Particle composition of high-pressure SF/sub6/ plasma with electron temperature greater than gas temperature

Particle Composition of High-Pressure SF_6 Plasma with Electron Temperature Greater than Gas Temperature

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***Abstract*—Numerical computation was performed to derive particle compositions of high-pressure SF_6 plasmas in which electron temperature T_e is greater than gas temperature T_g. The effective excitation temperature T^{ex} for each of particles was proposed to express the distribution of particles in excited states. Furthermore, the relation equations for chemical equilibrium in two-temperature plasma were developed on the basis of the second law of thermodynamics. At fixed T_g in the range of 6000–9000 K, the increasing T_e from 9000–20 000 K encourages the dissociation and ionization of particles, causing the composition of SF_6 plasma at 0.1 MPa to differ markedly from the composition under the condition of thermal equilibrium.**

***Index Terms*—Excitation temperature, high-pressure SF_6 plasma, two-temperature state.**

I. Introduction

In an SF_6 gas-blast circuit breaker, detailed investigations on a post-arc channel with temperatures below 6000 K after current zero are essential for a proper understanding of current interruption process. The post-arc channel is exposed to high electric field, because a transient recovery voltage (TRV) is applied to the circuit breaker during the time after current zero. The high electric field elevates the kinetic energy of electrons in the post-arc channel and then increases the electron temperature T_e. As a result, T_e become higher than gas temperature T_g in the post-arc channel [1]. This may change the particle composition of the post-arc channel from that under the condition of thermal equilibrium. It is therefore very important to investigate the particle composition of the post-arc channel in two-temperature states where T_e is greater than T_g.

Several papers have so far dealt with the particle compositions of Ar, H_2, or N_2 plasma in two-temperature state [2]. In calculating the compositions in some of the papers, multitemperature Saha's equation (MSE) was adopted to express chemical equilibrium in two-temperature state. However, recent work pointed out that MSE is invalid since it is not satisfied with the second law of thermodynamics [3].

The present paper describes calculation results for the particle composition of SF_6 plasmas in two-temperature state at a pressure of 0.1 MPa. First, in order to express the populations of the internal energy states of heavy particles, effective excitation temperature was proposed. Second, using the effective excitation temperature, relation equations for ionization and dissociation equilibriums in two-temperature state were induced on the basis of the second law of thermodynamics. Finally, particle compositions of SF_6 plasma in two-temperature state were theoretically calculated at a fixed T_g above 3000 K as a function of T_e.

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II. Equations for Calculating Particle Composition of SF_6 Plasma in Two-Temperature State

In the present calculation, it is assumed that SF_6 plasmas are in the following conditions. 1) The plasma is homogeneous. Although it is possible to expect the disturbance of homogeneity in the SF_6 decomposition products in pratical situation, we deal with homogeneous SF_6 plasmas as the fundamental approach in this paper. 2) Total pressure is 0.1 MPa. In calculating the composition, we took account of 23 species: SF_6, SF_5, SF_4, SF_3, SF_2, SF, SSF_2, FSSF, F_2, S_2, F, S, SF^-, F^-, S^-, F_2^+, S_2^+, F^{2+}, S^{2+}, F^+, S^+, SF^+, and electrons. 3) Electrons and heavy particles have Maxwellian energy distribution functions. 4) Temperatures of all the heavy particles are identical to gas temperature T_g. 5) Electron temperature T_e is greater than T_g. 6) The populations of the internal energy states of the heavy particles follow Boltzmann's law. 7) Chemical equilibrium between the particles is established.

A. Effective Excitation Temperature of Heavy Particle

Let us discuss the occupations of the internal energy states of heavy particles such as atoms, molecules, and ions. This is because the internal energy states of the heavy particles have great influence on chemical reactions such as ionization and dissociation. On the basis of Boltzmann's law, the population of the internal energy states of the heavy particle "A" is given by

$$\frac{n_A^{(u)}}{n_A^{(l)}} = \frac{g_u}{g_l} \exp\left(-\frac{\epsilon_u - \epsilon_l}{kT_A^{ex}}\right) \tag{1}$$

