

PRESSURE-DEPENDENT ELECTRON ATTACHMENT AND BREAKDOWN STRENGTHS OF UNITARY GASES, AND SYNERGISM OF BINARY GAS MIXTURES: A RELATIONSHIP

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ABSTRACT

The relationship between the pressure-dependent electron attachment rate constants (k) which have been observed in 1-C₃F₆ and in several perfluoroalkanes, and the uniform field breakdown strengths $(E/N)_{lim}$ in these gases is discussed. Measurements of the pressure dependence of k of OCS in a buffer gas of Ar are presented and the possible pressure dependence of $(E/N)_{lim}$ in OCS is discussed.

Uniform field breakdown measurements have been performed in C₃F₈, n-C₄F₁₀, and SO₂ over a range of gas pressures ($3 \leq P_T \leq 290$ kPa) and are reported. All three molecules have been found to possess pressure-dependent $(E/N)_{lim}$ values.

The various types of synergistic behavior which have been observed in binary gas dielectric mixtures are summarized and discussed. A new mechanism is outlined which can explain the synergism observed in several gas mixtures where the $(E/N)_{lim}$ values of the mixtures are greater than those of the individual gas constituents. Model calculations are presented which support this mechanism, and can be used to explain the pressure-dependent synergistic effects which have been reported in 1-C₃F₆/SF₆ gas mixtures.

KEYWORDS

Electron attachment and ionization coefficients; breakdown field strength; high voltage insulation; synergism in dielectric gas mixtures.

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INTRODUCTION

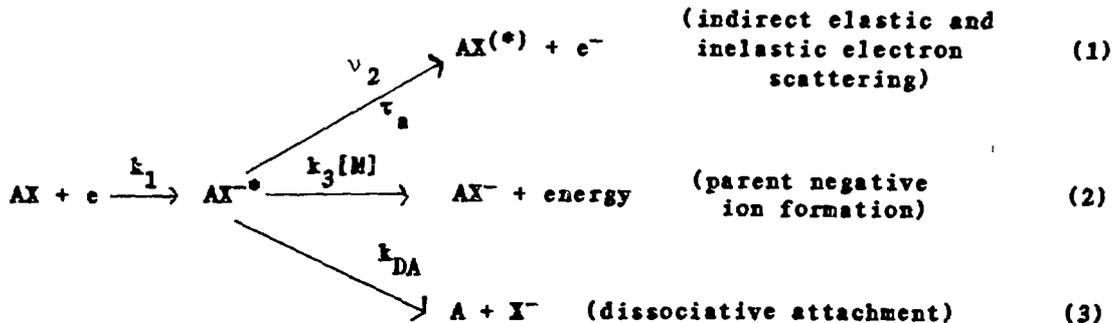
Several authors have analyzed the influences that electron attachment and ionization have on the uniform field breakdown strength, $(E/N)_{lim}$, of dielectric gases (e.g., Christophorou and coworkers, 1979, 1981b). McCorkle and coworkers (1984) have further demonstrated the importance of high electron attachment rate constants in achieving high dielectric strength.

The relationship between the pressure dependence of the electron attachment rate constant, k_a , and $(E/N)_{lim}$ of electronegative gases has been previously outlined (Hunter and coworkers, 1982). It was indicated that the increase in $(E/N)_{lim}$ which had been observed for $1-C_3F_6$ (Aschwanden and Biasiutti, 1981; Biasiutti, 1982; Hunter and coworkers, 1982) and C_3F_8 (Biasiutti, 1982) was due to an increase in stable negative ion production with increasing gas pressure resulting from stabilization of the parent negative ions in C_3F_8 , and the dimeric parent negative ions in $1-C_3F_6$, over the pressure range for which such stabilization processes are significant. In this paper, we report on measurements of k_a as a function of mean electron energy, $\langle \epsilon \rangle$, in OCS which has also been found to possess pressure-dependent electron attachment processes similar to those which have been observed in C_3F_8 and $n-C_4F_{10}$ (Hunter and coworkers, 1982; Hunter and Christophorou, 1984a).

Uniform field breakdown measurements have been performed as a function of gas pressure in C_3F_8 , $n-C_4F_{10}$, and SO_2 by Nakanishi and coworkers (1984). All these molecules have been found to exhibit a marked increase in $(E/N)_{lim}$ (corrected for gas compressibility) with increasing gas pressure which is believed to result from the pressure-dependent electron attachment processes in these gases over the pressure range of the $(E/N)_{lim}$ measurements. These findings and the influence that a pressure dependence in k_a has on the dielectric strength of binary gas mixtures, where one or both of the component gases possess pressure-dependent electron attachment processes at atmospheric pressures, are discussed.

