# 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 S F $_6$ 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 $T_e$.

## 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, $\epsilon$ 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, $T_A^{ex}$ depends on both of $T_e$ and $T_g$. We will therefore express $T_A^{ex}$ in terms of $T_e$ and $T_g$.

In the excitation process, a heavy particle "A" obtains energy of $\frac{3}{2} kT_e K_{eA} \nu_{eA}$ a second through collisions with electrons, where $K_{eA}$ and $\nu_{eA}$ 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 $\sum_{j(j \neq e)} \frac{3}{2} kT_g K_{jA} \nu_{jA}$ a second through collisions with various kinds of heavy particles, where $K_{jA}$ and $\nu_{jA}$ are the collision loss factor and the collision frequency between $j$ and "A," respectively. As a result, total energy $E_A^{rec}$ that the particle "A" obtains is given by

$$E_A^{rec} = \frac{3}{2} kT_e K_{eA} \nu_{eA} + \frac{3}{2} kT_g \sum_{j(j \neq e)} K_{jA} \nu_{jA}. \qquad (2)$$

This total energy $E_A^{rec}$ determines $T_A^{ex}$ 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 $\frac{3}{2} kT_A^{ex}$ and collides with "A" with a frequency of $\nu_{eA} + \sum_{j(j \neq e)} \nu_{jA}$. The assumptions enable us to set up the following equation:

$$\frac{3}{2} kT_A^{ex} K \left( \nu_{eA} + \sum_{j(j \neq e)} \nu_{jA} \right) = E_A^{rec} \qquad (3)$$

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 $K_{eA}, K_{jA}$, 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

$$T_A^{ex} = \frac{\nu_{eA} T_e + \sum_{j(j \neq e)} \nu_{jA} T_g}{\nu_{eA} + \sum_{j(j \neq e)} \nu_{jA}}. \qquad (4)$$

This equation expressing $T_A^{ex}$ in terms of $T_e$ and $T_g$ is newly proposed in the present paper. We named $T_A^{ex}$ 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

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

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

$$A + M \rightleftharpoons A^+ + e^- + M \qquad (5)$$
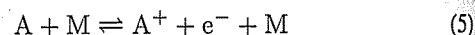
where M is an electron or a heavy particle colliding with "A." For this ionization equilibrium, the following expression is established on the basis of the second law of thermodynamics [3]:

$$\frac{\mu_A}{T_g} + \frac{\mu_M}{T_M} = \frac{\mu_{A^+}}{T_g} + \frac{\mu_e}{T_e} + \frac{\mu_M}{T_M} \qquad (6)$$

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, $\mu_x (x = e, A, A^+$ or M) is the chemical potential of each species. The chemical potential $\mu_x$ is expressed in terms of partition function $Z_x$, number density $n_x$, and volume of plasma $V$

$$\frac{\mu_x}{T_x} = -k \ln \left( \frac{Z_x}{n_x V} \right). \qquad (7)$$

We especially paid attention to partition function of "$A^+$" with reference to the ground state of "A." We expressed the partition function as $Z_{A^+} \times \exp[-(E_A - \Delta E)/kT_A^{ex}]$, where $E_A$ is the ionization potential of "A" and $\Delta E$ 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

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

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

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 $T_e$:

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

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 $T_g$

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

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

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

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 $T_A^{ex}$

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

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

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

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

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

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

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

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:

$$A + M \rightleftharpoons B + C + M \qquad (17)$$
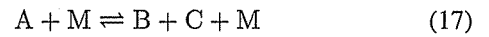
where A is a molecule, B and C are heavy particles, and M is an electron or a heavy particle. In a similar way as the procedure for deriving (15), we can obtain

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

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

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

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

where $P$ is the total pressure, $\Delta P$ the pressure correction term resulting from the electrostatic interactions [9], $z_{eff j}$ the effective charge number, $\varepsilon_0$ the permittivity of vacuum, and $e$ the electronic charge.

## III. CALCULATION PROCEDURE

Twenty-three simultaneous equations mentioned above were solved through the following procedures.
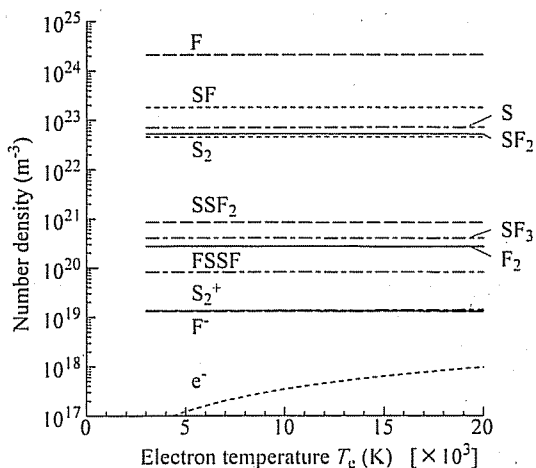
1) $T_e$ and $T_g$ are given.

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.

2) Initial values of number densities $n_j$ of each of species are given.

3) $T_j^{ex}$ is derived using (4) and $\Delta E$ is also calculated by (9).

4) The obtained $T_j^{ex}$ and $\Delta E$ 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 New-ton–Raphson method to newly derive number densities $n_j$.

