# DIELECTRIC PROPERTIES OF SULPHUR UNDER HIGH VOLTAGE STRESS

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#### UNDER HIGH VOLTAGE STRESS

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MASTER OF SCIENCE

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#### PREFACE

This work has been undertaken as part of a larger project of the Engineering Experiment Station, under Dr. Clark A. Dunn, to learn more about dielectric constants in general. This is a subject upon which adequate data is lacking, and for which, it is believed, the theories have overlooked some basic facts.

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#### REVIEW OF LITERATURE

#### A. Dielectric Constants

Between two electrically charged bodies, separated by a distance, r, there exists a force. In 1785, Coulomb determined that the magnitude of this force is:

$$\mathbf{F} = \frac{\mathbf{Q}_1 \, \mathbf{Q}_2}{\varepsilon \, \mathbf{r}^2} \tag{1}$$

where F is the force in dynes on either body owing to the presence of the other,  $Q_1$  and  $Q_2$  are the electrostatic charges in statcoulombs, r is the separation distance in centimeters, and  $\boldsymbol{\varepsilon}$  is a proportionality constant dependent upon the material separating the two charges. If E, the electrostatic field strength, is defined as the force acting upon a unit charge, then it may be expressed as:

$$E = \frac{Q}{\epsilon r^2}$$
 (2)

where E is in statuolts per centimeter. Some early experimenters used these relationships to determine the dielectric constant,  $\boldsymbol{\xi}$ , but other methods have been found to be more convenient.

Another approach is through the concept of capacity. One definition of the capacity of a condenser is the ratio of charge stored to the potential producing it.

$$C = \frac{Q}{V}$$
(3)

If a pair of parallel plates have a charge density of  $\tau$  units per unit of area and are separated by a centimeters, then, according to Gauss' Law:

$$E = \frac{4\pi\gamma}{\epsilon}$$
(4)

Letting V, the potential difference, equal Ea and substituting into equation (3), the capacity per unit area is

$$C = \frac{\varepsilon}{4\pi a}$$
(5)

Since capacity is directly proportional to dielectric constant, an easier method of measuring dielectric constants is by means of capacity measurements.

The concept of lines of electric flux is often used. These lines are assumed to eminate from a positive charge and terminate on a negative charge. The electrostatic flux density (lines of flux per unit area) is defined as:

# D-EE

D may also be thought of as displacement. This is described by Einer<sup>1</sup> as follows. The electric field causes the orbits of the electrons in the dielectric to be distorted. This distortion of the electron orbits is called displacement and the motion of the electrons in this action is called displacement current. In this case equation (6) still holds.

(6)

2

1D. F. Miner, Insulation of Electrical Apparatus, pp. 9-10.

### B. Correlation of Dielectric Constant

#### With Other Physical Properties

It has been shown that the ability of a dielectric substance to store energy is measured by its dielectric constant. Therefore, it would be of great significance to discover why a material has its particular dielectric constant and what factors may affect its constant.

Several theories are in existence and several attempts have been made to correlate dielectric constants with other physical properties. However, in spite of efforts expended in that direction, the material is quite sketchy and often contradictory.

Jones<sup>2</sup> states that, in general, the dielectric constant increases as the conductivity increases. Miner<sup>3</sup> draws up the following generalized table: Group Description Resistivity Dielectric Constant 0 to  $10^6$  ohm-cm. Ι Conductors 30-100 10<sup>6</sup> to 10<sup>12</sup> ohm-cm. II Semi-conductors 6- 30 1012 to 1020 ohra-cm. Good Insulators III 1- 6

There are many exceptions to the information given in this table, however. For example, pure water which is a good insulator has a dielectric constant of approximately 80, while sea water, a good conductor, is also nearly 80. According to Wainer<sup>14</sup>, special preparations of barium-strontium titanate, another insulator, have a dielectric constant of approximately 20,000. No reference is made in standard tables, nor in available literature to the exact dielectric

<sup>2</sup>H. L. Jones, <u>An Electron Theory of the Hall Effect and Susceptibility</u>, p. 7.

Miner, op. cit., p. 8.

<sup>4</sup>E. Wainer, "High Titania Dielectrics," <u>Transactions of the Electro-</u> <u>chemical Society</u>, LXXXIX (April, 1946), 337.

constant of any good conductor.

One of the few attempts at correlation with other physical properties was made by Addenbrooke<sup>5</sup>. He used the method of Blotzmann in his early experiments in 1873. This was to suspend a small dielectric sphere near a large charged metal sphere and measure the pull on the dielectric sphere. Then a small metal sphere with the same radius as the dielectric sphere was suspended an equal distance from the large sphere, and the pull on this sphere was similarly measured. Blotzmann showed experimentally that:

$$\frac{\text{Pull on dielectric sphere}}{\text{pull on metal sphere}} = \frac{\boldsymbol{\xi} - \mathbf{l}}{\boldsymbol{\xi} + 2}$$
(7)

where **E** is the dielectric constant of the material of the dielectric sphere. Addenbrooke tabulated energy stored,

$$\mathcal{E} = \frac{1}{2} \mathbf{Q} \mathbf{V} \tag{8}$$

the pull on the dielectric sphere, the melting point of the dielectric, and its boiling point. The dielectric constant was measured in the liquid state for all materials. No further information was given on the standardization of conditions. The data was tabulated with relative values, arbitrarily assigning hydrogen the unity value, since it is the lowest in each case. For melting and boiling points, oxygen was taken as a reference and assigned a value of two, since hydrogen does not follow accepted theory in these properties. Addenbrooke's work is summarized as follows:

<sup>&</sup>lt;sup>5</sup>G. L. Addenbrooke, "The Non-Metallic Elements - Connexions Between Their Dielectric and Other Physical Properties," The London, Edinburgh, and Dublin Philosophical Magazine and Journal of Science, XLVII (June, 1924), 945-965.

Material	Energy Stored	Attraction	Melt. Pt.	Boil, Pt.
Hydrogen	1.0	1.0	0.5	0,5
Oxygen	2.0	1,98	2.0	2,00
Nitrogen	2.4	2,27	2.3	2.33
Chlorine	7.7	6.1	5.4	6.0
Bromine	17.8	10.5	8.4	8.4
Iodine	27.7	13.3	12.2	11.4
Phosphorous	16.2	10.0	10.0	9.0
Sulphur	16.2	10.1	12.2	17.9
Selenium	32,2	14.3	15.6	24.0

Another correlation is suggested by Creighton and Koehler<sup>6</sup>. They state that the greater the dielectric constant of a solvent the less its percentage disassociation into ions. This analysis must, of course, be limited to a particular group of materials.

According to the work of I. Walden, cited by Creighton and Koehler<sup>7</sup>, the dielectric constant of a material increases by the addition of these chemical groups in this order: I, Br, Cl, F, NH<sub>2</sub>, CN, CO<sub>2</sub>H, CHO, CO, NO<sub>2</sub>, and OH. This is almost the same order in which Auwers found the substitution of these groups increased the ability of the solvent to prevent association of the molecules of the solute.

It has been noted that, in general, the highest dielectric constants listed are for compounds of elements occuring in the second group of the periodic table. The next highest dielectric constants are, in general, compounds of elements occuring in the fourth group. There are, of course, exceptions, but the number of compounds which follow this rule is striking.

<sup>6</sup>H. J. Creighton and W. A. Koehler, <u>Principles and Applications of</u> Electrochemistry, pp. 55-66.

<sup>7</sup><u>Ibid.</u>, pp. 55-66.

#### C. Variation of Dielectric Constant in

A Given Material

#### 1. General

The dielectric constant of a given material may be made to change by variation of many physical factors, such as temperature, pressure, and frequency. In considering these properties of dielectrics, other factors enter the picture which must be considered concurrently in workable theories. These other properties, not accounted for by the theory of perfect dielectrics, are called anomalous properties. Anomalous properties listed by Whitehead <sup>8</sup> are variation of conductivity, absorption, and losses higher than may be accounted for by conduction. In order to consider the theories involving all these phenomena, the subjects of polarization, polar molecules, and dipole moments must be first taken up.

#### 2. Dipoles and Dipole Moments

A dipole is defined by Ramo and Whinnery<sup>9</sup> as a pair of equal and opposite charges separated by a small distance. This is enlarged upon by Gerlach<sup>10</sup> who considers it as a group of negative charges whose center of gravity is separate from that of a group of positive charges of equal magnitude. He also makes a distinction between natural dipoles and those induced by an external field.

The dipole moment, or electric moment is the measure of force which a dipole may exert. According to Ramo and Whinneryll

8J. B. Whitehead, Lectures on Dielectric Theory and Insulation, pp. 17-19.

9S. Ramo and J. R. Mhinnery, Fields and Waves in Modern Radio, pp. 80-81.

10W. Gerlach, Translated by F. J. Fuch, <u>Matter</u>, <u>Electricity</u> and <u>Energy</u>, p. 58.

11 Ramo and Whinnery, loc. cit.

$$\mathbf{m} = \mathbf{Q} \mathbf{r} \tag{9}$$

where m is the magnitude of the dipole moment, 2 the value of one of the charges, and r is the distance between them. It is, however, a vector quantity and the direction is specified as being that of the line drawn from the negative to the positive charge. The potential at a given point due to this dipole is:

$$V = \frac{m \cos \theta}{\epsilon r^2}$$
(10)

where r is the distance to the dipole (the dipole separation is here considered as negligible with respect to r), and  $\Theta$  is the angle between the line drawn to the dipole and the dipole moment vector. A random arrangement of these dipoles would produce no net effect. An orderly arrangement, or even an arrangement which tends on the average to be in one direction, gives a material which is said to be polarized.

3. Polarization

#### a. General

Miner<sup>12</sup>, in considering this from the "displacement" viewpoint, states that the total displacement, or electric flux density, is the vector sum of the electric intensity, E, and  $4\pi$  times the polarization caused by the electric intensity.

$$D = E + 4 * P \tag{11}$$

Rice and Teller<sup>13</sup> explain that an elementary charge, e, producing a field  $e/r^2$ , will emit a certain number of lines of force. The number of lines of force crossing a sphere of radius, r, with the charge at its center is

 $\frac{e}{r_{L}} 4\pi r^{2} = 4\pi e$  (12)

<sup>12</sup>Miner, <u>op</u>. <u>cit.</u>, pp. 9-10.

13F. O. Rice and E. Teller, The Structure of Matter, p. 46.

