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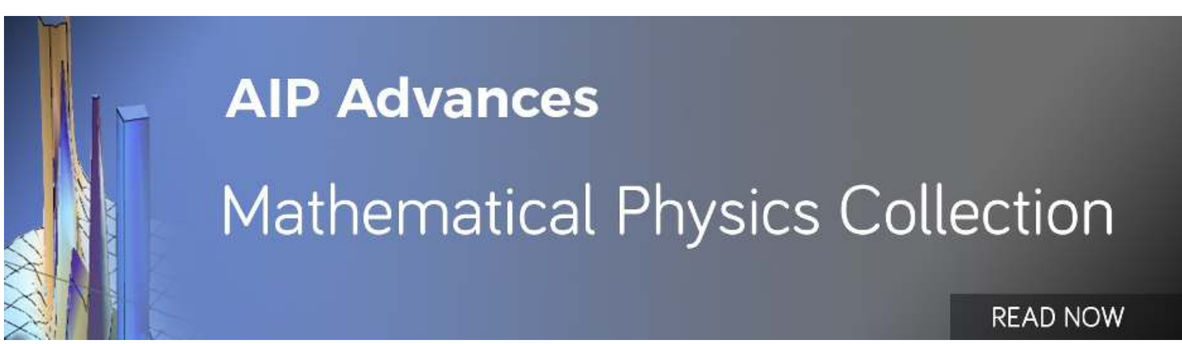

Comparison of dielectric breakdown properties for different carbon-fluoride insulating gases as SF_6 alternatives

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As a widely used insulating medium, sulfur hexafluoride (SF_6) is a greenhouse gas with very high global warming potential (GWP). Some carbon-fluoride gases have potential to replace SF_6 in insulating applications. In order to reveal their different dielectric performance, this paper is devoted to a comparative study of dielectric breakdown properties for SF_6 and four carbon-fluoride insulating gases i.e. CF_3I, C_2F_6, C_3F_8, and c-C_4F_8 mixed with CO_2, N_2, and CF_4 based on the numerical solution of Boltzmann equation. The electron energy distribution function (EEDF), reduced ionization coefficients α/N, reduced electron attachment coefficients η/N, and reduced critical electric field strength $(E/N)_{cr}$ are compared for various gas mixtures. Generally c-C_4F_8 presents the largest dielectric strength among the four carbon-fluoride insulating gases whichever buffer gas is mixed, while C_2F_6 presents the lowest dielectric strength. In terms of $(E/N)_{cr}$ and GWP, CF_3I is a good eco-friendly insulating medium. However, with the addition of buffer gases, the $(E/N)_{cr}$ of CF_3I mixtures declines more quickly than other mixtures. It is also found that the mixing of CF_4 makes insulating mixtures depend more linearly on the proportions of buffer gas than CO_2 and N_2. https://doi.org/10.1063/1.5043516

I. INTRODUCTION

Sulfur hexafluoride (SF_6) is a widely used insulating gas in high-voltage power apparatus e.g. gas-insulated switchgears (GIS) and gas-insulated transmission lines (GIL). However, due to the extremely high global warming potential (GWP) which is nearly 23900 times higher than that of CO_2 over a 100 year interval, SF_6 has been designated as one of the six greenhouse gases by the Kyoto Protocol.[1] Finding suitable SF_6 alternatives is therefore an urgent task.

During the past few decades, the searching of SF_6 replacements has been divided into two directions: one to mix SF_6 with buffer gases having low GWP, and the other to replace SF_6 with completely new eco-friendly gases. In the former way, various SF_6 mixtures were studied, such as SF_6-CO_2,[2,3] SF_6-N_2,[3,4] SF_6-CF_4,[5] and SF_6-He.[6] In the latter way, some carbon-fluoride compounds, such as CF_3I,[7–9] C_2F_6,[5,10] C_3F_8,[11,12] c-C_4F_8,[13–15] C_4F_7N,[16] $C_5F_{10}O$,[16–19] and $C_6F_{12}O$,[16] were found to present high dielectric strength. Compared with SF_6 mixtures, the carbon-fluoride gases show lower values of GWP and hence have potential to replace SF_6 and reduce the usage and emission of greenhouse gases in terms of GWP. However, as illustrated in Table I, most of the carbon-fluoride gases have relatively high boiling points, which consequently restricts their application in cold areas.

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TABLE I. Relative critical dielectric strength (E_{cr}), global warming potential (GWP) and boiling points (T_b) of various insulating gases at ambient pressure.

Gas	E_{cr}	GWP	T_b (°C)	Source.
SF_6	1.00	23900	-63.8	Ref. 17
N_2	0.36	0	-198	Ref. 17
CO_2	0.30	1	-78	Ref. 17
CF_4	0.42	6300	-128	Ref. 17
CF_3I	1.2	0.4	-21.8	Ref. 16
C_2F_6	0.77	9200	-78.2	Ref. 10
C_3F_8	0.96	7000	-37	Ref. 17
c-C_4F_8	1.1-1.3	8700	-6	Ref. 15, 17
C_4F_7N	2	1490	-4.7	Ref. 16
$C_5F_{10}O$	1.5-2.0	1	26.9	Ref. 16, 19
$C_6F_{12}O$	2.7	1	49	Ref. 16

For example, c-C_4F_8 is liquefied above -6 °C at ambient pressure, while the temperature in winter in some northern countries falls to -30 °C or even lower. Therefore, in order to increase the boiling points of insulating gases, it is necessary to mix them with buffer gases having low boiling points, such as CO_2, N_2, and CF_4.

