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Positive Synergy of SF_6 and HFO1234ze(E)

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ABSTRACT

We measured mixtures of SF_6 and HFO1234ze(E) ($C_3H_2F_4$) and determined the density-reduced critical electric field strength $(E/N)_{crit}$ in a pulsed Townsend experiment. HFO1234ze(E) is an environmentally friendly alternative to SF_6 since contrary to SF_6, its global warming potential is negligible. While SF_6 is unsurpassed in its efficiency of attaching thermal electrons, HFO1234ze(E) appears to moderate electron energies very efficiently. Electron attachment to HFO1234ze(E), on the other hand, is much weaker. We find a positive synergism in SF_6/HFO1234ze(E) mixtures in the sense that the mixture of both compounds has a higher electric field strength than each gas separately. A maximum $(E/N)_{crit}$ of 425 Td is reached for a mixture of 40% SF_6 and 60% HFO1234ze(E). A mixture of 90% HFO1234ze(E) and 10% SF_6 has a critical electric field strength of 360 Td, similar to pure SF_6. We attribute this effect to the high efficiency of HFO1234ze(E) in moderating down the electron energies, which enhances electron attachment to SF_6 and reduces electron impact ionization.

Index Terms — **gas discharges, gas insulation, SF_6, alternative gases**

1 INTRODUCTION

SF_6 is the current standard in industry for gaseous electrical insulation in medium and high voltage equipment [1]. Due to its high global warming potential (GWP) of 23,800 (compared to CO_2 at equal mass on a time horizon of 100 years) it is desirable to find gases or gas mixtures which have a lower GWP, while retaining the high electric strength of SF_6.

There is a long history of mixing SF_6 with other gases, in order to optimize their electric strength. Some of these mixtures are known to have higher electric strength than either of their components taken separately; this effect is often termed "positive synergy" [2]. An overview is given by Hunter and Christophorou in [3], featuring gases such as c-C_4F_8, SO_2, C_3F_8, OCS, and C_3F_6. However, all of these compounds are either highly toxic or have a high GWP themselves.

The main motivation for these investigations in the 1980s was mostly the improvement of the insulation gas, and the moderate benefit might not have outweighed the additional cost, considering for instance the more complicated gas handling. Today, the replacement of at least a large fraction of SF6 may become imperative, due to the strong global warming effect and technically unavoidable leakage. The search for suitable mixing gases can also be extended in new directions, as for instance partially fluorinated compounds, which are now commercially available and produced in large scale.

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In a recent publication on measurements of the compound R1225ye(Z) [4], we revisited literature on the three structurally similar compounds HFO1234ze(E), R1225ye(Z) (C_3HF_5) and C_3F_6 (boiling points at 100kPa of -19°C [5], -19.6°C [6], -29.6°C [7], respectively), all featuring a C=C-C backbone. While C_3F_6 is fully fluorinated, one and two hydrogen atoms remain in R1225ye(Z) and HFO1234ze(E). Despite the seemingly small difference, C_3F_6 is contrary to HFO1234ze(E) and R1225ye(Z) highly toxic [8].

Previous measurements of C3F6 [9], and our measurements of R1225ye(Z) [4] and HFO1234ze(E) [10] showed that these three compounds feature a lower ionization rate coefficient compared to SF_6 by a factor of 3 to 10 in their respective measurement range around the electric field strength. This presumably holds quite generally over a wider E/N range, and is very desirable for a gas intended for gaseous electrical insulation. The likely explanation is a very efficient moderation of electrons and thus a lower average electron energy in HFO1234ze(E) and R1225ye(Z) discharges, compared to SF_6 at equal field strength.

However, since also the attachment rate coefficient at critical field of these compounds is one to two orders of magnitudes lower than that of SF_6, their critical electric field strength is overall lower. HFO1234ze(E) and R1225ye(Z) owe their moderately high critical field strength not to a good electron attachment capability, but rather to their excellent electron energy moderation.

Besides dissociative attachment, C_3F_6 is known to exhibit a strong three-body electron attachment processes, leading to a pressure dependency of its critical field strength. We found a similar, yet weaker, pressure dependency in both HFO1234ze(E) [10] and R1225ye(Z) [4], indicating a much lower efficiency of the presumed three-body attachment process. We concluded that at

relevant application pressure this three-body component would have a measurable, yet limited effect.

2 METHOD

We use a pulsed Townsend experiment as described in [11], yielding characteristic current pulses from which we obtain the effective ionization rate coefficient. The measurement is performed for each gas mixture over a large range of reduced field strength values E/N. From this, we deduce the density-reduced field strength $(E/N)_{crit}$. The methods of analysis are described in previous works [10, 15].

3 RESULTS

The critical electric field strength of SF_6/HFO1234ze(E) mixtures is shown in Figure 1. Results at low pressures of 360-480 Pa (blue circles) agree hereby well with measurements at higher pressures of 4-12 kPa (red crosses).

