

GENERAL CHARACTERIZATION AND IONIZATION-RECOMBINATION PROPERTIES OF ELECTRONEGATIVE ELEMENTS PRESENT IN PLASMA DISCHARGES AT PLASMA CURRENT SWITCH OPERATION. PART I

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Based on literary sources, we present here the data on electron affinity values for a variety of negative atomic ions, and also, for some molecules and radicals being of interest for current experimental studies. Besides, the ionization potentials are given for nearly all ionization states of some electronegative elements (C, O, F, Cl). For the said elements, the ionization-recombination parameters and the dependences of the fractional values (ionic fraction) for ionization states and on the electron temperature within the framework of ionization equilibrium are given. A comparison is carried out between the calculated and experimental distributions of the fractions of multicharged C^{+4} ions versus electron temperature.

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INTRODUCTION

In the earlier papers [1 - 7] concerned with the problems of creation and research of a compact pulsed electron accelerator based on the plasma-filled diode, it has been indicated that, apart from the natural-origin elements, the spectrum of residual atmosphere inside the vacuum chamber may show up the elements of the electronegative Mendeleev's group. These elements may appear as a result of sputtering of key parts and details of the plasma guns involved in the experiment, and also, due to the external and internal gas puffing into the system for providing optimum conditions and results of filling the discharge gap of the plasma current switch (PCS) with plasma. The necessity of achieving this gas puffing effect was specified as early as at the moment of setting the problem, viz., creation of high-current electron accelerators (HCEA) and realization of first experiments. For example, it was suggested in ref. [1] that the parameters of the PCS, and eventually of HCEA, could be stabilized through keeping constant the charge-mass characteristic of the plasma bunch and the total number of particles contained in it. Further on [2 - 7], studies were made into the behavior of the switched current versus the plasma gun voltage, and the energy content of gun power, the residual gas density in the PCS chamber. It has been demonstrated that the stability increase in the PCS (and hence, the HCEA) operating parameters can be attained by increasing the amount and density of plasma in the discharge gap, and also, by making the plasma density distribution along the annular gap more uniform. At that, the switching current value can be increased either by enhancing the plasma injection into the discharge gap, or by applying additional gas injection, external or internal. Gases with low atomic number (H_2 , D_2 , He) are best suited to the external gas injection. The use of planar-type plasma guns with soft-dielectric surface breakdown, and also, the cable-type guns [8] may cause plasma contamination with atoms and ions of electronegative elements, e.g., fluorine, carbon and their compounds. In this connection, it appears of interest and advantage to trace their

effect on the PCS plasma, including the processes of ionization, recombination, and emission from plasma.

1. GENERAL DESCRIPTION OF ELECTRONEGATIVE ELEMENTS

The choice of electronegative elements C, O, F, Cl for consideration has been prompted in our case by experimental conditions, at which by reason of occurrence of some physical processes (desorption, decomposition, sputtering, dissociation, ionization, recombination) in the active vacuum volume there may appear (in controlled/uncontrolled way) and accumulate some amounts of the mentioned elements or their chemical species in both neutral (atoms, molecules) or ionized (positive and negative ions of different multiplicity) states. So, it stands to mention some of their general characteristics and properties.

The electron affinity [9], i.e., the energy released at production of negative ion from the neutral atom and electron ($A+e=A^-$), together with the ionization potential [10], are the important quantitative characteristics of the electronegativity of elements, which plays a large role in the molecular structure. In fact, the negative ion concept implies the atom, the molecule or the molecular group with a negative total charge. Free negative ions generally carry only unit excess negative charge. The negative ions in electrolytes and ionic crystals are most often multicharger. Directly in gas, the negative ions will be produced either as a result of dissociation of neutral molecules and molecular ions, or due to the transfer of one or a few electrons at heavy particle collisions during charge exchange. The additional electron is easiest to be captured by the atoms, which have single vacancies in outer shells, and therefore, the halogens (F, Cl, Br, I, At) should be strongly electronegative. The atoms with fully filled shells are poorly suited for negative ion production. Therefore, the inert gases are incapable of producing stable negative ions. For the negative ion to be stable, its binding energy should exceed the neutral atom-binding energy. Mathematically, it can be written as