As mentioned above, there are a number of works on the dielectric performance of SF_6 and carbon-fluoride mixtures. However, each of the previous works focused on only one specified insulating gas and did not clear the difference between different insulating gases, which is practical and crucial for engineers to select a SF_6 substitute among numerous insulating gas mixtures.

In order to reveal the different insulating performance between different gases, this paper presents a comparative study of dielectric breakdown properties for SF_6 and four carbon-fluoride insulating gases i.e. CF_3I, C_2F_6, C_3F_8, and c-C_4F_8 mixed with CO_2, N_2, and CF_4 based on the numerical solution of Boltzmann equation. The rest of the paper is organized as follows. In section II, the method to determine dielectric breakdown properties is described and the electron-impact cross sections used in the calculation is also presented. In section III, the dielectric breakdown properties including electron energy distribution functions (EEDF), reduced ionization coefficients α/N, reduced electron attachment coefficients η/N, and reduced critical electric field strength $(E/N)_{cr}$ for SF_6 and four carbon-fluoride gas mixtures are compared with each other. Finally, some remarks are concluded.

II. CALCULATION OF DIELECTRIC BREAKDOWN PROPERTIES

The insulating performance of gas mixtures is usually evaluated on the basis of their dielectric breakdown properties, such as electron-impact ionization and attachment coefficients, and critical electric field strength. The common theoretical approaches to obtain such properties comprise Monte Carlo method[13] and Boltzmann equation method.[20] Following our previous works[2,11] and also considering that the Monte Carlo method is much more time-consuming, the latter method i.e. Boltzmann equation analysis is adopted in this work.

A. Calculation method

Electrons in gas mixtures at room temperature are far from thermal equilibrium and thus their distribution function is far from Maxwellian. It is common practical to derive the electron energy distribution function (EEDF) from the solution of the Boltzmann equation describing the electron transport in gas mixtures as follows.[21]

$$\frac{\partial f}{\partial t} + \vec{v} \cdot \nabla f - \frac{e}{m}\vec{E} \cdot \nabla_v f = C[f] \quad (1)$$

Where f is the electron distribution in six-dimensional phase space, v the velocity, e the elementary charge, m the electron mass, E the electric field, ∇_v the velocity-gradient operator, and C represents the rate of change in f due to the elastic and inelastic collisions.

Assuming that the electric field and the collision probabilities are spatially uniform, a common approach to solve Boltzmann equation is to expand the electron distribution function in two terms of Legendre polynomials of spherical harmonics expansion. In high precision cases, six or more expansion terms are needed, but in many cases a two-term approximation can provide sufficient accuracy.[21]

After obtaining the EEDF, the reduced ionization coefficient α/N (also known as Townsend ionization coefficient) and reduced electron attachment coefficient η/N (also known as Townsend attachment coefficient) are calculated according to the following definitions.

$$\alpha/N = \frac{E/N}{P/N}\frac{m}{4\pi e}\int_0^{\infty}\sum_{k=ionization} x_k Q_k F_0 N_e \varepsilon d\varepsilon \tag{2}$$

$$\eta/N = \frac{E/N}{P/N}\frac{m}{4\pi e}\int_0^{\infty}\sum_{k=attachment} x_k Q_k F_0 N_e \varepsilon d\varepsilon \tag{3}$$

Where E/N is the reduced electric field and P/N is the reduced power gained by the electrons from the electric field.

The reduced critical electric field strength $(E/N)_{cr}$ is therefore determined when the formation and loss of electrons reach a balance. This means that the effective ionization coefficient (α-η)/N equals to zero.

B. Electron-impact cross sections

As described in Section II–B, the electron-impact collision cross sections are needed to solve the Boltzmann equation, and to obtain the EEDF and the electron swarm coefficients. In this work, the collisions between heavy particles as well as the photo-detachment and photo-ionization collisions are not considered due to their negligible effect on the dielectric breakdown properties of gas mixtures. The influence of electron-electron collisions becomes significant only when the ionization degree is above 10^{-6}.[21] However, the ionization degree of insulating gases at room temperature is so low that the electron-electron collisions can also be neglected. Therefore, only the interactions, including elastic, excitation, ionization, and attachment collisions, between electrons and neutral species are taken into account. It should be noted that the effect of ion kinetics[22] is not considered in this work because of the low gas temperature.

