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Citation: *Journal of Applied Physics* **117**, 143302 (2015); doi: 10.1063/1.4917210

View online: <http://dx.doi.org/10.1063/1.4917210>

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Prediction of the critical reduced electric field strength for carbon dioxide and its mixtures with copper vapor from Boltzmann analysis for a gas temperature range of 300 K to 4000 K at 0.4 MPa

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(Received 15 January 2015; accepted 29 March 2015; published online 10 April 2015)

The influence of copper vapor mixed in hot CO₂ on dielectric breakdown properties of gas mixture at a fixed pressure of 0.4 MPa for a temperature range of 300 K–4000 K is numerically analyzed. First, the equilibrium composition of hot CO₂ with different copper fractions is calculated using a method based on mass action law. The next stage is devoted to computing the electron energy distribution functions (EEDF) by solving the two-term Boltzmann equation. The reduced ionization coefficient, the reduced attachment coefficient, and the reduced effective ionization coefficient are then obtained based on the EEDF. Finally, the critical reduced electric field $(E/N)_{cr}$ is obtained. The results indicate that an increasing mole fraction of copper markedly reduces $(E/N)_{cr}$ of the CO₂–Cu gas mixtures because of copper's low ionization potential and large ionization cross section. Additionally, the generation of O₂ from the thermal dissociation of CO₂ contributes to the increase of $(E/N)_{cr}$ of CO₂–Cu hot gas mixtures from about 2000 K to 3500 K. © 2015 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4917210>]

I. INTRODUCTION

Electric power switchgear equipment, which is designed to interrupt normal load current and short circuit current, is particularly important in ensuring the safety and reliability of the power system.¹ Copper and its alloys are widely used in contacts on account of their excellent thermal and electrical conductivity.² Copper vapor inevitably enters the arcing chamber due to ablation of the contacts, and it may change the composition and properties of the arc and hot gas in the post-arc period.³ However, the influence of copper vapor on the interruption capability and dielectric breakdown properties is still not clear.¹

SF₆ has excellent insulation and arc-quenching capabilities. Since the 1960s, it has been widely used in high voltage circuit breakers. However, since SF₆ is a powerful greenhouse gas (its global warming potential is 23 900 times higher than CO₂)⁴ and is hard to degrade, its use has gradually been subjected to restrictions. As a result, there is increasing urgency to seek an environmentally friendly gas that can partially or wholly replace SF₆.

To deal with this problem, research directed towards seeking substitutes for pure SF₆ has been widely carried out. Current research is mainly focused on mixtures of SF₆ with gases such as CF₄,⁵ C₃F₈,⁶ N₂O,⁷ CF₃I,⁸ and on pure gases such as N₂, CO₂, and dry air. In recent years, CO₂ has attracted wide interest and extensive study due to its good interruption performance and its much lower global warming potential than SF₆. Tanaka⁹ calculated the thermodynamic and transport properties of CO₂, CO₂–O₂, and CO₂–H₂ mixtures for a temperature range of 300 to 30 000 K and at a

pressure range of 0.1 to 10 MPa. Uchii¹⁰ investigated the fundamental properties of CO₂ gas and its mixtures as an arc interruption medium in high-voltage gas circuit breakers (HV GCBs). Stoller¹¹ investigated CO₂, compared with air and SF₆, as an arc quenching and insulating medium for power equipment both theoretically and experimentally. Indeed, an HV GCB that uses CO₂ as the medium to extinguish the arc has been manufactured recently.

After current interruption, hot gases remain between the two electrodes in the arcing chamber at temperatures of several thousand kelvin.¹² The dielectric properties of hot gases differ from those of gases at room temperature. The trend of miniaturization of various electric power switchgear equipment sets higher requirements of the post-arc interruption capability. Most previous studies of gas dielectric breakdown properties have concentrated on room temperature values.^{13–16} For these reasons, it is important to study and better understand the dielectric properties of hot gas mixtures.

Studies on the influence of copper vapor on the properties of the arc and the interruption performance are helpful in developing a good understanding of the arc plasma and in improving the interruption capability.¹² Most previous work concentrated on the thermodynamic properties,⁹ transport coefficients,^{9,17} and radiation properties¹⁸ of different gases with copper vapor. However, there has been insufficient investigation of the influence of copper on the dielectric properties of CO₂ mixed with copper vapor.

In addition to the copper vapor, several factors can affect the electrical breakdown process during post-arc period under the action of the recovery voltage. Examples are the pressure and temperature distributions, the gas properties, and the electric field distribution, which is affected by the surface conditions and the motion of the electrodes. In the

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present work, we focus on the influence of the copper vapor concentration on the critical reduced electric field $(E/N)_{cr}$, which is fundamental data required for the evaluation and prediction of the electric breakdown characteristics.

