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Investigation of dielectric properties of cold C_3F_8 mixtures and hot C_3F_8 gas as Substitutes for SF_6

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Abstract. In order to reduce the global warming potential resulting from SF_6 widely used as an insulating and arc quenching medium, the substitutes need to be found. This paper focuses on different cold C_3F_8 mixtures (at room temperature) as an insulating gas and hot C_3F_8 gas (at temperatures of 300–3500 K) as an arc quenching medium, which seem to be a good replacement of SF_6. Firstly, the dielectric properties, including the reduced ionization coefficient α/N, reduced electron attachment coefficient η/N and reduced critical electric field strength $(E/N)_{cr}$, of the cold C_3F_8-CF_4, C_3F_8-CO_2, C_3F_8-N_2, C_3F_8-O_2 and C_3F_8-Ar mixtures are calculated numerically using the two-term approximation of the Boltzmann equation. The dependence of such dielectric properties on the buffer gas proportion is investigated. Among the various C_3F_8 mixtures, the C_3F_8-N_2 mixture has the lowest α/N and the C_3F_8-CF_4 mixture has the largest η/N, and moreover, the C_3F_8-N_2 mixture is the best insulator in terms of breakdown strength because it has the largest $(E/N)_{cr}$. Secondly, the $(E/N)_{cr}$ of hot C_3F_8 at temperatures up to 3500 K and various pressures is determined and compared with that of hot SF_6 gas. It is found that the hot C_3F_8 gas has much poorer dielectric performance than hot SF_6 because the $(E/N)_{cr}$ of C_3F_8 decreases significantly above room temperature.

1 Introduction

Sulfur hexafluoride (SF_6) that is nontoxic, nonflammable and chemically stable with high dielectric strength is widely used as an arc quenching gas in high-voltage circuit breakers (HVCB) and an insulating medium in gas-insulated substations (GIS) and gas-insulated lines (GIL). However, SF_6 has been identified as a greenhouse gas with an estimated global warming potential (GWP) that is nearly 24 000 times higher than that of CO_2 over a 100 year interval [1,2]. Also, the Kyoto Protocol has designated SF_6 as one of the gases whose release to the atmosphere needs to be limited [3]. Finding a suitable substitute has therefore been an urgent task. In the past two decades, due to the difficulty of finding the new compounds with the similar physical and chemical properties as SF_6, much attention has turned to the mixtures of SF_6 with different buffer gases (e.g. SF_6-N_2 [1,4,5], SF_6-CF_4 [6], SF_6-CO_2 [1,2,7], SF_6-He [1,4,5,8], SF_6-O_2 [5], et al.). Such mixtures can reduce the usage or emission of greenhouse gases in terms of GWP, but they are not excellent replacement as an insulator because the dielectric strength of such mixtures is inferior to that of pure SF_6. Consequently, it is necessary to investigate the other substitutes.

Some fluorinated compounds, such as C_3F_6 [9,10], C_3F_8 [11,12], c-C_4F_8 [13,14] and CF_3I [15,16], have been found to have much lower GWP and similar or even better dielectric performance than SF_6. For example, the GWP of c-C_4F_8 is 8700, only 36% of SF_6 [13], and the dielectric strength of c-C_4F_8 is about 1.27 times as large as that of SF_6 [17,18], proving that c-C_4F_8 is a good replacement of SF_6 in terms of dielectric strength and environment effect. Unfortunately, the liquefaction temperature of c-C_4F_8 is about −6 °C at ambient pressure, which means that c-C_4F_8 cannot be used in cold areas. Another fluorinated compound C_3F_8 having the low liquefaction temperature (−37 °C at 1 atm) seems to be a very promising candidate. It has good chemical stability, low toxicity and high dielectric strength (94% [17] as large as that of SF_6). What's more, the GWP of C_3F_8 is 7000 times that of CO_2 over a 100 year time period, which is lower than that of c-C_4F_8 which is 8700 over the same period [19]. It should be noted that the pressure dependence of electron attachment in C_3F_8 was reported by Moruzzi et al. [20], Hunter et al. [21] and Koch et al. [12], which is different from SF_6. Fortunately, according to the recent work by Koch et al. [12], the model developed for SF_6, which gives the basis for the prediction of breakdown behaviour, can be applied to C_3F_8.