Figure 1 presents the electron-impact cross sections of CF_3I, C_2F_6, C_3F_8, and c-C_4F_8 used in the work. The cross sections for CF_3I, C_2F_6, and c-C_4F_8 were compiled from the works by Kimura and Nakamura,[23] Christophorou and Olthoff,[10] and Yamaji and Nakamura[24] respectively. The excitation cross sections of C_3F_8 were determined according to the electron swarm experiment by Jeon,[25] and the other cross sections were compiled from Ref. 26. The cross sections of SF_6, CO_2, N_2, and CF_4 are consistent with our previous publication.[2,11,20] In order to make the calculated electron swarm coefficients agree better with experimental results, the excitation cross sections of C_2F_6 and C_3F_8 were adjusted following the method used by Kimura and Nakamura.[23]

III. COMPARISON OF DIELECTRIC BREAKDOWN PROPERTIES

The dielectric breakdown properties including the electron energy distribution functions (EEDF), reduced ionization coefficients α/N, reduced electron attachment coefficients η/N, and reduced critical electric field strength $(E/N)_{cr}$ of SF_6 and carbon-fluoride insulating gas mixtures are compared in this section. Some gases especially large molecular gases e.g. c-C_4F_8 present a pressure dependence of ionization and electron attachment processes. According to the report by Christophorou and Olthoff,[27] no pressure dependence was observed in the measurements of ionization coefficients α/N in c-C_4F_8 gas. However, the effective ionization coefficients (α-η)/N of c-C_4F_8 is pressure dependent in a certain pressure range. It should be noted that this pressure dependence was not considered in this work.

A. Electron energy distribution function (EEDF)

The EEDF of a gas is essential in gas discharge modeling because it is needed to compute reaction rates for electron collision reactions, such as electron-impact ionization and attachment.

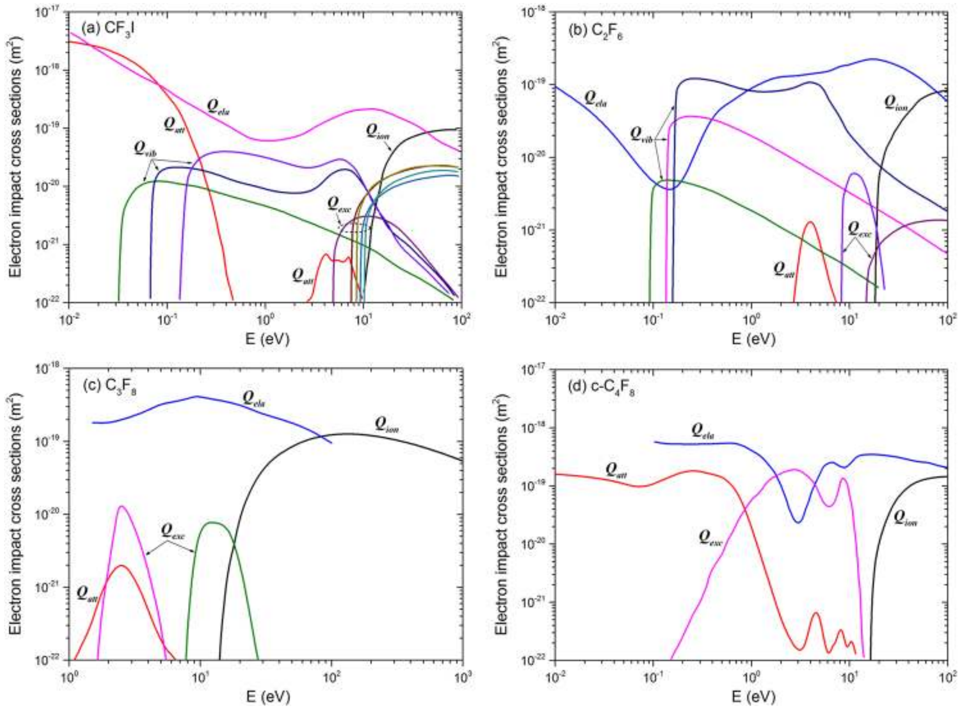

FIG. 1. Electron-impact cross sections of carbon-fluoride insulating gases i.e. CF_3I, C_2F_6, C_3F_8, and c-C_4F_8. Q_{ion}, Q_{att}, Q_{ela}, Q_{exc}, and Q_{vib} stand for electron-impact ionization, attachment, elastic, electronic excitation, and vibration excitation cross sections respectively.

Due to the departure from thermal equilibrium, the EEDF of gas mixtures at room temperature is far from Maxwellian.

Figure 2 describes the EEDF of C_2F_6 mixed with various proportions (in volume) of CO_2, N_2, and CF_4 at E/N of 300 Td. As observed in figure 2, the addition of buffer gases decreases the amounts of electrons with relatively low energy and increase the amounts of electrons with relatively high energy. It is known that electrons with higher energy colliding with neutral particles lead to more excited, positive and negative ionic species through electron-impact excitation, ionization, and attachment reactions. As a result, the mixing of buffer gases is expected to weaken the dielectric performance of gas mixtures.

Another observation is that the influence of buffer gases CO_2, N_2, and CF_4 on the EEDF is in the ascend order, which means CF_4 has a larger impact on the EEDF than N_2, and N_2 has a larger impact than CO_2. This can be attributed to their different dielectric strength E_{cr} as shown in Table I, i.e. $E_{cr}(CF_4) > E_{cr}(N_2) > E_{cr}(CO_2)$. This also indicates that the dielectric strength of a gas is associated with its EEDF. However, it is hard to deduce dielectric performance of a gas qualitatively only according to its EEDF because the dielectric properties e.g. ionization coefficient and electron attachment coefficient are the integrals of EEDF and corresponding cross sections from zero to infinite energy as formulated in equation (2) and (3).

