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IMPROVED UNITARY AND MULTICOMPONENT GASEOUS INSULATORS

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DISCLAIMER

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Improved unitary and multicomponent gaseous insulators are systematically designed according to detailed knowledge of fundamental electron-molecule interactions. Knowledge of the electron attachment and electron slowing-down properties of dielectric gases/mixtures as functions of electron energy is shown to be especially significant. On the basis of such knowledge it is possible to improve the gaseous dielectric's breakdown strength by effectively controlling the numbers and energies of the electrons present. Several unitary (e.g., C_4F_6, c-C_4F_8, and iso-C_4F_8) and multicomponent (e.g., $C_4F_6/SF_6/N_2$ and c-$C_4F_8/C_4F_6/SF_6/N_2$) gaseous systems have been tested and found to have better DC breakdown strength properties than SF_6. These findings are reported and discussed.

Introduction

Knowledge of fundamental electron-molecule interactions is being applied to the understanding and design of improved gaseous dielectrics for power transmission and other high voltage applications. The breakdown measurements reported in this paper have been performed in DC uniform fields for ease in correlation with the basic data on electron-molecule interactions. Our ultimate goal is to design and improve gaseous dielectrics and to develop design rules for their engineering applications.

In a gas under an electric field, free electrons may form an avalanche, which may lead to a Townsend or a streamer-type breakdown, or (in nonuniform fields) to corona. Advances in inhibiting breakdown can be made controlling both the numbers and energies of free electrons in the dielectric gas by a systematic application of fundamental knowledge of the underlying electron-molecule interaction processes. The free electrons in a gas have a distribution of energies extending over a considerable range. Since the many interactions between molecules and electrons that lead to breakdown possess various probabilities that depend on electron energy,[1] the events of importance are quite complex. In the Townsend and streamer breakdown models this complexity is circumvented by the use of "lumped parameters" (such as $\overline{\alpha} = \alpha - \eta$ when α is the first Townsend ionization coefficient, and η is the electron attachment coefficient) that are actually determined by the details of the complex fundamental interactions. These lumped parameters, useful as they are for engineering design, tell us little about the fundamental interactions that initiate (or inhibit) breakdown. Each fundamental electron-molecule interaction is described by the probability (per unit distance of electron travel through the gas) that this particular reaction occurs. This probability in a gas whose number density is N is $N\sigma$, where σ is a function of electron energy and is called the cross section for that particular electron-molecule interaction process. (The term cross section is derived from its unit of area, suggesting the interpretation of an effective target size; however, a given molecule actually has no one "size" because σ depends on the molecule, the kind of reaction, and the impacting electron's energy.)

It is the thesis of this work that breakdown can be inhibited by reducing the numbers of free electrons and by reducing the energies of those electrons remaining free. The number of electrons is to be reduced by using gas molecules which attach electrons to themselves, tying up, this way, free electrons in negative ions which are too large and too slow to participate in breakdown formation. Since different molecules possess different abilities to attach electrons at different energies, the best dielectrics should be multicomponent gas mixtures designed systematically according to fundamental knowledge so that the attachment cross sections are as large as possible over as wide an energy range as possible. In an insulator certain molecules should also be employed for the purpose of reducing the energies of free electrons not only because this will directly help inhibit ionization but also because basic physics dictates that electron attachment can be effected well only at lower energies ($\lesssim$ 1 eV), and the attachment process is thus enhanced by helping to confine free electrons to these lower energies where they are subject to effective capture.

On the basis of the above philosophy we have undertaken a program of studying breakdown strengths of gases/mixtures, first in DC uniform fields. Simultaneously, fundamental data are being compiled, and new data are being obtained on electron attachment and electron slowing-down cross sections as functions of electron energy. The breakdown and fundamental data are correlated in the design of gaseous dielectrics. Chemical and physical properties, the effects of nonuniform fields, rough surfaces, and particles, as well as AC and impulse tests, etc., are also being considered, but presently the emphasis is on the question, "What is it that makes a good gaseous dielectric?"

Experimental Apparatus and Procedure

Desired Measurements

The present breakdown measurements were confined to DC, very uniform electric fields, free of effects due to surface roughness and contaminants. Results under such conditions are the easiest to correlate with fundamental electron-molecule interactions and basic theories of breakdown, and they make it easier to understand effects of varying experimental parameters. Although large differences in dielectric strengths are being sought ultimately, considerable precision was desirable in breakdown measurements to reveal small differences which might lead to larger improvements.

