Dielectric characteristic of dichlorodifluoromethane (R12) gas and mixture with $\text{N}_2/\text{air}$ as an alternative to $\text{SF}_6$ gas

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Abstract: In this study, dichlorodifluoromethane (R12) gas and mixtures with nitrogen ($\text{N}_2$) and air at different pressures and mixing ratios have been investigated and showed good alternative to $\text{SF}_6$ gas in terms of high-voltage application. Mixed gases contain R12 gas, they offer good dielectric properties and possibility to be used in low-temperature environment. Synergistic effects and self-recoverability test have been performed. Dielectric strength under gas-uniform field showed in the order of $\text{SF}_6 > \text{R12} > \text{N}_2 > \text{air}$ in alternating current (AC), direct current (DC) and impulse was examined. Mixture R12/N$_2$ (80/20%) could reach over 0.90–0.95 times to that of $\text{SF}_6$ gas at 50 lbf/in$^2$ at AC and R12/air (70/30%) gives 0.80–0.90 to $\text{SF}_6$ gas. R12 gas presents brilliant self-recoverability. The optimal ratio to switch $\text{SF}_6$ gas is R12/N$_2$ (80/20%) and R12/air (70/30%) is based on the authors’ experimental condition and setup.

1 Introduction

$\text{SF}_6$ gas has the good dielectric strength and arc quenching ability. Therefore, used in the vast area of application such as gas-insulated switch (GIS) gears, breaker and gas-insulated transmission lines (GILs) and so on [1, 2]. $\text{SF}_6$ gas insulated equipment broadly used in power grids. $\text{SF}_6$ gas has been used for a very long time due to its properties like it is odourless, colourless, non-toxic, non-flammable, chemically stable and it has a low boiling point [3]. Table 1 shows the comparison of global warming potential (GWP) and atmospheric lifetime of $\text{SF}_6$ and dichlorodifluoromethane (R12). Due to its high GWP and atmospheric lifetime GIS manufacturers decided to restrict the use of $\text{SF}_6$ in Kyoto summit (1997) [5].

The high GWP of $\text{SF}_6$ leakage in the atmosphere causes severe harmful effects to the environment. In the past, different researchers tried different binary mixtures containing $\text{SF}_6$ [6–12]. Trends of emission of $\text{SF}_6$ from the electrical equipment started from the late-1990s. China started emission of about 70% of $\text{SF}_6$ from its electrical equipment sector [13]. Research on alternatives of $\text{SF}_6$ started from the late 1970s and which shows that dielectric strength of gas mixtures had improved from pure gas [14]. The best dielectric is those that control electrons below its excitation energy and ionisation process is less important than attachment process to prevent insulation from breakdown. In this way, buffer gases like nitrogen, oxygen and carbon dioxide can be advantageous because additive gases cannot attach electrons for all energy levels; e.g. all electronegative gases attachment becomes more difficult at an energy above 2 eV. So component gases are added to remove energy from electron which wants to get free from low energy attachment area. It is supposed that in the binary gas mixtures one gas should attach electron and other should de-energise electron. The role of additive gases is to de-energise electrons which are at a high energy level and bring them to a low energy level where attachment of electrons is easily possible [15].

In [15] dry air, nitrogen and carbon dioxide were examined but analysis shows that these gases lessen withstood voltage and there was a significant increase in the size of equipment. In [12, 16–23] mixtures of $\text{CF}_4/\text{I}/\text{CO}_2$ or $\text{N}_2$ have been studied and investigation show that it has high boiling point and toxic for reproduction. Mixtures of $\text{C}_2\text{F}_5\text{O}$, $\text{C}_2\text{F}_5\text{O}$ with air and $\text{CO}_2$ have been reviewed but research shows that it has high boiling point and high minimum operating temperature that for $\text{SF}_6$ [14, 24]. In [25] HFO1234ZeE has been analysed and it shows that it drops carbon dust on electrode and decreases dielectric strength. In [4, 26] has provided results for the breakdown voltage of R12 and a mixture of R12 and air at a range of pressures, but the authors did not give the detailed analysis. To use an alternative gas to $\text{SF}_6$ which has properties like high dielectric strength, non-toxic substance, non-flammable, chemically stable and economical should be available. Due to this reason, R12 gas in this paper has been analysed. Table 1 displays comparison of R12 gas with $\text{SF}_6$.

