

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

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Abstract: SF₆ 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 CO₂ and NO which was expected to release the internal energy of active species generated by discharge. SF₆ which was expected to attach electron was also effective to reduce the number of electrons CF₃I was also examined and its effect was the highest compared with other gases.

1 INTRODUCTION

SF₆ 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, SF₆ 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 SF₆ [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 N₂ 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 CO₂ compared with that of pure N₂. The number of supplied electrons also decreased by SF₆ which is expected to attach

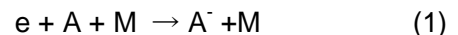
electrons [6]. Thus the authors paid attention to CF₃I which is expected to liberate the internal energy and to attach electrons emitted from the cathode.

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 CO₂, NO, SF₆ or CF₃I was changed and the number of supplied electrons was measured.

2.1 Electron attachment

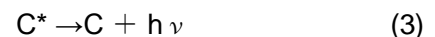
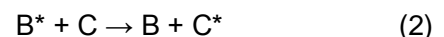
In general, electron is attached by the following reactions:



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 SF₆ and CF₃I were used.

2.1.1. De-excite of the internal energy

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



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:

$$\frac{d[B^*]}{dt} = k[B^*][C] \quad (4)$$

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 N_2 was 99.9995%. Pressure was changed from 0.1 to 0.6 MPa. Number density of mixing gas was changed from 8.0×10^{21} to 8.0×10^{25} particles/m³. The time delay of breakdown after voltage application was measured by the oscilloscope (Tektronix TDS3034B) using the high voltage probe (Tektronix P6015A).

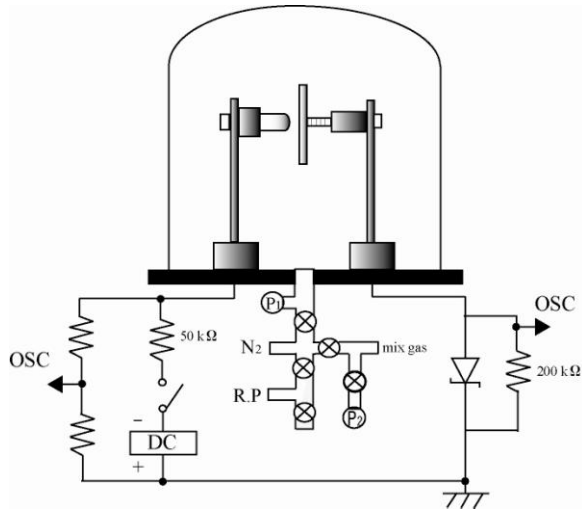


Figure 1: Experimental setup.

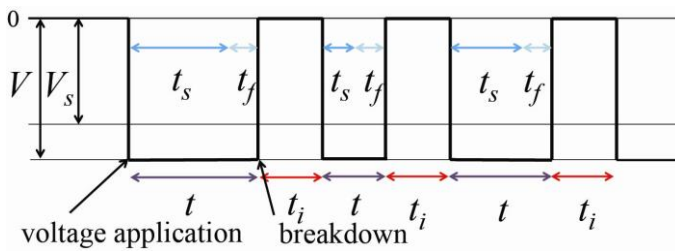


Figure 2: Time chart of repetitive discharge.

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_f 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_t denotes the number of times a delay time longer than t is observed. Then n_t/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:

$$\frac{n_t}{n_0} = \exp\left(-\frac{t - t_f}{t_0}\right) \quad (5)$$

In the above equation, n_t/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.

$$Y = \frac{1}{W t_s} \quad (6)$$

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

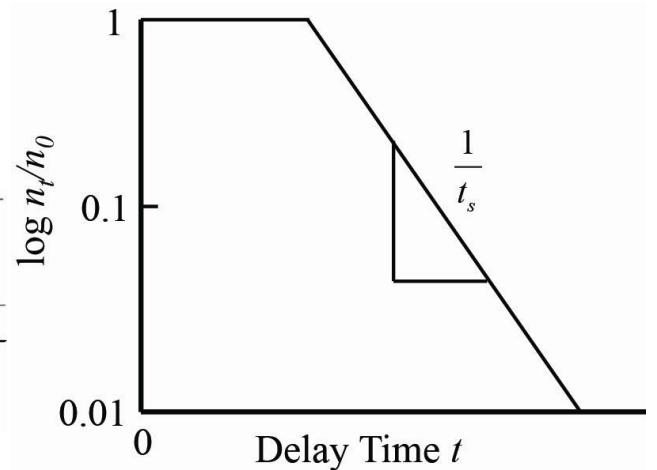


Figure 3: Laue plot.

