NUMBER OF SUPPLIED ELECTRONS UNDER REPETITIVE DISCHARGE IN HIGH PRESSURE NITROGEN-BASED GAS MIXTURE

Isao Nagashima, Torul Iwao and Motoshige Yumoto
Tokyo City University, 1-28-1 Tamazutumi, Setagaya, Tokyo 158-8557, Japan
*Email: <dure-su-tetete-ru@hotmail.co.jp>

Abstract: SF$_6$ gas is widely used in the electric power facility. However due to the global warming problems, alternation gases are examined. In this study, the breakdown delay time was measured in the nitrogen based gas mixture and the number of supplied electrons was evaluated. The number of supplied electrons decreased remarkably by mixing gases such as CO$_2$ and NO which was expected to release the internal energy of active species generated by discharge. SF$_6$ which was expected to attach electron was also effective to reduce the number of electrons. CF$_3$I was also examined and its effect was the highest compared with other gases.

1 INTRODUCTION

SF$_6$ gas is widely used in the electric power facility because of its excellent dielectric and arc-quenching properties. However, its GWP (Global Warming Potential) is very high. Therefore, SF$_6$ was designated as the exhausting reduction object at the third Conference of the Parties (COP3) held in 1997. Accordingly, many researches have been carried out on the substitute for SF$_6$ [1]. The authors have measured the breakdown delay time in nitrogen-based gas mixture.

However, it is widely known that a large amount of neutral activated species are generated by discharge in N$_2$ gas. Each of those species has high potential energy. Therefore, various influences on breakdown properties are observed. For example, there are many reports that the effective ionization coefficient increases [2] and a large number of electrons leading the following breakdown are supplied in low pressure [2] [3] [4]. It is shown that many electrons are supplied into the space for a long time under the condition of high pressure [5] [6]. From the result, the authors deduced that many neutral active species remained in space between electrodes for a long term even under high pressure. It is also suggested that electrons are supplied at the cathode surface by the gamma effect [7]. Therefore, accumulation of activated species generated by discharge should be the source of supplied electrons even under high pressure.

So a small amount of gas which was expected to release the internal energy of active species was mixed into nitrogen and the breakdown probability and the delay time was measured. From the results, the number of supplied electrons by mixing gases was evaluated [5] [6]. The results showed that the number of electrons supplied decreased remarkably by mixing NO or CO$_2$ compared with that of pure N$_2$. The number of supplied electrons also decreased by SF$_6$ which is expected to attach electrons [6]. Thus the authors paid attention to CF$_3$I which is expected to liberate the internal energy and to attach electrons emitted from the cathode.

2 EXPERIMENTAL SET UP AND METHOD

The purpose of this study is to make sure the contribution of the de-excitation effect and the attachment effect on the number of supplied electrons. So the particle number density of mixed gas such as CO$_2$, NO, SF$_6$ or CF$_3$I was changed and the number of supplied electrons was measured.

2.1 Electron attachment

In general, electron is attached by the following reactions:

$$e + A + M \rightarrow A^- + M$$  \hspace{1cm} (1)

where, e: electron, A: gas molecule A$: its negative ion and M: the third body molecule. When electron has high kinetic energy, third body: M is necessary to remove the surplus energy. In this study SF$_6$ and CF$_3$I were used.

2.1.1. De-excite of the internal energy

Activated species with the internal energy is de-excited by the following reactions:

$$B^* + C \rightarrow B + C^*$$  \hspace{1cm} (2)

$$C^* \rightarrow C + h\nu$$  \hspace{1cm} (3)

where, $B^*$: activated species with the internal energy, B: its ground state, C: quenching particle. As a result, activated species with a long life time will release its internal energy in a short time. Now, the number density of activated species decreases depending on the following equation:
\[
\frac{d[B^*]}{dt} = k[B^*][C]
\]  \hspace{1cm} (4)

where, \( k \) is the de-excitation reaction rate coefficient. Accordingly, its decreasing rate is proportional to the number density of the quenching molecule.

### 2.2 Experimental setup

The experimental setup is shown in figure 1. The tungsten hemisphere electrode of 5.0 mm in diameter – the brass plane electrode of 20.0 mm in diameter system was used and its gap length was 1.0 mm. The negative voltage was applied to the hemisphere electrode.

Prior to making measurement, the chamber was evacuated less than 1 Pa and gas was filled into the chamber. The purity of \( \text{N}_2 \) was 99.9995\%. Pressure was changed from 0.1 to 0.6 MPa. Number density of mixing gas was changed from \( 8.0 \times 10^{21} \) to \( 8.0 \times 10^{25} \) particles/m\(^3\). The time delay of breakdown after voltage application was measured by the oscilloscope (Tektronix TDS3034B) using the high voltage probe (Tektronix P6015A).