Electric power switchgear equipment usually adopts several atmospheres as its gas pressure. Thus, in the present work, we choose the fixed pressure of 0.4 MPa to study the influence of copper vapor on dielectric breakdown properties of hot CO₂ at the temperature range from 300 K to 4000 K. In the first part, equilibrium compositions of hot CO₂ with different copper fractions were calculated by the method of minimization of the Gibbs free energy under the assumption of local thermodynamic equilibrium. Then, by Boltzmann analysis, the influence of copper vapor on the dielectric breakdown properties of hot CO₂ gas was numerically predicted, by combining analysis of the electron energy distribution function (EEDF), the reduced ionization coefficient, the reduced attachment coefficient, and the reduced effective ionization coefficient.

II. CALCULATION METHOD AND PARAMETERS

The dielectric breakdown properties of hot gas differ from those of cold gas on account of the changes in the total number density, the gas composition, and the energy exchange rate caused by increasing temperature. Considering these differences, the present calculation predicts the dielectric breakdown property of hot CO₂ with copper in the following procedure: (1) Calculation of equilibrium compositions of hot CO₂ with copper; (2) computation of the electron energy distribution functions; (3) prediction of critical electric field strength.

In the present work, the computation of equilibrium compositions of the hot gas CO₂ with copper metallic vapor in the temperature range from 300 K to 4000 K and at a pressure of 0.4 MPa was performed with a method based on the mass action law, based on the assumption of equal electron and heavy-particle temperatures.¹⁹ The main species within the temperature range we considered are the following 6 gaseous species: CO₂, CO, O₂, O, Cu, and CuO. The data needed for the calculation were obtained from Ref. 20.

The electron energy plays an important role in the overall behavior of discharges, and the EEDF is the fundamental data for calculating the ionization and attachment coefficients. The EEDFs in hot gas mixtures were obtained by Boltzmann analysis. The method of solving the Boltzmann equation was introduced in the authors' previous work.^{5,21} In the present calculation, the influence of copper vapor on the EEDF and as a consequence on the critical reduced electric field $(E/N)_{cr}$ was quantitatively analyzed.

We neglected the effects of copper in the solid and liquid phases on the EEDF.¹² The fractional copper content X_{Cu} is defined as the sum of mole fractions of copper in all three states divided by the sum of mole fractions of all species

$$X_{Cu} = \frac{x_{Cu(l,s)} + x_{Cu}^g}{x_{Cu(l,s)} + \sum x_s^g},$$

in which, $x_{Cu(l,s)}$ is mole fractions of copper in the liquid and solid phases, x_{Cu}^g is mole fraction of copper in the gas phase,

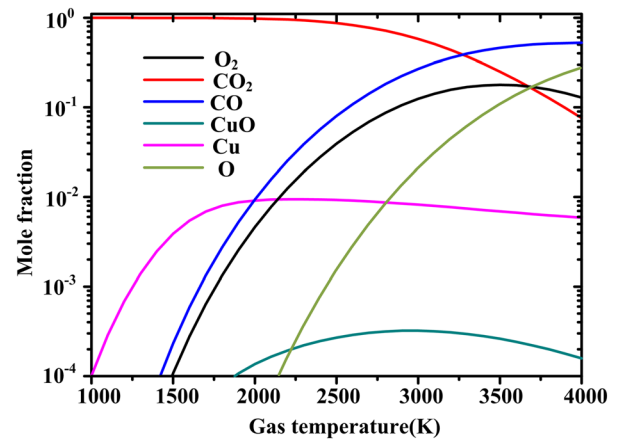


FIG. 1. Composition of hot 99%CO₂ with 1%Cu atoms considering the presence of CuO as a function of gas temperature at 0.4 MPa.

and $\sum x_s^g$ is the sum of mole fractions of all species in the gas phase.

Equilibrium compositions of hot 99%CO₂ with 1%Cu atom are shown in Fig. 1. Above 1700 K, the copper in the solid phase vaporizes completely. The mole fraction of CuO is in the range from 1×10^{-4} to 3.2×10^{-4} , while that of Cu is in the range from 5.9×10^{-2} to 9.4×10^{-2} . Therefore, compared with Cu, the effect of CuO is very small, and neglecting the presence of CuO will not introduce large errors.

The critical reduced electric field strength $(E/N)_{cr}$ is an important index to evaluate the dielectric breakdown properties in a gas or gaseous mixture. It is closely related to effective ionization coefficient, which is defined as the difference between the total ionization coefficient and the total attachment coefficient. A zero effective coefficient corresponds to the critical value for the dielectric breakdown phenomenon.

The electron impact cross sections for all the relevant species, namely, CO₂, O₂, CO, O, and Cu, are necessary to solve the Boltzmann equation. The electron impact cross sections for O,^{22,23} O₂,²⁴ CO₂,²⁵ CO,²⁶ and Cu¹² were taken from the literature, and are shown in Figures 2–6.

