# Research status of replacement gases for SF<sub>6</sub> in power industry

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🔟 Shuangshuang Tian, ២ Xiaoxing Zhang, ២ Yann Cressault, et al.





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Shuangshuang Tian,<sup>1</sup> Xiaoxing Zhang,<sup>1,a)</sup> Yann Cressault,<sup>2</sup> Juntai Hu,<sup>3</sup> Bo Wang,<sup>3</sup> Song Xiao,<sup>4</sup>

#### AFFILIATIONS

<sup>1</sup>Hubei Key Laboratory for High-efficiency Utilization of Solar Energy and Operation Control of Energy Storage System, Hubei University of Technology, Wuhan 430068, China

<sup>2</sup>LAPLACE (Laboratoire Plasma et Conversion d'Energie), UPS, INPT, Université de Toulouse, 118 route de Narbonne, CO F-31062 Toulouse Cedex 9, France

<sup>3</sup>State Grid Pingdingshan Power Supply Company, Pingdingshan 467001, China

<sup>4</sup>School of Electrical Engineering, Wuhan University, Wuhan 430072, China

#### <sup>a)</sup>Author to whom correspondence should be addressed: xiaoxing.zhang@outlook.com

#### ABSTRACT

 $SF_6$  is widely used in the industrial field due to its stable structure and excellent properties. It is mainly used in electrical insulation equipment. Due to the boiling point of  $SF_6$ , its use in extremely cold regions has been limited. It is harmful to the health of practitioners due to the toxicity of decomposition products. The gas has limited its wider use because of its strong greenhouse effect. As a result, researchers and electrical equipment manufacturing companies around the world are gradually searching for new environmentally friendly gases and have conducted research and exploration on theory and experiment. In this paper, the current status and existing problems of  $SF_6$  are summarized. The research contents and research methods of  $SF_6$  alternative gas direction are reviewed from the aspects of insulation performance, interrupter performance, and decomposition performance. The existing research results of the natural gas,  $SF_6$  mixed gas, perfluorocarbons, and  $C_nF_mX$ gas are summarized, and the future development trend of alternative gas for  $SF_6$  in the electrical industry is proposed.

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#### **I. INTRODUCTION**

In 1900, an artificial inert gas called sulfur hexafluoride  $(SF_6)$  was synthesized by two French chemists, Moissan and Lebeau.  $SF_6$  is composed of the most active elements in the halogen, fluorine (F) and sulfur (S) atoms.

The S atom is bonded by the sp3d2 hybrid orbit, and the molecular structure is an octahedron, in which the six F atoms are at the fixed position and the S atom is in the central position. S and F atoms are linked with covalent bonds, as shown in Fig. 1.<sup>1</sup> It has a stable chemical structure and strong insulation capacity, and the arc-extinguishing capacity of SF<sub>6</sub> is 100 times that of the air, and the insulation capacity is 2.5 times that of the air. It is a new ultra-high voltage dielectric insulation gas, which is superior to air and oil. Due to the excellent insulation properties and arc

extinguishing performance, it is used in electrical equipment as gasinsulation media, such as circuit breakers, high voltage transformers, gas sealed combination capacitors, high voltage transmission lines, and transformers.

High purity SF<sub>6</sub> is an ideal electronic etchant, which is widely used in microelectronic technology. SF<sub>6</sub> is also used as a refrigerant in the refrigeration industry, and the refrigeration range can be between -45 °C and 0 °C. SF<sub>6</sub> is used as an anti-adsorbent in the mining industry to replace oxygen in coal mine dust.

 $SF_6$  is widely used in metal melting (such as magnesium alloy melting furnace protective gas), aerospace applications, medical applications (x-ray machine and laser), meteorological tracer analysis, chemical applications (senior car tires and new fire extinguishers), and so on. With the development of science and technology, the fields covered by  $SF_6$  are constantly expanding more and more,



such as basic fields and science and technology fields. With the continuous development of the power grid and the gradual increase in the voltage level of the equipment, the usage of  $SF_6$  is also gradually increasing. However, since  $SF_6$  is a typical greenhouse gas and the demand for limiting its use is getting higher and higher, the research on alternative gases is going deeper and deeper. Of course, in addition to the greenhouse effect, the toxicity of  $SF_6$  decomposition products and the boiling point are also factors that limit their use.

This paper reviewed the research status of  $SF_6$  replacement gases in order to help researchers study the new substitute gases. This paper is organized as follows: Sec. II introduces the properties of SF<sub>6</sub>, which is the important reason that it can be used widely in the power industry, and points out the disadvantages, which is the reason for looking for substitute gases. Section III focused on the main evaluation indices of alternative gases, such as the gas insulation, arc extinguishing, and decomposing properties. Section IV lists the research results of natural gases, SF<sub>6</sub> mixtures, perfluorocarbons, and other gases based on the evaluation indices in Sec. III. Section V shows the conclusion and outlook about the SF<sub>6</sub> replacement gases.

#### II. SF<sub>6</sub>

#### A. The use and development of SF<sub>6</sub> in power industry

#### 1. Insulation properties

SF<sub>6</sub> is not permanently broken down as solid insulation, and the insulating properties can be comparable to those of oil if the pressure is large enough, so it can be used in gas-insulated equipment. Under severe discharge and high temperatures, pure SF<sub>6</sub> is relatively stable and self-recovery performance is better, and no harmful solid impurities are precipitated by gas decomposition. Camilli and Chapman<sup>2</sup> compared various gases (SF<sub>6</sub>, N<sub>2</sub>, CF<sub>2</sub>Cl<sub>2</sub>, CF<sub>3</sub>Cl, CF<sub>4</sub>, CHClF<sub>2</sub>, and CHF<sub>2</sub>) by testing the lightning impulse voltage and the 60 Hz AC voltage. SF<sub>6</sub> was found to be the most ideal insulating gas in terms of the boiling point and insulation properties. It can maintain good insulation properties even at lower pressure. Camilli *et al.*<sup>3</sup> also explored the feasibility of using gas-insulated media in

high-voltage transformers by analyzing the electrical properties and chemical properties. Compared with the transformer oil, the gas cooling capacity is relatively poor, so the application of SF<sub>6</sub> in the transformer may require an additional cooling system or increase in the volume of equipment. Menju *et al.*<sup>4</sup> studied the insulation properties of SF<sub>6</sub> in the gap between the ball electrode and the coaxial cylinder. By measuring the breakdown time T and the breakdown voltage V, it is considered that the VT curve (the abscissa represents T, and the ordinate represents V) of SF<sub>6</sub> is relatively flat, and the gas can well protect the device from over-voltage for a long time. This provides a theoretical basis for the application of SF<sub>6</sub> in gas-insulated transmission lines.

As the voltage level is gradually increased, the advantages of DC transmission equipment are obvious. The demand for gas-insulated equipment at DC voltage is driven. Menju and Takahashi<sup>5</sup> tested the DC breakdown characteristics of different electrodes and concluded that the combination of SF<sub>6</sub> gas and epoxy resin can be effectively used in the insulation system of a high voltage DC gas insulated substation. Rizk *et al.*<sup>6</sup> studied the movement characteristics of conductive particles between coaxial cylindrical electrodes and their effect on the DC breakdown voltage by experiments. They improved the model of particle motion in a coaxial field, considering that particles were subjected to charge accumulation and centrifugal forces.

For the microscopic parameters of gas discharge, Boyd and Crichton<sup>7</sup> measured the ionization coefficient and attachment coefficient of the SF<sub>6</sub> molecule, and these parameters provide important references for the study of the gas discharge process. In the range of 25 °C–125 °C, the temperature does not affect the electron attachment process of the gas molecules, but the increase in temperature would promote the diffusion process, thus changing the space charge distribution and eventually causing the breakdown voltage to rise.<sup>8</sup>

#### 2. Arc extinguishing properties

The interruption ability of SF<sub>6</sub> was significantly higher than that of air.<sup>9</sup> In 1956, Cromer and Friedreich<sup>10</sup> found that the interruption ability of SF<sub>6</sub> was significantly higher than that of air, the arc extinguishing capacity of plain-break arcs at AC voltage was about 100 times of that of air, and the limiting current value of the arc extinguishing current had a rough linear relationship with the arc length and the gas pressure. The SF<sub>6</sub> circuit breaker could cut off the current of 5000 A at 115 kV. In 1957, Leeds et al.<sup>11</sup> measured the current change in SF<sub>6</sub> during quenching. By installing a cross-blast and arc-splitter, the interrupting current can be increased as demonstrated by Kane and Colclaser in 1963<sup>12</sup> in a 69-kV SF<sub>6</sub> common-tank breaker. They concluded that SF<sub>6</sub> as the interrupter medium results in a perfect switching and interruption effect. In 1964, Easley and Telford<sup>13</sup> designed a new type of circuit breaker, which was quenched by a new jet-type interruption structure. The interruption and impulse withstand tests of the new circuit breaker reached the standards of the American Standards Association (ASA).<sup>1</sup>

In 1965, the first SF<sub>6</sub> 500 kV circuit breakers were started to be used in the USA.<sup>15</sup> 1550-kV basic insulation level (BIL) circuit breakers with SF<sub>6</sub> as the insulating gas also appeared one after another.<sup>16</sup> ASA (American Standards Association) reported the use of 115 kV-345 kV circuit breakers with SF<sub>6</sub> as the

arc-extinguishing medium in terms of the mechanical structure, working principle, and arc extinguishing performance, and extended it to the corresponding extra high voltage (EHV) transmission lines.<sup>17–21</sup>

Due to its superior performance, SF<sub>6</sub> was considered as an insulating medium and arc-extinguishing medium in electrical equipment.<sup>22</sup> SF<sub>6</sub> is today widely used in the power industry in gas insulated circuit breakers (GCBs), gas insulated switchgears (C-GISs), gas insulated circuits (GILs), and gas insulated transformers (GITs),<sup>23–30</sup> and the current production of SF<sub>6</sub> is mainly used in the power industry, accounting for about 80% of the total production.<sup>31</sup>

Since then, SF<sub>6</sub> gas-insulated equipment has been developed and perfected for decades. SF<sub>6</sub> occupies an important position in the gas-insulated equipment, and the theoretical and experimental research on SF<sub>6</sub> has never been interrupted. For example, SF<sub>6</sub> is used as the insulation medium and arc extinguishing medium in the 1000 kV GIS, and the rated short-time withstand current reaches 50 kA. With the continuous development of the power grid and the gradual increase in the voltage level of the equipment, the usage of SF<sub>6</sub> is constantly increasing despite its disadvantages.

#### **B.** The disadvantages

In the recent decades, it has been found that there are mainly three problems about the use of SF<sub>6</sub> in electrical equipment. The first one is the boiling point, which cannot be used in electrical equipment in extremely cold areas. The second one is toxicity since SF<sub>6</sub> may be decomposed in the equipment to produce some toxic gases such as SO<sub>2</sub>, SO<sub>2</sub>F<sub>2</sub>, SOF<sub>2</sub>, and S<sub>2</sub>F<sub>10</sub>, which can react with metals to produce various metal sulfides. Finally, SF<sub>6</sub> is a typical greenhouse gas. With the increase of usage, the impact on the environment cannot be neglected. The content of SF<sub>6</sub> in the atmosphere increased by 20% from 2011 to 2016, which is also the most important reason for the urgent need to find a new type of green gas instead of SF<sub>6</sub>. According to forecasts, by 2030, SF<sub>6</sub> emissions from the power sector alone will reach  $64 \times 10^6$  metric tons of carbon dioxide equivalent (MtCO2e).<sup>31</sup> So, limiting their use and emissions has become the current requirement for environmental protection and sustainable development.

#### 1. The boiling point

The boiling point of SF<sub>6</sub> is -65 °C (0.1 MPa). This temperature meets the operating requirements of most equipment, but liquefaction still persists in extremely cold regions, which needs a lower boiling point. In the initial gas-insulated equipment design and development phase, a variety of insulating gases were considered within the scope. All kinds of insulating gases were tested, and theoretical analysis was performed by research workers and electrical equipment companies such as General Electric (GE). The initial purpose of exploration has always been simpler, considering the insulating properties and the boiling point as the main purpose.

In 1980, Devins<sup>32</sup> from General Electric (GE) Company conducted research on the key parameters such as dielectric strength and boiling point of thousands of gases and screened out relatively high-performance insulating media. Most of the gases in the insulation strength and boiling point cannot reach the SF<sub>6</sub> level at the same time, but the impact on the environment is ignored. The research on the SF<sub>6</sub> gas mixture started earlier and almost at the same time as the research on SF<sub>6</sub> gas. It has been a hot topic, and it has been a breakthrough for the SF<sub>6</sub> gas mixture until 2000. The study found that the appropriate reduction in the SF<sub>6</sub> mixing ratio can meet the needs of equipment operation and solved the media liquefaction in the equipment. The SF<sub>6</sub> mixed gas was begun to be used in the GIL.

#### 2. The toxicity of decomposition products

Even if a new SF<sub>6</sub> gas insulated device can be non-toxic, it may contain toxic gases after a certain period of operation.<sup>33</sup> Despite the good chemical inertness, the decomposition of SF<sub>6</sub> still occurs under the arc with low-fluoride sulfides (SF<sub>2</sub> and SF<sub>4</sub>) produced by breaking the S-F bond. Under the conditions of high voltage and high temperature during discharge, SF<sub>6</sub> decomposes to form various free radicals, which can react with water, oxygen, and solid insulating material epoxy resin  $[(C_{11}H_{12}O_3)_n]$  in equipment to form SO<sub>2</sub>, SO<sub>2</sub>F<sub>2</sub>, SOF<sub>2</sub>, and so on. A very small amount of water molecules and oxygen molecules (ppm level) is inevitable in high purity SF<sub>6</sub>. Vanroggen *et al.*<sup>34</sup> noted that the reaction of  $SF_6$  with other gases can produce toxic low-fluoride sulfides, as well as oxide SOx, and even the highly toxic S<sub>2</sub>F<sub>10</sub>, HF, and others. Vijk explored the reaction of SF<sub>6</sub> with the metal in the circuit breaker by simulating the environment of the circuit breaker and measuring the consumption of different metal electrodes.<sup>35</sup> Due to the impurities contained in SF<sub>6</sub>, as well as metal and solid insulation materials in electrical equipment, the decomposition products also include CS<sub>2</sub>, WF<sub>6</sub>, and SiF<sub>4</sub>.

Some substances are toxic gases and cause health problems to workers in the power industry. Kraut and Lilis<sup>36</sup> reported that six electrical workers were exposed to SF<sub>6</sub> decomposition products during electrical service for more than 6 h (within 12 h) without protection. Most notably, the decomposition gas has a significant effect on the lung function. The threshold values (TLVs) of the main decomposition products SF<sub>4</sub>, S<sub>2</sub>F<sub>10</sub>, SO<sub>2</sub>, SOF<sub>2</sub>, SO<sub>2</sub>F<sub>2</sub>, and HF are also given. Pilling and Jones<sup>37</sup> described a series of symptoms of dyspnea, hemoptysis, and cyanosis when two power workers inhaled SF<sub>6</sub> decomposition products for on-site maintenance and pointed out that the main decomposition products of SF<sub>6</sub> has a direct effect on the alveolar capillary membrane, which might lead to pulmonary edema, hypoxemia, arrhythmia, and mild tissue acidosis based on the clinical observation. Table I shows the possible damage to the human body caused by the main gas decomposition products of SF<sub>6</sub>.

#### 3. The greenhouse effect

With the global warming, the use and emission of greenhouse gases have drawn more and more attention from all over the world. Obama pointed out in "Science" that if the greenhouse gas emission is not limited, the global average temperature may increase by  $4 \,^{\circ}$ C or more by 2100.<sup>41</sup> The Kyoto Protocol in 1997 classified the types of greenhouse gases. It was clear that SF<sub>6</sub> is one of six greenhouse gases. It was required that the use of SF<sub>6</sub> should be gradually reduced by 2020.<sup>42</sup> The GWP value of SF<sub>6</sub> is about 23 900 times that of CO<sub>2</sub> with a lifetime of 3200 years. The content of SF<sub>6</sub> gas in the atmosphere has increased by 8.7% per year.<sup>43</sup> So far, the SF<sub>6</sub> gas accounts for more than 15% of the total greenhouse gase reached 0.95 pg, which would reach 3.7 pg in 2050 keeping with the current

The decomposition gases	Health effects	Threshold limit value (TLV) (ppm)
SF <sub>2</sub>	Not toxic	
	Corrosive: Inhalation, skin, eye, and ingestion. It can cause	
SF <sub>4</sub>	lung damage, which affects the respiratory system; its toxicity	0.1
	its toxicity is similar to phosgene toxicity	
$S_2F_2$	Toxic and pungent smell. Destructive to the respiratory system	0.5
SOF <sub>4</sub>	Corrosive: Inhalation, skin, and eye	0.5
	Vascular-shock	
$S_2F_{10}$	Lungs, thorax, or respiration-acute pulmonary edema	0.01
	Blood—changes in the cell count (unspecified)	
	Strong irritation to the upper respiratory tract and lungs.	
SO <sub>2</sub>	It is mainly used in the upper respiratory tract. A large number of	2
	acute exposure can cause pulmonary edema and respiratory paralysis.	
SOF <sub>2</sub>	It can cause severe pulmonary edema and mucous membrane irritation.	1.6
$SO_2F_2$	Inhalation of this odorless gas can quickly result in death.	5
	Very hazardous in the case of skin contact (corrosive and irritant), eye contact (irritant	
HF	and corrosive), and ingestion. Inhalation of the spray mist may produce severe irritation	3
	of the spray mist may produce severe irritation of the respiratory tract,	
	characterized by coughing, choking, or shortness of breath.	
CS <sub>2</sub>	Extremely hazardous in the case of skin contact (irritant), ingestion, and inhalation.	10
CO	It combines with hemoglobin in the blood to cause hypoxia.	50
WF <sub>6</sub>	Very toxic and intensely irritating	0.1
	Inhalation, skin, eye, and mucous membrane contact. High concentration	
SiF <sub>4</sub>	exposures may produce pulmonary irritation, and low concentration	0.6
	exposures may produce generalized effects of fluorine exposure	0.6

TABLE I. Main clinical symptoms caused by decomposition products of SF<sub>6</sub>.<sup>38–40</sup> Notation: the TLV is a time-weighted average concentration at which no adverse health effects are expected, for exposure during 8 h/day, for up to 40 h/week, in ppm<sub>vol</sub>.

emission levels.<sup>44</sup> Since 1997, the USA, Japan, and the EU countries have formulated policies and measures to limit the use of SF<sub>6</sub>. California proposed that the use of SF<sub>6</sub> in the electrical field should be reduced year by year from 2020 and the EU plans to reduce SF<sub>6</sub> emissions to 2/3 of 2014 by 2030. Spain and Australia have increased the tax of SF<sub>6</sub> emissions. It can be said that the main reason for the research on SF<sub>6</sub> alternative gas still comes from its threat to the environment. In addition, the policy of restricting the use of SF<sub>6</sub> in various countries gradually makes the study of alternative gases a hot issue in the global power industry.