$$\left. \begin{aligned} E_1 + \sum_{i=1}^z E_i^- &> \sum_{i=1}^z E_i^0 \\ \text{or} \\ E_1 - \sum_{i=1}^z (E_i^0 - E_i^-) &> 0 \end{aligned} \right\}, \quad (1)$$

where E_i^0 is the binding energy of the i -th atomic electron prior to additional electron attachment, E_i^- is the binding energy of the same atomic electron after the attachment, E_1 is the binding energy of the additional electron after its attachment.

The left-hand side of inequality (1) is called the electron affinity of the atom. It is represented as the total energy difference of the ground states of the atom and

the ion, and is denoted as EA. In principle, EA is equal to the energy required for the most weakly attached electron to separate from the ion (detachment energy for ion). Table 1 lists the electron affinity values, which were obtained experimentally, theoretically and empirically by extrapolation for a number of negative atomic ions.

Table 2 gives the electron affinity values for a variety of molecules and radicals. The values were determined as the energy difference between the neutral molecule and the corresponding molecular ion.

Table 1

Electron affinity values for some negative atomic ions

Ion	H ⁻	B ⁻	C ⁻	N ⁻	O ⁻	F ⁻	Al ⁻	Si ⁻	Cl ⁻	Br ⁻	I ⁻
EA, eV	0.754	0.33	1.33	0.54	1.465	3.62	0.52	1.46	3.76	3.51	3.17

Table 2

Electron affinity values for a variety of molecules and radicals

Ion	H ₂ ⁻	O ₂ ⁻	OH ⁻	N ₂ ⁻	NO ₂ ⁻	NO ₃ ⁻	CH ⁻	CN ⁻	C ₂ ⁻	C ₃ ⁻	SF ₅ ⁻
EA, eV	0.9	0.44	1.78	<0	1.6	3.9	~1.6	3.1	~3.2	2.5	3.39

The above-given electron affinity data encourage us to point out the following.

1. The positive electron affinity points to the negative ion stability.

2. The experimental and theoretical data are considered reliable to the accuracy of 0.1 eV, except for Na⁻ and P⁻.

3. Determination of electron affinity for the molecules is a much more difficult problem than for atoms, because it calls for carrying out spectroscopic studies (measurements) with improved spectral resolution.

4. The affinity for two and more electrons, i.e., the energy of doubly- or multi-charged negative ion formation from the atom and two or more electrons, is always negative. For example, the formation of the O²⁻ ion from O+2e is supposed to take about 7 eV, whereas the formation of the S²⁻ ion from S+2e takes about 3.5 eV. In

the general case, the electric (nuclear) system, consisting of the nucleus and (z+k) electrons, i.e., in the form of the negative ion A^{-k}, at k>1, is always unstable and must break up with energy release.

5. Doubly charged negative ions were never observed in gases. The reason is that the second electron experiences a strong electrostatic repulsion. Therefore, the probability of the second electron attachment is negligibly small.

Among the parameters of certain elements, called as electron affinity, another parameter of importance that characterizes the special their features and electronegativity of the elements is the potential of ionization over all ionization states. Table 3 lists this parameter values [11] for four elements (C, O, F, Cl) being the subject of the present study.

