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High Voltage Research (Breakdown Strengths of Gaseous and Liquid Insulators)

Third Quarterly Report (Period Ending December 31, 1976)

L. G. Christophorou
D. R. James
R. Y. Pai
M. O. Pace
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MASTER

OAK RIDGE NATIONAL LABORATORY
OPERATED BY UNION CARBIDE CORPORATION FOR THE ENERGY RESEARCH AND DEVELOPMENT ADMINISTRATION

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HEALTH PHYSICS DIVISION

HIGH VOLTAGE RESEARCH (BREAKDOWN STRENGTHS OF GASEOUS AND LIQUID INSULATORS)

Third Quarterly Report
(Period Ending December 31, 1976)

L. G. Christophorou, D. R. James, R. Y. Pai, M. O. Pace
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TABLE OF CONTENTS

ABSTRACT

The systematic basic and applied work to improve gaseous dielectrics continued. The breakdown strengths of C_6F_{10} (perfluorocyclohexene) and C_6F_{12} (mixture of 1,2- and 1,3- perfluorodimethylcyclobutane) have been found to be, respectively, 2.3 to 2.4 and 1.9 to 2.2 times higher than SF_6 under identical experimental conditions. The breakdown strength of mixtures of C_4F_6, SF_6, c-C_4F_8, iso-C_4F_8, and C_3F_8 with N_2 have been studied systematically in various combinations in an effort to understand the role of the basic properties and effectiveness of each component of a multicomponent gas on the breakdown strength. A number of such mixtures (e.g., 20% C_4F_6, 20% SF_6, and 60% N_2) are better and cheaper than SF_6. Basic electron attachment data have been obtained for C_3F_8, and the overall effectiveness of inelastic processes has been assessed for a number of gases based on their electron transport coefficients. Work on our new high-pressure, high-voltage and variable temperature (-40 to +120°C) chamber for uniform-field breakdown tests proceeded well. Design of apparatus for breakdown testing with nonuniform fields and/or rough surfaces has begun; concentric-cylinder field electrodes of varied surface roughness are under fabrication. Plans are underway for an International Symposium on Gaseous Dielectrics to be sponsored by ORNL-ERDA-EPRI.

I. INTRODUCTION

This is the third quarterly report of a project begun at Oak Ridge National Laboratory (ORNL) to apply expertise on electron-molecule interactions and relevant basic physicochemical knowledge to the need for energy economy and independence by developing improved insulating materials for high voltage power transmission, with the potential of enormous savings. The first report (ORNL/TM-5604)[1] covered progress to June 30, 1976. The second report (ORNL/TM-5713)[2] covered progress to September 30, 1976. This report covers the quarter October 1-December 31, 1976.

The experience, knowledge, and capability of the Atomic, Molecular, and High Voltage Physics Group within the Health Physics Division especially in the area of fundamental electron-molecule interactions is being used to design better insulating gaseous systems. In this fundamental area the group has played a leading role for over a decade and a half, and its continuing vitality offers the capability to obtain desired data not yet available.

The best insulating systems are anticipated to be mixtures designed as to components and pressures using fundamental physicochemical data obtained primarily from studies on interactions of slow electrons with atomic and molecular gases. More specifically, the gaseous components should be chosen to act as a system which collectively provides the "best" effective combination of electron <u>thermalizing</u> (slowing down) and electron <u>scavenging</u> (forming relatively immobile negative ions) to inhibit breakdown by electrons. This involves many different but coupled energy-dependent interactions between the gas and the electrons, which possess a rather wide distribution of energies. Especially significant are the electron attachment and electron energy loss processes in the range from thermal to ∿ 3 eV (generally subexcitation energies). It is very important that electrons are captured or slowed down before they escape this energy range.

In addition to our fundamental goal to identify the best gaseous (and liquid) insulating systems, a number of other goals are being considered in current activities. One is the nontrivial problem of extending current uniform field results to the nonuniform field conditions characteristic of power apparatus, and ultimately to practical engineering design guidelines. Also being considered are chemical properties, economics, temperature effects, and numerous practical "spin-offs" from this research such as the detection and prevention or diversion of incipient breakdown. The effects of solid particles and breakdown products must also be studied for the best insulators.

During this quarter, two breakdown apparatuses have been in operation and have been employed to study unitary and multicomponent insulating gases at various pressures and electrode gaps, while improvements and additions in the apparatuses continued to be made. Design of a third apparatus has been underway to attain a variable temperature (-40 to 120°C) and also to attain the voltage, pressure, and gap necessary for breakdown studies at higher values of Pd (pressure times gap). Breakdown strength measurements have been performed for a number of unitary and multicomponent gases. Correlation has been assessed between fundamental electron-molecule interactions and molecular structure, and breakdown strength. A more detailed study of diverters has been carried on to develop them as protecting devices for power systems and experimental apparatus. Acquisition of basic data has proceeded well.

II. APPARATUS

The two breakdown apparatuses described primarily in the first report,[1] with some modifications given in the second report,[2] have been in operation during this quarter.

For apparatus 1 the new controller has been completed, debugged, and put into routine operation. The controller is compatible with the new 300 kV DC supply.

Apparatus 2 has been in routine operation this quarter. As it was anticipated in the last report, the mechanical stability of the electrodes has been greatly improved by installing the platform to support the lower electrode from the chamber sides rather than from the HV feedthrough (as shown in Fig. 5, Report 1[1]). An effective method of fast mixing of multicomponent gaseous mixtures has been developed. It was found that application of high voltage ($\sim$ 80 kV) on the electrodes at maximum gap separation ($\sim$ 1 inch) for $\sim$ 20 minutes greatly reduces the mixing time.

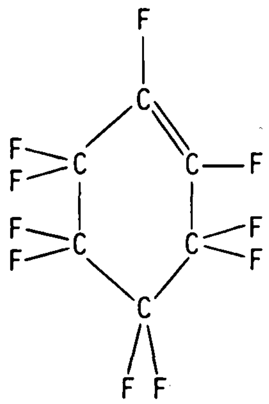
The new high-pressure, low-volume chamber for uniform field breakdown tests has been designed, and we expect its approval from the local pressure committee soon. This chamber, with the new 300 kV DC supply to be received next quarter, will provide a facility allowing breakdown tests at values of Pd much higher than attainable in the current apparatuses. New uniform field electrodes are being fabricated for this chamber.

The apparatus for the diverter experiment[2] for air has been completed and debugged after overcoming some difficulty with RF from the sparks. A plausible explanation is proposed in Section VI for the apparent two-mechanism breakdown in the diverter data presented in our previous report.[2] It will be tested experimentally in the next quarter. Presently, our diverter experiments are performed in air, but we are beginning to study diverters in other gases and gas mixtures.

