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Applying a Gas Mixture Containing c-C₄F₈ as an Insulation Medium

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ABSTRACT

This paper studies the possibility of applying a gas mixture containing c- C_4F_8 in the gas insulation of power equipment. Environmental aspects such as global warming potential, ozone depletion potential, recycling loss and toxicity are discussed. Insulation characteristics of gas mixtures such as c- C_4F_8/N_2 , c- C_4F_8/a ir, and c- C_4F_8/CO_2 are examined experimentally under a quasi-homogeneous or an inhomogeneous electric field condition. Most of the characteristics are compared with those of SF_6/N_2 , which is now widely studied as the most plausible alternative to pure SF_6 for gas insulation. The experimental results, together with the discussion, suggest that gas mixtures containing c- C_4F_8 are possible substitutes for SF_6 and SF_6/N_2 .

1 INTRODUCTION

 \mathbf{S}^{F_6} is now widely used in important electric power systems such as gas-insulated system (GIS) and gas circuit breaker (GCB). Intensive research made in the past has not succeeded in finding a substitute gas comparable to \mathbf{S}^{F_6} in terms of the integrated performance of electrical, physical and chemical properties. Recently, however, high global warming potential (GWP), explained below, or the greenhouse effect of \mathbf{S}^{F_6} gas has once more stimulated active studies of searching for a substitute gas or a gas mixture with lower environmental impact [1, 2].

According to these studies, it appears very difficult that any pure gas can bring a solution to the issue of desirable insulation ability and low environmental impact. The present research focuses on the possibility of applying a gas mixture instead of pure SF₆. Most of mixtures now under study contain SF₆ as a component, and with N₂ as the other, major, component. Mixtures composed of a strongly electronegative gas with high dielectric strength such as SF₆ and an ordinary gas (N₂, CO₂ or air) have the following advantages: reduction of the gas price; decrease of liquefaction temperature (especially useful in cold countries); and nonlinear increase of dielectric strength for a low mixing ratio of the electronegative gas, called synergism or synergistic effect.

We believe the wisest policy in the environmental aspect is to use pure SF_6 in the gas insulation and suppress its release and emission as low as possible [3]. However, gas mixtures have at least one additional advantage compared to pure SF_6 , *i.e.* reduction of the latent GWP of an insulating gas.

Because there exist many hydrofluorocarbon (HFC) and perfluorocarbon (PFC) with much lower GWP than SF₆, we suggest the possibility

of applying a mixture composed of one such gas with a reduced GWP. Furthermore, a component gas with higher boiling point than SF₆ may mitigate the difficulty of recovering the component gas via the liquid state. This paper discusses the application of c-C₄F₈ (perfluorocyclobutane, C-318) for a gas mixture in environmental, physical and electrical aspects. The principal part concerns the ac insulation characteristics of various gas mixtures in relation to the mixing ratio.

2 SELECTION OF A COMPONENT ELECTRONEGATIVE GAS

2.1 GENERAL

Various requirements have to be fulfilled to apply a mixture in gas insulation. At least one component gas will be an electronegative gas, or a halogenated gas, to realize high insulation ability. The fundamentally important requirements for the selection are non-toxicity, and no ozone destruction ability, or zero ozone depletion potential (ODP).

From the latter requirement, we must exclude compounds including chlorine or bromine, such as chlorofluorocarbon (CFC). The selection is thus limited to those containing only fluorine among halogens such as hydrofluorocarbon (HFC) and perfluorocarbon (PFC). Table 1 lists such candidate compounds with no, or unknown, toxicity, which have higher insulation ability than, or comparable to, SF_6 . The insulation ability is expressed by the relative dielectric strength to that of SF_6 .

Among these compounds, C_4F_{10} is not toxic, but has a relatively high boiling point and its GWP is not known. Toxicity and GWP are not known both for 1,3- C_4F_6 and CF_3OCF_3 . CF_3SF_5 is considered to have

Table 1. Candidate component gases.

	Dielectric strength	B.P.
SF ₆	1.0	-63*
C ₂ F ₆	0.78→0.79	-78*
C_3F_8	$0.96 \rightarrow 0.97$	-37
C ₄ F ₁₀	$1.36 \rightarrow 1.32$	-2
1,3-C ₄ F ₆	1.5	-6.5
CF ₃ OCF ₃	0.84	-59
CF ₃ SF ₅	$1.51 { o} 1.52$	-24 (-20)
c-C ₄ F ₈	1.25, 1.31	-6 (-8)

higher GWP than SF₆. We selected c-C₄F₈ as the most promising component gas of a mixture for gas insulation [4]. Other basic requirements are discussed further below.

