

The Prediction of the Dielectric Breakdown Properties of 6% C₄F₇N–94% CO₂ Mixtures at 300–4000 K and 0.1–3.2 MPa



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Abstract SF₆ is a greenhouse gas, therefore, we need to find a SF₆ substitute environment gases as an insulation and arc-quenching medium is an urgent for electrical engineer. C₄F₇N is a promising environmentally friendly insulating gas, while its boil temperature is high, therefore, CO₂ was added as a buffer gas. The dielectric strength of 6% C₄F₇N–94% CO₂ mixtures were evaluated at different pressure and temperature 0.1–3.2 MPa, and 300–4000 K. Firstly, the equilibrium compositions of 6% C₄F₇N–94% CO₂ mixtures at different gas pressures and temperatures up to 4000 K were calculated by the method of minimizing the Gibbs free energy. By Boltzmann equation analysis, the electron energy distribution function was obtained by the composition data. Finally, the critical reduced electric field ($(E/N)_{cr}$) of hot 6% C₄F₇N–94% CO₂ mixtures was determined, at which the rate of ionization is equal to attachment. The results show that in the gas temperature has an influence on 6% C₄F₇N–94% CO₂ mixing. The overall trend was that it first decreased with the increase of temperature, then rised, and finally falld. Further, the effect of barometric pressure on the $(E/N)_{cr}$ was also evident after 2000 K. The calculated results provided basic data for the post-arc breakdown of 6% C₄F₇N–94% CO₂, and have guiding significance for the engineering application of 6% C₄F₇N–94% CO₂.

Keywords Environmentally friendly insulating gas · Minimizing the Gibbs free energy · Boltzmann equation

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1 Introduction

SF₆ is mainly used for transmission and distribution equipment insulation and arc extinguishing in the power industry [1]. But given that SF₆ has an atmospheric lifetime of 3200 years (CO₂: 300–1000 years), in addition, its global warming potential (GWP) is 23,500 times greater than CO₂ [2]. Therefore, the realization of SF₆ free has become an important issue to be solved urgently for the green development of power grid. At present, the research on SF₆ alternative gases mainly focuses on traditional gases, SF₆ mixed gases and new environmental gases. Among them, new environment-friendly gases include CF₃I, C–C₄F₈, C₄F₇N, C₅F₁₀O and so on [3–6].

C₄F₇N has twice the insulation strength of SF₆ and its GWP is 2100 [7]. Its liquefaction temperature is about –4.7 °C, which is high, and the buffer gas is usually injected to meet the requirements of minimum operating temperature in engineering equipment, therefore, it is considered to be the most potential SF₆ alternative gas. Zhang et al. studied the breakdown strength and partial discharge characteristics of the C₄F₇N/CO₂ mixtures at alternating current voltages, and compared it with SF₆ [8]. Li et al. performed a breakdown experiment on C₄F₇N/N₂ gas mixture and measured the decomposition products using spherical electrodes at alternating current voltage [9]. The results shown that the main products were CF₄, C₂F₆, C₃F₈, CF₃CN, C₂F₄, C₃F₆ and C₂F₅CN after thirty times of breakdowns and the CF₄, C₂F₆ and CF₃CN contents were the highest. Li et al. analyzed the saturated vapor pressure characteristics of C₄F₇N-CO₂ mixtures based on the Antoine constants and the Antoine equation [10]. Then they discussed the application conditions at limits of environment temperature. Moreover, Zhang et al. first evaluated the electron neutral collision cross section of C₄F₇N. The set was verified by systematic comparison of Boltzmann equation analysis and experimental measurements of pure C₄F₇N, C₄F₇N/N₂ and C₄F₇N/Ar mixtures [11]. Under partial discharge (PD) and spark discharge conditions, the experimental values of decomposition products of C₄F₇N/CO₂ and C₅F₁₀O/air mixtures were also first presented by them. Then, ab initio molecular dynamics simulations of typical decomposition products were performed. In addition, the reliability of ab initio molecular dynamics simulation method in simulating electron induced ionization was verified [12]. GE, ABB and other companies have developed C₄F₇N as insulation medium in GIL, GIS and other products, and realized engineering application in Europe [13].

