Environment-friendly Insulating Gases for HVDC Gas-insulated Transmission Lines

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***Abstract*—Environment-friendly gas insulating mediums adapted to a DC gas-insulated transmission line (GIL) electric field condition is the key to the next generation of Environment-friendly HVDC GILs. In this paper, we review the literature on sulfur hexafluoride (SF_6) alternatives including the scientific understanding, control, and implementation of gas-solid systems in this type of power transmission. First, the structure-activity relationship between the molecular structure and physico-chemical properties of Environment-friendly insulating gases is presented. Then, the search and prediction of important physicochemical properties of gases are summarized. Subsequently, in view of the potential of environmental friendly insulating gases, the swarm parameters of gas discharge and breakdown properties in a quasi-uniform field, inhomogeneous field, and at the gas-solid interface, that need to be taken into account with industrialized DC GILs are discussed. The latest research progress on insulation characteristics, especially the polarity effect in DC gas–solid insulation systems, the sensitivity to the electrode surface state, and the non-uniformity of the electric field, and the influence of metal particles and their variation with air pressure, is highlighted. In addition, the heat transfer characteristics of insulating gases, related to DC GIL transmission with a large current-carrying capacity and the influence of alternative gases on the heat transfer characteristics are described. Finally, aiming at solving the contradiction of low environmental impact, high dielectric strength and low liquefaction temperatures in the selection of alternative gases, an coordinated regulation model for Environment-friendly gases in DC GILs is established. Considerations for future work on this topic are also presented.**

***Index Terms*—Environmental friendly insulating gas, gas-insulated transmission line, high-voltage direct-current transmission, sulfur hexafluoride.**

I. Introduction

With the advantages of less harm to the environment, smaller land occupation for energy collection offshore platforms and higher current-carrying power transmission [1], [2], the concept of Environment-friendly DC gas insulated transmission lines (GILs) have been developed in recent years. It is hoped that DC GILs can provide better stability in trunk network systems, increase transmission power, and address environmental problems.

Around 1940, sulfur hexafluoride (SF_6) was first used as an insulating gas for high-voltage devices in nuclear physics [3]. In 1972, SF_6 was used as the gas insulating medium in the first GIL launched at the Hudson Power Plant, USA [1]. At present, SF_6 is still the main insulating gas used in the power industry because of its superior technical performance to that of other gases. However, SF_6 gas also has some shortcomings [3]. SF_6 is a heavy gas with larger molecular weight and a higher liquefaction temperature than those of ordinary gases, so it is not suitable for use in very cold conditions. In addition, SF_6 gas is expensive, which makes it unsuitable for electrical equipment with large gas volumes. Moreover, the electrical strength of SF_6 gas is affected by a non-uniform electric field, conductive particles, and electrical surface roughness. As a greenhouse gas, the global warming potential (GWP) of SF_6 is 23,900 times that of carbon dioxide (CO_2) gas, and its life in the atmosphere is up to 3,200 years, which has a cumulative effect on global warming [4]. In the Kyoto Protocol signed by the parties to the United Nations Convention on Climate Change in 1997, SF_6 is listed as one of the six restricted greenhouse gases, which requires that the use of SF_6 is restricted by 2020.

Therefore, it is critical to find an Environment-friendly alternative gas to SF_6 for use as an insulating medium for high-voltage direct-current (HVDC) GILs to meet the Kyoto Protocol and protect the ecological environment. In addition, to understanding their gas discharge and gas-solid interface characteristics under DC electric fields, it is very important to conduct basic research on insulating gases for HVDC GILs [5].

To find Environment-friendly insulating gases suitable for DC GIL, the primary selection criteria areas are as follows: 1) Minimal environmental impact; 2) Non-liquefaction; 3) High dielectric strength, especially for DC conditions; 4) High heat dissipation.

Research on SF_6 substitutes has been performed for many years, but no gases better than SF_6 to fulfill the application criteria have yet to be found. Potential insulating gas candidates are introduced dry air, nitrogen (N_2), CO_2, perfluorocarbons (PFCs), hydrofluorocarbons (HFCs), and gas mixtures containing SF_6. These gases do not have a higher dielectric

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strength than that of SF_6, but their GWPs are lower and their boiling points are below 0 °C. Relatively well-understood gases such as dry air, N_2, CO_2, and SF_6/N_2 gas mixtures have been studied for use in GILs. Dry air, N_2, and CO_2 are environmentally friendly, easy to handle, and suitable for alternative dielectric (routine) tests in a factory. However, these gases have a lower dielectric performance than that of SF_6, so the size of the equipment has to be increased. Gas mixtures of SF_6/N_2 are also included in this review because they have already been used in gas-insulated systems for more than ten years and are effective to lower the GWP of systems [6]. Other alternative gases with low GWP possibly suitable for use in gas-insulated systems are now under study. These alternative gases include trifluoroiodomethane (CF_3I), Fluoronitrile (C_3F_7CN) and Fluoroketones ($C_5F_{10}O$: C5 F-ketone, $C_6F_{12}O$: C6 F-ketone), and are suggested to be used as gas mixtures with CO_2 or oxygen (O_2). These gases have excellent dielectric strength but their boiling points are higher than that of SF_6 [7]–[9].

The industrial applications of environmental friendly insulating gases are countable. The compressed air was used as the insulating medium inside GIL. To achieve the same insulation performance as pure SF_6 gas, the inflatable pressure would have to exceed a 1 MPa. CF_3I gas mixture if it was to be applied to replace SF_6 gas, and the results showed that the mixture of CF_3I and N_2 with a gas volume fraction of 10%–20% was suitable as an insulating medium in GIL. In 2017, GE company successfully developed 420 kV GIL filled with G3 (C_3F_7CN/CO_2) gas and put it into operation in Mannington, UK. The GIL used 4% C_3F_7CN/96% CO_2, and the gas pressure was 0.9 MPa.

Currently, there are still many problems in the application of environmentally friendly gases in practical projects.

1) At present, the synthesis ability of alternative gases is limited and the production cost is high, which limits the promotion and application of new gases.

2) Compared with SF6, all the new gas mixtures should increase the pressure when applied in the HV GIE to improve the insulation strength. Because of this, more stringent requirements are put forward for the mechanical strength, leak-proof level and welding technology of the equipment.

3) Although GIL does not require arc extinguishing, the compatibility of environmentally friendly gases with solid insulating materials and the effect of decomposition products on insulation after long-term operation are still under study.

The year of 2020 is the time node of the Kyoto Protocol to limit the use of SF_6. Here we review the latest research progress on alternative gases to SF_6, especially for use under DC conditions. An overview of recent prior research on SF_6 alternatives for gas-insulated equipment under DC stress is presented to provide inspiration for further study of DC insulation. The paper is organized as follows:

In Section II, physico-chemical properties of potential Environment-friendly gases including liquefaction characteristics, basic dielectric properties, thermo-physical properties and environmental protection-related characteristics of great concern are presented. Moreover, the effects of molecular and electronic structures on the macroscale liquefaction temperature and dielectric strength of gases, as well as the prediction of alternative gases based on structure–activity relationships and molecular design concepts is reviewed. In Section III, the gas insulation properties of substitute gases for SF_6 are introduced. In Section IV, the heat transfer performance of environmentally friendly gases and their temperature rise performance in GILs are discussed. In Section V, based on the presented research and our latest results for environmentally friendly insulating gases in DC GILs, a synergistic control model of alternative gas mixtures is proposed that considers liquefaction temperature, electrical strength, GWP, and temperature rise characteristics. Criteria for use of environmentally friendly insulating gases in DC GILs are also introduced. Finally, we present a brief summary and provide some suggestions for future work in Section VI.

II. Search and Prediction: Physico-Chemical Properties

Physico-chemical properties are the basic parameters of gases. The physical and chemical properties of the alternative insulating gases used in environmentally friendly DC GIL equipment include not only liquefaction temperature, dielectric strength, and heat transfer performance, but also factors related to environmental protection and safety, such as GWP, ODP, atmospheric retention time, and LC50. However, we have not yet found a gas that displays properties that meet all of the requirements for DC GILs at the same time. Therefore, although there are hundreds of alternative gases to SF_6, environmentally friendly insulating gases that can completely replace SF_6 are still under exploration. In recent years, with the development of quantum physics theory and advanced computing technology, theoretical research on alternative gases to SF_6 has started to improve the efficiency of the research process. By establishing the relationship between molecular microscale parameters and macroscale properties, theoretical prediction of the properties of unknown gases can be realized, and the optimization and design of alternative gases can be conducted. This section summarizes the structure–activity relationships and physicochemical properties in environmental and functional aspects of alternative gases to SF_6.

A. Global Warming Potential

In order to evaluate the potential contribution of each insulation gas to global warming, the greenhouse warming potential (GWP) was devised to aid in the identification of particularly effective greenhouse gases [10]. The GWP is defined as the radiative forcing due to an instantaneous release of 1 kg of a trace substance integrated over a chosen time horizon, relative to that of 1 kg of CO_2, and it has two important properties, the atmospheric lifetime (τ) and the radiative efficiency. It can be calculated according to the following formula:

$$GWP = \frac{\Delta F_{\mathrm{A}} \times \int_0^{\mathrm{T}} A(t)\mathrm{d}t}{\Delta F_{\mathrm{CO_2}} \times \int_0^{\mathrm{T}} R(t)\mathrm{d}t} \tag{1}$$

where T is the time horizon; ΔF_{A} is the radiative forcing resulting from a 1 kg increase of compound A; $A(t)$ is the time decay of a pulse of compound A, and $\Delta F_{\mathrm{CO_2}}$ and

$R(t)$ are the comparable quantities for the reference. Usually, the GWP was evaluated over a time horizon of 100 years assuming an exponential decay for the time evolution of the gas in the atmosphere with a rate of $1/\tau$, and the decay mechanism accounts for compound removal via reactions with OH only [11].

Some scholars determined the infrared absorption spectrum by measuring the reaction rate of the reaction of greenhouse gases with active species such as OH radicals, and calculated the gas lifetime and radiation efficiency, and then calculated the GWP of some greenhouse gases [12]. The previous alternative gas is often fluorocarbon, but some studies have shown that the greenhouse effect of C-F bonds is large [10], and compounds obtained by replacing one or more C-F bonds with C-O bonds have lower GWP [13]. Since 1970, the greenhouse effect of fluorocarbon bonds (CFCs), chlorocarbons, hydrofluorocarbons (HCFCs), and hydrofluorocarbons (HFCs) has gradually gained attention, and some scholars have carried out in-depth research [14]. G. Acerboni *et al.* have found that due to the short life span of C_2F_4, C_3F_6 and C_4F_6, the greenhouse effect of them could be almost ignored [15]. The atmospheric lifetime and *GWP* of CH_4 are much lower than greenhouse gases such as CO_2 and SF_6 [10]. Since halogenated olefins have a C=C bond, their lifetime is shorter than similar saturated compounds. Therefore, short-chain halogenated olefins could be used as substitutes for saturated halogenated hydrocarbons [15].