ELECTRON ATTACHMENT STUDIES

Recently, the gases C_3F_8 , $n-C_4F_{10}$, and $1-C_3F_6$ have been observed to possess pressure-dependent electron attachment processes in the mean electron energy range ($1 \leq \langle \epsilon \rangle \leq 5$ eV) in a high pressure buffer gas ($0.1 \leq P_T \leq 1.5$ MPa) of either Ar or N_2 (Hunter and coworkers, 1982, 1983; McCorkle and coworkers, 1983; Hunter and Christophorou, 1984a). Electron attachment to C_3F_8 and $n-C_4F_{10}$ has been interpreted as the result of stabilization of the excited parent negative ion in the following reaction scheme:



An electron associatively attaches to AX with a rate constant k_1 to form a transient parent negative ion, which can either autoionize with a decay constant ν_2 and a characteristic lifetime τ_a , be collisionally stabilized by another molecule at a number density $[M]^a$ (usually the buffer gas molecule in these experiments) with a rate constant k_3 to form a stable parent negative ion, or fragment to form negative ions and neutral radicals with a rate constant k_{DA} . In the absence of dissociative attachment and collisional detachment, competition between autoionization (Reaction 1) and collisional stabilization (Reaction 2) leads to a dependence of the observed attachment rate constant k_a on the total gas pressure (or number density of the buffer gas $[M]$) of the form:

$$k_a = \frac{k_1 k_3 [M]}{(\nu_2 + k_3 [M])} \quad (4)$$

Electron attachment to $1-C_3F_6$ is considerably more complex, and has been observed to be extremely dependent on gas temperature (k_a decreases as T increases for $\langle \epsilon \rangle \geq 0.1$ eV and $T > 300K$; McCorkle and coworkers, 1983) and on the partial $1-C_3F_6$ gas pressure as well as the total gas pressure (Hunter and coworkers, 1983). These observations were interpreted by Hunter and coworkers (1983) to be due to the pressure dependent formation and stabilization of the dimeric anion $(1-C_3F_6)_2^{-*}$ in the high pressure buffer gas.

Electron attachment measurements to OCS in Ar as a buffer gas have also shown that this molecule possesses a pressure-dependent k_a ($\langle \epsilon \rangle$), similar in magnitude and degree to that observed for C_3F_8 and $n-C_4F_{10}$ (Fig. 1). The pressure dependence of k_a for this molecule can be modeled--as those for C_3F_8 and $n-C_4F_{10}$ --using the reaction scheme (1) to (4). These results indicate that the $(E/N)_{lim}^a$ of OCS will be dependent on the gas pressure up to perhaps gas pressures of the order of one atmosphere. These measurements have not been performed at this time.

BREAKDOWN MEASUREMENTS

The observation that k_a ($\langle \epsilon \rangle$) in C_3F_8 , $n-C_4F_{10}$, and $1-C_3F_6$ are dependent on gas pressure led to the prediction that the $(E/N)_{lim}^a$ values in these gases are also pressure dependent, provided that the ionization rate constants, k_i , in these gases are independent of gas pressure, and that $(E/N)_{lim}^a$ is defined as the E/N value for which $(k_i - k_a) = 0$ (Hunter and coworkers, 1982). Measurements of $(E/N)_{lim}^a$ by Aschwanden and Biasiutti (1981), Biasiutti (1982), and Hunter and coworkers (1982) in $1-C_3F_6$ and by Biasiutti (1982) in C_3F_8 have indeed shown that the $(E/N)_{lim}^a$ of these gases is strongly dependent on gas pressure, particularly below 1 atm. The $(E/N)_{lim}^a$ measurements in C_3F_8 were repeated by Nakanishi and coworkers (1984) and extended to $n-C_4F_{10}$ over the gas pressure range $3 \leq P_T \leq 300$ kPa. They found that both gases exhibit a considerable variation in $(E/N)_{lim}^a$ with gas pressure even after the measurements were corrected for gas compressibility (Fig. 2). Previous electron attachment studies have shown that pressure dependent electron attachment processes also exist for N_2O (e.g., Phelps and Voshall, 1968; Chaney and Christophorou, 1969; Parkes, 1972; Shimamori and Fessenden, 1979) and SO_2 (Rademacher and coworkers, 1975; Lakdawala and Moruzzi, 1981). Dutton and coworkers (1973) and Biasiutti (1982) have shown that the $(E/N)_{lim}^a$ in N_2O is pressure dependent. Nakanishi and coworkers (1984) have measured the $(E/N)_{lim}^a$ in SO_2 over the pressure range $(7.8 \leq P_T \leq 290)$ kPa and have also found that $(E/N)_{lim}^a$ is dependent on gas pressure after correcting for gas compressibility. These measurements are given in Fig. 2 and show that $(E/N)_{lim}^a$ saturates in $n-C_4F_{10}$ at ~ 100 kPa, in C_3F_8 at ~ 150 kPa, and in SO_2 at ≥ 200 kPa. These measurements are summarized in Fig. 3 along with the previous pressure-dependent field strength measurements in these gases and in $1-C_3F_6$ and N_2O .