2.2 ENVIRONMENTAL ASPECTS

According to the evaluation reported by the Underwriters Laboratory, c-C₄F₈ is entirely nontoxic, classified as the safest rank of class 6 similarly as SF₆. As already mentioned, it has no ozone depletion potential.

The greenhouse effect is quantitatively represented by the GWP, which gives the warming effect of an emitted gas at unit quantity, over an integrated period, relative to the warming effect of CO₂. The GWP of SF₆ is 23900 for an evaluation period of 100 yr. On the other hand, the GWP of c-C₄F₈ is 8700, only 36% of SF₆ [5]. It corresponds to half as much warming effect as SF₆ at an identical pressure considering the difference of molecular weight between c-C₄F₈ (200) and SF₆ (146).

2.3 ALLOWABLE MIXING RATIO

The boiling point of c-C₄F₈ is reported as -6° C [6] or -8° C [7], depending on the literature. The application of c-C₄F₈ for gas insulation is inevitably as a gas mixture at a relatively low mixing ratio with an ordinary gas with low boiling point such as N_2 . The vapor pressure P_v (in MPa) of a gas with boiling point T_b is approximated as dependent on temperature T by the following equation [7]

$$P_v = \frac{1}{10} \exp \frac{A(1 - \frac{T_b}{T})}{R} \tag{1}$$

where A=87 J/mol K is a constant, and R=8.3 J/mol K is the gas constant. Thus, by assuming the ideal gas condition, liquefaction temperature T_{mb} of a gas mixture is given by

$$T_{mb} = \frac{T_b}{1 - \frac{\ln(10kP_v)}{10.5}} \tag{2}$$
 where k is the mixing ratio (ratio of partial pressures) for a total pres-

sure P.

Figure 1 represents the thus computed relation between T_{mb} and k. Japanese standards for gas-insulated equipment prescribe the range of ambient temperature between -20°C and 40°C. It can be seen in Figure 1 that in the case of $T_b = -6$ °C, the mixing ratio must be <14% and 11% for P=0.4 and 0.5 MPa, respectively, while it must be <15% and 12% when $T_b = -8$ °C.

2.4 LIQUEFACTION RECOVERY

Another advantage of a mixture with c-C₄F₈ concerns liquefaction recovery. We derive the release rate or loss rate at the liquefaction recovery of a gas mixture [4], with the ideal gas condition assumed in

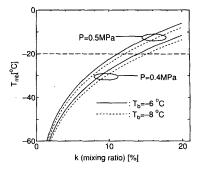


Figure 1. Relation of boiling point T_{mb} of gas mixtures as a function of mixing ratio k.

the derivation. The mixture is composed of gas A with a high boiling point and gas B with a low boiling point, which is compressed at pressure P_c and temperature T_c . The original mixing ratio is k and 1-k; respectively, for A and B. The liquefaction of gas A stops when the partial pressure of the residual gas A reaches its saturated vapor pressure; which depends on T_c . The mixing ratio of the residual gas state is represented similarly as k' and 1 - k'. The total gas volume V_0 of A is given at T_c as

$$V_0 = \frac{(1 - k')P_cVk}{1 - k} \tag{3}$$

where V is the volume of the residual gas mixture. The volume V_r of the residual gas A also at T_c is

$$V_r = k' P_c V \tag{4}$$

Thus, the release rate or the loss rate is given by the following simple expression

$$\lambda = \frac{V_r}{V_0} = \frac{\frac{k'}{1-k'}(1-k)}{k}$$
 (5)

Because $k'P_c = P_A(T_c)$, the saturated vapor pressure of A at T_c is

$$\lambda = \frac{(1 - k)P_A(T_c)}{k[P_c - P_A(T_c)]}$$
 (6)

As example, Figures 2(a) and (b) compare the release rate of SF₆ and c-C₄F₈, respectively. The boiling point of c-C₄F₈ is taken as T_b =-8°C, which corresponds to a more severe condition at the liquefaction recovery. When $T_c = -50$ °C, the release rate of c-C₄F₈ with k=0.1 (10%) is only 6.3 and 2.5% for P_c =2 and 5 MPa, respectively. On the other hand, the corresponding rate of SF₆ with k=0.1 amounts to 100 and 45%.