The calculated reduced effective ionization coefficient $(\alpha-\eta)/N$ in CO₂ at room temperature is shown in Fig. 7 with

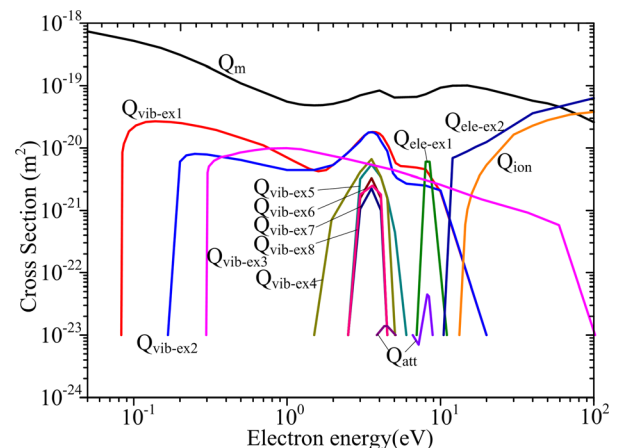


FIG. 2. Cross sections for CO₂.²⁵

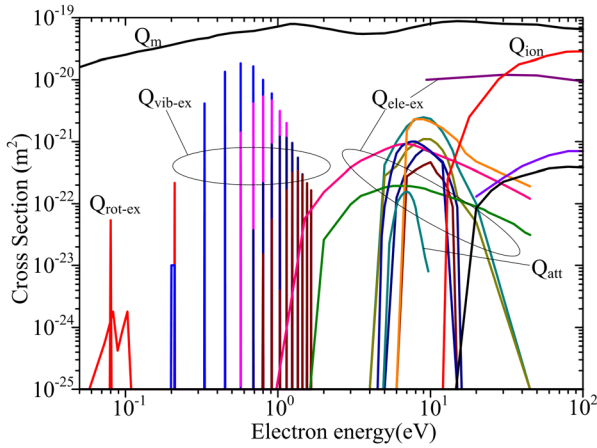


FIG. 3. Cross sections for O_2 .²⁴

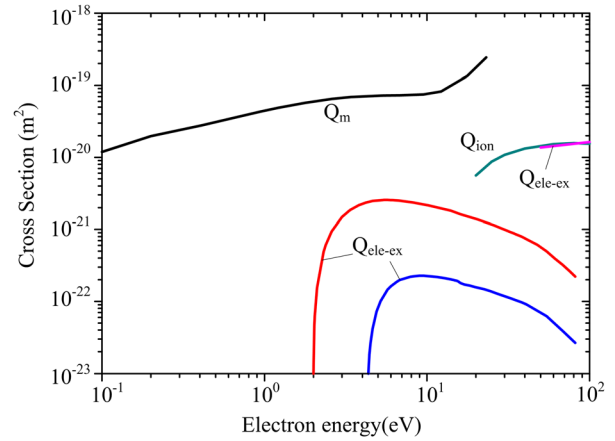


FIG. 6. Cross sections for O .^{22,23}

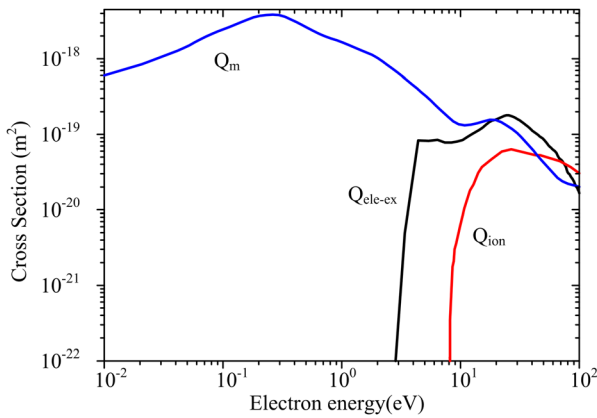


FIG. 4. Cross sections for Cu .¹²

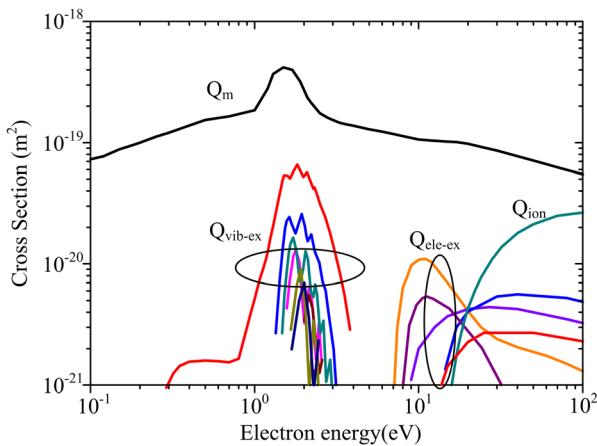


FIG. 5. Cross sections for CO .²⁶

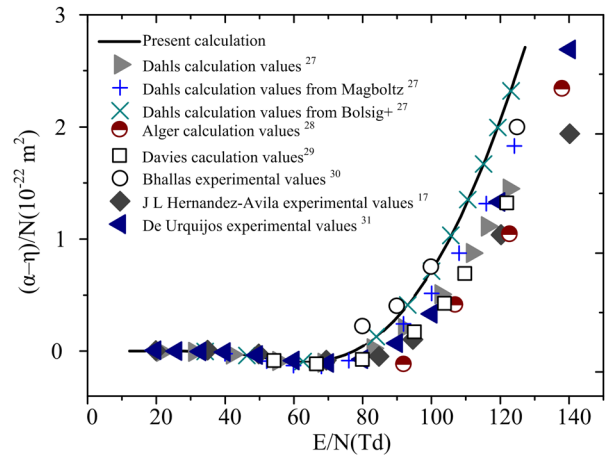


FIG. 7. Reduced effective ionization coefficient $(\alpha-\eta)/N$ of CO_2 as a function of E/N at room temperature, with results from the literature plotted for comparison.