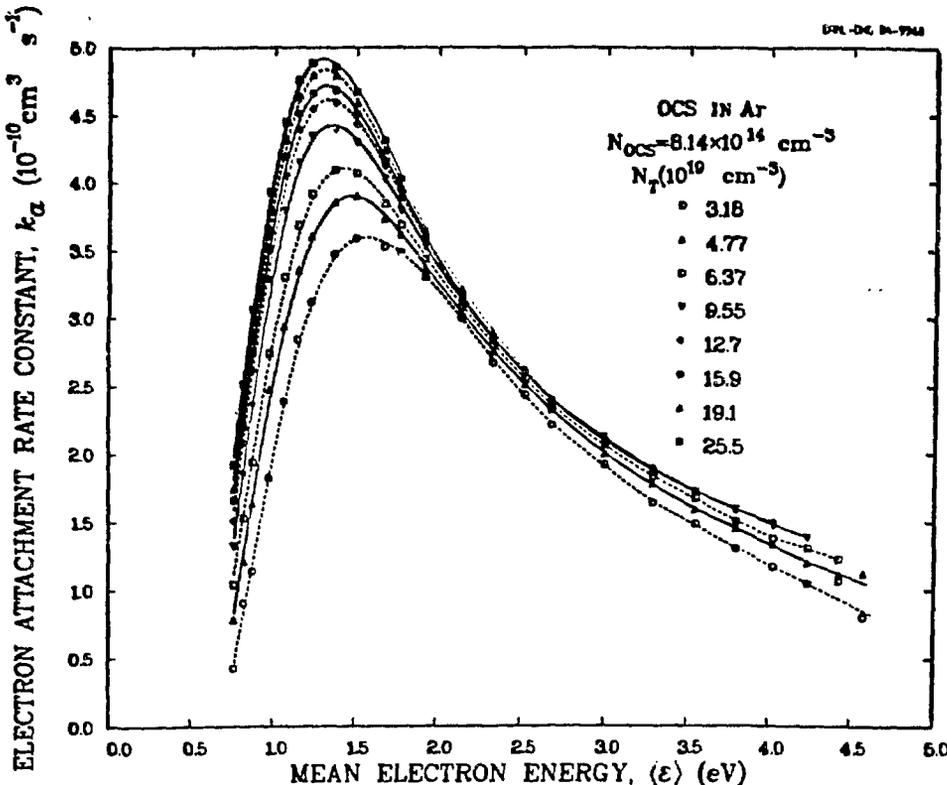


Fig. 1. Electron attachment rate constant, k_a , in OCS measured as a function of mean electron energy, $\langle \epsilon \rangle$, and total gas number density, N_T , at a fixed partial OCS number density, N_{OCS} , in a buffer gas of argon.

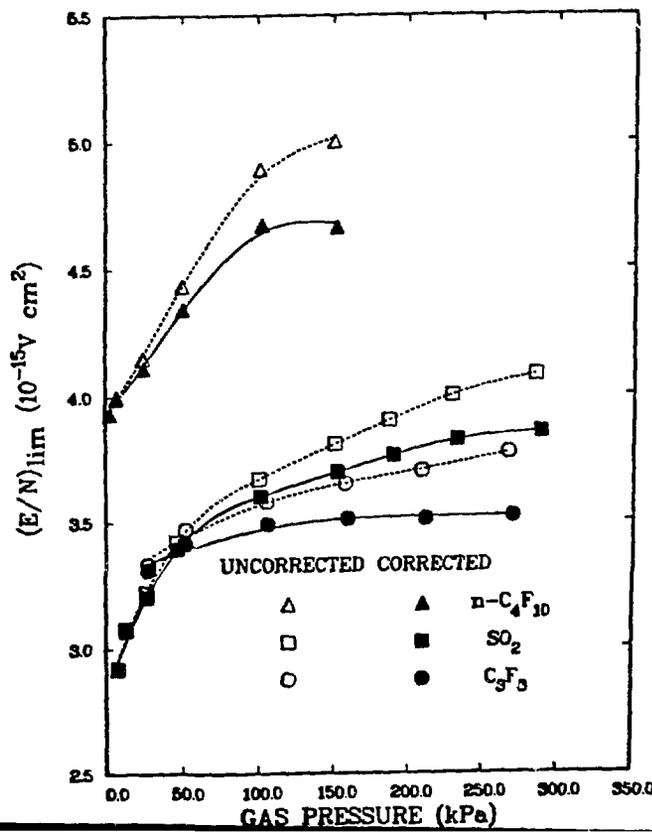


Fig. 2. Uniform field breakdown strength, $(E/N)_{\text{lim}}$, and $(E/N)_{\text{lim}} Z(P)$ of SO_2 , C_3F_8 , and $n\text{-C}_4\text{F}_{10}$ as a function of gas pressure, P , or $P/Z(P)$. Open symbols: uncorrected for gas compressibility $Z(P)$. Solid symbols: corrected for compressibility. (From Nakanishi and coworkers, 1984).