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The investigation into the dielectric performance of a certain gas is usually divided into two parts. One is for cold gases at room temperature which are applied as insulating medium [1,2,4,5]. The other is for hot gases at temperatures of 300–3500 K which are applied as arc quenching medium [6–8]. During the last few decades, there have been a few studies on the dielectric properties of cold C_3F_8 used as an insulator in plasma processes. Hunter et al. [21,22] studied the ionization processes and negative ion formation mechanism for C_3F_8 and, moreover, they measured the ionization coefficient and electron attachment coefficient of the gas. Spyrou et al. [23] obtained the rate constant for the total electron attachment to C_3F_8 using a swarm apparatus. They also investigated the effect of temperature on the dissociative and nondissociative electron attachment to C_3F_8. Moruzzi et al. [20], Hunter et al. [22] and Naidu et al. [24] used their data on electron attachment and ionization coefficients to deduce the effective ionization coefficient which is essential to determine the critical electric field strength. However, the pure C_3F_8 has a fatal disadvantage that it cannot be applied at high pressures, such as for high-voltage circuit breakers (commonly 0.6 MPa) because the liquefaction temperature of a gas increases with pressure. One way to solve this problem is mixing C_3F_8 with a low liquefaction temperature gas (e.g. N_2, CO_2, CF_4, ...) to obtain a mixture with a low liquefaction temperature and satisfactory insulation. The research on these mixtures is very rare. Kunhardat et al. [25] studied the DC breakdown of C_3F_8-Ar and C_3F_8-N_2 mixtures, and presented the Paschen curves for both mixtures. Hikita et al. [11] analyzed the partial discharge properties and breakdown mechanism of C_3F_8-CO_2 mixture through the experimental approaches. However, no data for dielectric properties of such mixtures have been reported. Furthermore, the above investigation of C_3F_8 is limited to the cases at room temperature. No researchers explored the dielectric breakdown properties of hot C_3F_8 during the dielectric recovery phase in HVCBs.

In this paper, the dielectric breakdown performance of cold C_3F_8 gas mixed with CF_4, CO_2, N_2, O_2 and Ar at room temperature is firstly investigated. The dielectric properties, including the reduced ionization coefficient α/N, reduced electron attachment coefficient η/N and reduced critical electric field strength $(E/N)_{\mathrm{cr}}$, where N refers to the particle number density, are determined by solving the Boltzmann equation numerically. Secondly, the dielectric properties of hot C_3F_8 gas are studied in the temperature range of 300–3500 K. The critical electric field strength of hot C_3F_8 at different pressures is presented and compared with that of hot SF_6 gas to determine whether C_3F_8 is a good arc quenching medium.

2 Calculation method and basic data

The dielectric properties concerning electrons, such as ionization coefficient and electron attachment coefficient, depend on the electron energy distribution function (EEDF) [26]. To obtain such electron swarm parameters, the Monte Carlo method [27,28] or Boltzmann equation method [1,7,29–31] is commonly used. This paper adopts the latter one. The EEDF is to be explicitly computed by the Boltzmann equation without assuming a given distribution function (such as Maxwellian and Druyvesteyn). In this paper, the EEDF of the mixtures is far from the above given functions. The corresponding calculation procedure has been described comprehensively in our previous work [7,32,33]. A brief introduction is presented as follows.

(i) *Solving the Boltzmann equation numerically in an approximate way.* To obtain the EEDF of C_3F_8 gas and its mixtures, a two-term approximation of the Boltzmann equation is adopted in this paper. The interactions including elastic, excitation, ionization and attachment collisions between electrons and neutral species are considered.

(ii) *Calculating ionization and electron attachment coefficients.* Once the EEDF of the gas mixtures is determined, the reduced ionization coefficient α/N and reduced electron attachment coefficient η/N can be calculated easily as discussed by Hagelaar et al. [26] and Holstein [34].

(iii) *Evaluating critical electric field strength.* The critical reduced electric field strength $(E/N)_{\mathrm{cr}}$ is determined when the ionization and electron attachment reach a balance. This means that the reduced effective ionization coefficient $(\alpha-\eta)/N$ equals to zero.

For hot C_3F_8 gas, the dissociation reactions will occur with the increase of temperature. The composition of the hot gas is therefore needed to be determined before solving the Boltzmann equation. As described in our published work [7,32,33], the Gibbs free energy method [35] is applied to obtain the equilibrium composition of hot C_3F_8 gas. The corresponding result is discussed in Section 4.1.