With the advancement of computer technology, the density functional theory (DFT) has been proven to calculate parameters such as radiation efficiency [16]. Some scholars combined the functional group contribution method to calculate the reaction rate of gas containing special functional groups with atmospheric OH radicals, atmospheric lifetime of gas, radiation efficiency, etc., and obtained their GWP [17]. Franck limited the molecular size to a maximum of 15 atoms and allowed only compositions of the elements: C, H, F, Cl, Br, N and S. And he calculated a variety of chemical gases. Finally, several candidate gases were obtained [11]. Some studies found that for molecules with the same structure, as the number of F atoms increases, the GWP value of the gas increases.

B. Intrinsic Dielectric Strength

The electrical strength (ES) of gases is determined by the microstructure of the gas molecules. The microscopic properties of gas could be used to screen and predict the electrical strength of the replacement gas, which can improve the efficiency of finding new replacement gases.

High electrical strength gas has similar molecular structure rules [18]. There is a significant relationship between ES and properties such as molecular weight, quantity, and chemical structure [19], and ES tends to increase with increasing molecular weight [20]. For hydrocarbon gases, many scholars have found that the main factors affecting the ES of saturated and unsaturated hydrocarbons are the number of C-H bonds [21], [22] and the number of C=C bonds [23].

Some scholars have studied the relationship between ES and parameters such as polarizability, ionization energy [24], [25], electron affinity [26], and light absorption spectrum integration (IOA) [27]. Based on the DFT calculation method, Franck *et al.* established a multivariate regression model of microscopic parameters and insulation properties [28]. Xiao and other scholars examined the relationship between the electronic affinity energy, molecular volume, LUMO energy level and other electrical properties of some organic gases, and found that a high electrical strength gas often has C=C double bonds and C≡N bonds [29].

Wang believes that the cross section of gas molecules is proportional to the surface electrostatic potential, and proposed a new structure-activity relationship (SAR) model [30]. It was found that CF_3 is the most effective functional group to improve ES due to its large surface area and polarizability, and SF_5CN and SF_5CFO were found to be the potent candidates to replace SF_6 [30]. Because molecular ionization energy and electron affinity energy are difficult to obtain [31], Wang and other scholars abandoned the ion calculation, and adopted parameters of physical meaning in neutral molecules to obtain a better performance SAR Model [32]. Li and other scholars also believed that the surface electrostatic potential is strongly correlated with the ES, and they found that ES will be stronger with the increase of the relative molecular mass of the halogen element [33].

After searching and screening, it was found that CF_3I, C_3F_8, c-C_4F_8 and other gases are expected to replace SF_6 as an environmentally friendly gas. In recent years, ALSTOM and 3 M have developed C_3F_7CN with good insulation properties, in which the gas molecule has a unique C≡N bond. ABB has proposed organic fluorinated gases with C=O bond such as $C_5F_{10}O$ and $C_6F_{12}O$.

C. Liquefaction

In practical applications, the choice of insulating gas type and mixing ratio is restricted by liquefaction temperature, which is also called the boiling point. Therefore, the liquefaction temperature should be focused on in the search and prediction process to ensure the practical applicability of an insulating gas.

Scholars from Tsinghua University measured the saturated vapor pressure of CF_3I in the temperature range of 243.15–393.15 K, and summarized an equation with four parameters that can accurately calculate the boiling point (T_{B}) of CF_3I. The saturated vapor pressure of SF_6 is much higher than the value of CF_3I at the same temperature [34]. The saturated vapor pressure of CF_3I/N_2 is higher than that of c-C_4F_8/N_2 for a certain mixed ratio, which means that CF_3I/N_2 mixtures can be used for higher operating pressures. It is found that CF_3I/N_2 gas mixture has a better potential as an alternative gas [35]. D. M. Xiao *et al.* ignored the effect of buffer gas and calculated the T_{B} of CF_3I/N_2 [36] and C_3F_8/N_2 [37] gas mixture using the Van Der Waals equation. The boiling point of CF_3I/CO_2(40%/60%) is about -5 °C at 0.5 MPa [38]. When the content of C_3F_7CN in C_3F_7CN/CO_2 gas mixture is about 5% and 10%, the T_{B} of the gas mixture is as low as -30 °C and -10 °C respectively [39], which is a powerful candidate to replace SF_6.

Wang found that the balance between ES and T_{B} can be achieved by minimizing the local polarity of the molecule [30]. Polarizability, ionization energy [24], [25], dipole moment and electron affinity [40] have important influences on the T_B. Based on the DFT calculation, Franck *et al*. observed strong correlation between the T_B and some predictors in non-polar molecules [28]. Li has proposed the effect of halogen substitution on the T_B of gas molecules and its variation [33].

Tu considered the interaction of various components in the gas mixture, and used the Peng-Robinson (PR) equation and the Van der Waals equation (PR-vdW method) to analyze the gas-liquid equilibrium data of SF_6/N_2, SF_6/CO_2 [41] and CF_3I/CO_2 [42] gas mixture. Yuan used the PR-vdW model to calculate the dew point temperature (T_B) of the binary gas mixture, with SF_6, C_3F_8, c-C_4F_8, CF_3I and C_3F_7CN as the insulating gas and N_2 or CO_2 as buffer gas. The dew point temperatures of the SF_6 substitute gases at different pressures and the upper limits of the insulating gas mole fraction at −30°C, −20 °C and −10 °C were obtained [43]. The relationship between pressure, temperature and the T_B of C_3F_7CN/N_2 and C_3F_7CN/CO_2 gas mixture is shown in Fig. 1.

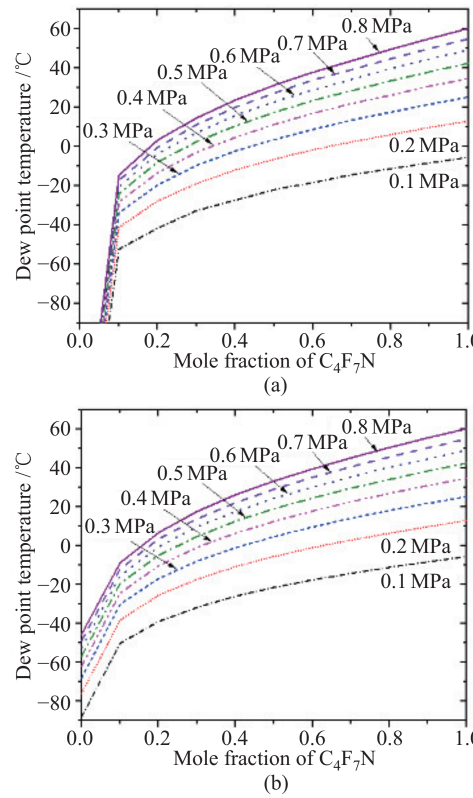


Fig. 1. Dew point temperatures of C_3F_7CN mixtures at different pressures. (a) C_3F_7CN/N_2. (b) C_3F_7CN/CO_2 [43].

D. Heat Dissipation

Convection is the dominant type of heat transfer in GILs. Therefore, the heat transfer performance is closely related to the gas flow. Heat transfer properties include many thermos-physical parameters such as gas density, thermal conductivity, capacity, and dynamic viscosity. According to Technical Brochure 218, WG 23/21/33-15 [44], Vermeer's constant is linked to the physical properties of gases, according to the following formula.

$$C = 0.1638 \left(\frac{\rho^2 \lambda^2 C_p}{\eta} \right)^{\frac{1}{3}} \tag{2}$$

where ρ is the density, λ is the thermal conductivity, C_p is the specific heat capacity and η is the dynamic viscosity.

For the convective heat transfer coefficient of gas mixtures, it can be calculated by the following formula.

$$C_{mix} = C_1(x_1)^{0.75} + C_2(x_2)^{0.75} \tag{3}$$

where C_1, C_2, x_1 and x_2 are the convection coefficient and volume fraction of two kinds of gas in the mixture.

It can be seen that the gases with high molecular weight, high thermal conductivity, high heat capacity and low dynamic viscosity are the good choices for insulating gases in large current-carrying-capacity DC GILs. The Vermeer's constant of C_3F_7CN, SF_6 and CO_2 at standard atmospheric pressure according to (3), are 13.8, 11.3 and 6.10, respectively [45]. This indicates that C_3F_7CN gas has a better heat dissipation performance than SF_6 and CO_2 gases. The heat dissipation of the gas mixture is a bit lower than pure SF6 but still significantly higher than CO_2.

E. Physicochemical Properties of Main Alternative Gases to SF_6

In recent years, the physico-chemical properties of the main alternative gases to SF_6 and buffer gases have been investigated extensively. Major parameters for these gases are summarized in Table I. In Table I, ozone depletion potential (ODP) is applied to assess the potential implications for ozone depletion of the gases. The 50% lethal concentration 50

TABLE I
PHYSICO-CHEMICAL PROPERTIES OF GASES

Molecular formula	Environmental and safety				Functional		
	GWP (IPCC 2013)	ODP	Atmos lifetime (years)	LC50, ppm (4 h)	Boiling point (°C)	Dielectric strength (SF_6)	Heat transfer coefficient
SF_6	23900	0	3400	–	−63	1	11.3
Dry Air	0	0	–	–	–	0.33	5.1
N_2	0	0	–	–	−195.8	0.3–0.4	5.83
CO_2	1	0	–	> 300000	−78.5	0.3–0.4	6.1
CF_4	6500	0	5000	895000 (0.25 h)	−128	0.39	–
c-C_4F_8	10300	0	3200	780000 (2 h)	−6.04	1.25	–
CF_3I	0.45	0.0001	< 0.005	160000	−22.5	1.3	–
C_4F_7N	2210	0	30	12000–15000	−4.7	2.1	13.8
$C_5F_{10}O$	1	0	0.014	> 20000	26	2	–
$C_6F_{12}O$	1	0	0.014	> 10000	49	2.8	–

(LC50) refers to the concentration of poisons that cause half of the animals to die in acute toxicity tests, which is conducive to quantitative comparison of toxicity of different gases.

Predicting the physico-chemical properties of gases using computational methods and then carrying out targeted research on gases predicted to have desirable properties represents an advance over traditional experimental research methods because this new approach can accelerate research progress.

III. Re-examination: Insulation Properties

At present, the main gas insulating medium used in AC transmission pipelines is SF_6. The insulation design of pipelines is also based on the discharge characteristics of SF_6 and the flashover voltage along the gas-solid interface. However, the insulation design for HVDC GILs is quite different from that of AC transmission pipelines. First, the discharge properties of gases vary at different voltage polarities. Subsequently, the electrode surface roughness and partial discharge (PD) characteristics also affect insulating behavior. The charge accumulation of insulating materials under DC voltage will directly affect the gas-solid insulation performance. Moreover, metal particles strongly influence the behavior of gas-solid interfaces.