Data Acquisition Techniques

The basic experiment begins with the evacuation of a clean chamber to a low pressure (on the order of 10^{-7} torr) and the introduction of an accurately measured quantity of the gas/mixture to be tested. The gap between uniform field electrodes is adjusted by a micrometer drive. The high voltage is set to some "base" value below the breakdown voltage. An automatic feedback system raises the applied voltage slowly and linearly as it is sampled repeatedly by a digital voltmeter. After breakdown in the test gap, a parallel diverter gap is broken down to relieve the test gap of voltage and to remove the power-supply-stored energy in tens of microseconds; on a longer time scale the input voltage to the supply is lowered back to the "base" level. Meanwhile, the digital voltmeter is held at the last voltage sample acquired before the breakdown began. After the data have been recorded, the entire cycle is repeated.

Chamber

Most data were obtained in a stainless steel system of approximately 30 liters volume with uniform field electrodes (described in the following section); unless otherwise noted, the description will be given for that system, although some data were acquired in a very similar prototype apparatus differing primarily in volume (approximately 10 liters) and electrodes (a sphere-plane pair with field deviation from uniformity on the order of 0 to 3%). Before the admission of the test gas, the chamber is pumped by cryogenic pumps, a titanium sublimation pump, a 200-liter/second ion pump, and a mechanical forepump. Test gas pressure is measured by a Wallace and Tiernan series 1500 absolute pressure gauge with a range of 0-10 atm and an accuracy of ± 0.066% of full scale. The chamber provides ports with windows for visual access and ultraviolet irradiation, as well as ports for test gas inlets. All data reported here were taken at room temperature (∿ 20°C).

Electrodes

The chamber contained the "uniform field" electrodes resembling discs 5-1/2 inches in diameter, machined on a computer-driven lathe and polished to approximately 0.1 micron surface roughness. The basic design of Pearson and Harrison[2] was followed for the electrode shape, but the numbers in their table of coordinates were smoothed and interpolated by us to provide a more detailed table for the computer. The design was intended (taking the adjacent chamber walls into account) to provide an electric field that was (1) uniform to within 0.2% in the central gap and (2) weaker along the electrode edges. These two conditions were imposed because highly effective insulating gases can be extremely sensitive to small irregularities of electric field.[3]

The test electrodes are irradiated through a sapphire window by a 60 W deuterium ultraviolet lamp. Electrode axes and surfaces are aligned by set screws on the high voltage electrode while the electrodes are in contact. After the test gas has been admitted, the gap is adjusted by driving the ground electrode toward the HV electrode with a micrometer drive feedthrough until electrical contact is first observed, and then driving the electrodes apart to the desired gap distance.

Gas mixtures were formed according to the partial pressures of the constituents, with those of lowest vapor pressure being introduced first; the partial pressures of subsequently introduced component gases were measured by the pressure increase each produced. It has been found that gases are mixed well in this apparatus by maintaining high voltage across a wide electrode gap for several minutes.

Electrical Apparatus

The electrical system of the apparatus is diagrammed in Figure 1. The heart of the electrical system is a digital voltmeter (Hewlett-Packard 3480) which samples the output of a 1000:1 divider probe approximately 20 times per second. When breakdown of the test gap is detected optically by a phototransistor spark detector, a locally built controller circuit commands the voltmeter to hold the last sample acquired before the breakdown, thus preserving a 4-digit value of breakdown voltage. This value is known to be somewhat lower than the true breakdown voltage (since the applied voltage was rising before breakdown), but the error is controllable and quite small. For a typical meter sampling rate of 20 $\sec^{-1}$ and a typical voltage rise rate of 100 V/sec, this meter sampling error is between 0 and +.5V.

The power supply provides zero to ± 125 kV DC with ripple less than 0.1%. Positive or negative polarity is selected by a switch submerged in an oil tank also containing the 140 MΩ, 1000:1 resistive voltage divider probe string. All measurements reported in this paper were made with the negative polarity.

The controller circuit generates a slowly rising (ramp) reference waveform selected to designate the desired high voltage variation. A feedback circuit compares the voltage divider output with this reference and operates a motor-driven variable autotransformer according to the error signal, making the high voltage follow the reference waveform. The resulting HV waveform is quite smooth because of the smooth operation of the variable autotransformer (two parallel Superior Electric powerstats loaded far below rated current and rotated by a precision stepping motor) with additional smoothing from the HV