Hence, the charge due to polarization, is said to be the source of  $4\pi$  lines of force. The term, susceptibility (k) is then defined as the factor relating polarization to the electric intensity causing it.

$$\mathbf{P} = \mathbf{K} \mathbf{E} \tag{13}$$

Susceptibility may be determined in terms of dielectric constant as follows:

$$D = E + 4\pi KE$$

$$D = E (1 + 4\pi K)$$

$$E = E (1 + 4\pi K)$$

$$E = 1 + 4\pi K$$

$$K = \frac{E - 1}{4\pi}$$
(14)

There is, apparently, more than one kind of polarization. Several theories exist, some of which are partially contradictory, and none of which explain all the phenomena observed. Miner<sup>14</sup> says that the dielectric constants of polar substances (those polarized even in the absence of an external field) change with temperature, field strength, and frequency. Whitehead<sup>15</sup> confirms that temperature and frequency affect the dielectric constants of polar substances, but says that field strength does not affect dielectric constant. Sidgwick<sup>16</sup> states that for small fields ordinarily used, the amount of orientation is small and proportional to field strength. For large fields approaching breakdown potentials, this is no longer true.

Various theories of polarization have been summed up by Morgan and Murphy<sup>17</sup>. They list four types of polarization:

1. Electronic Polarization

14 miner, op. cit., p. 18.

15 Whitehead, op. cit., p. 59.

<sup>16</sup>N. V. Sidgwick, Some Physical Properties of the Covalent Link in Chemistry, p. 130.

17E. J. Murphy and S. O. Morgan, "The Dielectric Properties of Insulating Materials," <u>Bell System Technical Journal</u>, XVI (October, 1937), 493-512.

- 2. Atomic Polarization
- 3. Dipole Polarization
- 4. Interfacial Polarization

The first two are known as instantaneous polarizations. They form and disappear in less than  $10^{-10}$  seconds. The last two take longer to occur and give rise to effects which are called absorption effects.

#### b. Electronic Polarization

Electronic polarization is due to the displacement of the center of gravity of the electrons with respect to the nucleus of the atom. This polarization depends upon the number of electrons per unit volume and the force binding them. An example is given of hydrogen, which has two electrons per molecule, and helium which has two electrons per atom. For the same number of electrons per unit volume, hydrogen has four times the susceptibility of helium. This shows that hydrogen electrons are less tightly bound, and are hence more easily polarized. Jones<sup>18</sup> points out that this also leads to greater conductivity on the part of the substance with the less tightly bound electrons. Materials which exhibit only this type of polarization should have dielectric constants which do not vary as frequency is increased up to the range of visible light. This is because of the low inertia of an electron. Electronic polarization is also said to be independent of temperature. Benzene is a material which exhibits only electronic polarization. The electronic polarization theory would explain why the dielectric constants of liquids are, in general, higher than for gases, since there are a greater number of electrons per unit volume in liquids. It would also explain why dielectric constants nearly always increase with pressure, regardless of state.

#### c. Atomic Polarization

Atomic polarization is due to the displacement of ions within a molecule.

18 Jones, op. cit., p. 21.

It occurs up to infra-red frequencies, and has a small positive temperature coefficient. Its upper frequency limit is lower than that of electronic polarization because of the greater inertia of the nuclei. Atomic polarization may be taken as fifteen per cent of the electronic polarization for an average, although it varies widely with the material under consideration.

#### d. Dipole Polarization

Dipole polarization is due to the lining up of the dipoles of a polar substance. Rice and Teller<sup>19</sup> caution that in considering a molecular dipole, only the effect of the external field has been considered, and not the effect of the neighboring dipoles. These other effects are not well defined and are hard to determine. However, their effect is often so small, that random thermal motion will cancel it. It may be seen that at lower temperatures the cancelling thermal motion may be insufficient, and the effect may enter in. The upper limit of dipole polarization is in the radio frequency range. Nost materials are polar. Water, for example, is highly polar, and this is the principal reason given for its high dielectric constant of nearly 80. Some polar radicals given in decreasing order of polar moment are NO2, CN, CO, OH, NH2, Cl, Br, I, and CH2. This is very similar to the dielectric constant list of Walden cited before. The reason for the correlation between this and the degree of association of a solvent is suggested by Creighton and Kochler<sup>20</sup>. When polar molecules are close together, they will tend to associate due to the attraction of opposite charges. The greater the polar moment, the greater the dielectric constant, and the greater the degree of association. A table of values is given in support of this, an excerpt from which follows:

<sup>19</sup>Rice and Teller, <u>op. cit.</u>, p. 50.
<sup>20</sup>Creighton and Koehler, <u>op. cit.</u>, p. 64.

Substance	Degree of Association	Dielectric Constant
Formamide	6.18	84
Water	3.81	81.7
Formic Acid	3.61	58
Methyl Alcohol	3.42	32+5
Ethyl Alcohol	2.74	21.7-27.4
Propionitrile	1.45	26.8
Acetone	1,26	21.3

Exceptions are also noted. Dipole polarization also has a negative temperature coefficient. As temperature decreases, there is less thermal agitation in the molecules, and therefore less agitation to oppose the orientation of the dipoles. Hence, with most polar substances, the dielectric constant tends to increase with decreasing temperature. The thermal agitation, in general, probably prevents a majority of the dipoles from aligning themselves with the field. However, as a number tend to align themselves somewhat, on the average, in their random motion, a net effect is produced. Exceptions are also noted here. Many polar materials have peaks and troughs in a plot of dielectric constant versus temperature, which this theory does not explain. Some nonpolar molecules are carbon disulphide, carbon tetrachloride, benzene, nitrous oxide, hexane, and oxygen. The symmetry of the molecules such as carbon tetrachloride may be seen (Fig. 1) as compared to a polar molecule such as water (Fig. 2).

A resonance effect in polarization is suggested by McCabe.<sup>21</sup> For simplicity, consider a polar particle suspended in a non-polar liquid. If an alternating voltage is applied, the particle will rotate in accordance with the field, but only at certain frequencies will its mass and moment of inertia allow it to resonate with the field. At these frequencies, it will undergo

<sup>&</sup>lt;sup>21</sup>R. P. McCabe, "A Survey of Particle Size Methods," <u>Ceramic Age</u>, L (September, 1947), 155-157, 190.

most complete re-orientation and have its maximum effect on the dielectric constant,

Interfacial or ionic polarization is caused by the accumulation of free ions at the boundaries between solid materials of different conductivities and dielectric constants. This effect occurs only up to radio frequencies. Hence, materials with impurities will show this type of polarization in the lower frequencies. The effect of temperature on interfacial polarization is not known.

Anomalous dispersion is defined as a decrease in dielectric constant with an increase in frequency over a particular band due to the inability to complete polarization in the period of one cycle. This is due to the inertia of the particle which must be aligned in order to complete polarization. Figure 3 is a typical curve given by Miner<sup>22</sup> for a hypothetical material exhibiting all four types of polarization.

e. Application to Solids

Discussions of polar theories are nearly always confined to liquids. Some solids are said to be polar, and others are said to be non-polar, but there seems to be hesitation to apply polar theories to solids in general.

This subject has been investigated by Smyth and Hitchcock<sup>23</sup> They found some substances with a sharp melting point, such as nitrobenzene, ethylene chloride, aniline, and phenol, show a sharp fall of dielectric constant at this point when freezing, and thereafter behave as non-polar or only slightly polar solids. The fall of dielectric constant is very sharp and accompanied

<sup>22</sup>Miner, <u>op</u>. <u>cit.</u>, p. 20.

<sup>23</sup>C. P. Smyth and C. S. Hitchcock, "Dipole Rotation in Crystalline Solids," Journal of the American Chemical Society, LIV (December, 1932), 4631-4647.





Carbon Tetrachloride Molecule FIGURE 1







by a sharp maximum in the loss angle, which may extend over only a few degrees of temperature. The explanation of dielectric constant change is a "freezing in" of the dipoles. No theory exists which enables the calculation of the losses. O ther substances such as hydrochloric acid and dimethyl sulphate, show only a small change of dielectric constant at the freezing point and behave as highly polar solids down to a certain transition temperature. This transition temperature for hydrochloric acid is at minus 17h degrees Centigrade (ninety degrees below its freezing point). At this temperature the dielectric constant falls abruptly as though this were its freezing point. For hydrochloric acid, the dielectric constant falls from 17.5 to 3.0 within a temperature range of one degree Centigrade and is accompanied by a sharp maximum in the loss angle. An explanation is given by Pauling<sup>24</sup> who says that dipoles are free to rotate in the solid state so long as their energy exceeds a value depending on their size and moment of inertia. He also does not give any explanation for the high loss at this point.

#### 4. Crystalline Structure

Crystalline structure has been recognized as a factor in determining dielectric constant. Some authors attribute the super-high dielectric constants to an unusual crystal structure, and unexpected variations in dielectric constant with temperature or other factors are sometimes said to be due to changes in crystalline structure. A few substances, such as sulphur, have a higher dielectric constant in the solid state than in the liquid, and this has also been attributed to the crystalline structure of the solid. However, very little has been done to correlate dielectric constant with the various types of crystalline structure, or to formulate laws of dielectric constant which involve

<sup>&</sup>lt;sup>24</sup>L. Pauling, "The Rotational Motion of Molecules in Crystals," <u>Physical</u> Review, XXXVI (August, 1930), 430-443.

crystalline structure. Hausner<sup>25</sup> suggests that some types of crystal lattices have polar properties, while others do not, even though the same molecules compose them.

Wainer<sup>26</sup> has developed a theory involving ionic radii. Atomic or ionic radius is a factor in determining crystal structure. He represents ions by rigid spheres, which do not indicate the physical size of the ions, but rather the area over which the ion may exert the particular influence under discussions on its neighbors. He redefines the term, polarization, as the ability of some particle in an electric field to exert a distorting force, and assumes that the negative ions are polarizable. The positive ions are acted upon and distorted by the negative ions. Capacity is therefore dependent upon the dimensions of the crystal lattice and the effective ionic radii. If the spacing is such that there is an overlapping of these imaginary spheres, the cause and effect relation is hindered and the dielectric constant is low. As the spacing is increased, the hindering is less, and a maximum in dielectric constant is obtained at a certain optimum distance where the spheres are tangent. As spacing is further increased, the interacting forces are less utilized, and the dielectric constant decreases. Figure 4 shows this mechanism for titanium dioxide. Supporting data is given for the titanates of magnesium, calcium, strontium, and barium, as well as for titanium dioxide.

Rochelle salts have been studied by Sawyer and Tower<sup>27</sup> in regard to their dielectric constant, losses, and piezoelectric properties. One sample gave a

<sup>25</sup>H. H. Hausner, "Research on Barium Titanate in the U.S.S.R.," <u>Ceramic Age</u>, L (September, 1947), 162-164.

<sup>26</sup>Wainer, <u>op</u>. <u>cit.</u>, pp. 343-353.

