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SOME TRANSPORT PROPERTIES IN PLASMAS CONTAINING ARGON AND FLUORINE

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Abstract. *In this paper some results of numerical evaluation of transport coefficients in plasmas in the mixtures of argon and fluorine are presented. These transport characteristics are given in the function of the temperature for low pressures ranging from 0,1 kPa to 1,0 kPa and for low temperatures between 500 K and 5 000 K in argon plasmas with 20% and 30% of the fluorine added. It is assumed that the system is kept under constant pressure and that a corresponding state of local thermodynamical equilibrium (LTE) is attained in it. The equilibrium plasma composition, necessary for the evaluations, was determined on the ground of the Saha equations for ionization processes and the law of mass action for the thermal dissociation of* F_2*, combined with the charge conservation relation and the assumption that the pressure remained constant in the course of temperature variations. The ionization energy lowering, required in conjunction with the Saha equations, was obtained with the aid of a modified expression for the plasma Debye radius proposed previously. A previously derived expression for the modified Debye radius, offering the possibility to treat the plasmas considered as weakly non-ideal in the whole temperature range, is used. The cut-off at the Landau length rather than of the smallest of ionic radii is introduced. This alteration in the evaluation procedure brings different considerable changes in the final numerical results for the all relevant quantities.*

Key words: *argon, fluorine, weakly non-ideal plasma, number density, electron and ion diffusion coefficients, electrical conductivity, thermal conductivity, collision frequency.*

1. INTRODUCTION

A very important technological problem, connected with a majority of practical applications of the noble-gas plasmas, is to improve their transport properties at lower temperatures. A simple way to solve this problem is the addition of a small amount of some different additives; this invariably leads to a significant increase in the electron and ion number densities. Theoretical investigations of various transport properties of low-

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temperature noble-gas plasmas with additives have aroused a continuous interest over a considerable span of time, due to numerous applications. Plasmas in mixtures of noble and other gases are of considerable interest from the fundamental standpoint and from numerous technological applications. Those formed in mixtures of noble gases and fluorine, in particular, are also of importance in different laboratory situations, for example, plasma etching and ion beam etching [1]. The basic characteristics of these plasmas in the investigation are the number densities of their constituents and the transport coefficients. Unfortunately, results of direct experimental measurements of these basic parameters are not always available, especially in the above conditions.

The equilibrium plasma composition was evaluated from a set of equations embracing the requirements of charge-conservation and conservation of total numbers of argon and fluorine nuclei, along with the equations for the law of mass action applied to the ionization stages of argon and fluorine and the dissociation processes of F_2 and F_2^+. The problem of the non-ideality according to standards criteria is circumvented by using an appropriately modified Debye radius r_D [2], rather than the conventional one, r_D [3-8]. In this evaluation procedure the cut off at the Landau length [9], rather than of the smallest of the ionic radii is used. This evaluation technique was already introduced in studies of noble gas plasmas with alkaline vapours as additive [2,3,4]. The transport coefficients were obtained here numerically for relatively low temperatures, from 500 K to 5 000 K and relatively low pressures, from 0,1 kPa to 1,0 kPa.

It was found that the use of the Landau length in the evaluation of the modified Debye radius (instead of the smallest ionic radii) is a better solution in the theoretical investigation.

2. OUTLINE OF THE THEORY

The composition and transport coefficients can be satisfactorily evaluated with the aid of the theory applicable to weak non-idealities, provided that in appropriately modified Debye radius, yielding a larger Debye number, is used. Thus, in the present paper, the equilibrium plasma composition, required for the evaluation of the transport coefficients, was obtained from the system of the Saha equations where the ionization energy lowerings were related to the modified Debye radius r_D^* given by

$$\left(\frac{1}{r_D^*}\right)^3=\frac{1}{r_D^3}-\frac{\pi}{3\varepsilon_0(kT)^4}\frac{\left(\sum_\alpha e_\alpha^4 n_\alpha\right)\left(\sum_\alpha e_\alpha^4 n_\alpha Q_\alpha^{'}\right)}{(4\pi\varepsilon_0)^3 r_D}, \tag{1}$$

Where

$$Q_\alpha^{'}=17{,}545-2\ln\frac{L}{r_D} \tag{2}$$

with L denoting the Landau length.