The dielectric properties of 6% C₄F₇N–94% CO₂ mixtures at 0.1–3.2 MPa and 300–4000 K were studied. First, by using the minimizing the Gibbs free energy, the equilibrium compositions of 6% C₄F₇N–94% CO₂ mixtures under different thermal states and gas pressure were calculated. By solving two-term Boltzmann equations, the electron energy distribution function (EEDF) was obtained. The value of the critical reduced electric field strength $(E/N)_{cr}$ was the electric field value corresponding to the reduced effective ionization coefficient $\alpha_{eff}/N = 0$. Finally, the $(E/N)_{cr}$ of 6% C₄F₇N–94% CO₂ mixture was determine. The calculated results provided basic data for the post-arc breakdown of 6% C₄F₇N–94% CO₂, and have guiding significance for the engineering application of 6% C₄F₇N–94% CO₂.

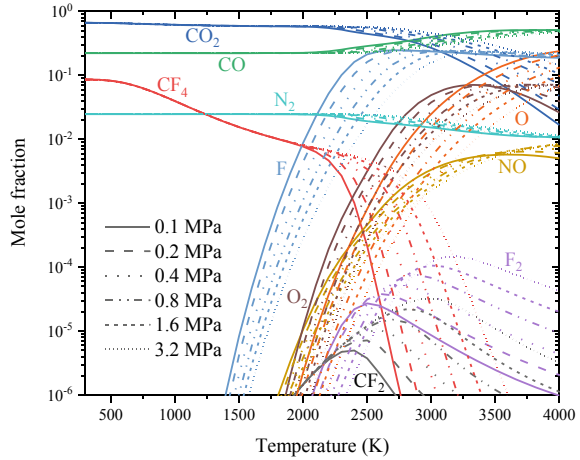
2 The Method of Calculation

Since the increase of gas temperature will lead to great variations in the total particle number density and gas composition, the dielectric breakdown methods of cold and hot gases are very different. The insulation properties of 6% C₄F₇N–94% CO₂ mixtures are evaluated by calculating the $(E/N)_{cr}$. The value of the $(E/N)_{cr}$ is the electric field (E/N) value corresponding to the effective ionization coefficient equals to zero. This means that the number of electrons produced by the collision ionization is balanced by the number of electrons lost by the collision attachment reaction. EEDF is important for obtaining ionization and attachment coefficients [14]. In this paper, Boltzmann equation is simplified by 2-term spherical harmonic approximation.

By minimizing the Gibbs free energy [15], the equilibrium composition of 6% C₄F₇N–94% CO₂ mixture at 0.1–3.2 MPa and 300–4000 K is calculated. The change of the composition of the 6% C₄F₇N–94% CO₂ mixture as gas temperature at different gas pressures is shown in Fig. 1. Ten species in 6% C₄F₇N–94% CO₂ mixtures are found, which concentrations are over 10⁻⁶, at the temperature range of 300–4000 K. There are CO₂, CO, CF₄, N₂, O₂, O, F, CF₂, NO and F₂, and is no C₄F₇N in the calculated component, which is due to the small amount of C₄F₇N, which will be quickly decomposed or react with other products. Among them, a large amount of CO, CF₄ and N₂ will be generated. Below 2000 K, the contents of CO₂, CO and N₂ are almost unchanged, and the content of CF₄ slowly declines with the increase of temperature. And, at this point, the content of these four gases does not change with the pressure. After 1500 K, F is decomposed, and the content of F also increases rapidly with the increase of temperature. Moreover, at the same temperature, the content of F decreases with the increase of air pressure. After 3000 K, the content is almost unchanged and does not change with the change of air pressure. At about 2000 K, O₂, O, CF₂, NO and F₂ increased with the increase of temperature. The content of O₂ is almost constant after 3000 K and does not change with the change of air pressure. When the temperature reaches a certain level, the contents of CF₂ and F₂ begin to decline. The change of components with temperature is very important for the calculation of the $(E/N)_{cr}$.

Solving the Boltzmann equation requires cross sections of all components in 6% C₄F₇N–94% CO₂ mixtures. Cross sections for all components (CO₂, CO, CF₄, N₂, O₂, O, F, CF₂, NO and F₂) are taken from the Lxcat website [16].