<sup>27</sup>C. B. Sawyer and C. H. Tower, "Rochelle Salt as a Dielectric," <u>Physical</u> <u>Review</u>, XLV (May, 1934), 654-655.

dielectric constant of 18,000. Valasek<sup>28</sup> found that the high dielectric constant and the piezoelectric properties both disappeared at the same temperature. Sawyer and Tower<sup>29</sup> obtained hysterisis loops on an oscilloscope for mechanically restrained and unrestrained crystals. The loops were larger for the unrestrained crystal, and all loops increased in an area with decreasing temperature. A maximum instantaneous value of dielectric constant of 200,000 was calculated from one of the loops, and the value for the overall loop excluding the saturation region was 10,500. Again correlation with crystal structure is suggested.

<sup>29</sup>Sawyer and Tower, <u>loc.</u> <u>cit</u>.

<sup>&</sup>lt;sup>28</sup>J. Valasek, "Infrared Absorption by Rochelle Salt Crystals," <u>Physical</u> Review, XLV (May, 1934), 654-655.





Ionic Separation Theory

## FIGURE 4

#### D. Anomalous Properties and Dielectric Losses

#### 1. Absorption

One of the anomalous properties is absorption which results in delayed charge or discharge. If a direct potential is applied to a condenser, there will be an initial rush of charging current plus a small leakage current. The leakage current will persist at a constant value, but the charging current normally ceases quickly. However, in some cases it persists for long periods of time, though small and decreasing. Similarly on discharging, there will be an initial rush of current which normally ceases quickly. When absorption has occurred, this current may also persist for long periods at small and decreasing values. If the discharge path is opened during this procedure, and later connected, the discharge current will be higher upon reconnecting than it was when the circuit was broken. This procedure may be continued revealing a logrithmic decrease in initial currents. Two types of absorption have been observed. Reversible absorption occurs when the material gives up its total charge, even though over a very long period of time. Irreversible absorption occurs when the material gives up only a portion of its total charge. The discharge current in this case decays at a rate which is dependent upon the applied potential. Some liquids and composite solids are said to exhibit this phonomena.

An important theory of absorption was given by Maxwell. He considered a dielectric as having properties of capacity and conductivity which are independent, but interact to produce absorption. The substance under consideration cannot be homogeneous, but must have impurities or layers of other substances intervening. As an example, consider a two layer dielectric between conducting plates. (See Fig. 5) Each layer has a thickness of unity. Layer one has a dielectric constant of one and a resistance of  $10^{12}$  ohms, while

layer two has a dielectric constant of two and a resistance of  $\mu \ge 10^{12}$  ohms. Neglecting conduction, at first, assume an electric flux density of one, which must be the same in both dielectrics, to be set up by the applied voltage. Since E, the voltage gradient, equals  $D/\epsilon$ , E<sub>1</sub> equals 1 and E<sub>2</sub> equals .5. These are the voltage gradients due to the dielectric constants alone. Since E is equal to V numerically in this case, the total applied voltage must be 1.5. Neglecting dielectric constants, the voltage of 1.5 will cause a conduction current to flow which will set up a different voltage gradient. The current will be the same in both dielectrics so the voltages across each will be in proportion to their resistances. V1 equals E1 which equals 0.3. V2 equals Ep which equals 1.2. Since D equals EE, the new electric flux densities are D<sub>1</sub> equal to 0.3 and D<sub>2</sub> equal to 2.4. This condition can not exist, since lines of flux eminating from a positive charge must always terminate on a negative. Therefore, there must be another charge present, which is not on the conducting plates. At first, unequal currents must have begun to flow in the dielectrics in accordance with the conditions first described. These caused a charge to accumulate at the boundary allowing the second set of conditions to exist. This charge, represented by  $D_2 - D_1$  is the absorbed charge. Whitehead 30 states that there is some experimental evidence to confirm this theory, but a great deal of evidence that does not confirm it. However, its importance is justified by its basic approach, and it may well be one of several factors involved in the problem.

Einer<sup>31</sup> advances the idea that unusual variations in conductivity may result in absorption. Gases and liquids do not always follow Ohm's Law.

30 Whitehead, op. cit., p. 45.

31<sub>Miner</sub>, op. cit., pp. 27-28.

An increase in electric field results in a smaller increase of current up to a certain point below breakdown. Then the current increases faster than the increase in electric field. No explanation for this is given. Gases and liquids which have high resistivity exhibit a current saturation effect. This is due to the ions, formed by pulling off electrons, being pulled to one of the plates.

#### 2. Dielectric Hysterisis

Dielectric hysterisis has been proposed to explain absorption effects as well as a loss in dielectrics which has been noted to increase with frequency and which is not due to conduction. Schwaiger and Sorenson<sup>32</sup> explain this as a time lag of current - that is, the charging current does not lead the applied voltage by ninety degrees, but by 90 degrees minus  $\boldsymbol{\delta}$ . Figure 6 shows a vector diagram of this condition.  $I_0$  is the theoretical capacitance current. To this is added  $I_n$ , a current which lags  $I_0$  by an angle  $\boldsymbol{\star}$  due to dielectric hysterisis. The resultant I leads the voltage by an angle  $\boldsymbol{\phi}$  and lags  $I_0$  by an angle  $\boldsymbol{\delta}$ .  $I_L$ , the conduction current in phase with the applied voltage must also be added giving  $I_1$ , the final resultant current lagging  $I_0$ by an angle  $\boldsymbol{\delta}_1$ . An increase in  $I_n$  will therefore increase the charging current,  $I_0 + I_c$ , and hence the capacity, as well as increasing the conductivity component,  $I_V + I_L$ .

Miner<sup>33</sup> states that closed loops have been obtained plotting polarization against electric intensity, but that the lag is only temporary and the loop would therefore be dependent on time. He also says that no saturation phenomena is found, although he has previously stated that dielectric constant

<sup>32</sup>A. Schwaiger and R. W. Sorenson, <u>Theory of Dielectrics</u>, pp. 36-38.
<sup>33</sup>Miner, op. cit., pp. 26-27.





FIGURE 5



Dielectric Loss Vector Diagram

FIGURE 6

does wary with field intensity. Whitehead<sup>34</sup> shows such loops attributed to Mouline, and says that others of electric intensity versus total charge have been plotted, but does not show these nor give the source. The analogy with a magnetic hysterisis loop is suggested by both authors, but the validity of the analogy is questioned. Dielectric hysterisis loops which plot field intensity versus total charge for barium titanate are reported by Wul and Goldman.<sup>35</sup> They show that the areas of the loops decrease with temperature. They were taken at fifty cycles. These loops are said to be one of the many unusual properties of this dielectric, which has a super-high dielectric constant.

Anderson and Keane<sup>36</sup> explain the residual charge occurring in the hysterisis loops in terms of resistivity. They first postulate that both resistivity and dielectric constant vary continuously between the electrodes. Poisson's equation is

$$\nabla \cdot \vec{E} \cdot \vec{e}$$
 (15)

where T is charge density. Since variation in only one direction this may be written

$$\frac{\partial}{\partial x} \cdot \overline{E} \cdot \overline{E}$$
 (16)

However, it has now been specified that the dielectric constant  $\boldsymbol{\xi}$ , also varies with the distance, x.

$$\frac{\partial}{\partial x} \left( \xi E \right) = T \tag{17}$$

34 Mnitchead, op. cit., pp. 58-59.

35B. M. Wul and I. M. Goldman, translated by H. H. Hausner, "Dielectric Hysterisis in Barium Titanate," <u>Comptes Rendus (Doklady) de l'Academie des</u> <u>Sciences de l'U.R.S.S., LI (January, 1946), 21-23.</u>

36A. Anderson and T. Keane, "A Study of Dielectric Hysterisis," The London, Edinburgh, and Dublin Philosophical Magazine and Journal of Science, XXIV (February, 1912), 437-443. Ohm's Law in slightly altered form may be written E equals  $\varphi$ , where E is the potential gradient in volts per centimeter,  $\boldsymbol{\iota}$  is the current density in amperes per square centimeter, and  $\boldsymbol{\rho}$  is the resistivity in ohm centimeters. The current density must be a constant, so the preceeding equation may take the form

$$L \frac{\partial}{\partial x} (\mathcal{E} \mathcal{P}) = \mathcal{T}$$
(18)

Integration between the limits  $\boldsymbol{\xi}, \boldsymbol{\beta}$  and  $\boldsymbol{\xi}_{2}, \boldsymbol{\beta}_{2}$ , the values at the respective plates, gives the total charge per unit area of the plates which has been absorbed in the dielectric.

$$q = \iota \left( \epsilon_{k} \rho_{k} - \epsilon_{j} \rho_{j} \right) \tag{19}$$

The residual charge always has the same sign as the primary charge. This leads the authors to the conclusion that the resistance of the dielectric is dependent upon electron density. Continued application of an electric field results in an electron drift which gives different values of resistivity at different points. The charges on the plates are given by:

 $Q_{1} = i \epsilon_{1} \rho_{1}$   $Q_{2} = i \epsilon_{1} \rho_{2}$  (20)

so the ratio of residual to initial charge is

$$\frac{9}{Q_1} = \frac{\xi_2 f_2 - \xi_1 f_1}{\xi_1 f_1}$$
(21)

The authors then state that it appears probable from experiment that  $\boldsymbol{\xi}_{1}$  equals  $\boldsymbol{\xi}_{1}$ , so that

$$\frac{\mathbf{q}}{\mathbf{Q}_{1}} = \frac{\mathbf{P}_{2} - \mathbf{P}_{1}}{\mathbf{P}_{1}}$$
(22)

When the field is removed, the residual charge disappears gradually and  $\rho_i$  approaches  $\rho_i$  in value. They conducted a series of experiments on sulphur and found that residual charge is dependent upon applied voltage and varied closely according to the law

$$\frac{\mathbf{q}}{\mathbf{Q}} = \frac{\mathbf{0.05V}}{\mathbf{V+10}} \tag{23}$$

#### 3. Complex Dielectric Constant

Another approach to the study of dielectric loss, is in the concept of complex dielectric constant. An explanation of this is given by Hamburger.<sup>37</sup> His view is of oscillating dipoles in a varying electric field. A difference in phase occurs between the dipole moment (and hence the polarization) and the electric field. Therefore the moment, the polarization, and the dielectric constant are complex.

$$\varepsilon = \varepsilon' - j\varepsilon''$$
 (24)

In a dielectric under alternating stress, the current has a component which leads the voltage by ninety degrees and results in stored energy. It also has a component in phase with the applied voltage which results in energy loss.  $\boldsymbol{\mathcal{E}}'$  is the ninety degree component, which is normally referred to as the simple dielectric constant and is that which is measured on a bridge. The in phase, or loss, component is  $\boldsymbol{\mathcal{E}}''$ .