To determine the insulation properties of environmental friendly gases, the research on the swarm parameter is reviewed in this section. Then, breakdown properties in the gas gap and at the gas-solid interface are reviewed.

A. Electron Swarm Parameter Measurements of Identified Alternative Gases

At low pressure, the γ process could be ignored in the study of insulation gas discharge. According to the Townsend discharge theory, when E/N (E is the electric field strength, and N is the number of gas molecules per unit volume) exceeds $E/N_{(\mathrm{lim})}$, the self-sustaining discharge condition is satisfied, and the $E/N_{(\mathrm{lim})}$ could be used to characterize ES [25]. Furthermore, studying the electron swarm parameters during gas discharge can guide the choice of the type and mixing ratio of the insulating gas.

Swarm parameters including the ionization coefficient, attachment coefficient, electron drift velocity and electron diffusion velocity can be obtained by measuring the current or luminous flux during discharge. The steady-state Townsend method (SST) [46], [47] and the pulse Townsend method (PT) [48] are two experimental methods. In addition, collision cross sections of gas can be used to calculate the electron swarm parameters, including the Monte Carlo simulation calculation and Boltzmann equation method, etc. [49].

1) Experimental Method

At present, compressed dry air and N_2 have been used in medium voltage switchgear [50]. Many scholars have experimented with conventional gases to obtain their $(E/N)_{\mathrm{lim}}$ [25], [51]–[54], where N_2 is 128.3 Td, CO_2 is 104.7 Td, and SF_6 is 361.4 Td (1 Td = 10^{-21} Vm^2). Urquijo *et al*. used the PT method to study the electron swarm parameters of CO_2 and its mixture with SF_6, N_2 and O_2 [55], [56]. The swarm parameters of SF_6/CO_2 and SF_6/N_2 gas mixtures were studied by the PT method [46], [55], [57]–[59], and it was found that the SF_6/N_2 gas mixture is more suitable [60], [61].

In view of the alternative gases on SF_6, some scholars measured the swarm parameters by PT method including the ionization coefficient, attachment coefficient and electron drift velocity of perfluoroalkanes n-C_NF_{2N+2} (N = 1–4) [62], [63]. It was found that the $E/N_{(\mathrm{lim})}$ of CF_4 is 123.8 Td [64], and the $E/N_{(\mathrm{lim})}$ of c-C_4F_8 is 439.5 Td [65]. Urquijo and Xiao studied the swarm parameters and $E/N_{(\mathrm{lim})}$ of CF_3I/N_2 [66] and c-C_4F_8/CF_4 [67] gas mixture respectively, which provided a basis for the selection of the mixing ratio. Nechml [68], Franck [69] and other scholars tested the swarm parameters of C_3F_7CN/CO_2 and C_3F_7CN/N_2 gas mixtures at low mixing ratio, and found that the mixing gas has a synergistic effect, which verifies the feasibility of using the C_3F_7CN gas mixture instead of SF_6. In addition, Franck also studied the swarm parameters of the $C_5F_{10}O/N_2$, $C_5F_{10}O/CO_2$ gas mixture at a low mix ratio [70].

Tu *et al*. have obtained the $E/N_{(\mathrm{lim})}$ of the C_3F_7CN/CO_2 and C_3F_7CN/N_2 gas mixture at different mixing ratios by the SST method. It was found that the ES of the C_3F_7CN/N_2 gas mixture is better than that of the C_3F_7CN/CO_2 gas mixture, and there is a synergistic effect between the two gas mixtures. When the ratio of C_3F_7CN reaches 6.8%, the coefficient of synergistic effect of the C_3F_7CN/CO_2 gas mixture reaches the maximum [71].

2) Calculation Method

The electron swarm parameters of the binary gas mixtures of CO_2/N_2 and CO_2/O_2 have been calculated by the scholars using the Monte Carlo and the Boltzmann equation calculation method. It was found that the $(E/N)_{\mathrm{lim}}$ of the CO_2/O_2 gas mixture is higher than that of the CO_2/N_2 gas mixture [60]. Using the Monte Carlo calculation and Boltzmann equation method based on two-term approximations [72], [73], Dincer, Xiao, and Li calculated the swarm parameters of SF_6/N_2 [74], SF_6/CO_2 [75], SF_6/CF_4 [76] and hot SF_6/CF_4 [77] gas mixtures.

For the new environmental gases, scholars calculated the swarm parameters of the CF_3I gas mixture by solving two-term approximation Boltzmann equations, and found that the $(E/N)_{\mathrm{lim}}$ of CF_3I/N_2 and CF_3I/air gas mixtures is the largest [78], [79]. Xiao *et al*. proposed a new group of collision cross sections, and the swarm parameters of the C_3F_8/N_2 gas mixture were calculated by solving the Boltzmann equation. The $(E/N)_{\mathrm{lim}}$ of C_3F_8 is 337 Td [80], while the $(E/N)_{\mathrm{lim}}$ of the C_3F_8/N_2 gas mixture is larger than that of C_3F_8/CO_2 [37]. Xiao and Li *et al*. calculated the swarm parameters of the c-C_4F_8 gas mixtures, and found that the $(E/N)_{\mathrm{lim}}$ of the c-C_4F_8/N_2 and c-C_4F_8/air gas mixtures are the largest, and the ES of the c-C_4F_8/N_2 gas mixture is comparable to SF_6/N_2 [81]. When the mixing ratio exceeds 80%, the ES of the c-C_4F_8 gas mixture is higher than that of pure SF_6 [82], which has potential to be an alternative gas. In addition, parameters such as electron density can be calculated by means of quantum code. The DM formula was modified by changing the weighting factors, and the collision cross sections of C_3F_7CN and $C_5F_{10}O$ were calculated, which provided a theoretical basis for further study of C_3F_7CN and

$C_5F_{10}O$ [83].

Table II summarize the $(E/N)_{\text{lim}}$ of insulation gases.

TABLE II
$(E/N)_{\text{LIM}}$ OF INSULATION GASES

Gas	$(E/N)_{\text{lim}}$ (Td)	Gas	$(E/N)_{\text{lim}}$ (Td)
N_2	128.3 [51]	C_3F_8	337.0 [80]
CO_2	104.7 [51]	c-C_4F_8	439.5 [65]
SF_6	361.4 [51]	C_3F_7CN	981.8 [68]
CF_3I	473.0 [66]	$C_5F_{10}O$	757.0 [70]
CF_4	123.8 [64]		

B. Breakdown Properties of Potential Gases

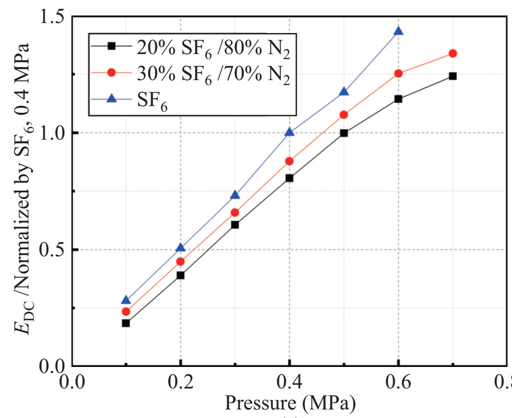
1) Breakdown Properties in Quasi-uniform Fields

Swarm parameters cannot reflect the breakdown at high pressure, so breakdown experiments are necessary.

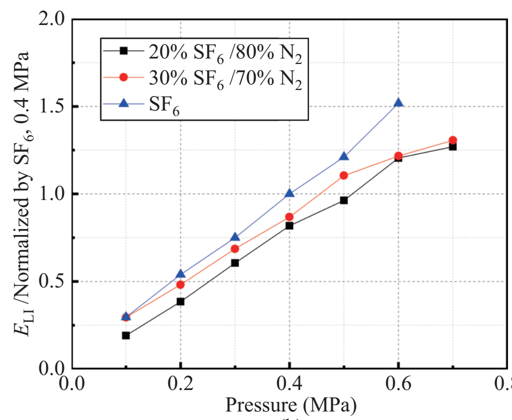
The study of SF_6 gas mixtures began in the 1970 s. The buffer gases used with SF_6 were primarily N_2, CO_2, CF_4, and inert gases. The gas mixtures were used as insulating mediums in high-voltage power equipment, thereby decreasing the amount of SF_6 needed. Because this approach directly lowers the amount of SF_6 used, the GWP of the gas mixture is decreased. As the amount of SF_6 is decreased, the cost and liquefaction temperature of the gas mixtures are also lowered. Therefore, gas mixtures can meet the requirements of gas insulation applications in high-pressure or low-temperature environments.

Woo *et al.* [84] studied the lightning impulse (LI) characteristics of SF_6/N_2 gas mixtures with different proportions under slightly non-uniform electric fields. The results showed that the breakdown voltage (V_B) of the 50% SF_6/50% N_2 gas mixture was 8%–12% higher than that of the 20% SF_6/80% N_2 gas mixture. Qiu and co-workers studied the breakdown characteristics of SF_6/N_2 and SF_6/CO_2 gas mixtures [85]–[88]. Their results showed that the breakdown field strength of SF_6/N_2 was higher than that of SF_6/CO_2 mixed gas under both slightly non-uniform and uniform electric fields. Zhou *et al.* [89] and Zhang *et al.* [90] studied the synergistic effects of SF_6/N_2 gas mixtures under LI conditions. The results showed that in quasi-uniform electric fields, the insulation strength of SF_6/N_2 increased with the SF_6 content in the gas mixture and tended to become saturated. Wang and colleagues studied the relationship between V_B and the proportion of SF_6 in SF_6/N_2 gas mixtures using ball–plate electrodes and power frequency voltages [91], [92]. Larin *et al.* [93] studied the synergistic effect of SF_6/CF_4 gas mixtures under uniform electric fields. They found that the promotion of ionization by SF_6 and the inhibition of ionization by CF_4 were the main reasons for the synergistic behavior of SF_6/CF_4 mixtures. Berg *et al.* [94] investigated the AC, DC, and LI breakdown characteristics of SF_6/CF_4 mixtures under uniform and slightly non-uniform electric fields. Their results showed that the breakdown field strength of the gas mixture depended linearly on the content of SF_6. The discharge characteristics of SF_6 gas mixtures under uniform, quasi-uniform, and slightly non-uniform electric fields are summarized as shown in Table III.

Tu *et al.* [95] studied the DC breakdown and LI characteristics of 30% SF_6/70% N_2 and 20% SF_6/80% N_2 gas mixtures under a uniform electric field. They found that the breakdown strength of 30% SF_6/70% N_2 at 0.6 MPa and 20% SF_6/80% N_2 at 0.7 MPa was comparable to that of SF_6 at 0.5 MPa, as shown in Fig. 2. At present, SF_6 gas mixtures are widely used in high-voltage power equipment [96].



(a)



(b)

Fig. 2. Breakdown field strength (E_{DC}, E_{LI}) of SF_6/N_2 mixtures normalized by SF_6, 0.4 MPa. (a) DC breakdown characteristics. (b) Lightning impulse breakdown characteristics [95].