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.

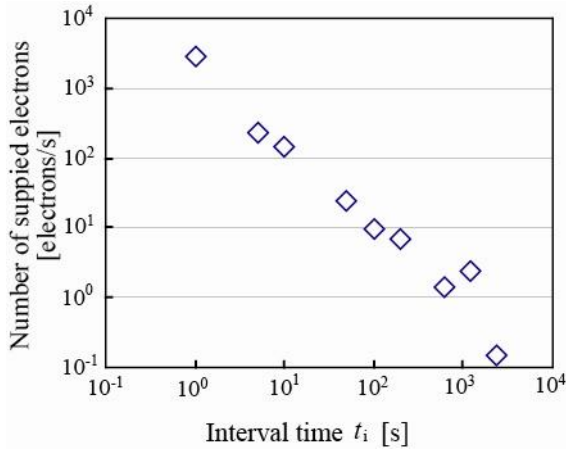


Figure 4: Dependence of the number of supplied electrons on the interval time in N_2 (0.5 MPa).

3.2 Laue plot of $\text{N}_2/\text{CF}_3\text{I}$

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 $\text{N}_2/\text{CF}_3\text{I}$ shows the convex curve extremely. The result obtained means that the test condition changes gradually by repeating breakdown. It is widely known that CF_3I is easily dissolved into its elements [9]. Accordingly, it was supposed that CF_3I 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_3I was obtained by changing gas every 10 measurements. Figure 9 is the Laue plot of CF_3I obtained by 30 measurements which shows roughly the straight line. The number of supplied electrons is calculated by using the straight Laue plot.

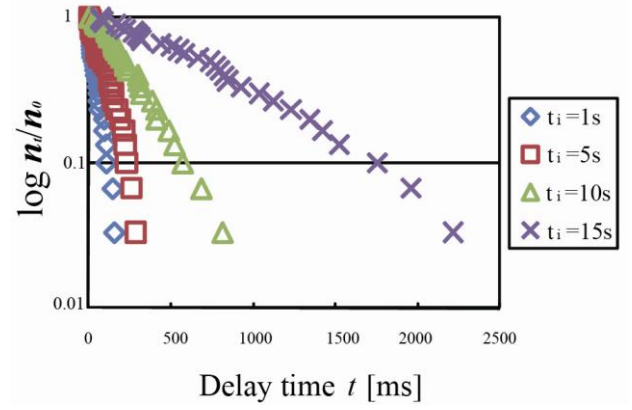


Figure 5: Laue plots for SF_6 obtained by 30 data (0.1MPa).

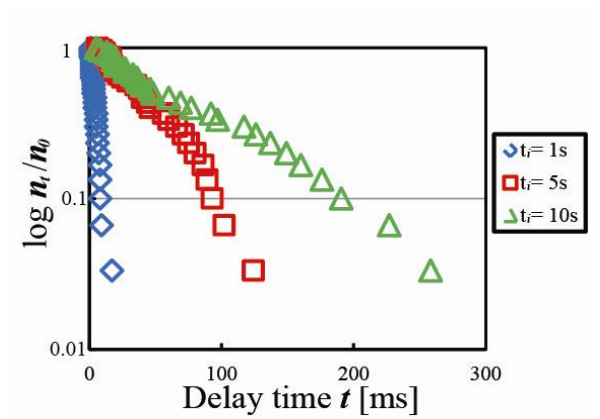


Figure 6: Laue plots for NO obtained by 30 data (0.1MPa).

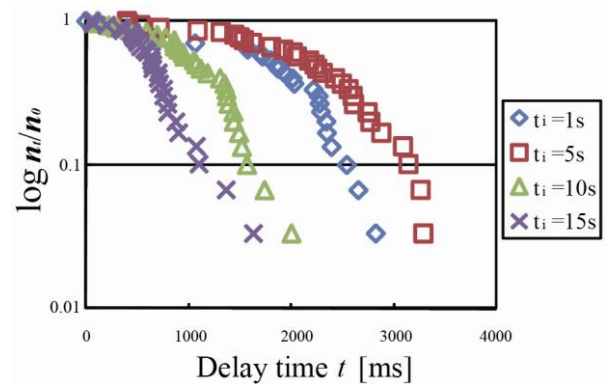


Figure 7: Laue plots for CF_3I obtained by 30 data (0.1MPa).