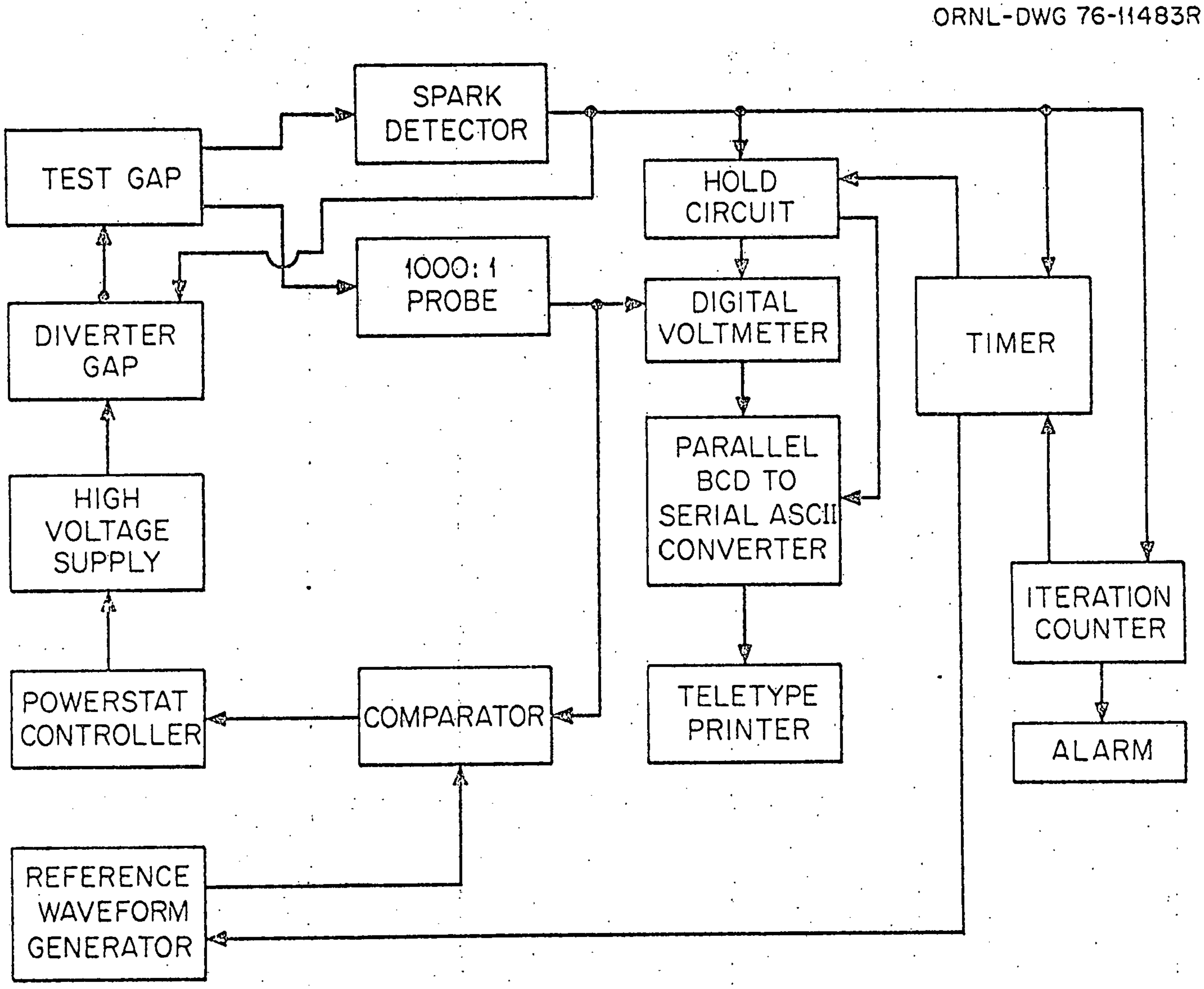


Fig. 1. Block diagram of apparatus for breakdown measurements.

power supply filter capacitor. Breakdown voltage values held by the voltmeter can be automatically recorded by a teletype printer. The timer automatically recycles the breakdown voltage measurement process after a (variable) delay of a few seconds. The counter can be used to halt the experiment after a desired number of breakdowns and to notify personnel.

The diverter gap is a trigatron pair of brass semispheres (each 60% of a 5-inch-diameter sphere) connected across the HV line between the power supply output capacitor and the test gap and voltage divider, as shown in Figure 2. In these experiments of negative polarity, only the grounded diverter main electrode has a trigger electrode, although provision is made for triggering both main electrodes. This trigger consists of 1/8-inch-diameter tungsten rod inside a 5/16-inch-outside diameter quartz rod, which is in turn fitted tightly in a hole drilled through the center of the trigatron electrode. The trigger is activated by a 50-kV pulse from a high performance auto ignition system (which can fire with a 2 millisecond period if required) upon command from the spark detector circuit.