TABLE III
STUDY ON BREAKDOWN PROPERTIES OF SF_6G AS MIXTURES

Authors	Gas	Voltage	Electric Field Property	Gas Pressure (MPa)
Su-Youl Woo *et al.* [84]	SF_6/N_2	LI	Quasi-uniform	0.3–0.7
Y.C. Qiu *et al.* [85]–[88]	SF_6/N_2 SF_6/CO_2	AC/DC	Quasi-uniform	0.1–0.4
Q.G. Zhang *et al.* [89]	SF_6/N_2	LI	Slightly non-uniform	0.1–0.4
W.J. Zhou *et al.* [90]	SF_6/N_2	LI	Slightly non-uniform	0.7
H. Wang *et al.* [91], [92]	SF_6/N_2	AC	Slightly non-uniform	0.1–0.4
Larin *et al.* [93]	SF_6/CF_4	–	theoretical analysis	–
J. Berg *et al.* [94]	SF_6/CF_4	AC/DC/LI	Uniform/Quasi-uniform	0.2–0.4

However, SF_6 gas mixtures still employ SF_6 as the main insulating medium, so their use only slows down the impact of SF_6 on the environment. At the same time, because of the lower insulation strength of its gas mixtures than that of pure SF_6, it is necessary to increase the gas pressure of the equipment, which may introduce additional costs to industrial production. At present, researchers are searching for new environmentally friendly insulating gases, such as CF_3I, C_3F_7CN, c-C_4F_8, and $C_5F_{10}O$, to achieve complete replacement of SF_6. However, such gases have high liquefaction temperatures and require mixing with buffer gases (CO_2 or N_2) when used under high-pressure conditions. The discharge characteristics of new environmentally friendly insulating gases are presented in Table IV.

Tu *et al*. studied the DC breakdown characteristics of CF_3I/N_2, C_3F_7CN/CO_2, and C_3F_7CN/N_2 gas mixtures [95], [99], [100]. The results for CF_3I/N_2 showed that the insulation performance of 20% CF_3I/80% N_2 at 0.2–0.25 MPa and 30% CF_3I/70% N_2 at 0.15–0.25 MPa was suitable for these mixtures to replace SF_6 in the DC GILs of distribution networks. For the insulation strength of the gas mixtures did not increase remarkably with the increase of ratios and pressure, it is suggested that the 10% SF_6/20% CF_3I/70% N_2 gas mixture at 0.2–0.25 MPa may be a suitable alternative gas to SF_6 in extremely cold regions. Under a uniform electric field, the DC breakdown field strengths of C_3F_7CN/CO_2 and C_3F_7CN/N_2 mixtures increased with the gas pressure. At 0.7 MPa, the negative DC breakdown field strengths of 4% C_3F_7CN/96% CO_2 and 8% C_3F_7CN/92% CO_2 reached 81.21% and 96.5% compared with SF_6 at 0.5 MPa, respectively. The negative DC breakdown field strength of 14% C_3F_7CN/86% N_2 gas mixture reached 87.3% compared with SF_6 at 0.4 MPa [106]–[108]. The DC breakdown electrical strength of environmental friendly insulating gases are shown in Fig. 3.

Under DC electric fields, the insulation properties of gases are sensitive to the surface roughness of the electrodes. The influence of electrode surface roughness on the breakdown strength of an insulating gas is directly related to the type of SF_6 alternative used. Multiple groups have studied the

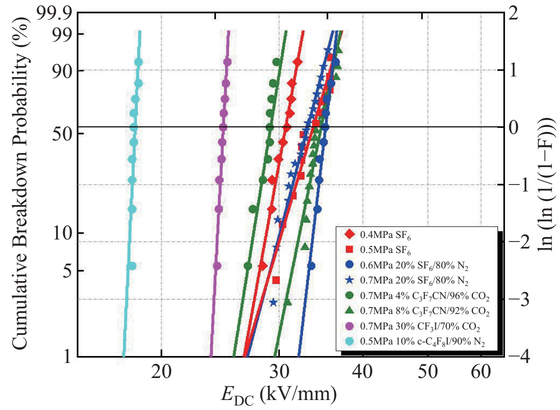


Fig. 3. The Weibull distribution of DC breakdown electrical strength of environmental friendly insulating gases.

influence of electrode surface roughness on the breakdown strength of SF_6 [112]–[114]. Farish *et al*. [115] theoretically calculated the influence of electrode surface roughness on the V_B of SF_6/N_2 mixtures. Their results showed that adding N_2 to SF_6 can lower the sensitivity of the insulating gas to the electrode surface roughness. Qiu *et al*. [116], [117] studied the influence of electrode surface roughness on SF_6/N_2 and SF_6/CO_2 gas mixtures. They found that although the addition of N_2 or CO_2 to SF_6 can decrease the sensitivity of the dielectric strength to surface roughness, the breakdown strength of SF_6 was still higher than that of SF_6/N_2. The breakdown strength of SF_6/CO_2 was slightly higher than that of SF_6 only when the electrode surface roughness reached a certain level. Tu *et al*. used plate–plate electrodes to measure the DC breakdown field strengths of SF_6, 6% C_3F_7CN/94% CO_2, and 20% SF_6/80% N_2 gas mixtures using electrodes with different surface roughness levels. The results showed that the DC breakdown field strengths of the insulating gases became saturated with rising gas pressure and the saturation was more obvious with the increase of electrode surface roughness. The 6% C_3F_7CN/94% CO_2 mixture was less sensitive to the electrode surface roughness than SF_6 and the 20% SF_6/80%

TABLE IV
STUDY ON BREAKDOWN PROPERTIES OF ENVIRONMENTALLY FRIENDLY INSULATING GASES

Authors	Gas	Voltage Property	Electric Field Property	Gas Pressure
Ngoc M N *et al*. [97]	CF_3I/N_2	DC	Quasi-uniform	Pressure-distance up to 5 mm bar
H. Katagiri *et al*. [38]	CF_3I/N_2 CF_3I/CO_2	LI	Slightly non-uniform	0.1 MPa
L. J. Chen *et al*. [98], [99]	CF_3I/N_2 CF_3I/CO_2	LI	Slightly non-uniform	0.1–0.2 MPa
Y. P. Tu *et al*. [95], [100]	CF_3I/N_2	DC	Uniform/	0.1–0.7 MPa
H. E. Nechmi *et al*. [101]	C_3F_7CN/CO_2	AC, LI	Slightly non-uniform	0.1–1 MPa
X. W. Li *et al*. [102]	C_3F_7CN/CO_2 CF_3I/N_2		Theoretical calculation	
Y. Kieffel *et al*. [103], [104]	C_3F_7CN/CO_2	AC, LI	Slightly non-uniform	0.1 MPa
J. Owens *et al*. [105]	C_3F_7CN/CO_2 C_3F_7CN/N_2 C_3F_7CN/Air	AC	Uniform	0.1–1 MPa
Y. P. Tu *et al*. [106]–[108]	C_3F_7CN/CO_2 C_3F_7CN/N_2	DC	Uniform	0.3–0.7 MPa
Yamamoto O *et al*. [109]	c-C_4F_8/Air c-C_4F_8/CO_2 c-C_4F_8/N_2	AC	Quasi-uniform Slightly non-uniform	0.1–0.4 MPa
P.Yan, J.Wang *et al*. [110], [111]	c-C_4F_8/N_2	AC, DC	Slightly non-uniform	0.15–0.5 MPa

N_2 gas mixture, as shown in Fig. 4.

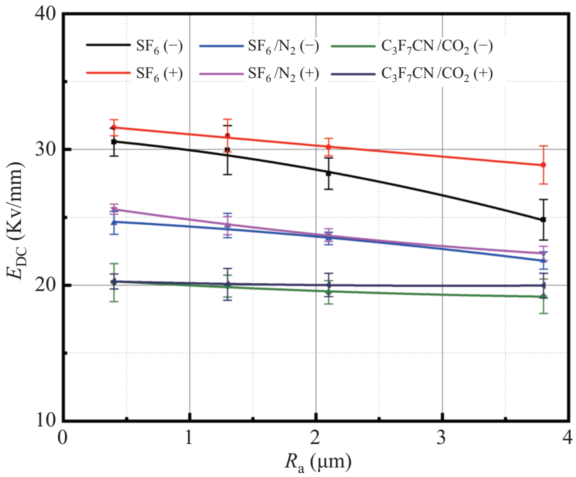


Fig. 4. Relationship between the positive and negative breakdown field strengths and electrode surface roughness for 6% C_3F_7CN/94% CO_2, 20% SF_6/80% N_2, and SF_6 at 0.4 MPa.

A GIL has a coaxial structure that is dominated by a quasi-uniform electric field. The influence of the non-uniformity of the electric field on the insulation performance of alternative gases to SF_6 should be examined.

Zhang *et al.* [118] studied the effect of electric-field non-uniformity on the discharge characteristics of SF_6/N_2 gas mixtures under power frequency electric fields. Under slightly non-uniform electric fields, the V_B of SF_6/N_2 tended to become saturated as the non-uniformity of the electric field increased and the influence of air pressure on V_B became weak. Zhang and colleagues studied the AC breakdown characteristics of CF_3I/N_2 mixed gas under different electric fields [119], [120]. They found that with increasing electric field utilization coefficient, the AC V_B of the CF_3I/N_2 mixture increased gradually with the linear growth rate of gas pressure. This result indicates that the more uniform the electric field, the better the AC breakdown characteristics of the CF_3I/N_2 mixture. Tu *et al.* measured V_B of 6% C_3F_7CN/94% CO_2 under slightly non-uniform electric fields, and compared the results with those for SF_6 and 20% SF_6/80% N_2. The electrodes used in their experiments are shown in Fig. 5.

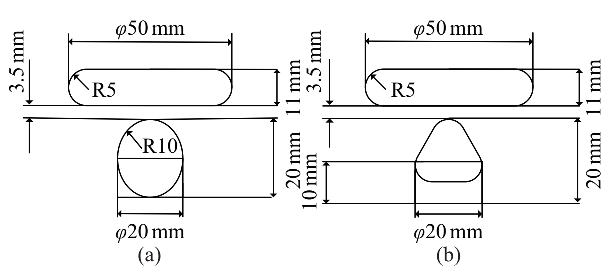


Fig. 5. Electrodes system for breakdown experiments under different electric-field non-uniformity (a) Hemispherical rod-plate electrodes and (b) cone-plate electrodes used to study the breakdown voltage of 6% C_3F_7CN/94% CO_2 under slightly non-uniform electric fields.