6) Procedures of 3–5 are iterated until $T_j^{ex}$ and $\Delta E$ 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 $10^{17}$ 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
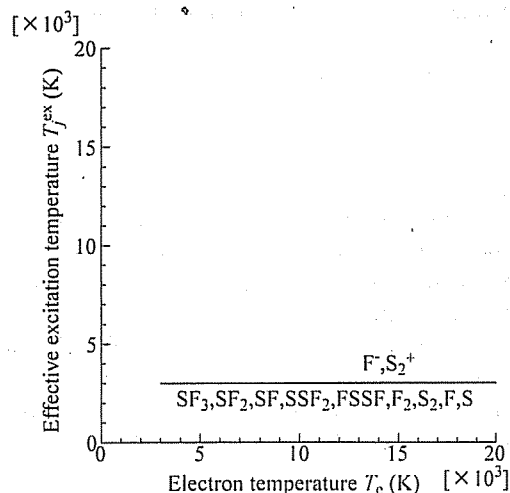


Fig. 2.   Effective excitation temperature of various species in two-temperature SF$_6$ plasma at 0.1 MPa as a function of electron temperature. Gas temperature is fixed at 3000 K.
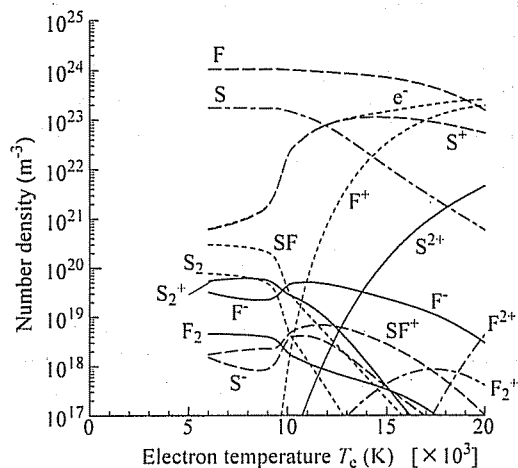


Fig. 3.   Particle composition of two-temperature SF$_6$ plasma at 0.1 MPa as a function of electron temperature. Gas temperature is fixed at 6000 K.

$\times 10^{20}$ to $9.24 \times 10^{22}$ 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 $10^{20}$ 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 $10^{22}$ 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.
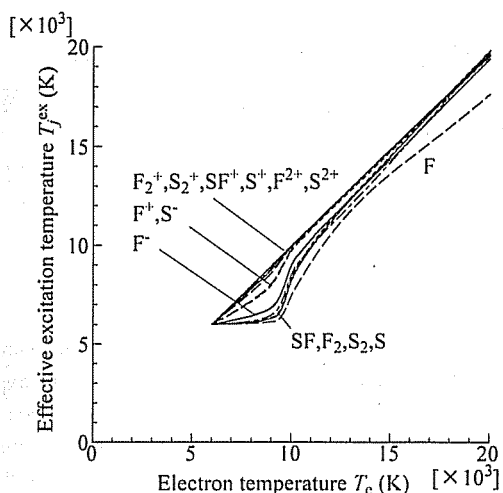
Fig. 4. Effective excitation temperature of various species in two-temperature SF6 plasma at 0.1 MPa as a function of electron temperature. Gas temperature is fixed at 6000 K.
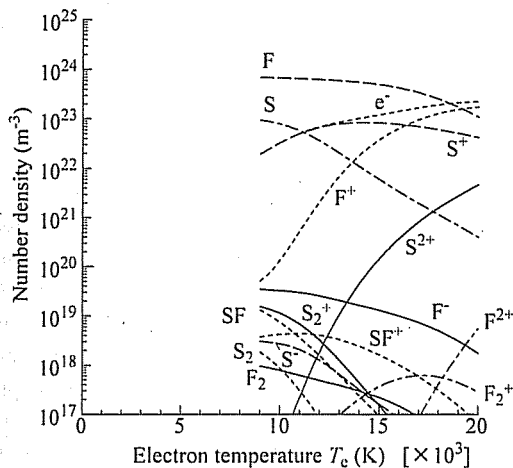


Fig. 5. Particle composition of two-temperature SF6 plasma at 0.1 MPa as a function of electron temperature. Gas temperature is fixed at 9000 K.
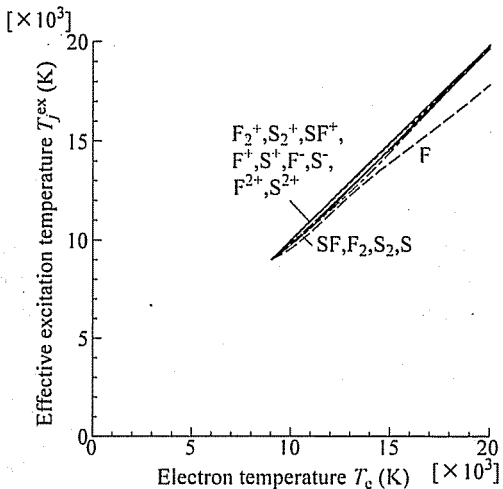


Fig. 6. Effective excitation temperature of various species in two-temperature SF6 plasma at 0.1 MPa as a function of electron temperature. Gas temperature is fixed at 9000 K.

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 SF6 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 SF6 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.

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