R12 possesses some pertinent features, such as (a) less GWP, (b) minimum atmospheric lifetime, but R12 contain chlorine that is ozone depleting. Here, R12 is employed with mixtures of $\text{N}_2$ and air that result in less depletion ozone layer. In this regard, several technical and literature studies report on the elimination of R12 harmful emission. For example, the authors in [27–29] presented R12 decomposition into hydrogen chloride that is collected as a harmless compound by neutralising hydrogen chloride.

In this study, R12 (base gas) with different ratios and pressures with a mixture of air and nitrogen gas is studied. The advantage of additive gases is to de-energise high energy electron and return them to lower energy level because at high energy level attachment of electrons is very difficult.

This paper discussed that the insulation strength of mixtures is greater than the weighted sum of the component gases also known as positive synergism [26]. Detailed analysis is given in this study of the synergistic effect of mixtures with air and nitrogen. Self-recoverability of R12 gas has been performed and shows that R12
gas insulation has been recovered to its original strength after breakdown.

2 Experimental arrangement and procedures

2.1 Experimental arrangement

The experiment circuit is presented in Fig. 1, the method of experiment is based on IEC60270 standard [30]. The circuit contains control desk (HV 9103) which has built-in variable voltage supply and peak voltmeter (HV 9150). The output of supply is 0–230 V and range of peak voltmeter is 100–1000 kV. Test transformer (HV 9105) output is 220 V–100 kV in a single stage, 200 kV in two stages and resistor is connected for protecting test transformer during the breakdown. The breakdown as it occurs, its voltage is measured by the voltmeter through measuring capacitor. Before starting the experiment, voltmeter and measuring devices were all calibrated by applying AC voltage of known value to avoid error and improve accuracy.

Testing vessel (HV 9134) for vacuum and gas is made of steel which has built-in pressure meter measure up to 6 bars. The electrode was made of aluminium and coated with nickel plating. Electrode diameter is 20 mm and the distance between the electrodes is varying from 0 to 30 cm.

2.2 Testing method

Before testing the vessel, the electrodes were cleaned with silk cloth dumped in alcohol to remove moistures and impurities in order to avoid errors in the experiments. The experiments have been performed in a dry place and at room temperature 20–25°C. The vessel was filled with gas and then removed the gas after 20 min to attain maximum vacuum. The duration of each experiment should be minimum 15–20 min to avoid error due to increasing temperature. To mix the gas properly 30–45 min was given [31].

3 Power frequency breakdown voltage testing

3.1 Mixtures of R12 and air

A gas is normally a perfect insulator but due to some free ions and electrons, by applying electric field free electron gain sufficient energy and due to that collision ionisation occur. The number of electrons produced in the path of a single electron travelling one centimetre is called Townsend first ionisation coefficient denoted by $\alpha$.

R12 is an electronegative gas and negative ions are formed by attaching electron to a neutral molecule and detachment occurs when positive ions are formed. Attachment and detachment could occur depending on the applied field. Detachment coefficient is denoted by $\eta$.

$$dN = N(\alpha - \eta)\, dx$$

where $N$ is the initial number of electrons, $dN$ is the electron travelled and $dx$ is the distance. When $\alpha > \eta$ (1) shows exponential growth and hence breakdown of gas occurs.

