Research and Analysis of Insulating Gas in Unified Test Conditions

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ABSTRACT: In order to study the insulation mechanism of SF₆ substitute gas, it is suggested to calculate the dielectric strength of insulating gas from the molecular structure. The dielectric strength of a typical gas is modeled by a power frequency breakdown test under a uniform electric field. The molecular parameters of insulating gases are calculated by the density functional theory method, and the effect of molecular structure parameters on the breakdown voltage of power frequency is studied. Based on the molecular structure parameters, which are closely related to the breakdown voltage of power frequency, a model of pressure and distance variation of AC breakdown voltage of insulating gas is established. The breakdown voltages of insulating gases (CF₃SO₂F) are also derived from the proposed model. The calculated breakdown voltage of power frequency of two gases is compared with the experimental value. The average error is just 2.6%. This model provides a basis for the future search for potential alternative insulating gases.

1. INTRODUCTION

Sulfur hexafluoride (SF₆) gas has been widely used in power equipment such as gas-insulated switchgear and gas-insulated transmission lines. It is an excellent insulator, with good arc extinguishing ability, stable chemical properties, nontoxicity, and noncorrosivenes.¹ However, due to the strong greenhouse effect of SF₆, it is necessary to study alternative gases for SF₆ because it is severely restricted under the “Paris Agreement”.²,³

Typical alternative gases such as c-C₄F₈, C₂F₃I, C₄F₇N, C₅F₁₀O, and C₆F₁₂O₄ and their mixed gases⁵ all have the disadvantage of high liquefaction temperature. It is necessary to have a new method that would form the theoretical basis for experimental screening of alternative gases with better performance.⁶

Compared with SF₆ gas, an alternative gas needs to meet basic requirements such as having a breakdown voltage higher than that of SF₆ gas, a liquefaction temperature lower than that of SF₆ gas, and GWP (global warming potential) lower than 5% of SF₆ gas under the same conditions. AC breakdown voltage has the advantages of being intuitive and reliable and is an important indicator of the performance of gas insulation in the field of high voltage. Due to the variety of gases available, it is difficult to carry out exhaustive tests. Therefore, it would be useful to establish a simulation model of power frequency breakdown voltage based on the characteristic parameters of gas molecules, which can theoretically predict the power frequency breakdown voltage of insulating gases.

Meurice’s team⁷ used density functional theory (DFT) to obtain a gaseous medium, the integrator optical spectra (IOA), in conjunction with electron energy loss spectroscopy. They analyzed the relationship between the dielectric strength of the insulating gas IOA. Olivet’s team⁸ used the Austin model and parametric model to study the ionization process and adsorption process of nine insulating gases occurring in different energy bands and studied the electron energy affinity and ionization power of the gas medium’s quantitative relationship with dielectric strength. Franck of ETH Zurich⁹–¹¹ used a new procedure to systematically identify and quantify novel molecular gases with low global warming potential for application in high-voltage insulation. However, the prediction error of insulation strength of polar molecules was large. Rong et al.¹² used neural networks and random forests to predict the electrical strength and boiling temperature of the substitutes for greenhouse gases. However, they were unable to describe the breakdown voltage of environmentally friendly gas with the change of gas pressure and spacing. Based on the analysis of carrier collision process, Chen¹³ selected five insulating gases with high replacement potential from 137 alternative gases. The effect of gas pressure on insulation strength was not considered.

Wang¹⁴ used the molecular characteristics of the insulating gas as parameters based on density functional theory and regressed to obtain a structure–activity relationship model to

Received: October 14, 2021
Accepted: February 25, 2022
Published: March 8, 2022

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determine the insulation strength of SF$_6$ gas. Since the AC breakdown voltage of different insulating gases has a saturation point which increases with pressure, the breakdown voltage does not increase linearly with the increase of pressure. So the relative SF$_6$ insulation strength is used to characterize the insulating ability of the gas, and AC breakdown voltage needs to be limited so that the saturation state is not reached when subject to change in a smaller gas pressure range.

This paper will carry out the power frequency discharge test of a typical insulating gas under a uniform electric field, which will be combined with the characteristics of the AC breakdown voltage of different insulating gases. Then a power frequency breakdown test research on insulating gas, the test results were not in good agreement due to inconsistent test conditions.

Breakdown voltage is the main parameter to characterize the performance of gas insulation, but it is affected by various factors during experimental measurement, including electric field uniformity ($f$), gas pressure ($p$, Pa), electrode spacing ($d$, mm), etc. (the product of the $p$ and $d$).