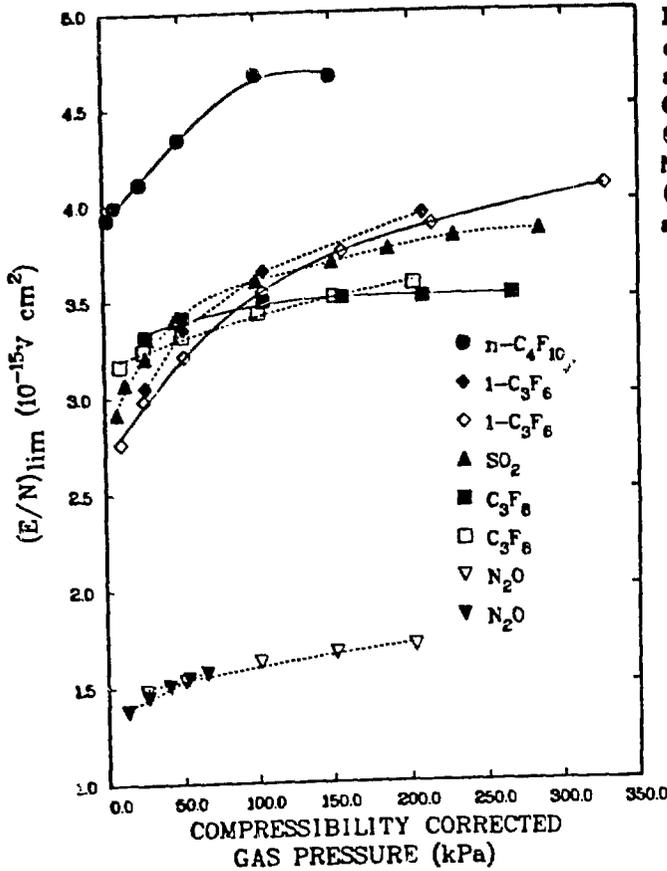


Fig. 9. The gas compressibility corrected $(E/N)_{lim}$ values of Nakanishi and coworkers (1984) in $n-C_4F_{10}$ - ●, C_3F_8 - ■, and SO_2 - ▲; of Bisiutti (1982) in C_3F_8 - □, $1-C_3F_6$ - ◇, and N_2O - ▽; of Hunter and coworkers (1982) in $1-C_3F_6$ - ◆; and of Dutton and coworkers (1973) in N_2O - ∇.

In contrast to C_3F_8 , $n-C_4F_{10}$, and $1-C_3F_6$ where parent and dimeric parent negative ion stabilization as a function of gas pressure is thought to be responsible for the pressure variation in $(E/N)_{lim}$, the apparent variation in k_a with pressure in N_2O and SO_2 may be also influenced by the competition between the formation of stable anions and the destruction of dissociative-attachment ions via ion-molecule reactions (Parkes, 1972; Dutton and coworkers, 1975a,b; Doussot and coworkers, 1982). Three-body parent negative ion formation occurs at near thermal energies in SO_2 (Rademacher and coworkers, 1975; Lakdawala and Moruzzi, 1981) but is insignificant at higher energies (20.6 eV).

SYNERGISTIC EFFECTS IN GAS MIXTURES

When an electronegative gas is mixed with another gas (electronegative or not), the $(E/N)_{lim}$ of the resultant mixture can vary considerably, depending on the electron scattering and ion-molecule reaction processes occurring within the electrically stressed mixture. Three basic types of behavior for the variation of the $(E/N)_{lim}$ with relative gas concentration have been identified and are illustrated in Fig. 4 with reference to three specific gas mixtures: $1-C_3F_6/SF_6$ (James and coworkers, 1980), N_2/SF_6 (James and coworkers, 1978), and CF_4/SF_6 (James and coworkers, 1980). The fundamental processes controlling the discharge characteristics in these mixtures have been described in detail by Christophorou and coworkers (1981a, 1982) and Chantry and Wootton (1981). The simplest case given in Fig. 4 is that labeled as curve C, and occurs when the breakdown strength of the gas mixture is very nearly given by the sum of the partial pressure weighted breakdown strengths of the constituent component gases. This behavior, where each gas acts independently of the other, has been observed in CF_4/SF_6 , $c-C_4F_8/SF_6$, and $2-C_4F_6/SF_6$ gas mixtures among others (James and coworkers, 1978, 1980; Wootton and coworkers, 1980).