Figure 5(a) depicts the hemispherical rod–plate electrodes. The hemisphere diameter was 20 mm, the spacing between the electrodes was 3.5 mm, and an unevenness coefficient of 1.2 was obtained through a simulation. Figure 5(b) displays the cone–plate electrodes with an electrode spacing of 3.5 mm and curvature of the cone regions of 5 and 3.5 mm; the unevenness coefficients (f) obtained from simulations were 1.4 and 1.6, respectively. The negative DC breakdown field strengths of 6% C_3F_7CN/94% CO_2, 20% SF_6/80% N_2, and SF_6 were obtained. The negative DC breakdown field strength of each gas system decreased linearly with increasing electrode unevenness. The breakdown field strengths SF_6 and C_3F_7CN/CO_2 showed similar declines and were lower than that of SF_6/N_2, as shown in Fig. 6.

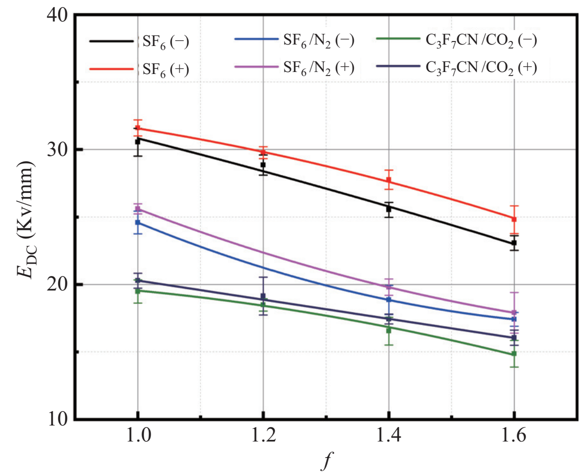


Fig. 6. Relationship between positive and negative breakdown field strengths and electric-field non-uniformity for 6% C_3F_7CN/94% CO_2, 20% SF_6/80% N_2, and SF_6 gas systems at 0.4 MPa.

2) Breakdown and Partial Discharge Properties in Non-uniform Electric Fields

In GILs, typical defects like needle-shaped protrusions or metallic particles lead to strong local non-uniform electric fields, resulting in a considerable decrease of the discharge inception voltage. The influence of defects on the PD and breakdown properties of gases under DC voltage is the main topic of this section. First, the influence of fixed protrusions on gas insulation behavior is described. Then, information about particle movement and the subsequent influence on PD and breakdown properties are discussed. Finally, the particle discharge properties under AC and DC voltages for potential alternative gases and SF_6 are compared. A highly non-uniform electric field distribution is generally simulated by using needle-plate or rod-plate electrodes with a small tip curvature radius or tip–plate electrodes. Non-uniform electric field distributions can also be emulated using a metal protrusion with a certain length attached to a spherical electrode [109], parallel plates [121], [122], or a smooth HVDC conductor [123], [124].

Many researchers have conducted experiments on the breakdown performance of SF_6 and its mixtures with CO_2, N_2, and PFCs [125] under highly non-uniform fields using the action of AC and impulse voltages [126]–[130]. DC breakdown characteristics under highly non-uniform fields have also been determined. Qiu and Kuffel found that the positive DC V_B of 20% SF_6/80% CO_2 and 20% SF_6/80% N_2 gas mixtures were both 43% higher than that of SF_6 gas at 0.22 MPa for

GILs [131]. Malik and co-workers studied the DC insulation characteristics of SF_6/N_2 gas mixtures using a needle-plate electrode system [132]. They found that the V_B of 0.1% SF_6/99.9% N_2 was 50% compared with SF_6 and 2.5 to 3 times that of N_2. The breakdown level of 5% SF_6/95% N_2 was 65% compared with SF_6. The V_B of 85% SF_6/15% N_2 with a wide electrode gap was almost the same as that SF_6 and even higher than that of SF_6 in some cases. Zhang *et al*. evaluated the DC V_B of SF_6/CF_4 gas mixtures to analyze the effects of electric field non-uniformity on breakdown characteristics at different mixing ratios and gas pressures [133]. They found that the V_B of a 20% SF_6/80% CF_4 gas mixture was 29.7% higher than that of CF_4 gas.

The insulation properties of SF_6-free gases under highly non-uniform fields are of current interest. To obtain the same dielectric strength of SF_6, it is necessary to raise the pressure by 3 to 4 times [134] because the dielectric strength values of conventional environmental friendly gases are only 20%–40% compared with SF_6 at the same pressure. Wada *et al*. reported the breakdown characteristics of CO_2 and N_2 under non-standard LI [135]–[137]. It was found that the DS of CO_2 was 1.1 to 1.6 times higher than that of a standard waveform, and the positive non-standard LI V_B of N_2 was 106%–167% of that under standard LI.

Extensive research on CF_3I gas mixtures with CO_2, N_2, and CF_4 as substitutes for SF_6 has been reported in recent years. However, few studies of the dielectric strength of such mixtures under DC voltage have been reported. Most work has focused on AC and standard LI breakdown characteristics. The groups of Kamarudin [7], Xiao [138]–[140], and Zhang [141], [142] have conducted much research in this area. The dielectric strength values of CF_3I/CO_2 mixtures containing 30%–40% CF_3I have exceeded that of SF_6 gas at 0.15–0.20 MPa and can be applied to cubicle-type gas-insulated switchgear (C-GIS). The $U_{50\%}$ value of a 35% CF_3I/70% N_2 mixture is lower than that of 20% SF_6/80% N_2. The AC V_B values of CF_3I/CO_2 gas mixtures are slightly higher than those of CF_3I/N_2 gas mixtures, and the AC withstand voltage level of 20% CF_3I/80% N_2 gas mixtures can reach that of SF_6. The ratios of the AC withstand voltages of a 30% CF_3I/70% N_2 gas mixture to those of SF_6 and a 20% SF_6/80% N_2 mixture are about 55% and 75%, respectively. It has been suggested that CF_3I/N_2 gas mixtures with 20%–30% CF_3I are suitable for use as the insulation medium in low-voltage electrical equipment. In addition, the AC withstand level of CF_3I/CF_4 gas mixtures is 59%–120% that of SF_6 and 15%–20% that of CF_3I/N_2 gas mixtures.

Considerable research has been conducted on PFC gases, including mixtures of c-C_4F_8, C_3F_8, and C_2F_6. Okabe *et al*. [143] measured the AC V_B of 10% PFC/90% CO_2 and 10% PFC/90% N_2 mixtures where the PFCs were CF_4, C_3F_8, and c-C_4F_8. They found that the breakdown characteristics of PFC/N_2 mixtures were better than those of PFC/CO_2 mixtures at a higher pressure range. The study of CF_3I gas mixtures with N_2, CO_2, and CF_4 showed that 20% and 10% were the optimal mixing ratios for c-C_4F_8/N_2 and c-C_4F_8/CO_2, respectively [144], [145]. The AC V_B of 20% c-C_4F_8/80% N_2 was 46%–90% that of 20% SF_6/80% N_2 under the same conditions and reached 57% that of SF_6 at 0.3 MPa. When 1% CO_2 was added to the 10% c-C_4F_8/90% N_2 gas mixture, its performance improved by 20%.

The C_3F_7CN and $C_5F_{10}O$ gases introduced by 3 M, USA, as gas dielectrics with high DS have attracted wide attention in the last few years [146]–[149]. Nechmi *et al*. measured the standard LI breakdown characteristics of C_3F_7CN/CO_2 mixtures with C_3F_7CN contents of 3.7%–20% [146]. The performance of 3.7% C_3F_7CN/96.3% CO_2 at 0.88 MPa was equal to that of SF_6 at 0.55 MPa, whereas the performance of this mixture at 1.04 MPa corresponded to that of SF_6 gas of 0.65 MPa. Zhang *et al*. [147] conducted AC breakdown tests using rod-plate and needle-plate systems. They found that in the range of 0.1–0.12 MPa, 20% C_3F_7CN/80% CO_2 with a small electrode gap performed better than SF_6 gas, giving this gas mixture potential to replace SF_6 in medium-voltage switchgear. Zhang *et al*. studied the AC V_B characteristics of C_3F_7CN/N_2 and C_3F_7CN/CO_2 gas mixtures [148]. The breakdown performance of C_3F_7CN/N_2 at 0.3–0.6 MPa reached 49%–63% compared with SF_6 gas at the lowest operating temperature of -25 °C and that of C_3F_7CN/CO_2 containing 2%–8% C_3F_7CN at 0.2 and 0.3 MPa reached 60%–71% and 56%–68% compared with SF_6, respectively. A study by ABB revealed that the DS of a $C_5F_{10}O/CO_2$ gas mixture at 0.7 MPa was slightly lower than that of SF_6 at 0.45 MPa and could reach the level of SF_6 at 0.6 MPa at a pressure of 1.0 MPa [149].

In 2018, Tu *et al*. [107] reported the DC breakdown characteristics of C_3F_7CN/N_2 gas mixtures at 126 kV using a needle-plate system. The C_3F_7CN/N_2 gas mixing ratios were 4:96 and 8:92, and the gas pressure ranged from 0.3 to 0.7 MPa. The results revealed that the DC breakdown characteristics of C_3F_7CN/N_2 gas mixtures were strongly affected by polarity. At the same gas pressure, for both types of gas mixtures, the negative V_B was much higher than the positive, and their difference increased with rising pressure. The magnitude of the negative V_B increased with pressure, whereas the positive V_B displayed the opposite behavior. The effect of polarity on V_B can be explained by the different discharge processes of C_3F_7CN gas under highly non-uniform electric fields. In addition, an interesting phenomenon was observed: the positive V_B with higher mixing ratio was lower than that of the gas mixture lower mixing ratio. Overall, these results provide a theoretical basis for the application of C_3F_7CN/N_2 gas mixtures in DC GILs.

Generally, rising pressure of a gas gap can reduce the average free path of electrons, hinder the development of collision ionization and effectively improve the breakdown voltage. However, under a highly non-uniform electric field, the V_B of the electronegative gas, whether AC, DC or impulse stress, may not increase linearly with the rise of pressure, but is greater than the corona partial voltage at lower pressure and tends to be consistent at higher pressure, showing a unique N-shape characteristic [150]. Scholars generally believe that this phenomenon of SF_6 and its substitute gases originates from the corona stabilization effect [151]–[155]. Corona discharge occurs at the tip of the needle electrode at lower voltages, resulting in a more uniform corona layer which improves the electric field distribution around the tip and thus increases the V_{B}. With increasing pressure, the diffusion of the corona layer is suppressed and a stable corona will not appear before breakdown, causing the reduction of the V_{B}. This phenomenon is more significant in positive polarity than in negative polarity, and is more significant under steady-state voltage than under impulse voltage [150].