where the scripts u and l denote the excitation levels, g is statistical weight, ϵ is excitation potential, and k is Boltzmann constant. The quantity T_A^{ex} is the so-called excitation temperature of the particle "A." The excitation temperature is considered to be close to T_{e} in low-pressure and low-temperature plasmas, where the excitation of the heavy particles results mainly from collisions with electrons. However, in high-pressure and high-T_{g} plasmas, the excitation of the heavy particle highly seems to result from collisions not only with electrons but also with heavy particles. In such plasmas, $T_{\mathrm{A}}^{\mathrm{ex}}$ depends on both of T_{e} and T_{g}. We will therefore express $T_{\mathrm{A}}^{\mathrm{ex}}$ in terms of T_{e} and T_{g}.

In the excitation process, a heavy particle "A" obtains energy of $\frac{3}{2}kT_{\mathrm{e}}K_{\mathrm{eA}}\nu_{\mathrm{eA}}$ a second through collisions with electrons, where K_{eA} and ν_{eA} are the collision loss factor and the collision frequency between an electron and the particle "A," respectively. In a similar manner, the particle "A" also acquires energy of $\Sigma_{j(j\neq \mathrm{e})}\frac{3}{2}kT_{\mathrm{g}}K_{j\mathrm{A}}\nu_{j\mathrm{A}}$ a second through collisions with various kinds of heavy particles, where $K_{j\mathrm{A}}$ and $\nu_{j\mathrm{A}}$ are the collision loss factor and the collision frequency between j and "A," respectively. As a result, total energy $E_{\mathrm{A}}^{\mathrm{rec}}$ that the particle "A" obtains is given by

$$E_{\mathrm{A}}^{\mathrm{rec}} = \tfrac{3}{2}kT_{\mathrm{e}}K_{\mathrm{eA}}\nu_{\mathrm{eA}} + \tfrac{3}{2}kT_{\mathrm{g}}\sum_{j(j\neq \mathrm{e})}K_{j\mathrm{A}}\nu_{j\mathrm{A}}. \tag{2}$$

This total energy $E_{\mathrm{A}}^{\mathrm{rec}}$ determines $T_{\mathrm{A}}^{\mathrm{ex}}$ of the particle "A."

Without distinction of electron and heavy particles colliding with "A," we regarded them as one kind of fictitious particle. Assume also that the fictitious particle has kinetic energy $\frac{3}{2}kT_{\mathrm{A}}^{\mathrm{ex}}$ and collides with "A" with a frequency of $\nu_{\mathrm{eA}} + \Sigma_{j(j\neq \mathrm{e})}\nu_{j\mathrm{A}}$. The assumptions enable us to set up the following equation:

$$\tfrac{3}{2}kT_{\mathrm{A}}^{\mathrm{ex}}K\left(\nu_{\mathrm{eA}} + \sum_{j(j\neq \mathrm{e})}\nu_{j\mathrm{A}}\right) = E_{\mathrm{A}}^{\mathrm{rec}} \tag{3}$$

where K is the collision loss factor between fictitious particle and "A." The collision loss factors usually depend on the temperatures and species of interest. However, there have been few experimental data available on the factors so far. Then, for the sake of simplicity, we assumed that $K_{\mathrm{eA}}, K_{j\mathrm{A}}$, and K are the same, although this assumption is rough and results in underestimation of the effect of collision with electrons. Substituting (2) into (3) and rewriting (3) yields

$$T_{\mathrm{A}}^{\mathrm{ex}} = \frac{\nu_{\mathrm{eA}}T_{\mathrm{e}} + \sum_{j(j\neq \mathrm{e})}\nu_{j\mathrm{A}}T_{\mathrm{g}}}{\nu_{\mathrm{eA}} + \sum_{j(j\neq \mathrm{e})}\nu_{j\mathrm{A}}}. \tag{4}$$

This equation expressing $T_{\mathrm{A}}^{\mathrm{ex}}$ in terms of T_{e} and T_{g} is newly proposed in the present paper. We named $T_{\mathrm{A}}^{\mathrm{ex}}$ as "effective excitation temperature." The effective excitation temperature greatly affects chemical reactions, as will be shown later.