Table 3

Ionization potentials for C, O, F, Cl

Carbon		Oxygen		Fluorine		Chlorine	
i	u _i , eV	i	u _i , eV	i	u _i , eV	i	u _i , eV
1	11.264	1	13.614	1	17.418	1	13.01
2	24.376	2	35.146	2	34.980	2	23.80
3	47.864	3	54.934	3	62.646	3	39.90
4	64.476	4	77.394	4	87.23	4	53.3
5	391.986	5	113.873	5	114.214	5	67.8±0.2
6	489.84	6	138.08	6	157.117	6	96.6±0.3
	-	7	739.114	7	185.139	7	114.2±0.1
	-	8	871.12	8	935.8±0.3	8	348.5
	-	-	-	9	1101.8	9	400.3±0.1
	-	-	-	-	-	10	455.3
	-	-	-	-	-	11	531.4
	-	-	-	-	-	12...17	-

2. IONIZATION-RECOMBINATION PROPERTIES AND PARAMETERS OF ELECTRONEGATIVE ELEMENTS

The problem of ionization (coronal) equilibrium belongs to the domain of recombination-ionization equilibrium and the charged-state equilibrium of the ions under consideration [12]. The time-varying spectral line intensity of highly ionized ions should be treated in terms of the rate of ionization and recombination [13]. The stationary populations of ions having different charges depend not only on the physics of atomic processes, but also on the ion motion in the plasma. At a low diffusion rate, the spatial profiles of ions will correspond to the coronal equilibrium, while at a quick ion motion through the plasma they will correspond to the non-coronal ionization.

We now mention some collective aspects of the processes of ionization and recombination. The set of conditions and parameters necessary for formulating the equation of balance in the many-particle system (i^+ , i , e^-) includes the following.

1. The most probable collisions are elastic collisions, which establish the Maxwellian distribution function.
2. The main collision types are: $e^- i^+$ (or $e^- p^+$, i.e., electrons and protons) + charge-exchange process.
3. Since $v_e > v_p$ (v_i), they have a greater impact on the ionization/excitation processes.
4. Pair collisions are more important than many-particle collisions. The three-body recombination is also of importance.

5. If the processes of photoionization and photoexcitation are neglected, then the optically thin plasma approximation is used.

The equilibrium equation includes the terms of two types: radiation (radiation-recombination) and collisional terms. Consideration is given to three ionic groups: 1) excited-state ions, but not metastable; 2) ground-state ions; 3) metastable-state ions. The equation describing the rate of ion density variation, $n(z)$, with the charge z has the form

$$\frac{dn(z)}{dt} = n_e [n(z-1)S(z-1, z) - n(z)\alpha(z, z-1) - n(z)S(z, z+1) + n(z+1)\alpha(z+1, z)], \quad (2)$$

where $S(z, z+1)$ is the effective ionization coefficient, $\alpha(z, z-1)$ is the effective recombination coefficient.

Equation (2) takes no account of the multiple ionization. Furthermore, the multiple ionization is regarded as unlikely, and the contribution of metastable levels is neglected.

The basic plasma parameter, which affects the time of achieving the ionization equilibrium steady-state, when ionization is balanced by recombination, is the plasma density. At the same time, the ionic charge state, attained through realization of ionization equilibrium, is determined by the electron temperature. Considering that at the ionization equilibrium state the rates of ionization and recombination are much the same, the time of steady-state equilibrium achievement can be estimated by the formula $t_{\text{ion. eq}} = 10^{12} \cdot n_e^{-1}$, s. For comparison, Table 4 gives the relaxation time values for some levels of hydrogen-like atoms.

Table 4

Relaxation times for some levels of hydrogen-like atoms

Θ		n_e	τ , s	τ , s	τ , s	τ , s
K	eV	cm^{-3}	p=1	p=2	p=3	p=4
$2.56 \cdot 10^5$	22.07	$1.0 \cdot 10^8$	$1.3 \cdot 10^{-1}$	$2.1 \cdot 10^{-9}$	$1.0 \cdot 10^{-8}$	$5.5 \cdot 10^{-6}$
		$1.0 \cdot 10^{12}$	$1.3 \cdot 10^{-5}$	$2.1 \cdot 10^{-9}$	$9.8 \cdot 10^{-9}$	$9.8 \cdot 10^{-10}$
		$1.0 \cdot 10^{15}$	$1.3 \cdot 10^{-8}$	$9.3 \cdot 10^{-10}$	$4.1 \cdot 10^{-10}$	$9.8 \cdot 10^{-13}$
		$1.0 \cdot 10^{18}$	$1.3 \cdot 10^{-11}$	$1.7 \cdot 10^{-12}$	$4.3 \cdot 10^{-13}$	$9.8 \cdot 10^{-16}$