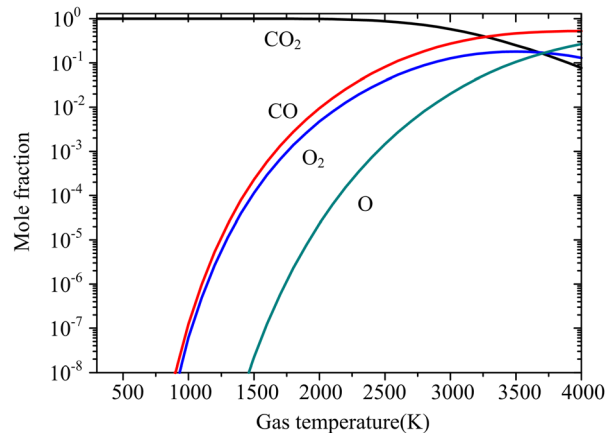


FIG. 8. Composition of hot CO_2 as a function of gas temperature at 0.4 MPa.

data from the literature plotted for comparison.^{17,27-31} Good agreement is found between the calculated and literature values for CO_2 .

III. SPECIES COMPOSITION OF HOT CO_2 WITH COPPER

Fig. 8 shows the composition of pure CO_2 in the temperature range from 300 K to 4000 K at 0.4 MPa. This figure indicates that the mole fractions of 4 species, namely, CO_2 ,

O_2 , CO , and O , do not change significantly at temperatures below 2500 K. CO_2 begins to dissociate into smaller particles, namely, CO and O_2 , around 2500 K. With a further increase of temperature, the mole fractions of CO and O increase to reach maximum values of about 52% and 26% at 4000 K, respectively. The mole fraction of O_2 increases to 18% at about 3500 K and drops slightly to 13% at 4000 K.

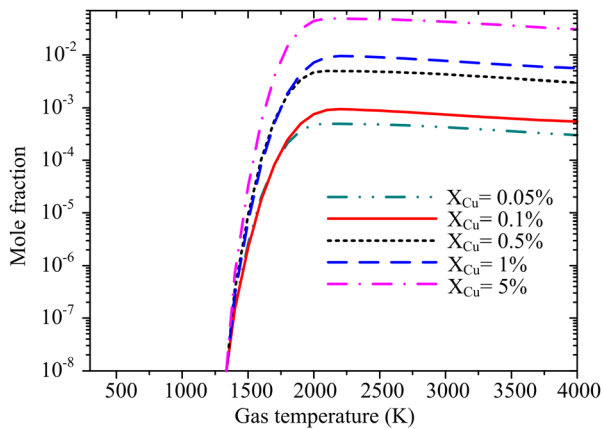


FIG. 9. Mole fraction of Cu atoms in the gas phase in hot CO₂ with different fractional Cu contents at a pressure of 0.4 MPa.

The mole fraction of copper atoms in the gas phase is shown in Fig. 9 for different X_{Cu} . It is concluded that mole fraction of Cu increases markedly from about 1300 K until its complete evaporation at around 2000 K. After that, because of the dissociation of CO₂, smaller particles, namely, CO, O₂, and O, are produced. Therefore, a slight decrease of the atomic copper mole fraction occurs as the temperature increases further to 4000 K.

Fig. 10 shows the composition of a hot gas mixture, neglecting the presence of CuO, at 0.4 MPa which can be obtained from the data shown in Figs. 8 and 9 by normalization. The effect on mole fractions of the species derived from CO₂ of adding 1% Cu is minimal, but even a small amount of copper can make a considerable difference to the dielectric breakdown properties of gas mixtures.

IV. EEDF IN HOT CO₂ WITH Cu

Fig. 11 illustrates the calculated EEDFs in a hot 99% CO₂-1% Cu gas mixture at an E/N of 100 Td for different gas temperatures. The EEDFs below 2500 K do not change markedly on account of the nearly constant compositions, as shown in Fig. 10. Above 2500 K, the EEDFs vary strongly with an increase of temperature. The values of the EEDF for electron energy below 1.75 eV increase strongly with the

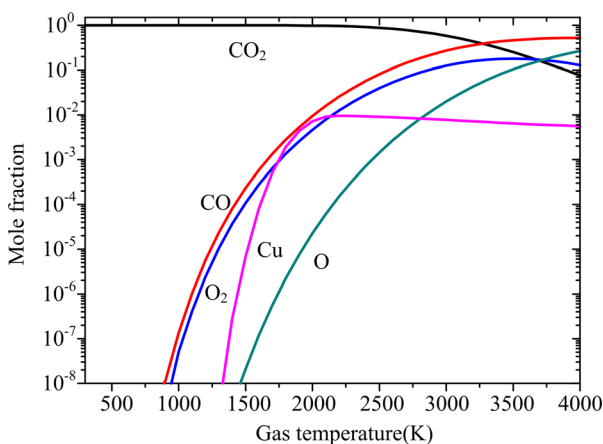


FIG. 10. Composition of hot 99%CO₂ with 1%Cu atoms neglecting the presence of CuO as a function of gas temperature at 0.4 MPa.