The EEDF at E/N of 300 Td for SF_6 and four carbon-fluoride insulating gases i.e. CF_3I, C_2F_6, C_3F_8, and c-C_4F_8 mixed with 50% CO_2, N_2, and CF_4 (in volume) is compared in figure 3 so as to discovery the difference between their EEDF. As seen from figure 3, SF_6 mixtures present more electrons with high energy and less electrons with low energy than other gas mixtures no matter which buffer gas is mixed. Besides, there are less electrons with low energy in the insulating gases mixed with CF_4 than the gases mixed with CO_2 and N_2.

It is also found that the EEDF of SF_6, CF_3I, and c-C_4F_8 mixtures are all observed a peak at around 1 eV, which means the quantity of electron with energy below 1 eV is reduced. This could probably affect the electron swarm parameters at low values of E/N because electrons obtain kinetic energy through the acceleration in the applied electric field.

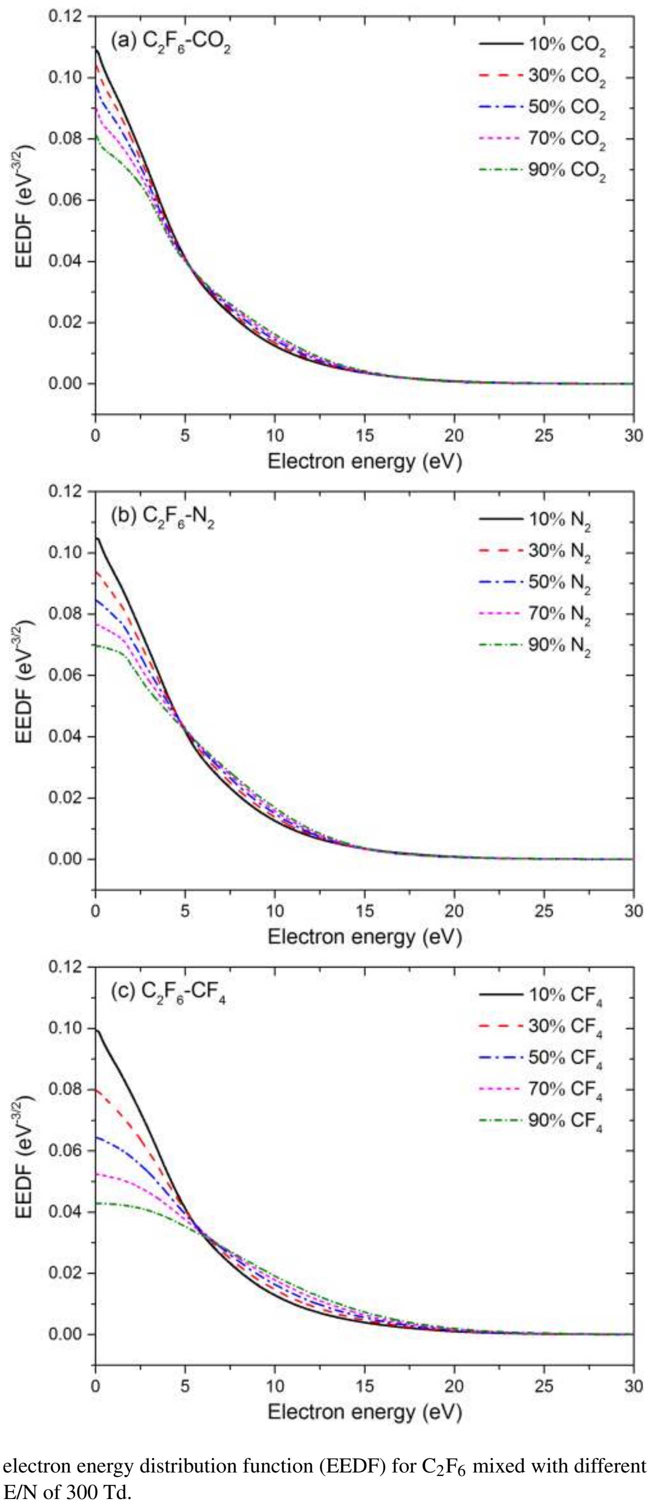

FIG. 2. Comparison of electron energy distribution function (EEDF) for C_2F_6 mixed with different proportions (in volume) of CO_2, N_2, and CF_4 at E/N of 300 Td.

B. Reduced ionization and electron attachment coefficients

Electron-impact ionization and electron attachment processes play an important role in Townsend discharge. Once obtained the EEDF, The reduced ionization coefficients α/N and reduced electron attachment coefficients η/N of SF_6 and carbon-fluoride gas mixtures are calculated according to equation (2) and (3). Consequently, the effective reduced ionization coefficient (α-η)/N is determined