In relevant national standards and IEC standards, the high voltage test conditions were specified, and the unified test conditions were determined (the unified test conditions are a temperature of 293 K, a 60% relative humidity (RH), a ventilated and cool place with avoidance of direct sunlight).

Therefore, this paper will determine the AC breakdown voltage of the insulating gas through the AC breakdown test under a uniform electric field. This will help provide data for the establishment of the calculation model on AC breakdown voltage based on characteristic molecular parameters.

### 2. TEST SETUP AND METHOD

Although many research teams in the world have conducted power frequency breakdown test research on insulating gas, the test results were not in good agreement due to inconsistent test conditions.

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#### 2.1. Test Setup.

The schematic diagram of the test setup is shown in Figure 1. The finite element calculation software COMSOL was used to simulate and analyze the experimental cavity and the test electrode. The test electrode was a plate electrode with a diameter of 15 mm, and the electrode spacing was less than 10 mm. The electric field distribution between the electrodes was uniform. The analysis results show that test electrode and experimental cavity could be used for the power frequency breakdown test under a uniform electric field.

In Figure 1, the resistance of the protective resistor was 50 kΩ. The highest output voltage of the transformer was 250 kV.

#### 2.2. Experiment Procedure.

According to the power frequency breakdown test requirements specified in GB/T 16927.1-2011 and IEC60060, the test steps are as follows.

1. After setting the electrode, close the cavity and evacuate the test cavity and the gas-filled pipe until the air pressure drops below 3 Pa.
2. Carry out the power frequency breakdown test 10–20 times and “burn-in” the electrode.

![Figure 1. Schematic diagram of the test setup.](https://example.com/figure1.png)

![Figure 2. Electric field distribution under the plate electrode and schematic diagram of the test electrode.](https://example.com/figure2.png)

![Figure 3. Illustration of the test chamber.](https://example.com/figure3.png)
(3) In the case of a laboratory temperature of 20 °C, fill the chamber with the gas to be tested, leave it to settle for 15 min, pump to 3 Pa with a vacuum pump, repeat three times, perform “washing”, and finally charge the gas to the test pressure. Let it stand for 15 min.

(4) Increase the voltage to the level for the gas to breakdown according to the power frequency discharge voltage test procedure specified in GB/T 16927.1-2011, and record the test breakdown voltage value.

(5) After completing the test of the pressure to be measured, elevate the test chamber to the next pressure and measure with a vacuum pump; repeat the above steps until the test is completed.

2.3. Test Result. With the above procedure, the power frequency breakdown test of SF6 gas was carried out under unified test conditions (temperature of 20 °C, 20–30% RH, plate electrodes, electrode spacing of 5 mm, and gas pressure of 0.1–0.5 MPa). The experimental value was compared for SF6 gas on the AC breakdown voltage with the reference value.\textsuperscript{24–28} The comparison result is shown is Figure 5.

\[ \epsilon\% = \left| \frac{E_{v,\text{ref}} - E_{v,\text{test}}}{E_{v,\text{test}}} \right| \times 100\% \]  \hspace{1cm} (1)

It can be seen from Figure 4, that the error between the effective value of the AC breakdown voltage of SF6 gas and the values reported in refs\textsuperscript{25} and 26 under uniform test conditions was 3%. The test shows that the platform was reliable and can be used to perform power frequency breakdown of typical gases. The AC discharge voltage of nine typical gases under uniform electrical field test conditions was measured using the platform above. The relationship between \( pd \) (\( p \) is absolute pressure) and AC breakdown voltage was determined by adjusting the gas pressure and electrode spacing. First, the electrode spacing was fixed at 2.5 mm to obtain the AC breakdown voltage under different gas pressures. Then the electrode spacing was changed to 5 and 7.5 mm, and the above steps were repeated. Finally, the AC breakdown voltage corresponding to the \( P_d \) was determined, as shown in Figures 6 and 7.

![Figure 4. Comparison between SF6 gas test values and literature values.](image1)

![Figure 5. AC breakdown voltages of four typical gases under high saturated vapor pressure.](image2)

To verify against the results of the reference, the referenced value and the test value were put into formula 1 to obtain the error between the two.