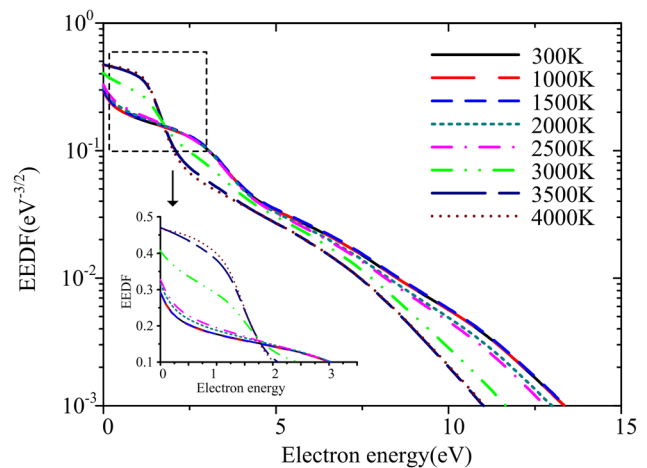


FIG. 11. Electron energy distribution function (EEDF) in hot CO₂ with 1% Cu atoms for different gas temperatures at 0.4 MPa and 100 Td.

growth of temperature, as shown in the inset of Fig. 11. Above 1.75 eV, in contrast, raising gas temperature causes a significant reduction in the EEDF. These can be attributed to the generation of CO. As shown in Figs. 2–6, CO has larger vibrational excitation cross sections than other species, which leads to efficient decay of the electron kinetic energy through frequent excitation collisions.

The influence of X_{Cu} on the EEDF in hot CO₂ and Cu gas mixtures is shown in Fig. 12. There are only small differences between the EEDF curves for different Cu concentrations. With an increase of X_{Cu} , however, the EEDF at energies below 3.7 eV rises, while the EEDF at energies below 3.7 eV decreases, as shown in the inset of Fig. 12. An increasing mole fraction of copper causes the average electron energy of the gas mixture to be reduced slightly. Compared with other species, Cu has a larger excitation cross section for electron impact, as shown in Figs. 2–6, which governs the probability of excitation collision between electron and heavy particles. Thus, electrons are more likely to collide with Cu atom and lose part of their energy.

As shown in Fig. 13, the EEDFs in hot CO₂ with 1% Cu at a fixed temperature of 2000 K and at a pressure of 0.4 MPa vary with E/N. The value of the EEDF at fixed electron

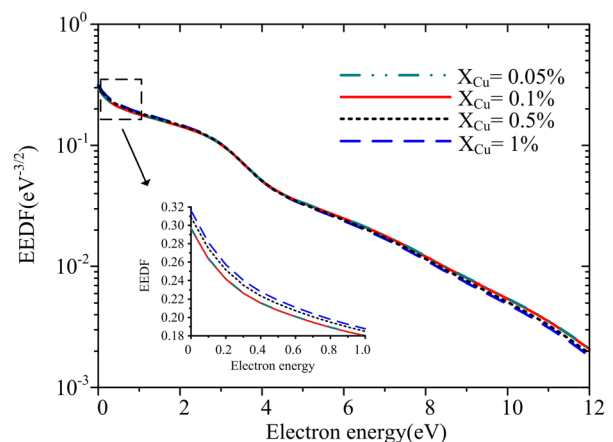


FIG. 12. Electron energy distribution function (EEDF) in hot CO₂ with different fractional Cu contents for 2000 K at 0.4 MPa and 100 Td.

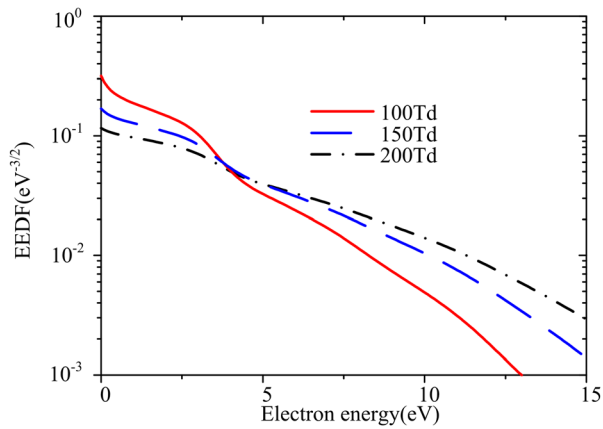


FIG. 13. Electron energy distribution function (EEDF) in hot 99% CO₂-1% Cu gas mixture for different E/N at 0.4 MPa and 2000 K.

energy is decreased by increasing E/N for electron energies below 4.4 eV, and the EEDF is increased for larger electron energies. This means that increasing E/N greatly increases the proportion of high kinetic energy electrons, due to acceleration of electrons by the high E/N.