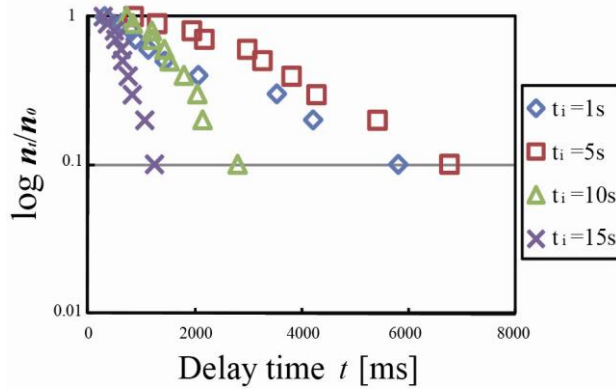


Figure 8: Laue plots for CF_3I obtained by 10 data (0.1MPa).

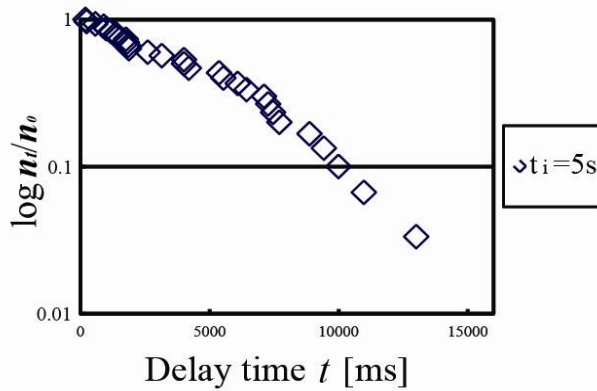


Figure 9: Laue plot for CF_3I obtained by 30 data (0.1MPa).

4 RESULTS AND DISCUSSION

4.1 Number of supplied electrons in $\text{N}_2/\text{CF}_3\text{I}$

Figure 10 shows the dependence of the number of supplied electrons on the number density of CF_3I . 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_3I . 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_3I 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 3×10^{23} particle/ m^3 .

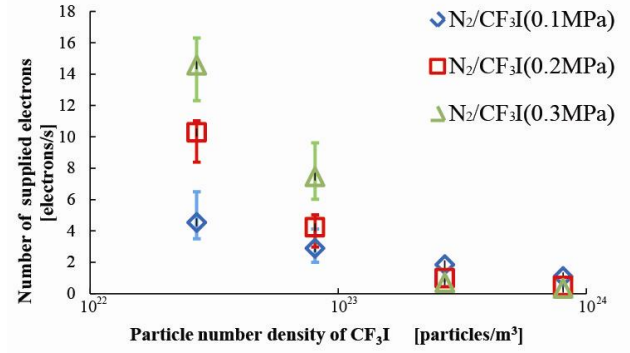


Figure 10: Dependence of the number of supplied electrons on the number density of CF_3I ($t_i = 5$ s).

From figure 8 and 9, dependence of the delay time on the interval time shows the different tendency compared with that of SF_6 and NO . That is to say, delay time becomes shorter under the larger interval time. From three results, it is conceivable that, the electron supplied process changes depending on the pressure, in case of CF_3I .

4.2 Dependence of mixed gases

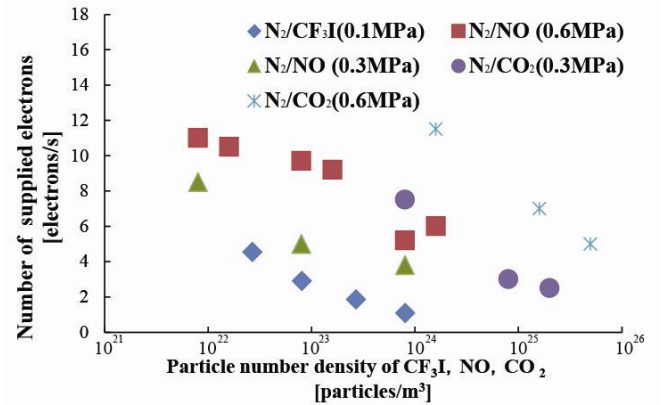


Figure 11: Dependence of the number of supplied electrons on the particle number density (NO , CO_2 or CF_3I).