To protect the instrumentation from the RF radiation of the breakdown, a copper mesh shielding cage surrounds the test chamber, diverter, voltage divider, and high voltage section of the power supply. Interlocks also protect personnel. All electrical connections for power and instrumentation inside the cage are made via capacitive feedthrough filters mounted through the cage wall. Water and gas exhaust connections are carried through the cage walls by wave guides. The single central ground point for the entire apparatus is formed by a copper plate mounted in the cage wall and accessible from both inside and outside the cage.

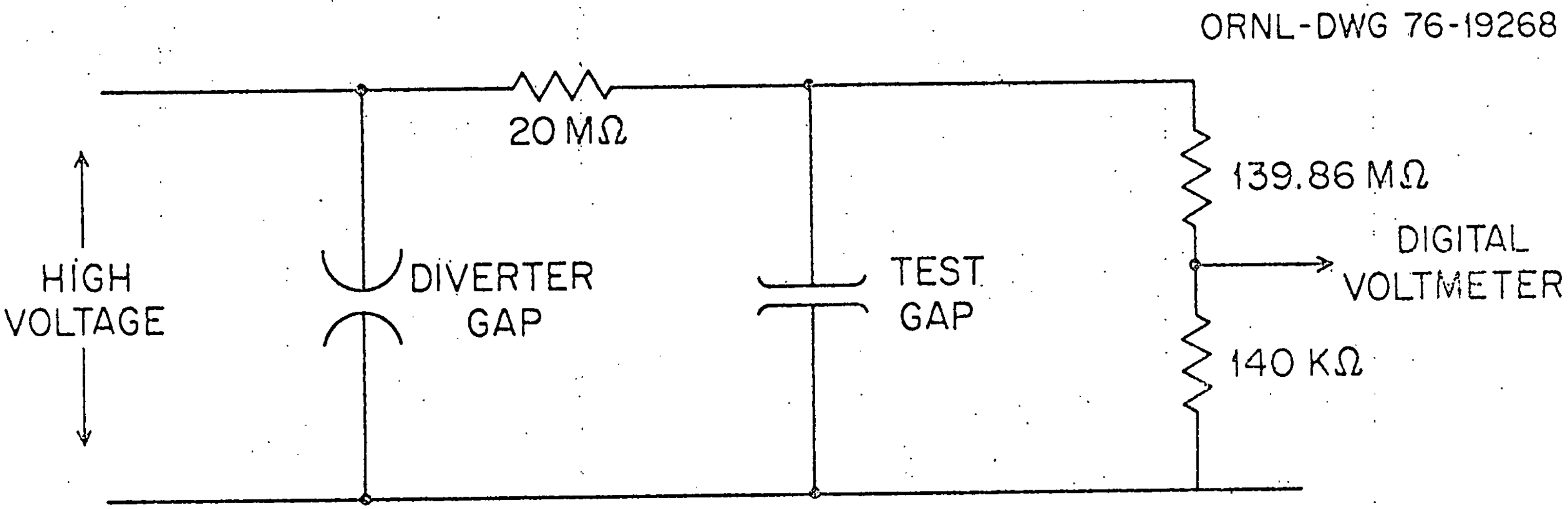


Fig. 2. High voltage test circuit.

Breakdown Results

The breakdown strengths of three fluorocarbon gases, measured for DC uniform fields as described earlier, are presented in Figure 3; each point represents an average of at least 10 breakdowns. Also shown in Figure 3 are breakdown voltages measured for SF_6 under identical experimental conditions. All three fluorocarbons are seen to have better dielectric strengths than SF_6, especially C_4F_6 (hexafluorobutyne) which in the E/P range indicated, is more than a factor of 2 better than SF_6.

The breakdown strengths of various mixtures of fluorocarbons, SF_6, and N_2 were systematically measured to construct an experimentally-based picture of the effectiveness of these gases as additives to N_2 and to correlate their effectiveness with their cross sections for electron attachment and electron slowing-down as functions of electron energy, as well as their individual breakdown strengths. Table 1 shows relative breakdown strengths obtained for $C_4F_6/SF_6/N_2$ mixtures; three of these mixtures were both better as dielectrics than SF_6 and cheaper than SF_6 (based on cost estimates of \$0.001/ft^3 for N_2, \$0.83/ft^3 for SF_6, and \$2.50/ft^3 for C_4F_6 [Ref. 4]). The results in Table 1 indicate that C_4F_6 is extremely effective as an additive to N_2. It should be noted that a mixture containing N_2 is expected to be better in comparison to SF_6 under nonuniform fields and impulse voltages than under DC uniform fields.