![Figure 6. AC breakdown voltages of five typical gases (low saturated vapor pressure).](image3)

In Figures 5 and 6, the gases were studied separately. The gas in Figure 5 was a conventional gas. The saturated vapor pressure of the conventional gas was relatively high. The high-pressure test was performed under a fixed discharge gap (2.5/5/7.5 mm),

![Figure 7. Correlation degree of influencing factors of gas molecular structure on insulation strength.](image4)
and the pd value was 0.25–5.25 mm MPa. The gas in Figure 6 had high insulation strength and is currently the gas with the most potential to replace SF₆, but its saturated vapor pressure is small (for example, the saturated vapor pressure of C₅F₁₀O at 293.15K is 0.1 MPa). It was not possible to obtain a higher pd value in 7.5 mm electrode spacing (0.1 MPa × 7.5 mm = 0.75 mm MPa).

Through the AC breakdown test of the typical gases under uniform test conditions, the breakdown voltage of the typical gases was obtained. The calculation model of the insulating gases was established based on characteristic molecular parameters.

3. CALCULATION OF AC BREAKDOWN VOLTAGE OF INSULATING GAS

Combining the power frequency breakdown test results under unified test conditions, and based on ref 14, research on the molecular structure parameters of insulating gases was carried out.

3.1. Molecular Feature Parameter Selection. The molecular structure parameters in refs 28–31 were used as a data set. These articles mainly analyzed the following structural parameters: molecular surface electrostatic potential PA, molecular volume, highest occupied orbital energy $E_{\text{HOMO}}$, lowest non-occupied orbital energy $E_{\text{LUMO}}$, polarizability $\alpha$, dipole moment $\mu$, molecular surface area $A_s$, and molecular surface average statistical deviation $\sigma_{\text{surf}}$. Average deviation of the electrostatic potential on the molecular surface is $\pi$, with molecular energy $E_v$.32

Relevance analysis was a new factor analysis method in gray theory, which analyzes the degree of correlation between multiple factors by comparing the geometric relationship of the system statistics series. Gray relational analysis (GRA) is a measure of the correlation of two or more factors. The degree of correlation indicates the degree of mutual restraint and influence between the various factors that affect the development of something.14 Through GRA analysis, the correlation degree $\gamma$ of the influencing factors of the gas molecular structure parameters affecting the insulation strength were sorted and selected.

(1) For the gas molecular parameter data sample, establish the gray incidence matrix.

$$X = \{x_0, x_1, ..., x_n\}$$

In the formula, $x_0$ represents the reference sequence, $x_i$ represents the comparison sequence, $i = 1, 2, 3...n$.

(2) Initial sequence dimensionless processing.

$$\bar{x}_i = \frac{1}{n} \sum_{k=1}^{n} x_i(k)$$

(3)

$$x'_i(k) = \frac{x_i(k)}{\bar{x}_i}$$

In the formula, $\bar{x}_i$ represents the average value of the $k$-th point $x_i(k)$ in each factor; $x'_i(k)$ represents the dimensionless value of $x_i(k)$ after initialization.

(3) Find the difference sequence (absolute difference of each point).

$$\Delta_0(k) = |x'_0(k) - x'_i(k)|$$

(5)

$$\Delta_0(k) = (\Delta_0(1), \Delta_0(2), ..., \Delta_0(n))$$

(6)

In the formula, $\Delta_0(k)$ is the sum of absolute differences of points.

(4) Calculate the weight of the comparison sequence relative to the reference sequence.

$$\varphi_0(k) = \frac{\min_{i=1}^{n} \Delta_0(k) + \rho \max_{i=1}^{n} \Delta_0(k)}{\Delta_0(k) \rho \max_{i=1}^{n} \Delta_0(k)}$$

(7)

$$\gamma_i = \frac{1}{n} \sum_{k=1}^{n} \varphi_0(k)$$

(8)

$$\gamma = \frac{\gamma_i}{\sum_{k=1}^{n} \gamma_i}$$

(9)

In the formula, $\min_{i=1}^{n} \Delta_0$ is the minimum value of the two poles, $\max_{i=1}^{n} \Delta_0$ is the maximum value of the two poles, and $\rho$ is the resolution coefficient; usually it is taken as 0.5. It is to prevent the absolute error value from being too large and the distortion to the difference between the correlation coefficients.15 The value of $\gamma_0$ is the degree of gray correlation, and $\gamma_i$ is the weight of each factor.16

Through the above-mentioned GRA method, the correlation degree $\gamma_0$ of the influence factors of the gas molecular structure parameters affecting the insulation strength, boiling point temperature, and GWP was calculated, and they were sorted as shown in Figure 8.