As the necessary input data to the Boltzmann solver, the electron impact collision cross sections for neutral species in the C_3F_8 mixtures are required. Christophorou et al. [36,37] reviewed the research on electron interactions with C_3F_8 and provided the recommended values for various cross sections which are adopted in this paper. As for the excitation cross sections of C_3F_8, we used the recent results reported by Jeon [38]. The same data for CF_4, CO_2 and O_2 used in our previous work [7] are followed in this paper. The corresponding cross sections for N_2 and Ar are compiled from the online database [39–42]. For hot C_3F_8 gas, the cross sections of dissociative products are required. For C, C_2, F, F_2, CF, CF_2, CF_3 and CF_4, the cross sections compiled from our published work for hot SF_6-CO_2 mixtures [7] are used. For C_3, C_2F_4 and C_2F_6, the data presented in another published work for hot CF_4 gas [33] is used. Following our previous work [7,32], the unavailable ionization cross sections are calculated based on the Deutsch-Märk (DM) formalism [43]. Figure 1 presents the calculated results of ionization cross sections for C_5, C_3F, and C_3F_4 with electron energy up to 1000 eV.

The calculated result for α/N in pure C_3F_8 gas at room temperature is compared with the experimental results by Moruzzi et al. [20], Hunter et al. [22] and Naidu et al. [24] in Figure 2. Generally, the present calculation

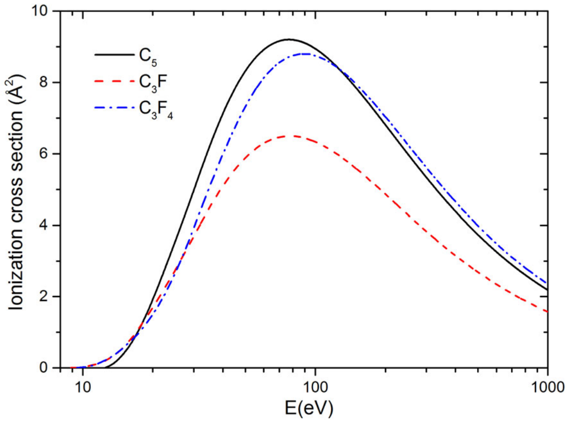

Fig. 1. Ionization cross sections of C_5, C_3F, and C_3F_4 calculated by DM method.

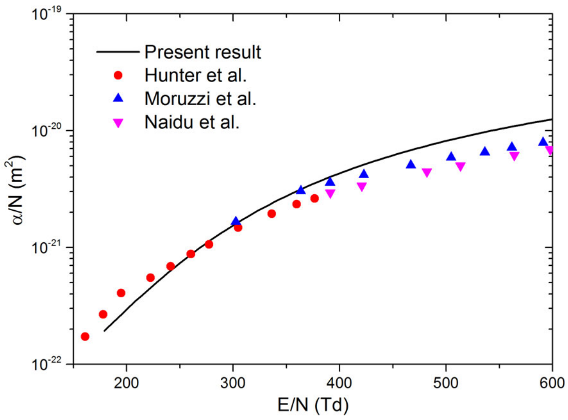

Fig. 2. Calculated values of α/N in pure C_3F_8 gas at room temperature as function of E/N and the comparison with the results measured by Moruzzi et al. [20], Hunter et al. [22] and Naidu et al. [24].

agrees well with the measurements. The departure at high values of E/N is probably due to the missing of high-energy threshold excitation cross sections which provide the high-energy dissipation channel and thus reduce the ionization coefficient at high values of E/N. In our calculation, the threshold of excitation cross sections is up to 7.5 eV as measured by Jeon [38]. Fortunately, this slight disagreement will not affect the final determination of the critical electric field strength because the breakdown of C_3F_8 gas usually occurs below 400 Td [36] at which the mean electron energy is below 7.0 eV according to our calculation.

3 Results and discussion for various cold C_3F_8 mixtures

3.1 Reduced ionization coefficient α/N of C_3F_8 mixtures

Figures 3–7 describe the calculated values of reduced ionization coefficient α/N in different C_3F_8 mixtures with CF_4, CO_2, N_2, O_2 and Ar respectively as function of reduced electric field strength E/N. Due to the acceleration of electrons in the electric field, ionization processes in C_3F_8 mixtures are enhanced with the increase of E/N, no matter whichever buffer gas is mixed. However, the dependence of α/N on buffer gas proportion is different for different mixtures. For C_3F_8-N_2 mixture as displayed in Figure 5, the value of α/N is generally reduced with the addition of N_2, while for the other mixtures, the opposite