Dependence of the number of supplied electrons on particle number density of CO_2 , NO and CF_3I 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_3I 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_3I needs the smallest number density to obtain the same effect.

Table 1 shows the rate coefficients for quenching of $\text{N}_2(\text{A}^3\Sigma_u^+)$ [9] [10]. The state of $\text{N}_2(\text{A}^3\Sigma_u^+)$ should be quenched for a short time under the high pressure test condition. However, many data concerned about $\text{N}_2(\text{A}^3\Sigma_u^+)$ 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_3I 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_3I . 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_3I at 0.2 and 0.3 MPa shows a little different tendency. It is widely known that CF_3I dissociates easily by discharge [9]. CF_3I is a molecule consisting of halogen atoms. Therefore, CF_3I generates many halogen atoms by dissociation. Accordingly, the electrons supply processes of CF_3I at 0.2 and 0.3 MPa may be a little difference compared with that of CO_2 , NO and CF_3I at 0.1 MPa. It is necessary to compare the results of SF_6 at the next stage.

Table 1: Deexcitation reaction rate constant for $\text{N}_2(\text{A}^3\Sigma_u^+)$ [10] [11].

Gas	De-excitation Reaction rate constant [m^3/s]
N_2	1×10^{-24}
CO_2	2×10^{-19}
NO	7×10^{-17}
CF_3I	2×10^{-16}

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_3I , 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_3I , 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_3I 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_3I , it is assumed that the behavior of CF_3I 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

- [1] T.Roknohe, Y.Yagihashi, K.Aoyama, T.Oomori and F.Endou: "Development of SF_6 -free 72.5 kV GIS", *IEEE Trans. Power Deliv.*, Vol.22, No.3, pp1869-1876(2007).
- [2] S.C.Haydon and O.M.Williams: "The experimental value of the Townsend first ionization coefficient in nitrogen at low value of E/p ", *J.Phys.D:Appl.Phys.*, Vol.5, pp.L79-L81 (1972).
- [3] V Lj Markovic, M M Pejovic and Z Lj Petrovic: "Kinetics of activated nitrogen states in late afterglow by the time-delay method", *J.Phys.D:Appl.Phys.*, Vol.27, pp.979-984(1994).
- [4] Momcilo M Pejovic, Emilija N Zivanovic and Milic M Pejovic: "Kinetics of ions and active species in the afterglow and their influence on the memory effect in nitrogen at low pressures", *J.Phys.D:Appl.Phys.*, Vol.37, pp.200-210 (2004).
- [5] Y.Kuroki and M.Yumoto: "Repression of primary electrons by mixing the NO or SF_6 with N_2 under high pressure", *T.IEE-A of Japan.*, Vol.127, No.12, pp.777-783(2007) (in Japanese).
- [6] F.Kobayashi, S.Hamano, T.Iwao and M.Yumoto: "Analysis of primary electron supply process and suppression of primary electron in high pressure nitrogen", *Proc. XVI International Conference on Gas Discharges and their Applications*, pp.345-348 (2008).

- [7] F.Kobayashi, S.Hamano, T.Iwao and M.Yumoto: "Decrease of Primary Electron Number by NO, CO₂ and SF₆ Mixing at the Breakdown in High Pressure Nitrogen", *Electrical Engineering in Japan*, Vol.172, No.3, pp.1-7 (2010).
- [8] M.M.Pejovic, G.S.Ristic and J.P.Karamarkovic: "Electrical breakdown in low pressure gases", *J. Phys. D:Appl. Phys.*, Vol.35, pp.R91-R103 (2002).
- [9] T.Takeda, S.Matsuoka, A.Kumada and K.Hidaka: "Breakdown Characteristic of CF₃I gas under steep front square pulse application", *Proc. Technical Meeting of Electronic Discharge*, IEEE Japan, ED-08-41, pp.25-30(2008).
- [10] J.T. Herron: "Evaluated chemical kinetics data for reactions of N(²D), N(²P), and N₂(A³Σ_u⁺) in the gas phase", *J. Phys. Chem. Ref. Data*, Vol.28, No.5, pp1453-1483(1999).
- [11] V Guerra, P.A.Sa and J.Loureiro: "Role played by the N₂(A³Σ_u⁺) metastable in stationary N₂ and N₂-O₂ discharges", *J. Phys. D: Appl. Phys.*, Vol.34, pp.1745-1755 (2001).