Based on the above reasons, it has become an important research subject in the power industry to look for a new type of environmentally friendly gas with good insulation ability and safety. Research institutes across the world are dedicated to finding suitable gas alternatives to  $SF_6$  for use in gas-insulated power equipment such as GISs and GILs.

#### **III. EVALUATION INDICES OF ALTERNATIVE GASES**

Alternative gases should be safe, harmless, and environmentally friendly and should meet the needs of equipment operation, such as the boiling point, insulation properties, interrupter performance, and decomposition characteristics. At present, the gas with alternative potential is mainly studied in terms of the insulation performance, the arc-extinguishing performance, and the decomposition characteristics. The summary of the research is shown in Fig. 2.

Based on the environmental parameter (GWP), insulation properties, safety, and other aspects, the main alternative gases and basic parameters are shown in Table II. The research focuses mainly on natural gases (N<sub>2</sub>, CO<sub>2</sub>, He, Ar, and so on), SF<sub>6</sub> gas mixtures, perfluorocarbons (c-C<sub>4</sub>F<sub>8</sub>, C<sub>3</sub>F<sub>8</sub>, and C<sub>2</sub>F<sub>6</sub>), CnFmX gases, and other gases such as HFOs.

#### A. Insulation performance

The macroscopic parameters of the insulation performance mainly include the breakdown voltage, the partial discharge (PD) inception voltage, and the volt-second characteristic of different gases tested, which can be measured in the test. The voltage type (DC, AC, or lightning impulse voltage), electric field uniformity, gas pressure, and mixing ratio of the insulated gas are all important factors to judge the insulation properties, as shown in Fig. 3. The experimental method of breakdown voltage is shown in Fig. 4. The measurement of partial discharge (PD) inception voltage needs to be combined with the voltage measurement device while measuring the impedance to collect the PD signals presented in the oscilloscope.

Among the influencing factors, the electric field uniformity is determined by the electrode structure, and the non-uniformity factor f of the electric field is given by



FIG. 2. Evaluation indices of alternative gases and research resource.

$$f = \frac{E_{max}}{E_{av}}.$$
 (1)

field distribution around the electrodes can be calculated by software such as COMSOL. Different gases have different sensitivities to electric field uniformity.

In Eq. (1),  $E_{max}$  represents the maximum electric field strength and  $E_{av}$  represents the average electric field strength. The electric The influence of the gas mixture ratio on the insulation properties mainly refers to the synergistic effect of the mixed gases. When

TABLE II. Physical and environmental characteristics of common dielectric gases.<sup>45–50</sup>

Туре	Insulation strength relative to SF <sub>6</sub>	Boiling temperature (°C) (1 bar)	Lifetime (year)	GWP
SF <sub>6</sub>	1	-63.9	3 200	23 500
N <sub>2</sub>	0.36	—196	0	0
$CO_2$	0.30	-78.5	Infinity	1
Air	0.30	<183	Infinity	≈0
CF <sub>4</sub>	0.39	-186.8	6 300	50 000
$C_2F_6$	0.78-0.79	-78	10 000	9 200
$C_3F_8$	0.96-0.97	—37	2 600	7 000
c-C <sub>4</sub> F <sub>8</sub>	1.25, 1.31	-6(-8)	3 200	8 700
CF <sub>3</sub> I	1.23	-22.5	0.005	1-5
$C_4F_7N$	2.2	-4.7	0	2 1 0 0
$C_5F_{10}O$	2.0	27	0	1
$C_6F_{12}O$	2.5	49	0	1
CF <sub>3</sub> NSF <sub>2</sub>	2.41	—6		
HFO1234zeE	0.98	-9	12	0.02
R12	0.9	-29.8	12	2 400





two kinds of gases (at least one is a kind of gas with good insulation performance) are mixed, the insulation performance of the mixed gas shows three different variations with the increase in the insulating gas content: negative synergistic effect, linear relationship, and positive synergistic effect. Mixed gas with a negative synergistic effect is rare, which is not suitable as an insulating gas. The mixed gases with a positive synergistic effect will show great advantages such as the same insulating property as pure gas at a certain mixing ratio. Therefore, finding the characteristics of the synergistic effect is helpful to find the most suitable mixing ratio,

$$C = \frac{k(V_m - V_1)}{(1 - k)(V_m - V_2)},$$
(2)

where  $V_1$  and  $V_2$  are the breakdown voltage of the two pure gases 1 and 2 ( $V_1 > V_2$ ) in kV,  $V_m$  is the breakdown voltage of the mixed gas



**FIG. 4**. Basic synergy relationship of mixed gas: negative synergistic effect C > 1 (curves 1 and 2), linear relationship C = 1 (curve 3), and positive synergistic effect C < 1 (curves 4 and 5).

in kV, *k* is the mixing ratio of gas 1, and *C* represents the degree of synergy of two gases. A smaller value of *C* indicates that the two gases have the better synergistic effect.<sup>51</sup> The basic synergy relationship of mixed gas: negative synergistic effect is shown in Fig. 4.

The microscopic parameters for characterizing the gas insulation properties can also be measured experimentally or calculated theoretically. The experimental measurement process is shown in Fig. 5, which is generally performed under a steady-state Townsend (SST) test or a pulsed Townsend (PT) test (generally, the gas pressure is small at about 100 Pa).<sup>52</sup> The measured parameters include the effective ionization coefficient  $\alpha$  of the molecule and the effective adhesion coefficient  $\eta$ . It is generally considered that the critical electric field  $E_{cr}/N$  can be obtained when the two coefficients are equal, which can be used as a reference value for measuring gas breakdown. According to Townsend discharge theory,  $\alpha$  and  $\eta$ can be obtained by changing the electrode spacing *d* and measuring repeatedly current *I*, according to the following equation:

$$I = I_0 \left[ \frac{\alpha}{\alpha - \eta} e^{(\alpha - \eta) d} - \frac{\eta}{\alpha - \eta} \right].$$
(3)



In theory, the electron swarm parameters (such as effective ionization coefficient, electron drift velocity, and particle diffusion coefficient) between gaseous molecules and electrons at the gas microlevel are theoretically calculated to comprehensively evaluate the insulating properties of the gas. The first step to obtain these parameters is the calculation of the collision cross section. More information about collisions in the main gases can be found in Ref. 53. There are two main methods of calculation: (1) the Monte Carlo simulation, which is the method with many iterations and computational complexity, and (2) solving the Boltzmann equation using some approximations. Indeed, the transport state and energy distribution of electrons in the discharge process should be clear. When the electron density is unevenly distributed, the electron diffuses from a high concentration point to a low concentration point, which is caused by the thermal motion of electrons. At the same time, under the action of the electric field, it accelerates the movement along the electric field to cause the collision ionization to form an electronic collapse. This complex state of motion results in different distributions of electron energy in the discharge space. In order to determine the transport state of charged particles, a self-consistent model need to be established that reflects the interaction between the electric field and the particles. In this complex model, both the elastic and the inelastic collision between electrons and gas molecules should be properly considered and the effect of the external electric field on charged particles should be taken into consideration. Therefore, some simplifications and approximations are usually needed to obtain electron swarm parameters and transport parameters during gas discharge. In a weakly ionized gas, the velocity distribution function f(r, v, t) of electrons in a six-dimensional phase space obeys Boltzmann's equation,

$$\frac{\partial f}{\partial t} + v \cdot \nabla_r f + a \cdot \nabla_v f = C[f(r, v, t)], \tag{4}$$

where a = eE/m is the acceleration of the electron under the action of an electric field, in which *e* and *m* denote the charge and mass of the electron, respectively. *E* is the electric field strength; *v* is the electron velocity; and *r* is the electron position parameter. C[f(r, v, t)] refers to the collision process of electronic collision process, including the elastic collision and inelastic collision.

The general solution of Eq. (4) is obtained using the Legendre polynomial sphere expansion to construct a series of equations equivalent to Eq. (4) and to introduce the correlation coefficients. In the case of higher order expansion, it is usually necessary to introduce at least six parameters. However, for the second order, the equation can be greatly simplified,<sup>54,55</sup> but the corresponding assumptions need to be made.

- (1) The distribution of the electric field in space is uniform.
- (2) Collisions are stable over the mean free path dimension.
- (3) The velocity distribution of the electron velocity distribution function *f* in the direction of the electric field is distributed symmetrically.
- (4) In the time dimension, only the distribution of the electric field is considered stable and the electrons are in a state of stable distribution or high-frequency oscillation.

By assuming that the equation is simplified, the electron energy distribution function (EEDF) is obtained by approximately solving the Boltzmann equation, and the ionization coefficient and the attachment coefficient are obtained. When the ionization coefficient and the attachment coefficient are equal, the breakdown field strength is the critical breakdown field strength, (E/N)cr, and the parameters such as the electron drift speed and the average energy can also be obtained.<sup>56,57</sup>

In addition, some parameters are calculated based on quantum chemical calculation, such as the molecular ionization energy (IE), binding energy, surface area, and molecular polarization properties, and the gas insulation properties can also be predicted.<sup>58</sup>

#### B. Arc extinguishing performance

The main parameters for the experimental measurement are the gas arc time and the cut-off current value. Tests are generally carried out in the interrupter of the circuit breaker. According to Mayr's arc model,<sup>59</sup> the time constant and power loss coefficient of arc extinguishing can be obtained by changing the arc voltage (V) and arc current (I), based on the following formula:

$$\frac{1}{g}\frac{dg}{dt} = \frac{1}{\theta} \bigg\{ \frac{Vi}{N_0} - 1 \bigg\},\tag{5}$$

where v is the arc voltage, *i* is the arc current, *g* is the arc conductance,  $\theta$  is time constant, and  $N_0$  is the power loss coefficient.

To judge the arc performance of the gas and finally to evaluate the arc extinguishing performance, the thermodynamic parameters and transport characteristics of the plasma formed during the arc process have to be determined depending on its thermodynamic state. When the temperature is very high, the plasma has a uniform thermodynamic temperature, and it is in a thermodynamic equilibrium state. At this point, the particle velocity is in accordance with the Maxwell-Boltzmann distribution, the population density of the ground state and excited state is in accordance with Boltzmann distribution, and the ionization process can be established based on the Saha equation. However, the complete thermodynamic equilibrium plasma can hardly exist in both natural and laboratory environments. For arc plasma, it is usually assumed that it is in the local thermodynamic equilibrium. Under this state, the particle collisions replace the radiation and occupy the dominant position in various reactions. The electron temperature is approximately equal to the temperature of heavy particles. The population density approximates the Boltzmann distribution, and the Saha equation is also approximately qualified. However, with the decrease in the arc temperature, the electron density decreases and the electron collisions gradually decay. The electron temperature deviates from the temperature of the heavy particles, and the arc plasma is in a nonthermodynamic equilibrium state. In addition, in the arc plasma, the rate of chemical reactions, such as decomposition, ionization, recombination, and adsorption, is limited. When the relaxation time of chemical reactions is less than the characteristic time of physical movement, such as particle convection and diffusion, the plasma reaches the local chemical equilibrium; otherwise, it is in the nonchemical equilibrium state.<sup>60,61</sup> In the study of non-chemical equilibrium, the speed of chemical reactions is the standard to judge the degree of chemical non-equilibrium. Therefore, the rate constant of chemical reactions becomes the key parameter to study the chemical reaction process. At present, an effective means of obtaining the reaction rate constant is by means of quantum chemical calculation.  $^{62}$ 

Due to the non-chemical equilibrium, the original statistical physics method cannot accurately describe the particle distribution and internal processes of the plasma. Instead, a control equation containing both the chemical reaction process and the physical process needs to be established for each particle so that the modeling and the computational difficulty are greatly increased. Most of the calculations are based on the local thermodynamic equilibrium and the two-temperature model under chemical equilibrium.<sup>63</sup> The calculation modeling is shown in Fig. 6. Under equilibrium conditions, there are two theories for the calculation of equilibrium compositions: one is based on the Saha equation and the Guldberg-Waage equation, combined with the Dalton partial pressure law, the conservation of chemical metrology and the conservation of charge to obtain the equilibrium compositions, and the other is solved by the minimum Gibbs free energy. Both of these computational theories are mathematically equivalent with all particles in the gaseous phase. The Saha equation is no longer applicable if the non-gaseous phase (e.g., solid, liquid, and molten) particles are considered, and the minimum Gibbs free energy method is applied. This method simplifies the modeling process without considering specific ionization and decomposition reactions, and it is widely used in the solution of equilibrium particle components.64,65

#### C. Decomposition characteristics

Insulation defects in the device can cause decomposition of the gas and further reaction with traces of impurities in the device to produce more complex products. The detection of discharge or high temperature decomposition products and the theoretical calculation of the decomposition process of mixed gas have also become an important issue in studying alternative gases. The decomposition characteristics are considered for the following reasons: First, the decomposition process and decomposition mechanism directly affect the gas self-recovery performance. Second, the products of decomposition, especially the gas and solid products that may be generated by breakdown, partial discharge, and electric arcs under different conditions, may have an impact on the environment and the safety of the human body. Finally, the decomposing characteristics of the alternative gas also provide a test method for the maintenance and operation after it has been applied to the equipment. The exploration of the decomposition process is also generally carried out through a combination of experiments and theoretical calculations. In the recent years, the influence of metal particles (such as Cu, Fe, and Ag), a small amount of gas impurities (such as H<sub>2</sub>O and O<sub>2</sub>), and temperature on the insulation properties has been deeply researched and explored. Even SF<sub>6</sub> decomposition has become an important diagnostic method for detecting SF<sub>6</sub> gas-insulated equipment. Under different insulation defects, SF<sub>6</sub> decomposition products showed different rules. The use of chemical means (GC-MS, infrared, UV, and nano-sensors) to detect these decomposition gases has also become an important study to determine the internal fault of the device and improve equipment maintenance.66

For the experiment, the gas in the chamber after the breakdown or partial discharge is collected and detected by the testing equipment such as the GC-MS or infrared spectrometer, as shown in Fig. 9. The main components of the decomposed gas can be obtained qualitatively or quantitatively. Generally, the





FIG. 7. Measurement process of decomposition products.

possible products can be obtained through the qualitative analysis of the mass spectra or spectra, and then, the content of the products can be determined through the calibration of the standard gas. Through the XRD (x-ray diffraction) detection on the electrode surface, the atoms involved in the solid reaction can be clear during the gas decomposition process. The measurement process is shown in Fig. 7.<sup>79</sup>

In theoretical calculations, based on density functional theory (DFT), energy changes in chemical reactions, frequency

characteristics, and transition state search, the basic properties of decomposition products are calculated by means of chemical molecular calculation software (Gaussian or Material Studio). In addition, using molecular dynamics in a constant temperature and constant volume system, we can calculate the decomposition process of gases at different temperatures to obtain the concentration of various free radicals. The reaction of gas molecules with H<sub>2</sub>O or O<sub>2</sub> and the route of product generation are also important parts of the research.

Properties	Indices	Require values	Method
	Breakdown voltage	High	Measure
	Partial discharge inception voltage	High	Measure
	Synergistic effect	Low (less than 1)	Calculation based on the measured voltage
	Electron drift velocity	Low	Calculation
Insulation performance	Diffusion coefficient	Low	Calculation
-	Effective ionization coefficient	Low	Calculation or measure
	Ionization coefficient	Low	Calculation or measure
	Attachment coefficient	High	Calculation or measure
	Critical electric field E <sub>cr</sub> /N	High	Calculation
	Molecular ionization energy	High	Calculation or measure
	Interrupting current	High	Measure
	Time constant of arc	Low	Measure
	Specific heat	High	Calculation
Arc-extinguishing performance	Thermal conductivity	High	Calculation
	Electrical conductivity	Low	Calculation
	Net emission coefficient	Low	Calculation
	Conductance of the decaying arc	Low	Calculation
	Toxicity of decomposition products	Low	
Decomposition characteristics	Formation rate of products	Low	Measure
• 	GWP and ODP of products	Low and 0	

TABLE III. Evaluation indices of alternative gases to have good properties.

#### D. The main evaluation indices

The main evaluation indices of the insulation performance of gas-insulated media are shown in Table III, which may be helpful to evaluate the properties of new gases.

#### IV. THE RESEARCH RESULTS OF ALTERNATIVE GASES

#### A. Natural gases (N<sub>2</sub>, CO<sub>2</sub>, and air) and mixed gases

The natural gases with much lower liquefaction temperature and lower GWP than SF<sub>6</sub> have got relatively stable physical and chemical properties and are cheap. Therefore, it is better to use natural gases in gas-insulated equipment, especially in medium- and low-voltage equipment. Currently, natural gases used as insulating gases are N<sub>2</sub>, CO<sub>2</sub>, and air. Although the insulation performance is weaker than that of SF<sub>6</sub>, these gases are initially considered as gases for insulation due to the simple molecular structure, and theoretical studies also started earlier. Until the introduction of alternative gases, these gases were focused and studied in more detail, both theoretically and experimentally. The related equipment is designed to produce and put into use. However, due to the relatively low insulation performance, it is necessary to take reasonable measures to improve the insulation properties such as increasing the gas pressure of the gas chamber, expanding the size of the equipment, and adopting the gas-solid insulation method.

#### 1. Breakdown voltage

Husain and Nema<sup>80</sup> extended the Townsend breakdown formula to obtain Paschen curves for air, N<sub>2</sub>, and SF<sub>6</sub>, completing the evaluation of the three gas breakdown voltages. Hara *et al.*<sup>81</sup> studied the breakdown behavior of N<sub>2</sub> and N<sub>2</sub>/O<sub>2</sub> gas mixtures in the presence of metallic particles at the solid–insulator interface of solid insulators. The effects of the cone insulator angle, particle size, and pressure were analyzed. Mizuno *et al.*<sup>82</sup> proposed a combination of the gas and solid insulation method. By wrapping rubber on the surface of the electrode, the breakdown voltage of N<sub>2</sub> can be increased by 1.5 times at 0.2 MPa, and the results can be used in gas-insulated switch devices. Philp<sup>83</sup> experimentally measured the breakdown voltage of the N<sub>2</sub> + CO<sub>2</sub> mixture in a ball grid electrode, which is only one-third that of SF<sub>6</sub>.