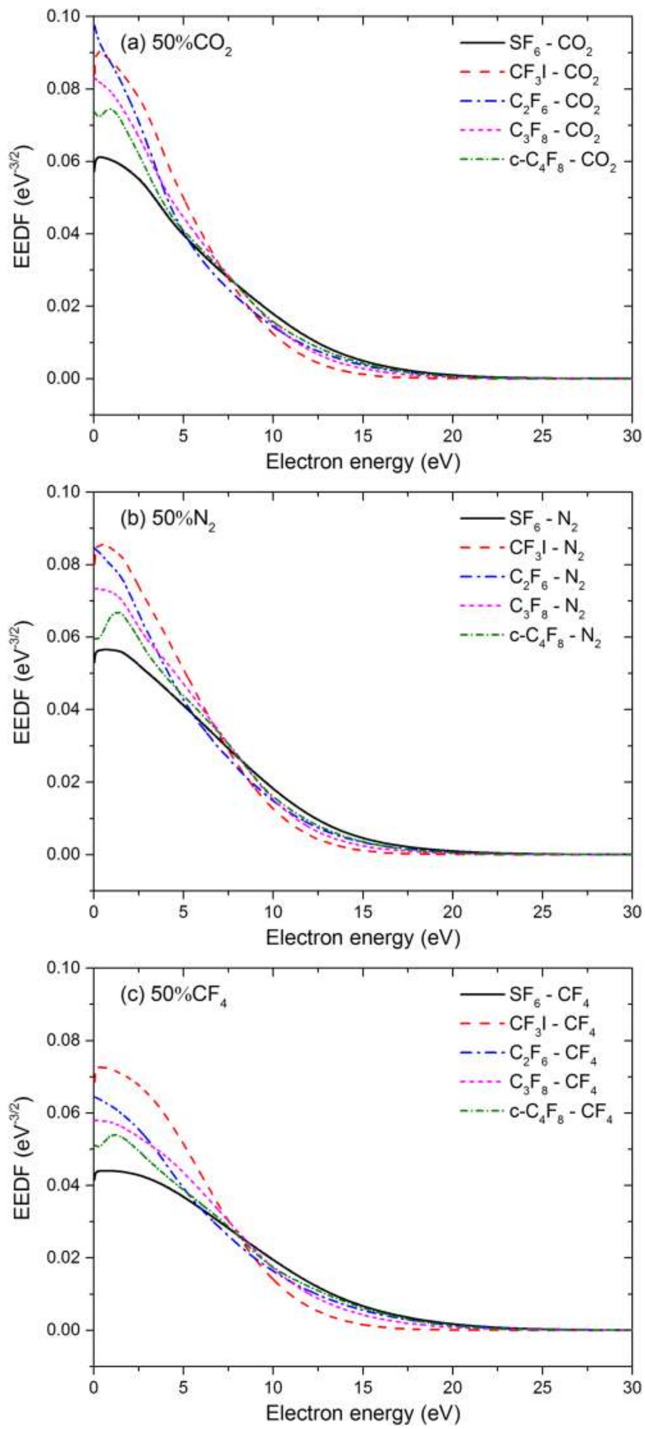

FIG. 3. Comparison of electron energy distribution function (EEDF) for SF_6 and carbon-fluoride insulating gases (CF_3I, C_2F_6, C_3F_8, and c-C_4F_8) mixed with CO_2, N_2, and CF_4 (50% in volume) at E/N of 300 Td.

for a gas, which characterize its ability of generating net electrons in gas discharge. The intersection point of the two curved lines for α/N and η/N respectively corresponds to a critical condition. The comparison of α/N and η/N for various gas mixtures is presented in figure 4 and 5.

Figure 4 illustrates the values of α/N and η/N for C_2F_6-CO_2, C_2F_6-N_2, and C_2F_6-CF_4 mixtures with various proportions of buffer gases in volume. It is seen that the ionization coefficients