The expression (1) was derived [2] from Poisson equation for the potential of electrostatic field around a given ion in which, apart from the basic linear (Debye-Huckel) term, the first non-linear correction is also retained. The quantity L in (2) is a cut-off length instead of the ionic radii. This choice is more acceptable physically in the context of a plasma [6,7], and seems more justifiable *a posteriori* as it yielded final results.

The evaluation of the electron diffusion coefficient under above conditions was based on the expression given by [10]

$$D_e = \frac{4\pi}{3n_e}\int_0^\infty \frac{d}{d\upsilon}\left(\frac{\upsilon^5}{\nu_e(\upsilon)}\right) f_e^{00}(\upsilon)d\upsilon, \tag{3}$$

where $f_e^{00}(\upsilon)$ is the isotropic part of the Maxwellian electron velocity distribution function, and n_e is the velocity - dependent overall electron collision frequency.

The evaluation of the electrical conductivity in multi-component plasmas under the above conditions was based on the well-known expression given by [10]

$$\sigma_e = \frac{2e^2 n_e m_e^{3/2}}{3\sqrt{2\pi}}(kT)^{-5/2}\int_0^\infty \frac{\upsilon^4}{\nu(\upsilon)}\exp\left(-\frac{m_e\upsilon^2}{2kT}\right)d\upsilon. \tag{4}$$

The thermal conductivity is evaluated on the basis of Refs. 10. and 12.

$$\lambda_e = \frac{n_e m_e^{5/2}}{3\sqrt{2\pi T}}(kT)^{-3/2}\int_0^\infty \frac{\upsilon^6}{\nu_e(\upsilon)}\left(\frac{m_e\upsilon^2}{3kT}-\frac{3}{2}\right)\exp\left(-\frac{m_e\upsilon^2}{2kT}\right)d\upsilon, \tag{5}$$

An important criterion permitting to infer that $f_e^{00}(\upsilon)$ is Maxwellian is that the dimensionless parameter δ, determined by the frequencies of electrons collision with electron and heavy particles, according to

$$\delta = \sum_h \frac{m_e}{m_h}\frac{\nu_{eh}}{\nu_{ee}} << 1 \tag{6}$$

(h being here the index enumerating the heavy particles; $h = 1,2,3,4$ in the cases studied); $\nu_{eh} = \nu_{ei} + \nu_{en}$. With $\delta << 1$, the Maxwellian electron distribution function can be reliably assumed [12].

The knowledge of effective collision frequencies for encounters between electron and heavy particles (neutrals and ions) is also of interest. The electron collision frequency in noble-gas plasmas with different additives was evaluated previously by the authors of the present paper [3-8], and here this quantity is taken in its usual form

$$\nu_e(\upsilon) = \upsilon\left[\sum_{ion} n_i Q_{ei}(\upsilon) + \sum_{atom} n_a Q_{ea}(\upsilon)\right]; \tag{7}$$

the summation is extended over all the heavy species present. The quantities n_i and n_a in (2) are the ion and atom number densities, while $Q_{ei}(\upsilon)$ and Q_{ea} denote the pertaining velocity-dependent momentum-transfer cross sections. The influence of the plasma non-ideality on the $e - i$ collision processes is accounted for by the use of modified Debye radius (rather than r_D in the Coulomb logarithms. The diffusion coefficients of ions are evaluated with the aid of Einstein relation with the expression from [11] in modified form [5-8].

3. RESULTS AND COMMENTS

In plasmas formed in mixtures of argon and fluorine under the conditions specified above, a large variety of charged and neutral species exists: e, Ar, F, F_2, F_2^+, A_r^+ and F^+. (Higher ionization stages are excluded by the temperature range assumed.) For the applications in etching technology, ionic components are of primary interest, but the analysis of the transport properties requires the knowledge of number densities of all the components. A previously derived expression for the modified Debye radius r_D^* [2], offering the possibility to treat the plasmas considered as weakly non-ideal in the whole temperature range, is used and the cut-off at the Landau length, rather than of the smallest of the ionic radii is introduced.