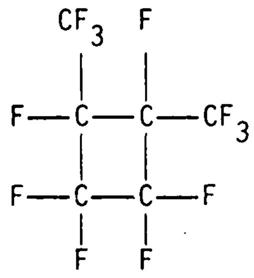
Design of apparatus for "practical conditions testing," viz., testing with nonuniform fields and/or rough surfaces has begun. Electrodes have been designed and put into fabrication for coaxial concentric cylinder geometries of varied inner radii, varied material, and varied surface roughness. Uniform field electrodes for varied surface roughness are under fabrication. In the next quarter we will begin to use these electrodes to study the most promising gases/mixtures.

III. BREAKDOWN STRENGTHS OF UNITARY GASES

During this quarter, we measured the dielectric strengths of two additional fluorocarbons: C_6F_{10} (perfluorocyclohexene) and C_6F_{12} (perfluorodimethylcyclobutane). Both of these compounds have relatively low vapor pressures at room temperature, on the order of 100 torr. The C_6F_{12} is cyclic with two CF_3 methyl groups attached to the ring. The C_6F_{10}, also cyclic, has one double bond. Their structures are illustrated below.



Perfluorocyclohexene



1,2-Perfluorodimethylcyclobutane

Their breakdown strengths are compared in Fig. 1 with those of C_4F_6 (hexafluorobutyne) and SF_6. The points for each gas deviate slightly from linearity as Pd is increased. The nonlinearity is to be expected since large gap separations were used in the sphere-plane electrode system, where the electric field becomes substantially nonuniform. Since P was small, large gaps of several millimeters were necessary to achieve the Pd's in the range 0.5 to 2.0 atm-mm. The C_4F_6 and SF_6 data were also taken at low pressure (see Fig. 1). Table I gives the approximate strengths relative to SF_6 = 1.0.

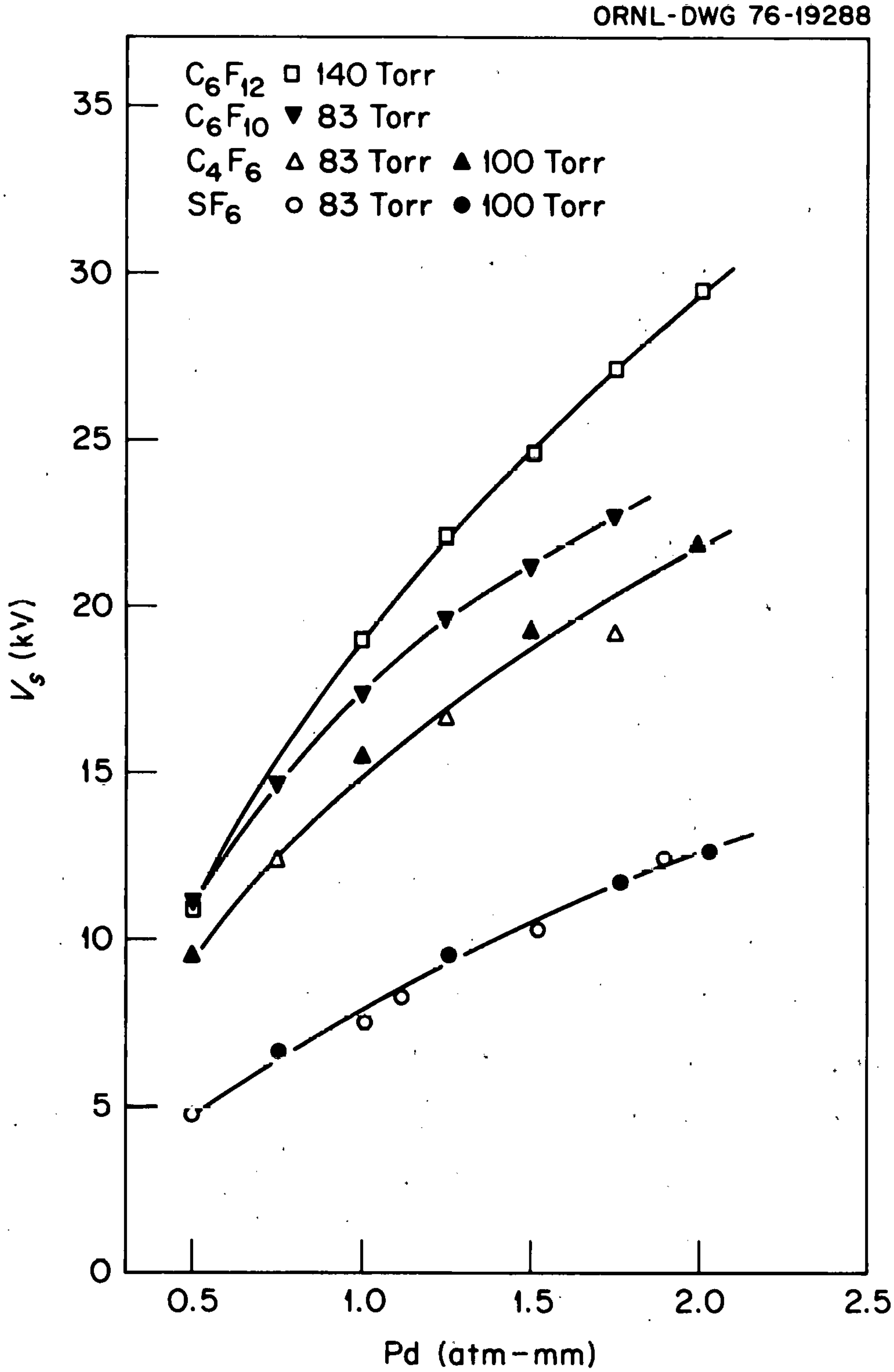


Fig. 1. Breakdown voltages versus Pd measured for C_6F_{12}, C_6F_{10}, C_4F_6, and SF_6 at the indicated pressures (sphere-plane electrodes).

TABLE I

Relative Breakdown Strengths of Some Unitary Gases*

Gas	Relative Strength
C_6F_{12}†	2.3—2.4
C_6F_{10}	1.9—2.2
C_4F_6	1.7—1.9
SF_6	1.0

*Sphere-plane geometry.

†Mixture of 1,2- and 1,3-perfluorodimethylcyclobutane.

These compounds, although too low in pressure to be used as insulators by themselves, are possible candidates as additives to buffer gases. The electron attachment cross sections as a function of electron energy for C_6F_{10} and C_6F_{12} will be investigated in the future.

Gases other than fluorocarbons are being investigated for potential use in gaseous insulation also.

IV. BREAKDOWN STRENGTHS OF GAS MIXTURES

As an initial step in our efforts to develop multicomponent gaseous mixtures with properties (cost, dielectric strength, etc.) superior to those of SF_6, we have made a series of measurements on mixtures of various electronegative gases with nitrogen. Nitrogen, aside from its inertness, abundance, and low cost, has the advantageous property of resisting breakdown by impulse more strongly than SF_6. Mixtures of N_2 and SF_6 have shown this improvement,[3] although the static dielectric strength is less than that for pure SF_6.