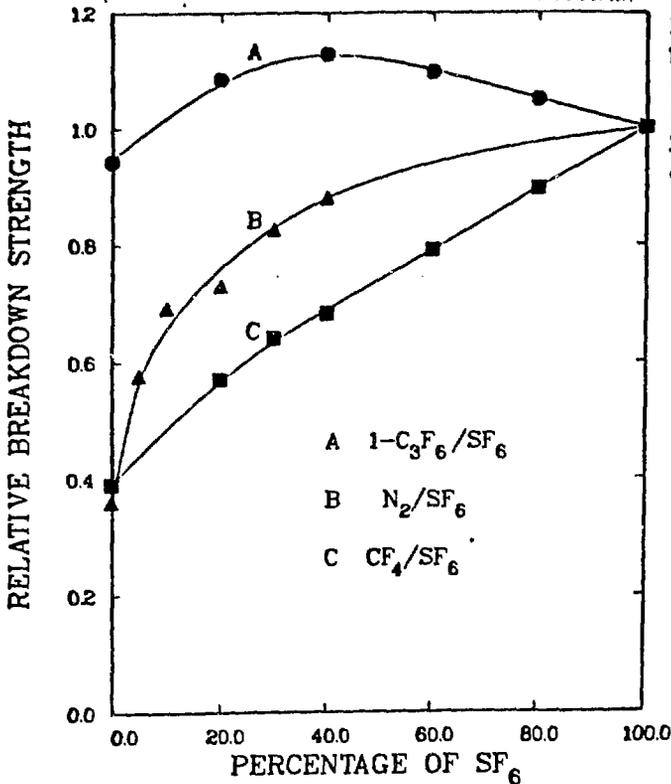


Fig. 4. Relative uniform field breakdown strengths of (A) 1-C₃F₆/SF₆ (James and coworkers, 1980), (B) N₂/SF₆ (James and coworkers, 1978), and (C) CF₄/SF₆ (James and coworkers, 1980) (P_T = 66.7 kPa).

A second type of behavior (curve labeled B in Fig. 4) shows a marked increase in $(E/N)_{lim}$ in the gas mixture when a highly attaching gas whose k_a is pressure independent is admixed into a weakly attaching (whose k_a is also pressure independent) or a nonattaching gas. The increase in $(E/N)_{lim}$ of the resultant gas mixture is much larger than that predicted by summing the partial pressure weighted $(E/N)_{lim}$ of the individual gas components. This effect has been termed a synergistic effect and, as has been shown by Kline and coworkers (1979) and Chantry and Wootton (1981), can be predicted using the Wieland approximation (Wieland, 1973). The $(E/N)_{lim}$ in SF₆/N₂ gas mixtures has been accurately predicted using this technique, which involves summing the partial pressure weighted apparent ionization coefficients $\bar{\alpha}$ (defined as $\bar{\alpha} = \alpha - \eta$, where α and η are the ionization and attachment coefficients) of the individual gas constituents. The most reliable approach for predicting the breakdown strengths of these gas mixtures is, of course, by Boltzmann equation analysis. However, this approach requires a detailed knowledge of the elastic, inelastic, and electron attachment and ionization cross sections of the individual gases if the accuracy of the technique is to be fully exploited. Such knowledge is at present limited, or even nonexistent, for most mixtures of interest.

A third type of synergistic behavior observed in a few gas mixtures occurs when the $(E/N)_{lim}$ of the mixture is larger than that of the individual component gases (curve A in Fig. 4). This effect, which we define as positive synergism, cannot be predicted using the Wieland approximation, nor has it been demonstrated to occur in any Boltzmann equation analysis of the breakdown behavior in gas mixtures. We propose a mechanism which can account for this behavior based on the observation that in gas mixtures where this effect occurs, the electron attaching properties of one or both of the constituent gases are a function of the total gas pressure of the mixture.

The variations in the relative breakdown strength with gas composition in gas mixtures in which this phenomenon is known to occur are summarized in Fig. 5. These mixtures include SO₂/SF₆ (Wootton and coworkers, 1980), OCS/SF₆ (Christophorou and coworkers, 1979; Wootton, 1982), C₂F₈/SF₆ (Wootton, 1982), 1-C₃F₆/SO₂ (Wootton and coworkers, 1980), and 1-C₃F₆/c-C₄F₈ (James and coworkers,

1980). An extensive series of breakdown measurements have been performed in $1-C_3F_6/SF_6$ gas mixtures (James and coworkers, 1980; Wootton and coworkers, 1980; Biasiutti and coworkers, 1983) (Fig. 6) and have shown that the breakdown strength in these mixtures is strongly dependent on the total gas pressure, and that the positive synergism observed at lower gas pressure ($P_T \sim 68$ kPa) decreases in magnitude with increasing gas pressure.

We propose that the positive synergistic effects which have been observed in these binary gas mixtures can be explained as resulting from an increase in stable negative ion production in the gas mixture as compared to the pure component gases. Let us assume that in a binary gas mixture, one component, AX, attaches electrons via a three-body attachment process and another, BC, attaches electrons dissociatively. The total electron attachment in the gas mixture at a given E/N value is the sum of the dissociative electron attachment to BC and the pressure-dependent electron attachment to AX, which is given by the reaction scheme (1), (2), and (4). This scheme assumes that dissociative attachment to AX is negligible and that the stabilizing molecule, M, is either AX or BC. Dissociative attachment to AX can easily be included in this reaction scheme, and has been excluded only for simplicity. The rate of electron attachment in the mixture due to AX alone is then,

$$k_{AX} = \frac{k_1(k_3[AX] + k_N[BC])}{(v_2 + k_3[AX] + k_N[BC])} \quad (5)$$

where k_N is the rate constant for collisional stabilization of AX^{-*} in reaction (2) by BC. The total rate of electron attachment, including the rate constant, k_{DA} , for dissociative attachment to BC for the mixture is

$$k_A = \frac{(k_{AX}[AX] + k_{DA}[BC])}{N_T} \quad (6)$$

where $N_T = [AX] + [BC]$.