The plasma equilibrium composition, as well as some transport coefficients were obtained here numerically, for relatively low temperatures (from 500 K to 5 000 K) and relatively low pressures (from 0,1 kPa to 1,0 kPa). The temperature dependence of the evaluated important transport characteristics, with pressure as parameter, in Tables 1. to 4. is given. The tables show the temperature dependence of evaluated electron number densities, electron collision frequencies, electron and ion diffusion coefficients and electrical and thermal conductivities for the argon plasma with 20% and 30% of F_2 respectively. The given values for these transport properties show the temperature variations, for the two pressures studied, in the argon plasma with admixed fluorine.

In the physical situation considered presently, the electron number density is entirely determined by the processes of thermal ionization including the ionization energy lowerings. The number densities of electrons tend to increase with the temperature, and one can easily grasp the significance of the relations between the ionization and dissociation energies of the neutral species present. The energy of the first ionization of fluorine (17,42 eV) is 1,11 times higher that of argon (15,75 eV); the energy of dissociation of fluorine molecule (1,60 eV) is 2,10 times smaller than that of F_2^+ (3,30 eV). On the other hand, the energy of the first ionization of fluorine atom is 1,11 times higher than that of fluorine molecule.

An inspection of the Tables discloses that the increase of the pressure raises the electron number density. This is an easily recognizable feature of the Saha equation. It is of interest to mention that the ratio of F^+ number densities for the two pressures considered 0,1 kPa and 1,0 kPa is about unity at temperatures around 1 000 K (the same goes for the electron number densities). At lower temperatures the ionization of fluorine is not massive and that of argon is very small. The overall degree of ionization is very small and this quantity is very little sensitive to pressure variations. Again, all these details are amenable to the specificities of the Saha equation and the ionization energy lowerings.

Under these conditions, the electron-heavy particles scattering dominate. The values ν_{eh} are higher for the pressure of 1,0 kPa (about one order of magnitude larger) for both presence fluorine in the plasma mixture.

It is seen that, at higher temperatures, the electron diffusion coefficients are about four orders of magnitude larger than the corresponding overall diffusion coefficients of ions. In the whole temperature range examined, both D_e and D_i are lower in argon plasma with 30 % of fluorine as additive. Also, both diffusion coefficients show a higher sensitivity to pressure variations at temperature above 1 500 K. The diffusion coefficients decrease very rapidly with the increase of temperature in the extreme low-temperature region (below 1 500 K). In the higher temperature region (above 1 500 K) one can notice only a slight increase with temperature in both coefficients. The main reason for this seems to be the thermal dissociation of F_2. The collisions with the ions are relatively insignificant in whole temperature region considered here, as the ionization processes become important at temperatures above 5 000 K.

The ratios $D_e(p_1) / D_e(p_2)$ and $D_i(p_1) / D_i(p_2)$ are about 10 for the both percentages of the fluorine in the plasma mixtures. The electron and ion diffusion coefficients in the argon plasmas with the 20% fluorine as additives are the 1,5 times higher than these quantities in the argon plasmas with the 30% fluorine. An inspection of the Tables discloses that the increase of the pressure raises the number densities of electron and ion constituents, the increase being obviously more noticeable for the neutrals with the pressures.

Table 1. Electron number density n_e , collision frequency ν_{eh}, diffusion coefficients D_e and D_i, electrical and thermal conductivities σ_e and λ_e in argon plasma with 20% fluorine on pressure p = 0,1 kPa

