Thermodynamic Properties at High Temperatures and Particles Present After Arc Extinction in CO\textsubscript{2}-CF\textsubscript{3}I Mixture

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Keywords: arc, CO\textsubscript{2}, CF\textsubscript{3}I, temperature-reduction factor, principal particles after an arc extinction

From the viewpoint of the environmental issues, SF\textsubscript{6} circuit breakers are expected to adopt the gas not having greenhouse effect as the quenching medium. At the present stage, CO\textsubscript{2} is promising quenching-gas. Furthermore, CO\textsubscript{2} admixed with an other gas is also anticipated to improve the current interrupting capability of the circuit breaker. Meanwhile, CF\textsubscript{3}I is discussed as an insulating gas alternative to SF\textsubscript{6} in the research field of the gas insulated switchgear. Thus the present research aimed at obtaining the fundamental characteristics of a high-temperature CO\textsubscript{2}-CF\textsubscript{3}I mixture to find out the admixing influence of CF\textsubscript{3}I to CO\textsubscript{2}.

Firstly, thermodynamic properties at 0.1 MPa were calculated in a temperature range of 300–30,000 K for various CF\textsubscript{3}I-concentrations \( X_{\text{CF3I}} \). In this determination, the concentration \( X_{\text{CF3I}} \) was defined as the ratio of the total masses of the particles related with CF\textsubscript{3}I to the sum of those of all particles. The calculation results revealed that, for instance, the specific heat for \( X_{\text{CF3I}} \) of 0.9 or less showed the higher magnitude around 6,000–9,000 K, but this higher magnitude disappeared for \( X_{\text{CF3I}} \) above 0.9.

Secondly, from the thermodynamic properties, relative magnitude \( \hat{\beta} \) of temperature-reduction factor was elucidated for different \( X_{\text{CF3I}} \). Figure 1 reveals \( \hat{\beta} \) at 0.1 MPa as a function of the temperature for different \( X_{\text{CF3I}} \). At the same temperatures in the range from 9,000 to 15,000 K, \( \hat{\beta} \) shows no significant dependence on \( X_{\text{CF3I}} \). However, \( \hat{\beta} \) at the same temperature in the range of 6,000 to 9,000 K increases markedly with \( X_{\text{CF3I}} \) from 0.9 to 1, but mostly remains 1 irrespective of \( X_{\text{CF3I}} \) in the range from 0 to 0.7. Above-mentioned dependence of \( \hat{\beta} \) on \( X_{\text{CF3I}} \) provides us the following suggestion: admixing CF\textsubscript{3}I to CO\textsubscript{2} at concentrations above 0.9 have a possibility of decaying the temperature of the extinguishing arc more rapidly, in particular at temperatures of 6,000–9,000 K.

Finally, the principal particles present after an arc extinction were obtained in consideration of the phase transformation of iodine molecules at 0.1 MPa. Figure 2(a) shows the number density of the gas-phase particle at 300 K as a function of \( X_{\text{CF3I}} \) in the range from 0 to 1, while Fig. 2(b) presents the amount of the solid-phase \( I_2(s) \). As seen in this figure, principal particles present after the arc extinction proved to be CO\textsubscript{2}, CF\textsubscript{4} and CO in the gaseous phase and \( I_2 \) in the solid state for \( X_{\text{CF3I}} \) of 0.01 to 0.94, while to be C\textsubscript{2}F\textsubscript{6} and CF\textsubscript{4} in the gas phase and \( I_2 \) in the solid state for \( X_{\text{CF3I}} \) of 0.94 to 1. Experiments were performed and the results supported the validity of the calculation result.
Thermodynamic Properties at High Temperatures and Particles Present After Arc Extinction in CO2-CF3I Mixture

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Fundamental investigations were carried out to find out the admixing influence of CF3I to CO2. Firstly, thermodynamic properties at 0.1 MPa were derived in a temperature range of 300–30,000 K for various CF3I-concentrations $X_{\text{CF3I}}$. Secondly, from the thermodynamic properties, a temperature-reduction factor was determined as a function of the temperature for different $X_{\text{CF3I}}$. This determination led to the suggestion that admixture of CF3I to CO2 at $X_{\text{CF3I}}$ above 0.9 can produce a rapid effect on decay in the temperature of an extinguishing arc. Finally, the principal particles present after an arc extinction were obtained in consideration of the phase transformation of iodine molecules at 0.1 MPa. For instance, the principal particles proved to be CO2, CF4 and CO in the gaseous phase and I2 in the solid phase for $X_{\text{CF3I}}$ of 0.01–0.94.