Fig. 1 Mole fractions of 6% C_4F_7N -94% CO_2 mixture with gas temperature at 0.1–3.2 MPa



3 Results and Discussion

3.1 α_{eff} and the $(E/N)_{cr}$ in 6% C_4F_7N -94% CO_2 Mixture at Room Temperature

As the electron kinetics model is adopted, the calculated $(E/N)_{cr}$ is independent of pressure, and the displayed α_{eff} is also independent of pressure. The α_{eff} of 6% C_4F_7N -94% CO_2 at room temperature is shown in Fig. 2. At lower field strengths (< 40 Td), α_{eff} is almost zero. At 40–80 Td, α_{eff} is less than 0 and decreases as E/N increases. At larger electric field, α_{eff} increases with E/N increasing. Moreover, when $E/N = 99.71$ Td, $\alpha_{eff} = 0$, which is the $(E/N)_{cr}$ of 6% C_4F_7N -94% CO_2 at room temperature.

3.2 α_{eff} and the $(E/N)_{cr}$ in 6% C_4F_7N -94% CO_2 Mixtures at High Temperature

Figure 3 shows that α_{eff} of in 6% C_4F_7N -94% CO_2 mixture at 1.6 Mpa and different gas temperatures. The overall trend of α_{eff} is almost constant at lower E/N , then decreasing and then increasing. At 1000–2000 K, the value of α_{eff} increases with the increase of temperature when the E/N is higher, so the $(E/N)_{cr}$ decreases with the increase of temperature. When the temperature is between 3000 and 4000 K, α_{eff} at 3000 K is less than that at 4000 K before 80 Td. And the $(E/N)_{cr}$ for 3000 K is less than that of 4000 K.

α_{eff} of in 6% C_4F_7N -94% CO_2 mixture at 3000 K and different gas pressures is shown in Fig. 4. As the electric field becomes larger, α_{eff} first drops and then rises.

Fig. 2 α_{eff} of in 6% $\text{C}_4\text{F}_7\text{N}$ -94% CO_2 mixture at room temperature and different gas pressures

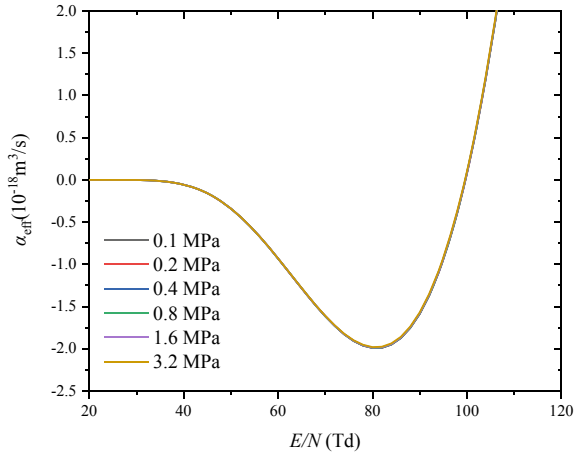
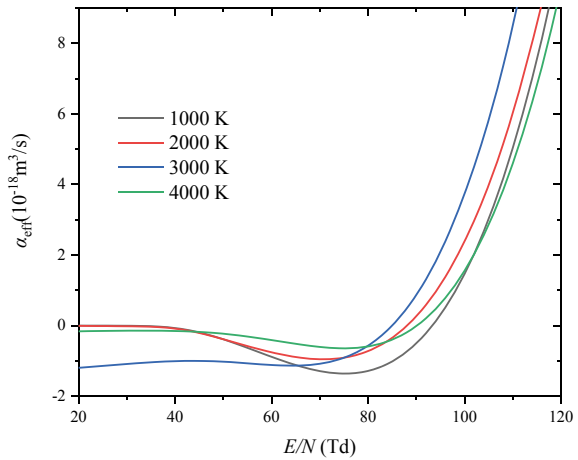