Fig. 2 illustrates the AC breakdown characteristic of R12 gas and its mixtures with air. The distance between electrodes is 6 mm. In this paper, experimentation on (a) pure R12, (b) R12 + 50% air, (c) R12 + 40% air, (d) R12 + 30% air, (e) R12 + 20% air and (f) R12 + 10% air has been performed. Fig. 2 shows that by adding 50% of R12 to air increases breakdown strength of air rapidly; however, by increasing percentage of R12, the breakdown strength does not bring the same change. This is due to the fact that R12 has a high attachment cross-section at low energies of 0–0.5 eV [15]. Thus when a small amount of R12 was added almost all the low energy electrons attached caused a rapid increase in the breakdown voltage.

The gap distance also effects dielectric strength. The increase in the distance between the electrodes also increases the breakdown voltage which can be verified from (2). When the distance between electrodes is increased then it needs high potential for balancing the electric field between the electrodes

$$E = f \times \frac{V}{D}$$

where $f$ is the non-uniformity constant, $D$ is the distance between electrodes and $V$ is the applied voltage.

Fig. 3 shows that the dielectric strength increases by increasing the gap distance between the electrodes. The figure also gives a comparison of R12 with SF$_6$ in AC environment. Dielectric strength of R12 is 0.90 times of SF$_6$.

Fig. 4 contains Paschen curve for different ratios of R12/air in the PD (pressure × distance) range of 0.2–2.5 Mpa*mm. Here, the
distance from one electrode to the other is \(D\) and \(P\) is the pressure. With the increase in pressure, the breakdown voltage also increases, however, there is a steady decrease in the slope of the curve in the sphere–sphere electrode. The effects are deviations from Paschen's law as shown in Fig. 4. Breakdown strength on ideal gas has to be linear in theory.

Fig. 5 illustrates the breakdown characteristic of R12/air in DC environment. DC has been generated by Greinacher voltage doubler circuit in the range of 0–140 kV. The best dielectric strength of R12/air (70/30%) has been achieved in DC.

Fig. 6 gives the breakdown characteristic of R12/air in impulse environment. Impulse voltage has been generated by impulse generated circuit. Positive impulse is applied then breakdown voltage increases with pressure for certain pressure value \(P_m\). The best dielectric strength of R12/air (70/30%) has been achieved in impulse.

The best dielectric strength of R12/air (70/30%) has been achieved in AC, DC and impulse based on our experimental condition. By adding air, the dielectric strength of the mixture is greater than that of pure R12 called positive synergistic effect. The advantage of adding air is that it decreases its boiling temperature and also GWP. GWP of R12 is ten times less than \(SF_6\) gas as a result R12 is the best alternative of \(SF_6\) gas.

3.2 Mixture of R12 and nitrogen gas

Nitrogen gas is an inert gas, which is the most stable available gas and it has zero GWP and very low boiling temperature. By adding nitrogen to the R12 gas will decrease its boiling point and GWP. Experiment shows that adding a small amount of R12 will significantly increase dielectric strength of nitrogen gas and further addition will not bring a rapid increase in breakdown voltage. It is due to the attachment of low energy electron because attachment of high energy electron is very difficult. In Fig. 7, breakdown strength of R12 with a different mixing ratio of nitrogen by varying pressure has been studied in this paper. Fig. 7 shows that best dielectric strength can be achieved at R12/N\(_2\) (80/20%) at 50 lb/in\(^2\), which is 0.95 times that of \(SF_6\). Fig. 8 gives the Paschen curve of R12/N\(_2\) at different ratios and pressures.

3.3 Insulation characteristic of base and mixed gases

Tables 2 and 3 show the insulation characteristic of the base and mixed gases. Table 2 shows the insulation characteristic of R12/air and Table 3 gives the insulation characteristic of R12/N\(_2\). These tables depict standard deviation, mean and the coefficient of variation from the experimental values to show the variability of

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and as carbon is a good conductor of electricity therefore, it bridges the insulation. This is a drawback of R12 used in high-voltage application, but this drawback can be removed by the certain method of preventing of carbonisation given in the literature [33].