Our findings on binary and tertiary gaseous systems are summarized in Table II. The data on pure SF_6, N_2, and C_4F_6 are also listed in Table II for reference. It is clear that some gas combinations have higher static dielectric strength than pure SF_6 and are also cheaper. One particular mixture—20% C_4F_6, 20% SF_6, and 60% N_2—has a dielectric strength approximately 10% higher than pure SF_6. If we assume[4] costs of 83¢/cu. ft. for SF_6, 250¢/cu. ft. for C_4F_6, and 0.1¢/cu. ft. for N_2, we find that this mixture costs about 80% of the cost of SF_6. With the relatively large concentration of N_2, we would also expect the impulse-withstand limit of this mixture to be better than that of pure SF_6.

For 30% C_4F_6, 20% SF_6, and 50% N_2, we calculate a cost 10% higher than pure SF_6, but the dielectric strength for this mixture is 30% higher than that of pure SF_6. This combination should also possess better impulse characteristics than SF_6.

From Table II we see that mixtures of 20% C_4F_6 with N_2 are at least as good as pure SF_6. These mixtures must of course be tested under actual transmission line conditions before definite conclusions can be drawn.

TABLE II

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

Gaseous Mixture			Total Pressure (torr) (1 atm = 760 torr)	$\Delta V_s (\frac{kV}{atm\text{-}mm})$
N_2	C_4F_6	SF_6		
100%	—	—	500	2.98
—	—	100%	500	8.65
—	100%	—	500	19.76
90%	10%	—	500	6.77
80%	20%	—	500	8.71
70%	30%	—	500	10.18
80%	—	20%	500	6.53
80%	10%	10%	500	7.60
60%	20%	20%	500	9.51
50%	30%	20%	500	11.51

*Plane-plane, uniform-field geometry.

To understand the role and effectiveness of each component of a multicomponent gas, we studied a number of gas mixtures with three and four additives while keeping the buffer gas the same, i.e., N_2. We considered it desirable to combine N_2 (N_2 slows effectively subexcitation electrons because of its negative ion resonance at $\sim$ 2.3 eV[5]) with gases having good electron-attaching properties over as wide an energy range as possible (see examples in Fig. 8 of second quarterly report). Given a combination of gases, we proceeded to selectively remove one particular electron-attaching component at a time and to thus investigate the effect of electron capture in different energy regions on the breakdown strength of the mixture. We have used additives of 10% each of C_4F_6, SF_6, c-C_4F_8, and C_3F_8, with N_2 as the remaining buffer gas. By removing one additive at a time from the mixture, we found the relative importance of each of these compounds in a buffer of N_2. The slopes of the breakdown voltage versus Pd for a number of such mixtures are summarized in Table III.

From the data in Tables II and III, it is clear that:

1. C_4F_6 is extremely effective as an additive. Replacement of 10% of C_4F_6 by N_2 has a dramatic effect on the dielectric strength of the mixture. A decrease of nearly one-quarter is observed. It is furthermore seen that with 10% of C_4F_6 in the mixture the dielectric strength of any of the combinations is greater than 90% of the breakdown strength of the full combination of the four additives with with N_2.

TABLE III

Relative Breakdown Strengths* of Some Four-Component Gaseous Mixtures

Gaseous Mixture					$\Delta V_s(\frac{kV}{atm\text{-}mm})$	Relative ΔV_s
N_2	C_4F_6	SF_6	$c\text{-}C_4F_8$	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.

2. c-C_4F_8 is not so effective as an additive. When we replaced the 10% of c-C_4F_8 by N_2, the dielectric strength dropped by only less than 1%.

3. Strikingly, C_3F_8 seems to be more effective than SF_6 although its electron attachment rates below $\sim$ 0.8 eV (see Fig. 5 in Section V) are more than one thousand times lower in magnitude than those of SF_6.

The superior properties of C_4F_6 and perhaps also the strikingly good properties of C_3F_8 seem to indicate the importance of the electron attachment processes at energies $\gtrsim$ 1 eV (see Fig. 8 of Ref. 2 and Fig. 5 of Section V of this report). More work, however, is necessary to assess quantitatively the significance of this on breakdown. It is, for example, curious that iso-C_4F_8 and c-C_4F_8 do not increase the dielectric strength in low-percentage mixtures with N_2 as much as an equal amount of SF_6 does, although in the pure form they are both stronger dielectrics than pure SF_6.

The above observations are further substantiated by the data in Figs. 2, 3, and 4 where the breakdown voltage of various mixtures of attaching gases with N_2 are shown. The breakdown voltage of pure N_2 is shown for reference.

With 10% additive to N_2 (Fig. 2), the SF_6 mixture is close to, but lower than, the C_4F_6 mixture; mixtures with iso-C_4F_8 and c-C_4F_8 are significantly lower. As we increase the attaching gas ratio to 20% (Fig. 3), the C_4F_6 mixture becomes much better relative to the SF_6 mixture, and the iso-C_4F_8 mixture equals the SF_6 mixture. At 30% (Fig. 4), the C_4F_6 mixture is better still, and the iso-C_4F_8 mixture surpasses the SF_6 mixture.