Figure 8. Procedure for the selection of molecular characteristic parameters and establishment of calculation model of power frequency breakdown voltage.

The four strongly correlated molecular characteristic parameters used to characterize the gas on AC breakdown voltage were obtained through the above correlation analysis. $A_s$ is the total surface area of gas molecules; $\sigma_{\text{surf}}$ is the balance of positive and negative electrostatic potential and electrostatic potential of the product of the standard deviation; $\Pi$ is the statistical average deviation of the electrostatic potential on the molecular surface, and $E_v$ is the total energy of the molecule.

The relationship between the selection of molecular characteristic parameters and the calculation model of AC breakdown voltage is shown in Figure 8.
3.2. Model Building. Ten kinds of gas constituent elements were input into the molecular structure calculation software Gaussian 09W, and the visualization wave function software was Multiwfn. The molecular structure and molecular orbital data were calculated, and the molecular characteristic parameters are shown in Table 1.

Based on the molecular structure parameters and breakdown voltage data in Table 1, the structure–activity relationship model was obtained using a multivariate nonlinear fitting method, which is given in eq 10 below.

\[
\sigma_{b} = \left( A_{s} + B_{s}v_{\sigma_{tot}} + C + D_{s}pd \right) (0.5 < pd < 1.5) \tag{10}
\]

\[U_{b} = \begin{cases} 
U_{A} = AA_{s} + Bv_{\sigma_{tot}} + CII & (p = 0.5) \\
U_{I} = AA_{s} + Bv_{\sigma_{tot}} + CII + DE_{v}e^{pd} & (0.5 < pd < 1.5)
\end{cases}
\]

\[U_{b} = \text{AC breakdown voltage of an insulating gas}; A_{s} \text{ is the total surface area of gas molecules}; v_{\sigma_{tot}} \text{ is the balance of positive and negative electrostatic potential and electrostatic potential of the product of the standard deviation}; II \text{ is the statistical average deviation of the electrostatic potential on the molecular surface}; E_{v} \text{ is the total energy of the molecule}; p \text{ is the gas pressure of the insulating gas}; d \text{ is the electrode spacing}; A \text{ is the accelerated ionization capacity of the unit molecular surface area}; B \text{ is the electron capacity (adsorption coefficient) captured by the molecule due to the imbalance of the electrostatic potential}; C \text{ is the degree of symmetry due to the molecular structure, or the degree of difficulty of being ionized (ionization coefficient)}; \text{ and } D \text{ is the coefficient to be determined.}

We selected the same 10 kinds of insulating gases under unified test conditions, and the power frequency discharge voltage under pd of 0.5 mm MPa was calculated and is shown in Table 1.

The calculation flowchart is shown in Figure 9.

We used four gas characteristic molecular parameters as input into the AC breakdown voltage model (eq 10). The undetermined coefficients of the AC breakdown voltage expression was obtained through the nonlinear least-square method. The undetermined coefficients of the power frequency discharge voltage calculation model under the conditions were \( A = 19.9384, B = 0.0557, C = -40.4732, D = 0.0128, \) and \( a = 1.2282. \)

Therefore, the expression of the AC breakdown voltage calculation model becomes

\[
U_{b} = 19.9384A_{s} + 0.0557v_{\sigma_{tot}} - 40.4732II (pd = 0.5)
\]

\[
U_{b} = 19.9384A_{s} + 0.0557v_{\sigma_{tot}} - 40.4732II + 0.0128E_{v}e^{2.282pd} (0.5 < pd < 1.5) \tag{11}
\]

At pd = 0.5 mm MPa, it was found through curve fitting that the main factor affecting the AC breakdown voltage of the gas was the molecular structure of the gas itself. At 0.5 < pd < 1.5 mm MPa, the AC breakdown voltage of the gas was affected by the mean free stroke of the molecule and the molecular structure. In this case, the AC breakdown voltage of the gas exhibited an exponential increase with the increase of pd. At pd > 1.5 mm MPa, the AC breakdown voltage of gas was affected by the roughness of the electrode and other factors. The AC breakdown voltage of gas increases with the increase of pd, but it no longer showed an exponential trend. Formula 11 proposed in this paper is no longer applicable.