Here p is the principal quantum number according to the Bohr-Sommerfeld theory, equal to the sum of two (azimuthal n_θ and radial n_r) quantum numbers. The $p=1, 2, 3, 4$ values correspond to the states K, L, M, and N; Θ and n_e are, respectively, the reduced temperature and density of electrons. The relaxation time values are inversely related to z^4 . At that, the time of steady-state ionization equilibrium achievement must be considerably shorter than the required energy confinement time, i.e., the relationship $t_{\text{ion. eq}} \text{ should be compared with the Lawson criterion } nT_E > 10^{14} \text{ cm}^3 \cdot \text{s}.$

In a number of works [14 - 21], within the ionization equilibrium approximation for different ionization states, the ionic fractions were determined as functions of electron temperature for different elements. For example, the data are reported for H in [14], for C in [15], for N, O, Xe in [16], for Ar and Ti in [17], for Kr in [18] and for multicomponent gas-metal (Ar+Ti) plasma of a pulsed reflex discharge in [19]. The authors of free-standing works [20, 21] give the data for the element

groups comprised of 11 and 28 names respectively. For tasks set in the given case, the work [21] is the most acceptable. Based on the above-given literary sources [14 - 21], we have obtained the following information data for certain electronegative elements (such as carbon, oxygen, fluorine, chlorine), in the form of dependences $\chi_i = f(T_e)$ of fractional values (ionic fractions), where $\chi_i = -\log(N_i/N_{\text{total}})$, on the electron temperature within the ionization equilibria (Figs. 1-5). Table 5 gives distribution of investigated energy ranges by ionization states for different electronegative elements shown in Figs. 1 to 5.

Note that the left-hand branch of the $\chi_i = f(T_e)$ curves at $i > 1$ relates to the recombination part of the ionization equilibrium process, while the right-hand part at $i > 1$ – to the ionization part. Figs. 1-5 show the temperature dependences of ionic fractions $\chi_i = f(T_e)$ for carbon (see Fig. 1) over the ionization states $i=I-VI$, for oxygen (see Fig. 2) over $i=I-VIII$, for fluorine (see Fig. 3) over $i=I-IX$, for chlorine over $i=I-IX$ (see Fig. 4) and over $i=X-$

XVII (see Fig. 5). Fig. 2,a,b illustrates the work data of two corporate authors [20, 21] for oxygen. The difference between their results varies within 15 to 40%.

Table 5

Distribution of the studied energy ranges by ionization states for various electronegative elements

Ionization state		i=1	2	3	4	5	6	7	8	9	10	11
Elements and Energy ranges $\Delta\varepsilon$	C	0.86±	0.86±	2.41±	5.52±	7.07±	39.7±					
	$\Delta\varepsilon$, eV	16.38	31.90	55.17	319	862	862					
	O	0.86±	0.86±	3.97±	6.29±	7.84±	16.38±	31.90±	70.68±			
	$\Delta\varepsilon$, eV	16.38	39.7	47.41	62.93	163.79	551.72	3189.7	6293			
	F	0.86±	0.86±	3.96±	6.29±	7.84±	16.38±	31.89±	39.65±	78.44±		
	$\Delta\varepsilon$, eV	8.62	31.89	55.17	70.68	78.44	318.96	706.8	4741.4	8620.7		
Cl	0.86±	0.86±	2.41±	3.96±	6.29±	7.84±	16.38±	24.14±	47.71±	62.93±	78.44±	
$\Delta\varepsilon$, eV	6.29	24.14	47.41	55.17	62.93	78.44	163.79	396.55	474.1	551.7	629.3	
i		12	13	14	15	16	17					
Cl	86.2±	163.8±	241.4±	319±	319±	551.7±						
$\Delta\varepsilon$, eV	784.5	1637.9	4741	7844	47414	47414						