$$\mathcal{E}''=\frac{1}{WRC_{o}}$$
 (25)

where **W** is  $2\pi$  times the frequency,  $C_0$  is the capacity of the condenser in a vacuum, and R is the equivalent shunt resistance. Energy loss is expressed as a phase angle,  $\emptyset$ , so that

$$\tan \phi = \frac{\varepsilon''}{\varepsilon'}$$
(26)

Debye<sup>38</sup> gives the following equations for calculating  $\boldsymbol{\varepsilon}'$  and  $\boldsymbol{\varepsilon}''$ 

$$E' = E_{a} + \frac{E_{e} - E_{m}}{1 + [(E_{a} + 2)^{2}/(E_{a} + 2)^{2}]w^{2}\gamma^{2}}$$
 (27)

$$\boldsymbol{\xi}^{\boldsymbol{\pi}} : \left(\boldsymbol{\xi}_{\boldsymbol{\sigma}} - \boldsymbol{\xi}_{\boldsymbol{\varpi}}\right) \frac{\left[\left(\boldsymbol{\xi}_{\boldsymbol{\sigma}} + \boldsymbol{2}\right)\right] \left(\boldsymbol{\xi}_{\boldsymbol{\sigma}} + \boldsymbol{2}\right)^{2}\right] \boldsymbol{w}^{\boldsymbol{\tau}}}{1 + \left[\left(\boldsymbol{\xi}_{\boldsymbol{\sigma}} + \boldsymbol{2}\right)^{2}\right] \left(\boldsymbol{\xi}_{\boldsymbol{\sigma}} + \boldsymbol{2}\right)^{2}\right] \boldsymbol{w}^{\boldsymbol{\tau}} \boldsymbol{\gamma}^{2}}$$
(28)

<sup>37</sup>F. Hamburger, Jr., "Polar Molecules, Their Contribution to Energy Loss in Dielectrics," <u>Physical Review</u>, XXXV (May, 1930), 1119-1124.

38P. Debye, Polar Molecules, p. 91.

where  $\boldsymbol{\xi}_{\bullet}$  is the dielectric constant under direct voltage,  $\boldsymbol{\xi}_{\bullet}$  is the dielectric constant at a very high frequency approaching infinity, and  $\boldsymbol{\gamma}$  is the relaxation time - the time required for all molecules to revert to their random state after removing the impressed field.  $\boldsymbol{\xi}_{\bullet}, \boldsymbol{\xi}_{\bullet}$  and  $\boldsymbol{\gamma}$  vary with temperature, so the values used must be for a given temperature. Crane<sup>39</sup> states that, in practice,  $\boldsymbol{\xi}_{\bullet}$  and  $\boldsymbol{\xi}_{\bullet}$  are usually taken as those values on each side of a region of anomalous dispersion.

Skilling<sup>40</sup> introduces complex dielectric constant in this manner. Consider an electric wave moving through a dielectric in the "y" direction, and whose value is given by

$$E_{y} = E_{max} \mathcal{L}^{Jwt} \mathcal{L}^{\pm} \mathcal{I}^{\chi}$$
(29)

where t is time, and x is distance, and  $\Gamma$  is the propagation function whose value is

$$\int -jw\sqrt{x} \varepsilon \left(1 + \frac{r}{Jw} \varepsilon\right)$$
(30)

Y is the conductivity. The complex dielectric constant is defined as

$$E\left(1+\frac{r}{Jw}E\right) \tag{31}$$

This expression also occurs in the equation of intrinsic impedance

$$N = \sqrt{\frac{1}{E(1+\frac{1}{1})E}}$$
(32)

The more familiar forms are observed by setting  $\mathbf{Y}$  equal to zero in these expressions.

The "Q" of a dielectric, analogous to the "Q" of a tuned circuit, is defined as

$$Q - \frac{\omega \epsilon}{r}$$
 (33)

<sup>39</sup>A. H. Crane, <u>The Dielectric Constant of Petroleum Wax</u>, p. 23. <sup>40</sup>H. H. Skilling, Fundamentals of Electric Waves, p. 127.

#### 4. Variation of Losses With Frequency

Dielectric loss has been shown to increase with increasing frequency for some materials, decrease with increasing frequency for others, and remain constant with frequency for others. Theories have been developed to show why any one of those conditions should exist. Whitehead<sup>h1</sup> explains the hypothesis of K. W. Wagner to show why all of these conditions may exist.

Considering a dielectric to be non-homogeneous, an ideal two layer dielectric is studied as before, for simplicity. An absorption constant, h, is defined.

$$h = \frac{\varepsilon_1 \tau_2 - \varepsilon_2 \tau_1}{\varepsilon_1 \varepsilon_2 (\tau_1 - \tau_2)^2}$$
(34)

and the theory advanced that dielectric loss is proportional to

$$\frac{hwt}{1+hw^2t^2}$$
(35)

A curve may be plotted showing how loss would then vary as is done in Figure 7. It is seen that the tangent of the loss angle reaches a maximum value of  $\frac{h}{2\sqrt{1+h}}$  when wt is  $\sqrt{1+h}$ . This peak may be very broad in some cases. The conclusion is drawn that, for many materials, the value of h is such that very extreme frequencies must be used to get on a different portion of the curve. Hence, over a normal range of frequencies only a single characteristic is noted.

Whitehead, op. cit., pp. 92-102.

#### E. Dielectric Breakdown

When a dielectric is subjected to increasing electric fields, a point is reached where conductivity suddenly increases. The reasons for this are not known, but many theories have been developed. There may be several factors which account for the different types of breakdown.

#### 1. Gases

Breakdown in gases is best understood. There is no appreciable conduction or absorption prior to breakdown and failure occurs at certain definite values of potential gradient. Breakdown voltage is directly proportional to the pressure of the gas. The mechanism of breakdown is explained by Schwaiger and Sorenson42 The current is a movement of electrons, which, in a vacuum, would follow the lines of electric flux. In a gas they can not do this, as they continually collide with gas molecules which deflect them and cause them to follow a zigzag path. When a certain critical gradient is reached, the electrons strike the molecules with sufficient force to dislodge other electrons. The process becomes cumulative until an appreciable current is flowing. Miner43 states that breakdown occurs at a constant value of gradient for gases, but Schwaiger and Sorenson state that the potential gradient required for breakdown is less for greater spacing of the electrodes. This is explained by the longer path for electrons and greater number of collisions which must then occur. There is a time lag in building up this electron avalanche and the lag decreases with greater field strength. Hence the punc-

44Schwaiger and Sorenson, loc. cit.

<sup>42</sup> Schwaiger and Sorenson, op. cit., pp. 20-24.

<sup>43</sup>miner, op. cit., p. 30.

ture strength is greater for a shorter applied stress.

#### 2. Liquids

The breakdown of liquid dielectrics is more complex, and no rules have been laid down which apply to all of them. Attempts have been made to consider liquids as highly compressed gases, but experimental evidence does not support this theory. In general, the breakdown voltage of liquids is not high enough in comparison with gases, as it would have to be were this the case. The liquids differ among themselves in breakdown properties.

#### 3. Solids

The breakdown of solid dielectrics is more complex still. At the critical voltage gradient, the current increases rapidly and burns or punctures the dielectric. The source of charged particles, whose flow constitutes this current, is not definitely known. The critical gradient decreases with increasing temperature and is not affected by changes in pressure below the region of the elastic limit.

Wagner<sup>45</sup> proposes thermal causes for breakdown of solid dielectrics. Again impurities, or non-uniform composition must be considered. Some areas of the dielectric may have lower resistance than others, causing a non-uniform current distribution. The areas of lower resistance are heated more rapidly. If the heat is not all conducted away, the spot becomes hotter and, therefore, probably has lower resistance still, since most dielectric materials have a negative temperature coefficient of resistance. This effect becomes cumulative until breakdown occurs. Joffe<sup>46</sup> states that thermal breakdown may occur in a

46A. F. Joffe, The Physics of Crystals, pp. 154-159.

<sup>45&</sup>lt;sub>K. W. Wagner, "The Physical Nature of the Electrical Breakdown of Solid Dielectrics," <u>Transactions of the American Institute of Electrical Engineers</u>, XLV (June, 1926), 983-997.</sub>

homogeneous material. He cites the work of a number of experimenters who have shown this. Breakdown occurs near the center of the dielectric, since less heat is conducted away from this region, and it takes an appreciable time to occur, since some time is required to heat the dielectric.

Another group of theories assumes a flow of ions in the dielectric. High temperatures would cause ion formation, but at ordinary temperatures, collision ionization would be necessary to account for breakdown. Joffe<sup>47</sup> develops a theory showing that exceeding a critical ion density causes breakdown. Hoover<sup>48</sup> states that there is an equilibrium between electrons leaving the molecules and ions receiving electrons. His theory is that breakdown occurs when ions take on sufficient velocity and the equilibrium is destroyed, causing current flow.

Disruptive theories, analogous to mechanical breakdowns, are also advocated. Miner<sup>49</sup> notes that dielectrics which have a low atomic weight and few electron orbits, have higher dielectric strengths. The forces holding the electrons are greater, and more energy is required to disrupt the molecules. Materials of high atomic weight, are poorer insulators relatively. The destruction of the lattice arrangement of crystalline substances may also be a cause. Sodium chloride is cited, which is an insulator in crystalline form, but a conductor otherwise. Breakdown may also be affected by mechanical stress. Breakdown voltage has been reduced to ten per cent of its nominal value by mechanical stress near the elastic limit. Cracks in a material often cause the breakdown voltage to be less. Another factor is electrical fatigue, which seems to be

47 Ibid., pp. 162-170.

48p. L. Hoover, "The Mechanism of Breakdown of Dielectrics," <u>Transactions</u> of the American Institute of <u>Electrical Engineers</u>, XLV (June, 1926), 983-997. <sup>49</sup>Miner, op. cit., p. 35.

analogous to mechanical fatigue. A repetition of impulse voltages will tend to lower the breakdown voltage if the impulses are eighty per cent of the single impulse breakdown voltage according to Peek.<sup>50</sup> The damage in general increases with time and is permanent, although some materials have shown an improvement after a "rest" period.

Moon and Morcross<sup>51</sup> have proposed that each of these three classes of theories is true in its own temperature range. They showed the three types of breakdown occurring in glass, and summarized their results in the graph of Figure 8. Investigation of other materials showed these regions to vary in temperature range, as in the following table.