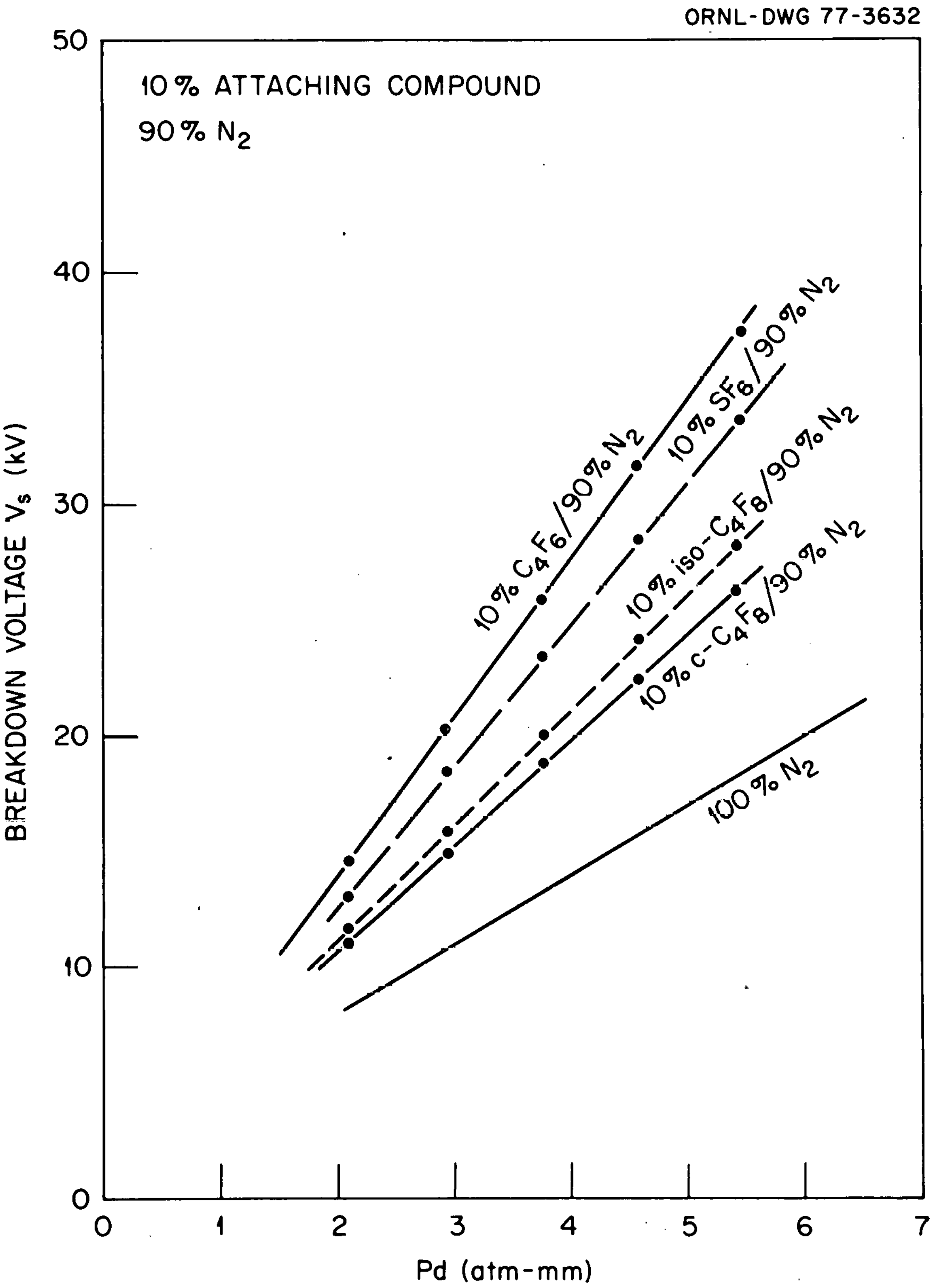


Fig. 2. Breakdown voltages versus Pd measured for mixtures of 90% N_2 with 10% C_4F_6, 10% SF_6, 10% c-C_4F_8, and 10% iso-C_4F_8 compared with 100% N_2.

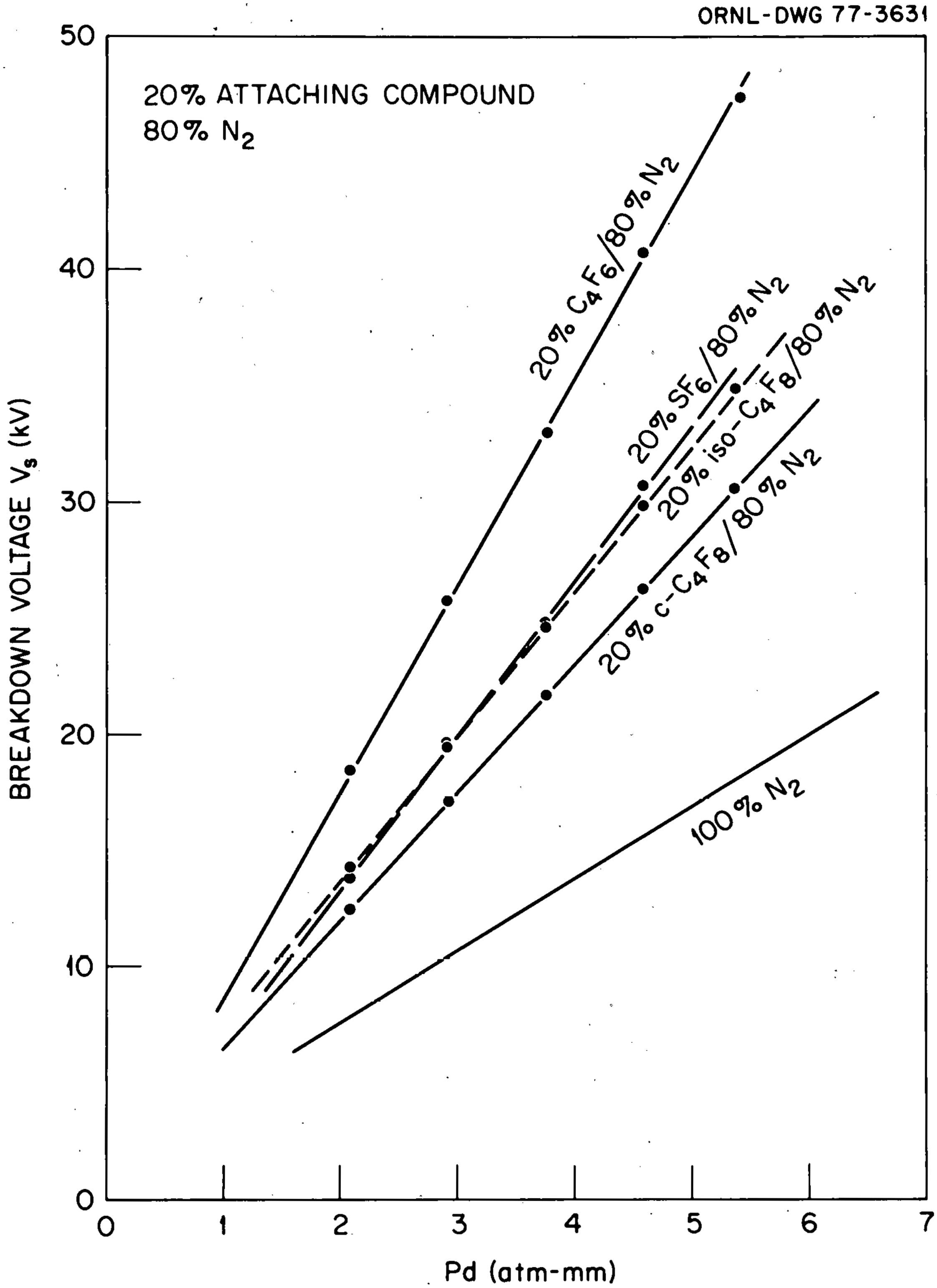


Fig. 3. Breakdown voltages versus Pd measured for mixtures of 80% N_2 with 20% C_4F_6, 20% SF_6, 20% c-C_4F_8, and 20% iso-C_4F_8 compared with 100% N_2.