#### 2.3 Number of supplied electrons

The time chart of test procedure is shown in figure 2. Where the applied voltage: \( V \), the static breakdown voltage: \( V_s \), the statistical time delay: \( t_s \), the formative time delay: \( t_f \), and the delay time is the sum of \( t_i \) and \( t_s \). The voltage was applied repeatedly after the passage of the interval time: \( t_i \) and the delay time: \( t \) was measured. In this experiment, interval time was 1, 5, 10 and 15 s.

Suppose that a voltage is applied \( n_0 \) times to a gap, and the delay time: \( t \) is measured every time. Let \( n_i \) denotes the number of times a delay time longer than \( t \) is observed. Then \( n_i/n_0 \) can be represented as shown in Figure 3, so called Laue plot, and the portion with a negative gradient can be expressed as follows:

\[
\frac{n_i}{n_0} = \exp(- \frac{t-t_f}{t_0})
\]  \hspace{1cm} (5)

In the above equation, \( n_i/n_0 \) represents the probability of breakdown with a delay time of \( t \) or longer. The statistical delay time: \( t_s \) is the inverse of the gradient at the straight line. The following relationship exists between the number of supplied electrons: \( Y \) appearing in the gap per unit time, the probability \( W \) that primary electron accelerated by the electric field will cause breakdown, and the gradient of the Laue plot.

\[
Y = \frac{1}{Wt_s}
\]  \hspace{1cm} (6)

Here a probability \( W \) of 1 is assumed when the applied voltage: \( V \) is \( 1.1V_s \) or higher. Accordingly, \( 1.2V_s \) was applied in this test. Therefore, the number of supplied electrons: \( Y \) can be calculated from the gradient of the Laue plot.
3 PRELIMINARY EXPERIMENT

3.1 Number of supplied electrons in nitrogen

Figure 4 shows the number of supplied electrons obtained under 0.5MPa in pure nitrogen by changing the interval time from 1 to 2400s. The number of supplied electrons reduces to 0.1 s\(^{-1}\). It is reported that the number of supplied electrons reached to a constant value under the fewer active species [8] even if changing interval time. Therefore, the back ground of supplied electrons is 0.1 s\(^{-1}\) or less under the experimental condition.

Figure 4: Dependence of the number of supplied electrons on the interval time in N\(_2\) (0.5 MPa).

3.2 Laue plot of N\(_2\)/CF\(_3\)I

Figures 5 and 6 show the Laue plots of N\(_2\)/SF\(_6\) and N\(_2\)/NO. Each Laue plot was obtained by 30 measurements under 0.1MPa and varying the interval time. Laue plots of N\(_2\)/SF\(_6\) and N\(_2\)/NO show roughly the straight line. Figure 7 which is the Laue plots of N\(_2\)/CF\(_3\)I shows the convex curve extremely. The result obtained means that the test condition changes gradually by repeating breakdown. It is widely known that CF\(_3\)I is easily dissolved into its elements [9]. Accordingly, it was supposed that CF\(_3\)I was dissociated by discharges and the composition in the test chamber changed gradually. Then, the Laue plots were obtained by 10 measurements for each interval time. The result is shown in figure 8. Figure 8 shows the straight line on each condition and its reproduce in high. Therefore, the Laue plot of CF\(_3\)I was obtained by changing gas every 10 measurements. Figure 9 is the Laue plot of CF\(_3\)I obtained by 30 measurements which shows roughly the straight line. The number of supplied electrons is calculated by using the straight Laue plot.

Figure 5: Laue plots for SF\(_6\) obtained by 30 data (0.1MPa).

Figure 6: Laue plots for NO obtained by 30 data (0.1MPa).

Figure 7: Laue plots for CF\(_3\)I obtained by 30 data (0.1MPa).
4 RESULTS AND DISCUSSION

4.1 Number of supplied electrons in N$_2$/CF$_3$I

Figure 10 shows the dependence of the number of supplied electrons on the number density of CF$_3$I. Each plot was obtained by four measurements. Each plot in figure 10 means the average and the error bars in the figure show the maximum and the minimum of each measurement. The author judged that each plot obtained in the different pressure has meaningful difference from the width of dispersion.