V. REDUCED IONIZATION AND ATTACHMENT COEFFICIENTS IN HOT CO₂ WITH COPPER

In this section, the influence of gas temperature and mole fraction of Cu on the reduced ionization coefficients, the attachment coefficients, and the effective ionization coefficients of CO₂-Cu gas mixture is discussed.

A. α/N , η/N , and $(\alpha-\eta)/N$ in a hot 99% CO₂-1% Cu mixture

As shown in Fig. 14, for temperatures below 1500 K, there is little dependence on temperature of the reduced ionization coefficients α/N of a 99% CO₂-1% Cu gas mixture. As temperature increases above 1500 K, the copper begins to be evaporated into the gas phase as shown in Fig. 10, and affects α/N significantly. The reduced ionization coefficient for 2000 K is substantially higher, as a result of the higher

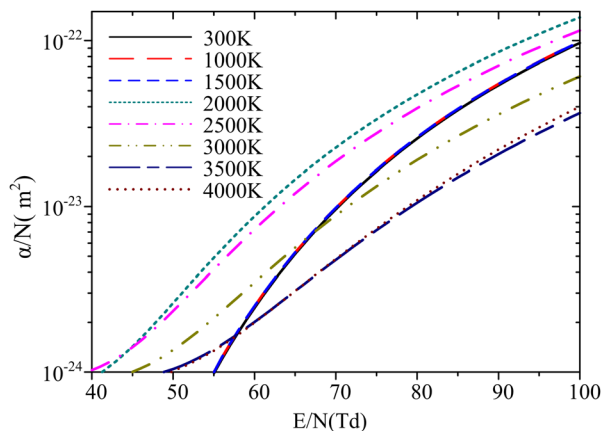


FIG. 14. Reduced ionization coefficient α/N in hot CO₂ with 1% Cu atoms as a function of E/N for different gas temperatures at 0.4 MPa.

TABLE I. Ionization potential of the main species in CO₂-Cu mixtures.

Species	CO ₂	CO	O	O ₂	Cu
P _{ion} (eV)	13.30	14.20	13.62	12.06	7.73

Cu mole fraction. The ionization potential of Cu is 7.73 eV and much lower than other species considered, as shown in Table I. As the gas temperature increases further up to 3500 K, there is a clear reduction of α/N . This phenomenon results from the generation of CO and O₂ for the following reasons. On the one hand, as stated earlier, the average electron energies for 2500 K or higher temperature are much lower than those for temperatures below 2500 K because of the large vibrational excitation cross sections of CO. On the other hand, the ionization cross sections of CO and O₂ are clear smaller than CO₂, which makes the ionization reactions less likely to occur above 2500 K. For the temperature range from 3500 K to 4000 K, mole fractions of Cu and CO do not change much, so the two curves of α/N for 3500 K and 4000 K are very close to each other.

The reduced attachment coefficient η/N in hot CO₂ with 1% Cu atoms is presented as a function of E/N for different gas temperatures in Fig. 15. Generally, an increasing temperature leads to a slight reduction of η/N for the temperature range from 2000 K to 4000 K and at E/N below 80 Td. However, the growth of η/N curves for 3000 K to 3500 K outpaces those of other temperatures over 80 Td. Because of the thermal dissociation of O₂, we see that mole fraction of O₂ drops slightly at 4000 K in Fig. 10. The dissociation of O₂ decreases the electron attachment efficiency of the mixture.

Based on the calculated α/N and η/N , the reduced effective ionization coefficient $(\alpha-\eta)/N$ is easy to obtain, and is shown in Fig. 16. For the temperature range from 2000 K to 3500 K, the value of $(\alpha-\eta)/N$ increases strongly with temperature because of the strong decrease in α/N and the only slight reduction in η/N . For gas temperatures lower than 1500 K, $(\alpha-\eta)/N$ is higher than for 2000 K and 2500 K on account of the relatively low mole fraction of copper below 1500 K.

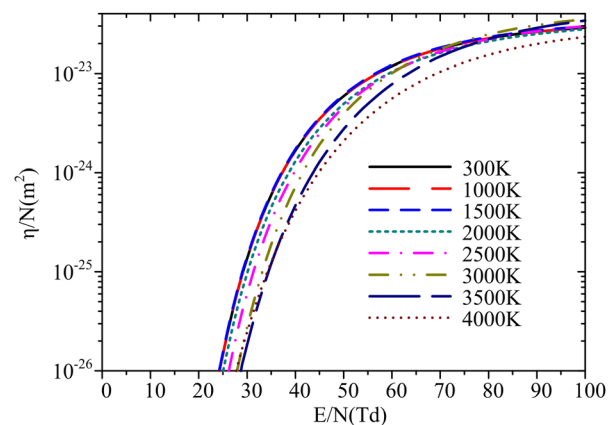


FIG. 15. Reduced attachment coefficient η/N in hot CO₂ with 1% Cu atoms as a function of E/N for different gas temperatures at 0.4 MPa.