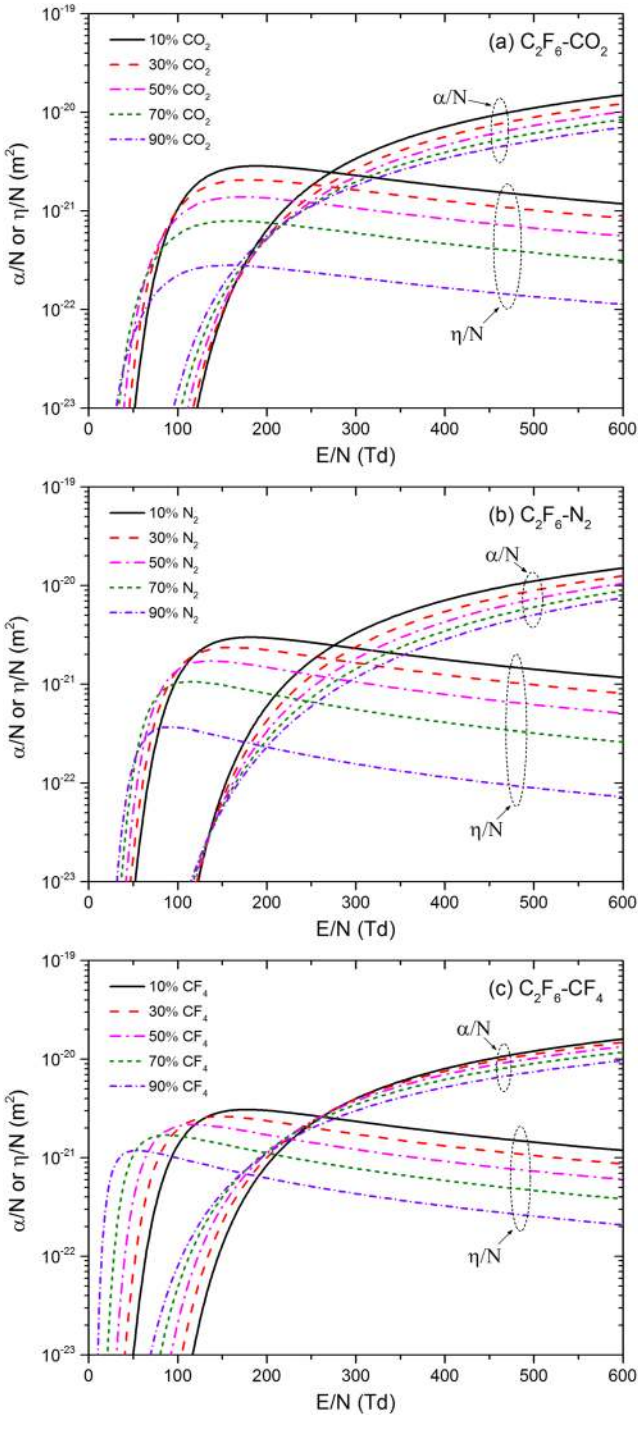

FIG. 4. Comparison of reduced ionization coefficients α/N and reduced electron attachment coefficient η/N for C_2F_6 mixed with different proportions (in volume) of CO_2, N_2, and CF_4.

increase with the values of E/N because electrons obtain more energy at larger E/N, which make gas molecules easier to ionize colliding with such electrons. However, as shown in figure 4, the electron attachment coefficients rise and then fall with the increase of E/N. This can be explained as follows. On the one hand, electrons with higher energy is easier to generate electron-attached anions during colliding with gas molecules, which results in larger values of η/N. On the other hand, electrons with higher energy transport more quickly than those with lower energy, which makes gas

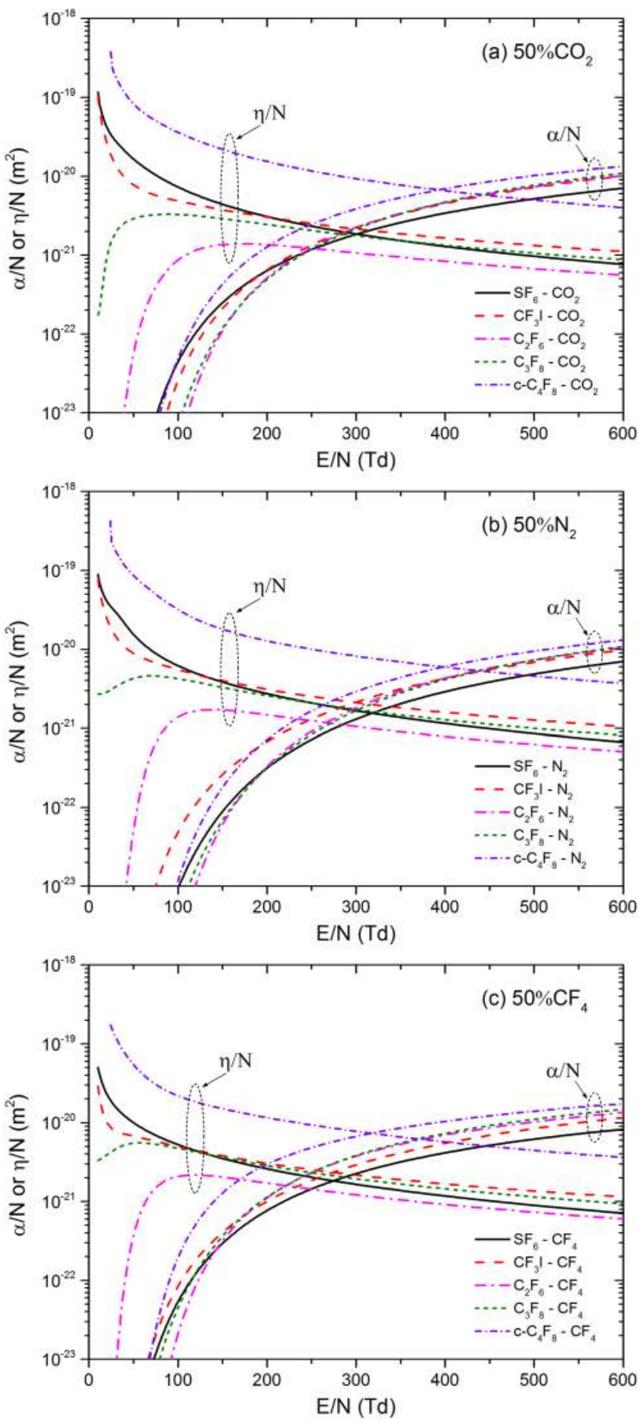

FIG. 5. Comparison of reduced ionization coefficients α/N and reduced electron attachment coefficient η/N for SF_6 and carbon-fluoride insulating gases (CF_3I, C_2F_6, C_3F_8, and $c\text{-}C_4F_8$) mixed with CO_2, N_2, and CF_4 (50% in volume).

molecules more difficult to capture them, resulting in lower values of η/N. Therefore, there exists a peak in the graph of η/N for C_2F_6 mixtures. This peak corresponds to the balance of these two mechanism.