INSULATION CHARACTERISTICS

3.1 **GENERAL**

Several papers already were published on the insulation strength of pure c-C₄F₈ and its mixtures [8-10]. Reference [8] compares the discharge characteristics of c-C₄F₈ with SF₆ in parallel/plane electrodes, with ac and impulse voltages. The sparkover voltage of c-C₄F₈ is $\sim 1.3 \times$ higher than that of SF₆ at atmospheric pressure for ac (electrode separation \leq 37.5 mm). For lightning impulse voltage with the electrode separation $\lesssim 12.5$ mm, c-C₄F₈ is 1.3 to 1.4× higher than SF₆. At higher

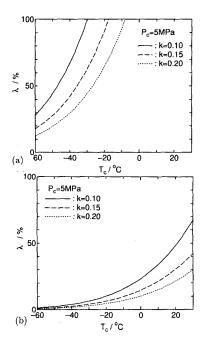


Figure 2. Release rate as function of compression temperature T_{c_r} (a) SF_6/N_2 , (b) $c-C_4F_8/N_2$.

pressures, this ratio somewhat increases with pressure, but remains 1.3 to 1.4 to $0.24\,\mathrm{MPa}$.

It is well known that a mixture with a strongly electronegative gas shows a nonlinear increase of sparkover voltage (synergistic effect) when it is mixed with an ordinary gas. Sparkover voltage of c-C₄F₈/N₂ is reported for a parallel/plane electrode system (dc, negative) and a co-cylindrical configuration (dc, positive and negative) in [9]. It reports that sparkover voltage of c-C₄F₈/N₂ is almost linear in relation to the mixing ratio. However, the experimental condition is limited to low pressure, P=0. 066 MPa (corresponding to Pd=0.6 MPa.mm) for the former measurement and 0.1 MPa for the latter one.

In the following, we investigate the insulation characteristics of $c-C_4F_8$ and its mixture gases experimentally for a wider range of Pd in a quasi-homogeneous field condition. Discharge characteristics in an inhomogeneous field condition are studied also.

3.2 EXPERIMENTAL

The experiment was conducted in a cylindrical pressure tank with 126 mm diameter and 320 mm length, and equipped with a window made of fused glass for observing the discharge. Experimental conditions are as follows: Gases, c-C₄F₈, SF₆, N₂, CO₂ and air; Gas mixtures, c-C₄F₈/N₂, c-C₄F₈/CO₂, c-C₄F₈/air and SF₆/N₂; Mixing ratio (partial pressure of electronegative gas), 0, 5, 10, 20 or 100 vol%; Gas pressure, 0.1, 0.2 and 0.4 MPa.

We employed a sphere/plane electrode system as a quasi homogeneous electric field. Figure 3(a) shows the configuration consisting of a sphere with 30 mm diameter and a plane with 50 mm diameter, both made of aluminum. The electrode separation d was adjusted to 2, 5 and

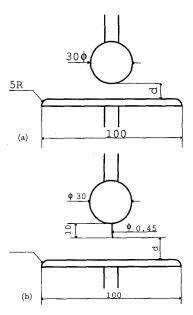


Figure 3. Two types of electrodes, (a) sphere to plane, (b) needle to plane.

10 mm by using a micrometer, which was connected to the plane electrode and installed at one end of the pressure tank. The average surface roughness of the sphere electrode is $\sim 10~\mu \rm m$. This corresponds to the roughness in conductors used for commercial gas insulated equipment such as gas insulated line (GIL) and GIS. The field uniformity is represented quantitatively by the field utilization factor $\xi=E_a/E_{\rm max}$, where $E_{\rm max}$ is the maximum field strength at the surface of the sphere electrode and E_a the average field strength. The value calculated by a charge simulation method is 0.92, 0.81 and 0.67, respectively, for electrode separations of 2, 5, and 10 mm.

An inhomogeneous field condition was realized by attaching a hemispherically capped steel needle to the sphere electrode, as shown in Figure 3(b). The diameter of the needle is $0.45~\mathrm{mm}$ and the length is $10~\mathrm{mm}$. The electrode separation between the needle tip and the plane was selected as $10~\mathrm{or}~15~\mathrm{mm}$.