Table 2 shows measurements made on three- and four-component mixtures in which N_2 was the primary gas. The effect of removing any one additive (and replacing it by N_2) was investigated. C_4F_6 was found to be the most effective additive at the 10% level, followed in decreasing order of effectiveness by C_3F_8, SF_6, and c-C_4F_8.

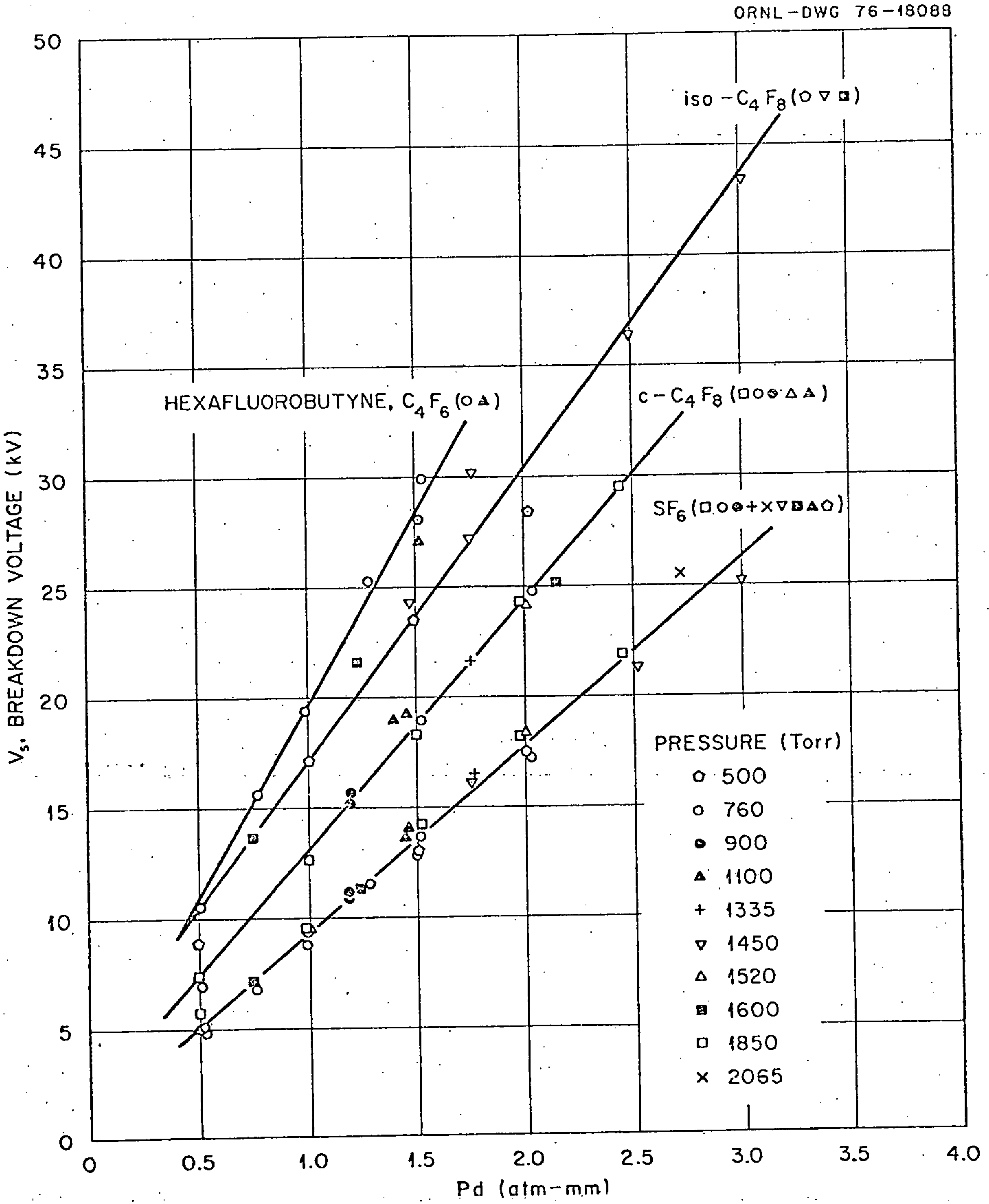


Fig. 3. Measured breakdown strengths of single gases (unitary systems).

TABLE 1

Relative Breakdown Strengths* of Some Two- and Three-Component Gaseous Mixtures

Gaseous Mixture			$\frac{\Delta V_s}{\Delta(Pd)}(\frac{kV}{atm\text{-}mm})$
N_2	C_4F_6	SF_6	
100%	—	—	2.98
—	—	100%	8.65
—	100%	—	19.76
90%	10%	—	6.77
80%	20%	—	8.71
70%	30%	—	10.18
80%	—	20%	6.53
80%	10%	10%	7.60
60%	20%	20%	9.51
50%	30%	20%	11.51

*Plane-plane, uniform-field geometry. The total pressure was 500 torr.