Meijer *et al.*<sup>84</sup> compared the breakdown voltage of N<sub>2</sub> and CO<sub>2</sub> on the AC voltage of a plate electrode by experiments. Although the difference between the breakdown values of the two gases is not significant, CO<sub>2</sub> is more stable and the breakdown voltage of CO<sub>2</sub> linearly increases with the pressure value. In order to test the ability of the gas to withstand over-voltage in the field, Ueta et al.81 tested the breakdown characteristics of CO2 and N2 under nonstandard lightning impulse voltage and proposed a method to evaluate the performance of CO2 gap insulation of non-standard lightning impulse waves. Pfeiffer and Schoen<sup>88</sup> experimented and studied the development of the streamer current before the breakdown of N2 and N2O mixed gases. It is believed that the use of the mixed gas in large-scale high-pressure equipment has obvious advantages. Pelletier et al.<sup>89</sup> measured the breakdown field strength of N<sub>2</sub> and He mixed gases under a uniform field and obtained the standard formula of the breakdown voltage and mixing ratio, Olivier's equation. Lim and Bae<sup>90</sup> studied the flashover voltage and the partial discharge inception voltage of dry air, N2, O2, and mixed gas on

the surface of the GIS insulator. Considering economy and insulation performance, it shows that dry air is suitable for gas insulated switchgear.

Rokunohe et al.<sup>91</sup> found that the insulation property of air at 0.5 MPa is greater than that of pure N<sub>2</sub> and CO<sub>2</sub>, and is comparable to that of  $N_2/O_2$  (the mixing radio of  $O_2$  is 20%). They pointed out that the dielectric strength of air at 0.6 MPa is 95% of  $SF_6/N_2$  (the mixing radio of SF<sub>6</sub> is 5%). Pontiga found that N<sub>2</sub> and O<sub>2</sub> mixed gases produced N<sub>2</sub>O under negative corona discharge.<sup>92</sup> Saitoh et al.93 studied the partial discharge and breakdown characteristics of air and N<sub>2</sub> at the plate electrode. The partial discharge voltages of the two gases are almost the same, but the breakdown voltage of air is larger than pure N<sub>2</sub>. Without changing the size of the equipment, using 1.0 MPa N<sub>2</sub> and solid insulation can replace 0.5 MPa SF<sub>6</sub>.<sup>94</sup> Zhang et al.<sup>95</sup> proposed an improved design method of air gap and interface insulation. Based on the small or no SF<sub>6</sub> experiment in medium-pressure C-GIS, a low-pressure N2 gas tank was used as the insulation gas to replace SF<sub>6</sub>. A low-pressure N<sub>2</sub> gas tank 40.5 kV-1250 A three-phase C-GIS was developed. It has a vacuum interrupter and the same area covered as the SF<sub>6</sub> C-GIS.

#### 2. Electron swarm parameters

Zhao et al.<sup>96,97</sup> calculated the critical breakdown field (E/N)cr of CO2 mixed O2 and H2. Compared to pure CO2 and 50% CO2-50% H<sub>2</sub>, the 50% CO<sub>2</sub>-50% O<sub>2</sub> mixture has better breakdown characteristics. The (E/N)<sub>cr</sub> of the 50% CO<sub>2</sub>-50% O<sub>2</sub> mixed gas almost does not change when the gas temperature is up to 2500 K. Although there is a significant decrease with the temperature rising to 3500 K, it is still higher than that of pure CO<sub>2</sub>. Chen et al.<sup>98</sup> calculated the critical breakdown strength of CO<sub>2</sub> and mixed gases (CO<sub>2</sub>, CO<sub>2</sub>/O<sub>2</sub>, CO<sub>2</sub>/CH<sub>4</sub>, CO<sub>2</sub>/He, and CO<sub>2</sub>/N<sub>2</sub>) over a temperature range of 300 K-4000 K. The results show that the mixtures  $CO_2/O_2$  and CO<sub>2</sub>/CH<sub>4</sub> have a high critical breakdown field, which makes it possible to further improve the insulation properties of pure CO<sub>2</sub>. Yousfi et al.<sup>99</sup> measured the electron swarm coefficients (drift velocity, diffusion coefficient, and effective ionization coefficient) of N<sub>2</sub>, CO<sub>2</sub>, and O2, and mixed gases using a pulse Townsend discharge. The hindering effect of the two gases on the electrons was verified by solving the Boltzmann equation. Moruzzi and Price<sup>100</sup> measured the ionization coefficient of air in the range of 90-260Td. With the addition of a small amount of CO<sub>2</sub>, the adsorption of air was significantly enhanced, but the electron attachment process of N<sub>2</sub> could not be detected. Raju and Gurumurthy<sup>101</sup> calculated the parameters of N<sub>2</sub> by solving the Boltzmann equation numerically. The electron average energy, drift velocity, and mean effective field intensity  $(E_{avef})$ are obtained from the distribution function, and the field strength of the electron population is described when the electric and magnetic fields coexist. Kucukarpaci and Lucas<sup>102</sup> calculated the electron swarm parameters of CO2 and N2 over a large electric field (14-3000 Td) using the Monte Carlo algorithm using the latest collision cross section parameters, and the calculated results were consistent with the experimental measurements.

#### 3. Interruption property

Stoller<sup>103</sup> tested the interrupting current of CO<sub>2</sub> as an interrupter medium in circuit breakers and calculated it by using a hydrodynamic model and found that the thermal disruption performance of CO<sub>2</sub> is lower than that of SF<sub>6</sub> but higher than air. Optimizing the circuit breaker structure may result in the CO<sub>2</sub> substitution of SF<sub>6</sub> as an arc extinguishing medium. Jonsson *et al.*<sup>104–107</sup> tested the ability of air to break in a 630 A/24 KV medium voltage load short-circuit switch. Optimization of the contact and nozzle structure helps improve the ability to interrupt, while the length and diameter of the nozzle have little effect on the ability to interrupt. Uchii *et al.*<sup>108</sup> tested the ability of CO<sub>2</sub> to break off in a GCB (gas circuit breaker). At the same gas pressure, the thermal shutdown capacity of CO<sub>2</sub> is about 50% of SF<sub>6</sub>. By blowing more heat through the improved mixing chamber, the cut-off capacity of CO<sub>2</sub> can be increased by 30%.

Chen *et al.*<sup>109,110</sup> established the arc model to simulate the interruption ability of the vacuum-CO2 mixed circuit, and the results show that with the combination of vacuum interrupters, the current interruption capacity of CO2 gas can be increased. Matsumura et al.<sup>111</sup> studied the current interruption ability of CO<sub>2</sub> mixed with N<sub>2</sub>, O<sub>2</sub>, He, and air. It was found that the conductance of the residual arc of the CO<sub>2</sub> mixture of O<sub>2</sub> or He (the mixing ratio of CO<sub>2</sub> is 20%) decreased more rapidly and the breaking capacity improved significantly. In theoretical calculation, Zhong et al.<sup>112</sup> studied the thermal physical properties of plasma and the effect of the copper vapor from the electrode surface of the circuit breaker on the CO2-N<sub>2</sub> mixture. The behavior of gas during the quenching process was analyzed based on the thermodynamic parameters and transport characteristics. The existence of copper vapor in the equipment was also analyzed. The influence of copper vapor on the thermodynamic properties of the mixed gas was evidently influenced by the ionization properties of the copper atom itself, atomic density, as well as partition function.

The advantages of natural gas are low cost, harmless to the environment, and no security threat to ecology and human resources. At present, there are already finished products in middle and low voltage equipment (ABB, Schneider, and other companies) using conventional gas as an insulation medium. The main properties are shown in Table V of the Appendix. CO<sub>2</sub> is likely to be used as an arc extinguishing medium in circuit breakers. The use of conventional gas in the switch equipment needs to optimize the structure of the equipment reasonably, and the mechanical structure and electrical performance have greater space to improve. Simply relying on increasing the pressure and increasing the size will reduce the safety of the equipment and increase the production cost of the equipment. It is also an effective means to use the solid insulation material. However, solid insulation materials exist in the atmosphere for a long time and are difficult to degrade. Adding solid insulation materials to the equipment will undoubtedly increase the environmental burden on the other hand. However, this kind of gas still has a great prospect of development and use, which has a great exploration value.

#### B. Mixtures of SF<sub>6</sub> with natural gases

Although the study of the SF<sub>6</sub> gas mixture started earlier, there is no interruption in the exploration of insulation and arc extinguishing ability. At this stage, research focuses more on practicality. The SF<sub>6</sub> gas mixture, which generally refers to the mixture of SF<sub>6</sub> and some buffer gas with relatively poor insulation and a very low boiling point (air, N<sub>2</sub>, CO<sub>2</sub>, N<sub>2</sub>O, CF<sub>4</sub>, H<sub>2</sub>, and some inert gases), not only meets the equipment insulation requirements but also reduces the amount of  $SF_6$ .

#### 1. Breakdown voltage

Malik and Qureshi<sup>113,114</sup> compared the breakdown voltage of SF<sub>6</sub> mixed with N<sub>2</sub>, CO<sub>2</sub>, and air. It was considered that SF<sub>6</sub> mixed N2 was the most ideal mixed gas. The mixture is non-toxic and non-flammable, and when the  $SF_6$  mixing ratio reaches 50%–60%, the insulation performance can reach 85%-90% of the pure SF<sub>6</sub>. When the lightning impulse voltage was applied, the addition of a small amount of SF<sub>6</sub> to N<sub>2</sub> increased the positive lightning impulse breakdown voltage of N<sub>2</sub>. This is due to the positive space charge resulting in an anode shield.<sup>115</sup> Farish et al.<sup>116</sup> investigated the effect of the surface roughness of the SF<sub>6</sub> and N<sub>2</sub> mixed gas on the breakdown voltage with a coaxial electrode. Compared with pure SF<sub>6</sub>, the mixed gas is not sensitive to the electric field roughness. Graf and Boeck<sup>117</sup> studied the sensitivity of the SF<sub>6</sub>/N<sub>2</sub> mixed gas to nonuniform electric fields by setting protrusions on the electrodes. The mixed gas is more sensitive to non-uniform fields than SF<sub>6</sub> at the same dielectric strength. Woo et al.<sup>118</sup> measured the breakdown performance of the SF<sub>6</sub>/N<sub>2</sub> mixture at the lightning impulse voltage and AC voltage. As the electric field uniformity increased, the slope of the breakdown voltage decreased. In the lightning impulse test, the positive breakdown voltage was higher than the negative breakdown voltage. Guerroui and Lemzadmi<sup>119</sup> by experiment measured the initial partial discharge voltage of the SF<sub>6</sub> and N<sub>2</sub> mixed gas under a non-uniform field. The initial voltage increases with an increase in pressure, and the positive polarity was higher than the negative polarity. Sato et al.<sup>120</sup> designed a gas insulated bus with  $SF_6/N_2$  as gas-insulation. Compared with 0.4 MPa pure  $SF_6$  gas insulated busbars, if a 0.6 MPa 10%SF<sub>6</sub>-90% N<sub>2</sub> mixed gas is used, the diameter of the tank needed to be increased by 15%. Hoshina et al.<sup>121</sup> tested the insulation properties of the  $SF_6/N_2$  mixture on a full-scale gas-insulated busbar model and analyzed the effect of contaminated particles on the breakdown characteristics of AC voltage and impulse voltage. The mixed gas at 0.3 MPa and 0.5 MPa showed a positive synergistic effect, and the partial discharge inception voltage of the mixed gas showed a negative synergistic effect. Guo et al.<sup>122</sup> studied the lightning impulse breakdown voltage of the SF<sub>6</sub>/N<sub>2</sub> mixture, which showed a negative synergistic effect at low gas pressure. Rizk and Eteiba<sup>123</sup> measured the breakdown voltage and time of a mixture of SF<sub>6</sub> and N<sub>2</sub> under the coaxial electrode and validated it theoretically. Mizobuchi et al.<sup>124</sup> observed the streamer development of SF<sub>6</sub>-N<sub>2</sub> mixed gas discharge by experimental tests and verified by numerical calculation. The electric field was kept constant during the development of the streamer by single steep square high voltages in the measurements. At different mixing ratios, the development trend of the flow-injection process was similar. Jiasen et al.<sup>125</sup> tested multi-channel discharge characteristics of SF<sub>6</sub>-N<sub>2</sub> or SF<sub>6</sub>-Ar gas with different mixing ratios in a coaxial distorted electric field using 40 kV-78 kV pulse power. The results showed that the discharge channels of the SF<sub>6</sub>-Ar or SF<sub>6</sub>-N<sub>2</sub> mixture decreased with the increase in the SF<sub>6</sub> mixing ratio and then tended to be saturated. Hirata et al.<sup>126</sup> tested the creeping discharge characteristics of the mixed gas. The flashover voltage of 3% SF<sub>6</sub> mixture was lowest with a positive polarity impulse voltage. The mixtures of SF<sub>6</sub> and N<sub>2</sub> gases have been used in GIL. The first GIL that used the SF<sub>6</sub>/N<sub>2</sub> gas mixture as the insulation medium

developed by Siemens was put into operation at Geneva International Airport in Switzerland in 2001.<sup>127</sup> Currently, the SF<sub>6</sub>/N<sub>2</sub> gas mixture GIL has been successfully applied in the 245 kV–550 kV line.

The SF<sub>6</sub>-air mixture has a more stable corona for positive DC voltages.<sup>128</sup> Li *et al.*<sup>129</sup> measured the breakdown characteristics of the gap between coaxial electrodes of various impulse power sources for SF<sub>6</sub> (1%) with air and N<sub>2</sub> gas mixtures in the range of 100 kPa–500 kPa. The results showed that the influence of the waveform on the breakdown voltage was great. Under the coaxial electrode, little change was caused in insulation properties with adding 1% of the SF<sub>6</sub> in the air, which is different from the situation under a non-uniform field.

Qiu et al.<sup>130-132</sup> compared the dielectric properties of SF<sub>6</sub>/N<sub>2</sub> and SF<sub>6</sub>/CO<sub>2</sub> mixtures and concluded that the dielectric properties of the SF<sub>6</sub>/N<sub>2</sub> mixed gas are better than that of SF<sub>6</sub>/CO<sub>2</sub> in uniform or quasi-uniform fields, but SF<sub>6</sub>/CO<sub>2</sub> on the contrary is better than SF<sub>6</sub>/N<sub>2</sub> under the non-uniform fields. By testing the breakdown voltage of 50% SF<sub>6</sub> mixed CO<sub>2</sub> and N<sub>2</sub> at 60 Hz alternating current, it was found that adding a small amount of CO<sub>2</sub> into the 50% SF<sub>6</sub>/N<sub>2</sub> mixture can significantly improve the breakdown voltage. The breakdown voltage of 50% SF<sub>6</sub>-49% N<sub>2</sub>-1% CO<sub>2</sub> is 1.35 times that of 50% SF<sub>6</sub>-50% N<sub>2</sub> and 1.15 times that of pure SF<sub>6</sub>.<sup>133</sup> By testing the breakdown behavior of mixed gases under the rodplate electrode, it was found that the breakdown voltages of SF<sub>6</sub>- $N_2$  and SF<sub>6</sub>-air were similar under the negative impulse voltage.<sup>12</sup> The SF<sub>6</sub>-CO<sub>2</sub> breakdown voltage is higher than the other two. As the gas pressure is higher than 0.1 MPa, the three kinds of mixed gases produced a leader before the breakdown. With the increase in the SF<sub>6</sub> mixing ratio and pressure, the leader showed a gradual development.

Cookson and Wootton<sup>135</sup> tested the corona and breakdown voltage at AC voltage with a mixture of SF<sub>6</sub> and H<sub>2</sub>. The addition of a very small amount of SF<sub>6</sub> (0.002%) to H<sub>2</sub> increases the breakdown voltage of pure H<sub>2</sub> by 70%. The starting voltage of pure SF<sub>6</sub> is about 2.7–3 times that of pure H<sub>2</sub>. Adding a small amount of SF<sub>6</sub> (within 1%) to H<sub>2</sub> does not affect the corona onset voltage. Farish *et al.*<sup>136</sup> tested the breakdown voltage of the SF<sub>6</sub> and H<sub>2</sub> mixed gas at negative polarity lightning impulse voltage. For H<sub>2</sub>, the breakdown voltage of the negative lightning impulse is 60%–100% higher than that of the positive polarity. However, adding a small amount of SF<sub>6</sub> (within 1%) to H<sub>2</sub> increases the rate of increase in breakdown voltage less than the positive polarity.

Akbar and Malik<sup>137</sup> measured the breakdown voltage of several mixed gases at DC voltage. The measurement results showed that the mixed gas containing N<sub>2</sub>O was less sensitive to electrode surface defects. Park *et al.*<sup>138,139</sup> tested SF<sub>6</sub>/CF<sub>4</sub> mixed gas insulation performance, including the lightning impulse test, AC frequency test, and partial discharge test in a 25.8 kV three-phase GIS tank. The experimental results showed that the breakdown voltage increased with the SF<sub>6</sub> content, which showed that the mixing ratio of SF<sub>6</sub> had little effect on the starting voltage. Berg *et al.*<sup>140–142</sup> studied the insulation properties of SF<sub>6</sub> and CF<sub>4</sub> gas mixtures at DC voltage, 60 Hz AC voltage, and standard lightning impulse voltage. The SF<sub>6</sub> and CF<sub>4</sub> mixed gas was more suitable than the SF<sub>6</sub>–N<sub>2</sub> mixed gas to be used as an arc extinguishing medium. Lee *et al.*<sup>143</sup> compared the breakdown characteristics of N<sub>2</sub>, SF<sub>6</sub>, and CF<sub>4</sub> in high-voltage bushing at low temperatures. Due to its outstanding insulating properties

and low boiling point,  $\mathrm{CF}_4$  has shown its obvious superiority in cold environments.

Siddagangappa *et al.*<sup>144</sup> proposed a simple calculation formula to predict the breakdown voltage of SF<sub>6</sub> and mixed gases under a uniform electric field. Nema *et al.*<sup>145</sup> proposed a breakdown voltage calculation formula of the SF<sub>6</sub> mixed gas under a uniform and non-uniform electric field, which was used to infer the insulating properties of the mixed gas. The calculation results were consistent with the experimental test results. The limitation of the formula is that the parameters in different gas formulas are different and not universal.