Material	Disruptive	Ionic	Thermal	lst Transf. Temp.	2nd Tran <b>sf.</b> T <b>emp.</b>
	Kv/cm	K <b>v/cm</b>	Kv/cm	degrees C.	degrees C.
Fused quartz	5000	1815	560	- 31	270
Pyrex	4800	1050	200	- 20	140
G-1 Glass	3100	1200	102	✤ 22	150
Cover Glass	-	730	60	+ 20	165
Lime Glass	4500	355	32	- 33	217
Celluloid (125)	2500	420		- 10	140
India ruby mica	10600	-	-	+100	-

Another possible mechanism of solid breakdown is explained by Del Mar, Davidson, and Marvin.<sup>52</sup> A particle of higher dielectric constant is imbedded in the dielectric. It will concentrate lines of electric flux to itself, creating points of excess electric flux density. Decomposition takes place and conducting needles form. An insignificant foreign particle may gradually but

<sup>50</sup>F. W. Peek, <u>Dielectric Phenomena in High-Voltage Engineering</u>, p. 243.

<sup>51</sup>P. H. Moon and A. S. Norcross, "Three Regions of Dielectric Breakdown," <u>Transactions of the American Institute of Electrical Engineers</u>, XLIX (April, 1930), 755-765.

<sup>52</sup>W. A. Del Mar, W. F. Davidson, and R. H. Harvin, "Electric Strength of Solid and Liquid Dielectrics," <u>Transactions of the American Institute of Elec-</u> trical Engineers, XLVI (June, 1927), 1049-1061. cumulatively destroy the insulation in this way. Confirmation is found in examination of many solid dielectrics in which breakdown has occurred. The punctures resemble a series of needle points thrust through the dielectric.



Wagner's Dielectric Loss Theory

FIGURE 7



FIGURE 8

#### EXPERIMENTAL PROCEDURE

#### A. Purpose and Background

It was decided to conduct an investigation into the dielectric properties of sulphur. Sulphur was chosen for study since it is one of the few chemical elements which are good insulators, it has an interesting crystalline structure, and it is easily obtainable. The study of a chemical element was thought desirable since this simplified structure might make easier the interpreting of the results. Other chemical elements which are insulators are phosphorous and boron. Phosphorous is not easy to handle and boron is not readily obtainable.

Sulphur is the sixteenth element in the periodic table, and has an atomic weight of thirty-two. Data on its structure is given by Hackh and Grant.<sup>1</sup> In the solid state it exists as a molecule composed of eight atoms in a ring arrangement as shown in Figure 9. Below ninety-six degrees Centigrade, the crystalline form is the rhombic, octahedral pattern of Figure 10. The inter-axial angles,  $\prec$ ,  $\beta$ , and  $\gamma$  are all ninety degrees. The axes, a, b, and c are all of different length. Above ninety-six degrees Centigrade and up to its melting point of one hundred fourteen degrees Centigrade, it exists in the monoclinic, prismatic form of Figure 11. The interaxial angles,  $\prec$ , and  $\checkmark$  are ninety degrees, but  $\beta$  is an acute angle. The axes, a, b, and c, are of unequal length.

Mellor<sup>2</sup> gives the following data on the electrical resistivity of sulphur. R. Therlfall found the resistivity of crystalline sulphur at seventyfive degrees Centigrade to be  $6.8 \times 10^{25}$  ohm-centimeters and that this value

1. W. D. Hackh and J. Grant, Chemical Dictionary, pp. 262-264.

<sup>2</sup>J. W. Mellor, <u>A</u> <u>Comprehensive</u> <u>Treatise</u> <u>on</u> <u>Inorganic</u> <u>and</u> <u>Theoretical</u> <u>Chemistry</u>, pp. 77-79.

decreased with increasing temperature. H. Heumann gives  $2 \times 10^{18}$  to  $2 \times 10^{19}$  ohm-centimeters as the resistivity at ordinary temperatures. Liquid sulphur has increasing conductivity up to one hundred sixty degrees Centigrade according to D. H. Black. It then falls to a minimum at one hundred eightyfive degrees, and thereafter increases. This behavior corresponds to viscosity changes. The resistivity at one hundred sixty-three degrees is given as 7.5 x  $10^{10}$  ohm-centimeters. Values at low temperatures are not given.

Mellor<sup>3</sup> also gives the following data on the dielectric constant of sulphur. It is typical of much of the information found on dielectric constants in general. L. Blotzmann measured the dielectric constants of sulphur crystals in the directions of the three axes and found them to be respectively h.773, 3.970, and 3.811. F. Rosetti and M. Faraday gave 2.24 for the dielectric constant; A. Wullner 2.88 to 3.21; F. Rosetti, 1.81; J. E. H. Gordon, 2.5793; J. J. Thomson, 2.4; C. B. Thwing, 2.69; M. Lefebore, 2.7; W. Schmidt, 3.95 for a wavelength of seventy-five meters for freshly cast and after ageing, 3.90; and R. Fellinger gave 4.05 and 3.60 for infinite wavelength.

The electrical moment of the sulphur molecule was measured by Taylor and Rideal who found it to be 7.5 x  $10^{-18}$  electrostatic units.

The tests consisted of measuring the dielectric constant of sulphur and, where possible, the losses, under varying temperature, frequency, and electrostatic stress. The method used was to measure the capacity of a condenser with a dielectric of sulphur, and compare it with the capacity of the same condenser with an air dielectric. The tests were made, not with the object of determining values of dielectric constant which are absolutely correct, but to obtain values which are relatively correct, so that the variations in

<sup>3</sup>Ibid., pp. 77-79.



dielectric constant may be studied and reasons for these variations deduced. Further the work has been done with a view of seeking material which requires additional study, and possibly special equipment not now available, so that others who follow in this project may be guided toward more fruitful study.

#### B. Equipment and Procedure

Capacity was measured on a General Radio Type 821-A Twin-T Measuring Circuit. It is a null-type instrument for measuring impedances in the range of 460 kilocycles to 40 megacycles. Both susceptance and conductance are measured. The dial is calibrated directly in micromicrofarads of capacity, however. The conductance is read from a dial calibrated in micromhos for frequencies of 1, 3, 10, and 30 megacycles. At other frequencies, a ratio may be applied to obtain the true conductance. The error is given as plus or minus 0.1% plus 2 micromicrofarads for capacity, and 2% of the reading plus 0.1% of the full scale value for conductance. The circuit is shown in Figure 12. There are two parallel paths from the generator to the detector, a-b-c, and a-d-c. No energy is transferred from the generator to the detector when the transfer impedances (defined as the ratio of input voltage to output current when the output terminals are short-circuited) are equal and opposite. The circuit is initially balanced to a null. The condenser to be measured is connected across the "Unknown" terminals. The conductance and susceptance condensers are again adjusted for a null, and the difference from the original readings gives the desired values.

As the signal source, a United States Army Frequency Meter BC-221-M, manufactured by the Bendix Radio Corporation, was used. This instrument gives an output over the range of .125 to 20 megacycles. A Signal Corps Receiver, BC-348-R was used for a detector. Its frequency range is .2 to .5 megacycles and 1.5 to 18 megacycles. A crystal filter in this receiver was helpful in reducing noise and making the null more easily discernable. However, the amount of noise output from the receiver was still too great to permit maximum accuracy, so a filter was connected to the receiver output. This filter tuned sharply to about 1,000 cycles, so that adjustment of the receiver output to

this frequency resulted in a signal which contained an extremely small amount of noise. The output was observed both on earphones and on an oscilloscope, a Du Mont Type 146 which has a three-inch tube. It was found that a phase shifting circuit to put a circular pattern on the oscilloscope further aided in more accurate detection of the null.

The experiment required applying a high direct voltage to the test condenser from a rectifier having a voltage output variable between 108 and 15,000 volts. This could not have been done directly without damage to the impedance measuring circuit. A special circuit to accomplish this is shown in Figure 13. A radio frequency transformer was necessary to isolate the measuring circuit, and a capacitor kept the high voltage from appearing on the transformer. The circuit was also protected by fuses as shown. The readings taken with this procedure were recorded and corrected by placing a previously calibrated variable capacitor in place of the test capacitor and repeating the readings. Since the proper variable conductance could not be obtained, an ammeter was used to measure leakage current directly.

The variation of temperature was best accomplished by using an immersiontype air condenser. A beaker could then be filled with the dielectric material and the test condenser immersed in it, and heating and cooling were thus facilitated.

It was found that the air content of powdered sulphur was quite high and reduced the apparent dielectric constant by a large amount. The amount of this air would vary with temperature, which would invalidate the results on a relative basis. It was decided, therefore, to first melt the sulphur and allow it to solidify in the condenser in a hard form.

The test condenser was carefully inspected for very nearly equal plate spacing. It was then boiled in distilled water to remove contaminating materials.

However, other sources of error are still present. Due to the construction of the condenser, all the capacity is not between the plates themselves. However, this is a constant factor. The spacing was probably not exactly the same throughout, which would lead to unequal stress on the dielectric material. This also was a constant factor, but might have lead to making larger the variation in dielectric constant which it is desired to study, could it have been corrected for. Air was probably present to some degree in the solidified sulphur. However, this effect must have been negligible, since the sulphur was remelted many times during the course of the experiment, and the dielectric constant would return to its original value on solidifying. For the same reason, if any chemical reaction between the condenser plates and the sulphur had resulted, its effect was negligible on the dielectric constant. The effect of impurities which may have been in the sulphur is not known. It has been pointed out previously that several theories of the behavior of dielectric constants are based on the presence of impurities, even the extremely minute amounts of impurities that still exist in the purest substances that may be chemically produced. Therefore, this effect can not be disregarded in the analysis of the results. The sulphur tested was commercially obtained "Chemically Pure" sulphur, and no attempt was made at further purification. Any inaccuracy in the calibration of frequency of the signal generator would have been negligible since dielectric constant varied very little over very wide ranges of frequency. The same may be said for any variation in the output voltage of the signal generator with frequency. Another source of error was the uneven heating of the sulphur. This effect would have been appreciable in determining the absolute value of dielectric constant, since the thermometer did not read the exact value of temperature of the sulphur between the plates of the condenser. Again results were relatively correct, however, and readings could be repeated.







FIGURE 13

### C. Data

#### Table I

#### Dielectric Constant versus Temperature

Frequency constant at 0.5 megacycles, conductivity too low to measure in all cases, capacity in air 56.7 micromicrofarads.