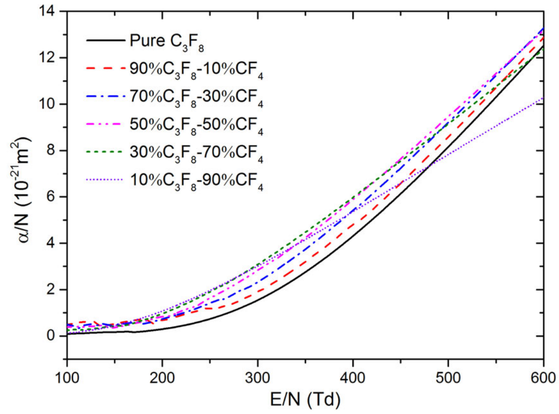

Fig. 3. α/N of C_3F_8-CF_4 mixtures with various buffer gas proportions as function of E/N.

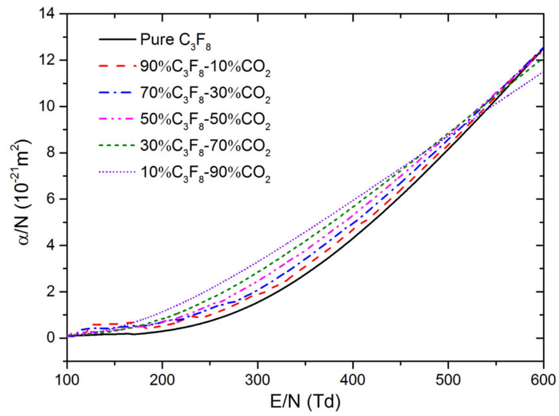

Fig. 4. α/N of C_3F_8-CO_2 mixtures with various buffer gas proportions as function of E/N.

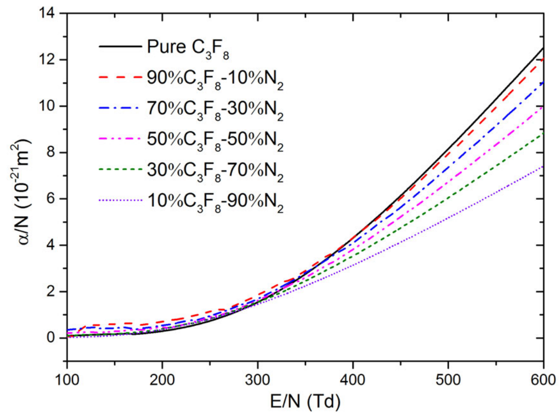

Fig. 5. α/N of C_3F_8-N_2 mixtures with various buffer gas proportions as function of E/N.

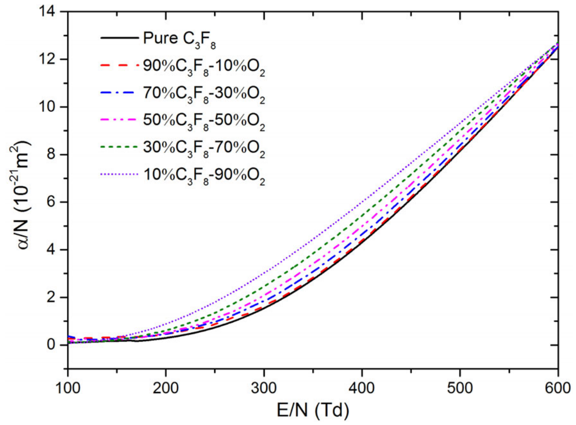

Fig. 6. α/N of C_3F_8-O_2 mixtures with various buffer gas proportions as function of E/N.

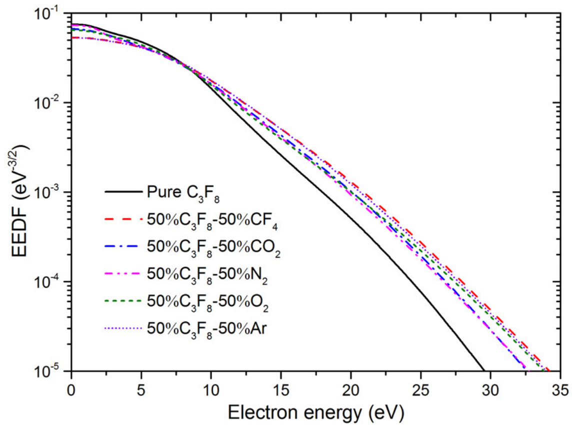

Fig. 8. EEDF of pure C_3F_8 gas and its mixtures with 50% buffer gas at E/N of 400 Td.