#### 2. Electron swarm coefficients

Maller and Naidu<sup>146</sup> measured the ionization and attachment coefficients of the SF<sub>6</sub> mixed air and nitrogen. In the meantime, using the calculation method, the breakdown field strength of the three mixed gases of SF<sub>6</sub>-N<sub>2</sub>, SF<sub>6</sub>-air, and SF<sub>6</sub>-CO<sub>2</sub> was calculated in the pressure range from 100 kPa to 500 kPa, and the insulation properties were predicted.<sup>147</sup> Govinda Raju calculated the breakdown field strengths of SF<sub>6</sub>/N<sub>2</sub> and SF<sub>6</sub>/N<sub>2</sub>O mixed gases by solving Boltzmann parameters.<sup>148</sup> Kline *et al.*<sup>149</sup> calculated the electron swarm parameters of SF<sub>6</sub> mixed with He and N<sub>2</sub>. The critical breakdown field strength of the mixed gas was obtained from the ionization coefficient and the attachment coefficient. Dincer and Govinda Raju<sup>150</sup> measured the ionization and attachment coefficients of SF<sub>6</sub> and N<sub>2</sub> mixed gases at steady state Townsend discharge. Govinda Raju et al.<sup>151</sup> used the steady-state Townsend apparatus to measure the ionization of the SF<sub>6</sub> and N<sub>2</sub> mixed gas and obtained the critical breakdown field strength of the mixed gas. The breakdown field strength does not show a simple linear relationship with the increase in the SF<sub>6</sub> content, and a synergistic effect existed. Lee<sup>152</sup> measured the relative parameters of a mixture of  $SF_6$  and  $CO_2$ . Fréchette<sup>153</sup> used the SST apparatus to measure the ionization coefficients of the SF<sub>6</sub>/N<sub>2</sub> and SF<sub>6</sub>/CC<sub>12</sub>F<sub>2</sub> mixed gas to obtain the limited breakdown field strength. For the SF<sub>6</sub>/CCl<sub>2</sub>F<sub>2</sub> mixed gas, the ionization coefficient was not linear with the SF<sub>6</sub> mixing ratio, and there was a synergistic effect. The breakdown strength of the mixed gas with a SF<sub>6</sub> mixing ratio of 25% was 5% higher than that of pure CCl<sub>2</sub>F<sub>2</sub>. Using the latest collision cross section data, Phelps and Van Brunt<sup>154</sup> calculated the electron swarm parameters (transport, ionization, adhesion, and diffusion coefficients) of SF<sub>6</sub> and N<sub>2</sub>, O<sub>2</sub>, and He mixtures by solving two Boltzmann equations approximately. The electron swarm process of the SF<sub>6</sub> and CO<sub>2</sub> mixture under PT discharge was calculated, as well as the ionization, adsorption, and dissociation coefficients.<sup>155</sup> Adding a small amount (2%–5%) of SF<sub>6</sub> into CO2 under the condition of pulsed Townsend discharge has no obvious effect on the electron drift speed and diffusion coefficient function. However, it had a significant impact on the effective ionization process. Under the same mixing ratio, the critical breakdown strength of SF<sub>6</sub>-CO<sub>2</sub> was lower than that of SF<sub>6</sub>-N<sub>2</sub>.<sup>156</sup> Urquijo measured the electron drift, diffusion, and effective ionization processes and attachment coefficients for SF<sub>6</sub>/CHF<sub>3</sub> and SF<sub>6</sub>-CF<sub>4</sub> mixed gases under PT discharge conditions. The electron drift and diffusion process had no relation with the SF<sub>6</sub> content, but the ionization coefficient depends on the SF<sub>6</sub> content. The critical breakdown fields of both gases were lower than SF<sub>6</sub>-N<sub>2</sub>.<sup>157</sup> Xiao et al.<sup>158</sup> and Liu and Xiao<sup>159</sup> calculated the electron swarm parameters of SF<sub>6</sub> and CF<sub>4</sub> using the Monte Carlo method. With the increase in the SF<sub>6</sub> content,

the breakdown voltage increased the effective ionization coefficient decreased. The electron diffusion coefficient of CF4 was larger than SF<sub>6</sub>. The improved Monte Carlo algorithm was used to calculate the electron swarm parameters of the SF<sub>6</sub>/N<sub>2</sub> mixed gas, and the calculation was further improved.<sup>160</sup> Zhao et al.<sup>161</sup> calculated the critical breakdown field strength of mixed gas of 0.01-1.6 MPa SF<sub>6</sub> and N<sub>2</sub> with the temperature of 300-3000 K. The minimum Gibbs free energy was used to calculate the equilibrium composition of particles. The electron energy distribution function was obtained using various particle data analyzed by the Boltzmann equation. At higher gas temperatures, the addition of N<sub>2</sub> to SF<sub>6</sub> gas reduced the kinetic energy of the electrons and increased the breakdown field strength. Increasing the N<sub>2</sub> concentration effectively increased the (E/N)<sub>cr</sub> of SF<sub>6</sub>-N<sub>2</sub> mixtures at gas temperatures around 2000-3000 K. The same method was used to calculate the SF<sub>6</sub>-CF<sub>4</sub> gas mixture. At low temperatures, the breakdown field of pure SF<sub>6</sub> was higher than that of the mixed gas, but the breakdown strength of the mixed gas was higher than that of SF<sub>6</sub> at temperatures greater than 4000 K (0.4 MPa).<sup>162</sup> For the SF<sub>6</sub>-CO<sub>2</sub> mixed gas, a large amount of CO<sub>2</sub> could reduce the number of high-energy electrons. When the temperature was higher than 1740 K, the addition of CO<sub>2</sub> to SF<sub>6</sub> could improve the insulation performance. However, adding too much CO<sub>2</sub> at low temperatures may cause the insulation performance to decrease. At 0.8 MPa, the dielectric properties of the mixed gas increased with an increase in pressure.<sup>163</sup> Larin discussed the synergistic effect of SF<sub>6</sub>/CF<sub>4</sub> gas mixtures based on solving the Boltzmann equation.164

#### 3. The interruption performance

Lee and Frost<sup>165</sup> studied the interruption ability of SF<sub>6</sub> and its mixed gas and evaluated the arc extinguishing performance of 15 gases and mixed gas at 60 Hz AC. It was showed that the arc extinguishing performance of these gases and mixed gases was lower than that of pure SF<sub>6</sub>. Tomita et al.<sup>166</sup> studied the interruption capability of 20% SF<sub>6</sub> and 80% Ar mixed gas in circuit breakers. The development of arc quenching was analyzed, and the electron density was measured. The arc extinguishing speed of mixed gas was faster than that of pure Ar. Gleizes et al.<sup>167</sup> calculated the transport parameters of the SF<sub>6</sub> and N<sub>2</sub> mixed gas plasma at temperatures from 1000 K to 15 000 K. With the temperature below 8000 K, the conductivity of the 50% SF<sub>6</sub>-N<sub>2</sub> mixed gas was similar to SF<sub>6</sub>, and much larger than pure N<sub>2</sub>, which was due to the lower ionization coefficient of the S atom. At temperatures greater than 4000 K, the thermal conductivity of the mixed gas was greater than that of pure SF<sub>6</sub>. In order to evaluate the arc-extinguishing ability of SF<sub>6</sub>-CF<sub>4</sub> and SF<sub>6</sub>-C<sub>2</sub>F<sub>6</sub> mixed gas, Chervy et al.<sup>168,169</sup> calculated the main parameters of the plasma based on the local thermodynamic equilibrium, which mainly referred to the thermodynamic parameters and transport parameters. The arc-extinguishing performance of SF<sub>6</sub>-CF<sub>4</sub> and SF<sub>6</sub>-C<sub>2</sub>F<sub>6</sub> mixed gases was better than that of SF<sub>6</sub>-N<sub>2</sub>, and SF<sub>6</sub>-CF<sub>4</sub> was superior to SF<sub>6</sub>-C<sub>2</sub>F<sub>6</sub>. The thermodynamic properties of CF4 with a better ability to interrupt were more stable than those of C<sub>2</sub>F<sub>6</sub>. When the mixing ratio of SF<sub>6</sub> exceeded 50%, the interruption performance of the mixed gas approached SF<sub>6</sub>. With the mixing ratio of SF<sub>6</sub> below 25%, the performance is close to CF<sub>4</sub>. 40% SF<sub>6</sub> mixed with CF<sub>4</sub> is a more appropriate mixing ratio. The arc extinguishing performance of the SF<sub>6</sub>-CO<sub>2</sub> mixture used in circuit breakers was evaluated by calculating the mixed gas

transport parameters (thermal conductivity and conductivity).<sup>170</sup> Wang et al.<sup>171</sup> calculated the effective ionization coefficient and the critical breakdown field strength of the SF<sub>6</sub> and He mixed gas at a high temperature (300 K-3500 K) in a high voltage circuit breaker. The mixed gas showed better interruption ability because of the He with a higher ratio thermal and thermal conductivity. CF<sub>4</sub> also had a good arc-extinguishing performance to be used in equipment such as a 115 kV/40 kA high voltage circuit breaker with 50%SF<sub>6</sub>/50%CF<sub>4</sub> at 0.7 MPa gas pressure applied in the Manitoba hydropower station of Canada. SF<sub>6</sub>/CF<sub>4</sub> circuit breakers with a rated voltage of 550 kV, a nominal current of 4 kA, and a breaking capacity of 40 kA operated at the Dorsey Converter Station.<sup>172</sup> In the plasma calculation, Yang *et al.*<sup>173,174</sup> calculated the thermodynamic properties and transport parameters of the plasma of SF<sub>6</sub> mixed with N<sub>2</sub> and CO<sub>2</sub> at high temperatures, as well as the influence of metal particles. In the non-thermodynamic equilibrium, the plasma behavior has been analyzed and discussed. Under the non-thermodynamic equilibrium condition, the difference of the conductivity between the mixed gas and N2 was larger. The thermal conductivity of mixed gases also appeared significantly different, especially below 2000 K.

#### 4. Decomposition characteristics

Pradayrol et al.<sup>175</sup> examined the major decomposition products of 50%SF<sub>6</sub> mixed 50%CF<sub>4</sub> at 50 Hz AC, including SOF<sub>2</sub> + SF<sub>4</sub>, SOF<sub>4</sub>, SO<sub>2</sub>F<sub>2</sub>, S<sub>2</sub>F<sub>10</sub>, S<sub>2</sub>OF<sub>10</sub>, S<sub>2</sub>O<sub>2</sub>F<sub>10</sub>, and S<sub>2</sub>O<sub>3</sub>F<sub>6</sub>. The addition of CF4 to SF6 had little effect on the major decomposition products. When H<sub>2</sub>O and O<sub>2</sub> were present, the formation of the major products  $(SF_4 + SOF_2)$  and  $S_2F_{10}$  was reduced. In the presence of solid insulation, the products of the mixed gases were similar to pure SF<sub>6</sub>. When H<sub>2</sub>O, O<sub>2</sub>, and insulators exist, they can interfere with each other.<sup>176</sup> Under negative corona discharge,  $O_2$  and  $H_2O$ (less than 0.2% H<sub>2</sub>O and less than 1% O<sub>2</sub>) enhanced the production of all sulfur oxyfluoride. At lower pressures, product generation increased. The presence of 50%CF4 inhibited the production of all compounds.<sup>177</sup> SF<sub>6</sub>/O<sub>2</sub> under spark discharge decomposed SOF<sub>4</sub>, SO<sub>2</sub>F<sub>2</sub>, SOF<sub>2</sub>, and SO<sub>2</sub>.<sup>178</sup> The main products of SF<sub>6</sub> and N<sub>2</sub> included SOF<sub>4</sub>, SO<sub>2</sub>F<sub>2</sub>, SF<sub>4</sub> + SOF<sub>2</sub>, SO<sub>2</sub>, S<sub>2</sub>OF<sub>10</sub>, S<sub>2</sub>F<sub>10</sub>, S<sub>2</sub>O<sub>2</sub>F<sub>10</sub>, S<sub>2</sub>O<sub>3</sub>F<sub>6</sub>, SF<sub>5</sub>NF<sub>2</sub>, NF<sub>3</sub>, and (SF<sub>5</sub>)<sub>2</sub>NF during AC corona discharge. Adding a small amount of water molecules would affect the rate of formation of all products. The production of SF<sub>5</sub>NF<sub>2</sub>, NF<sub>3</sub>, and (SF<sub>5</sub>)<sub>2</sub>NF was minimal compared to other products. N<sub>2</sub>O and CO<sub>2</sub> were also produced at higher pressures.<sup>179,180</sup> SF<sub>6</sub> and N<sub>2</sub> produce the same decomposition when AC spark discharges.<sup>181</sup> When a small amount of CO2 was added to the mixture of SF6 and N2, the generation of SOF<sub>4</sub> + SO<sub>2</sub>F<sub>2</sub>, COF<sub>2</sub>, CO, and S<sub>2</sub>F<sub>10</sub> was reduced under negative DC corona.<sup>182</sup> Since SF<sub>6</sub> and N<sub>2</sub> have been used in the GIL, their decomposition products in the case of partial discharges and their discharge characteristics were helpful for the detection of partial discharges and insulation monitoring. However, the decomposition products of mixed gases are more complicated than pure SF<sub>6</sub>, and the difficulty in insulation monitoring is further increased.

Mixed gases are suitable for equipment in extremely cold areas, such as northern China. However, the recycling and reuse of mixed gases still need to be improved and optimized for existing SF<sub>6</sub> equipment.<sup>183,184</sup> ABB proposed a standard for the use of SF<sub>6</sub> gas mixtures in equipment, proposing that the assessment of the environmental



impact needs to take into account the time period. The mixed gas should be reused in the field without generating other gas components and be easy to recycle separation. The existing  $SF_6$  recovery equipment needs to be rebuilt and upgraded to recover the mixed gas.<sup>185</sup>

The use of  $SF_6$  gas mixtures in electrical equipment has the obvious advantage of lowering the boiling point and maintaining good insulation properties. The main properties of  $SF_6$  mixtures are shown in Table VI of the Appendix.

# C. Mixtures of perfluorocarbons (PFCs) with natural gases

In order to completely replace SF<sub>6</sub>, some electronegative gases with a low greenhouse effect and reliable insulation have become the main research objects. Compared with SF<sub>6</sub>, the PFCs have better insulating properties and the GWP value is lower relative to SF<sub>6</sub>. The typical electronegative gases studied today are  $c-C_4F_8$ ,  $C_3F_8$ ,  $C_2F_6$ , and CF<sub>4</sub> based on the boiling point, safety, and insulation properties. The molecular structures are shown in Fig. 8. The insulating gas and the buffer gas are generally mixed (N<sub>2</sub>, CO<sub>2</sub>, etc.), which reduces the gas cost and lowers the boiling point of the mixed gas under the premise of ensuring the dielectric strength.

Takuma et al.<sup>186</sup> calculated the boiling point of the mixed gas as a function of the mixing ratio according to the van der Waals formula and tested the breakdown voltage of the mixed gas of c-C<sub>4</sub>F<sub>8</sub> and N<sub>2</sub>. The insulation performance of the c-C<sub>4</sub>F<sub>8</sub> gas under a uniform electric field is 1.18–1.25 times that of the SF<sub>6</sub> gas. Although the price of  $c-C_4F_8$  is ten times that of SF<sub>6</sub>, the actual usage is much smaller than that of SF<sub>6</sub>. The use of c-C<sub>4</sub>F<sub>8</sub> as an arc-extinguishing medium may produce carbon particles that affect the insulation properties. Adding a buffer gas such as CO<sub>2</sub> and  $N_2$  can ease the formation of particles. Wada *et al.*<sup>187</sup> tested the breakdown behavior of the four perfluorocarbon gases, c-C<sub>4</sub>F<sub>8</sub>, l-C<sub>3</sub>F<sub>6</sub>, C<sub>3</sub>F<sub>8</sub>, and C<sub>2</sub>F<sub>6</sub>, under AC voltage at the plate-plate electrode and combined them with CO<sub>2</sub>, N<sub>2</sub>, and CF<sub>4</sub>, respectively. It was found that PFCs and buffer gas mixtures have shown a positive synergistic effect. At the same time, the gas mixture was analyzed in terms of the boiling point and GWP. When the ambient temperature is above -20 °C, the insulation performance of the

mixed gas can reach 70% that of  $SF_6$  and the GWP is reduced by 30%.

Zhao *et al.*<sup>188</sup> studied the synergistic effect of the c-C<sub>4</sub>F<sub>8</sub>/N<sub>2</sub> mixed gas at lightning impulse voltage. The effect of gas pressure on the synergistic effect is not significant, and the obvious polar effect appears in the discharge characteristic of c-C<sub>4</sub>F<sub>8</sub>/N<sub>2</sub>. The breakdown voltage under negative impulse was lower than that under positive impulse. This was because the polarity of the lightning impulses affects the speed and kinetic energy of free electrons. The free electrons in a negative lightning strike had a higher velocity, making the c-C<sub>4</sub>F<sub>8</sub> to absorb fewer free electrons, so the breakdown voltage under negative lightning impulses was lower than the breakdown voltage under negative lightning impulses.