It is also observed in figure 4 that the values of α/N and η/N are both raised at low E/N and reduced at high E/N with the addition of whichever buffer gases. In general, the mixing of buffer gases weakens the insulating performance of gas mixtures due to the low dielectric strength of buffer gases.

The intersection point of two curved lines for α/N and η/N respectively corresponds to a critical condition for dielectric breakdown, which will be discussed in section III C for critical dielectric strength. As seen from figure 4, the intersection point is shifted towards a low E/N with the increase of content of buffer gases.

The three parts of figure 5 describe the comparison of α/N and η/N for SF_6, CF_3I, C_2F_6, C_3F_8, and c-C_4F_8 mixed with three buffer gases CO_2, N_2, and CF_4 respectively. The mixing ratios are all 1:1 in volume. As seen from figure 5, c-C_4F_8 mixtures have much larger α/N and also larger η/N than the other gas mixtures. According to the intersection point of α/N and η/N, c-C_4F_8 mixtures also present much larger critical dielectric strength no matter which buffer gas is mixed. The α/N of C_2F_6 is almost the same as that of C_3F_8, which is consistent with their ionization energy as listed in Table II.[27,28] However, as shown in figure 5, the electron attachment ability of C_2F_6 is much poorer than C_3F_8, which thus makes the dielectric performance of C_2F_6 much weaker than that of C_3F_8. Although CF_3I has lower ionization energy than C_3F_8 as shown in Table II, the values of α/N for CF_3I at high E/N is close to those of C_3F_8. Meanwhile, the η/N for CF_3I are slightly larger than that of C_3F_8, which makes CF_3I present better dielectric performance.

C. Reduced critical electric field strength

As discussed in section III B, the intersection point of the graphs for α/N and η/N respectively is a critical point which is associated with critical dielectric breakdown for a gas. The reduced electric field E/N corresponding to this critical point is called reduced critical electric field strength $(E/N)_{cr}$, which means a dielectric breakdown will occur as long as the applied electric field strength is higher than $(E/N)_{cr}$. Accordingly, the values of $(E/N)_{cr}$ are determined when the ionization of gas mixtures is completely balanced by the electron attachment.

In order to reveal the difference of $(E/N)_{cr}$ between SF_6 and carbon-fluoride insulating gases, figure 6 compares the $(E/N)_{cr}$ for various SF_6, CF_3I, C_2F_6, C_3F_8, and c-C_4F_8 mixtures with CO_2, N_2, and CF_4. Obviously, the insulating gases composed of large carbon-fluoride molecules, such as c-C_4F_8, always present high dielectric strength. This can be attributed to the very high electron affinity of fluorine atom as illustrated in Table III.[29] It is also found that the gases composed of halogen group atoms, such as CF_3I, have very high insulating performance due to the excellent electron attachment ability of halogen atoms such as iodine and fluorine.

CF_3I has another advantage as its global warming potential (GWP) is much lower than that of other carbon-fluoride gases. Moreover, CF_3I has lower boiling point than c-C_4F_8, which means CF_3I needs fewer buffer gases to mix with than c-C_4F_8 to make sure the mixtures can be applied at high pressures and in cold areas. However, with the addition of buffer gases, the dielectric performance of CF_3I mixtures declines more quickly than c-C_4F_8. Hence, more CF_3I and less c-C_4F_8 are needed to achieve a given dielectric strength under the same conditions.

Another interesting observation is the dependence of $(E/N)_{cr}$ on the proportions of buffer gases. As seen from figure 6, the $(E/N)_{cr}$ of CF_3I is always linearly proportional to the content of buffer gas whichever gas is mixed, while the graph describing the relationship between $(E/N)_{cr}$ of c-C_4F_8 and the content of buffer gases is a curved line instead of a straight line. Likewise, the $(E/N)_{cr}$ of C_2F_6 and C_3F_8 mixtures with CO_2 and N_2 also depends nonlinearly on the content of buffer gases. The type of buffer gas also affects this dependence. Compared with CO_2 and N_2, CF_4 makes insulating mixtures depend more linearly on the proportions of buffer gas. This could be explained by the synergistic effect between the buffer gases and the carbon-fluoride insulating medium

TABLE II. Ionization energies (IE) of SF_6 and carbon-fluoride insulating gases.[27,28]

gas	IE (eV)	gas	IE (eV)
SF_6	15.32	CF_3I	10.28
C_2F_6	13.60	C_3F_8	13.38
c-C_4F_8	< 16	CO_2	13.78
N_2	15.58	CF_4	14.70