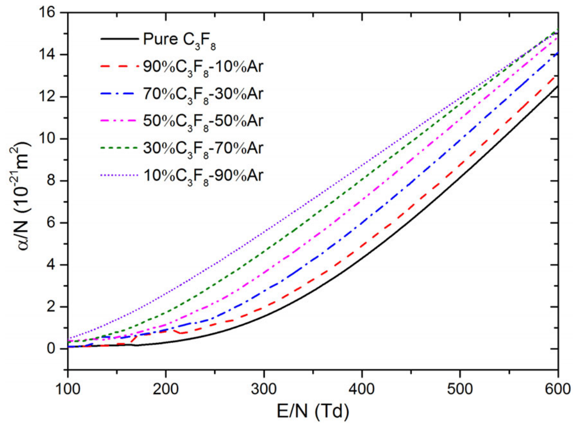

Fig. 7. α/N of C_3F_8-Ar mixtures with various buffer gas proportions as function of E/N.

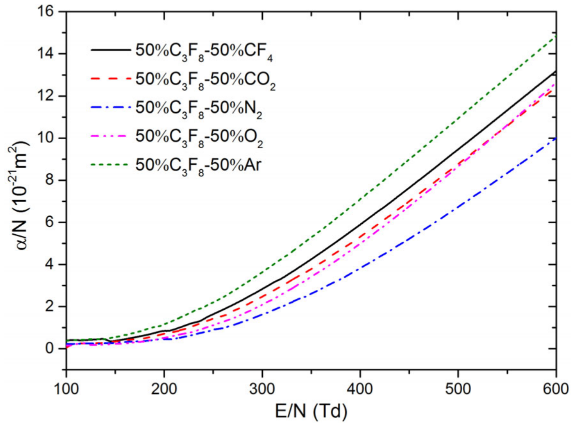

Fig. 9. α/N of different C_3F_8 mixtures with 50% buffer gas as function of E/N.

dependence on the buffer gas is observed. This can be attributed to the EEDF of mixtures together with the corresponding ionization cross sections. Figure 8 shows the EEDF of various C_3F_8 mixtures at E/N of 400 Td. The electrons having energy larger than 10 eV are referred as electrons with high energy because the ionization potential of the gases considered in the paper is larger than 10 eV. It is found that the pure C_3F_8 has the smallest amount of electrons with high energy and the C_3F_8-N_2 mixture ranks second. Therefore, although C_3F_8 [36] has the larger ionization cross section than CF_4, CO_2, O_2 and Ar [39–42], the pure C_3F_8 gas has the weaker ionization ability than its mixtures with such buffer gases. As for the C_3F_8-N_2 mixture, the balance between the EEDF and the ionization cross sections for C_3F_8 and N_2 leads to the different result in Figure 5.

In order to compare the different ionization performance of different mixtures, the α/N in various C_3F_8 mixtures is illustrated in Figure 9. Obviously, the ionization reactions in C_3F_8-N_2 are the poorest among the various mixtures due to the relatively low ionization cross section of N_2 and its high excitation cross sections which provide the extra energy dissipation channels and thus restrain the ionization processes.

3.2 Reduced electron attachment coefficient η/N of C_3F_8 mixtures

Figures 10–14 show the variation of reduced electron attachment coefficient η/N as function of E/N for C_3F_8-CF_4, C_3F_8-CO_2, C_3F_8-N_2, C_3F_8-O_2 and C_3F_8-Ar mixtures respectively. In contrast with α/N shown in Figures 3–7, the value of η/N drops in general with E/N because the electrons with higher energy gained through the acceleration in the higher electric field make the electron attachment to heavy particles more difficult. It is also seen that unlike α/N the dependence of η/N on buffer gas proportion behaves consistently for all the mixtures. With the increase of buffer gas percentage, the content of C_3F_8 which has the largest electron attachment cross section is reduced, and thus the η/N of mixtures falls significantly.