T, K	n_e, m^{-3}	ν_{eh}, s^{-1}	D_e, m^2/s	D_i, m^2/s	σ, $(\Omega\text{m})^{-1}$	λ, W/mK
500	$7,0 \times 10^{-11}$	$1,2 \times 10^{8}$	4.0×10^{0}	$1,2 \times 10^{-4}$	$1,6 \times 10^{-26}$	$1,5 \times 10^{-31}$
1000	$7,8 \times 10^{-11}$	$2,4 \times 10^{10}$	$6,1 \times 10^{-1}$	$1,7 \times 10^{-5}$	$9,2 \times 10^{-29}$	$1,8 \times 10^{-33}$
1500	$2,0 \times 10^{-2}$	$3,0 \times 10^{10}$	$7,4 \times 10^{-1}$	$2,2 \times 10^{-5}$	$1,9 \times 10^{-20}$	$5,3 \times 10^{-25}$
2000	$1,0 \times 10^{5}$	$3,3 \times 10^{10}$	$8,8 \times 10^{-1}$	$2,5 \times 10^{-5}$	$8,4 \times 10^{-14}$	$3,1 \times 10^{-18}$
2500	$1,1 \times 10^{9}$	$3,6 \times 10^{10}$	$1,1 \times 10^{0}$	$3,1 \times 10^{-6}$	$8,7 \times 10^{-10}$	$4,0 \times 10^{-14}$
3000	$5,7 \times 10^{11}$	$3,8 \times 10^{10}$	$1,2 \times 10^{0}$	$3,4 \times 10^{-5}$	$4,2 \times 10^{-7}$	$2,3 \times 10^{-11}$
3500	$4,9 \times 10^{13}$	$4,0 \times 10^{10}$	$1,2 \times 10^{0}$	$3,9 \times 10^{-5}$	$3,4 \times 10^{-5}$	$2,2 \times 10^{-9}$
4000	$1,4 \times 10^{15}$	$4,1 \times 10^{10}$	$1,3 \times 10^{0}$	$4,5 \times 10^{-5}$	$9,5 \times 10^{-4}$	$2,0 \times 10^{-8}$
4500	$2,0 \times 10^{16}$	$4,2 \times 10^{10}$	$1,7 \times 10^{0}$	$5,1 \times 10^{-5}$	$1,3 \times 10^{-2}$	$1,1 \times 10^{-6}$
5000	$1,6 \times 10^{17}$	$4,3 \times 10^{10}$	$1,9 \times 10^{0}$	$5,6 \times 10^{-5}$	$1,0 \times 10^{-1}$	$9,3 \times 10^{-6}$

Table 2. Same as Tab.1, but in argon plasma with 20% fluorine; p = 1,0 kPa

T, K	n_e, m^{-3}	ν_{eh}, s^{-1}	D_e, m^2/s	D_i, m^2/s	σ, $(\Omega\text{m})^{-1}$	λ, W/mK
500	$1,7 \times 10^{-10}$	$9,8 \times 10^{10}$	5.4×10^{-1}	$1,5 \times 10^{-5}$	$4,8 \times 10^{-29}$	$4,5 \times 10^{-34}$
1000	$2,5 \times 10^{-10}$	$2,0 \times 10^{11}$	$7,2 \times 10^{-2}$	$2,1 \times 10^{-6}$	$3,4 \times 10^{-29}$	$6,5 \times 10^{-34}$
1500	$6,2 \times 10^{-2}$	$2,5 \times 10^{11}$	$7,4 \times 10^{-2}$	$2,2 \times 10^{-6}$	$7,0 \times 10^{-21}$	$2,0 \times 10^{25}$
2000	$3,2 \times 10^{5}$	$3,3 \times 10^{11}$	$8,8 \times 10^{-2}$	$2,4 \times 10^{-6}$	$2,7 \times 10^{-14}$	$1,0 \times 10^{-18}$
2500	$3,5 \times 10^{9}$	$3,6 \times 10^{11}$	$1,1 \times 10^{-1}$	$3,1 \times 10^{-5}$	$3,9 \times 10^{-11}$	$1,8 \times 10^{-15}$
3000	$1,7 \times 10^{12}$	$3,8 \times 10^{11}$	$1,2 \times 10^{-1}$	$3,4 \times 10^{-6}$	$1,3 \times 10^{-7}$	$7,3 \times 10^{-12}$
3500	$1,6 \times 10^{14}$	$4,0 \times 10^{11}$	$1,4 \times 10^{-1}$	$3,9 \times 10^{-6}$	$1,1 \times 10^{-4}$	$7,1 \times 10^{-9}$
4000	$4,5 \times 10^{15}$	$4,1 \times 10^{11}$	$1,5 \times 10^{-1}$	$4,4 \times 10^{-6}$	$3,1 \times 10^{-4}$	$2,3 \times 10^{-8}$
4500	$6,3 \times 10^{16}$	$4,2 \times 10^{11}$	$1,7 \times 10^{-1}$	$5,1 \times 10^{-6}$	$4,2 \times 10^{-3}$	$3,5 \times 10^{-7}$
5000	$5,2 \times 10^{17}$	$4,3 \times 10^{11}$	$1,9 \times 10^{-1}$	$5,6 \times 10^{-6}$	$3,4 \times 10^{-2}$	$3,2 \times 10^{-6}$

The obtained values for n_e led us to conclude with certainty that the assumption on LTE was well met in both plasmas. For example, in the argon plasma, it is generally accepted [11] that LTE exists if $n_e > 5 \times 10^{21}$ m^{-3}, and this was found to be valid within the whole temperature range considered and for all the pressures taken. Although this requirement is not easy to satisfy in plasmas with heavy particles having very large masses; the Maxwellian $f_e^{00}(\upsilon)$was assumed correctly in (3), (4) and in (5).