**Keywords:** arc, CO2, CF3I, temperature-reduction factor, principal particles after an arc extinction

1. Introduction

Sulfur hexafluoride (SF6) is widely used as an arc-quenching gas in a gas circuit breaker and the SF6 has a global warming potential of as high as 23900. From the viewpoint of the environmental issues, the circuit breaker is expected to adopt the gas not having greenhouse effect as the quenching medium.

At the present stage, CO2 is a promising quenching-gas, especially for high-voltage circuit breakers. Furthermore, CO2 admixed with an other gas is also expected as the alternative quenching gas, because the adding the other gas to CO2 may cause a circuit breaker to have higher interrupting capability than use of the pure CO2. On the other hand, CF3I is discussed as an insulating gas alternative to SF6 in the research field of gas insulated switch gear. This is based on the following features: (1) the CF3I shows a dielectric strength similar to SF6 and (2) the CF3I possesses a global-warming-potential below 5. With the above background, Taki et al. have reported experimental results with respect to current interrupting performance for CO2 with additional gas CF3I.

However, regarding to an arc discharge burning in the CO2-CF3I mixture, data on fundamental characteristics of a high-temperature CO2-CF3I mixture are very scare. One of the fundamental characteristics are equilibrium compositions that show the number densities of molecules, atoms, ions and an electron as a function of a temperature. Thus, as the first stage of the present research project, we have revealed the calculation results of the equilibrium composition for the high-temperature CO2-CF3I mixture at 0.1 MPa, on the assumption that all particles are present in gas phases.

The present paper describes the results for the second stage of this research project. Firstly, thermodynamic properties of the high-temperature CO2-CF3I mixture that are one of the fundamental properties are evaluated at 0.1 MPa for different CF3I concentrations. Secondly, use of the thermodynamic properties allowed determination of the dependence of a temperature-reduction factor on the CF3I concentration. The temperature-reduction factor provides the suggestions, regarding the effect of inclusion of CF3I to CO2 on the temperature decay of an interrupting arc. Finally, principal particles present after an arc extinction in CO2-CF3I mixture are obtained as a function of the CF3I concentration. To derive the particles, an attention was paid to take in consideration of particles in the liquid and the solid phases. Furthermore, the equilibrium composition at a pressure of 0.1 MPa was derived in a temperature range from 300 to a sufficient high temperature. This is because the calculation condition corresponds to the following: once after CO2 and CF3I molecules decompose to atoms and further ionize to mono-atomic ions due to a sufficient high temperature, the temperature of CO2-CF3I mixture decreases to the temperature $T$. The calculation result shows the composition at this temperature $T$. Especially, the calculation result at 300 K corresponds to the composition formed after the arc extinction in CO2-CF3I mixture.

2. Thermodynamic Properties at High Temperatures

2.1 Calculation Method

In the first stage of the present research project, we have so far obtained the equilibrium composition of a high-temperature CO2-CF3I mixture at 0.1 MPa. This determination was made at temperatures up to 30,000 K for various CF3I concentrations $X_{\text{CF3I}}$. On the assumption that a thermal equilibrium was established and that all particles were present in the gaseous phase. In this determination, the concentration $X_{\text{CF3I}}$ was defined as the ratio of

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the total masses of the particles related with CF$_3$I to the sum of those of all particles.