In addition, the values of η/N in different C_3F_8 mixtures with 50% buffer gas are presented in Figure 15 as function of E/N. It can be observed that the C_3F_8-CF_4 mixture is superior to the other mixtures in terms of electron attachment performance, and the C_3F_8-N_2 mixture comes in last. This is because CF_4 has the larger electron attachment cross section that the other buffer gases, and N_2 has no electron affinity (EA) [44] which means that it

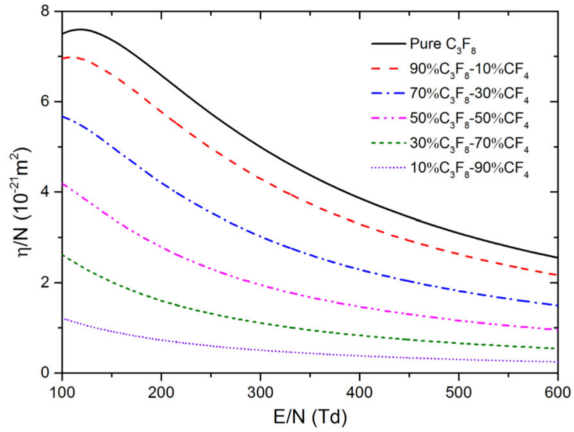

Fig. 10. η/N of C_3F_8-CF_4 mixtures with various buffer gas proportions as function of E/N.

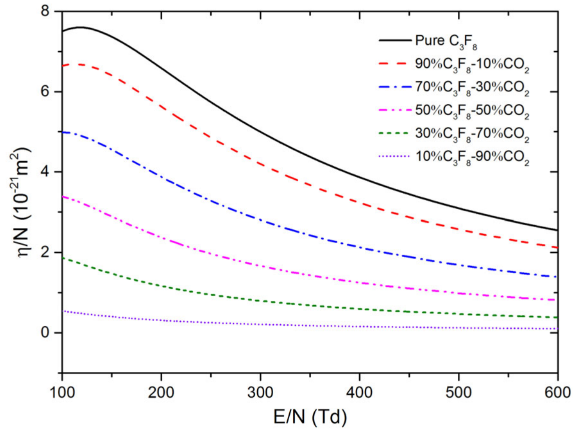

Fig. 11. η/N of C_3F_8-CO_2 mixtures with various buffer gas proportions as function of E/N.

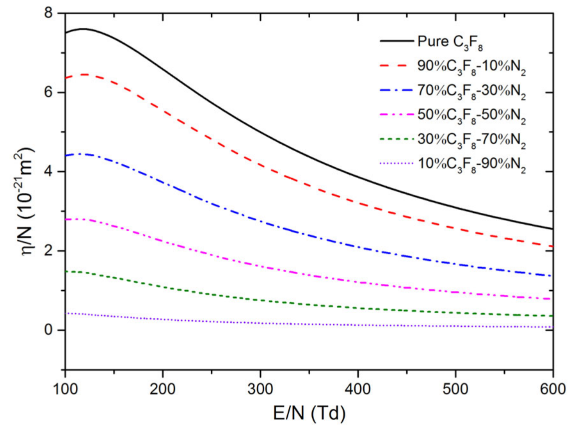

Fig. 12. η/N of C_3F_8-N_2 mixtures with various buffer gas proportions as function of E/N.

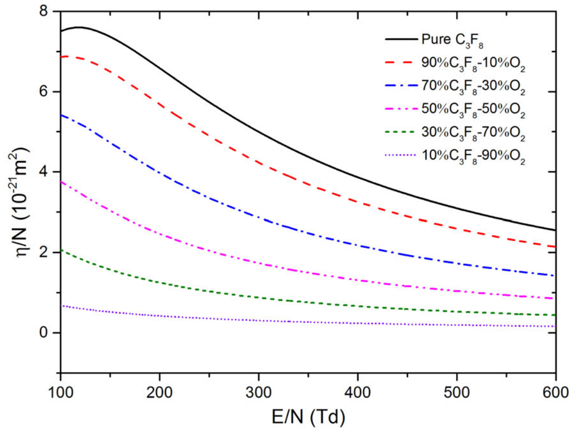

Fig. 13. η/N of C_3F_8-O_2 mixtures with various buffer gas proportions as function of E/N.

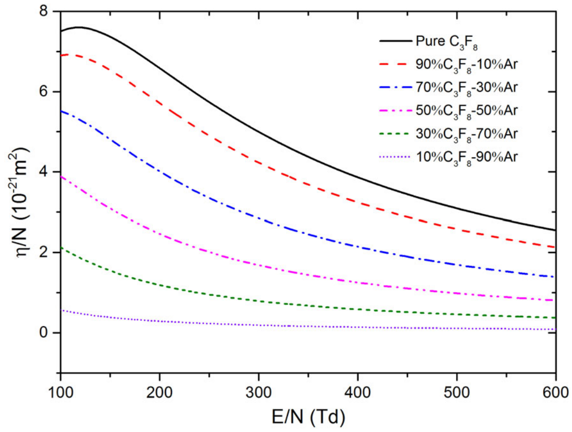

Fig. 14. η/N of C_3F_8-Ar mixtures with various buffer gas proportions as function of E/N.