We applied 60 Hz ac voltage 10 times for each of the experimental conditions to obtain average sparkover voltage or corona inception voltage. Registering a voltage waveform on a digital oscilloscope, we identified the phase or the polarity of the voltage at the instant of sparkover. We also identified the corona inception by monitoring current flowing thorough a shunt resistance, which was inserted between the plane and the ground.

3.3 SPARKOVER UNDER A QUASI-HOMOGENEOUS FIELD CONDITION

3.3.1 SPARKOVER VOLTAGE OF PURE GASES

Figure 4 shows the sparkover voltage of the sphere/plane electrode system for $c-C_4F_8$, SF_6 , N_2 , CO_2 and air as a function of gas pressure.

The electrode separation d=10 mm, and the other separations examined give similar characteristics. The measurement for c-C₄F₈ was conducted only for 0.1 and 0.2 MPa, since the liquefaction occurred at 0.32 MPa when the ambient temperature was 15°C. As seen in the Figure, the sparkover voltage of each gas increases almost linearly with pressure. The sparkover voltage for c-C₄F₈ is 1.3 to 1.35× higher than SF₆ at a same pressure, which is consistent with the already reported data [8].

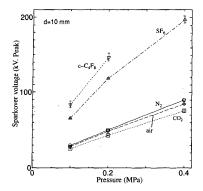


Figure 4. Sparkover characteristics of pure gases under quasi-homogeneous field condition.

3.3.2 SPARKOVER VOLTAGE OF GAS MIXTURES

Figure 5 shows the sparkover voltage as a function of the gas-mixing ratio ranging to 30%. Figures 5(a), (b) and (c) correspond to the gap separation of 2, 5 and 10 mm, respectively. Each of these Figures includes the data for four kinds of gas mixtures at gas pressures of 0.1, 0.2 and 0.4 MPa. Each plot in the Figures shows average sparkover voltage with an error bar indicating the maximum and the minimum voltages obtained during the ten voltage applications.

It can be seen from Figure 5 that the gas mixtures containing $c\text{-}C_4F_8$ have an insulation ability comparable to SF_6/N_2 . Furthermore, when the electrode separation is large (d=10 mm) and the gas pressure is high (P=0.4 MPa), they are often superior to SF_6/N_2 . This result is important from the practical point of view, since GIL is designed at a larger electrode separation with a utilization factor $\xi < 0.67$, and at pressures $\geqslant 0.4 \text{ MPa}$.

3.3.3 VOLTAGE POLARITY AT SPARKOVER

Table 2 shows the number of sparkovers, which occur at positive polarity for ten trials. Sparkovers at positive polarity occurred only for d=2 mm, where the field was most uniform, and most of them were at the lowest pressure of 0.1 MPa. When d=5 and 10 mm, all the sparkovers took place at negative polarity. This result differs from the sparkover characteristics of SF₆/N₂ reported in [11, 12], where the impulse sparkover voltage is lower for positive than negative polarity.

3.3.4 MAXIMUM ELECTRIC FIELD STRENGTH AT SPARKOVER

Figures 6(a) and (b) indicate the maximum field strength $E_{\rm max}$ at sparkover as a function of gas pressure for SF₆/N₂ and c-C₄F₈/air, re-

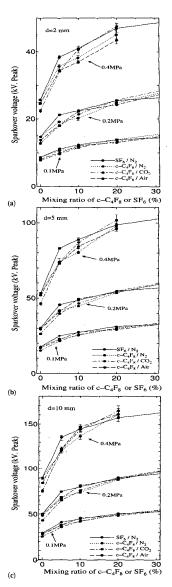


Figure 5. Sparkover characteristics of gas mixtures under quasi-homogeneous field condition, (a) d=2 mm, (b) d=5 mm and (c) d=10 mm.

spectively, where the mixing ratio and the electrode separation are parameters. It increases almost linearly with gas pressure in all cases examined. Note that $E_{\rm max}$ of the gas mixtures with c-C₄F₈ is almost independent of the electrode separation. The other gas mixtures containing c-C₄F₈ investigated in the present study show much the same characteristics. These results mean that the sparkover voltage of a gas mixture with c-C₄F₈ depends strongly on the maximum field strength. Thus, in a quasi-homogeneous gap configuration, we can estimate the sparkover voltage of a gas mixture with a different gap spacing $d, V_s(d)$, by using the experimentally obtained $E_{\rm max}$ and the calculated field utilization factor ξ as

Table 2. Polarity of voltage at sparkover (*d*=2 mm). The mark (-) denotes that all the sparkover occurred in the negative phase.