B. Equations for Chemical Equilibrium in Two-Temperature State

Table I indicates ten dissociation and ten ionization equilibrium reactions taken into account in the present calculation. Equations for the reactions will be derived below.

TABLE I
CHEMICAL REACTION TAKEN INTO ACCOUNT IN THE CALCULATION. "M" INDICATES AN ELECTRON OR A HEAVY PARTICLE

$\mathrm{SF_6 + M \rightleftharpoons SF_5 + F + M}$	(4.00 eV [4])	(5)
$\mathrm{SF_5 + M \rightleftharpoons SF_4 + F + M}$	(2.27 eV [4])	(6)
$\mathrm{SF_4 + M \rightleftharpoons SF_3 + F + M}$	(3.47 eV [4])	(7)
$\mathrm{SF_3 + M \rightleftharpoons SF_2 + F + M}$	(2.92 eV [4])	(8)
$\mathrm{SF_2 + M \rightleftharpoons SF + F + M}$	(4.01 eV [4])	(9)
$\mathrm{SF + M \rightleftharpoons S + F + M}$	(3.52 eV [4])	(10)
$\mathrm{SSF_2 + M \rightleftharpoons SF_2 + M}$	(3.90 eV [4])	(11)
$\mathrm{FSSF + M \rightleftharpoons SF + SF + M}$	(3.71 eV [4])	(12)
$\mathrm{F_2 + M \rightleftharpoons F + F + M}$	(1.60 eV [4])	(13)
$\mathrm{S_2 + M \rightleftharpoons S + S + M}$	(4.37 eV [4])	(14)
$\mathrm{F^- + M \rightleftharpoons F + e^- + M}$	(3.40 eV [4])	(15)
$\mathrm{S^- + M \rightleftharpoons F + e^- + M}$	(2.08 eV [4])	(16)
$\mathrm{F + M \rightleftharpoons F^+ + e^- + M}$	(17.4 eV [5])	(17)
$\mathrm{S + M \rightleftharpoons S^+ + e^- + M}$	(10.4 eV [5])	(18)
$\mathrm{F^+ + M \rightleftharpoons F^{2+} + e^- + M}$	(35.6 eV [5])	(19)
$\mathrm{S^+ + M \rightleftharpoons S^{2+} + e^- + M}$	(23.4 eV [5])	(20)
$\mathrm{F_2 + M \rightleftharpoons F_2^+ + e^- + M}$	(15.8 eV [6])	(21)
$\mathrm{S_2 + M \rightleftharpoons S_2^+ + e^- + M}$	(8.30 eV [6])	(22)
$\mathrm{SF + M \rightleftharpoons SF^+ + e^- + M}$	(10.1 eV [6])	(23)
$\mathrm{SF^- + M \rightleftharpoons SF + e^- + M}$	(2.10 eV [4])	(24)

1) Ionization Equilibrium: Let us discuss the ionization equilibrium in two-temperature state

$$\mathrm{A + M \rightleftharpoons A^+ + e^- + M} \tag{5}$$

where M is an electron or a heavy particle colliding with "A." For this ionization equilibrium, the following expression is established on the basis of the second law of thermodynamics [3]:

$$\frac{\mu_{\mathrm{A}}}{T_{\mathrm{g}}} + \frac{\mu_{\mathrm{M}}}{T_{\mathrm{M}}} = \frac{\mu_{\mathrm{A^+}}}{T_{\mathrm{g}}} + \frac{\mu_{\mathrm{e}}}{T_{\mathrm{e}}} + \frac{\mu_{\mathrm{M}}}{T_{\mathrm{M}}} \tag{6}$$

where the temperature T_{M} of M is T_{e} when M is an electron, while T_{M} is T_{g} when M is a heavy particle. Also, $\mu_x(x = \mathrm{e, A, A^+}$ or M) is the chemical potential of each species. The chemical potential μ_x is expressed in terms of partition function Z_x, number density n_x, and volume of plasma V