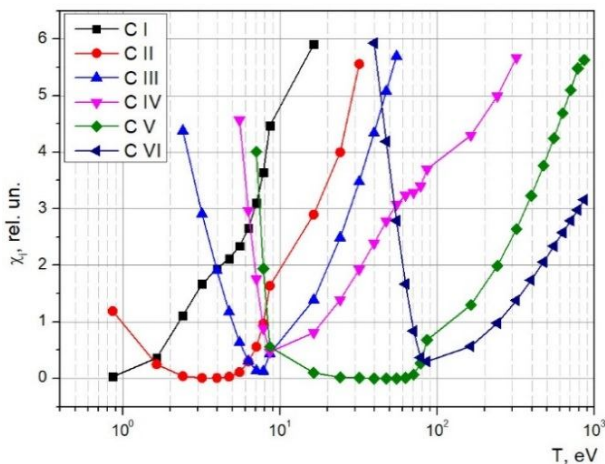


Fig. 1. Carbon ionic fractions versus electron temperature for ionization states ranging from I to VI, according to J.M. Shull and M. van Steenberg [20]

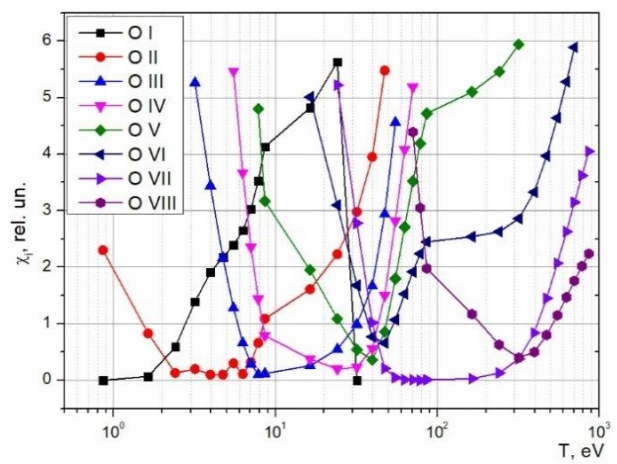


Fig. 2b. Oxygen ionic fractions versus electron temperature for ionization states ranging from I to VIII, according to J.M. Shull and M. van Steenberg [20]

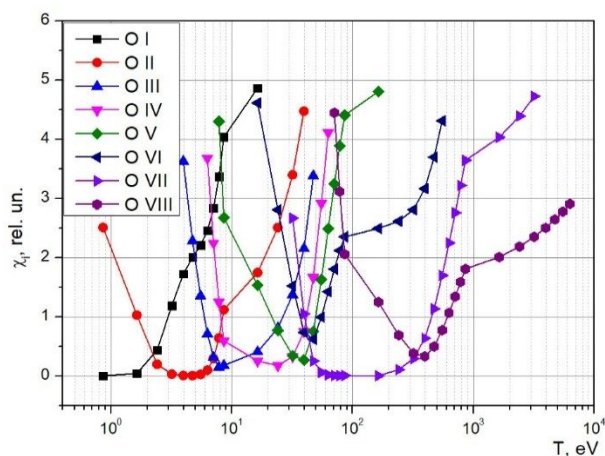


Fig. 2a. Oxygen ionic fractions versus electron temperature for ionization states ranging from I to VIII, according to P. Mazzotta et al [21]

