without having the opportunity to measure the whole lateral intensity distribution, allowing the Abel inversion to be executed. Such measurements have been performed, e.g. in [5–8], applying the wall-stabilized helium arc with traces of N_2 or CO_2, where the radiation emitted side-on through a 1.5 mm diameter hole in the central disk of the arc has been measured. In these experiments the Boltzmann plot method [30] was applied for temperature determination. Applying ionic lines, T-values of about 14000 K were determined, while using atomic lines significantly lower temperatures were obtained ($T \cong 10000\,\mathrm{K}$). The higher "ionic" temperatures were confirmed by end-on measurements on the arc axis.

In order to explain these discrepancies, we calculated the population density of selected CI levels along the arc radius, assuming the temperature and carbon density distributions shown in Fig. 3. The results are presented in Fig. 6. Subsequently the "averaged" population densities were calculated

$$\overline{\rho}_m = \frac{1}{R}\int_0^R \rho_m(r)\mathrm{d}r, \tag{2}$$

where $R = 2$ mm, is the radius of the arc column. On the basis of these averaged population densities the effective temperatures of the plasma were evaluated

$$T_{\mathrm{eff}} = \frac{E_2 - E_1}{k\ln\left(\overline{\rho}_1/\overline{\rho}_2\right)}, \tag{3}$$

where E_1, E_2 are the excitation energies of the selected levels, $\overline{\rho}_1$, $\overline{\rho}_2$ — the corresponding averaged population densities. The temperature values obtained in this manner correspond to the results obtained by the use of the Boltzmann plot method.

The calculations were accomplished applying the LTE as well as the pLTE model. In both cases the calculations were performed with and without the presence of the demixing effect in the plasma. For comparison, analogous calculations were done for the argon plasma, but only in the case of the uniform distribution of

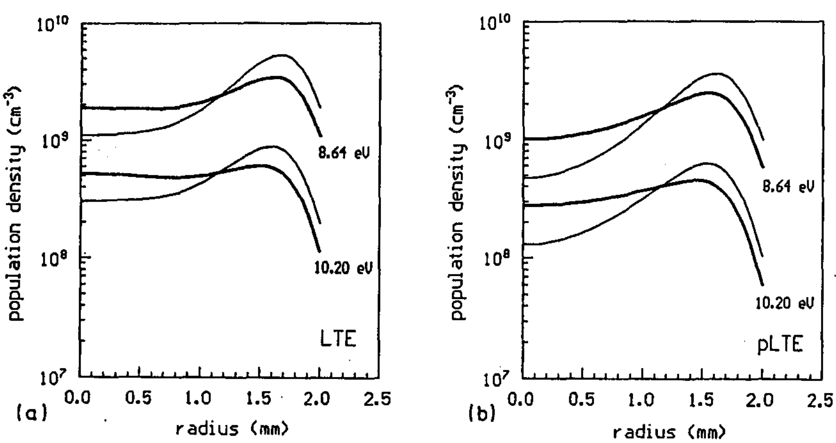

Fig. 6. Population density distributions of selected CI levels in the helium plasma resulting from the temperature and carbon admixture distributions shown in Fig. 3. The thin lines represent the case of demixing occurrence, while the thick lines represent the case of uniform carbon distribution.

TABLE

Effective temperatures (defined by Eq. (3) obtained from "averaged" population densities of selected CI and CII levels for different plasma compositions and for different equilibrium approximations (LTE, pLTE).

	E_2 [eV]	E_1 [eV]	Effective temperature [K]					
			Ar + CO_2		He + CO_2 without demixing effect		He + CO_2 with demixing effect	
			LTE	pLTE	LTE	pLTE	LTE	pLTE
CI	4.18	7.48	10310	10320	10720	10660	10170	10140
	4.18	13.12	10440	10450	11510	11420	10910	10820
	8.85	10.35	10500	10510	11890	11780	11230	11100
	9.76	12.48	10550	10560	12290	12180	11680	11520
CII	16.33	22.87	10810	10810	13590	13590	13530	13530
	20.48	27.38	10830	10840	13660	13660	13620	13620
assumed temperature interval (see Fig. 3)			8000–11000		8000–14000		8000–14000	

the admixture for reasons explained earlier. The evaluated population density distributions of CI and CII levels in the argon plasma show a typical shape reflecting the assumed temperature distribution, without any characteristic features. The same refers to the population density of CII levels in the helium plasma. In Table the effective temperatures obtained in this way applying different pairs of energy levels are quoted. As expected the evaluated effective temperatures, obtained on the basis of calculated populations of excited levels of ionized carbon, are very close to the assumed temperature value at the arc axis (for the argon as well as for the helium plasma). This effective temperature is only slightly influenced by the selected levels applied for T-evaluation as well as by the inclusion of the demixing effect. The model applied in calculations (LTE, pLTE) does not influence the obtained effective temperatures based on populations of CII levels.

All the above drawn conclusions hold for the effective temperatures obtained from CI level populations in the case of the argon plasma with CO_2 admixture. This is not surprising since also in this case the plasma volume around the arc axis yields the main contribution to $\overline{p}_m$ evaluated from Eq. (2).

On the contrary the results for the helium plasma are more differentiated. The effective temperatures depend strongly on the excitation energy of the levels chosen for T-determination. The inclusion of the demixing effect leads to an essential decrease in the effective temperature since the contribution from plasma layers outside the arc axis is more pronounced in this case. The applied model (LTE, pLTE) remarkably influences the resulting temperature — the pLTE results are always below those obtained at the LTE assumption.

The comparison of evaluated effective temperatures with the presumed temperature distributions reveals that in the case of the helium plasma the T-values based on levels of CII are close to the temperature around the arc axis ($r \leq (0.5\,\mathrm{mm})$, while the T-values obtained from CI levels are characteristic for plasma layers lying outside the arc axis $r \in (1.1–1.6\,\mathrm{mm})$.

4. Concluding remarks

Our calculations of parameters of noble gas plasmas containing very small admixtures of CO_2 show that traces of these admixtures may influence considerably the plasma parameters, particularly in the case of the helium plasma. In helium dominated plasmas, the normal temperatures of atomic carbon lines depend strongly on the CO_2 admixture. These temperatures, at fixed total pressure of the plasma, are shifted towards lower values compared with those for plasmas produced from decomposition of CO_2 or for "pure" carbon plasmas.

Our calculations of particle density distributions in the arc plasma show that the high temperature layers around the arc axis are mainly contributing to the ionic spectrum, while the cooler boundary layers are responsible for the atomic line radiation. From the evaluated data it is evident that temperature determination, based on measured intensity ratios of spectral lines originating from two subsequent ionization stages, often applied in diagnostic of plasmas, may be highly questionable in the case of plasmas showing strong temperature gradients.

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