$$\frac{\mu_x}{T_x} = -k\ln\left(\frac{Z_x}{n_xV}\right). \tag{7}$$

We especially paid attention to partition function of "$\mathrm{A^+}$" with reference to the ground state of "A." We expressed the partition function as $Z_{\mathrm{A^+}} \times \exp[-(E_{\mathrm{A}} - \Delta E)/kT_{\mathrm{A}}^{\mathrm{ex}}]$, where E_{A} is the ionization potential of "A" and ΔE represents a lowering of ionization potential. This is because the populations of the excited states of "A" follow Boltzmann law for the temperature of T_A^{ex}. By substituting (7) into (6), we get

$$\frac{n_e n_{A^+}}{n_A} = \frac{Z_e Z_{A^+}}{Z_A V} \exp\left(-\frac{E_A - \Delta E}{kT_A^{ex}}\right) \tag{8}$$

$$\Delta E\ [\mathrm{eV}] = 6.96 \times 10^{-9} \left(n_e [\mathrm{m}^{-3}]\right)^{1/3}. \tag{9}$$

For the derivation of number densities through (8), the calculation of partition functions Z_A, Z_{A^+}, and Z_e is necessary. We will therefore discuss the dependence of the partition functions on T_e, T_g, and T^{ex}.

a) Partition function for electron Z_e: Translational motions of electrons depend only on T_e, because T_e indicates the kinetic energy of electrons. Thus, Z_e in two-temperature state is expressed as a function of T_e:

$$Z_e = 2Z_e^{tr} = 2V\left(\frac{2\pi m_e kT_e}{h^2}\right)^{3/2} \tag{10}$$

where Z_e^{tr} is the translational partition function of the electron, h is Planck's constant, and m_e is the mass of an electron.

b) Partition function for heavy particle Z_A and Z_{A+}: Heavy particles have three energy states: translational, rotational–vibrational, and electronic states. Translational motions of heavy particles depend on T_g. Translational partition function Z_A^{tr} is thus calculated using T_g

$$Z_A^{tr} = V\left(\frac{2\pi m_A kT_g}{h^2}\right)^{3/2}. \tag{11}$$

We assumed that rotational–vibrational motions of heavy particles depend on T_g, because these motions are due to the kinetic motions. In this case, rotational–vibrational partition function Z_A^{rv} is written by

$$Z_A^{rv} = Z_A^{rv}(T_g). \tag{12}$$

We calculated Z_A^{rv} for diatomic molecules and for polyatomic molecules in accordance with the methods suggested by Burhorn [7] and Herzberg [8]. On the other hand, since the internal energy state of "A" is governed by T_A^{ex}, electronic partition function Z_A^{ele} is obtained as a function of T_A^{ex}

$$Z_A^{ele} = Z_A^{ele}(T_A^{ex}) = \sum_i g_i \exp\left(-\frac{\epsilon_i}{kT_A^{ex}}\right) \tag{13}$$

where g_i and ϵ_i are statistical weight and excitation potential, respectively. These values are given in [4]–[6]. Use of (11)–(13) induces us to have

$$Z_A = V\left(\frac{2\pi m_A kT_g}{h^2}\right)^{3/2} Z_A^{rv}(T_g) Z_A^{ele}(T_A^{ex}). \tag{14}$$

In a similar manner, the partition function for the ion "A^+" can be expressed as

$$Z_{A^+} = V\left(\frac{2\pi m_{A^+} kT_g}{h^2}\right)^{3/2} Z_{A^+}^{rv}(T_g) Z_{A^+}^{ele}(T_{A^+}^{ex}). \tag{15}$$

Finally, substitution of (10), (44), and (15) into (8) yields a expression as a function of T_e, T_g, and T^{ex}

$$\frac{n_e n_{A^+}}{n_A} = 2\left(\frac{2\pi m_e kT_e}{h^2}\right)^{3/2} \frac{Z_{A^+}^{rv}(T_g) Z_{A^+}^{ele}(T_{A^+}^{ex})}{Z_A^{rv}(T_g) Z_A^{ele}(T_A^{ex})} \times \exp\left(-\frac{E_A - \Delta E}{kT_A^{ex}}\right). \tag{16}$$

This is the equation that we newly developed for ionization equilibrium in two-temperature state. Note that (16) includes the effective excitation temperature. Equation (16) was applied to express each of 10 ionization equilibriums (15)–(24) shown in Table I.