When a breakdown occurs, due to temperature increase and overvoltage if insulation was not damaged is called self-recoverability of insulation. A power AC frequency breakdown tests were performed, the testing circuit is shown in Fig. 1. For every 1 min, 20 shots of breakdown were applied to the insulation gas and a minute decay in breakdown voltage was observed which can be ignored. The by-products formed by decomposition of R12 are carbon monoxide, carbon dioxide, hydrogen fluoride, hydrogen chloride [32]. A small amount of carbon is formed on the electrode and as carbon is a good conductor of electricity therefore, it bridges the insulation. This is a drawback of R12 used in high-voltage application, but this drawback can be removed by the certain method of preventing of carbonisation given in the literature [33].

Thus, R12 gas shows very good self-recoverability (Fig. 9).

The result obtained from the experiments. A standard deviation shows us how the data has varied or dispersed from its mean. In the tables, these values are calculated for different proportions. As we can see from Table 3, the standard deviation of R12 and N₂ (70/30%) gives us the lowest value, which shows that there is a rapid change in value. This is also demonstrated by the low value of the coefficient of variation to the mean as well.

### Table 2 Insulation characteristic of R12 and air

<table>
<thead>
<tr>
<th></th>
<th>BG</th>
<th>MG</th>
<th>R12 Air</th>
</tr>
</thead>
<tbody>
<tr>
<td>S.D</td>
<td>9.16</td>
<td>9.6</td>
<td>11.2</td>
</tr>
<tr>
<td>μ</td>
<td>44.4</td>
<td>54.6</td>
<td>57.9</td>
</tr>
<tr>
<td>CV</td>
<td>0.20</td>
<td>0.17</td>
<td>0.19</td>
</tr>
<tr>
<td>Max, kV</td>
<td>57.3</td>
<td>68.9</td>
<td>72</td>
</tr>
<tr>
<td>Min, kV</td>
<td>28.2</td>
<td>35.3</td>
<td>35.7</td>
</tr>
</tbody>
</table>

BG, base gas; MG, mixed gas; BGR, base gas ratio; S.D, standard deviation; μ, mean; CV, coefficient of variation.

### Table 3 Insulation characteristic of R12 and N₂

<table>
<thead>
<tr>
<th></th>
<th>BG</th>
<th>MG</th>
<th>R12 Nitrogen</th>
</tr>
</thead>
<tbody>
<tr>
<td>S.D</td>
<td>11.0</td>
<td>12.3</td>
<td>5.22</td>
</tr>
<tr>
<td>μ</td>
<td>46.1</td>
<td>51.3</td>
<td>59.7</td>
</tr>
<tr>
<td>CV</td>
<td>0.23</td>
<td>0.24</td>
<td>0.08</td>
</tr>
<tr>
<td>Max, kV</td>
<td>60</td>
<td>69.3</td>
<td>70.1</td>
</tr>
<tr>
<td>Min, kV</td>
<td>27</td>
<td>33</td>
<td>51.2</td>
</tr>
</tbody>
</table>

BG, base gas; MG, mixed gas; BGR, base gas ratio; S.D, standard deviation; μ, mean; CV, coefficient of variation.

### Fig. 9 Insulation self-recoverability of R12 gas

When two gases are mixed and they show non-linear behaviour then this non-linearity will be used to describe its synergistic effect. There are four types of synergistic effects which are: positive synergistic effect, negative synergistic effect, linear relation effect and synergistic effect [31]. The connection between the synergistic effect index C, mixing ratio and breakdown voltage is shown in the following equation:

\[ V_m = V_1 + \frac{(V_2-V_1)}{(1+k)} \quad v_1 > v_2 \]  

(3)

V₁ and V₂ represent the breakdown voltages of the pure gases, \( V_m \) is the breakdown voltages of mixed gas, \( k \) is the mixing ratio and \( C \) is the synergistic effect index. Using (3), the synergistic effect of R12/air and R12/N₂ has been calculated and given in Tables 4 and 5, respectively. Detail analysis is given in Tables 3 and 4 of R12/air and R12/N₂.