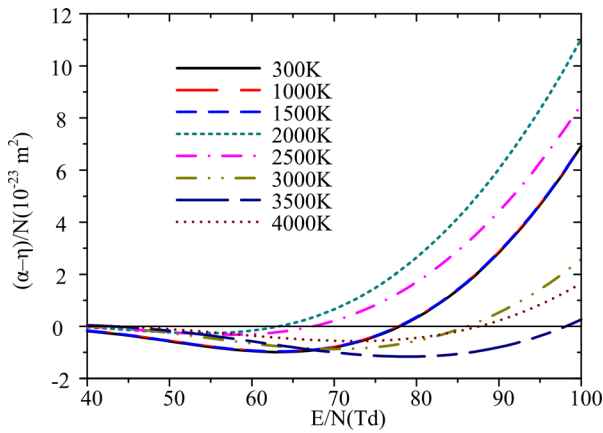


FIG. 16. Reduced effective ionization coefficient $(\alpha-\eta)/N$ in hot CO_2 with 1% Cu atoms as a function of E/N for different gas temperatures at 0.4 MPa.

B. α/N , η/N , and $(\alpha-\eta)/N$ in a hot CO_2 with different fractional Cu mixture

Figs. 17 and 18 show the calculated reduced ionization coefficient α/N and reduced attachment coefficient η/N in hot CO_2 with different fractions of Cu at 0.4 MPa and 2000 K. An increase of the Cu fraction from 0.05% to 1% leads to a noticeable increase in the values of α/N . However, η/N slightly declines with increasing mole fraction of Cu. This can be explained by noting that Cu has zero attachment cross section. Further, an increase of the copper mole fraction leads to a decreased oxygen mole fraction, which makes attachment less likely to happen.

Fig. 19 shows the reduced effective ionization coefficient in hot CO_2 with different fractions of Cu at 0.4 MPa and 2000 K. An increasing mole fraction of copper leads to an obvious reduction in $(\alpha-\eta)/N$ of the gas mixtures. Based on the data shown in Fig. 19, we can determine that the value of $(E/N)_{cr}$ decreases by 18.3%, from 77.5 Td to 63.3 Td, as X_{Cu} increases from 0.05% to 1%. Thus, it is clear that increasing X_{Cu} will markedly increase the probability of dielectric breakdown occurring in the gas mixture at a fixed gas pressure.

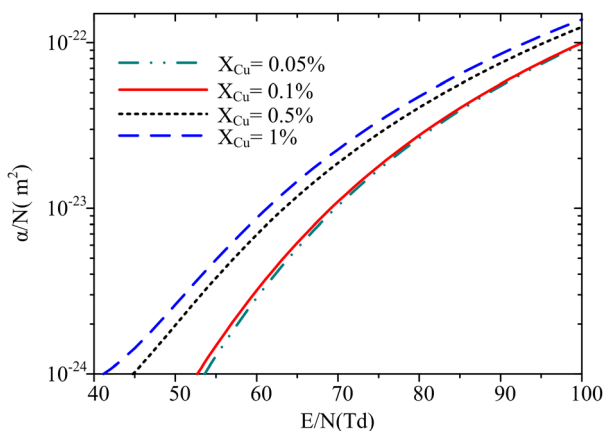


FIG. 17. Reduced ionization coefficient α/N in hot CO_2 with different fractional Cu contents as a function of E/N at 0.4 MPa and 2000 K.

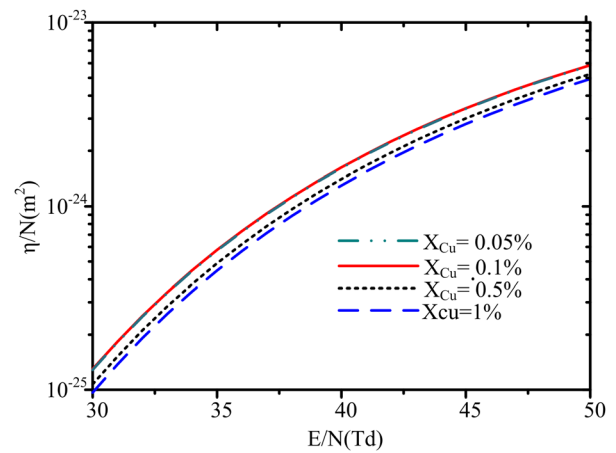


FIG. 18. Reduced attachment coefficient η/N in hot CO_2 with different fractional Cu contents as a function of E/N at 0.4 MPa and 2000 K.