2) Dissociation Equilibrium: Let us discuss the following dissociation equilibrium:

$$\mathrm{A} + \mathrm{M} \rightleftharpoons \mathrm{B} + \mathrm{C} + \mathrm{M} \tag{17}$$

where A is a molecule, B and C are heavy particles, and M is an electron or a heavy particle. In a similar way as the procedure for deriving (15), we can obtain

$$\frac{n_B n_C}{n_A} = \left(\frac{2\pi m_B m_C kT_g}{m_A h^2}\right)^{3/2} \times \frac{Z_B^{rv}(T_g) Z_C^{rv}(T_g)}{Z_A^{rv}(T_g)} \frac{Z_B^{ele}(T_B^{ex}) Z_C^{ele}(T_C^{ex})}{Z_A^{ele}(T_g)} \times \exp\left(-\frac{E_A^d}{kT_A^{ex}}\right) \tag{18}$$

where E_A^d is the dissociation potential of "A." The above expression was adopted to express each of dissociation equilibriums (5)–(14) shown in Table I.

C. Other Governing Equations for Two-Temperature Plasma

Besides the above-mentioned (16) and (18), three equations for Dalton's law of partial pressure, charge neutrality, and conservation law of stoichiometric equilibrium were set up. The equation for Dalton's law in two-temperature state is written by

$$P + \Delta P = n_e kT_e + \sum_{j(j \neq e)} n_j kT_g \tag{19}$$

$$\Delta P = \frac{kT_e}{24\pi}\left[\frac{e^2}{\varepsilon_0}\left(\frac{n_e}{kT_e} + \sum_{j(j\neq e)} z_{\mathrm{eff}j}^2 n_j kT_g\right)\right]^{3/2} \tag{20}$$

where P is the total pressure, ΔP the pressure correction term resulting from the electrostatic interactions [9], $z_{\mathrm{eff}j}$ the effective charge number, ε_0 the permittivity of vacuum, and e the electronic charge.

III. Calculation Procedure

Twenty-three simultaneous equations mentioned above were solved through the following procedures.

1) T_e and T_g are given.

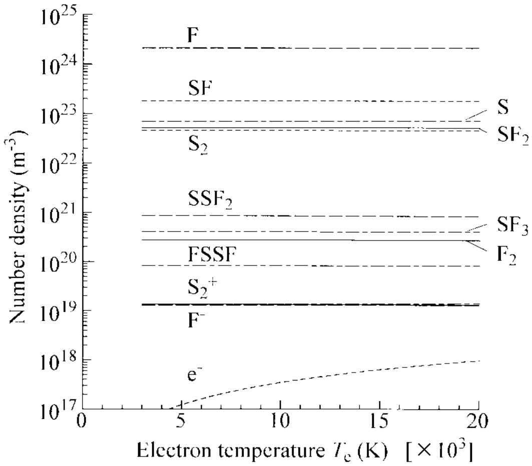

Fig. 1. Particle composition of two-temperature SF_6 plasma at 0.1 MPa as a function of electron temperature. Gas temperature is fixed at 3000 K.

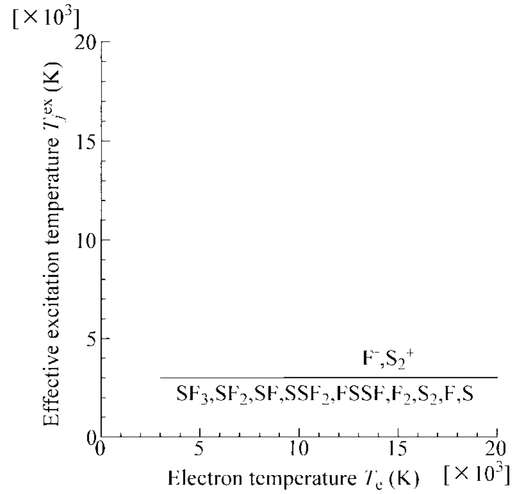

Fig. 2. Effective excitation temperature of various species in two-temperature SF_6 plasma at 0.1 MPa as a function of electron temperature. Gas temperature is fixed at 3000 K.