Positive synergism can result from an increase in the rate of stable negative ion production which can occur when k_1 is larger than k_3 ; i.e., when the rate, k_N , of collisional stabilization of AX^{-*} by BC is greater than the rate, k_3 , of collisional stabilization of AX^{-*} by AX. Similar reaction schemes can be envisioned incorporating collisional detachment (Dutton and coworkers, 1975a,b) and charge transfer reactions, which may also result in enhanced stable negative ion formation, if the resultant anions are more stable (and hence the rate constant for detachment is decreased) than AX^- . The validity of the proposed reaction scheme [Eqs. (4)-(6)] has been tested by performing a model calculation of the dependence of the positive synergism on the relative composition and total pressure for $1-C_3F_6/SF_6$ gas mixtures. The following relations were assumed for this system and the calculations shown in Fig. 7: $k_1 = 1.43 \times k_N$; $v_2/k_3 = 10^{19} \text{ cm}^3 = N_c$; $v_2 = 10^{10} \text{ s}^{-1}$; and $k_N = 5 \times k_3$ (N_c is the critical number density; i.e., the number density of AX at which the rate of autoionization of AX^{-*} is equal to the rate of collisional stabilization). The values of v_2/k_3 and v_2 were chosen to approximately model the pressure dependence of the electron attachment processes at $\langle \epsilon \rangle \sim 3$ eV in $1-C_3F_6$ (Hunter and coworkers, 1983), and the ratios k_1/k_N and k_N/k_3 were chosen to model the variation of $(E/N)_{lim}$ in the $1-C_3F_6/SF_6$ gas mixtures given in Fig. 6. It can be seen in Fig. 7 that as $N_T \rightarrow \infty$, the synergistic effects disappear and the $(E/N)_{lim}$ of the gas mixture becomes independent of gas pressure (i.e., independent of k_3 and k_N).

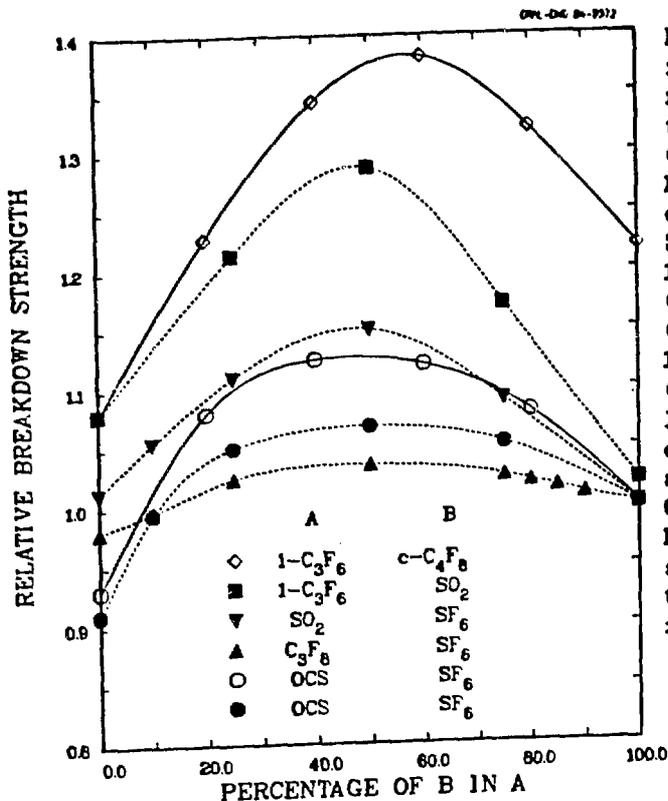


Fig. 5. Uniform and quasi-uniform field breakdown strengths of the following gas mixtures, relative to that of SF₆ (≡1). 1-C₃F₆/c-C₄F₈ - ◇ (James and coworkers, 1980) (P_T = 66.7 kPa); 1-C₃F₆/SO₂ - ■ (Wootton and coworkers, 1980) (P_T = 152 kPa); SO₂/SF₆ - ▼ (Wootton and coworkers, 1980) (P_T = 152 kPa); OCS/SF₆ - ○ (Christophorou and coworkers, 1979) (P_T = 66.7 kPa); OCS/SF₆ - ● (Wootton, 1982) (P_T = 152 kPa); C₃F₈/SF₆ - ▲ (Wootton, 1982) (P_T = 152 kPa). The larger synergistic effect, which is observed for the 1-C₃F₆/c-C₄F₈ mixture as compared to the 1-C₃F₆/SO₂ and 1-C₃F₆/SF₆ mixtures, is due to the higher rate constant, k_N, for stabilization of 1-C₃F₆⁻ by c-C₄F₈ than that for SF₆ and SO₂ as outlined in the text.