The non-uniformity of electrodes will influence the electric field. In addition, the increase of a needle-plate (or rod-plate) gas gap distance and the enlarging of the length of a metal protrusion under a quasi-uniform background electric field equal to reducing the gap distance can enhance the non-uniformity, leading to the decrease of the $V_{B.}$ It is reported that the 50% double exponential impulse V_{B} of SF_6 at 0.5 MPa decreases linearly when the non-uniformity coefficient of the electric field f ranges from 30 to 70 [123]. Zhou pointed out that the test results with different types of electrodes (cone-plate and needle-plate electrodes) under highly non-uniform electric fields were divergent [151], which was similar to the literature [11]. The increase of V_{B} of the C_3F_7CN/CO_2 mixture shows a saturation trend with the rise of pressure at the cone-plate electrodes with lower f value, but shows a remarkable N-shape characteristic at the needle-plate electrodes, whose reason lies in the strength of the corona stabilization effect. In addition, Feng stated that the N-shape characteristics would also appear with the increase of the electrodes gap distance [156].

According to the relationship between DS and volume fraction, the binary gas mixture can be divided into positive synergistic effect, synergistic effect, linear relationship and negative synergistic effect. The positive synergistic effect makes the mixture's breakdown behavior better than that of single insulation gases, which helps to reduce the insulation distance and manufacturing cost of the device. Many scholars have studied the synergistic effects of various alternative gases in order to find the optimal with the highest DS. AC breakdown tests of 30%$CF_3I/CO_2/N_2$ and 30%$SF_6/CO_2/N_2$ ternary mixtures [142] show that no synergistic effect is observed with the increase of CO_2 content. For PCF mixtures and SF_6 mixtures, the synergistic effect of CO_2-based mixtures is better than that of N_2-based mixtures [144]. The synergistic effect of C_4F_8 and buffer gas is in the order of $SF_6/N_2 < C_4F_8/N_2 < C_4F_8/CO_2 < C_4F_8$/Air [109]. In addition, Zhang found that the synergistic effect of SF_6/N_2 gas mixtures under negative polarity LI was much stronger than that of the positive polarity [124]. The above experimental research on synergistic effect has made many achievements, but it is still necessary to further deepen the research on the micro-mechanism of the synergy effect.

Under the positive polarity voltage stress, the zone around the rod electrode where the electric field is concentrated is ionized first and then gathers the positive space charge, reducing the electric field around the rod electrode and strengthening the electric field of the external space. So the discharge is easy to develop forward and the positive V_{B} is low; when the negative polarity voltage is just the opposite, the V_{B} is higher [122], [141], [143]. However, under a highly non-uniform electric field, the results of electronegative gas gap may be contrary to the more common, namely the polarity reversal phenomenon. Zhang found that the polarity reversal of SF_6 gas appeared under the action of VFTO and LI under a highly non-uniform electric field [109].

3) Partial Discharge Properties

At present, the research on the PD of SF_6 substitute gases has focused on positive and negative half-cycle partial discharge initial voltage (PDIV$^+$ and PDIV$^-$, respectively) and 50% lightning impulse partial discharge initial voltage (LI PDIV $_{50\%}$); little research on DC PD behavior has been reported. However, with the recent large-scale construction of HVDC transmission systems, it has become necessary to focus on the performance of SF_6 substitute gases under DC PD.

To evaluate the PD behavior of SF_6 substitute gases, simultaneous measurements of the PD current and the corresponding light emission phenomena are widely used to study the discharge process from PD initiation to breakdown in highly non-uniform fields. Hayakawa and co-workers studied the PD and breakdown mechanisms of SF_6/N_2 gas mixtures under positive LI [157]. It is reported that there are two types of impulse PD modes: streamer discharge and leader discharge. And it was found that SF_6/N_2 gas mixtures showed a good synergistic effect. The PDIV $_{50\%}$ value of a mixture containing 2% SF_6 at 0.1 MPa reached 60% compared with SF_6 and 240% compared with N_2.

The PD behavior of conditional gases and new environmentally friendly gases including PFCs and CF_3I has been investigated [158]–[160]. Saitoh *et al.* [158] found that the LI PD ability of air at 0.25 MPa along with that of N_2 gas at 0.5 MPa was equivalent to that of SF_6 at 0.1 MPa, which means that both air and N_2 are suitable to replace SF_6 gas in C-GIS. The PDIV of C_4F_8/N_2 was equal to or higher than that of SF_6/N_2, and the PDIV of c-C_4F_8/CO_2 was 20%–30% higher than those of c-C_4F_8/N_2, C_4F_8/CO_2, and c-C_4F_8/air mixtures [109]. Under the same conditions, the PD performance of 10% PFC/90% CO_2 mixtures was superior to that of equivalent 10% PFC/90% N_2 mixtures [144]. The PDIV of c-C_4F_8 gas was 1.2–1.4 times higher than that of SF_6 gas. The PD synergistic coefficient C of C_4F_8 and N_2 gas mixtures is from 0.27 to 0.58. The PDIV of a 15% c-C_4F_8/85% N_2 gas mixture was 65% compared with SF_6 gas and the PDIV of 20% c-C_4F_8/80% N_2 was equal to that of 20% SF_6/80% N_2 and 0.70 times that of SF_6 gas. From the perspective of PD, a mixture of c-C_4F_8 and N_2 with 10%–20% C_4F_8 may be suitable to replace SF_6 gas in GILs.

Results show that the PDIV$^+$ and PDIV$^-$ of CF_3I/CO_2 were 1.2 and 1.0–1.4 times higher, respectively, than that of CF_3I/N_2 under AC voltages. That is, the synergistic effect of the former was larger than that of the latter [161]. Therefore, CF_3I/CO_2 is more suitable than CF_3I/N_2 as an SF_6 substitute in low and medium-voltage electrical equipment. The optimal CF_3I content was 25%–30% [162]. In addition, comparison of CF_3I/CO_2 and SF_6/CO_2 showed that the PDIV of CF_3I/CO_2 was 0.9–1.1 times that of SF_6/CO_2 with the same mixing ratio and the performance of CF_3I/CO_2 was slightly better than that of SF_6/CO_2. The PDIV of 30% CF_3I/70% CO_2 was about 70% compared with SF_6 [163]. The C value of CF_3I/CO_2 was about 0.53 and PDIV$^+$ of CF_3I/CO_2 with a CF_3I content of 30%–40% at 0.2 MPa reached that of SF_6 at 0.1 MPa. Therefore, CF_3I/CO_2 is promising to replace SF_6 gas in low-voltage electrical equipment, such as C-GIS [143].

The PDIV of 15% C_3F_7CN/CO_2 was equal to that of SF_6 at 80–120 KPa and slightly lower than that of 20% C_3F_7CN/CO_2 under the same conditions [148]. The PDIV$^-$ of C_3F_7CN/N_2 gas mixtures with 2%–12% C_3F_7CN at 0.3–0.6 MPa reached 62.8%–80.4% compared with SF_6 gas at a minimum operating temperature of -25 °C. The PDIV of C_3F_7CN/CO_2 gas mixtures with 2%–8% C_3F_7CN in the range of 0.1–0.3 MPa reached average values that were 78%–86% compared with SF_6 at 0.3 MPa and about 66%–80% compared with SF_6 at 0.15 MPa [149]. For electrode structures consisting of protrusion on conductor (POC) and protrusion on enclosure (POE) configurations, the PDIV of a 4% C_3F_7CN/96% CO_2 gas mixture was 76%–81% of that of SF_6 in POC mode and 78%–84% of that of SF_6 in POE mode [121]. In addition, the PDIV of 4% C_3F_7CN/96% CO_2 at 0.5 MPa was equivalent to that of SF_6 gas at 0.3 MPa. For application in GIE, the pressure or mixing ratio of C_3F_7CN needs to be increased.

In 2018, Tu *et al*. reported the DC PD characteristics of C_3F_7CN/N_2 gas mixtures at 0.5 MPa using needle-plate electrodes. The results are summarized in Table V. The mixing ratios of C_3F_7CN/N_2 were 4:96 and 8:92. For the C_3F_7CN/N_2 gas mixtures, PDIV$^-$ was higher than PDIV$^+$. The PDIV values of the C_3F_7CN/N_2 gas mixtures were higher than those of SF_6 gas under the same conditions. In addition, PDIV$^+$ of 4% C_3F_7CN/96% N_2 was higher than that of 8%C_3F_7CN/92%N_2, whereas PDIV$^-$ showed the opposite behavior.

TABLE V
DC PDIV OF C_3F_7CN/CO_2 MIXTURE AND SF_6 AT 0.5 MPA

Gases	PDIV$^+$ (kV)	PDIV$^-$ (kV)
SF_6	21.77	27.27
4%C_3F_7CN/96%CO_2	32.53	49.23
8%C_3F_7CN/92%CO_2	24.03	56.75

4) *Influence of Particles on Insulation Properties*

Particle jumping height, particle lift-off electric field, and crossing electric field are parameters used in basic investigations to compare substitute gases with SF_6. The relationship between particle movement (jumping height, lift-off, and crossing) and the electric field strength on the enclosure inner surface has been evaluated. In general, particle lift-off, jumping height, and crossing to the HV conductor were similar for SF_6 and other gases at low electric field strength. The lowest electric field strength was achieved for N_2. Compared with spherical metal particles, linear particles cause more electric field distortion and have the greatest effect on insulation degradation [164]–[166]. Distortion of the electric field near the tip of a linear particle causes corona discharge [167], [168], which ionizes numerous gas molecules in the region, as shown in Fig. 5. The ions generated by the micro discharge not only interacted with the metal particles, but also caused a large-scale jet motion under the action of a strong electric field to generate an ion wind. The interaction of electricity and the ion wind affected not only the movement of linear metal particles, but also gas breakdown.

Studying the breakdown characteristics caused by metal particles under different gases is an important test method for the engineering practicality of SF_6 replacement gas. S. A. Ward *et al*. compared the sensitivity of SF_6/N_2 and SF_6/air mixed gas to the local electric field strength surge caused by metal particles under DC voltage and proposed a better performance SF_6 mixed gas component [168]. Wang. *et al*. performed a comparative analysis of the discharge sensitivity of free metal particles in C_3F_7CN/CO_2 and SF_6/N_2 gas mixtures under a DC electric field [169]. It indicated that although the breakdown voltage induced by metal particles is similar in different ratios of gas mixtures, the DSP values of particle discharge sensitivity of 4% C_3F_7CN/96% CO_2 and 20% SF_6/80% N_2 are significantly lower than those of other ratios of gases.

C. *Flashover Properties of Gas-insulator Interface*

Surface flashover properties of gas-insulator interfaces are important for the design of gas-insulated systems. Moreover, under the stress of a DC voltage, surface charge accumulation results in distortion of the electric field, which could be specific cause of surface flashover. This section summarizes information on the basic flashover properties of insulators in substitute gases for SF_6 by presenting the latest experimental results in this area of research. The flashover performance of substitute gases for SF_6 has only been evaluated in a few of the flashover studies under AC and LI voltages. To be comparable with commercial GIL conditions, DC flashover experiments of downsized insulators with different substitute gases and the flashover studies of spacers at 0.1 MPa under electric field-temperature stress and strong LI should be conducted. Finally, the latest research progress of SF_6/N_2 and C_3F_7CN/CO_2 gas mixtures in $\pm$ 100 kV DC GILs is described. In addition, the influence of particles at gas-insulator interfaces on their insulation properties is discussed.