2) Initial values of number densities n_j of each of species are given.
3) T_j^{ex} is derived using (4) and ΔE is also calculated by (9).
4) The obtained T_j^{ex} and ΔE are substituted into 20 simultaneous equations for ionization and dissociation equilibriums corresponding to (16) and (18).
5) All 23 simultaneous equations are solved using the Newton–Raphson method to newly derive number densities n_j.
6) Procedures of 3–5 are iterated until T_j^{ex} and ΔE converge.

IV. Results and Discussions

Fig. 1 shows the calculated particle composition of SF_6 plasma as a function of T_e at fixed $T_g = 3000$ K. The composition at $T_e = 3000$ K corresponds to that under the condition of thermal equilibrium at 3000 K. An increase in T_e from 3000–20 000 K brings about the little change in the composition. Fig. 2 shows the derived T_j^{ex} of each of the heavy particles as a function of T_e under the same condition as that for Fig. 1. All of T_j^{ex} almost equal T_g. The results shown in Figs. 1 and 2 arise from the following fact. At $T_g = 3000$ K, the electron density n_e is as low as of the order of $10^{17}\,\mathrm{m}^{-3}$. Thus, collision frequencies between heavy particles and electrons are very much lower than those between heavy particles. In other words, the heavy particles collide more frequently with the other heavy particles than with electrons. As a result, T^{ex} of the heavy particle remain T_g regardless of T_e. As a consequence, T_e hardly affects dissociation and ionization reactions, thereby resulting in no change in the composition.

Fig. 3 represents the obtained particle composition at fixed $T_g = 6000$ K as a function of T_e. No significant change in composition is found with increasing T_e from 6000–9000 K. In contrast to this, raising T_e to temperatures above 9000 K leads to marked dissociation and ionization of heavy particles, causing the composition to differ from that under the condition of thermal equilibrium. For example, n_e rises from 6.24

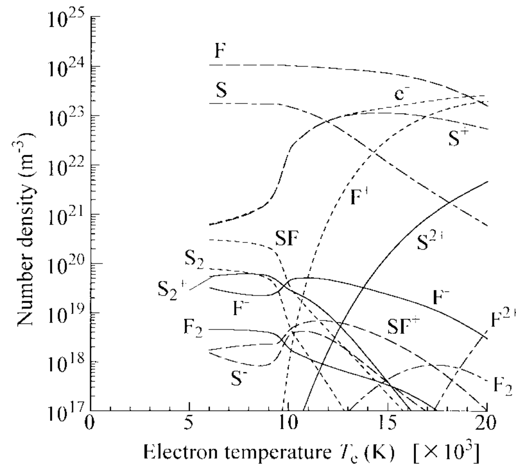

Fig. 3. Particle composition of two-temperature SF_6 plasma at 0.1 MPa as a function of electron temperature. Gas temperature is fixed at 6000 K.

$\times\ 10^{20}$ to 9.24 $\times\ 10^{22}\,\mathrm{m}^{-3}$ with an increase of T_e from 6000–12 000 K. It can be also seen in Fig. 3 that n_e is almost equal to S^+ density at $T_e < 15\,000$ K. This indicates that the ionization of S mainly produces the electron. Fig. 4 represents derived T_j^{ex} as a function of T_e. Let us discuss T_S^{ex} of S that mainly produce the electron. As seen in Fig. 4, T_S^{ex} is almost as high as T_g at $T_e < 8000$ K, while T_S^{ex} agrees with T_e at $T_e > 10\,000$ K. The results shown in Figs. 3 and 4 are interpreted as follows. At $T_e < 8000$ K, n_e is as low as the order of $10^{20}\,\mathrm{m}^{-3}$ so that S excites or ionizes mainly by collision with heavy particles. Thus, T_S^{ex} is kept constant at T_g, while n_e exceeds $10^{22}\,\mathrm{m}^{-3}$ at $T_e > 10\,000$ K. Hence, S excites or ionizes mainly by collision with electrons and thus T_S^{ex} increases to T_e. Consequently, the particle composition depends on T_e markedly, and differs from that under the condition of thermal equilibrium.

Fig. 5 shows the particle composition at fixed $T_g = 9000$ K as a function of T_e. As can be seen in this figure, an increase in T_e induces the dissociation and ionization of all particles, thus causing the number densities of the particle to vary greatly.