Tables 3. and 4. show some of the more interesting results obtained from eq. (4), in combination with eqs. (1) and (2). They represent the temperature dependence, with pressure as parameter. To interpret the behaviour of the values shown, one should take into account that both n_h and $\upsilon Q_h(\upsilon)$ in eq. (4) depend on temperature. As an estimate, one might assume that $\upsilon Q_h(\upsilon)$ is proportional to $T^{1/2}$ in the case of neutral particles, and to $T^{-3/2}$ for ions; this corresponds, respectively, to the "billiard-ball" model for the former type of collisions, and to the well-known Spitzer-Harm formula for the latter.

Table 3. Same as Tab. 1, but in argon plasma with 30% fluorine; p = 0,1 kPa

T,K	n_e,m^{-3}	ν_{eh},s^{-1}	D_e, m^2/s	D_i, m^2/s	σ, (Ωm)$^{-1}$	λ,W/mK
500	$6,9 \times 10^{-11}$	$1,2 \times 10^{8}$	4.0×10^{0}	$1,2 \times 10^{-4}$	$1,6 \times 10^{-26}$	$1,5 \times 10^{-31}$
1000	$7,4 \times 10^{-11}$	$3,6 \times 10^{10}$	$3,9 \times 10^{-1}$	$1,2 \times 10^{-5}$	$5,9 \times 10^{-29}$	$1,1 \times 10^{-33}$
1500	$1,8 \times 10^{-2}$	$4,4 \times 10^{10}$	$4,9 \times 10^{-1}$	$1,3 \times 10^{-5}$	$1,1 \times 10^{-20}$	$3,1 \times 10^{-24}$
2000	$9,4 \times 10^{4}$	$5,1 \times 10^{10}$	$6,0 \times 10^{-1}$	$1,7 \times 10^{-5}$	$5,0 \times 10^{-14}$	$1,8 \times 10^{-18}$
2500	$1,0 \times 10^{9}$	$5,4 \times 10^{10}$	$7,3 \times 10^{-1}$	$2,1 \times 10^{-5}$	$5,0 \times 10^{-10}$	$2,3 \times 10^{-14}$
3000	$5,3 \times 10^{11}$	$5,7 \times 10^{10}$	$8,1 \times 10^{-1}$	$2,4 \times 10^{-5}$	$2,5 \times 10^{-7}$	$1,4 \times 10^{-11}$
3500	$4,6 \times 10^{13}$	$6,3 \times 10^{10}$	$9,3 \times 10^{-1}$	$2,7 \times 10^{-5}$	$2,0 \times 10^{-5}$	$1,3 \times 10^{-9}$
4000	$1,3 \times 10^{15}$	$6,1 \times 10^{10}$	$1,0 \times 10^{0}$	$3,1 \times 10^{-5}$	$5,9 \times 10^{-4}$	$4,4 \times 10^{-8}$
4500	$1,8 \times 10^{16}$	$6,3 \times 10^{10}$	$1,2 \times 10^{0}$	$3,5 \times 10^{-5}$	$8,1 \times 10^{-3}$	$6,8 \times 10^{-7}$
5000	$1,5 \times 10^{17}$	$5,9 \times 10^{10}$	$1,3 \times 10^{0}$	$3,8 \times 10^{-5}$	$7,3 \times 10^{-2}$	$6,8 \times 10^{-6}$