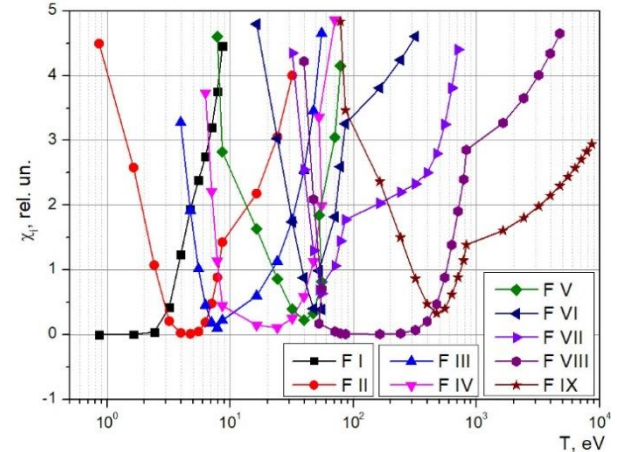


Fig. 3. Fluorine ionic fractions versus electron temperature for ionization states ranging from I to IX, according to P. Mazzotta et al [21]

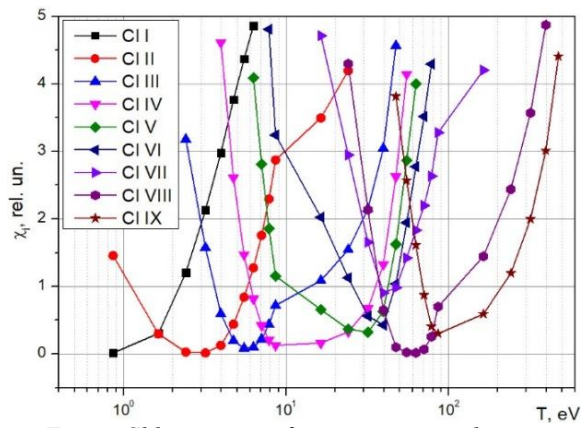


Fig. 4. Chlorine ionic fractions versus electron temperature for ionization states ranging from I to IX, according to P. Mazzotta et al [21]

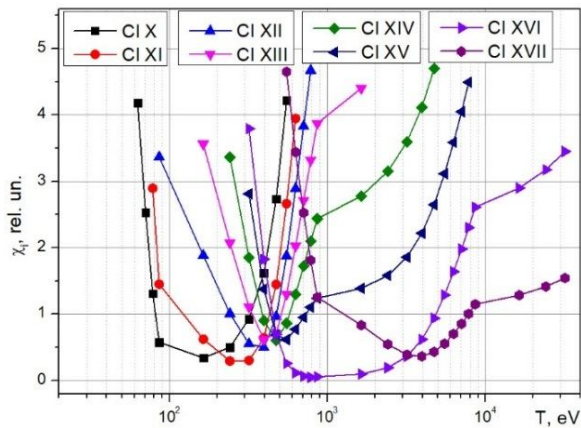


Fig. 5. Chlorine ionic fractions versus electron temperature for ionization states ranging from X to XVII, according to P. Mazzotta et al [21]

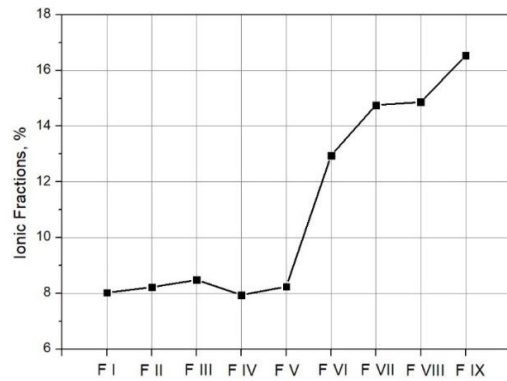


Fig. 6. Fluorine ionic fractions distribution in different ionization states in the temperature range from 10^4 to 10^9 K (0.862...86207 eV)

The distributions of fluorine/chlorine ionic fractions over different ionization states were also obtained in the temperature range $10^4 \dots 10^9$ K ($\sim 1 \dots 10^4$ eV) (Figs. 6-8). In this case, with the use of data from ref. [20] and Fig. 1, calculations were performed to determine the fraction distributions of the multicharged ions C^{+4} in the energy range from 5.52 to 55.17 eV. The resulting relationship $N_{i=4}/N_{total}=f(T)$ (Fig. 9,a) has made it possible to determine (from the half width of the curve in the region of $Ni/N_t > 1\%$ and at ΔT from 23 down to 13 eV) the average C^{+4} fraction value which is $\sim 15\%$, even though the maximum fraction value in the narrow energy range (≤ 8 eV) may reach $\sim 70\%$.