1) *Basic Properties*

The dielectric strength of clean and particle-contaminated gas–insulator interfaces with different types of gases and gas mixtures have been investigated. This section reviews such interfaces under clean conditions. The spacers were made of epoxy resin filled with aluminum oxide.

In 2016, Xiao *et al*. [170] studied the flashover characteristics of insulators in c-C_4F_8 under AC voltage stress. They found that the flashover voltage of c-C_4F_8 at 0.1 MPa was 1.2 times that of SF_6 gas. The flashover characteristics of c-C_4F_8 insulators showed more obvious polarity than those of SF_6. The impact breakdown voltage of negative lightning was much larger (about 1.3 times larger) than that of positive lightning. Regarding the surface flashover performance of c-C_4F_8/CO_2 insulators, the results showed that the content and pressure of c-C_4F_8 are important factors affecting the surface flashover performance of c-C_4F_8/CO_2 insulators under power frequency and LI voltages. At 0.10 MPa, the interfacial flashover voltage of 30% c-C_4F_8/70% CO_2 was 80% compared with SF_6, and at 0.30 MPa, the interfacial flashover voltage of 30% c-C_4F_8/70% CO_2 was about 1.4 times that of SF_6 at 0.10 MPa. It is noteworthy that after many experiments, the damage of insulators in a c-C_4F_8 gas environment was more severe than that in CO_2 and SF_6 environments. When using c-C_4F_8, attention should be paid to the protection of insulators.

In 2016, Xiao *et al*. found that under 0.1 to 0.3 MPa, the relationship between the power frequency flashover voltage of insulators in the mixture of CF_3I/N_2 and SF_6/N_2 is as follows: 30% CF_3I/70% N_2 >30% SF_6/70% N_2 >20% SF_6/80% N_2 [171].

The flashover voltages for a 30% CF_3I/70% N_2 gas mixture were similar to those of SF_6/N_2 mixtures with 20% and 30% SF_6 gas.

The flashover characteristics of insulators in CF_3I/N_2 gas mixtures under DC voltages in the pressure range of 0.4 to 0.7 MPa were studied experimentally by Tu *et al*. [106]. The flashover voltage of the insulator in the CF_3I/N_2 mixture was much lower than those in SF_6 and SF_6/N_2 gas systems. It is therefore considered that CF_3I/N_2 gas mixtures are not suitable for use in high-pressure DC GIE.

Wang *et al*. found that the breakdown and flashover characteristics of c-C_4F_8/N_2 gas mixtures under DC voltages were lower than those of pure SF_6 gas when the mixing ratio was 10%–20% [110]. Thus, c-C_4F_8/N_2 gas mixtures are very promising to replace SF_6 as the insulating gas if the gas pressure or insulating distance is increased. However, the problem of decomposition and precipitation of carbon particles in c-C_4F_8 during repeated discharge will cause a short-circuit of the air gap around insulators, which will damage the insulation structure.

Polarity effect of flashover properties of insulators was investigated in C_3F_7CN/CO_2 gas mixtures under DC voltage [172]. It shows the positive and negative DC flashover voltages of C_3F_7CN/CO_2 mixtures increased with increased gas pressure. The negative DC flashover voltages of 4% C_3F_7CN/ 96% CO_2 and 8% C_3F_7CN/92% CO_2 mixtures at 0.7 MPa were 96.06% and 101.50% compared with SF_6 at 0.5 MPa.

In order to study the flashover properties of spacers under the combined electric stress and thermal stress, Ma *et al.* set up an experimental platform, which consists of a coaxial cylindrical test model and an external-circulation oil heating system. The DC superimposed lightning impulse voltage is tested with 110 kV spacers with a SF_6/N_2 gas mixture, at 0.1 MPa. The experimental results in Fig. 7 show that with the increase of pre-added DC voltage duration time, the flashover voltage under composite voltage is significantly lower than that under single lightning impulse voltage. The flashover voltage of the insulator with clean surface decreases by 10.34% compared with that of the insulator with impulse voltage acting alone when the surface charge accumulates and saturates [173].

Based on the results of downsized insulators in different types of Environment-friendly gases, we performed recent

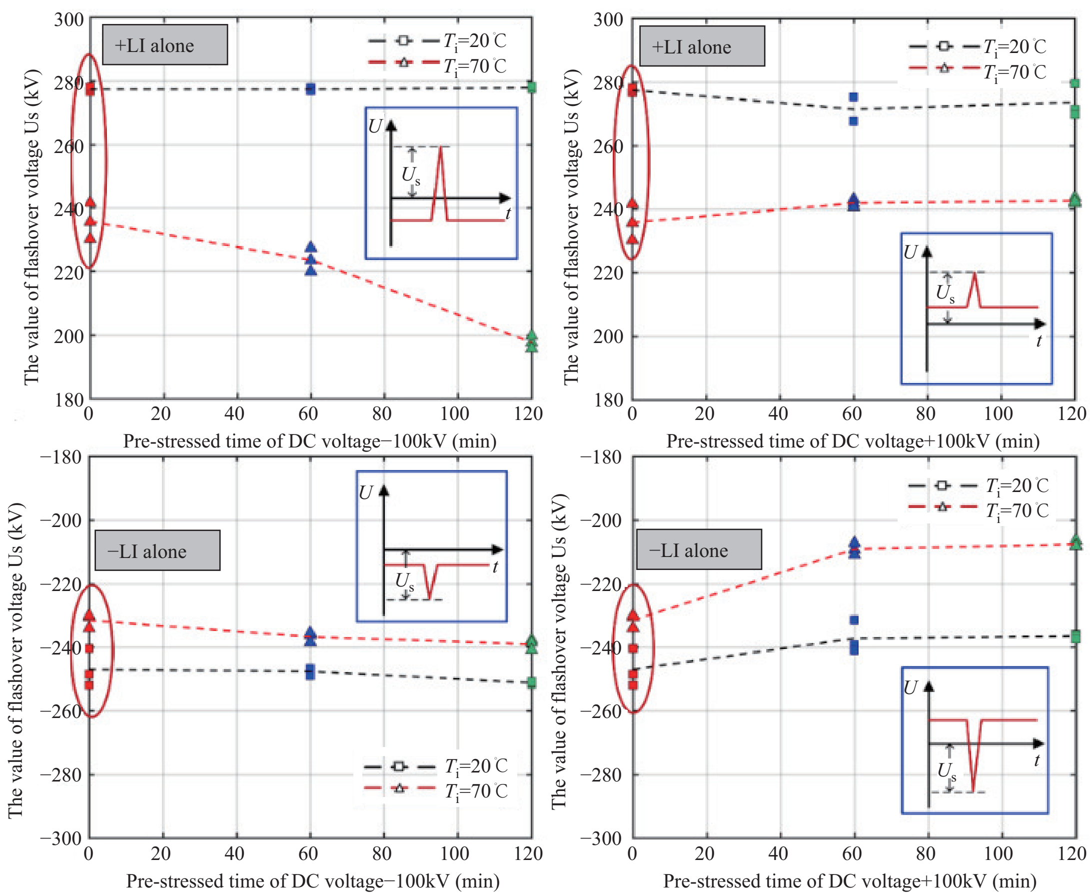


Fig. 7. Flashover traces of insulators after DC superimposed impulse tests [173].

studies into the flashover properties of spaces in $\pm$ 100 kV DC GIL prototype filled with 0.7 MPa, SF_6/N_2 gas mixture and 0.7 MPa, C_3F_7CN/CO_2 gas mixtures under combined voltage of DC with lightning impulse. The test scheme is to apply a positive 100 kV voltage to the DC GIL prototype, maintain the voltage for 2 hours, and then apply a negative lightning impulse voltage to the prototype to flashover for three consecutive flashover times. The time between the two continuous pulses should not be less than 1 minute. The flashover voltage should exceed over 5.5 times that of the rated voltage based on the 100 kV DC voltage.

Superimposed LI voltage test results in Fig. 8 show that the flashover voltage of DC insulators in a 20% SF_6/80% N_2 gas mixture at 0.7 MPa reached 915 kV [174]. The flashover voltage of an insulator in a 4% C_3F_7CN/96% CO_2 gas mixture at 0.7 MPa was 717 kV. These results both exceeded the assessment standard of 650 kV. These novel gas mixtures can pass positive DC voltage and negative LI tests. These environmentally friendly gases can meet the insulation requirements under DC voltage. The main assessment results for these mixtures are presented in Table VI.

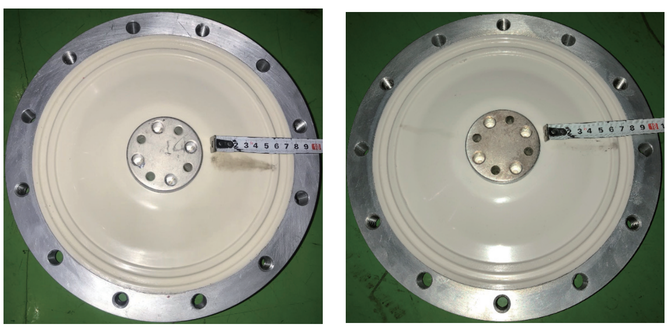


Fig. 8. Flashover traces of insulators after DC superimposed impulse tests.

TABLE VI
TEST RESULTS OF DC SUPERIMPOSED LI VOLTAGE OF INSULATORS IN ENVIRONMENT-FRIENDLY GAS MIXTURES

Gas Type	Flashover Voltage (kV)
20%SF_6/80%N_2	915
4%C_3F_7CN/96%CO_2	717

2) *Influence of Particles on Flashover Properties*

The influence of these defects on insulation properties are important aspects for design, manufacturing conditions and service of gas-insulated systems. When there is metal particle fixing, adsorping or sliding along the surface of an insulator, the flashover voltage will be seriously lowered [175]–[179]. Investigation results, comparing different types of gases, are reported in the literature [180]–[184], including SF_6, air, N_2 and CO_2 under AC and LI voltages. Under DC voltage, the SF_6 is still the main research area for metal particles. The comparative study of different new environmentally friendly gases is rarely reported.

The position(s) at which the particles attach to the surface of the insulator and the particle shape and size are important factors affecting the surface flashover of the insulator. For cone-type insulators, when the attached particles are located on the side near the high-voltage conductor, the flashover voltage is the lowest. At a certain position on the surface, the longer the linear particle or larger the diameter of the spherical metal particle, the lower will be the flashover voltage of the insulator. The adhesion of metal particles causes charge accumulation and distortion of the surrounding electric field, which is a necessary condition for initiating the lead and streamer. The current research primarily focuses on the influence of fixed metal particles on the surface charge and the flashover voltage along the insulator surface. However, the mechanism of the evolution of the surface charge on the insulator surface is not clear during the whole process of the development of the surface discharge, and the cooperative induction mechanism between particles and charges on the surface flashover remains to be established. In addition, the corresponding movement conditions should be considered, such as adsorption and sliding along the surface, which causes a greater influence. Moreover, the random disturbance of particle motion and the interaction of surface charge are still unclear, especially in different gases.