3.3. Reliability Analysis. Figure 10 shows the comparison between the calculated and tested values of 10 insulating gases on AC breakdown voltage (N\(_{2}\), SF\(_{6}\), N\(_{2}\)O, C\(_{5}\)F\(_{10}\)O, c-C\(_{4}\)F\(_{8}\), C\(_{4}\)F\(_{7}\)N, C\(_{3}\)F\(_{8}\), CO\(_{2}\), SO\(_{2}\)F\(_{2}\), CF\(_{4}\)). It can be observed from Figure...
weight gases (N₂, CO₂, N₂O) present a high-frequency insulating gas under uniform test conditions. Low molecular model can accurately replace SF₆ gas. The liquefaction temperature of CF₃SO₂F gas at 0.1 MPa is only 11.3%.

Two synthetic gases were considered to have the potential to replace SF₆ (CF₃SO₂F). According to reports, the GWP values of these two gases are both less than 20% of SF₆. Therefore, the gas has a stable presence in the atmosphere for less than 40 years, which is much lower than the 3400 years of SF₆ gas. The liquefaction temperature of CF₃SO₂F gas at 0.1 MPa is −22 °C. These characteristics showed that these two gases are environmentally friendly gases.

In order to verify the accuracy of the calculation model, it is necessary to establish a molecular structure model. We used the molecular structure calculation software Gaussian 09W and the molecular visualization software Multiwfn to calculate the molecular characteristic parameters and molecular energy of CF₃SO₂F gas. The calculation results are shown in Table 2.

Table 2. Calculation Parameters of CF₃SO₂F Gas

| gas type     | A_v (nm²) | v_C² (kJ/mol)² | II (eV) | E_v (au) |
|--------------|-----------|----------------|---------|---------|
| CF₃SO₂F     | 1.230     | 216.863        | 0.346   | 983.267 |

Formula 11 was used to calculate the AC breakdown voltage of CF₃SO₂F gas, with the comparison diagram of the test value measured by our team, as shown in Figure 11.

As can be seen from the above figure, the AC breakdown voltage of CF₃SO₂F gas shows a linear increase with the increase of pd. However, the AC breakdown voltage of CF₃SO₂F gas, relative to that of SF₆ gas, shows a downward trend with increasing pd (in an uniform electric field, the power frequency voltage of CF₃SO₂F gas increases at a rate lower than that of SF₆ gas with pressure and gap). The sensitivity of the gas to electric fields is slightly lower than that of SF₆.

The deviation between the calculated value of the CF₃SO₂F gas AC discharge voltage and the test value was only 1.7%. At 0.5–0.7 mm MPa, the calculated value and the test value have a consistent upward trend, although there is a certain error between the calculated value and test value. The dispersion of the power frequency breakdown test and the calculated value is more in line with the increasing trend of the test value. At 0.7–1.5 mm MPa, it can be seen that the growth trend of the calculated value and the test value were relatively slow. The proposed method can predict the AC breakdown voltage of insulating gas more accurately.

5. RESULTS AND DISCUSSION

In this paper, based on DFT and a multiple linear regression method, a calculation model of AC breakdown voltage of insulating gas using molecular characteristic parameters was proposed. The AC breakdown voltage test (condition) on the insulating gas was carried out for the purpose of verification. The calculation results were consistent with the test values. In summary, the following conclusions can be obtained.
The five main influencing parameters of $A_\nu$, $\sigma_\text{ion}$, $\Pi$, $E_\nu$, and $pd$ were obtained, and the multivariate nonlinear model was used to implement the least-squares fitting. The obtained prediction model also conformed to the theoretical analysis result, and the fitting error was 2.6%.

An AC breakdown test platform was built. The AC breakdown test of $\text{CF}_3\text{SO}_2\text{F}$ gas under uniform test conditions was consistent with the calculated results. The error was within 1.5 mm MPa, which is small, but it became excessive after 1.5 mm MPa. Therefore, this method was only suitable for $pd$ values in the range of 0.5–1.5 mm MPa.

If parameters (the interaction force between molecules and their calculation methods) could be substituted into the calculation model for fitting, the accuracy of the calculation model will be further improved. Future research will be carried out to improve the prediction model and to search for new environmentally friendly insulating gas molecules.

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**Notes**

The authors declare no competing financial interest.

**ACKNOWLEDGMENTS**

The work was partially supported by the National Natural Science Foundation of China (U1966211). The authors are sincerely grateful to the mentioned agencies.

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