Use of the determined equilibrium compositions allowed calculation of the thermodynamic properties at the pressure of 0.1 MPa: a mass density $\rho$, an enthalpy $h$ and a specific heat $C_P$ at a constant pressure, as mentioned below. The mass density $\rho$ is determined from the number density $n_i$ of the species $i$

$$\rho = \sum_{i=1}^{N} m_i n_i,$$

where $m_i$: a mass of species $i$, $N$: the total number of the considered particles. The enthalpy $h$ is expressed by

$$h = \frac{1}{\rho} \sum_{i=1}^{N} \left[ \frac{5}{2} kT + kT^2 \frac{\partial}{\partial T} \left( \ln Z_i^{(\text{int})} \right) + \Delta_f H_i^\circ \right] n_i$$

where $k$: the Boltzmann constant, $T$: a temperature, $Z_i^{(\text{int})}$: internal partition function of species $i$, $\Delta_f H_i^\circ$: a standard enthalpy of formation of specie $i$. The specific heat $C_P$ at a constant pressure $P$ is obtained from:

$$C_P = \frac{\partial h}{\partial T} \bigg|_{P=\text{const}}.$$
X_{\text{CF}_3\text{I}} in the range from 0 to 0.7.

3.3 Discussion  The dependence of $\tilde{\beta}$ on $X_{\text{CF}_3\text{I}}$ provides us the following suggestions, regarding the extinguishing-arc established in the axial gas-flow: (1) adding CF_{3}I to CO_{2} can result in more rapid decay in the arc temperature, especially at temperatures of 6,000 to 9,000 K and (2) this rapid effect distinctly manifests for the cases of $X_{\text{CF}_3\text{I}}$ of 0.9 or above.

Let us consider the circuit breaker adopting the axial flow of a quenching gas as an arc extinction manner. In comparison to a thermal-conduction and a radiation powers, the convection power lost from the arc causes the arc temperature to attenuate around an interrupting-current zero, furthermore dominating an arc-interruption capability. Moreover, the arc around the current zero in the thermal re-ignition region is considered to have the above-mentioned temperatures of 6,000–9,000 K. In consideration of these two situations on the convection loss and the arc temperature, the above-stated dependence of $\tilde{\beta}$ on $X_{\text{CF}_3\text{I}}$ gives us the following suggestion: use of the CO_{2} mixed with CF_{3}I at the concentration above 0.9 enables the circuit breaker to have larger arc-interruption performance in the thermal re-ignition region than use of the pure CO_{2}, whereas the CO_{2}-CF_{3}I mixture at the concentrations lower than 0.9 produces no pronounced effect on the arc-interruption performance.

4. Particles Present After Arc Extinction

Let consider other quenching-gases such as SF_{6}, air and H_{2}. Equilibrium compositions of these gases reveal that even if these molecules decompose to other particles due to a high temperature of an arc, the decomposed particles recombine to the original molecules at 300 K after the arc extinction. However, as described in the present section in detail, the mixture gas consisting of CO_{2} and CF_{3}I shows different phenomena from the other quenching gases and, in other words, the once-decomposed molecules do not necessarily return to the original molecules, even if the temperature returns to 300 K. Concerning this phenomenon, this section will describe the calculation procedure and result. Especially, this calculation takes in consideration of not only the gas-phase particles but also the liquid-phase and the solid-phase particles. This is because this calculation is focused on the main particles not at high-temperatures but at the normal temperature of 300 K. Finally, the calculation result will be compared with measurement results.

4.1 Calculation Method

4.1.1 Particle species  A literature provides us the enormous species of neutral and ionic molecules. Referring to this literature led us to take account of all of neutral and ionic molecules in gas phase, formed from C, O, F and I. These neutral and ionic molecules are 37 and 8 sorts respectively, as listed in Table 1. In this table, a neutral iodine-molecule in the gas phase is denoted by I_{2}(g). Furthermore, we took into consideration of 4 atoms, 11 mono-atomic ions and an electron in gas phase and these species are also listed in Table 1. Among the particles in gas phases, an iodines molecule has a boiling and a melting temperature of 457.5 and 386.8 K at a pressure of 0.1 MPa. These temperature are higher than 300 K. Thereby, in addition to the gas-phase particles, I_{2}(l) and I_{2}(s) in a liquid and a solid state were employed as the species taken into account.