The obtained positive synergy is striking, and we measure a density-reduced critical field strength $(E/N)_{crit}$ of 425 Td for the optimal mixture of 60% HFO1234ze(E) and 40% SF_6, while at roughly 10% of SF_6 and 90% of HFO1234ze(E) the same strength as pure SF_6 is obtained.

Breakdown measurements in SF6/C3F6 from [2] (black squares) show a very similar trend. The maximum, however, is less pronounced.

The synergy coefficient, i.e. the ratio between the actual $(E/N)_{crit}$ and the linear interpolation of the strength of the pure compounds by mole fraction (assuming 360Td for SF_6 and 200Td for HFO1234ze(E)), is given as the green dashed line. The most significant increase is observed at 10-20% SF_6, where the coefficient reaches values around 1.75.

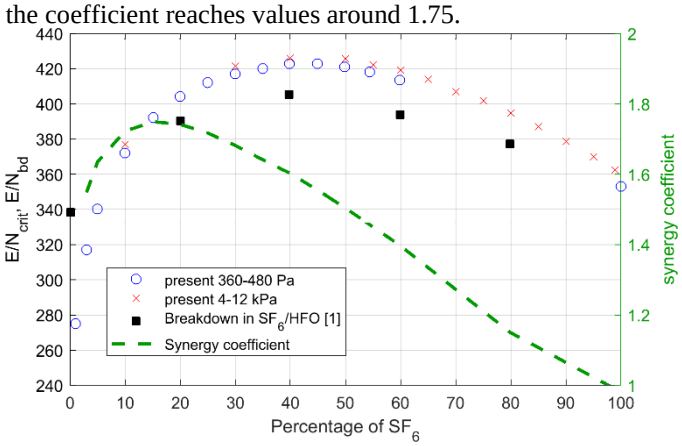

Figure 1. Critical electric field strength $(E/N)_{crit}$ of mixtures of SF_6 with HFO1234ze(E). In blue circles, a measurement series at pressures of 360-480 Pa is shown. In red crosses, we used higher, variable pressures from 4-12 kPa. In black squares, breakdown results $(E/N)_{bd}$ of James *et al* [2] for the mixture of SF_6 with C_3F_6 are given at a pressure of 67 kPa. The synergy coefficient is plotted with scale on the right-hand side.

4 DISCUSSION

The present results confirm that positive synergism occurs in SF_6/HFO1234ze(E) mixtures, in direct analogy to older results for SF_6/C_3F_6 mixtures. For the latter it was suggested that three-body attachment [8] mechanisms were the origin of the positive synergy [3], yet this explanation does not seem to fit with HFO1234ze(E), for which the three-body electron attachment is comparatively weak. Furthermore, we observe the positive synergy at low pressures, where pressure dependent effects are negligible.

Our results rather indicate that the observed positive synergy is simply due to the good moderation of electron energies by compounds with C=C-C backbone. In a gaseous electrical discharge, electrons constantly gain kinetic energy by acceleration in the electric field, and lose energy in collisions with neutral molecules. Inelastic and elastic processes transfer some of the electron energy to the gas, and thus moderate the electron energy down. The balance between acceleration and deceleration results in an electron energy distribution function (eedf), which depends only on the applied reduced electric field strength *E/N*, and the gas properties. In particular, the inelastic processes such as vibrational excitation of a molecule by electron impact, differ strongly in between gases. It appears now that the eedf of gases with C=C double bonds is particularly low. Fitting with this, the early comparative studies of insulation gases [13, 14] suggested an improved performance of gases with double bond compared to saturated gases. When mixing such gases with SF_6, the lower mole fraction of SF_6 is over-compensated by lower mean electron energies, leading to both lower ionization and a higher attachment efficiency of SF_6 molecules.

5 CONCLUSION

A significant reduction of SF_6 in medium pressure equipment could be achieved without loss of electric strength, by replacing a large fraction of SF_6 with HFO1234ze(E). This could have an immediate environmental benefit. In high voltage equipment, the insulation gas is utilized at higher pressures, and the moderate vapor pressure of HFO1234ze(E) limits the amount of SF_6 that could be replaced [12]. Here, a further candidate for a moderating gas with C=C-C backbone could be the isomer HFO1234yf, which has a better (i.e. lower) boiling point of -30ºC, yet higher flammability compared to HFO1234ze(E).

It further seems worthwhile to also investigate synergism of HFO1234ze(E) with alternative gases such as C_4F_7N and $C_5F_{10}O$. Due to the low boiling points of these compounds, they are deployed in mixture with either synthetic air or CO2, and an additional moderating gas such as HFO1234ze(E) in the mix would not necessarily complicate gas handling by much.

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