TABLE 2

Relative Breakdown Strengths* of Some Four-Component Gaseous Mixtures

Gaseous Mixture					$\frac{\Delta V_s}{\Delta(Pd)}(\frac{kV}{atm\text{-}mm})$	Relative $\frac{\Delta V_s}{\Delta(Pd)}$
N_2	C_4F_6	SF_6	c-C_4F_8	C_3F_8		
60%	10%	10%	10%	10%	8.39	100 %
70%	10%	10%	—	10%	8.32	99.2%
70%	10%	—	10%	10%	8.00	95.4%
70%	10%	10%	10%	—	7.80	93.0%
70%	—	10%	10%	10%	6.75	80.4%

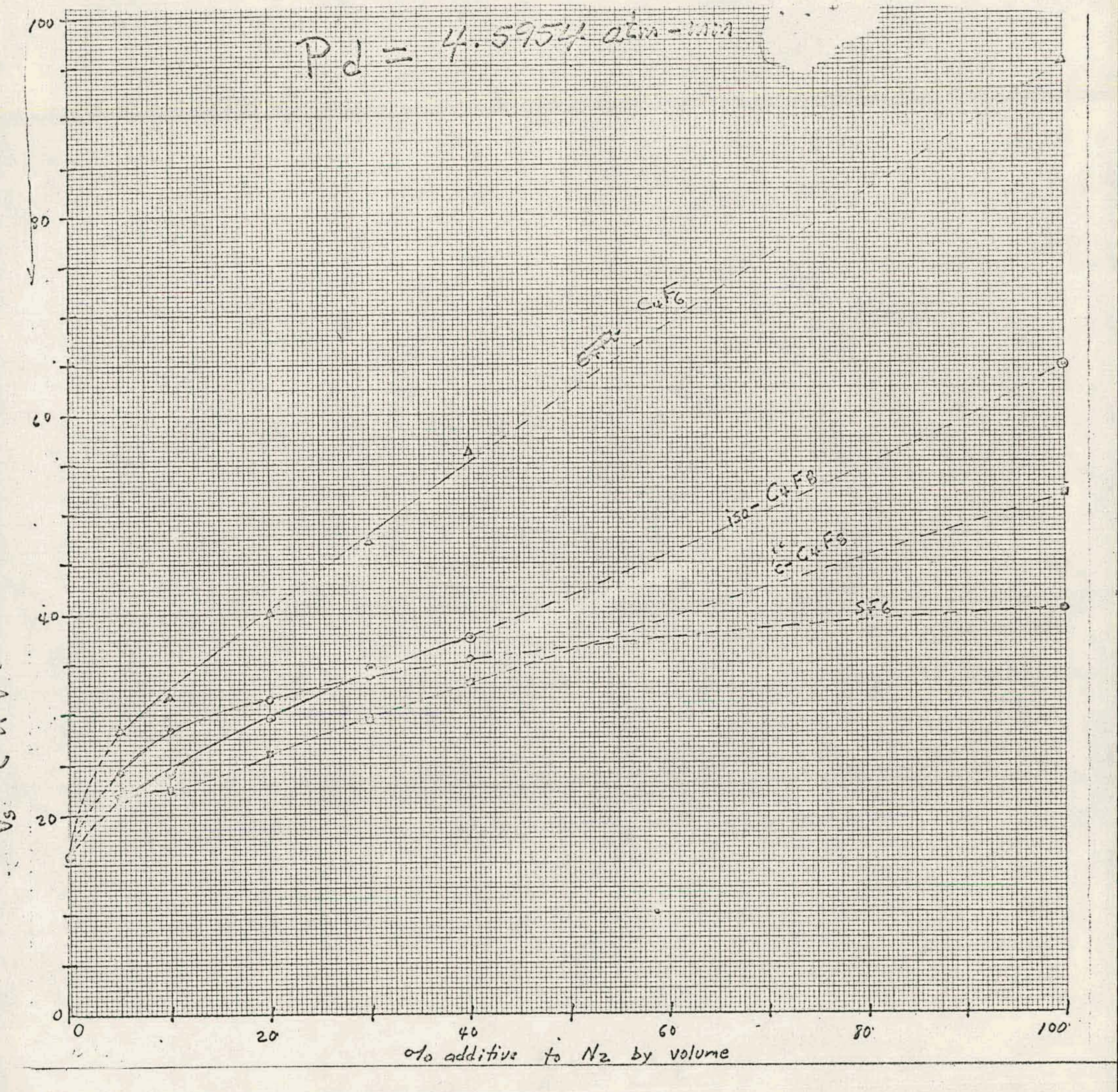
*Plane-plane, uniform-field geometry. The total pressure was 500 torr.