4.1.2 Saturation vapor pressure  As mentioned later in detail, our calculation method utilizes the saturated vapor pressure of the iodine molecule to determine the number density of the iodine molecule in each phase. Thereby the saturation vapor pressure $P^{(\text{sat})}_{I_{2}}$ of the iodine molecule as a function of the temperature was calculated in accordance with

$$P^{(\text{sat})}_{I_{2}} = P_{0} \exp \left( \frac{\Delta_{l}G - \Delta_{g}G_{E}}{RT} \right), \quad \text{......(5)}$$

where $P_{0}$: a standard pressure, $R$: a gas constant, $T$: the temperature, $\Delta_{l}G$: a standard Gibbs energy of formation in gas phase. In addition to these quantities, $\Delta_{g}G$ is a standard Gibbs energy of formation in liquid phase, if $T$ is higher than a melting point. If not, $\Delta_{g}G$ is a standard Gibbs energy of formation in solid phase. Referring the literature enabled us to obtain $\Delta_{l}G_{E}$ and $\Delta_{g}G$ of the iodine molecule as a function of $T$.

4.1.3 Equations  The following assumptions were made: (i) a thermal and a chemical equilibriums are established; (ii) the volumes of the liquid and the solid particles are neglected and thus these never affect the total pressure and (iii) the transformation temperature between I_{2}(l) and I_{2}(s) is 386.8 K, independent of the partial pressure. The equilibrium composition under the condition of the total pressure of 0.1 MPa was calculated for different CF_{3}I concentrations, in consideration of the phase transformation of the iodine molecule. This calculation was performed in the temperature range from 300 K to a sufficient high temperature. The sufficient high temperature was set to be 10,000 K. The detailed procedure is given below.

The following equations were expressed in terms of the number densities $n_{i}$ of the gaseous species $i$:

(i) 37 Guldberg-Waage’s equations for dissociation reactions;
(ii) 19 Saha’s equations for ionization reactions;
(iii) a charge neutrality equation;
(iv) Unsöld’s equation.
(v) Dalton’s law of partial pressures; In addition, the following equations were expressed in terms of not only $n_{i}$ of the gaseous particles but also the number amounts $n_{i}(l)$ of I_{2}(l) and I_{2}(s):
(vi) two equations defining composition of C, O, F and I;
(vii) an expression of CF_{3}I concentration $X_{\text{CF}_3\text{I}}$.\[\text{Various Properties of High Temperature CO}_2\text{-CF}_3\text{I Mixture}\]
The CF3I concentration $X_{CF3I}$ was defined as the ratio of the total mass $M_{CF3I}$ of the particles related with CF3I to the sum of those of all particles: $X_{CF3I} = \frac{M_{CF3I}}{\sum m_{i} n_{i}}$, where $m_{i}$ is the mass of the particle species $i$.

(2) All particles were temporary assumed to be present in the gas state. This assumption yielded

(viii) additional equations: $n_{I2(g)} = n_{I2(s)} = 0$.

Solving the simultaneous equations composed of above-listed equations (i)–(viii) allowed to derive the number densities of the individual particles in the gas phases.

From the derived number densities, the partial pressure $P_{I2(g)}$ of $I_2(g)$ at the temperature $T$ was determined:

$$P_{I2(g)} = \frac{n_{I2(g)} P_{i}}{\sum_{j \in \text{gas}} n_{j}}$$

where $P$: the total pressure of 0.1 MPa. If $P_{I2(g)}$ at $T$ is lower than or equal to the saturation vapor pressure $P_{i}^{(sat)}$ at the same $T$, this fact reveals that a correction for including the phase change is unnecessary. Therefore, the obtained number densities at $T$ are the final solutions.

(3) If $P_{I2(g)}$ at $T$ is higher than the saturation vapor pressure $P_{I2}^{(sat)}$ at the same $T$, this situation shows that the correction calculation is required to determine the compositions in consideration of the phase transformation of the iodine molecule. Thus the correction mentioned below were performed.

(ix) The equation in the item (v) were modified to two equations:

$$n_{I2(g)} = \frac{P_{i}^{(sat)}}{kT}$$

and

$$\sum_{j \in \text{gas}} n_{j} - n_{I2(g)} = \frac{P - P_{i}^{(sat)}}{kT}$$

(x) Furthermore, the equations written in the item (viii) were changed into either

$n_{I2(s)} = 0$ (T ≥ $T_{mI2}$) ................................. (9)

or

$n_{I2(g)} = 0$ (T < $T_{mI2}$), ................................. (10)

depending on $T$, where $T_{mI2}$ represents the melting point of the iodine molecule.