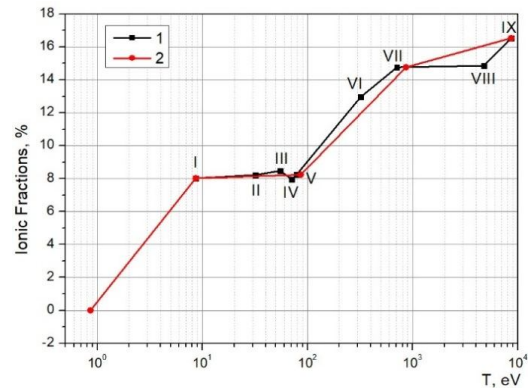


Fig. 7. Fluorine ionic fractions distribution in different ionization states versus electron temperature: 1 – the black line is plotted by the computational points; 2 – the red line is plotted by the averaged temperature ranges

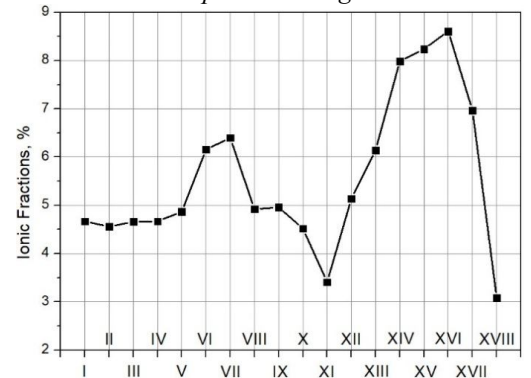


Fig. 8. Chlorine ionic fractions distribution in different ionization states at electron temperatures ranging from 10^4 to 10^9 K

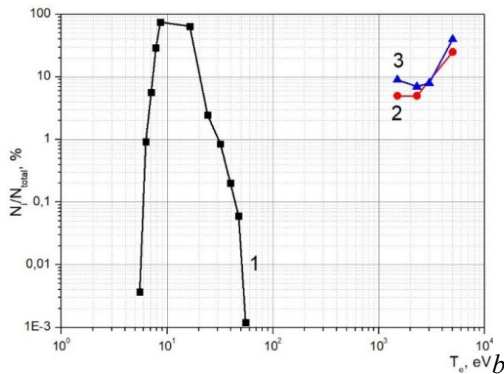
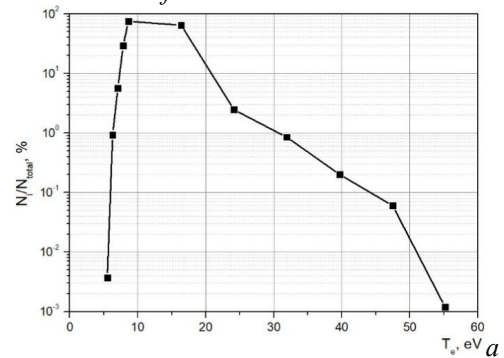


Fig. 9. Temperature dependence of the relative amount of multicharged C^{+4} ions, calculated with regard to the data of Fig. 1,a; Comparison between the carbon multicharged ionic fractions as functions of temperature, calculated with regard to the data of [20] and Fig. 1, and determined by experiment [22] (b)

The studies [22] into the special features of the energy-mass composition of the plasma of the nanosecond coaxial vacuum surface discharge have revealed the presence of differently charged carbon ions (C^{+1} , C^{+2} , C^{+3} , C^{+4}). A correlation was made between the experimental fractions of the multicharged C^{+4} ions with the computational estimates. The results of the correlation are presented in Fig. 9,b. On the left, curve 1 shows the calculated C^{+4} ionic fraction distribution obtained with the use of the data from ref. [20] under the assumption of the ionization equilibrium. At the right top of Fig. 9,b the curves show the experimental results of work [22] at different polarities of the electrode-dielectric (high pressure polyethylene) pair (+ : curve 2, - : curve 3).