Table 4. Same as Tab. 1, but in argon plasma with 30 % fluorine; p = 1,0 kPa

T,K	n_e,m^{-3}	ν_{eh},s^{-1}	D_e, m^2/s	D_i, m^2/s	σ, (Ωm)$^{-1}$	λ,W/mK
500	$1,6 \times 10^{-10}$	$9,3 \times 10^{10}$	5.6×10^{-1}	$1,6 \times 10^{-5}$	$4,8 \times 10^{-29}$	$4,5 \times 10^{-34}$
1000	2.3×10^{-10}	$3,2 \times 10^{11}$	$5,0 \times 10^{-2}$	$1,5 \times 10^{-6}$	$2,0 \times 10^{-30}$	$3,8 \times 10^{-35}$
1500	$5,8 \times 10^{-2}$	$4,4 \times 10^{11}$	$4,9 \times 10^{-2}$	$1,4 \times 10^{-6}$	$3,6 \times 10^{-21}$	$1,0 \times 10^{-26}$
2000	$3,0 \times 10^{5}$	$5,3 \times 10^{11}$	$5,9 \times 10^{-2}$	$1,7 \times 10^{-6}$	$1,6 \times 10^{-14}$	5.9×10^{-19}
2500	$3,3 \times 10^{9}$	$5,4 \times 10^{11}$	$7,3 \times 10^{-2}$	$2,1 \times 10^{-6}$	$7,8 \times 10^{-10}$	$3,6 \times 10^{-14}$
3000	$1,7 \times 10^{12}$	$5,4 \times 10^{11}$	$8,1 \times 10^{-2}$	$2,4 \times 10^{-6}$	$8,7 \times 10^{-8}$	$4,9 \times 10^{-19}$
3500	$1,5 \times 10^{14}$	$5,3 \times 10^{11}$	$9,3 \times 10^{-2}$	$2,7 \times 10^{-6}$	$7,8 \times 10^{-6}$	$5,1 \times 10^{-10}$
4000	$4,2 \times 10^{15}$	$5,5 \times 10^{11}$	$1,0 \times 10^{-1}$	$3,1 \times 10^{-6}$	$2,1 \times 10^{-4}$	$1,5 \times 10^{-8}$
4500	$5,9 \times 10^{16}$	$6,3 \times 10^{11}$	$1,2 \times 10^{-1}$	$3,5 \times 10^{-6}$	$2,6 \times 10^{-3}$	$2,2 \times 10^{-7}$
5000	$4,8 \times 10^{17}$	$5,9 \times 10^{11}$	$1,3 \times 10^{-1}$	$3,8 \times 10^{-6}$	$2,3 \times 10^{-2}$	$2,1 \times 10^{-6}$

The electrical and thermal conductivities are seen to increase with temperature, which is an easily comprehensible feature ascribable mainly to the increasing thermal ionization.

4. CONCLUSION

From all that was said above, it can be concluded that in the low-pressure and low-temperature thermal argon plasmas with fluorine as additive, the relations between the ionization energies of the atomic species present (neutral atoms, singly charged ions and molecules) is by far the most significant physical factor in determining the plasma composition and its transport properties. All the above results indicate that the added fluorine is indeed quite important in determining all transport coefficients. The above outlined specificities in the transport coefficients behaviour are mainly due to the multiplier n_e in eq. (3). In the physical situation considered presently, the electron number density is entirely determined by the processes of thermal ionization. Special attention should be given to the fact that the suggested revision of the plasma Debye radius broadens the scope of applicability of the results derived for the weakly non-ideal plasma, and enables the more accurate evaluation of local plasma parameters in a multitude of ionized systems. The problem of the weak plasma non-ideality is satisfactorily circumvented by adopting the Landau length instead of the smallest ionic radii as the cut-off length.

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NEKE TRANSPORTNE OSOBINE U PLAZMI ARGONA I FLUORA

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Dati su neki rezultati numeričkog izračunavanja transportnih koeficijenata u plazmi smeše argona i fluora. Ove transportne karakteristike su date u funkciji temperature za niske pritiske (od 0,1 kPa do 1,0 kPa) i za niske temperature (od 500 K do 5 000 K) u argonovoj plazmi sa dodatkom 20 % i 30% fluora. Uzeto je da je sistem na konstantnom pritisku u stanju lokalne termodinamičke ravnoteže. Korišćen je modifikovani izraz za Debye-ev radijus, što je omogućilo da posmatrana plazma bude tretirana kao slabo neidealna u celom temperaturnom intervalu. Uzeta je Landau-ova dužina u izrazu za modifikovan Debye-ev radijus umesto manjeg jonskog radijusa. Ovakav izbor pri izračunavanju daje različite promene u konačne numeričke rezultate.