Whitman<sup>189</sup> studied the impulse breakdown characteristics of  $C_3F_8$  and compared it to air. At both positive and negative impulse voltages, the breakdown voltage of  $C_3F_8$  was about 2.5 times that of air with both electrodes (ball and plate electrodes). Okubo *et al.*<sup>190</sup> tested the partial discharge inception voltage and breakdown voltages of  $C_3F_8$  and  $C_2F_6$  with the needle plate electrode. The synergistic effect of the  $C_3F_8/N_2$  and  $C_2F_6/N_2$  mixed gases was lower than that of  $SF_6/N_2$ . The breakdown voltage of the mixed gas is arranged in descending order of  $SF_6/N_2$ ,  $C_3F_8/N_2$ , and  $C_2F_6/N_2$ .

de Urquijo and Basurto<sup>191</sup> used the PT test to measure the C-C<sub>4</sub>F<sub>8</sub> electronic parameters such as the ionization coefficient, attachment coefficient, and diffusion coefficient, resulting in a critical breakdown field strength of 439.5 Td (1 Townsend =  $10^{-17}$  Vcm<sup>2</sup>), which is larger than that of SF<sub>6</sub> (369Td). Itoh *et al.*<sup>192</sup> calculated the electron transport parameters of the mixed gas of SF<sub>6</sub> and c-C<sub>4</sub>F<sub>8</sub>. The insulation characteristics of the mixed gas are better than that of pure gases. Wu et al.<sup>193</sup> and Liu et al.<sup>194</sup> calculated the critical breakdown field strength of the mixed gases of c-C<sub>4</sub>F<sub>8</sub> and N<sub>2</sub> under pulsed discharges using Monte Carlo. The calculated results showed that the insulating properties of the mixed gases of c-C<sub>4</sub>F<sub>8</sub> and N<sub>2</sub> were comparable to those of the SF<sub>6</sub>/N<sub>2</sub> mixed gas, but the GWP value decreased significantly. Liu calculated the insulation properties of the mixed gas of c-C<sub>4</sub>F<sub>8</sub> and CO<sub>2</sub> in the same way. As a result, the critical breakdown strength of the mixed gas of c-C<sub>4</sub>F<sub>8</sub> and CO<sub>2</sub> was greater than that of the mixed gas with  $SF_6/CO_2$ . Li et al.<sup>19</sup> calculated electronic swarm parameters of the c-C<sub>4</sub>F<sub>8</sub> mixed gas The critical breakdown field strength of c-C<sub>4</sub>F<sub>8</sub>-N<sub>2</sub> and c-air were

similar and significantly higher than other mixed gases. The insulation performance of  $C_4F_8$  can reach levels of pure  $SF_6$  when the content exceeds 80%. Deng *et al.*<sup>196</sup> used the same calculation method to analyze the insulation properties of  $C_3F_8$  mixed  $N_2$  and  $CO_2$ . Under the same mixing ratio, the critical breakdown strength of the  $C_3F_8-N_2$  gas mixture is greater than that of  $C_3F_8-CO_2$ . The  $C_3F_8-N_2$  gas mixture containing 20%  $C_3F_8$  can reach 60% of the dielectric strength of pure  $C_3F_8$ . Xiaoling *et al.*<sup>197</sup> proposed a method for evaluating the electronegativity of a gas by calculating the electron transport parameters of the molecule. The electron attachment property of the molecule was obtained by the Boltzmann equation under steady state Thomson discharge. It was found that the ability of  $c-C_4F_8$  to attach electrons was still weaker than that of  $SF_6$  and its electronegativity coefficient was at least three orders of magnitude lower than that of  $SF_6$ .

Koch and Franck<sup>198</sup> studied the characteristics of the C<sub>3</sub>F<sub>8</sub> gas during partial discharge and breakdown. It is found that the discharge characteristics of the SF<sub>6</sub> gas were approximately similar from the results of discharge capacity, flow propagation time, and time interval, which may be related to (E/N)cr, and ionization coefficients were closely related. The same model was used to predict the breakdown voltage and PD voltage of the CF<sub>4</sub> gas.<sup>199</sup> By calculating the breakdown parameters of C<sub>3</sub>F<sub>8</sub>-CF<sub>4</sub>, C<sub>3</sub>F<sub>8</sub>-CO<sub>2</sub>, C<sub>3</sub>F<sub>8</sub>-N<sub>2</sub>, C<sub>3</sub>F<sub>8</sub>-O<sub>2</sub>, and C<sub>3</sub>F<sub>8</sub>-Ar of C<sub>3</sub>F<sub>8</sub> in the temperature range 300-3500 K, Wang et al.<sup>200</sup> found that the ionization coefficient of C<sub>3</sub>F<sub>8</sub>-N<sub>2</sub> was higher, and the attachment coefficient of C<sub>3</sub>F<sub>8</sub>-CF<sub>4</sub> was the largest. Under a normal temperature, the insulation property of C<sub>3</sub>F<sub>8</sub>/N<sub>2</sub> was outstanding. However, the critical breakdown strength of C3F8 decreased with the increase in temperature. The same method was used to calculate the critical breakdown field of the CF4 plasma and predict the insulating properties.<sup>201</sup>

Lisovskiy *et al.*<sup>202</sup> measured the DC breakdown voltage of CF<sub>4</sub> at low pressure and calculated the electron swarm parameter of the gas at a small PL value (PL < 2 Torr cm, where P is pressure and L is distance). Wang *et al.*<sup>203,204</sup> calculated the thermodynamic parameters and transport parameters of the main PFCs (CF<sub>4</sub>, C<sub>2</sub>F<sub>6</sub>, and C<sub>3</sub>F<sub>8</sub>) in the temperature range of 300–30 000 K, including the electron diffusion coefficient, viscosity, thermal conductivity, and conductivity, extending to the third order approximate (viscosity of the second order) Chapman–Enskog method. Theoretical calculations were made to analyze the arc-extinguishing properties of the three gases. The thermodynamic properties of olefinic gases (C<sub>2</sub>F<sub>2</sub>, C<sub>2</sub>F<sub>4</sub>, C<sub>3</sub>F<sub>6</sub>, etc.) containing C=F double bonds were also analyzed.

Kang *et al.*<sup>205</sup> studied the decomposition products of  $c-C_4F_8$ and mixed gas with N<sub>2</sub> at 50 Hz AC corona discharge. The products of pure  $c-C_4F_8$  gas discharge mainly included  $CF_4$ ,  $C_2F_4$ ,  $C_3F_6$ ,  $C_2F_6$ , and  $C_3F_8$ , and  $C_2F_3N$  was produced in the mixed gas.

The insulation properties of  $c-C_4F_8$ ,  $C_3F_8$ , and  $C_2F_6$  all reach or exceed the level of SF<sub>6</sub>, and the GWP values are obviously smaller than those of SF<sub>6</sub> (8700, 7000, and 9200, respectively). However, due to the limitation of the boiling point, it is necessary to mix with the buffer gas. With the addition of the buffer gas, the environmental impact obviously decreases, and the insulating property of the mixed gas also decreases. The main properties of PFCs mixtures are shown in Table VII of the Appendix. There is no obvious advantage in dielectric strength, which restricts the research and exploration process.

#### D. C<sub>n</sub>F<sub>m</sub>X mixtures

#### 1. Mixtures of CF<sub>3</sub>I with natural gases

One of the F atoms in CF<sub>4</sub> is replaced by the iodine atom from the CF<sub>3</sub>I molecule, as shown in Fig. 9, and the performance of the molecule is greatly changed. Initially, CF<sub>3</sub>I was considered as an alternative to the Halon 1301 fire extinguishing agent because CF<sub>3</sub>I has no effect on the environment and is non-flammable and non-explosive.<sup>206,207</sup>

The initial study found that the  $CF_3I$  gas has good electron attachment capacity. The calculation of the electron swarm parameter showed that the critical breakdown field strength was higher and even higher than SF<sub>6</sub>. From 2010, one after another, researchers began to explore the possibility to apply it in electrical equipment.

The AC breakdown voltage of the CF<sub>3</sub>I/N<sub>2</sub> mixed gas in the non-uniform field was investigated, and the effects of pressure, electrode spacing, and mixing ratio were analyzed. When the mixing ratio was 30%, the breakdown voltage of the CF<sub>3</sub>I/N<sub>2</sub> mixture was about 85%–90% of that of SF<sub>6</sub> at a pressure of 0.3 MPa, but the cost of gas was three times higher than the SF<sub>6</sub> one.<sup>208</sup> At the same time, the initial voltage of partial discharge of the CF<sub>3</sub>I/CO<sub>2</sub> mixed gas is 0.9–1.1 times that of SF<sub>6</sub>/CO<sub>2</sub>. The PD performance of the CF<sub>3</sub>I/CO<sub>2</sub> mixed gas, with a CF<sub>3</sub>I volume fraction of 30%–70%, was about 0.74 times of that of pure SF<sub>6</sub>. The CF<sub>3</sub>I/CO<sub>2</sub> mixed gas showed a good synergistic effect, with a value of 0.53.<sup>209</sup>

Toyota *et al.*<sup>210</sup> used square pulse voltage as a transient overvoltage to detect the (V-t characteristic) relationship between the breakdown voltage and the breakdown time of the CF<sub>3</sub>I–N<sub>2</sub> and CF<sub>3</sub>I–air mixture at the plate electrode. The test results showed that the breakdown voltage of pure CF<sub>3</sub>I was 1.2 times that of SF<sub>6</sub> at the same pressure. The breakdown voltage of the CF<sub>3</sub>I–N<sub>2</sub> and CF<sub>3</sub>I–air mixture increased with the increase in the CF<sub>3</sub>I mixing ratio. When the mixing ratio of CF<sub>3</sub>I reached 60%, the dielectric strength of the mixed gas can reach the level of pure SF<sub>6</sub>.

Yuan *et al.*<sup>211</sup> measured the gas–liquid equilibrium data (CF<sub>3</sub>I + CO<sub>2</sub>) at 243.150 K. The experimental data were modified by Peng–Robinson (PR) on the classical van der Waals formula to get the rule that the vapor pressure of the CF<sub>3</sub>I and CO<sub>2</sub> gas mixture and its mole fraction change with the operating environment of GIS/GIL.



Kamarudin et al.<sup>212</sup> tested the breakdown performance of a mixture of CF<sub>3</sub>I and CO<sub>2</sub> (30%:70% mixing ratio) under three kinds of electrodes (rod, plate, and coaxial electrodes) under a lightning impulse voltage. The tests were conducted, and it was assumed that CF<sub>3</sub>I gas mixtures could be used in medium voltage (MV) equipment without involving arc extinguishing equipment. Youping et al.<sup>213</sup> tested the breakdown characteristics of the mixed gas of CF<sub>3</sub>I and N2 in a uniform electric field and concluded that the decomposition products of CF<sub>3</sub>I would affect the insulation properties. The insulation performance of 30% CF<sub>3</sub>I-70% N<sub>2</sub> at 0.1 MPa was similar to that of 20% SF<sub>6</sub>-80% N<sub>2</sub> by DC voltage and lightning impulse voltage. Widger et al.<sup>214</sup> tested the insulation properties of a 30% CF<sub>3</sub>I-CO<sub>2</sub> mixture in a ring-grid cabinet with a vacuum circuit breaker. It was demonstrated by laboratory tests that the gas mixture at this ratio met the requirements for medium-voltage equipment. Chen measured the 50% breakdown voltage of the 30/70% CF<sub>3</sub>I/N<sub>2</sub> mixture at the lightning impulse voltage of a coaxial electrode. In order to explore the feasibility of the CF<sub>3</sub>I-CO<sub>2</sub> mixture in a 400 kV gas-insulated power transmission line (GIL), the experiment was conducted under the equivalent similarity model with the same electric field distribution.<sup>215</sup> The possibility of the CF<sub>3</sub>I/CO<sub>2</sub> mixture for insulation equipment was analyzed from the breakdown performance. It was considered that this gas was not suitable for the arc extinguishing medium, but it can be considered for use in the GIL.  $^{216}$  The breakdown voltage of  $CF_3I$  mixed with  $CO_2$  and  $N_2$ showed a ternary mixed gas and no significant synergistic effect, and under the same conditions, the CF<sub>3</sub>I and CO<sub>2</sub> mixed gas has better breakdown characteristics than CF<sub>3</sub>I-N<sub>2</sub>. This phenomenon was explained by measuring the decomposition products, and calculations showed that CO2 could provide C atoms for the self-recovery of CF<sub>3</sub>I molecules.<sup>2</sup>

de Urquijo<sup>218</sup> measured the electron swarm parameters of the CF<sub>3</sub>I and N<sub>2</sub> mixed gas, including the ionization coefficient, attachment coefficient, drift velocity, and diffusion coefficient. The (E/N)cr of CF<sub>3</sub>I was about 437Td, much larger than that of SF<sub>6</sub>. The breakdown field of the 70%CF<sub>3</sub>I-N<sub>2</sub> mixture is equal to that of SF<sub>6</sub>. Kawaguchi et al.<sup>219</sup> calculated these parameters using the Monte Carlo algorithm and found good agreement with the experimental results. Kimura and Nakamura<sup>220</sup> calculated the electron swarm parameters of the mixed gas of CF<sub>3</sub>I with CO<sub>2</sub>. Li et al.<sup>221</sup> calculated the electronic swarm parameters of CF<sub>3</sub>I mixed with a variety of gases such as CF<sub>4</sub>, CO<sub>2</sub>, N<sub>2</sub>, O<sub>2</sub>, and air. CF<sub>3</sub>I mixed with N<sub>2</sub> and air has more significant advantages, and when the mixing ratio of CF<sub>3</sub>I exceeded 70%, the breakdown field can exceed pure SF<sub>6</sub>. Yun-Kun and Deng-Ming<sup>222</sup> and Deng and Xiao<sup>223</sup> have done a similar calculation. Combining the boiling point and the greenhouse effect, the feasibility of using CF<sub>3</sub>I in high-pressure equipment was analyzed. It was considered that the mixed gas containing 70% CF<sub>3</sub>I had the same dielectric strength as SF<sub>6</sub>. Li<sup>224</sup> established the model to calculate the discharge process of CF<sub>3</sub>I and calculated the time and process of the formation of the streamer current from the electron avalanche to the electron reaching the cathode.

Taki *et al.*<sup>225</sup> studied the interruption capability of CF<sub>3</sub>I. First, the arc time constant and arc power loss coefficient were obtained by using Mayr's equation. The order of the arc time constant was SF<sub>6</sub>, CF<sub>3</sub>I, CO<sub>2</sub>, H<sub>2</sub>, Air, and N<sub>2</sub>, while that of the arc loss coefficient was H<sub>2</sub>, SF<sub>6</sub>, CO<sub>2</sub>, Air, N<sub>2</sub>, and CF<sub>3</sub>I; the short circuit fault interruption performance of CF<sub>3</sub>I is tested, which can reach 90%

of SF<sub>6</sub>. When the volume ratio of CF<sub>3</sub>I exceeded 20%, the interruption ability of CF<sub>3</sub>I mixed with CO<sub>2</sub> approached pure CF<sub>3</sub>I levels. The Breaker Terminal Failure (BTF) interruption capabilities of CF3I and the mixed gas were tested. The BTF interruption capacity of CF<sub>3</sub>I-CO<sub>2</sub> (30%-70%) was about 67% of SF<sub>6</sub>. Yokomizu et al.<sup>228</sup> calculated the electrical conductivity and thermal conductivity of CF<sub>3</sub>I mixed with CO<sub>2</sub> at high temperatures and calculated transient parameters during quenching. When the temperature was above 10 000 K, the conductivity was greatly affected by the content of CF<sub>3</sub>I. At 7000 K, the thermal conductivity was greatly affected by the content of CF<sub>3</sub>I. The calculated results showed that the decay rate of the CF<sub>3</sub>I mixed gas with CO<sub>2</sub> was weaker than that of  $SF_6$ , and the breakoff capacity was worse than  $SF_6$  as a whole. Cressault et al.<sup>229</sup> calculated the plasma thermodynamic parameters (mass density, enthalpy, and specific heat) and transport parameters (viscosity, thermal conductance, and conductance) of CF<sub>3</sub>I mixed with CO<sub>2</sub>, N<sub>2</sub>, and air. The calculated results showed that CF<sub>3</sub>I mixed CO<sub>2</sub> is superior to the other two mixed gases because CO<sub>2</sub> plasma has better heat dissipation ability under a high temperature.

Ngoc et al.<sup>230</sup> tested the DC breakdown voltage of CF<sub>3</sub>I and mixed N<sub>2</sub> at a quasi-uniform electric field (ball-ball electrode). The breakdown voltage of pure CF<sub>3</sub>I was greater than that of SF<sub>6</sub>, but the synergistic effect with N2 was not obvious. With the increase in the CF<sub>3</sub>I mixing ratio, the breakdown voltage also increased. When the mixing ratio of CF<sub>3</sub>I was less than 50%, the breakdown voltage of the mixed gas was lower than the SF<sub>6</sub>-N<sub>2</sub> mixed gas with the same ratio. After the breakdown experiment, it was found that solid iodine was deposited on the electrode surface. Kasuya et al.<sup>231,232</sup> tested the decomposition products of the CF<sub>3</sub>I-CO<sub>2</sub> (30%-70%) mixed gas during quenching and improved iodine deposition by adding adsorbents. Continuous breakdown of CF3I for 1300 cycles resulted in a 11% reduction in breakdown voltage.<sup>233</sup> The solid iodine also was generated by CF<sub>3</sub>I-CO<sub>2</sub> gas mixture breakdown at the lightning impulse voltage.<sup>234-236</sup> Jamil et al.<sup>237</sup> quantitatively detected C<sub>2</sub>F<sub>6</sub>, C<sub>2</sub>F<sub>4</sub>, C<sub>2</sub>F<sub>5</sub>I, and a few C<sub>3</sub>F<sub>8</sub>, CHF<sub>3</sub>, C<sub>3</sub>F<sub>6</sub>, and CH<sub>3</sub>I within 224 h of CF<sub>3</sub>I partial discharge under a non-uniform field. The presence of H<sub>2</sub>O and O<sub>2</sub> would have an impact on the formation of decomposition products during discharge of CF<sub>3</sub>I and accelerates the decomposition of CF<sub>3</sub>I and produce toxic substances such as CF<sub>2</sub>O.

Although  $CF_3I$  has high insulation properties, it cannot be used alone in equipment due to the boiling point. Under the premise of ensuring the boiling point, the mixed gas with the buffer gas added cannot reach the level of pure  $SF_6$ , and there is no obvious arc extinguishing advantage. After the gas discharge, the obvious solid main atoms are generated (I and C), which is very detrimental to gas selfrecovery in the use of the device. The oxidation of materials due to the presence of iodine could not be ignored. Its acute toxicity is acceptable, but this gas is genotoxic, so the safety of practitioners is a threat in the long-term operation of the device. Although a great deal of exploration has been done, all these achievements are carried out in the laboratory, and it is unlikely that the gas will be used in the equipment at present.

#### 2. Mixtures of C<sub>4</sub>F<sub>7</sub>N with natural gases

In addition to the above gases, the bond in the carbon-fluorine bond is broken and replaced by atoms such as N to form some new



derivatives such as C<sub>4</sub>F<sub>7</sub>N, the molecular structure of which is shown in Fig. 10.