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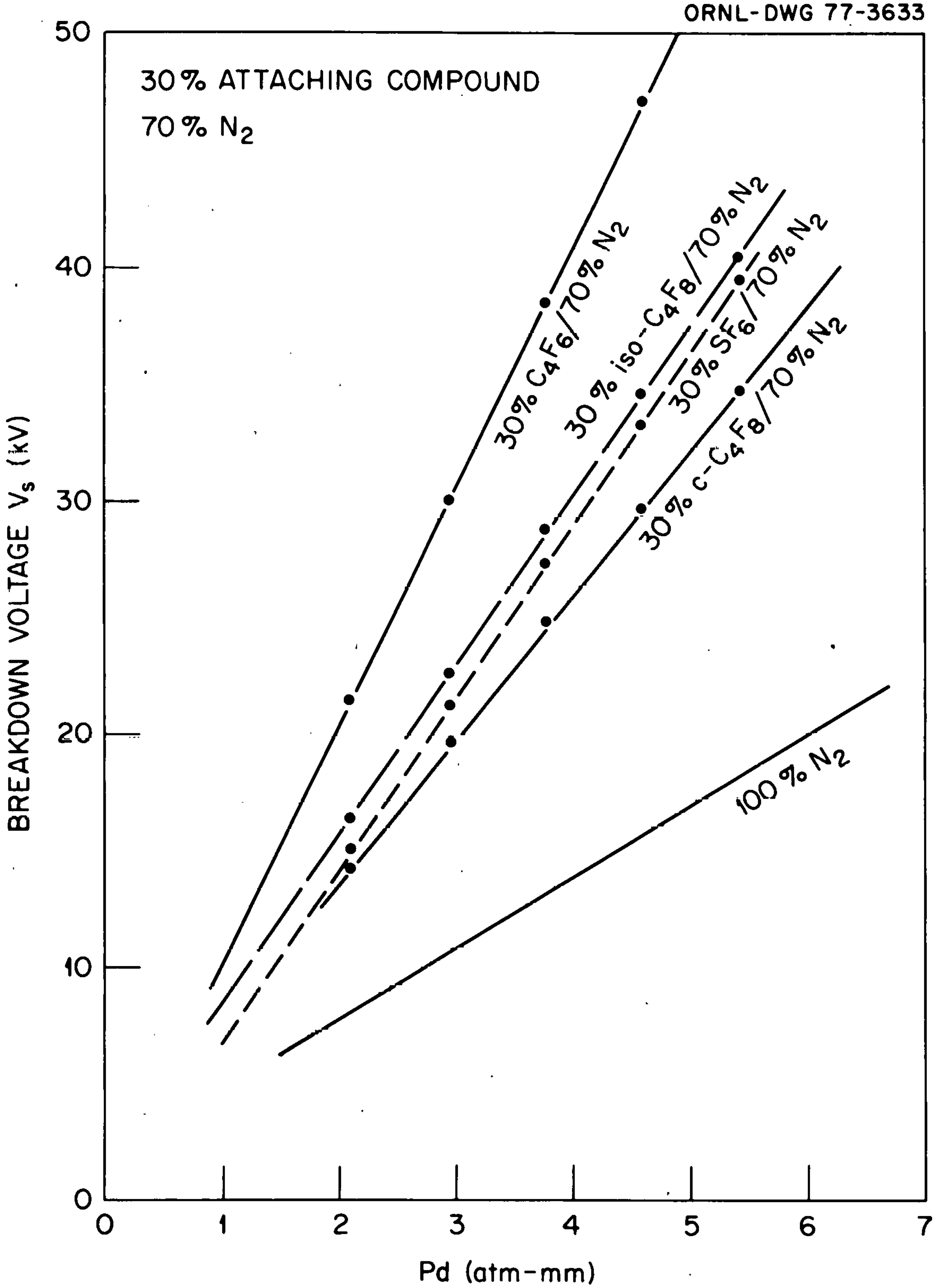


Fig. 4. Breakdown voltages versus Pd measured for mixtures of 70% N_2 with 30% C_4F_6, 30% SF_6, 30% c-C_4F_8, and 30% iso-C_4F_8 compared with 100% N_2.

Experiments are in progress with N_2 and C_3F_8 as buffer gases.

The systematic study of mixtures to determine the effect of the contributions of each of the electron-attaching components and their cross sections and relative concentrations to increasing breakdown strength is continuing. This basic and systematic knowledge will enable the design of superior mixtures for gaseous dielectrics.

V. BASIC STUDIES

In this quarter we have continued to measure the electron-attachment rates as a function of mean electron energy and the corresponding cross sections as a function of electron energy for fluorocarbon molecules, viz., C_3F_8, C_4F_6, and C_3F_6. As mentioned in our previous quarterly report, the electron affinity of C_3F_8 is small, yet it is as good a dielectric as SF_6. Figure 5 shows the measured electron-attachment rate as a function of mean electron energy for C_3F_8. If we compare this attachment rate with that of SF_6, we see that at mean electron energies below $\sim$ 0.8 eV the attachment rate for C_3F_8 is more than three orders of magnitude lower.

Electron beam studies[6] have shown that C_3F_8 dissociatively attaches electrons in the energy range 2.3 to $\sim$ 4 eV. Our studies of C_3F_8 in argon indicate that the attachment rate has a maximum in this energy range. It would seem that either the electron-attachment properties of C_3F_8 in this energy range are very important in controlling its breakdown strength (in spite of their relatively small magnitude) and/or that C_3F_8 possesses strong inelastic scattering cross sections at subexcitation energies.

In conjunction with breakdown measurements, we made preliminary measurements of the electron-attachment rates of C_3F_6 in the mean electron energy range 0 to $\sim$ 0.8 eV. These rates are about an order of magnitude lower than the rates for SF_6 and other fluorocarbons studied.

The physical properties of small quantities of C_4F_6 have prevented us from measuring absolute attachment rates for this gas to better than ±20%. This uncertainty may be the best that we can obtain with our techniques for this molecule.