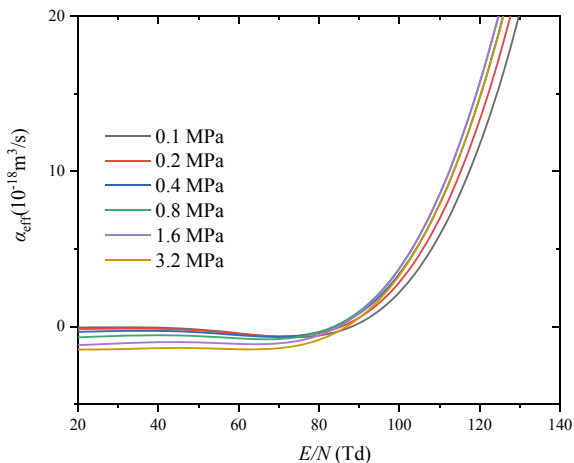
Fig. 3 The α_{eff} of in 6% $\text{C}_4\text{F}_7\text{N}$ -94% CO_2 mixture at 1.6 MPa and different gas temperatures



α_{eff} decreases with increasing air pressure before 80 Td. After 80 Td, α_{eff} increases with increased air pressure. At 3000 K, the $(E/N)_{\text{cr}}$ varies between 80 and 90 Td, but with little regularity. This is due to the different ionization and attachment rates of the decomposition products in $\text{C}_4\text{F}_7\text{N}$ - CO_2 mixture.

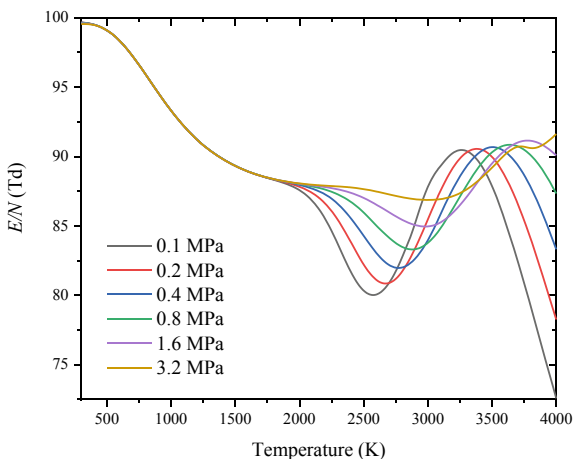
Except for 3.2 MPa, the overall trend of the $(E/N)_{\text{cr}}$ under other gas pressure is roughly the same, first decreasing, then increasing, and then decreasing with the increase of temperature. Before 2000 K, $(E/N)_{\text{cr}}$ does not change with the change of gas pressure, but decreases slowly with the increase of temperature. After 2000 K, $(E/N)_{\text{cr}}$ first decreases, then increases and then decreases with the increase of temperature. With the increase of pressure, the variation trend of $(E/N)_{\text{cr}}$ with temperature slows down, and the temperature at the inflection point of $(E/N)_{\text{cr}}$ rising and falling

Fig. 4 Effective ionization coefficient α_{eff} of in 6% $\text{C}_4\text{F}_7\text{N}$ -94% CO_2 mixture at different gas pressures and 3000 K



increases. When the pressure is 3.2 MPa, it is completely consistent with other pressure values and trends before 2000 K. With the further increase of temperature, it first continues to decline until the temperature reaches 3000 K, then rises, then falls and rises again. These trends are mainly caused by the formation of O_2 , CF_2 , F_2 and NO at 2000 K, and the rapid decline of F_2 and CF_2 at 2500–3000 K (Fig. 2).

Fig. 5 $(E/N)_{\text{cr}}$ in hot 6% $\text{C}_4\text{F}_7\text{N}$ -94% CO_2 mixtures at 0.1–3.2 MPa



4 Conclusions

The dielectric breakdown properties of 6% C₄F₇N–94% CO₂ mixtures at 0.1–3.2 MPa and 300–4000 K were studied in this paper. By using the minimizing the Gibbs free energy, the equilibrium compositions of 6% C₄F₇N–94% CO₂ mixtures under different thermal states and gas pressure were calculated. Then, by solving two-term Boltzmann equations, the EEDF was obtained. The value of the $(E/N)_{cr}$ was the electric field value corresponding to the $(\alpha-\eta)/N = 0$. Except for 3.2 MPa, the overall trend of the $(E/N)_{cr}$ under other gas pressure is roughly the same, first decreasing, then increasing, and then decreasing with the increase of temperature. Finally, the $(E/N)_{cr}$ of 6% C₄F₇N–94% CO₂ mixture was determine. The calculated results provided basic data for the post-arc breakdown of 6% C₄F₇N–94% CO₂, and have guiding significance for the engineering application of 6% C₄F₇N–94% CO₂.

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