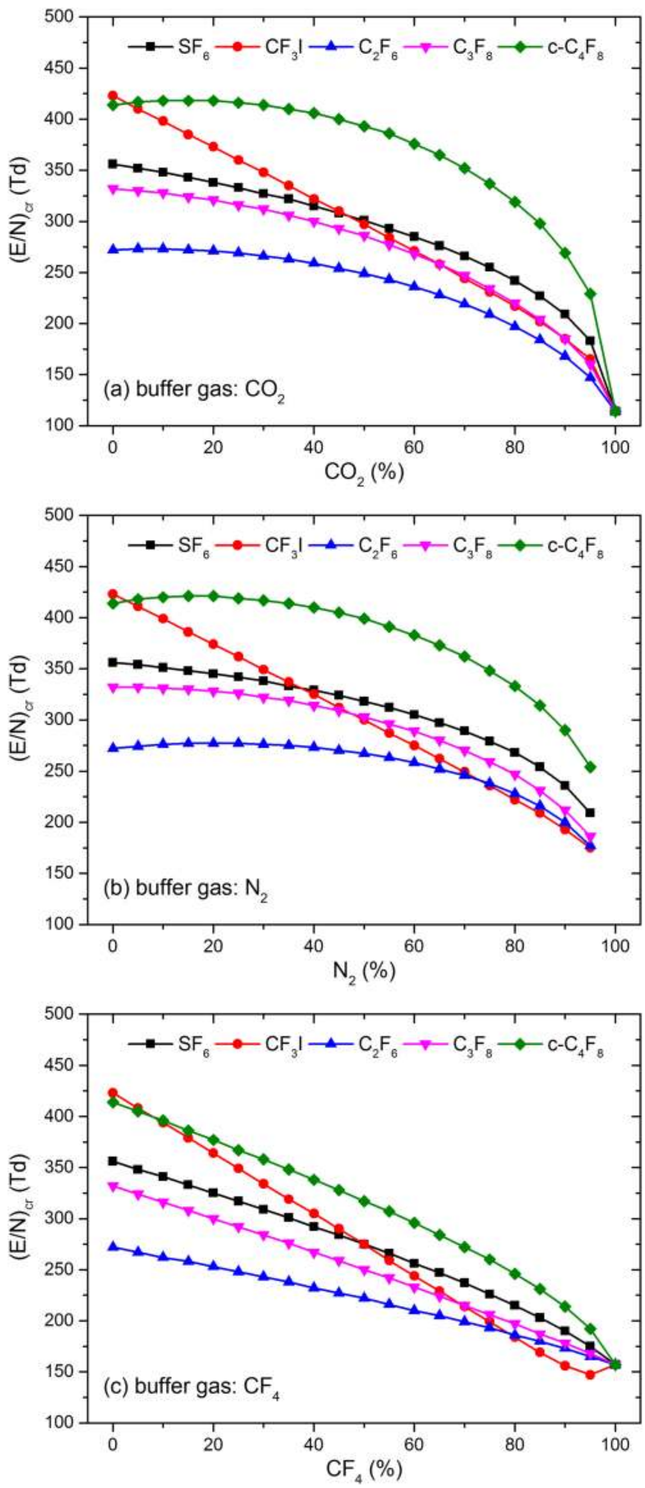

FIG. 6. Comparison of reduced critical electric field strength $(E/N)_{cr}$ for SF_6 and carbon-fluoride insulating gases (CF_3I, C_2F_6, C_3F_8, and c-C_4F_8) mixed with CO_2, N_2, and CF_4.

studied in this work. The synergy between CF_4 and carbon-fluoride gases e.g. c-C_4F_8 is less than that between the other two buffer gases (CO_2 and N_2) and primary gases, which makes the $(E/N)_{cr}$ of CF_4 mixtures decrease more linearly than CO_2 or N_2 mixtures with the increase of buffer gas content.

TABLE III. Electron affinities (EA) of selected atoms.[29]

atom	EA (eV)	atom	EA (eV)
C	1.26	N	< 0
O	1.46	S	2.08
F	3.40	Cl	3.61
Br	3.36	I	3.06

IV. CONCLUSIONS

In this paper, the dielectric breakdown properties of SF_6 and carbon-fluoride insulating gases i.e. CF_3I, C_2F_6, C_3F_8, and c-c-C_4F_8 mixed with CO_2, N_2, and CF_4 are calculated based on the two-term solution of Boltzmann equation. The electron energy distribution functions (EEDF), reduced ionization coefficients α/N, reduced electron attachment coefficients η/N, and reduced critical electric field strength $(E/N)_{cr}$ are compared for various SF_6 and carbon-fluoride mixtures. The following conclusions could be drawn.

a) Generally, the mixing of buffer gases weakens the dielectric performance of gas mixtures because the dielectric strength of buffer gases is much poorer than that of primary insulating gases.
b) Among the three buffer gases, CF_4 has a largest impact on the EEDF of gas mixtures. The mixing of CF_4 reduces more electrons with low energy than CO_2 and N_2. In addition, as a buffer gas, CF_4 makes insulating mixtures depend more linearly on the content of buffer gas than CO_2 and N_2.
c) Among the four primary carbon-fluoride gases, c-C_4F_8 presents the largest dielectric strength in general no matter which buffer gas is mixed, while C_2F_6 presents the lowest dielectric strength. Moreover, the $(E/N)_{cr}$ of c-C_4F_8 mixtures is nonlinearly proportional to the content of buffer gas whichever buffer gas is mixed.
d) In terms of critical dielectric strength and global warming potential, CF_3I is a good eco-friendly insulating medium. However, the $(E/N)_{cr}$ of CF_3I mixtures declines more quickly than other gas mixtures with the addition of buffer gases, which means more CF_3I are needed to achieve a given dielectric strength under the same conditions. It should be noted that the lethal concentration at 50% mortality (LC50) of CF_3I is 160,000,[16] which is large enough to restrict the widely usage of CF_3I in industry.

ACKNOWLEDGMENTS

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