IV. RE-EXAMINATION: THERMAL PROPERTIES

Current flowing through gas-insulated equipment generates heat. The associated temperature rise not only reduces the maximum current-carrying capacity of the conductor, but also affects the performance of the insulation material and reduces the service life of the equipment [185]. The SF_6 gas used in GILs exhibits high thermal conductivity and allows heat to dissipate to keep the temperature rise in GILs at an acceptable level. If SF_6 is substituted with another gas, it is necessary to examine the thermal properties of the new gas. There is a major difference between AC and DC GILs in terms of the heat source. Unlike the AC GILs, when the current passes through the internal conductor in a DC GIL, no skin effect occurs. Therefore, the equivalent resistance of the DC GIL decreases. In addition, no eddy current is induced in the enclosure. This allows the DC GILs to have a higher current capacity than that of equivalent AC GILs. At the same time, this feature also provides more design space for the potential improvement of the heat transfer performance of alternative gases to SF_6.

The physical model of convective heat transfer in GIL is shown in Fig. 9. The heat transfer characteristics and temperature rise effected in GILs can be determined via experimental testing and numerical calculations.

In 2016, GE Grid took the temperature rise test in their novel 420 kV GIL filled C_3F_7CN/CO_2 gas mixture [186]. Due to the large share of CO_2 in the mixture, the thermal performance of g3 gas is dominated by the gas CO_2. Investigations have demonstrated a reduction of approximately 10% of heat dissipation when using the same filling pressure as in SF6.

Based on of the studies described above, Chen. *et al*. established a temperature rise test platform for experimental studies and a three-dimensional (3D) simulation model for GILs using the thermal–fluid coupled finite element method [187]. Combining these two research methods, the corresponding heat transfer performance and temperature rise experiments were carried out. The results in Fig. 10 indicates that increasing

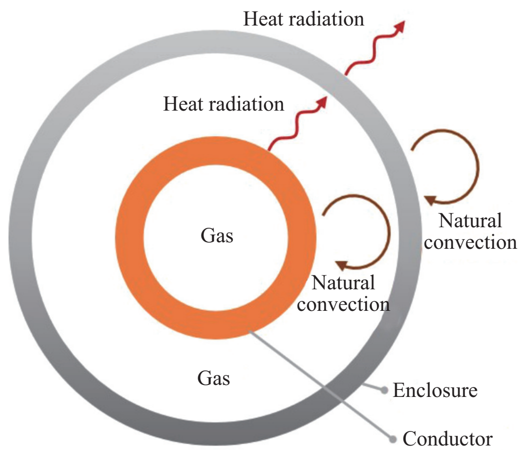


Fig. 9. Heat transfer mechanism of GIL bus bar.

the C_3F_7CN gas component ratio or pressure of the insulating gas mixture could improve the heat dissipation and current-carrying capacity of GILs. The relationship between the heat transfer characteristics of AC and DC GIL was considered. A DC GIL has a higher current capacity than the equivalent AC GIL, offering a good solution for high-capacity energy transmission.

V. Multi-Performance Cooperative Control Model for Insulation Gas Mixtures

Higher insulation performance usually requires gases with strong electronegativity, but most such gases are toxic with unstable chemical properties and low saturated vapor pressure. There are trade-offs between high dielectric strength and liquefaction temperature and high dielectric strength and low toxicity of gases. For the above reasons, although there are hundreds of alternative gases to SF_6, environmentally friendly insulating gases that can completely replace SF_6 are still being explored.

Based on the GWP of alternative gases, the dew point temperature distribution of gas mixtures, and their insulation characteristics, a coordinated control model of alternative gases to SF_6 was established for several environmentally friendly insulating gases, as shown in Fig. 11.

In terms of environmental friendliness, the goal is to lower the GWP to less than 30% of that of SF_6. According to the operational experience of HVDC GILs using alternative gases and the insulation performance of potential alternative gases, the insulation performance of alternative gases to SF_6 generally reaches 70%–80% that of SF_6. Charge accumulation of insulators in HVDC GILs induces electric field distortion and the flashover of cone-type insulators along the surface will be induced. As a result, the flashover voltage drops by about 10% [188]. Therefore, the insulation strength of SF_6 alternatives needs to be more stringently controlled in HVDC GILs than in the case of AC GILs. It has been suggested that the substitute gases for SF_6 should be screened according to the breakdown field strength of the gas and maximum field strength of DC flashover at 0.4 MPa with 95% of the insulation strength of SF_6 as the lower limit.

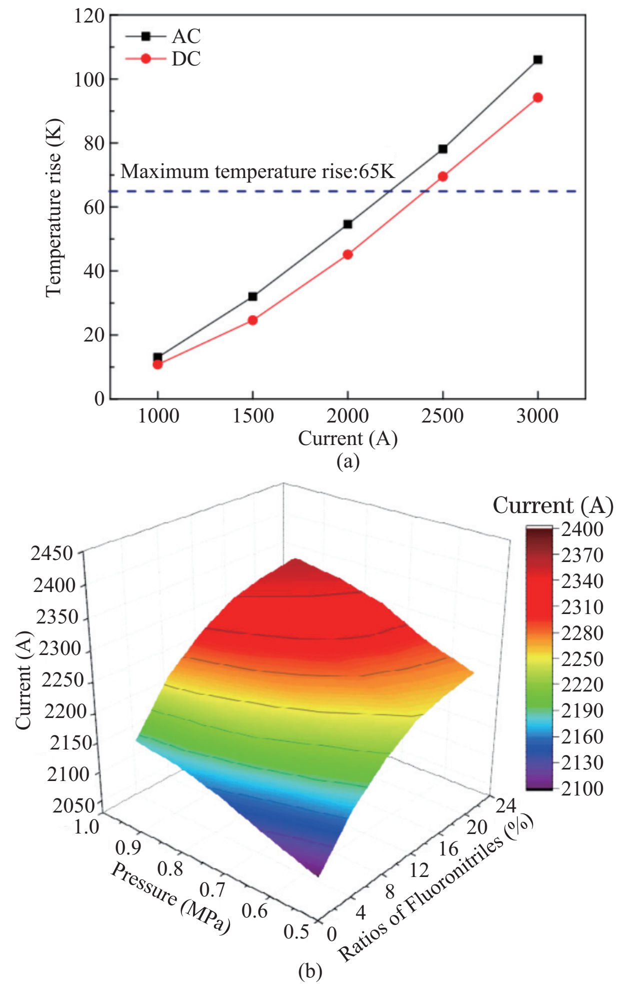


Fig. 10. Analysis on temperature rise of an environmentally friendly GIL and current-carrying capacity design. (a) Comparison of temperature rises of AC and DC-GIL conductors at different load currents. (b) Relationships between the current-carrying capacity of the GIL conductors and the gas pressure and ratio of C_3F_7CN [187].

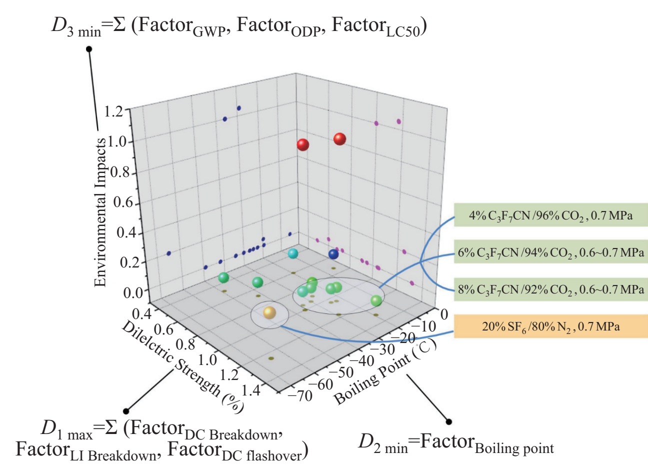


Fig. 11. Multi-performance cooperative control model for alternative gases to SF_6.

According to IEC 62271–1, the application environments of environmentally friendly alternative gases can be divided into cold areas, normal outdoors, and normal indoors. The liquefaction temperature of a 20% SF_6/80% N_2 gas mixture at 0.7 MPa is −63.4 °C, which means that this mixture can be used in cold areas. Meanwhile, 4% C_3F_7CN/96% CO_2 at 0.7 MPa can be used in DC GILs under most conditions to realize the complete replacement of SF_6 gas because the liquefaction temperature of this mixture is −34.1 °C. The insulation performance of 6%–8% C_3F_7CN/92%–94% CO_2 mixtures at pressures of 0.6–0.7 MPa is better than that of a 4% C_3F_7CN/96% CO_2 gas mixture. The insulation performance of SF_6 gas is 101% to 123% at 0.4 MPa. Because of its liquefaction temperature range of −19.4 to −25.5 °C, it is recommended that 6%–8% C_3F_7CN/92%–94% CO_2 mixtures are applied under indoor conditions or in areas with high temperatures. If both SF_6/N_2 and C_3F_7CN/CO_2 gas mixtures can meet the application requirements, the use of the C_3F_7CN/CO_2 mixture is preferred. Application principles and selection suggestions for alternative gases to SF_6 for use in environmentally friendly HVDC GILs are listed in Table VII.

TABLE VII
THE TYPICAL APPLICATIONS

Application temperature (°C)	Gas	Gas Pressure (MPa)
−50	20% SF_6/80% N_2	0.7
−30	4% C_3F_7CN /96% CO_2	0.7
−10	8% C_3F_7CN /92% CO_2	0.6–0.7

VI. CONCLUSION

This paper reviewed the research methods and properties of environmentally friendly gases for DC gas-insulated equipment. In particular, the progress of research on the main alternative gases to SF_6 was described in detail. A multi-performance cooperative control model for alternative gases was established, and the gas application recommendations for DC GILs were given. We hope that this review with inspire the further directed exploration of alternative gases to SF_6.

Despite the existing research, there are still many topics that should be considered in future studies, including, but not restricted to the areas mentioned below.

1) Electron affinity and molecular ionization energy are difficult to obtain accurately and there is no obvious quantitative relationship between the dielectric strength of an insulating gas and microscale parameters such as ionization energy and electron affinity. It is still difficult to establish an accurate mathematical relationship between microscale parameters and macroscale electrical performance with clear physical meaning.

2) The existing results show that a mixture of electronegative gas and buffer gas can provide improved insulation and liquefaction performance to realize alternatives to SF_6. The synergistic effect of mixed gases and the microscale mechanism of this effect need to be further explored.

3) Searching for appropriate environmentally friendly insulating gases is a systematic project that needs to be considered from many aspects, including insulation properties, liquefaction, and heat dissipation. It is necessary to consider the optimization of the comprehensive performance of gases under multifactor coupling conditions.

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