Solving the simultaneous equations listed in the items (i)–(iv), (vi)–(vii) and (ix)–(x) provided us the equilibrium composition with inclusion of the amounts $n_{I2(s)}$ and $n_{I2(g)}$.
Similar calculations were carried out for different $X_{CF_3I}$. Figure 7(a) shows the number density of the gas-phase particles at 300 K as a function of $X_{CF_3I}$ in the range from 0 to 1. Figure 7(b) indicates the same density, especially focused on the concentration range from 0.94 to 1. Meanwhile, Fig. 7(c) presents the amount of the solid-phase $I_2(s)$ as a function of $X_{CF_3I}$ in the range from 0 to 1. Concerning these figures, the detailed descriptions (1)–(4) are given below.

(1) $X_{CF_3I}$ of 0.01: As seen in these figures (a) and (c), admixing CF$_3$I to CO$_2$ at a slight concentration of 0.01 yields CO$_3$ at 10$^{25}$ m$^{-3}$ and also produces CF$_2$O, CO, I$_2$(g) and I$_2$(s) of about 10$^{22}$ m$^{-3}$ order after the decompositions.

(2) $X_{CF_3I}$ in a range from 0.01 to 0.94: Increasing $X_{CF_3I}$ from 0.01 to 0.94 raises the number densities of CF$_2$O and CO from 10$^{25}$ to 10$^{23}$ m$^{-3}$ order by three orders and also grows the amount $I_2(s)$ in the same way. However the same rise in $X_{CF_3I}$ never increase the number density $n_{I_2(g)}$ of I$_2$(g) and thus $n_{I_2(g)}$ remains at a constant of 1.17 × 10$^{25}$ m$^{-3}$.

(3) $X_{CF_3I}$ in a range from 0.94 to 1: It is found from Figs. 7(b) and (c) that a further augment in $X_{CF_3I}$ from 0.94 to 1 allows CO$_2$ and CO to be absent and also introduces slight variation in $n_{CF_2O}$ and $n_{I_2(s)}$. In contrast, the same rise in $X_{CF_3I}$ results in very extreme grow in the number density $n_{CF_2O}$ of C$_2$F$_6$ from 5.50 × 10$^{25}$ to 2.30 × 10$^{23}$ m$^{-3}$ by three orders. This phenomena reveals that the particles of C$_2$F$_6$, CF$_4$ and I$_2$(s) are present at the densities of 10$^{23}$ m$^{-3}$ or above, being as principal ones at 300 K for the case of $X_{CF_3I} = 1$. Whereas increasing drastically with $X_{CF_3I}$ from 0.99 to 1, the density of C$_5$ is 6.3 × 10$^{22}$ m$^{-3}$ at $X_{CF_3I} = 1$. This density is denoted by an open circle, but being lower than those of C$_2$F$_6$, CF$_4$ and I$_2$(s).

(4) I$_2$(s): Recall from Fig. 7(c) that I$_2$(s) shows increase in $n_{I_2(g)}$ from 1.60 × 10$^{22}$ to 2.41 × 10$^{25}$ m$^{-3}$ with $X_{CF_3I}$ from 0.01 to 1, while the number density of I$_2$(g) remains at a constant of 1.17 × 10$^{22}$ m$^{-3}$. This indicates that almost all iodine molecules at 300 K exist in the form of the solid.

4.3 Discussion

4.3.1 Validity Measurements have been performed to find out the particles species present after an arc extinction in the CO$_2$-CF$_3$I mixture [10]. The explanations related to these measurements are given below.