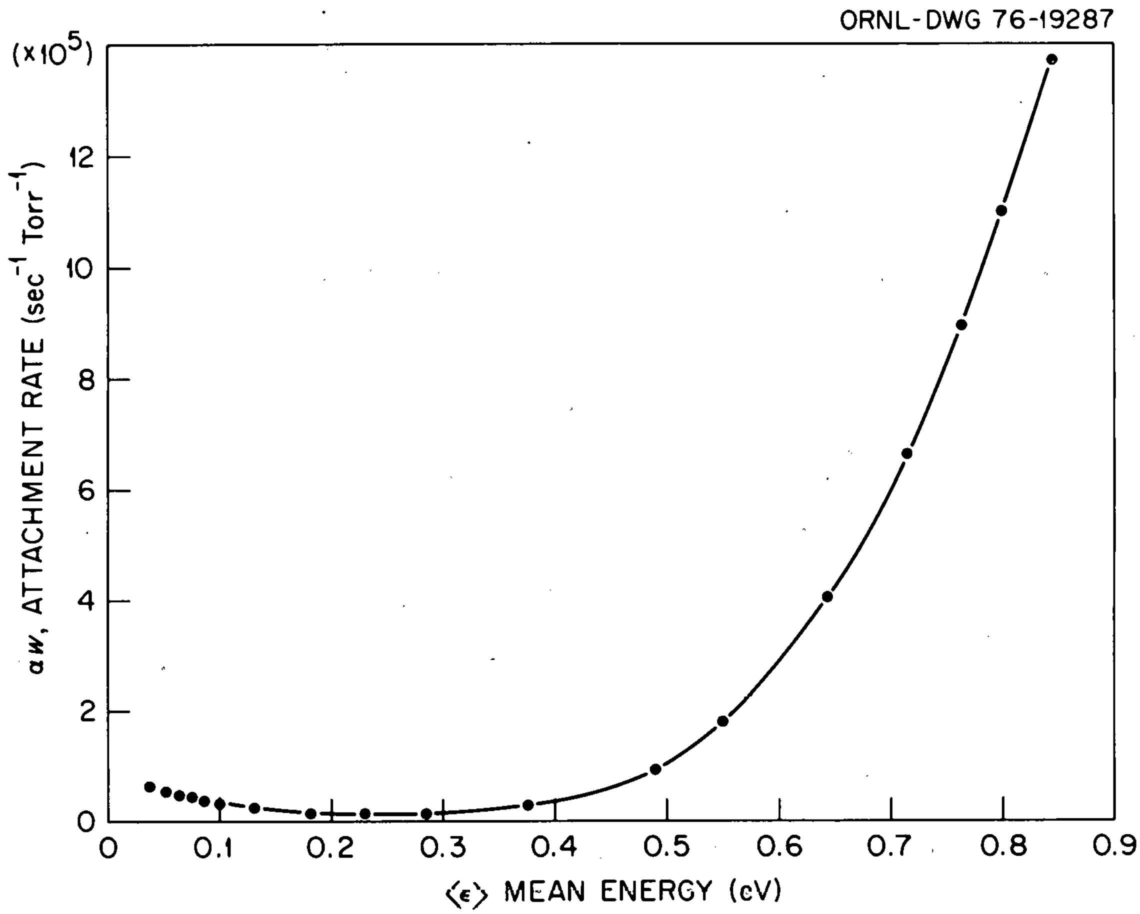
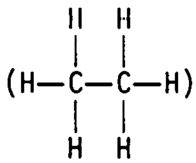
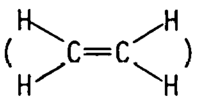


Fig. 5. Electron-attachment rate versus mean electron energy for C_3F_8.

In the next quarter, we plan to measure the attachment rates as a function of energy for other perfluorocarbons of interest, such as perfluorocyclohexene (C_6F_{10}), perfluoro-1,3-dimethylcyclohexane (C_8F_{16}), etc.

During this quarter also, efforts have been made to obtain other basic data which reflect the <u>overall</u> effect of inelastic processes. For a given electric field, E, and total pressure, P, i.e., for a given value of E/P (or E/N, where N is the number density), the mean electron energy, $\langle\varepsilon\rangle$, should be lower the more effective the inelastic processes are. A good measure of $\langle\varepsilon\rangle$ is the characteristic energy $\langle\varepsilon\rangle_M \equiv 3/2\ e\ D_L/\mu$, where e is the electron charge, and D_L/μ is the ratio of the lateral diffusion coefficient D_L to the electron mobility μ. Identification of $\langle\varepsilon\rangle$ with $3/2\ e\ D_L/\mu$ assumes that the shape of the electron energy distribution is Maxwellian. Although this assumption is not strictly valid, the functions D_L/μ versus E/P and $\langle\varepsilon\rangle$ versus E/P are closely related and can help indicate which compounds are more effective electron thermalizers and thus better suited for use as buffer gases in multicomponent gaseous mixtures. From published data (mostly collected by Christophorou[5]) we obtained the information in Fig. 6. It is seen that for a given value of E/P, the electrons have lower energies ($\langle\varepsilon\rangle$ is lower) for systems with double and triple bonds [compare data for ethane (), ethylene (), and acetylene (H—C=C—H)]. This is clearly demonstrated in Fig. 7 where the mean scattering cross sections, $\langle\sigma\rangle$, are plotted as