Tables 4 and 5 specify that the \( C \) value of R12/air and R12/N₂ declines when the increase in pressures is at the same ratio as that of the mixing gas. \( C \) value indicates synergistic effects. When the gas pressure increases beyond 10 lb/in² at 0.70–0.80 mixing ratios, the \( C \) value becomes negative and synergistic effect changes to positive synergistic effects. Thus as pressure increases, the \( C \) value becomes less. The more evident the synergistic effect is, the better the benefits of using mixed gases as insulating gas. The advantage of additive gases is to de-energise high energy electron and return them to lower energy level because at high energy level attachment of electrons is very difficult. The mixing ratio of R12/air (70/30%) at 45 lb/in² gives the best optimal value. R12/N₂ the mixing ratio (80/20%) shows less value of \( C \) and it gives a better result than pure R12.

- \( C > 1 \) shows negative synergistic effect.
- \( C = 1 \) shows linear relation effect.
- \( 0 < C < 1 \) shows synergistic effect.
- \( C < 0 \) shows positive synergistic effect.

### 6 Liquefaction temperature

Liquefaction factor is an important point for choosing an alternative of SF₆. Gas insulation strength degrades due to decreasing temperature because its pressure gets low when temperature decreases. R12 gas has −29.8°C boiling point at atmospheric pressure as shown in Fig. 10. Calculated from (4) which is higher than SF₆, −63°C [34]. Such a high temperature is a disadvantage of R12 gas. Nitrogen has −196°C boiling point at atmospheric pressure.
atmospheric pressure, which is very low than R12 gas. If nitrogen and R12 gases were mixed then disadvantages of high liquefaction point can be eliminated. Mixed gas liquefaction temperature is shown in Fig. 11, which suggests that if the ratio of base gas was increased liquefaction temperature is increased

\[ p = \exp \left( A \left( \frac{1 - T_b/T}{R} \right) \right) \]  

where \( p \) is the gas boiling point pressure; \( T_b \) is the liquefaction temperature at atmospheric pressure (K); \( T \) is the liquefaction temperature of the base gas in mixed gases; \( R \) is gas constant (2 Cal deg.\(^{-1}\) mol\(^{-1}\)); and \( A \) is constant (21 Cal deg.\(^{-1}\) mol\(^{-1}\)).

### 7 Conclusion

Alternative to SF\(_6\), the gas used in power equipment for insulation has been examined in this paper. Dielectric characteristic of R12 gas with mixture of nitrogen and air shows good alternative to SF\(_6\). Under the quasi-uniform electric field, power frequency breakdown characteristics of gases with different pressures are in order of SF\(_6\) < R12 < N\(_2\) < air. When the gas pressures are 45 and 50 lb/in\(^2\) the dielectric strength of mixtures of R12 and air/N\(_2\) reaches to 0.90–0.95 times of SF\(_6\).

Self-recoverability test has been performed, R12 shows good self-recoverability potential. The breakdown has been done after every 1 min and its dielectric strength slightly decreases which can be ignored. Mixed gases show good synergistic effects so it is advantageous to use mixed gases as compared to pure one. R12/air (70/30%) at 45 lb/in\(^2\) shows good synergism index value. R12/N\(_2\) (80/20%) shows decent synergism index value at 50 lb/in\(^2\). Mixed gases have low GWP and boiling point as compared to pure one.

Liquefaction temperature of an R12 gas is high which is a disadvantage of R12 gas. Low liquefaction gases such as nitrogen and air have been added to R12 gas then its liquefaction temperature will decrease.

While this study has provided the fundamental characteristics of R12 with a mixture of air and nitrogen. However, in the subsequent
study R12 gas with another mixture of gases such as (a) carbon dioxide and (b) helium can be investigated due to low GWP and high dielectric strength. Furthermore, another gas from Freon family, namely R134 gas has 94% less GWP and low cost as compared to SF6 can be systematically analysed.

8 References