Gas ratio	0.1	0.2	0.4
SF ₆ (20%) / N ₂	9	-	-
SF ₆ (10%) / N ₂	1	- 1	5
SF ₆ (5%) / N ₂	-	3	1
c-C ₄ F ₈ (20%) / N ₂	5	-	-
c-C ₄ F ₈ (10%) / N ₂	-	4	5
$c-C_4F_8(5\%) / N_2$	6	2	2
c-C ₄ F ₈ (20%) / CO ₂	7	-	-
$c-C_4F_8(10\%) / CO_2$	8	2	-
c-C ₄ F ₈ (5%) / CO ₂	4	7	<u> </u> -
c-C ₄ F ₈ (20%) / air	1	-	1
c-C ₄ F ₈ (10%) / air	6	1	-
c-C ₄ F ₈ (5%) / air	<u>-</u>	1	<u> </u>

$$V_s(d) = \xi dE_{\text{max}} \tag{7}$$

3.4 DISCHARGE CHARACTERISTICS UNDER INHOMOGENEOUS FIELD CONDITION

Figure 7 shows corona inception and sparkover voltages for $c\text{-}C_4F_8/N_2$, $c\text{-}C_4F_8/CO_2$ and SF_6/N_2 , where d=10 mm and the mixing ratio is 10%. The sparkover voltage for $c\text{-}C_4F_8/N_2$ and SF_6/N_2 increases almost linearly with gas pressure, in which the value for $c\text{-}C_4F_8/N_2$ is higher by 1.1 to 1.2× than for SF_6/N_2 . On the other hand, $c\text{-}C_4F_8/CO_2$ shows the highest sparkover voltage for the pressure ranging from 0.1 to 0.2 MPa, but it shows saturation for higher pressures. The corona inception voltage of $c\text{-}C_4F_8/N_2$ is almost the same with or higher than that of SF_6/N_2 , while $c\text{-}C_4F_8/CO_2$ shows the corona inception voltage by 20 to 30% higher than the other mixtures.

It is well known that the sparkover characteristic of an electronegative gas is governed by the corona stabilization effect in an inhomogeneous gap when the gas pressure is comparatively low. We infer that the saturation in the sparkover characteristic of $c\text{-}C_4F_8/CO_2$ is caused by the mitigated stabilization effect. In the case of $c\text{-}C_4F_8/N_2$ and SF_6/N_2 , the corona stabilization is considered still effective under the experimental conditions in this study.

4 DISCUSSION

4.1 OTHER POINTS IN APPLICATION

We here discuss possible disadvantages when applying a gas mixture containing $c\text{-}C_4F_8$. An evident disadvantage of $c\text{-}C_4F_8$ is its high price, several to $\sim 10\times$ higher than SF₆ at present. However, this is considered mainly due to the fact that $c\text{-}C_4F_8$ is not used in large quantities although the use of this gas has recently increased for semiconductor etching. There is enough possibility of reducing the price, when using a large amount of $c\text{-}C_4F_8$ in the power industry.

Another possible disadvantage compared with SF_6 is that $c-C_4F_8$ may generate conductive dusts or particles by decomposition due to the presence of carbon in the molecular structure. They might deteriorate the insulating performance of gas-insulated systems. However, if

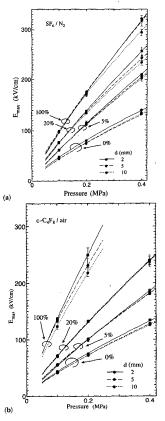


Figure 6. Characteristics of the maximum sparkover field strength as function of the gas pressure under quasi-homogeneous field condition, (a) SF_6/N_2 , (b) $c-C_4F_8/air$.

 $c\text{-}C_4F_8$ is used only for such switchgear components without current-interrupting functions, there exists no or only a rare chance of discharges that might decompose $c\text{-}C_4F_8$. Furthermore, an ordinary gas such as N_2 , CO_2 or air composing the larger portion of a mixture is considered to mitigate the generation of conductive dusts by decomposition.