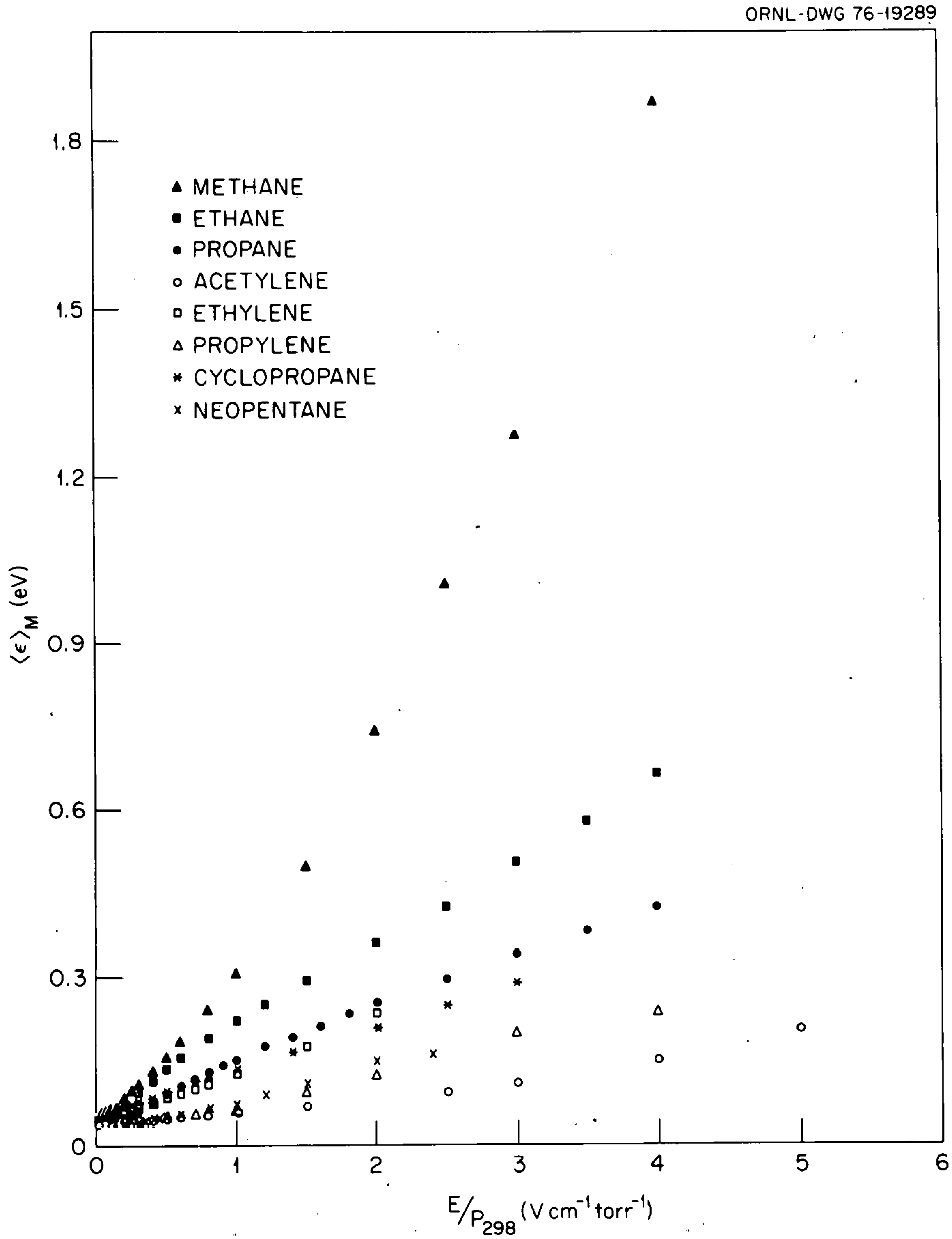


Fig. 6. Mean electron energy versus E/P_{298} for methane, ethane, propane, acetylene, ethylene, propylene, cyclopropane, and neopentane.

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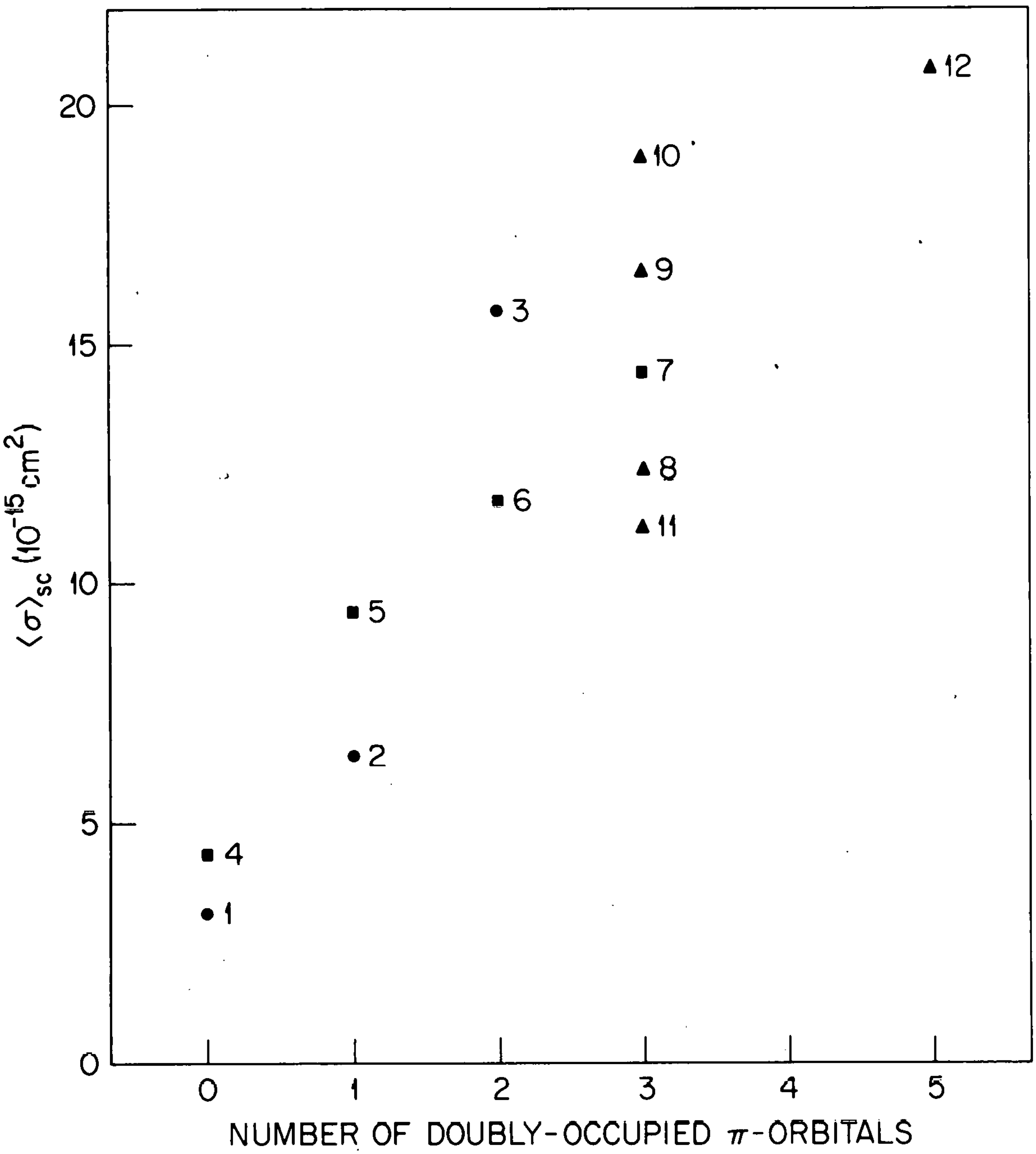

Fig. 7. Mean scattering cross sections as a function of number of doubly occupied π-orbitals. Data based on the work of L. G. Christophorou, R. P. Blaunstein, and D. Pittman, Chem. Phys. Letters 22, 41 (1973).

a function of the number of doubly occupied π-orbitals of the molecule, i.e., the number of double bonds. Although these cross sections are for epithermal electrons, they are still quite revealing; the larger the number of double bonds in the molecule, the larger are the electron scattering cross sections and thus the lower the mean electron energy and, therefore, the better the gas from the dielectric point of view.

In Fig. 8, D_L/μ is plotted as a function of E/P for some diatomic and triatomic gases (data taken from Christophorou[5]). All molecular gases in this figure possess negative-ion states in the subexcitation region. It is interesting to note that the electron energy (as exemplified by the D_L/μ values) is low over a wider E/P range for CO_2 and CO than for N_2O and N_2. For the former two the negative-ion states lie lower than for the latter two. (It should, of course, be noted that CO is slightly polar, and CO_2 possesses a quadrupole moment which could enhance the scattering at low energies.)

Unquestionably, the existence of negative-ion resonances (as many as possible and over as wide an energy range above 0.0 eV as possible) would enhance scattering and lower the electron energy and would thus yield a better gas for use as a buffer component in a multicomponent gaseous insulator. In this regard, a combination of gases with a number of negative-ion resonances covering a wide range of energies may be more advantageous than a single component.