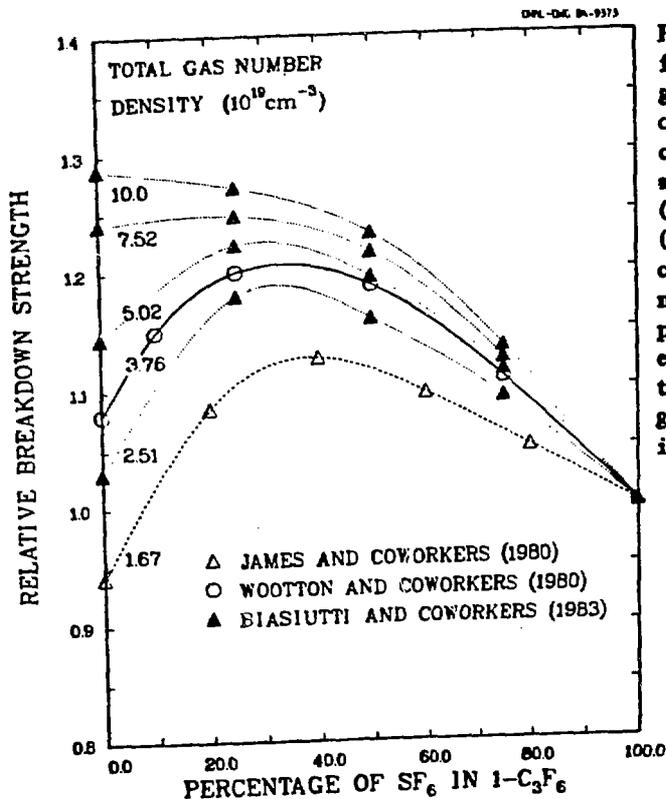


Fig. 6. Dependence of the uniform field breakdown strength in 1-C₃F₆/SF₆ gas mixtures, relative to that of SF₆, on the partial pressure of the constituent gases and total pressure, as measured by James and coworkers (1980) - △, Wootton and coworkers (1980) - ○, and Biasiutti and coworkers (1983) - ▲. The lines have no significance. The reaction scheme proposed in this paper can be used to explain the apparent differences in the breakdown strengths in 1-C₃F₆/SF₆ gas mixtures which had been observed in these studies.

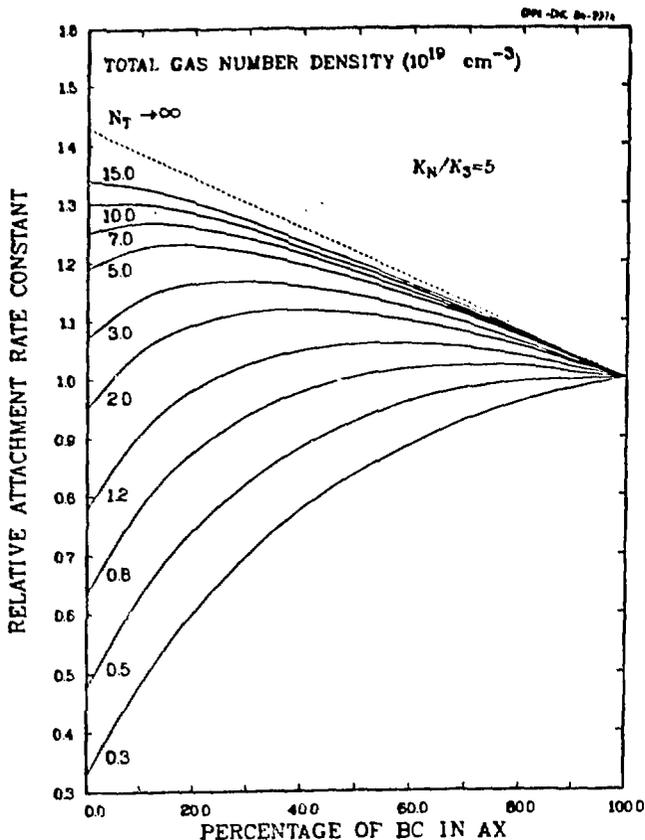


Fig. 7. Model calculations of the pressure dependence of the electron attachment rate constant at an E/N value close to breakdown of gas mixtures, with one gas, AX, possessing a pressure-dependent breakdown field strength (e.g., $1\text{-C}_3\text{F}_6$, C_3F_8) in another attaching gas, BC (e.g., SF_6 , $c\text{-C}_4\text{F}_8$), with a high rate constant for collisionally stabilizing AX^- . The numerical values of the rate constants are given in the text and were chosen to approximately model the electron attachment and breakdown field strength characteristics in $1\text{-C}_3\text{F}_6$ and $1\text{-C}_3\text{F}_6/\text{SF}_6$ gas mixtures. In this case $k_N = 5 \times k_3$.