The breakdown strengths measured for single additives (C_4F_6, iso-C_4F_8, c-C_4F_8, SF_6) to N_2 are shown in Figure 4. Although for each case the dielectric strength of the mixture increases more rapidly with increase in additive at small quantities of the additive, the others do not suffer from the marked saturation effect evident for the SF_6 additive. C_4F_6 is the best additive for any proportions, while SF_6 is only better than iso-C_4F_8 and c-C_4F_8 at low proportions of additive. Preliminary results on mixtures of these gases with a buffer gas composed of equal quantities of N_2 and CO_2 led to similar conclusions.

Discussion

The electron attachment cross sections measured in a separate experiment for SF_6, c-C_4F_8, C_4F_6, and iso-C_4F_8 for electron energies up to 1.4 eV are shown in Figure 5 along with the calculated maximum cross section for s-wave capture. As exemplified by this maximum value as a function of energy, basic physics limits attachment cross sections to smaller values at higher energies; therefore, if breakdown is to be controlled by electron attachment, either the electrons must be captured before the electric field accelerates them above the energy range 0.0 to $\sim$ 2 eV, or gases must be added to effectively slow down electrons which reach higher energies, thus returning them to the energy range where they are likely to be captured. It is noted that although N_2 does not capture electrons itself, it is nevertheless a good buffer gas because it possesses a negative ion resonance at $\sim$ 2.3 eV which is very effective in slowing-down electrons around this energy.[1]

The attachment cross sections in Figure 5 begin to reveal a picture of effective breakdown control, although further details, both on electron attachment and on the inelastic processes that slow down electrons, must still be obtained. Figure 5 suggests



Breakdown strengths for N_2/single additive mixtures

Fig. 4

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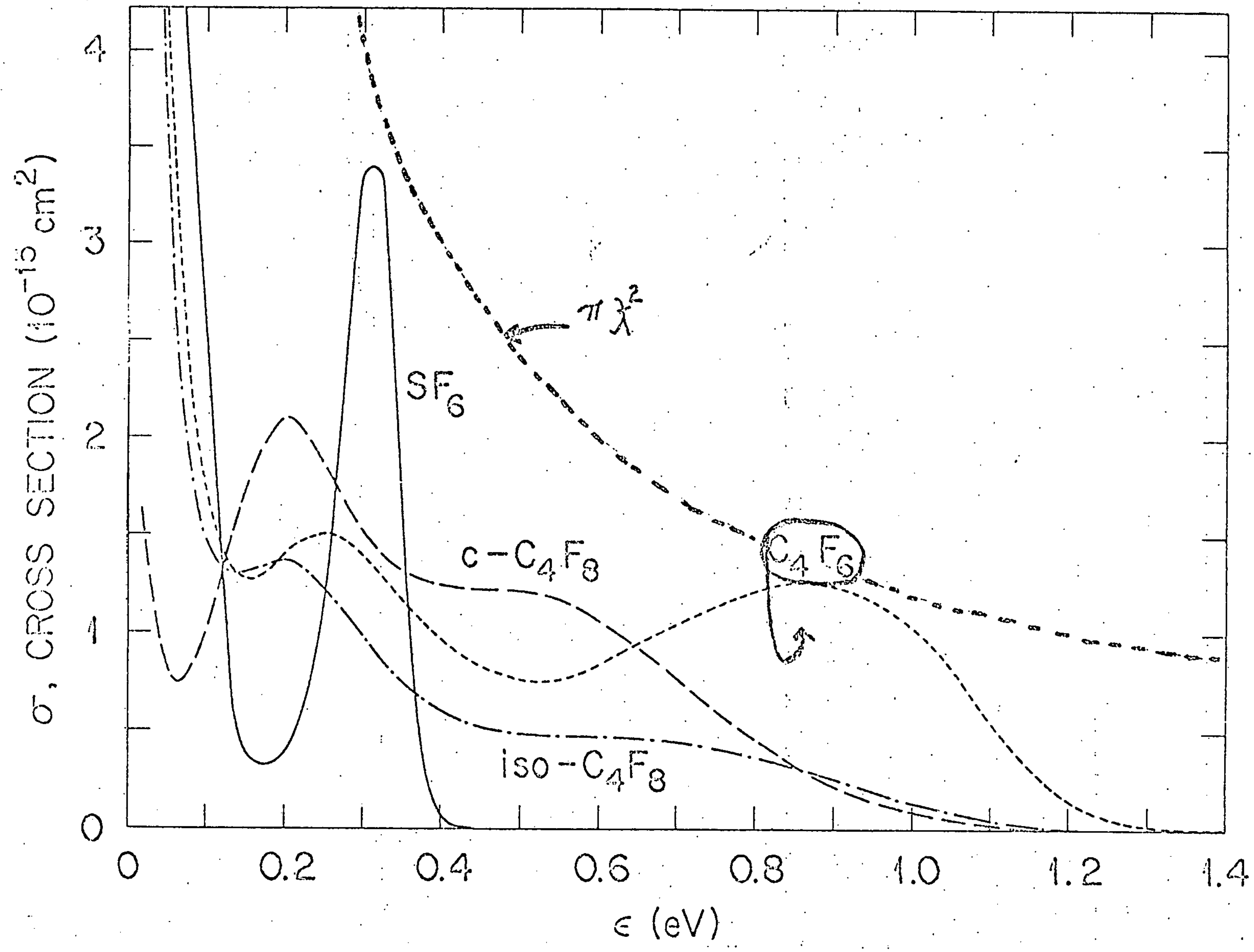