In 2015, Alstom and 3M collaborated to test the power frequency dielectric strength of the C<sub>4</sub>F<sub>7</sub>N- CO<sub>2</sub> mixture in a 145 kV GIS. The study found that when the content of  $C_4F_7N$  is 18%–20%, it can achieve the same dielectric strength as pure SF<sub>6</sub>.<sup>47</sup> The frequency and lightning impulse breakdown of the C<sub>4</sub>F<sub>7</sub>N/CO<sub>2</sub> mixed gas under different electric field types were tested under the influence of the mixing ratio, electrode distance, and gas pressure by Nechmi et al. from the French National Academy of Sciences AMPERE Lab.<sup>240</sup> It was found that the synergistic effect of C<sub>4</sub>F<sub>7</sub>N and CO<sub>2</sub> at a concentration of 3.7% was obvious. At present, the C<sub>4</sub>F<sub>7</sub>N/CO<sub>2</sub> mixed gas (g3) is tested in a 420 kV GIL and 245 kV current transformers. Owens<sup>241</sup> conducted the breakdown test of  $C_4F_7N$  with  $CO_2$ ,  $N_2$ , and dry air mixtures at different pressures on a plate-plate electrode and found that the mixture had the best dielectric strength with  $CO_2$ . Preve *et al.*<sup>242</sup> tested the dielectric strength of the C<sub>4</sub>F<sub>7</sub>N/dry air mixture and found that its insulation properties were superior to SF<sub>6</sub> at impulse voltage but slightly inferior to SF<sub>6</sub> at AC voltage. Kieffel et al.<sup>243</sup> also conducted a continuous 100-s breaking test on  $C_4F_7N/CO_2$  (the mixing ratio of  $C_4F_7N$  is 4%), with an average arcing time of 12 ms, which was lower than that of pure  $SF_6$  (15 ms) at the same pressure.

C<sub>4</sub>F<sub>7</sub>N has a very high dielectric strength and is currently the most promising alternative to SF<sub>6</sub> in existing gases. However, the presence of a cyano-group ( $C \equiv N$ ) in the molecule can be a hazard to the safety of the gas used in the equipment and the toxicity of the pure gas is not yet demonstrated, especially regarding neurotoxicity. Its main decomposition products and reaction with the molecules in the environment have also drawn attention.<sup>244,245</sup> For C<sub>4</sub>F<sub>7</sub>N, there may be some highly toxic by-products, such as CO and PFIB (C<sub>4</sub>F<sub>8</sub>), after 100 C-O 630 A/24 kV, the LC50 (4 h on mice) in an 11-l epoxy bulb of C<sub>4</sub>F<sub>7</sub>N/air or C<sub>4</sub>F<sub>7</sub>N/CO<sub>2</sub> mixture is inferior to 225 ppm.<sup>246</sup> Sulbaek Andersen et al.<sup>247</sup> measured the possible chemical reactions of C<sub>4</sub>F<sub>7</sub>N with OH, Cl, and O<sub>3</sub> in the atmosphere and found the products to be COF<sub>2</sub> and CF<sub>3</sub>COF. The GWP of the gas was evaluated based on the infrared absorption spectrum. Due to the low content of the gas, the decomposition products did not adversely affect the environment. Blázquez et al.24 calculated the reaction of gas molecules with OH radicals using the

quantum chemical calculation method and obtained the reaction rate at 278–358 K.

The calculation of the molecular structure based on DFT mainly refers to the binding energy, ionization energy and molecular orbital gap, and decomposition process of the gas molecules.<sup>249</sup> The breakdown voltage of  $C_4F_7N/N_2$  at power frequency voltage for 30 times does not change much, but a series of products are produced, such as  $CF_4$ ,  $C_3F_8$ ,  $CF_3CN$ ,  $C_2F_4$ ,  $C_2F_5CN$ , and  $C_3F_6$ .<sup>250</sup> It is noteworthy that the formed  $CF_3CN$  and  $C_2F_5CN$  are highly toxic gases and the toxicity of the gas  $C_4F_7N$  also needs further research and confirmation.<sup>251</sup>  $C_4F_7N$  would further react with H and OH, and the main production are  $CF_2HCN$ ,  $CFH_2CN$ ,  $CF_3CHFCN$ , and  $CF_3CH_2CN$ .<sup>252</sup> The computational decomposition process was studied, and the reaction of the molecules within the device with the metal Cu and Al surface was considered.<sup>253,254</sup>

The gas is currently a research hotspot and also has great potential for the gas medium used in high-voltage equipment. Due to the current use of less gas, the price of gas is relatively expensive. The gas greenhouse effect is low for high voltage applications and still high for medium voltage applications, while maintaining a low mixing ratio can still meet the equipment requirements of the insulation operation. However, due to the cyano group (C=N) contained in the molecular structure, the free radical may react with others or atoms and even the metal in the device during discharge to generate toxic substances such as HCN and CuCN. This section needs further research and exploration to avoid the health threats.

# 3. Mixtures of perfluoroketones ( $C_5F_{10}O$ and $C_6F_{12}O$ ) with natural gases

In 2015, ABB proposed to consider the use of ketones  $C_5F_{10}O$  or  $C_6F_{12}O$  (boiling points of 26.5 °C and 49 °C, respectively) in combination with the buffer gas as an insulating medium.<sup>255</sup> The molecular structures are shown in Fig. 11.

For the insulation properties, Mantilla et al.<sup>255</sup> found that the 50% positive lightning impulse breakdown voltage of a mixture of 5% C<sub>5</sub>F<sub>10</sub>O and dry air at 0.7 MPa reached that of pure SF<sub>6</sub> at 0.4 MPa. Simka and Ranjan<sup>256</sup> tested the breakdown frequency field of C<sub>5</sub>F<sub>10</sub>O and dry air under a quasi-uniform electric field and found that the critical breakdown strength of the mixed gas containing 5.2%  $C_5F_{10}O$  at 0.7 MPa can reach 95% of pure SF<sub>6</sub> at 0.45MPa % and 80% of pure SF<sub>6</sub> at 0.6 MPa. For the engineering applications, Saxegaard *et al.*<sup>257</sup> found that the lightning impulse withstand level increased by 60% and the power frequency voltage level increased by 44%, by filling a 12 kV air switch cabinet with the C<sub>5</sub>F<sub>10</sub>O-air mixture. Hyrenbach and Zache<sup>258</sup> has produced a switchgear insulated with the C<sub>5</sub>F<sub>10</sub>O mixed gas, which was commissioned in 2015 in a substation of Zurich. The switchboard passed the IEC62271-200 standard requirements of the full voltage test, and at this stage, C<sub>5</sub>F<sub>10</sub>O mixed with air can be used for medium voltage equipment (6–40.5 kV). The switchboard with  $C_5F_{10}O$  as the insulating medium is developed by ABB, and the gas chamber is made of stainless steel to prevent the metal materials inside the equipment from being oxidized.

By calculating the thermodynamic parameters and transport parameters of plasma at a high temperature of  $C_5F_{10}O$  gas molecules,



FIG. 11. C<sub>5</sub>F<sub>10</sub>O and C<sub>6</sub>F<sub>12</sub>O.

it is considered that it has a better arc extinguishing property than SF<sub>6</sub> from the nature of transport. Part of the particle structure is complex, so its parameters (such as partition function and polarizability) have no reference data, which are calculated by computational chemistry.<sup>259</sup> However, Zhong *et al.*<sup>260</sup> calculated the thermodynamic properties of  $C_5F_{10}O$  mixed with CO<sub>2</sub> and O<sub>2</sub>. The calculation results showed that due to decomposition of  $C_5F_{10}O$  in the process of quenching, the gas at the end of quenching cannot completely recover. The main products were CF<sub>4</sub>, CO<sub>2</sub>,  $C_4F_6$ , and graphite carbon. O<sub>2</sub> were added to the mixture without affecting the thermodynamic parameters and transport parameters. Therefore, the existing equipment is used in a mixture of  $C_5F_{10}O$  and air.

Tatarinov *et al.* explored the decomposition products of  $C_5F_{10}O$ -dry air mixed gases under the application of 10 kV dielectric barrier discharge (DBD) and found that  $C_4F_{10}$ ,  $C_6F_{14}$ ,  $C_3F_6$ ,  $C_5F_{12}$ , and other substances were produced.<sup>261</sup> Based on the optimization of the molecular model, the carbonyl carbon atom is the active site of C5-PFK molecules, and the decomposition of C5-PFK may take place centered on the carbonyl carbon atom.<sup>262–264</sup>

At the same time, the impact of C5 on the environment and personal safety also attracted attention. ABB tested the gas composition of the GIS with AIRPLUS gas ( $C_5F_{10}O$  and air) during normal operation and gas leakage.<sup>265</sup> The mixture is not to be a threat to the environment or human body, but the LC50 (4 h) for mice after breaking tests lead to 2100 ppm, which is far lower than the one of gas mixture before breaking.<sup>246</sup>

For  $C_6F_{12}O$ , because of the high boiling point of the gas, the mixing ratio is seldom used in the equipment. Under the frequency test, adding 3%  $C_6F_{12}O$  to  $N_2$  can increase the breakdown voltage of pure  $N_2$  almost by 1 time.<sup>79</sup> The products after breakdown are PFCs and thus have no effect on the insulation properties. The content is low, which will not affect the environment. It is considered that the gas is more suitable for use in medium and low voltage equipment. By adding a small amount of  $C_6F_{12}O$  in  $N_2$ , the insulation level of the equipment is improved. Existing studies have shown that the  $C_6F_{12}O/CO_2$  mixed gas has the potential for application in a 10 kV switchgear.<sup>266,267</sup>

The gas insulation far exceeds SF<sub>6</sub>, but the problem of boiling point has also become very prominent. At present,  $C_5F_{10}O$  is expected to be the main insulating gas for medium voltage equipment. The main properties of  $C_nF_mX$  mixtures are shown in Table VIII of the Appendix.

#### E. Others

Since C atoms and F atoms have good electronegativity, the gas molecules, formed by the combination of C and F with other atoms such as H, Cl, S, and N, generally also have good insulation properties. Devins in 1980 summarized the basic insulation properties of gases,<sup>32</sup> such as  $CF_nCl_{3-n}$ ,  $CHF_nCl_{3-n}$ , and  $C_nH_{2n+2}$ ; their insulation properties gradually increased with the increase in n, but the boiling point was also gradually obvious. However, some gases have the greenhouse effect, and some were toxic gases such as  $CF_3CN$ . Therefore, there was not much gas available for reference at the moment.

CF<sub>3</sub>NSF<sub>2</sub> was also considered to have high-performance gas insulation, the insulation performance of about 2.41 times the SF<sub>6</sub>, and the boiling point of -6 °C. Cheng *et al.* calculated the reaction mechanism of this molecule with OH.<sup>268</sup> The literature for the study of this gas is still relatively less, and the field of its application is relatively vague now. The gas products available are difficult to supply. There is not much literature on the experimental research of its various properties.

 $CCl_2F_2$  (R12) is a Freon gas with a GWP of 2400 that is far less than that of SF<sub>6</sub>. The experimental study found that the insulation strength of the gas was about 90% of that of SF<sub>6</sub>. The mixed gas showed good self-stability through mixed air and N<sub>2</sub> breakdown experiments.<sup>48</sup> However, this gas has a large damage to the ozone layer, with the ozone depletion potential (ODP) up to 1.0.

HFO1234zeE (CF<sub>3</sub>CH=CHF) is non-toxic, non-flammable, and non-destructive of the ozone layer, with a GWP value of less than 0.02 and the lifetime of 1 day. It is generally used as a refrigerant. Through the application of AC and positive and negative lightning impulse voltage, the breakdown test was studied under the uniform field and extreme non-uniform field. HFO1234zeE showed very similar insulating properties to SF<sub>6</sub>. It can therefore be considered as a substitute for medium-voltage equipment.<sup>49</sup> For the electron swarm parameter of the HFO1234zeE molecule under PT discharge, the gas was found to have strong electron attachment capacity.<sup>269</sup> In medium- or high-voltage applications, the HFO1234zeE/N<sub>2</sub> mixture would show more excellent electrical strength in the range of 100 kPa to 1 MPa than currently obtained at 10 kPa. HFO1234zeE has comparable electrical strength to SF<sub>6</sub> at 130 kPa. There is no synergistic effect in the HFO1234zeE/N<sub>2</sub> and HFO1234zeE/CO<sub>2</sub> mixtures.<sup>270</sup> The main characteristics of this gas demonstrates that it can be used as a dielectric medium for the MV switchgear, but it cannot be used in breaking.<sup>271</sup>

Chen<sup>272</sup> analyzed the molecular structure of OCCHCN and concluded that it was not easily ionized and excited based on quantum chemistry calculation, but there was no experimental data to validate it.

The insulation properties of gases can be predicted to filter potential alternative gases based on the calculation results. The parameters that affect the gas insulation performance are the relative molecular mass M, the dipole moment  $\mu$ , the polarizability  $\alpha$ , and so on. The ionization collisions increase the number of carriers, thereby weakening the gas dielectric strength. The electron attachment collision does not increase the number of carriers but can slow down carriers, which is helpful to strengthen the dielectric strength. Molecules with higher ionization energy are more susceptible to ionization collisions. More electronegative molecules attach electrons more easily, so insulating gas should have higher ionization energy (IE) and electronegativity.

 $Yu^{58}$  builds the new structure-activity relationship (SAR) models to calculate the molecular insulation properties by combining the surface area, polarizability, electronegativity, and molecular hardness of a molecule or functional group. Through the calculation of the existing molecules, it is found that the functional group CF<sub>3</sub> has outstanding dielectric strength in the molecular structure due to its large surface area and polarizability. At the same time, taking into account the impact of the boiling point, the authors believe that SF<sub>5</sub>CN and SF<sub>5</sub>CFO have greater potential to replace SF<sub>6</sub>.

#### V. CONCLUSION AND OUTLOOK

#### A. Conclusion

Table IV shows the main alternative gases and their properties. The main studies of several major alternative gases now draw the following conclusions:

- (1) For natural gas, the insulation performance is only 40% of  $SF_6$  or even lower, but safety and low cost become the advantages. By optimizing the structure of the equipment, increasing pressure, and combining solid insulation, the natural gas could be used in medium and low voltage equipment.
- (2) The research results of  $SF_6$  mixtures are quite mature.  $SF_6/N_2$  is a relatively stable combination for the moment and has been used in GILs and switchgear cabinets. However, studies have shown that  $CO_2$  shows obvious thermodynamic properties at high temperatures despite the slightly lower insulating properties of  $CO_2$  mixtures, and the sensitivity of the mixed gas to the electric field uniformity is lower than that of the  $N_2$  mixed gas. For interrupter properties, the  $SF_6$ - $CF_4$  mixed gas has also been used in circuit breakers. As for decomposition performance, the decomposition products of the mixed gas are more complicated than pure  $SF_6$ . However, the reduced use of  $SF_6$  also inhibits the production of toxic sulfur compounds.
- (3) PFCs have some advantages over SF<sub>6</sub> in terms of GWP, and both theoretical calculations and experimental measurements show that the insulating properties of pure gas are superior to that of SF<sub>6</sub>. Affected by the boiling point, the insulating properties of the mixture cannot reach the level of SF<sub>6</sub>.
- (4) Despite its superior gas-insulating properties and no greenhouse effect, the decomposition of CF<sub>3</sub>I after discharge is the

**TABLE IV**. Basic properties of main alternative gases. G presents that the property is better than that of  $SF_6$ . NG presents that the property is worse than that of  $SF_6$ . E presents that the property is comparable to that of  $SF_6$ . – presents that the gases were studied but have no real application.

Gases	Boiling point	GWP/ODP	Insulation	Interruption	Decomposition	Application
SF <sub>6</sub>	Good	Not good	Very good	Very good	Not good (toxic but in low concentration)	GIS/GIL/GCB/GIT
$N_2$	G	G	NG	NG		C-GIS(72 kV)
Air	G	G	NG	NG		C-GIS(40 kV)
CO <sub>2</sub>	G	G	NG	NG		40 kV C-GIS 70 kV GCB
$SF_6 + N_2$	G	G	Е	NG	G	245-550 kVGIL
$SF_6 + CO_2$	G	G	Е	NG	G	
$SF_6 + CF_4$	G	G	Е	Е	G	550 kV GCB
c-C <sub>4</sub> F <sub>8</sub> mixtures	NG	G	Е	NG	NG	
C <sub>3</sub> F <sub>8</sub> mixtures	NG	G	Е	NG		
C <sub>2</sub> F <sub>6</sub> mixtures	NG	G	NG	NG		
$CF_4$	NG	G	NG	NG		
CF <sub>3</sub> I mixtures	NG	G	Е	NG	NG	
$C_4F_7N + CO_2$	NG	G	Е	Е	NG in MV equipment than SF <sub>6</sub>	420 kV GIL/245 kV CT
$C_5F_{10}O + air$	NG	G	Е	Е	NG in MV equipment than SF <sub>6</sub>	MV GIS(6-40.5 kV)
C <sub>6</sub> F <sub>12</sub> O mixtures	NG	G	Е		Ğ	C-GIS(12 kV)
HFO1234zeE	G	G	E	NG	G	C-GIS (24 kV)

Gases	Breakdown voltage/partial discharge voltages	Voltage/	electrode	P (MPa)	Reference
	The breakdown voltage can be increased by 1.5 times at 0.2 MPa by composite solid insulation.	AC/roo	l-plane	0.1-0.6	82
N <sub>2</sub>	The breakdown voltage is lower for positive polarity waveforms in the presence of a bias voltage.	Non-staı rod-j	ndard-LI plane	0.7	87
	The breakdown voltage of $\rm N_2$ at 1.0 MPa can be equal to that of 0.5 MPa $\rm SF_6.$	LI/spheri	cal-plane	0.6–2.0	94
	The breakdown values of $\text{CO}_2$ are more stable than those of $N_2$ .	AC and LI J	plane-plane	0.7-1.1	84
$CO_2$	The breakdown voltage is lower for positive polarity waveforms in the presence of a bias voltage.	Non-star rod-j	idard-LI/ plane	0.7	86
Air	The dielectric strength of air is higher than that of $N_2$ and $CO_2$ at 0.5 MPa based on the initial flashover voltage and the 50% flashover voltage.	LI/coaxial- and para	cylindrical llel plate	0.1-0.5	91
	The partial discharge voltages of air and $N_2$ are almost the same, but the breakdown voltage of air is larger than that of pure $N_2$ .	LI/rod-plane		0.1-0.4	93
N <sub>2</sub> O/N <sub>2</sub>	The dielectric strength for 20/80% $N_2O/N_2$ at 0.6 MPa showed a synergy effect.	DC/need	le-plane	0.1-1.0	88
N <sub>2</sub> -He	The breakdown voltage of $N_2$ -He rises rapidly and then follows a linear behavior with the mixing ratio of $N_2$ .	AC/plate-plate		<0.6	89
N <sub>2</sub> /CO <sub>2</sub>	The breakdown voltage of the $50\%N_2/50\%CO_2$ mixture is only one-third that of SF <sub>6</sub> .	LI/sphere-plane		01-30	83
Gases	Electron swarm parameters	Method	E/N (Td) T (K)	P (MPa)	Reference
	Small additions of $CO_2$ (5%) produced a significant attachment in the air- $CO_2$ mixtures.	Measure	90–260 	10–100 Torr	100
	In the mixtures of $CO_2$ with both 50% $O_2$ and 50% $H_2$ , (E/N) <sub>cr</sub> clearly shows higher values than in pure $CO_2$ .	Calculation	0–200 300–3500	0.1 MPa	96
Mixtures	Adding a small amount of $O_2$ to $CO_2$ can effectively improve the value of $(E/N)_{cr}$ with a clear synergistic effect.	Calculation	0–200	0.1 MPa	97
	The reduced critical breakdown field of $\mathrm{CO}_2$ with $\mathrm{O}_2$ and $\mathrm{CH}_4$ show higher values.	Calculation	0–150 300–4000	0.1 MPa	98
	The values of $(\alpha - \eta)/N$ for $CO_2 - N_2$ mixtures are about 30 times smaller than those for $CO_2 - O_2$ mixtures.	Measure/ calculation	0.01–1000 293–298	0.6–600 Torr	99
Gases	Interruption ability	Met	hod	P (MPa)	Reference
Air/CO <sub>2</sub>	For a fixed pressure, the gases were ranked in terms of arc interruption performance as follows, in increasing order: air, $CO_2$ , and $SF_{6}$ .	Measure/calculation 300–4000 K		0.5	103
Air	A low nozzle-to-contact diameter ratio is favorable in terms of requiring less upstream overpressure for successful interruption of air.	Measure/calculation Load currents: 300–900 A		0.05–1.4 bar (upstream)	107
CO <sub>2</sub>	The thermal interruption capability of $CO_2$ could be estimated to be about 50% of that of $SF_6$ .	Mea 84 kV	sure GCB		108

TABLE V. Basic properties of natural gases and mixtures.