Temp.	Capacity	Dielectric	Temp.	Capacity	Dielectric
Deg. Cent.	uuf	Const.	Deg. Cent.	uuf	Const.
115	150,2	2.649	7	156.0	2.751
110	152.3	2.686	2	155.8	2.748
105	153.8	2.712	- 4	155.6	2.744
100	154.9	2.732	- 9	155.3	2.739
95	156.0	2.751	-15	154.9	2.732
85	156.2	2.755	-21	154.2	2.720
75	156.2	2.755	-26	153 <b>.1</b>	2.700
65	156.2	2.755	-32	151.0	2,663
55	156.3	2.757	-37	150.3	2.651
45	156.3	2.757	-43	150.0	2.646
35	156.3	2.757	-46	150.0	2.646
25	156.3	2.757	-51	150.2	2.649
18	156.3	2.757	-57	150.4	2.653
13	156.2	2.755	- <b>,</b>	·	

#### Table II

#### Dielectric Constant versus Temperature

Frequency constant at 1.5 megacycles, conductivity too low to measure in all cases, capacity in air 56.7 micromicrofarads.

Temp. Deg. Cent.	Capacity uuf	Dielectric Const.	Temp. Deg. Cent.	Capacity uuf	Dielectric Const.
115	150.1	2.647	7	156 <b>.1</b>	2 <b>.753</b>
110	152.2	2.684	2	155.9	2.750
105	154.0	2.716	- 4	155.6	2.744
100	155 <b>.1</b>	2.735	- 9	155.3	2.739
95	156.0	2.751	-15	154.9	2.732
85	156.2	2.755	-21	154.3	2.721
75	156.2	2.755	-26	153.2	2.702
65	156.2	2.755	32	151.0	2.663
55	156.3	2.757	-37	150.4	2.653
45	156.3	2,757	-1.3	150.1	2.647
35	156.3	2.757	-16	150.0	2.616
25	156.3	2.757	-51	150.1	2.647
าอิ	156.3	2.757	-57	150.3	2.651
13	156.2	2.755			

#### Dielectric Constant versus Temperature

Frequency constant at 2,5 megacycles, conductivity too low to measure in all cases, capacity in air 56,7 micromicrofarads.

Capacity	Dielectric	Temp.	Capacity	Dielectric
uuf	Const.	Deg. Cent.	uuf	Const.
150.1	2.647	7	156.1	2.753
152.4	2.688	2	155.9	2.750
153.9	2.714	- 4	155.6	2.744
155.1	2.735	- 9	155.3	2.739
156.1	2.753	-15	154.9	2.732
156.2	2.755	-21	154.2	2.720
156.2	2.755	-26	153 <b>.1</b>	2,700
156.2	2,755	-32	150.9	2.661
156.3	2.757	-37	150.3	2.651
156.3	2.757	-43	150.0	2.646
156.3	2.757	-46	150.0	2.646
156.3	2.757	-51	150.2	2.649
156.2	2.755	-57	150.3	2,651
156,2	2.755			
	Capacity uuf 150.1 152.4 153.9 155.1 156.1 156.2 156.2 156.2 156.3 156.3 156.3 156.3 156.3 156.3	Capacity         Dielectric           uuf         Const.           150.1         2.647           152.4         2.688           153.9         2.714           155.1         2.755           156.1         2.755           156.2         2.755           156.3         2.757           156.3         2.757           156.3         2.757           156.3         2.757           156.3         2.757           156.3         2.757           156.2         2.755           156.3         2.757           156.3         2.757           156.3         2.757           156.2         2.755           156.2         2.755	CapacityDielectricTemp.uufConst.Deg. Cent. $150.1$ $2.647$ 7 $152.4$ $2.688$ $2$ $153.9$ $2.714$ $-4$ $155.1$ $2.735$ $-9$ $156.1$ $2.753$ $-15$ $156.2$ $2.755$ $-21$ $156.2$ $2.755$ $-26$ $156.2$ $2.755$ $-32$ $156.3$ $2.757$ $-37$ $156.3$ $2.757$ $-43$ $156.3$ $2.757$ $-46$ $156.3$ $2.757$ $-51$ $156.2$ $2.755$ $-57$	Capacity uufDielectric Const.Temp. Deg. Cent.Capacity uuf $150.1$ $2.647$ 7 $156.1$ $152.4$ $2.688$ 2 $155.9$ $153.9$ $2.714$ $-4$ $155.6$ $155.1$ $2.735$ $-9$ $155.3$ $156.1$ $2.753$ $-15$ $154.9$ $156.2$ $2.755$ $-21$ $154.2$ $156.2$ $2.755$ $-26$ $153.1$ $156.2$ $2.755$ $-32$ $150.9$ $156.3$ $2.757$ $-37$ $150.3$ $156.3$ $2.757$ $-46$ $150.0$ $156.3$ $2.757$ $-51$ $150.2$ $156.3$ $2.757$ $-51$ $150.2$ $156.3$ $2.757$ $-57$ $150.3$ $156.2$ $2.755$ $-57$ $150.3$ $156.2$ $2.755$ $-57$ $150.3$ $156.2$ $2.755$ $-57$ $150.3$ $156.2$ $2.755$ $-57$ $150.3$

#### Table IV

#### Dielectric Constant versus Temperature

Frequency constant at 4.0 megacycles, conductivity too low to measure in all cases, capacity in air 56.7 micromicrofarads.

Temp. Deg. Cent.	Capacity uuf	Dielectric Const.	Temp. Deg. Cent.	Cap <b>acity</b> u <b>uf</b>	Dielectric Const.
115	149.9	2.644	7	156.1	2.753
110	152.3	2.686	2	155.9	2.750
105	153.9	2.714	- 4	155.6	2.744
100	155.3	2.739	- 9	155.3	2.739
95	156.0	2.751	-15	154.9	2.732
85	156.1	2.753	-21	154.3	2.721
75	156.1	2.753	-26	153.2	2.702
65	156.2	2.755	-32	151.0	2.663
5 <b>5</b>	156.2	2.755	-37	150.3	2.651
45	156.2	2.755	-43	159.1	2.647
35	156.2	2.755	-46	150.0	2.646
25	156.2	2.755	-51	150.1	2.647
18	156.2	2.755	-57	150.3	2.651
13	156.2	2.755	2.1		-

#### Dielectric Constant versus Temperature

Frequency constant at 6,0 megacycles, conductivity too low to measure in all cases, capacity in air 56.7 micromicrofarads.

Temp.	Capacity	Dielectric	Temp.	Capacity	Dielectric
Deg. Cent.	uuf	Const.	Deg. Cent.	uuf	Const.
115	149.9	2.64	7	155.9	2.750
110	152.3	2.686	2	155.7	2.746
105	153.9	2.714	- 4	155.5	2.743
100	155.4	2.741	- 9	155.2	2.737
95	155.9	2.750	-15	154.9	2.732
85	156.1	2.753	-21	154.2	2.720
75	156.1	2.753	-26	153.2	2.702
65	156.1	2.753	-32	151.1	2.665
55	156.2	2.755	-37	150.3	2.651
45	156.2	2.755	-43	150.1	2.647
35	156.2	2.755	-46	150.0	2.646
25	156.2	2.755	-51	150.1	2.647
18	156.2	2.755	-57	150.3	2.651
13	156.1	2.753			-

#### Table VI

#### Dielectric Constant versus Temperature

Frequency constant at 8.0 megacycles, conductivity too low to measure in all cases, capacity in air 56.7 micromicrofarads.

Temp. Deg. Cent.	Capacity uuf	Dielectric Const.	Temp. Deg. Cent.	Capacity uuf	Dielectric Const.
115	149.7	2,640	7	155.7	2.746
110	152.1	2.683	2	155.5	2.743
105	153.7	2.711*	- 4	155.2	2.737
100	155.1	2.735	- 9	154.9	2.732
95	155.6	2.744	-15	154.5	2.725
85	155.8	2.748	-21	154.0	2.716
75	155.9	2.750	-26	153.1	2.700
65	155.9	2.750	-32	151.1	2.665
55	156.0	2.751	-37	150.3	2.651
45	156.0	2.751	-43	150.0	2.645
35	156.0	2.751	-46	149.9	2.644
25	156.0	2.751	-51	150.2	2.649
18	156.0	2.751	-57	150.3	2.651
13	155.9	2.750	2.1		-

#### Table VII

### Dielectric Constant versus Direct Voltage

Temperature constant at 25 degrees Centigrade, capacity in air 57.1 micromicrofarads, frequency 1.5 megacycles.

D. C. Fotentia Volts	al Capacity uuf	Dielectric Const.	Tot. Charge uucoulombs	D. C. Current uamps
0	157.6	2.760	0	0
108	157.6	2.760	+ 17,021	0.5
324	157.6	2.760	51,062	0.5
540	157.6	2.760	85,104	0.5
765	157.6	2.760	120,564	0.5
972	157.6	2.760	153,187	0.5
1170	157.6	2.760	184, 392	0.5
1296	157.6	2.760	204,250	0,5
1413	157.6	2.760	222,689	0.5
1458	157.6	2,760	229,781	0.5
1512	157.6	2.760	238,291	0.5
1566	157.5	2.758	246,799	10.0
1.620	157.5	2.758	255, 304	20.0
1566	155.0	2.714	246,866	1.0,0
1512	155.3	2.720	238,488	0.5
1458	156.1	2.734	230.081	0.5
1395	157.1	2.751	220,215	0.5
1296	157.6	2.760	204,637	0.5
1188	157.6	2.760	187,616	0.5
972	157.6	2.760	153,575	0.5
756	157.6	2.760	119,533	0.5
522	157.6	2.760	82,655	0.5
324	157.6	2.760	51,450	0.5
108	157.6	2.760	17,408	0.5
0	157.6	2,760	÷ 387	0
- 108	157.6	2.760	- 16,634	- 0.5
- 324	157.6	2,760	- 50,676	_ 0.5
- 540	157.6	2,760	- 84,718	- 0.5
- 756	157.6	2.760	- 118,759	- 0.5
- 972	157.6	2.760	- 152,801	- 0.5
-1188	157.6	2.760	- 186,842	- 0.5
-1305	157.6	2,760	- 205,282	- 0.5
-1404	157.6	2.760	- 220,884	- 0.5
-1458	157.6	2.760	- 229, 394	- 0.5
-1521	157.6	2.760	- 239,323	- 0.5
-1566	157.6	2.760	- 246,415	-10.0
-1620	157.5	2,758	- 254,923	-20.0
-1566	155.0	2.714	- 246,585	-10.0
-1512	155.2	2.718	- 238,110	- 0.5
-1458	155.8	2.729	- 229,713	- 0.5
-11,04	156.2	2.736	- 221,289	- 0.5
-1314	157.0	2.750	- 207,195	- 0.5
-1188	157.6	2.760	- 187, 375	- 0.5