4.2 SYNERGISM IN SPARKOVER VOLTAGE

An important point in applying a mixture for gas insulation is its nonlinear characteristic (synergism) of the sparkover voltage relative to mixing ratio. The nonlinear characteristic can be expressed by the following empirical formula, which one of the authors has proposed in a previous paper [13].

$$V_m = V_2 + \frac{k(V_1 - V_2)}{[k + (1 - k)C]} \qquad V_1 > V_2 \qquad (8)$$

where V_1 and V_2 are the sparkover voltage of pure gases 1 and 2, respectively, V_m is the sparkover voltage of the gas mixture, k is the mixing ratio and C a constant ($0 \leqslant C \leqslant 1$). When C=1, V_m increases linearly from V_2 to V_1 with k, and nonlinearly when $0 \leqslant C < 1$. In a particular case that C=0, V_m will show a step-like increase with k, as

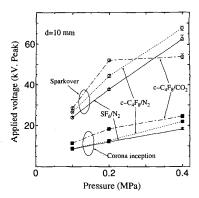


Figure 7. Discharge characteristics of gas mixtures under inhomogeneous field condition.

shown in Figure 8. Thus, we can estimate the degree of nonlinearity, that is, synergism, from the value of C; the smaller C means a more pronounced synergism.

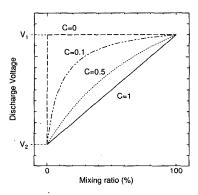


Figure 8. Concept of synergism with the empirical formula.

Figures 4(a), (b) and (c) indicate that the sparkover voltage of a mixture containing c-C₄F₈ increases more or less nonlinearly with gas mixing ratio k. We have selected C so as to give the best fit on these V_m ,k characteristics. Figure 9 demonstrates V_m ,k graphs so obtained for c-C₄F₈/N₂ as an example. It is noteworthy that the above empirical formula with thus selected C very well agrees with the measured characteristic.

Table 3 shows the values of C for all the gas mixtures examined, where C is between 0.1 and 0.17 for SF_6/N_2 , and between 0.26 and 0.53 for the mixtures containing c- C_4F_8 . Figure 10 demonstrates the value of C in relation to Pd for the four kinds of gas mixtures. It can be seen in this Figure that C is represented relative to Pd by a unique curve corresponding to each gas mixture. Although C is lowest for SF_6/N_2 among the four mixtures, it remains almost constant when Pd is increased. On the other hand, C markedly decreases with increasing Pd for gas mixtures with c- C_4F_8 , meaning that larger nonlinearity is expected for the condition of higher Pd in practical gas insulated equipment.

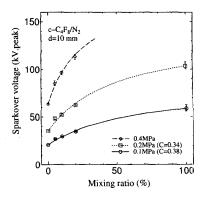


Figure 9. Best fit curves drawn by using the empirical formula.

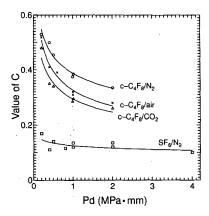


Figure 10. Relations of value of C as function of Pd.

Table 3.	Values	of	C
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d, mm	2			5			10		
Gas mixture	0.1	0.2	0.4	0.1	0.2	0.4	0.1	0.2	0.4
SF ₆ /N ₂	0.17	0.11	1 :				0.14	0.14	0.10
c-C ₄ F ₈ /N ₂	0.53	0.50	-	0.45	0.38	-	0.38	0.34	-
c-C ₄ F ₈ /CO ₂	0.48	0.35	-	0.34	0.29	۱ - ۱	0.29	0.26	- '
c-C ₄ F ₈ /Air	0.52	0.41	'	0.39	0.31	<u> </u>	0.32	0.28	- 1

5 CONCLUSION

We have studied fundamentally the possibility of applying a gas mixture containing c-C₄F₈ as an insulation medium in GIL. The toxicity, the environmental aspects and the recycling loss have been discussed. The proposed gas mixture has advantages such as lower environmental impact and easier liquefaction recovery when compared to a mixture with SF₆. We have measured the 60 Hz discharge characteristics of three mixtures containing c-C₄F₈ in sphere/plane and needle/plane electrode systems. It has been shown that compared with SF₆/N₂, these gas mixtures have reasonably good insulation strength under quasi-homogeneous electric field conditions, and is even superior to SF₆/N₂ under inhomogeneous conditions. The nonlinear characteristic of sparkover voltage in relation to mixing ratio has been discussed by the empirical formula for the gas mixtures. It has been found that nonlinearity, or synergism, increases with Pd for gas mixtures con-

taining c- C_4F_8 . We conclude that a gas mixture containing c- C_4F_8 is a possible candidate alternative to SF_6/N_2 , or even pure SF_6 , especially, from the viewpoint of environmental impact.

ACKNOWLEDGMENT

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