Number of supplied electrons decreases depending on the number density of CF$_3$I. Now, there is a tendency that the decrease rate of the number supplied electrons depends on the pressure. At 0.1 MPa, the decrease rate is small and it decrease linearly against the number of density of CF$_3$I apparently. On the other hand, the number of supplied electrons obtained at 0.2 and 0.3 MPa decreases drastically and each curve intersects around $3 \times 10^{23}$ particle/m$^3$.

4.2 Dependence of mixed gases

Figure 11: Dependence of the number of supplied electrons on the particle number density (NO, CO$_2$ or CF$_3$I).

Dependence of the number of supplied electrons on particle number density of CO$_2$, NO and CF$_3$I are shown in Figure 11. The results were obtained by changing the total gas pressure.

The number of supplied electrons for each gas mixture decreases depending on the particle number density. Its decrease rate, is about the same. Therefore the number of supplied electrons decreases linearly apparently. Accordingly, it is seen that each data distributes in parallel. The similar tendency is obtained in case of CF$_3$I at 0.1 MPa. Now, for obtaining the same number of supplied electrons, the number density of CO$_2$ needs about 2 order of magnitude large than that
of NO. This tendency is the same at 0.3 and 0.6 MPa. As compared with 3 mixed gases, CF$_3$I needs the smallest number density to obtain the same effect.

Table 1 shows the rate coefficients for quenching of N$_2$(A$^3\Sigma_u^+$) [9] [10]. The state of N$_2$(A$^3\Sigma_u^+$) should be quenched for a short time under the high pressure test condition. However, many data concerned about N$_2$(A$^3\Sigma_u^+$) have been reported. So, to discuss the mechanism quantitatively, table 1 is shown for one reference. From table 1. It is shown that the rate coefficient of CF$_3$I is the highest compared with other gases. The magnitude of the rate coefficient of NO is about 2 order larger than that of CO$_2$. Compared with the result shown in figure 4 and that of figures 10 and 11, it is clear that the number of supplied electrons decreases remarkably by mixing a small amount of CO$_2$, NO or CF$_3$I. The effect of mixed gas shown in figure 11 denotes the similar tendency of the quenching rate coefficient shown in table 1. From these results, it is assumed that the number of supplied electrons depends on the quenching reactions. The author pointed out that the gamma effect at the cathode surface was one of the important process for electron supply under repetitive breakdown in enclosed condition [7]. Therefore, it is deduced that the number of activated species generated by discharge may be quenched by mixing gas and the number of supplied electrons may decrease.

However, dependence of the number of supplied electrons on the number density of CF$_3$I at 0.2 and 0.3 MPa shows a little different tendency. It is widely known that CF$_3$I dissociates easily by discharge [9]. CF$_3$I is a molecule consisting of halogen atoms. Therefore, CF$_3$I generates many halogen atoms by dissociation. Accordingly, the electrons supply processes of CF$_3$I at 0.2 and 0.3 MPa may be a little difference compared with that of CO$_2$, NO and CF$_3$I at 0.1 MPa. It is necessary to compare the results of SF$_6$ at the next stage.

<table>
<thead>
<tr>
<th>Gas</th>
<th>De-excitation Reaction rate constant [m$^{-3}$/s]</th>
</tr>
</thead>
<tbody>
<tr>
<td>N$_2$</td>
<td>1×10$^{-24}$</td>
</tr>
<tr>
<td>CO$_2$</td>
<td>2×10$^{-19}$</td>
</tr>
<tr>
<td>NO</td>
<td>7×10$^{-17}$</td>
</tr>
<tr>
<td>CF$_3$I</td>
<td>2×10$^{-16}$</td>
</tr>
</tbody>
</table>

5 SUMMARY

The authors paid their attention to the nitrogen based gas mixture as alternative gas of SF$_6$. However, nitrogen generates a large amount of activated species by discharge. Therefore, many electrons are supplied for a long time even under the condition of high pressure. These results mean that many activated species remain in the space between electrodes. To reduce the number of supplied electrons, a small amount of gas, such as CO$_2$, NO and CF$_3$I, was mixed into nitrogen. It is expected that these gases may de-excite the activated species. Delay time of breakdown was measured and the number of supplied electrons was calculated. The test was carried out by changing the particle number density of mixed gases. The results are as follows:

In the case of CF$_3$I, decrease rate of the number of supplied electrons varied depending on pressure.

Dependence of the number of supplied electrons on the number density of CF$_3$I at 0.1 MPa showed a similar tendency to that of CO$_2$ and NO. It is deduced that the quenching reactions of activated species may be the important processes for electron supply.

In the case of CF$_3$I, it is assumed that the behavior of CF$_3$I may change depending on pressure. It is necessary to compare the results of SF$_6$. The test will be carried out in near future.

6 REFERENCES