The mixture composed of CO$_2$ and CF$_3$I particles was filled at a pressure of 0.1 MPa in an arc quenching chamber with a volume of 0.05 m$^3$. The CF$_3$I concentration was adjusted to be 0.33, 0.71 and 1. In this arc quenching chamber, a fixed and a moving contacts were arranged as shown in Fig. 8 and this distance between the contacts were about 50 mm under a fully open condition. For each CF$_3$I concentration, a sinusoidal damping current with a frequency of 60 Hz was generated from a capacitor through a reactor to produce an arc and the peak current of the first-half cycle was raised to 300, 500, 800, 1000, 1200, 1500 and 1700 A in this order. By way of example, Fig. 9 indicates the arc current and the voltage between the contacts measured for $X_{CF_3I}$ of 1 and the first peak value of 1,000 A, presenting that the arc burned during
particles of C2F6 and CF4. Unfortunately, the data on neither
6(b), the I2(g) for
lation. This issue will be addressed in future.
of the arc chamber was made to find out whether I2(s) formed
chromatograph-mass spectrometer (GS-MS) and a CO detec-
produced particles in the gas phase were analyzed with a gas
one cycle. After this sequence of the arc-establishments, the
valid.
The compositions derived from the theoretical calculation are
C3F8, C3F6 nor C2F5I are published in the literature (10) so
4.3.2 Thermodynamic properties From the Fig.
the temperature range in which the solid or the liquid iodine-
molecules exists. As seen in this figure, the phase change
the gas into the liquid or the solid occurs at temperatures
around 400 K for all of XCF3-I. This temperature is very much lower than those of an interrupting arc in ex-
cess of several thousands Kelvin. This suggests that the
phase-transformation phenomena never affect the interrupt-
arc. Therefore, even if thermodynamic properties are calculated from compositions without consideration of the
phase change, the properties derived in the previous section 2
are useful for finding out the characteristics of the interrupt-
ing arc.
4.3.3 Solid-phase iodine As mentioned in the sec-
tion 4.2.2, the majority of iodine molecules proved to be
present in the solid state after the arc extinction. The pro-
duced I2(s) are considered to attach to the surface of arc con-
tacts and to deposit on the various-component surfaces inside
an enclosure with the quenching chamber. This phenomenon
has been confirmed in the experiment described in the sec-
tion 4.3.1. In terms of the residual I2(s), practical usages of
the CO2-CF3I mixture as the arc quenching gas in a circuit
breaker are discussed below. The I2(s) attached to the con-
tacts may affect on a contact-resistance as well as the open-
ning and closing motions of the contacts. The I2(s) deposited
on the components may corrode these components and may
produce a degraded effect on an electrical-insulation perfor-
performance. These undesirable phenomena manifest more
pronouncedly at larger XCF3-I, because the amount I2(s) grows
extremely with XCF3-I as found in Fig. 7. Thus a method is
expected to achieve that the enclosure is free from the I2(s)
contamination even after the arc interruptions. By way of ex-
ample of the method, not I2(s) but I2(g) is considered to be
moved outside the enclosure before completion of the phase
transformation from I2(g) into the liquid and the solid phases.
5. Conclusions
Fundamental properties of CO2-CF3I mixture were
obtained in view of the arc-quenching gas. Firstly, the

![Fig. 9. Example of waveforms of arc current and voltage between contacts (CF3-I concentration: 1, Peak current of the first half-cycle: 1,000 A)](image)

![Fig. 10. Example of chromatogram obtained after arc extinctions](image)

![Table 2. Particle species detected after arc extinctions in CO2-CF3-I mixture and their relative concentrations (unit: arbitrary unit)](image)

![Fig. 11. Temperature range in which liquid or solid iodine molecule is present](image)
thermodynamic properties of the high-temperature CO$_2$-CF$_3$I mixture at 0.1 MPa were determined for different the CF$_3$I concentrations $X_{CF_3I}$. For instance, the specific heat for $X_{CF_3I}$ of 0.9 or less showed the higher magnitude around 6,000–9,000 K, but this higher magnitude disappeared for $X_{CF_3I}$ above 0.9. Secondly, from the thermodynamic properties, one of the temperature-reduction factor was elucidated for different $X_{CF_3I}$.

On the basis of this factor, admixing CF$_3$I to CO$_2$ at concentrations above 0.9 proved to have a possibility of decaying the temperature of the extinguishing arc more rapidly, in particular at temperatures of 6,000–9,000 K. Finally, principal particles present after the arc extinction more rapidly, in particular at temperatures of 6,000–9,000 K. But this higher magnitude disappeared for $X_{CF_3I}$ of 0.9 or less showed the higher magnitude around 6,000–9,000 K. The present research was partly supported by the New Energy and Industrial Technology Development Organization (NEDO) of Japan.

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References


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