Electron Attachment Cross Section vs Electron Energy

Fig. 5

that all four molecules are good dielectrics because of their high electron attraction cross sections. It also indicates that C_4F_6 has the best dielectric strength of the four by virtue of its large cross section to substantially higher energies compared with SF_6, c-C_4F_8, and iso-C_4F_8. The better dielectric strength properties of the two C_4F_8 forms compared with SF_6 may similarly be due to the fact that they can attach electrons over a wider energy range than SF_6. It would appear from the data in Figure 5 that effective electron attachment in the energy range ∿ 0.5 to ∿ 1.5 eV is very significant in controlling breakdown. It is not yet certain as to why the iso-C_4F_8 is a better dielectric than c-C_4F_8 when the latter has a larger cross section in the 0.2 to 1.0 eV range than the former. However, it should be noted the iso-C_4F_8 has a much larger attachment cross section than the c-C_4F_8 below 0.2 eV, and it contains a double bond which is likely[5] to provide inelastic interactions with free electrons to slow them down to energies where capture is more efficient.

One striking feature of the SF_6 scattering cross section in Figure 5 is that its value is relatively quite small above ∿ 0.4 eV, while those for the fluorocarbons are substantial up to ∿ 1.2 eV. This may suggest that SF_6 is a poorer dielectric compared with these fluorocarbons because it loses control of free electrons once they attain an energy of $\gtrsim$ 0.4 eV, and they continue accelerating in the applied electric field.

The breakdown strengths of mixtures of SF_6 and the above-mentioned fluorocarbons each with N_2 were shown in Figure 4. The better dielectric each fluorocarbon is in its pure form, the better it is as an additive to N_2 (see Figures 3 and 4). There is relatively little departure from linearity for the mixture breakdown strength versus proportion of each accitive. SF_6 appears in small amounts to be as good an additive to N_2 as the fluorocarbons, and even better than the two C_4F_8 forms. However, increasing the proportion of SF_6 additive provides relatively little gain; i.e., the benefit of adding SF_6 seems to "saturate." This point is still under investigation, but it appears that this behavior may be due to the fact that (see Figure 5) a modest amount of SF_6 could attach almost all electrons at low energies (0 to ∿ 0.4 eV), so that additional amounts of SF_6 serve little purpose. On the other hand, the fluorocarbons attach electrons over a wider part of the free electron energy distribution and increasing their proportions provides additional attachment. It is stressed that it is important to attach electrons over a fairly wide energy range and that it would be advantageous to use (in addition to electron attaching molecules) molecules which can slow down electrons that have attained enough energy to escape the effective attachment energy range, thus returning them to the energy range where they are more readily attached.

Although the present breakdown data are for uniform fields without surface roughness or particles, they suggest that SF_6 may tend to be an inferior dielectric in locally nonuniform field (e.g., around surface roughness and particles) to gases able to attach electrons over a wider energy range than SF_6. This suggestion is made because the enhanced field at a rough surface or at a particle would shift the free electron energy distribution to higher energies and thus away from the relatively narrow low-energy range where SF_6 attaches electrons strongly. Hence, free electron control by a gas/mixture which attaches electrons over a wider energy range.

In conclusion, the design of improved gaseous dielectrics is being pursued by building an understanding of how to control both the number density and energy distribution of free electrons in the gaseous dielectric through application of fundamental detailed knowledge of electron-molecule interactions underlying the breakdown process.

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