## Table VII (Continued)

D.	C. Potential Volts	Capacity uuf	Dielectric Const.	Tot. Charge uucoulombs	D. C. Current uamps
		ז ביז ג	2.760	יין בי און אין די	0 4
	- 756	157 6	2.760		
	- []0	15/20	2.760	- 119,292 St 250	
	- 540	101+0 107 6	2.760	= 09,290 51,200	= 0.9
	- )24	15/+0	2 760	- <u>51,209</u>	- 0.5 0.5
	- 100	15(•0	2.700	- 101e)1 -	- 0.5
	109	15/00	2. (00	- <u>1110</u>	0 0 r
	★ TOO	15/+0	2.760	+ 10,007	+ U.5
	524	15/00	2.100	50,911 81. of 0	0.5
	540	15/.0	2.100	04,959	0.5
	(05	15/.0	2.100	120,419	0.5
	90 <u>1</u>	15/.0	2.100	154,400	0.5
	1100	15/.0	2.700	107,003	0.5
	1295	157.6	2.760	204,104	0.5
	1/10/1	157.0	2.760	221,125	0.5
	1512	157.6	2.760	238,140	0.5
	1566	157.5	2.758	246,654	10.0
	1620	157.5	2.758	255,159	20.0
	1566	154.9	2.713	246,724	10.0
	1512	155.3	2.720	238,348	0.5
	1458	155.7	2,727	229,951	0.5
	1413	156.1	2.734	222,936	0.5
	1296	156.7	2.744	204,637	0.5
	1188	157.6	2.760	187,665	0.5
	972	157.6	2.760	153,623	0.5
	756	157.6	2.760	119,582	0.5
	522	157.6	2.760	82,703	0.5
	324	157.6	2.760	51,498	0.5
	108	157.6	2.760	17,457	0,5
	0	157.6	2.760	+ 436	0
	- 972	157.6	2.760	- 152,75 <b>1</b>	- 0.5
	-1458	157.6	2.760	- 229, 345	- 0,5
	-1566	157.6	2.760	- 246, 365	-10.0
	-1620	157.5	2.758	- 254,873	-20.0
	-1566	154.9	2.713	- 246,438	-10,0
	-1512	155.2	2.718	- 238,066	- 0,5
	-1458	155.8	2.729	- 229,669	- 0.5
	-1404	156.2	2.736	- 221,245	- 0.5
	-1296	157.1	2.751	- 204.326	- 0.5
	- 972	157.6	2.760	- 153.345	- 0.5
	0	157.6	2.760	- 158	0

### Table VIII

## Dielectric Constant versus Direct Voltage

Temperature constant at 70 degrees Centigrade, frequency 1.5 megacycles, capacity in air 57.1 micromicrofarads.

D.	C. Potential	Capacity	Dielectric	Tot. Charge	D. C. Current
	Volts	uuf	Const.	uucoulombs	uamps
	0	157.5	2.758	0	0
	108	157.5	2.758	17.010	0.5
	333	157.5	2,758	52.147	0.5
	51.0	157.5	2,758	85.050	0.5
	765	157.5	2.758	120,1.88	0.5
	972	157.5	2.758	153,090	0.5
	1188	157.5	2.758	187,110	0.5
	1296	157.5	2.758	201.120	0.5
	1/10/1	157.5	2.758	221,130	0.5
	14.67	エンT●ン コピア ピ	2 758	231,052	0.5
	1501	エンイルン コピア ピ	2 758	230,058	
	1566	157 1	2 756	216.643	20.0
	1500	エン/#44 コピレ O	20173	238 211	10.0
	1148	エン4●フ コビビ つ	2 790	220 825	
	1450	エラフ <b>・</b> フ コピム コ	<i>∠</i> . <i> </i> ∠∪ ○ 737	2270000	
	1404	190 <b>, 3</b>	4.121	2219426	
	1350	12(11	2. ()L	212,900	
	1207	15(•5	2,750	203,050	0.5
	1100	157.5	2.750	107,450	0.5
	972	157.5	2.758	153,438	0,5
	756	157.5	2.758	119,418	0,5
	531	157.5	2.758	83,980	0.5
	324	157.5	2.758	51,378	0.5
	108	157.5	2.758	17,358	0.5
	0	157.5	2.758	+ 348	0
	- 108	157.5	2.758	- 16,662	- 0.5
	- 3 <b>3</b> 3	157.5	2.758	- 52,099	- 0.5
	- 540	157.5	2.758	- 84,072	- 0,5
	- 756	157*5	2.758	- 118,722	- 0.5
	- 981	157•5	2.758	- 154,159	- 0.5
	-1188	157.5	2.758	- 186,762	- 0.5
	-1296	157.5	2.758	- 203,772	- 0.5
	-1404	157*5	2.758	- 220,782	- 0.5
	-1458	157.5	2.758	- 229,287	- 0.5
	-1512	157.5	2.758	- 237,792	-10.0
	-1566	157.4	2.756	- 246,294	-20.0
	-1512	154.8	2.711	- 237,865	-10.0
	-1458	155.2	2.718	- 229.495	- 0.5
	-1395	155.8	2.729	- 219.698	- 0.5
	-1350	156.6	2.743	- 212,669	- 0.5
	-1296	157.0	2.750	- 204.202	- 0.5
	-1188	157.5	2,758	- 187.219	- 0.5
	- 981	157.5	2.758	- 154.617	- 0.5
	- 756	157.5	2.758		
	- 51	157 K	2 758	- 82 7ho	
		-21+2		- 0,0,144	- 0.9

# Table VIII (Continued)

D. C. Potential	Copacity	Dielectric	Tot. Charge	D. C. Current
AOT 02	uut	00113.0-		, and the second s
- 324	157.5	2,758	- 51,139	- 0.5
- 108	157.5	2.758	- 17,119	- 0.5
0	157•5	2.758	- 109	0
+ 108	157•5	2.758	+ 16,901	+ 0.5
324	157.5	2.758	50,921	0.5
558	157.5	2,758	87,776	0,5
765	157.5	2.758	120,379	0.5
972	157.5	2.758	152,981	· 0.5
1188	157.5	2.758	187,001	0.5
1296	157.5	2.758	204,011	0.5
1413	157.5	2.758	222,439	0.5
11,58	157.5	2.758	229,526	0.5
1512	157.5	2.758	238,031	10.0
1566	157.4	2.756	246,533	20.0
1512	154.8	2.711	238,104	10.0
1458	155.1	2.716	229,737	0.5
1404	155.7	2.727	221, 345	0,5
1350	156.5	2.741	212,916	0.5
1305	157.1	2.751	205,860	0.5
1197	157.5	2.758	188,871	0.5
972	157.5	2.758	153,434	0.5
756	157.5	2.758	119,414	0.5
522	157.5	2.758	82,559	0.5
324	157.5	2,758	51,374	0.5
108	157.5	2.758	17,354	0.5
0	157.5	2.758	<ul> <li>3144</li> </ul>	0
- 756	157.5	2.758	- 118,726	- 0.5
-1458	157.5	2.758	- 229,291	- 0.5
-1512	157.5	2.758	- 237,796	-10.0
-1566	157.4	2.756	- 246,298	-20.0
-1512	154.8	2.711	- 237,869	-10.0
-1458	155.2	2.718	- 229,499	- 0.5
-1404	155.7	2.727	- 221,105	- 0.5
-1359	<b>156.5</b>	2.741	- 214.080	- 0.5
-1296	157.1	2.751	- 204,202	- 0.5
-1188	157.5	2.758	- 187,213	- 0.5
- 756	157.5	2.758	- 119,173	- 0.5
e	157.5	2.758	- 103	0

#### Table IX

### Dielectric Constant versus Direct Voltage

Temperature constant at minus 55 degrees Centigrade, frequency 1.5 megacycles, capacity in air 57.1 micromicrofarads.

D.	C. Potential	Capacity	Dielectric	Tot. Charge	D. C.	. Current
	VOT 0B	uui	CONSU.	uucouromos	، ج	uunµs. Ъ**
					c.	0
	0	152.0	2.662	0	0	0
	108	152.0	2.662	16,416	40	40
	324	152.0	2.662	49,248	45	45
	540	152.0	2,662	82 <b>,0</b> 80	50	50
	756	152.0	2.662	114,912	60	60
	972	152.0	2.662	147,744	80	80
	1134	152.0	2.662	172,368	100	1.00
	1188	151.9	2.660	180,573	110	110
	1134	149.5	2.618	172,436	110	110
	1080	149.9	2 <b>.625</b>	164,352	105	105
	1026	150,8	2.641	156,233	100	100
	972	151.7	2.657	148 <b>,0</b> 65	90	90
	864	152.0	2.662	131,666	80	80
	765	152.0	2.662	116,618	70	70
	540	152.0	2,662	82,418	60	60
	324	152 <b>.0</b>	2,662	49,586	50	50
	108	152.0	2.662	16,754	30	30
	0	152.0	2,662	+ 338	0	0
	- 108	152.0	2.662	- 16,078	-110	- 40
	- 324	152.0	2.662	- 48,910	- 70	- 50
	- 549	152.0	2.662	- 83,110	- 60	- 50
	- 765	152.0	2.662	- 115,942	- 70	- 70
	- 972	152.0	2.662	- 147,406	-100	-100
	-1134	152.0	2.662	- 172,030	-120	-120
	-1188	151.9	2.660	- 180,235	-145	-160
	-1134	149.4	2.616	- 172,100	-170	-185
	-1080	149.7	2.622	- 164,024	-175	-175
	-1026	150.5	2.636	- 155,919	-160	-160
	- 981	151.3	2.650	- 149,129	-150	-150
	- 004	152.0	2.662	- 131,386	-140	-140
	- (50	152+0	2.062	- 114,970	-125	-125
	- 540	152.0	2.662	- 62,136	-110	-110
	224	152.0	2,062	- 49,306	-100	-100
	- 100	152.0	2.002	— <u>тој</u> ц(ц	- 60	- 15
	9 108	152.0	2,002	- 50	0	С ГО
	20) 	152.0	2.002	+ 10,350	+270	+ 50
	510	152.0	2 662	47 <b>5</b> 170	120	100
	756	152.0	2 662	116 8¢6	120	100
	972	152 A	2 662	1)7 626	120	100
	113	152 0	2 662	172 310	130	120
	1188	יבע-גע זהוס	2.660	180.515		יעב חו\ר
	113		2 676	170 280	1 20	1 20 1 40
	+++	⊥47 <b>44</b>	C OTO	100 <b>6</b> 21T	JO L	という