VI. CRITICAL REDUCED ELECTRIC FIELD STRENGTH $(E/N)_{cr}$ IN HOT CO_2 WITH COPPER

Fig. 20 presents the critical reduced electric field strength $(E/N)_{cr}$ in hot CO_2 with different fractions of Cu at 0.4 MPa. In the range from 300 K to 1500 K, $(E/N)_{cr}$ is almost independent on temperature. From 1500 K to 2000 K, the copper in the solid phase gradually vaporizes. This increases the ionization reactions and slightly reduces the average electron energy of the gas mixtures. Thus, the $(E/N)_{cr}$ values in hot CO_2 decline for this range of temperature. Above 2200 K, a drastic increase in $(E/N)_{cr}$ results from the generation of O_2 . Then, with a further increased temperature, the $(E/N)_{cr}$ values decrease again as a consequence of the dissociation of O_2 . At $X_{Cu}=1\%$, for example, $(E/N)_{cr}$ decreases by 21.5% from 77.8 Td to 61.1 Td when gas temperature is raised from 300 K to 2200 K, then increases up to 98.1 Td at 3500 K and finally decreases to 87.6 Td at 4000 K. This trend is quite different from the cases of SF_6 ,²⁸ $\text{SF}_6\text{-N}_2$,²⁹ and air.¹² For relatively low gas temperatures, the $(E/N)_{cr}$ of those gases show only a slight dependence on gas temperature, while raising temperature further leads to a significant reduction of the values of $(E/N)_{cr}$. Further, an increased mole fraction of copper vapor from erosion of

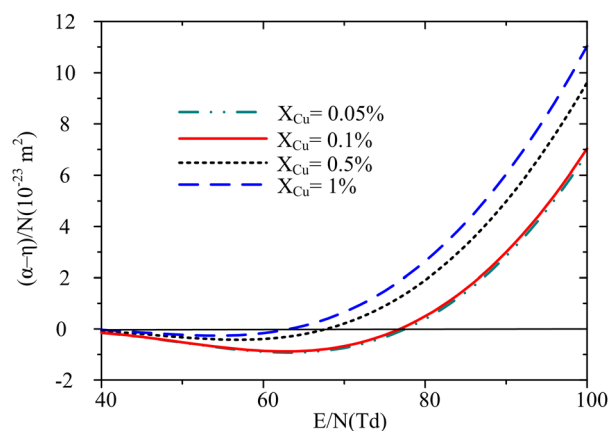


FIG. 19. Reduced effective ionization coefficient $(\alpha-\eta)/N$ in hot CO_2 with different fractional Cu contents as a function of E/N at 0.4 MPa and 2000 K.

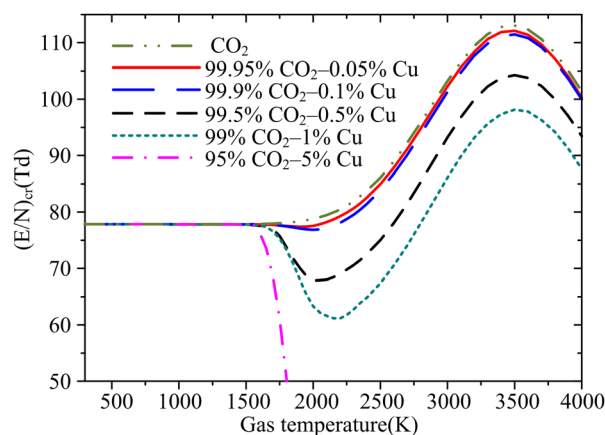


FIG. 20. Critical reduced electric field strength $(E/N)_{cr}$ in hot CO_2 with different fractional Cu contents as a function of gas temperature at 0.4 MPa.

contacts leads to a marked reduction of $(E/N)_{cr}$ of the gas mixtures. At $X_{\text{Cu}} = 5\%$, $(E/N)_{cr}$ sharply declines from 77.8 Td to a very low value when temperatures are increased above 1500 K, because of the increase in the ionization reactions caused by copper's large ionization cross section.

VII. CONCLUSIONS

The influence of copper vapor on dielectric breakdown properties of hot CO_2 for the temperature range from 300 K to 4000 K at a fixed pressure of 0.4 MPa has been analyzed by solving the two-term Boltzmann equation. It can be concluded that although Cu has a relatively large excitation cross section, which leads to a slight reduction in the EEDF, its low ionization potential and large ionization cross section significantly increase the importance of the ionization reactions.

The results reveal that the critical electric field strength $(E/N)_{cr}$ decreases as temperature increases from 1500 K to 2000 K and then increases with temperature up to 3500 K, and an increasing mole fraction of copper makes the decrease of $(E/N)_{cr}$ more severe. At $X_{\text{Cu}} = 0.1\%$, $(E/N)_{cr}$ drops from 77.8 Td to the minimum value of 76.9 Td, while at $X_{\text{Cu}} = 1\%$, the $(E/N)_{cr}$ drops to 61.1 Td at 2200 K. Therefore, the formation of copper vapor from the erosion of contacts is likely to increase the probability of the occurrence of post-arc electric breakdown. The dielectric strength data presented here can also serve as fundamental data for the evaluation of the post-arc dielectric breakdown properties of electric power switchgear equipment.

ACKNOWLEDGMENTS

The work was supported by National Natural Science Foundation of China (51221005 and 51222704).

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