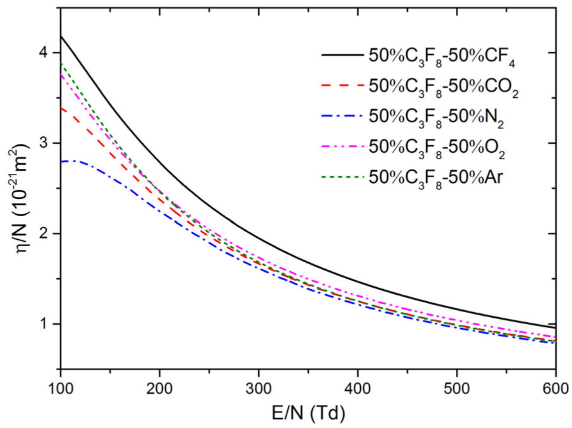

Fig. 15. η/N of different C_3F_8 mixtures with 50% buffer gas as function of E/N.

is impossible to get attached electrons. It is notable that Ar also has no positive EA [44] but it has larger threshold of excitation cross sections than N_2 [39–42]. This results in the larger η/N of C_3F_8-Ar than C_3F_8-N_2, especially at relatively low E/N, as shown in Figure 15.

3.3 Critical reduced electric field strength $(E/N)_{cr}$ of C_3F_8 mixtures

The critical reduced electric field strength $(E/N)_{\rm cr}$ which is the dielectric breakdown criterion is determined when the ionization of mixtures is completely balanced by electron attachment. This means that the effective ionization

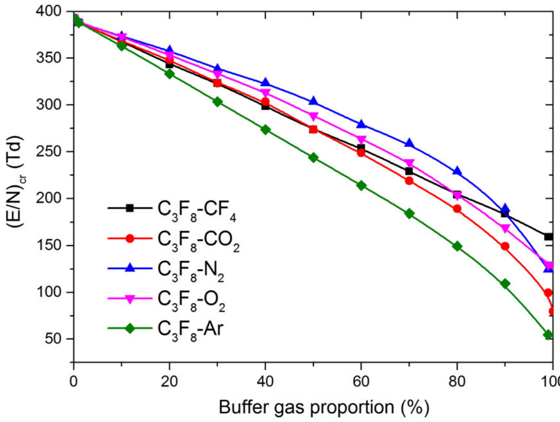

Fig. 16. $(E/N)_{\rm cr}$ of different C_3F_8 mixtures as function of buffer gas proportion.

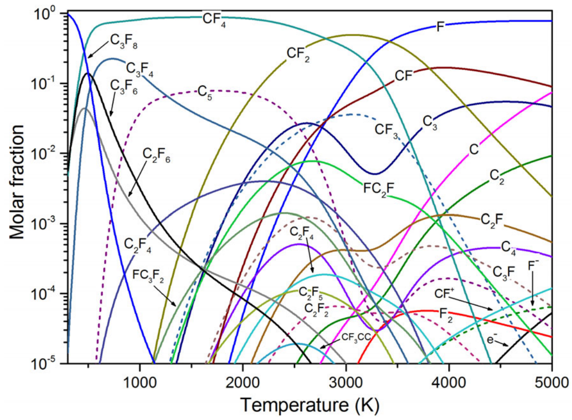

Fig. 17. Equilibrium composition of hot C_3F_8 gas in the temperature range of 300–5000 K at 0.4 MPa.

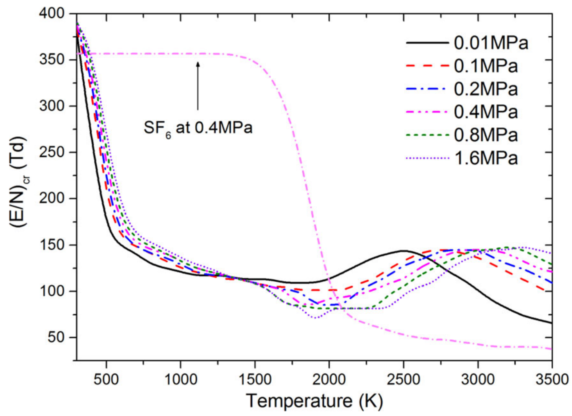

Fig. 18. $(E/N)_{\rm cr}$ of hot C_3F_8 gas at temperatures of 300–3500 K and various pressures with the comparison of $(E/N)_{\rm cr}$ of hot Sf$_6$ at 0.4 MPa.

coefficient $(\alpha - \eta)/N$ equals to zero. Once the determination of α/N and η/N, the $(\alpha - \eta)/N$ can be obtained accordingly.