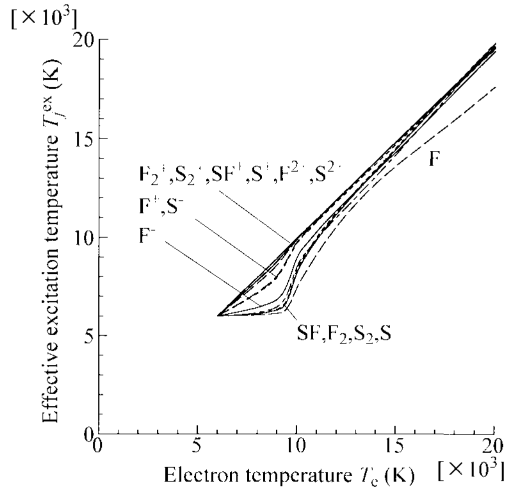

Fig. 4. Effective excitation temperature of various species in two-temperature SF_6 plasma at 0.1 MPa as a function of electron temperature. Gas temperature is fixed at 6000 K.

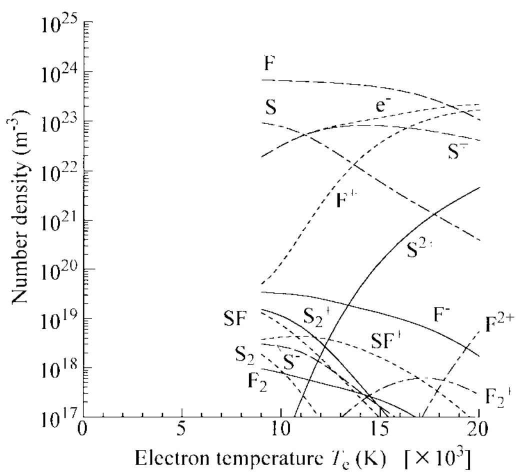

Fig. 5. Particle composition of two-temperature SF_6 plasma at 0.1 MPa as a function of electron temperature. Gas temperature is fixed at 9000 K.

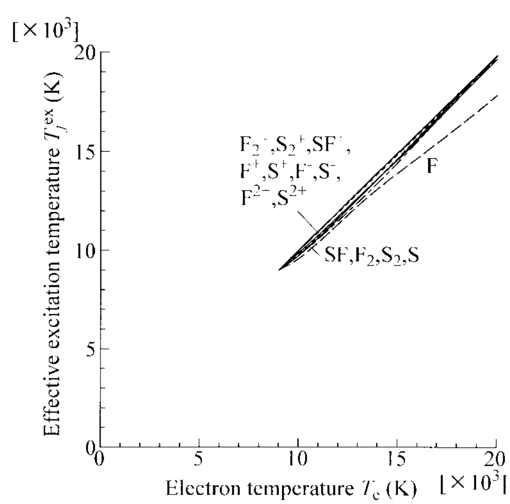

Fig. 6. Effective excitation temperature of various species in two-temperature SF_6 plasma at 0.1 MPa as a function of electron temperature. Gas temperature is fixed at 9000 K.

Fig. 6 represents T_j^{ex} at $T_g = 9000$ K. The effective excitation temperatures of almost all the species rise linearly with T_e. This indicates the particle composition depends almost only on T_e.

V. Conclusion

Particle composition of SF_6 plasma at 0.1 MPa in two-temperature state was theoretically calculated, in which electron temperature T_e is higher than gas temperature T_g. In order to simply express the populations of the internal energy states of heavy particles, the effective excitation temperature was proposed considering collision frequency. At fixed T_g in the range of 6000–9000 K, a rise of T_e from 9000–20 000 K encourages the dissociation and ionization of particles, resulting in the composition of SF_6 plasma to differ markedly from the composition under the condition of thermal equilibrium.

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Yasunori Tanaka, for a photograph and biography, see this issue, p. 990.

Yasunobu Yokomizu (M'91), for a photograph and biography, see this issue, p. 990.

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Toshiro Matsumura (M'95), for a photograph and biography, see this issue, p. 990.