## **TABLE VI**. Basic properties of $SF_6$ mixtures.

Gases	Breakdown voltage/partial discharge voltages	Voltage/electrode	P (MPa)	Reference
	$SF_6-N_2$ mixtures containing 50%–60% of $SF_6$ have a dielectric strength of up to 85%–90% that of pure $SF_6$ .	AC/DC/LI Plate-plate/rod-sphere	0.1-0.5	113
	A simplified streamer criterion proposed for $SF_6$ can be used to calculate the discharge inception levels in $SF_6-N_2$ mixtures.	DC Sphere-plane/ sphere-sphere	0.1-0.5	114
	The addition of 0.1% of SF <sub>6</sub> into $N_2$ results in an increase in the breakdown voltage at pressures below 2.5 bars.	PI Rod-plane	0.1-0.5	115
$SF_6 + N_2$	The dielectric strength of the 50% $SF_6/N_2$ mixture is about 8–12% higher than that of the 20% $SF_6$ gas mixture.	LI and AC Sphere-plane/rod-plane	0.3-0.7	118
о <u>-</u>	The increase in corona onset voltages with the increase in the amounts of $SF_6$ . The values of positive polarity are higher to those obtained with negative polarity.	DC Tip-plane	0.3-1.5	119
	An increase in the GIS tank diameter of about 15% with the 10% $SF_6-N_2$ mixture at 0.6 MPa compared to pure SF6 at 0.4 MPa.	LI/SI/AC Coaxial cylinder	0.2-0.6	120
SF <sub>6</sub> + CO <sub>2</sub>	$N_2$ –SF <sub>6</sub> mixtures with a SF <sub>6</sub> content of 1%–20% and pressure of 0.71 MPa–1.24 MPa are more sensitive to fixed protrusions than pure SF <sub>6</sub> at 0.5 MPa of the same dielectric strength.	AC/LI/SI Needle-plane	0.5-1.24	144
	The negative synergistic effect of the $SF_6/N_2$ mixture appeared with low gas pressure.	voltagesVoltage/electrodeP (Na dielectricAC/DC/LI Plate-plate/rod-sphere0.1 Plate-plate/rod-sphere0.1 Plate-plate/rod-sphereun be used to mixtures.DC Sphere-plane/ sphere-sphere0.1 OC Sphere-plane/ sphere-sphereucrease rs.PI Rod-plane Sphere-plane/rod-plane0.3 OC OC OC Sphere-plane/rod-planevease in vease in owith the 10% at 0.4 MPa.DC Tip-plane 	0.2-0.5	122
$SF_6 + CO_2$	With a single large protrusion on the electrode, the breakdown strength of the $50\%$ SF <sub>6</sub> –CO <sub>2</sub> mixture is slightly higher than that of SF <sub>6</sub> .	AC Cylindrical	0.1-0.4	132
	With adding $CO_2$ to the $N_2/SF_6$ gas mixture, the breakdown values rise significantly, particularly at high pressures.	AC/LI Needle-plane	$\begin{array}{c c} P (MPa) \\ 0.1-0.5 \\ 0.1-0.5 \\ 0.1-0.5 \\ 0.1-0.5 \\ 0.1-0.5 \\ 0.3-0.7 \\ 0.3-1.5 \\ 0.2-0.6 \\ 0.5-1.24 \\ 0.2-0.6 \\ 0.5-1.24 \\ 0.2-0.5 \\ \hline 0.1-0.5 \\ 0.1-0.6 \\ e \\ 0.1-0.5 \\ \hline 0.1-0.5 \\ 0.05-0.3 \\ 0.2-0.4 \\ 0.3-1.0 \\ \hline 0.1-0.4 \\ 0.05-0.3 \\ \hline \end{array}$	133
SEc + Air	The rate of rise impulse wave voltage has a significant effect on the breakdown voltage of $1\%$ SF <sub>6</sub> /air mixture.	LI/SI Coaxial cylinder/rod-sphere	0.1-0.5	129
516 1 711	SF <sub>6</sub> -air mixtures have corona stabilization and is higher than the corresponding values for SF <sub>6</sub> -CO <sub>2</sub> and SF <sub>6</sub> -N <sub>2</sub> mixtures.	DC Rod-plane	0.1-0.5	128
	The PD inception voltages have hardly any effect on mixed rate of $SF_6$ in 25.8 kV GIS	AC/LI Sphere-sphere	0.05-0.3	138
$SF_6 + CF_4$	$SF_6$ - $CF_4$ mixtures under uniform fields have breakdown voltages that vary linearly with the $SF_6$ content. Under highly non-uniform fields, a corona stabilization may occur under positive impulse, positive DC and AC voltages leading to high breakdowns for the mixtures 10% SF6-90% CF4 to 25% SF_6-75% CF_4.	AC/LI/DC Sphere-sphere	0.2-0.4	140
	The AC corona inception occurred at lower voltage levels than DC. The average time to breakdown corresponding to negative lighting impulse breakdown voltages are generally shorter than those of positive	AC/DC/LI0.1-0.511Plate-plate/rod-sphere0.1-0.511Sphere-plane/ sphere-sphere0.1-0.511Pl Rod-plane0.1-0.511Sphere-plane/rod-plane0.3-0.711DC Tip-plane0.3-1.511DC Tip-plane0.3-0.712AC/LI/SI0.2-0.612AC/LI/SI0.5-1.2414LI Rod-plane0.2-0.512AC Cylindrical0.1-0.413Needle-plane0.1-0.512AC Cylindrical0.1-0.512DC Rod-plane0.1-0.512DC Rod-plane0.1-0.512AC/LI Sphere-sphere0.05-0.313AC/LI/DC Sphere-sphere0.2-0.414AC/DC/LI Sphere-sphere0.3-1.014AC Rod-plane/ rod-rod0.1-0.413AC Rod-plane/ rod-rod0.1-0.413AC Rod-plane/ rod-rod0.1-0.413AC Rod-plane0.1-0.414LI Rod-plane0.1-0.415AC Rod-plane/ rod-rod0.1-0.415	142	
$SF_6 + N_2$ $SF_6 + CO_2$ $SF_6 + Air$ $SF_6 + CF_4$ $SF_6 + H_2$	The addition of 0.002% SF <sub>6</sub> to $H_2$ resulted in a substantial increase in AC breakdown voltage of typically 70%.	AC Rod-plane/ rod-rod	0.1-0.4	135
$SF_6 + H_2$	The increase in negative breakdown voltage with the addition of small quantities (<1%) of $SF_6$ is smaller than for positive-impulse conditions	LI Rod-plane	0.05-0.3	136

## TABLE VI. (Continued.)

Gases	Electron swarm parameters	Method	E/N (Td) T (k)	P (MPa)	Reference
$\frac{SF_6 + N_2}{SF_6 + N_2O}$	The breakdown is governed by the mean energy of $SF_6$ for low concentrations of $SF_6$ up to 10%, and only for still lower concentrations, the mean energy of electrons corresponds to that of $N_2$ .	Calculation	700 273		148
$SF_6 + N_2$	The variation of the effective ionization coefficient is non-linear with the mixture ratio.	Measure SST	182–258 293	0–100 Torr	150
$SF_6 + N_2$	The variation of the effective ionization coefficient does not vary linearly with the ratio of the two gases in the $SF_6-N_2$ mixture. With an increasing $SF_6$ content in the mixture, a reduction in the apparent secondary ionization coefficient is observed.	Calculation	100-450 V cm <sup>-1</sup> Torr <sup>-1</sup>	5–40 Torr	151
	The rate of increase in the attachment coefficient with an increase in the percentage of $SF_6$ in the $SF_6$ -CO <sub>2</sub> mixtures was much larger than the rate of change in the first ionization coefficient.	Measure (SST) sphere-sphere/ rod-rod		0.4	152
	$CO_2$ in SF <sub>6</sub> – $CO_2$ mixtures can change the transport properties, but the effect of detachment is relatively slight on the properties of electrical discharge.	Calculation (PT) parallel plate	270-370		155
$SF_6 + CO_2$	The influence of SF <sub>6</sub> in the SF <sub>6</sub> –CO <sub>2</sub> mixture is strongly apparent in the values for the effective ionization coefficients. The value of critical field strength $E/N_{cr}$ is smaller than that measured for the SF <sub>6</sub> –N <sub>2</sub> mixtures.	Measure (PT)	100–700 293–302	P (MPa)       P          Image: state sta	156
	At temperatures above 1750 K, an addition of $CO_2$ to $SF_6$ gas can enhance dielectric breakdown performances, while at low temperatures, too much CO2 added into mixtures can reduce dielectric breakdown abilities.	Calculation	 300–3500		162
	After addition $CF_4$ to $SF_6$ , the electron drift velocity in $SF_6$ and $CF_4$ increases, effective ionization coefficient decreases with the increase in the $SF_6$ content, and diffusion of $CF_4$ is stronger than that of $SF_6$ .	Calculation (Monte Carlo)	150–500 293 K	1 Torr	158
$SF_6 + CF_4$	For higher gas temperatures (i.e., $T > 2200$ K at 0.4 MPa), the (E/N)cr in SF <sub>6</sub> -CF <sub>4</sub> mixtures are obviously higher than that in pure SF <sub>6</sub> .	Calculation (Boltzmann)	300-3000	 0-100 Torr 5-40 Torr 0.4  0-0.001 0.01-1.0 1 Torr 0.01-1.6 1 Torr 0.01-1.6 P (MPa) 0.1 0.1 0.01 and 1.6	162
	The mixture of SF <sub>6</sub> and CF <sub>4</sub> exhibits a suppressed CF <sub>4</sub> ionization and an increased SF <sub>6</sub> ionization in the mixture relative to pure CF <sub>4</sub> and SF <sub>6</sub> .	Calculation	140–600 293 K	1 Torr	159
$SF_6 + He$	The addition of He to the gaseous mixture decreases the reduced critical electric field.	Calculation (Boltzmann)	 300–3500	0.1–1.6	171
Gases	Interruption ability	Me	thod	P (MPa)	Reference
$SF_6 + N_2$	Under two-temperature assumption, the electron conductivity of the $SF_6-N_2$ mixture is distinctly lower than that of pure $N_2$ .	Calculation 300-4	LTE/NLTE 0 000 K	0.1	173
	Mixing $CO_2$ with $SF_6$ at a certain ratio, compared to the case of pure $CO_2$ , the largely preserved thermal cooling effect of $SF_6$ can favor the thermal recovery of circuit breakers during current interruption.	case Calculation in LTE 300–30 000 K		0.01 and 1.6	170

#### TABLE VI. (Continued.)

Gases	Interruption ability	Method	P (MPa)	Reference
	The arc interruption capabilities of the $SF_6-CO_2$ mixture increase greatly with an increase in $SF_6$ concentration and are about 40%, 45%, and 70% that of $SF_6$ at a $SF_6$ content of 0%, 20%, and 50% in the applied puffer GCB test prototype and at a short-circuit current of 10 kA.	Measure short-circuit current: 10 kA		174
$SF_6 + CF_4$	The net radiation of a SF <sub>6</sub> -CF <sub>4</sub> or SF <sub>6</sub> -C <sub>2</sub> F <sub>6</sub> mixture is always greater than that of pure SF <sub>6</sub> . The SF <sub>6</sub> /CF <sub>4</sub> mixture has interruption properties very close to those of pure SF <sub>6</sub> as long as the proportion of SF6 is about 50%.	Calculation in LTE 300–30 000 K	0.1	168,169
	An SF <sub>6</sub> /CF <sub>4</sub> filled circuit breaker often retains its breaking capacity down to the lowest ambient temperatures.	Method       P (         Measure       Measure         Ire about       short-circuit         current: 10 kA       Image: Calculation in LTE         ivys       Calculation in LTE         ng as       300–30 000 K         Measure       0.1         TFT/SFT       0.1         i of       Measure (LTS)         Method       P (         AC sparks discharge       Point-insulator-plane         2, S2OF10,       DC/AC corona discharges         ssayed.       Point-plane         ured       Spark discharges         Point-plane       0.1         Corona discharges       0.1         Point-plane       0.1	0.5,0.7	172
SF <sub>6</sub> + Ar	The diameter of the Ar/SF <sub>6</sub> (20%) arc was smaller than that of the Ar arc. Before interrupting the arc current, the electron density of the Ar/SF <sub>6</sub> (20%) arc was larger than that of Ar. After the current interruption, however, the electron density of the Ar/SF <sub>6</sub> (20%) arc decreased rapidly.	Measure (LTS)	0.1	166
Gases	Decomposition products	Method	P (MPa)	Reference
$SF_6 + CF_4$	The gaseous by-products of the spark decomposition of mixtures included $SOF_2 + SF_4$ , $SOF_4$ , $SO_2F_2$ , $S_2F_{10}$ , $S_2OF_{10}$ , $S_2O_2F_{10}$ , and $S_2O_3F_6$ with $O_2$ and $H_2O$ .	AC sparks discharge Point-insulator-plane	0.1	176
$SF_6 + N_2$	The decomposition products SOF <sub>4</sub> , SO <sub>2</sub> F <sub>2</sub> , SF <sub>4</sub> + SOF <sub>2</sub> , SO <sub>2</sub> , S <sub>2</sub> OF <sub>10</sub> , $S_2F_{10}$ , $S_2O_2F_{10}$ , $S_2O_3F_6$ , SF <sub>5</sub> NF <sub>2</sub> , NF <sub>3</sub> , and (SF <sub>5</sub> ) <sub>2</sub> NF were assayed.	DC/AC corona discharges Point-plane	0.4	179
$SF_6 + O_2$	The yields of $SOF_4$ , $SO_2F_2$ , $SOF_2$ , and $SO_2$ have been measured as a function of $O_2$ content in $SF_6/O_2$ mixtures.	Spark discharges Cylindrical	0.133	178
$SF_6 + CO_2$	The presence of $CO_2$ in $SF_6$ and $SF_6-N_2$ mixtures leads to a considerable increase in the formation of ( $SOF_4 + SO_2F_2$ ) and decreased formation of $S_2F_{10}$ .	Corona discharges Point-plane	0.4	182

biggest drawback, which also limits its use in equipment. The solid iodine and carbon deposited on the surface of electrodes, which is detrimental to the gas self-healing, so the gas is not suitable for use in the interrupter medium. Its genotoxicity is also a major obstacle.

- (5)  $C_3F_7N$  has a great insulating property so that the mixed gas (at a very low mixing ratio) can still reach the insulation level of SF<sub>6</sub> for high voltage equipment. Mixed gases are currently used in GIS and circuit breakers. It is noteworthy that the decomposition of gas discharge may produce some of the toxic gases or solid products, which requires further study and verification.
- (6) Perfluoroketones, mainly  $C_5F_{10}O$ , have similar insulation properties of  $C_4F_7N$ . However, using mixed gas is more suitable for medium voltage equipment due to the higher boiling point than  $C_4F_7N$  and the too high value of GWP of  $C_4F_7N$ . Air is used in the mixture to prevent oxidation inside the equipment. The decomposition product after

discharge will not affect the environment and the human body.

(7) For other gases such as HFO1234zeE, some research studies have not reached a mature conclusion, so their potential for substitution remains to be further studied.

#### **B. Outlook**

Through the above research on the current alternative gases, the research on alternative gases mainly focuses on the insulation performance, the arc-extinguishing performance, and the decomposition characteristics.