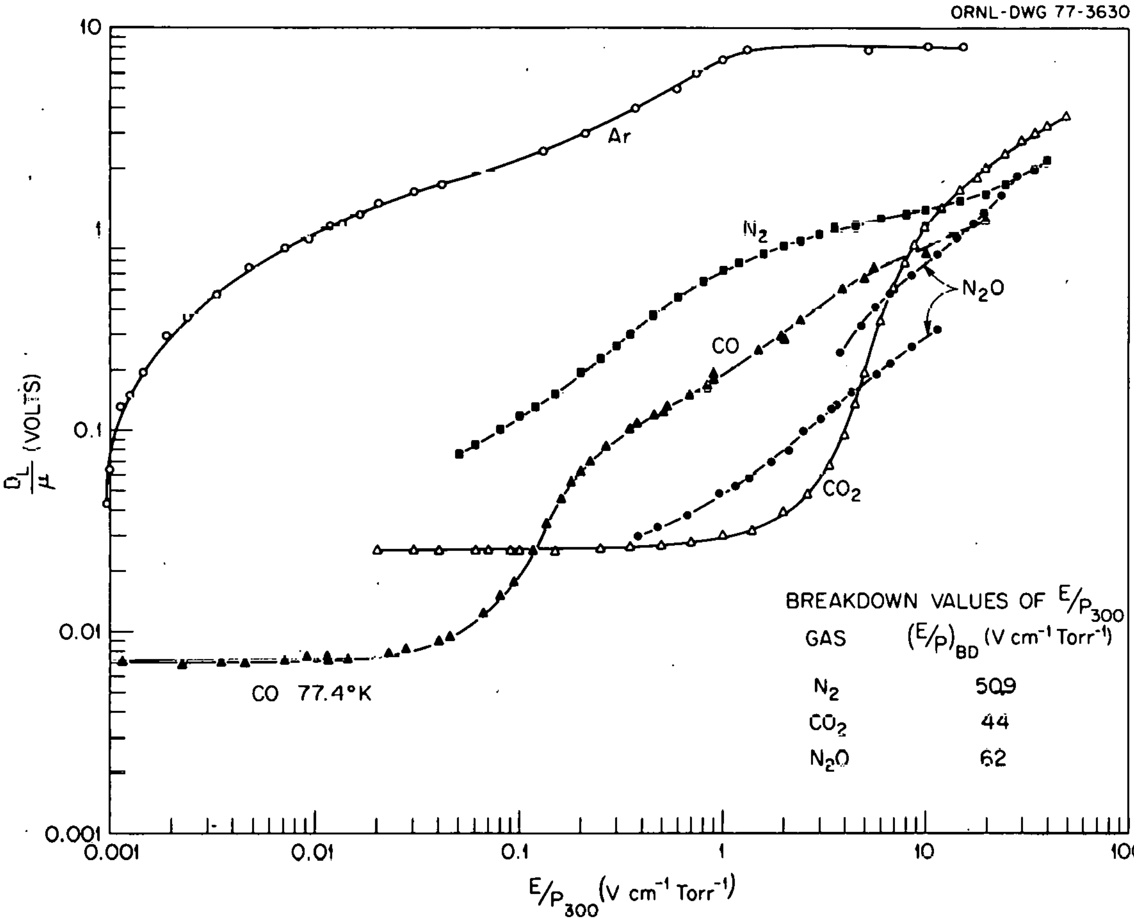

Fig. 8. D_L/μ versus E/P_{300} for Ar, N_2, CO_2, N_2O, and CO. All data are for 300°K except for CO which are for T = 77.4°K.

VI. APPLIED STUDIES

Applied studies is a new section in this report in which we describe the results of our investigation of the best gases/mixtures under more practical conditions (than DC uniform fields with extremely smooth surfaces). Thus, Sections III, IV, V, and VI give results for the three major respective facets of our program: breakdown strengths under ideal conditions (and their correlation with basic physics) (Sections III and IV on unitary and multicomponent systems, respectively); the basic physics (Section V); and the breakdown strengths under more practical conditions as well as special gaseous-insulator applications (and their correlation with basic physics and ideal conditions) (Section VI).

For testing the best gases/mixtures under conditions closer to practice, electrodes have been designed and are being fabricated for breakdown tests in coaxial cylinder geometry of various inner radii, roughness, and materials. The effects of radii and roughness will be compared with breakdown theory. Uniform field electrodes are also being fabricated for testing with variable roughness so its effect can be isolated from the effect of macroscopically nonuniform fields produced by cylindrical geometry. The best gases from uniform field experiments will be tested with nonuniform fields and rough surfaces.

In the diverter studies, data in air were given in the last report.[2] The reported[2] apparent two-mechanism breakdown is believed to be associated with a gap of two high field regions (near the two electrodes) and an intervening low field region. Triggered gap tests are planned with electrodes (sphere-sphere) producing such conditions, and also with electrodes (sphere-plane) giving only one higher field gap region.

VII. INTERNATIONAL SYMPOSIUM ON GASEOUS DIELECTRICS

At the request of Thomas Garrity of ERDA, preliminary plans have been made for holding a conference entitled "International Symposium on Gaseous Dielectrics" on research and development of high voltage transmission systems using gaseous dielectrics as insulators. Topics to be discussed include investigations into the breakdown phenomenon of dielectrics in uniform and nonuniform fields, development of methods to reduce the effect of particle contamination, and experimentation with new gases and gas mixtures. The conference will bring together key individuals from leading research centers, industrial laboratories, and funding agencies to review and discuss the progress and the problems of current interest and to seek solutions to the latter.

ORNL will act as host to the three-day conference to be held at the Hyatt Regency Hotel in Knoxville, Tennessee, on March 6-8, 1978, with an expected attendance of about 150. The conference is to be sponsored in part by registration fees and in part by ORNL, ERDA, and EPRI. A request for funding will be submitted to ERDA in the next quarter.

VIII. CONTACTS

Robert Pai delivered a paper from the group at the 29th Annual Gaseous Electronics Conference (October 18-20, 1976) in Cleveland, Ohio. An abstract for a paper from the group has also been submitted to the IEEE Electrical/Electronic Insulation Conference (September 26-29, 1977) in Chicago, Illinois.

Randy James and Marshall Pace, on December 8 and 9, 1976, visited the High Voltage Laboratory at Westinghouse Research Laboratories, Pittsburgh, Pennsylvania, the High Voltage Laboratory of MIT, and the Delta Ray Corporation, Burlington, Massachusetts. They had quite profitable discussions of mutual research interests at these laboratories and were instructed at Delta Ray on our 300 kV supply being completed there.

Profitable discussions were had with T. F. Garrity and N. P. Laguna of ERDA, J. A. Phillips of TVA, and L. L. Radcliffe of Oak Ridge Operations during their on-site visit in December 1976.

L. G. Christophorou has been invited to give a lecture entitled "Elementary Electron-Molecule Interactions and Negative-Ion Processes at Subexcitation Energies and Their Significance in Gaseous Dielectrics" at the 13th International Conference on Ionization Phenomena in Gases (Berlin, September 1977).

We were visited in October by Professor Barry Weedy from the University of Southampton, England. Professor Weedy was here as the National Science Foundation Distinguished Visiting Professor at The University of Tennessee. Professor J. L. Franklin of Rice University also visited the group in October.

IX. REFERENCES

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