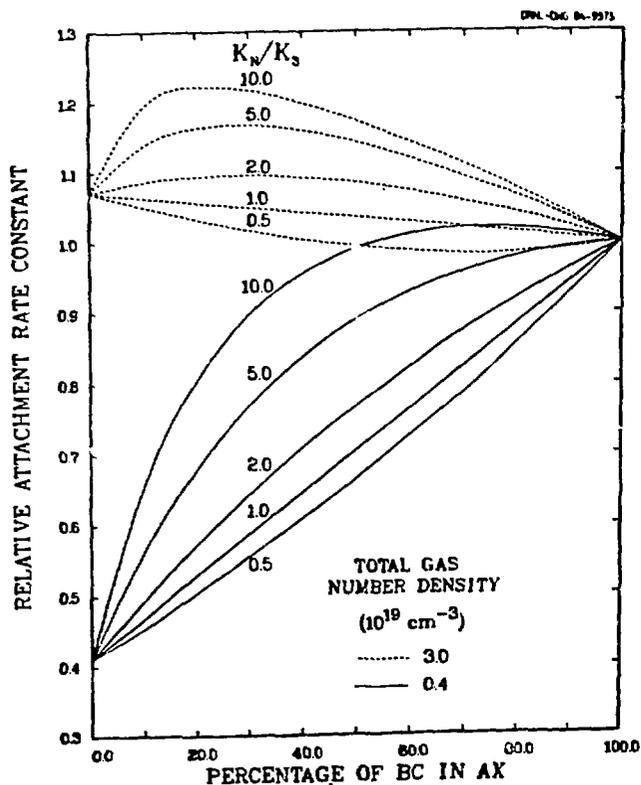


Fig. 8. Model calculations for the conditions given in Fig. 7 except at two different total gas number densities, and for different relative rate constants of stable negative ion formation enhancement, i.e., different values of the ratio k_N/k_3 . It is assumed that electron attachment in BC is independent of gas pressure.

The effect on the rate of attachment in the mixture by changes in the relative rates of stable negative ion production (i.e., the ratio k_N/k_3) at two total number densities is shown in Fig. 8. It may be noted that from the calculations given in Fig. 8 negative synergistic effects may be observed when the rate of stable negative ion formation is less in the mixture than for pure AX (i.e., $k_N < k_3$). Such effects have not been observed in uniform fields to the authors' knowledge.

To make a comparison between the pressure variation in the attachment rate constants predicted in Figs. 7 and 8 and the experimental breakdown measurements given in Figs. 5 and 6, it is necessary to assume that the electron energy distribution function in the gas mixture under electrical stress is unaltered by changes in the relative amounts of one gas in the other at a constant E/N value. This assumption implies that without the aforementioned additional stable negative ion production mechanisms, the rate of negative ion production in the gas mixture will be a simple linear addition of the rates of electron attachment of the component gases. This situation is shown in Fig. 8 when $k_N/k_3 = 1$. A further implication of this assumption is that the rate of ionization in the mixture is a simple linear addition of the rates of the component gases. The calculations in Figs. 7 and 8 have been made at an E/N value close to breakdown while the dielectric strength of a gas mixture was defined as the E/N at which $(\alpha - \eta)/N_T = 0$ (or $k_1 - k_a = 0$), where α , k_1 , and k_a are the ionization coefficient and ionization and attachment rate constants, respectively, and η is the attachment coefficient. In general, as the E/N near breakdown for a particular gas mixture is increased, η/N decreases and α/N increases, and hence the enhancement of k_a in the gas mixture will tend to be masked in the breakdown measurements. Nevertheless, a comparison between the results given in Figs. 5 to 8 clearly indicates that the positive synergism observed in a number of gas mixtures can be explained using the reaction scheme outlined above. A more detailed account of these investigations will appear elsewhere (Hunter and Christophorou, 1984b).

DISCUSSION

The work outlined in this paper has demonstrated the relationship between the pressure-dependent electron attachment rate constants and the pressure dependent $(E/N)_{lim}$ values observed in several electronegative gases. The $(E/N)_{lim}$ increases with gas pressure due to the increase in the rate of electron attachment with gas pressure. Further, the positive synergism that has been observed to occur in binary gas mixtures comprised of one of these gases along with another gas possessing only non-pressure-dependent electron attachment processes can also be explained in terms of the pressure dependence in k_a of one of the component gases.

The observation of positive synergism requires that (i) at least one of the electronegative components of the mixture possesses pressure-dependent electron attachment processes; (ii) the other component has a higher rate constant for stabilizing transient anions, or alternatively, can reduce collisional detachment processes in AX by charge transfer or ion-molecule reactions to form stable negative ions; and (iii) in the total gas pressure range of interest, the dielectric strengths of the two components are not too dissimilar.

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