(1) For insulation properties, the experimental parameters measured are relatively easy to obtain. The gas insulation can be evaluated by comparing with the SF<sub>6</sub>. Setting different electrodes, voltage type, pressure, and mixing ratio has become common experimental means to comprehensively investigate the insulation properties of gas. The inception voltage

## TABLE VII. Main properties of PFCs and mixtures.

Gases	Breakdown voltage/partial discharge voltage comment	Voltage/el	ectrode	P (MPa)	Reference
${c-C_4F_8/1-C_3F_6/} \\ C_3F_8/C_2F_6 + N_2/ \\ CO_2/CF_4$	Positive synergism was observed for gas mixtures with a PFC gas $(c-C_4F_8, 1-C_3F_6, C_3F_8 \text{ and } C_2F_6)$ and any of $N_2$ , CO <sub>2</sub> , and CF <sub>4</sub> gases.	AC Rod	-plane	0.10-0.40	180
c-C <sub>4</sub> F <sub>8</sub> /N <sub>2</sub>	The sparkover voltage of $c-C_4F_8$ is about 1.3 times and 1.3–1.4 times higher than SF <sub>6</sub> .	AC/ Parallel-	LI plane	0.1	186
c-C <sub>4</sub> F <sub>8</sub> /N <sub>2</sub>	The breakdown voltage of the c-C <sub>4</sub> F <sub>8</sub> /N <sub>2</sub> mixture under negative lighting impulse is lower than that under positive polarity. c-C <sub>4</sub> F <sub>8</sub> /N <sub>2</sub> mixtures show the synergistic effect.	LI Sphere-plane		0.1-0.3	188
$\frac{C_{3}F_{8}/N_{2}}{C_{2}F_{6}/N_{2}}$	In $C_3F_8/N_2$ and $C_2F_6/N_2$ mixtures, the synergistic effect in electrical insulation performance was less remarkable than that in SF <sub>6</sub> /N <sub>2</sub> mixtures.	AC Needl	e-plane	0.1	190
C <sub>3</sub> F <sub>8</sub> + air	Both breakdown ratios of $C_3F_8$ to air approach a minimum of about 2.5 at the sphere-plane electrode system.	LI Sphere point-p	-plane/ blane	0.1	189
	$CF_4$ has a smaller critical field strength and is less attaching than $SF_6$ .	AC/DO Point-p	C/LI blane	0.1-0.4	199
CF <sub>4</sub>	At large values, $pL > 2$ Torr cm, the rate of growth of breakdown voltage with pressure essentially increases due to the attachment.	DC Plane-plane		0.1–10 Torr	202
Gases	Electron swarm parameters	Method	E/N (Td) T (K)	P (MPa)	Reference
c-C <sub>4</sub> F <sub>8</sub>	A critical field strength E/Ncr of $c-C_4F_8$ is 439.5 td.	Measure (PT) 12 Td-43 Td	330–600 292–300 K	1–7.5Torr	191
$c-C_4F_8 + SF_6$	The limiting E/N of 20% SF <sub>6</sub> and 80% c- $C_4F_8$ can exceed the values for the respective pure gases.	Calculation	282–707 273 K	1 Torr	192
$c-C_4F_8 + CO_2$	The limiting field $(E/N)_{lim}$ of the c-C <sub>4</sub> F <sub>8</sub> /CO <sub>2</sub> mixtures is higher than that of SF <sub>6</sub> /CO <sub>2</sub> mixtures.	Calculation (Monte Carlo)	150–450 293 K	1 Torr	194
$c-C_4F_8 + N_2$ $c-C_4F_8 + air$	c-C <sub>4</sub> F <sub>8</sub> -N <sub>2</sub> and c-C <sub>4</sub> F <sub>8</sub> -air have very similar (E/N)cr values, higher than those of the other three mixtures (CO <sub>2</sub> , CF <sub>4</sub> , and O <sub>2</sub> ), and superior even to that of pure SF <sub>6</sub> for c-C <sub>4</sub> F <sub>8</sub> concentrations above 80%.	Calculation	150–600 300 K	0.1	195
$C_3F_8 + N_2$	The limiting field strength of the $C_3F_8-N_2$ gas mixture is greater than that of $C_3F_8-CO_2$ .	Calculation	1-1000 293	1 Torr	196
CF <sub>4</sub>	Replacing $SF_6$ with $CF_4$ brings an increase in the critical reduced electric field strength for temperatures above 2200 K.	Calculation	0–1000 T 300–35 0	0.01-1.6	201
$C_{3}F_{8} + N_{2}$	The $C_3F_8-N_2$ mixture has the largest (E/N)cr of the cold $C_3F_8-CF_4$ , $C_3F_8-CO_2$ , $C_3F_8-N_2$ , $C_3F_8-O_2$ , and $C_3F_8-Ar$ . The hot $C_3F_8$ gas gas has much poorer dielectric performance than hot SF6 because the (E/N)cr of C3F8 decreases significantly above room temperature.	Calculation	100–600 300–3 500	0.01-1.6	200
Gases	Decomposition	Meth	od	P (MPa)	Reference
$c-C_4F_8 \\ c-C_4F_8 + N_2$	The main identified gaseous by-products are $CF_4$ , $C_2F_6$ , $C_2F_4$ , $C_3F_8$ , and $C_3F_6$ for pure c- $C_4F_8$ gas and $CF_4$ , $C_2F_6$ , $C_2F_4$ , $C_3F_8$ , $C_3F_6$ , and $C_2F_3N$ for the c- $C_4F_8/N_2$ mixture.	50 Hz ac coror Needle-	na discharge plane	0.1	205

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Gases	Breakdown voltage/partial discharge voltages	Voltage/	electrode	P (MPa)	Reference
	The optimum ratio of $CF_3I/N_2$ to replace $SF_6$ gas as an insulating medium is 30% at 0.3 MPa based on the breakdown characteristics.	AC Nee	dle-plate	0.1-0.3	208
	Under 0.1 MPa and 0.2 MPa, $30\% CF_3 I/70\% N_2$ present similar insulation properties as $20\% SF_6/80\% N_2$ .	DC Plane	/LI -plane	0.1 and 0.2	213
CF <sub>3</sub> I/N <sub>2</sub>	Based on the 50% breakdown voltage of the $\rm CF_3I/N_2$ gas mixture. The $\rm CF_3I$ mixture has the potential to replace $\rm SF_6$ as an insulation medium in GIL.	LI Co cyline	oaxial drical	0.1-0.3	215
	The breakdown voltage of $CF_3I-N_2$ mixtures increases proportionally to the $CF_3I$ content without the synergistic effect. The breakdown voltages for mixtures of 50% $CF_3I$ are lower than with $SF_6$ at the same ratio.	DC Sphe	re-sphere	0.1-0.6	230
	When the RMU vacuum circuit breakers are insulated with a $30\%$ :70% CF <sub>3</sub> I-CO <sub>2</sub> mixture, no disruptive discharges are detected for a 50 impulse test series.	LI R	MU	0.14	214
CF <sub>3</sub> I/CO <sub>2</sub>	The 50% breakdown strength of the 30:70% mixture of $CF_3I-CO_2$ for the coaxial electrode was more than two times higher than air.	LI Rod plane- coaxial c	-plane/ plane/ ylindrical	0.1	212
	The PDIV+ of CF <sub>3</sub> I/CO <sub>2</sub> is higher than that of SF <sub>6</sub> /CO <sub>2</sub> when k is10%–40%. The PDIV+ of CF <sub>3</sub> I/CO <sub>2</sub> (30%–40%) at 0.15–0.2 MPa can reach the level of pure SF <sub>6</sub> at 0.1 MPa.	AC Nee	AC Needle-plate		209
CF <sub>3</sub> I-air	Pure $CF_3I$ gas has 1.2 times as high a dielectric strength as pure $SF_6$ gas at the same pressure. V-t characteristics in $CF_3I$ -air gas mixtures are almost the same as those in $CF_3I$ -N <sub>2</sub> gas mixtures.	Steep-front square voltage Rod-plane		0.1	210
$C_4F_7N + CO_2$	The content of $C_4F_7N$ was 18%–20%, and it can achieve the same dielectric strength as pure $SF_6$ .	AC inhomogeneous field		0.15, 0.19	47
$C_4F_7N + CO_2$	The synergistic effect of $C_4F_7N$ and $CO_2$ at a concentration of 3.7% is obvious, and the mixture in uniform electric field reaches 72% of of the dielectric breakdown of pure SF <sub>6</sub> at 0.55 MPa.	AC/LI plane-plane/ sphere-plane/rod-plane/ sphere-sphere		0.1-1.04	240
$C_4F_7N + CO_2/n_2/air$	The dielectric breakdown strength of the gas mixture increases at higher pressures and increased $C_4F_7N$ concentration.	Disk ele	ectrodes	0.1–1.1	241
$C_5F_{10}O + air$	The air PFK-max mixture at 0.7 MPa has about 95% the withstand capability of 0.45 MPa $SF_6$ .	Step Sphere	-DC -sphere	0.1-0.7	250
$\frac{C_5F_{10}O + air}{C_6F_{12}O + air}$	The breakdown performance of the investigated mixtures can approach $SF_6$ values at increased total filling pressures.	AC Sphere	/LI -sphere	0.1-0.7	249
Gases	Electron swarm parameters	Method	E/N (Td) T (K)	P (MPa)	Reference
$CF_3I + N_2$	The limiting field strength of $CF_3I$ was found to be $E/Nlim = 437$ td. For the $CF_3I-N_2$ mixture with 70% $CF_3I$ , this $E/Nlim$ value was found to be essentially the same as that for pure $SF_6$ .	Measure PT	100-850 293-301	0.4–20 Torr	218
$CF_{3}I + CF_{4}/$ $N_{2}/CO_{2}/$ $O_{2}/air$	The (E/N)cr of CF <sub>3</sub> I–N <sub>2</sub> and CF <sub>3</sub> I–air are obviously higher than that of SF <sub>6</sub> –N <sub>2</sub> at the CF <sub>3</sub> I ratio higher than 65% and even higher than 65% and even higher than that of pure SF <sub>6</sub> at the CF <sub>3</sub> I ratio ratio higher than 70%.	Calculation	0-600 300		221
$\overline{\begin{array}{c} CF_{3}I+N_{2}/\\ CO_{2} \end{array}}$	For the mixtures with 70% $CF_3I$ , the values of (E/N)lim are essentially the same as that for pure $SF_6$ .	Calculation SST	100–1000 293	1 Torr	222

### TABLE VIII. Main properties of C<sub>n</sub>F<sub>m</sub>X mixtures. DBD-Dielectric Barrier Discharge. 1 Torr = 133.3Pa.

### TABLE VIII. (Continued.)

	Breakdown voltage/partial discharge voltages	Voltage/e	electrode		
Gases	Electron swarm parameters	Method	E/N (Td) T (K)	P (MPa)	Reference
CF <sub>3</sub> I + Ar/ Xe/He/ N <sub>2</sub> /CO <sub>2</sub>	Among the mixtures of $CF_3I$ with Ar, Xe, He, N <sub>2</sub> , and $CO_2$ , $CF_3I-N_2$ present the greatest insulation strength with 20%–90% $CF_3I$ content based on (E/N)cr.	Calculation SST	100–800 293	1 Torr	223
Gases	Interruption ability	Met	hod	P (MPa)	Reference
	The interruption performance of the $CF_3I/CO_2$ mixture approximated that of pure $CF_3I$ when the ratio of $CF_3I$ exceeds 20%.	Measure/SLF frequency of oscillating current: 45 kHz.		0.2 (upstream)	218
$CF_3I + CO_2$	In BTF interruption, $CF_3I-CO_2$ is superior to $CF_3I-N_2$ and the performance approximates to that of pure CF3I when the proportion of CF3I exceeds 30%.	Measure/BTF Peak of current: 3.0 kA.		0.2	225
	The arc conductance attenuated more rapidly for $CO_2$ mixed with $CF_3I$ at concentrations above 0.9 than for pure $CO_2$ .	Calculatio 300–30	on in LTE 000 K	0.1,0.5	228
$CF_3I + CO_2/$ N <sub>2</sub> /air	$CF_3I$ has a rather small low-temperature electrical conductivity (lower than $SF_6$ ).	Calculation LTE 300–50 000 K		0.1-3.2	229
$C_4F_7N + CO_2$	The arcing time is stable over the 100 operations, and the average arcing time is about 12 ms compared to a typical value of 15 ms for $SF_6$ .	Measure 420 kV disconnector		0.55	243
$C_5F_{10}O + air$	The pressure peak is up to 30% higher compared to $SF_6$ and the peak for $SF_6$ is reached in 17 ms, while for the mixture, it is reached in 10–12 ms.	Measure GIS short circuit current: 40 kA			253
C <sub>5</sub> F <sub>10</sub> O	The arc interruption performance and electric strength of C5-PFK are comparable to or better than those of $SF_6$ .	Calculation 300–30 000 K		0.1-1.0	254
Gases	Decomposition characteristics	Met	hod	P (MPa)	
CF <sub>3</sub> I	The partial discharge by-products of $C_2F_6$ , $C_2F_4$ , $C_2F_5I$ , $C_3F_8$ , CHF <sub>3</sub> , $C_3F_6$ , and CH <sub>3</sub> I were obtained.	Exper AC/need	iment le-plane	0.1	237
	CF <sub>3</sub> I gas generates iodine after current interruption.	Exper SLF an	iment d BTF	0.2	231
CF <sub>3</sub> I-CO <sub>2</sub>	Iodine is deposited on the electrode along with carbon and oxygen after $CF_3I-CO_2$ gas mixtures discharge under a positive impulse polarity.	Experi LI/rod	iment -plane	0.1-0.2	234
$CF_3I + H_2O$	The decomposition products of $CF_3I$ include the main components $C_2F_6$ , $I_2$ and other components such as $C_2F_4$ , $C_2F_5I$ , $C_3F_8$ , HF, $H_2$ , $COF_2$ , $CF_3H$ , and $CF_3OH$ with a small amount of $H_2O$ .	Calculation			238
$CF_3I + O_2$	The breakdown decomposition rate of $CF_3I$ is accelerated, and $COF_2$ is produced in the presence of $O_2$ .	Measure and Calculation/AC			239
$C_4F_7N + air$ $C_4F_7N + CO_2$	CO and $C_4F_8$ are produced after the breaking test, and the LC50 (4 h on mice) after 100 C–O 630 A/24 kV in an 11-l epoxy bulb is inferior to 225 ppm.	Mea 100 C-O 63	sure 30 A/24 kV	0.174	246
$C_4F_7N + air$	The sole atmospheric degradation products appear to be NO, $COF_2$ , and $CF_3C(O)F$ .	Measu calculation	re and /296 ± 1 K	700 Torr	247

#### TABLE VIII. (Continued.)

	Breakdown voltage/partial discharge voltages	Voltage/electrode		
Gases	Interruption ability	Method	P (MPa)	Reference
$C_4F_7N + CO_2$	The decomposition rate of $C_3F_7CN$ and generation rate of the products increase with the increase in ambient temperature.	Calculation (2 000–3 000 K)	0.1	249
$C_4F_7N + Cu/Al$	The adsorption energy of $C_3F_7CN$ adsorbed on Cu (1 1 1) and Al (1 1 1) is both below 0.8 eV.	Calculation		253
$\frac{C_5F_{10}O + Air}{N_2/CO_2}$	After the DBD processing of two test-gases, a large quantity of of toxic $C_3F_6$ was found in pure $C_5F_{10}O$ .	Measure DBD	0.1	258
$C_5F_{10}O + Air$	CO and $C_4F_8$ are produced after the breaking test, and the LC50 (4 h on mice) after 100 C–O 630 630 A/24 kV in an 11-l epoxy bulb is inferior to 2100 ppm.	Measure 100 C–O 630 A/24 kV	0.125	246
$C_5F_{10}O + N_2$	The amount of decomposition products of $C_5F_{10}O$ (CF <sub>4</sub> , $C_2F_6$ , $C_3F_6$ , $C_3F_8$ , $C_4F_{10}$ , and $C_6F_{14}$ ), increased with the increased number of discharges.	Measure calculation AC/sphere-sphere	0.11	262
$C_6F_{12}O + N_2$	The breakdown decomposition products of $C_6F_{12}O + N_2$ were $CF_4$ , $C_2F_6$ , $C_3F_6$ , $C_3F_8$ , $C_4F_{10}$ , and $C_5F_{12}$ .	Measure calculation AC/sphere-sphere	0.1	79

measurement for partial discharge is already there. The calculation of the discharge process of PD or discharge by Townsend discharge theory has also been verified.

The measurement process is generally at a low pressure (1 Torr), and there is a deviation of the calculation results of the gas insulation performance under high pressure. The calculated value is derived from the collision cross section of gas molecules. According to the collision cross section data, the corresponding parameters can be calculated, and the critical breakdown field strength can be obtained from the ionization coefficient and the attachment coefficient. At present, the data of the collision cross section of gas are limited and are constantly replenished and improved. It is difficult to measure and calculate the collision cross section, which is an important factor that hinders the new gas research. The current micro-parameter calculations are based on some assumptions and simplifications. There is still a need for further research and exploration on the microscopic calculation of gas discharge to macroscopic processes.

(2) Extinguishing performance: Gas arc performance can be obtained through the interruption test in the laboratory or the circuit breaker equipment. By optimizing the structure of the arc extinguishing device, changing the size of the contact, and adding solid insulating materials, the gas arc extinguishing effect can be improved. According to the data in Table IV, it can be seen that there is not any arc-extinguishing medium, so looking for gases with excellent interruption performance is still the difficult point at present.

For the calculation of the arc extinguishing performance, the calculation of the arc conductance through thermodynamic performance, transport parameters, and radiation has been a relatively mature process. However, the calculation process is still in an ideal condition. The details of the calculation are still worth exploring. For example, the difference between the electron temperature and the heavy ion temperature in Non Local Thermodynamic Equilibrium (NLTE) and various particles of the chemical reaction rate in nonchemical equilibrium need to be considered in the calculation of equilibrium composition. In-depth exploration of the theory is still a research direction.

(3) For decomposition performance: The decomposition products of various gases in different discharge conditions can be measured by the experiment. However, for the theoretical calculation, at present, only the possible reaction path can be specified to calculate the reaction energy, and the calculations have some subjectivity. The chemical process is not calculated on the physical conditions, so the rate of formation of decomposition products cannot be calculated quantitatively according to the experimental conditions of discharge.

So far,  $SF_6$  is still the most stable and reliable insulation and interrupter medium. The existing alternative gas as an insulation medium can be used in different equipment. In contrast, the interrupter medium needs further exploration. The new gas is still in constant exploration.

The gas-insulated medium is not a single kind of gas. Different gases may be used as the insulating medium in different equipment. Because there is no single insulating gas that can completely replace the use of  $SF_6$  in electrical equipment, mixed gases have become the mainstream of development. There are still many unexplored areas for the gas currently studied. The molecular structure of the ideal alternative gas by computational chemistry can be designed. The calculation can predict the boiling point, insulation properties, and other aspects of the new insulating gas, which is a way to look for alternative gases.

At present, there is still no gas that can substitute for  $SF_6$  completely, and the gas with good insulating property needs to be mixed with the buffer gas (except HFO1234zeE and CF<sub>3</sub>I, which can be used alone as an insulating medium in the MV equipment). A series of problems (such as the leakage, the supplementary gas, the recovery, and the decomposition) brought by the use of the mixed gas need to be considered in the future, and the relevant industries are still being explored. Research on alternative gases still faces greater challenges and requires significant time and economic investment.

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#### APPENDIX

Tables V–VIII contain the current research conclusions and references of major environmentally friendly gases.

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