The $(E/N)_{\rm cr}$ of various C_3F_8 mixtures is presented in Figure 16 as function of buffer gas proportion. It can be seen that the values of $(E/N)_{\rm cr}$ descend dramatically with the addition of buffer gas because of the reduction of C_3F_8 which has the better dielectric ability than the other gases considered in this paper. Also as found in Figure 16, the C_3F_8-Ar mixture is the poorest insulator no matter how much argon gas is mixed due to its much better ionization performance than the other mixtures as shown in Figures 7 and 9. Moreover, the C_3F_8-N_2 mixture is observed to have the largest $(E/N)_{\rm cr}$ when the buffer gas percentage is lower than 90% although the η/N of the C_3F_8-N_2 mixture is the lowest. This proves that the C_3F_8-N_2 mixture is the first choice as an insulating medium compared with other C_3F_8 mixtures.

4 Results and discussion for hot C_3F_8 gas

4.1 Equilibrium composition of hot C_3F_8 gas

As a necessary step to determine the dielectric breakdown properties of hot gases, the equilibrium compositions of hot C_3F_8 gas at temperatures of 300–5000 K and various pressures are calculated. The results are consistent with the previous work [45]. Figure 17 illustrates the result at 0.4 MPa. It is notable that when the temperature increases from room temperature, C_3F_8 starts to decompose quickly, mainly into CF_4, C_3F_4, C_3F_6, C_2F_6. In the temperature range of 500–2800 K, the dissociative product CF_4 dominates the mixture, while at temperatures of 2800–3400 K, the further dissociation leads CF_2 to become the key particle. With the further increase of temperature, the atomic F rules the mixture.

4.2 Critical reduced electric field strength $(E/N)_{\rm cr}$ of C_3F_8 gas

In order to determine whether C_3F_8 is a good arc quenching medium, the critical reduced electric field strength $(E/N)_{\rm cr}$ of hot C_3F_8 gas is calculated at different pressures and presented in Figure 18 with the comparison of that of SF_6 at 0.4 MPa [7]. Unfortunately, compared with SF_6, the $(E/N)_{\rm cr}$ of hot C_3F_8 gas falls sharply when the temperature increases from room temperature. This can be attributed to the fact that C_3F_8 decomposes above room temperature, while SF_6 start to decompose above 160 K [46]. The dielectric strength of the dissociative products of C_3F_8 (i.e. CF_4) is inferior to that of pure C_3F_8. This result indicates that although the cold C_3F_8 gas at room temperature can withstand strong electric filed strength, the hot C_3F_8 gas has much poorer dielectric performance than hot SF_6 when used as an arc quenching medium. Consequently, It might be impossible to replace SF_6 with C_3F_8 gas in the arc quenching apparatuses, such as HVCBs.

5 Conclusions

The dielectric properties, including the reduced ionization coefficient α/N, reduced electron attachment coefficient η/N and reduced critical electric field strength $(E/N)_{\rm cr}$, of cold C_3F_8 gas mixed with CF_4, CO_2, N_2, O_2

and Ar at room temperature are calculated using a two-term approximation of the Boltzmann equation. The dependence of such dielectric properties on reduced electric field strength E/N and buffer gas proportion is investigated. In order to reach the better dielectric performance, the value of α/N in the mixture should be lower and the η/N should be larger. Among the mixtures considered in this paper, the C_3F_8-N_2 mixture has the lowest α/N and the C_3F_8-CF_4 mixture has the largest η/N. Furthermore, the C_3F_8-N_2 mixture has the largest $(E/N)_{cr}$ which is determined when the value of α/N equals to η/N. Therefore, the C_3F_8-N_2 mixture is the best insulator in terms of breakdown strength compared with other C_3F_8 mixtures.

The equilibrium compositions of hot C_3F_8 gas at temperatures of 300–3500 K are calculated. The reduced critical electric field strength $(E/N)_{cr}$ of hot C_3F_8 at different pressures is determined and compared with that of hot SF_6 gas. It is found that the hot C_3F_8 gas has much poorer dielectric performance than hot SF_6 because the $(E/N)_{cr}$ of C_3F_8 decreases significantly above room temperature.

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