RESULTS AND CONCLUSIONS

The conducted investigations are mainly aimed at evaluating the impact of some factors, which cause the plasma-bridge formation and decay in the PCS electrode gap, on the efficiency of the PCS operation in attaining the maximum current values and voltage multiplication. Among these factors is the electronegativity of certain elements that enter into the weight composition of plasma in the form of ionized and neutral particles.

The main results of the present work can be summarized as follows.

1. General characterization has been given for several electronegative elements (C, O, F, Cl) that present some features of interest for the current studies. The data include the electron affinity values and the values of ionization potentials in different ionization states.

2. With due regard to the ionization-recombination properties and the parameters of the electronegative elements under consideration, studies have been made to determine the distributions of fluorine and chlorine ionic fractions in different ionization states in the context of ionization equilibrium. At that, the electron temperature was varied in the range from 1 to 10^4 eV.

3. Comparison has been made between the calculated and experimental data on the distribution of multicharged C^{+4} ionic fractions as a function of electron temperature.

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**ОБЩАЯ ХАРАКТЕРИСТИКА И ИОНИЗАЦИОННО-РЕКОМБИНАЦИОННЫЕ СВОЙСТВА
ЭЛЕКТРООТРИЦАТЕЛЬНЫХ ЭЛЕМЕНТОВ, ПРИСУТСТВУЮЩИХ В ПЛАЗМЕННЫХ
РАЗРЯДАХ ПРИ РАБОТЕ ПЛАЗМЕННЫХ КОММУТАТОРОВ ТОКА. ЧАСТЬ I**

Е.И. Скибенко, В.Б. Юферов, А.Н. Озеров, И.В. Буравілов

На основе литературных источников приведены данные по величинам сродства к электрону для ряда отрицательных атомарных ионов и для некоторых молекул и радикалов, представляющих интерес для проводимых экспериментальных исследований. Приведены также потенциалы ионизации почти по всем ионизационным состояниям для некоторых электроотрицательных элементов (С, О, F, Cl). Для них же приведены ионизационно-рекомбинационные параметры и зависимости фракционных значений (ionic fraction) по ионизационным состояниям от температуры электронов в рамках ионизационного равновесия (ionization equilibria). Проведено сравнение расчетных и экспериментальных данных по распределениям доли многозарядных ионов C^{+4} от температуры электронов.

**ЗАГАЛЬНА ХАРАКТЕРИСТИКА ТА ІОНІЗАЦІЙНО-РЕКОМБІНАЦІЙНІ ВЛАСТИВОСТІ
ЕЛЕКТРОНЕГАТИВНИХ ЕЛЕМЕНТІВ, ПРИСУТНІХ У ПЛАЗМОВОМУ РОЗРЯДІ ПРИ РОБОТІ
ПЛАЗМОВИХ КОМУТАТОРІВ СТРУМУ. ЧАСТИНА I**

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На основі літературних джерел наведені дані по величинам спорідненості до електрону для ряду негативних атомарних іонів і для деяких молекул і радикалів, що представляють інтерес для експериментальних досліджень, що проводяться. Наведено також потенціали іонізації майже за всіма іонізаційними станами для деяких електронегативних елементів (С, О, F, Cl). Для них же наведені іонізаційно-рекомбінаційні параметри і залежності фракційних значень (ionic fraction) по іонізаційним станам від температури електронів у рамках іонізаційної рівноваги (ionization equilibria). Проведено порівняння розрахункових і експериментальних даних за розподілом частки багатозарядних іонів C^{+4} від температури електронів.