### Table IX (Continued)

D.	C. Potential	Capacity	Dielectric	Tot. Charge	D. C.	Current
	Volts	uuf	$Const_{\bullet}$	uucoulombs	ູບ	lamps.
					a*	b***
	1080	149.6	2.620	164,307	110	110
	1026	150.4	2.634	156,207	100	100
	972	151.4	2.651	148,058	90	90
	864	152.0	2.662	131,675	80	80
	765	152.0	2.662	116,627	70	70
	540	152.0	2.662	82,427	60	60
	324	152.0	2.662	49,595	50	50
	108	152.0	2.662	16,762	30	30
	0	152.0	2.662	+ 347	0	0
	- 108	152.0	2.662	- 16,069	-100	- 40
	- 324	152.0	2 <b>.</b> 66 <b>2</b>	- 48,091	- 75	- 50
	- 540	152.0	2.662	- 81,733	- 65	- 65
	- 756	152.0	2,662	- 114,565	- 70	- 70
	- 972	152.0	2.662	- 147, 397	-100	-100
	-1134	152.0	2.662	- 172,021	-125	-125
	<b>-11</b> 88	151.9	2.660	- 180,226	-135	-150
	-1134	149.4	2.616	- 172,091	-160	<b>1</b> .80
	-1080	149.6	2 <b>.620</b>	- 164,018	-175	-180
	-1026	150.2	2.630	- 155,923	-150	-150
	- 972	150.9	2.643	- 147,793	-140	- <u>1</u> 40
	- 864	151.5	2.653	- 131,464	130	-130
	- 765	152.0	2.662	- 116, 441	-120	-120
	- 540	152.0	2 <b>.662</b>	- 82,241	-110	<b>-1</b> 10
	- 324	152.0	2.662	- 49,409	-100	-100
	- 108	152.0	2,662	- 16,577	- 50	- 60
	0	152.0	2.662	- 161	0	0

\* Initial Current Reading.
\*\* Gurrent reading at a time later when no change was noticed for a period of one minute.

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The following additional data was also observed. In Tables I through VI, results were the same whether the sulphur was in the process of heating or cooling. Frequency was not increased above 8 megacycles, since a resonance in the measuring circuit invalidated all readings from that point to the upper limit of frequency.

Observations were made similar to those of Tables VII through IX at other frequencies in the range of the equipment, and the changes in dielectric constant were of the same magnitude. No results appreciably different from those shown were obtained by varying frequency.

An attempt was made to observe a dynamic hysterisis loop on an oscilloscope. A sinusoidal voltage of 150 volts peak and of 60 cycles was used. This produced only a straight line as temperature was varied from 115 degrees Centigrade to minus 55 degrees Centigrade.

Data similar to that shown in Table VII was attempted with molten sulphur. No change in dielectric constant could be observed as voltage was increased to breakdown, or upon decreasing the voltage from any point below breakdown.

No change in dielectric constant could be observed by decreasing the voltage after reaching a point below the maxima in Tables VII to IX under those conditions. A small increase in voltage above these maxima resulted in breakdown.











MADE TO ULSUAL

![](_page_61_Figure_0.jpeg)

![](_page_62_Figure_0.jpeg)

![](_page_63_Figure_0.jpeg)

#### ANALYSIS OF RESULTS AND CONCLUSIONS

The theories which have been advanced to explain the dielectric constants of materials are not completely satisfactory. The data upon which these theories are based is apparently sketchy, or is not generally available. Precise values of dielectric constants under a given set of conditions are not common in the literature. There are contradictions in the available data, as well as in the theories. There is data which the more commonly accepted theories do not explain. Further a tendency is apparent to add more and more involved hypotheses to the commonly accepted theories in order to explain new facts that are discovered. Mathematical analysis has, in many cases, become quite complicated, and still leaves some facts unexplained. It is believed, therefore, that some very basic facts remain to be discovered, and that the solution to this problem demands more data which is not now available, and possibly a correlation of this data from new approaches.

The behavior of the dielectric constant of sulphur is not like that of most materials. As a solid it exhibits a higher dielectric constant than as a liquid. It is believed that this is due to its crystalline structure, and that some crystal structures may, in themselves, have polar properties, or exhibit something similar to polar properties. If this is the case, some change in crystal structure or in its method of influence must take place as temperature is lowered to minus 46 degrees Centigrade. The dielectric constant at this point is very nearly that of the liquid. Whatever the contribution of crystalline structure, it has ceased to have an appreciable effect at this point. Below this point, the dielectric constant increases with decreasing temperature as is expected from normal polar theories. An investigation of crystal structure as a correlation with this data, and other data that may be later obtained on this project, should yield some useful information.

The variation of dielectric constant with frequency indicates the beginning of a region of anomalous dispersion around eight megacycles. This effect can be explained by the ending of the interfacial polarization component, which, in turn, indicates the presence of impurities. As has been previously stated, the equipment did not permit investigation over the range of this region of anomalous dispersion.

The phenomena observed at values just below breakdown potential is suggestive of the beginning of the complete breakdown of crystalline structure which occurs with the arc. A distortion taking place at this point would result in the lessening of the polar, or similar, contribution of the crystal lattice. It is to be noted that the result could only be obtained at values very close to breakdown. As breakdown is approached, the material approaches the limit of its storage capability. A decrease in voltage then results in a sharp decrease in dielectric constant, followed by a gradual rise to the initial value. As soon as the condenser is caused to return energy to the circuit, after reaching this peak point, the dielectric constant of the sulphur decreased sharply, possibly indicating inability to return to its original structure. All of the charge stored at a given voltage is not returned on decreasing to this voltage. That this is a strain on the material was indicated in the low temperature test. The voltage was increased to a value near breakdown, and then the first decrease in voltage, on many of the tests, resulted in a current which increased steadily until the sulphur finally broke down. when breakdown did not occur on this decrease in voltage, the sulphur was able to be restored to its original dielectric constant after further voltage decreases. The sharp decrease in dielectric constant may then be related to the anomalies in leakage current, the reversal of charge transfer, or both. Studies of this phenomenon in other materials, and of crystalline structure, should aid in

explaining more fully the mechanism of this process. In computing the total stored charge, a small residual charge at the return zero voltage is evident. In each case it is noted to be larger in the direction in which the voltage was first increased. The changes which occur near breakdown result in the storing of a small amount of charge which is not given up with the removal of voltage. This may be analogous to the permanent deformation caused by straining a material beyond its elastic limit. The relative values of these residual charges check fairlyclosely, but may be questioned because their small magnitude is in the region of error. The changes in dielectric constant however, prove the existence of the small residual charges, whatever their value, and the recurrence of nearly the same values on numerous tests may be indicative of some degree of accuracy. This process was noted to occur only near breakdown potentials, and, therefore, is thought to be different from dielectric hysterisis, although a similar loop may be plotted. A more plausible explanation seems to be the exceeding of the elastic limit of the crystal structure.

The unusual variations of leakage current at low temperature, confirmed in several trials, are also believed to be closely related to crystal structure. It was noted that the leakage current was consistently higher in one direction than in the other. Non-bilateral resistors are commonly explained in terms of the work functions of crystals, an involved subject the study of which has not been attempted here. It can also be seen that the properties observed here are more unusual than those of a simple non-bilateral resistor. The differences in current with increasing and decreasing voltage and the time delay of current in reaching a steady value, indicate additional unusual conditions. The unexpected current behavior at low temperature was not noted to produce any effect on dielectric constant changes. Whether it is an important factor in the problem under consideration, therefore, is not known. Investiga-

tions of other materials should show its importance and give additional clues as to its effects.

It is believed that much additional and valuable information may be gained by further research on this project. More data is badly needed on the subject. It is thought that an investigation of dielectric constant at high direct voltage with a material of high dielectric loss would be particularly important in formulating a better explanation of the material discussed herein. It would also be interesting to attempt to obtain this type of data on a material for which a dynamic hysterisis loop may be observed. Research by a crystallographer with a view to aiding this project should give much needed information.

The obtaining or building of some additional equipment would be of great value in this study. A special immersion condenser, with smooth evenly-spaced plates, and which was so constructed as to be sure that nearly all the capacity and loss was occurring in the dielectric material under test, would result in better measurements. Materials used in its construction would have to remain rigid over wide ranges of temperature, and not contaminate or react chemically with the dielectric material. The measuring instrument most often mentioned in the literature on dielectric constant research is the heterodyne beat instrument. A coil is placed in parallel with the test condenser and the circuit is made to oscillate at its natural resonant frequency. A similar coil and calibrated variable condenser are also made to oscillate and tuned to the frequency of the unknown, by mixing the signals, and tuning for a zero difference frequency. It is believed that such an instrument would facilitate further study.

#### BIBLIOGRAPHY

- Addenbrooke, G. L. "The Hon-Metallic Elements Connexions Between their Dielectric and other Physical Properties." The London, Edinburgh, and <u>Dublin Philosophical Magazine and Journal of Science</u>, XLVII (June, 1924), 945-965.
- Anderson, A. and Keane, T. "A Study of Dielectric Hysterisis." The London, Edinburgh, and Dublin Philosophical Magazine and Journal of Science, XXIV (February, 1912), 437-443
- Crane, A. H. The Dielectric Constant of Petroleum Wax. Stillwater, Oklahoma: A Thesis Submitted to Oklahoma Agricultural and Mechanical College, 1948.
- Creighton, H. J. and Koehler, N. A. Principles and Applications of Electrochemistry. New York: John Wiley and Sons, Inc., 1943
- Debye, Peter. Polar Molecules. New York: Dover Publications, 1945
- Del Mar, W. A., Davidson, W. F., and Marvin, R. H. "Electric Strength of Solid and Liquid Dielectrics." Transactions of the American Institute of Electrical Engineers, XLVI (June, 1927), 1049-1061
- Gerlach, W., Translated by Fuch, F. J. Matter, Electricity, and Energy. New York: D. Van Nostrad and Company, 1928
- Hackh, I. J. D. and Grant, J. Chemical Dictionary. Philadelphia: P. Blakiston's Son and Company, Inc., 1937
- Hamburger, Jr., F. "Polar Holecules, Their Contribution to Energy Loss in Dielectrics." <u>Physical Review</u>, XXXV (May, 1930), 1119-1124
- Hausner, H. H. "Research on Barium Titanate in the U.S.S.R." <u>Ceramic Age</u>, L (September, 1947), 162-164, 190
- Moover, P. L. "The Mechanism of Breakdown of Dielectrics." Transactions of The American Institute of Electrical Engineers, XIN (June, 1926), 983-997
- Joffe, A. F. The Physics of Crystals. New York: McGraw-Hill Book Company, Inc., 1928
- Jones, H. L. An Electron Theory of the Hall Effect and Susceptibility. Stillwater, Oklahoma: Oklahoma Engineering Experiment Station of the Oklahoma Agricultural and Mechanical College, 1948
- McCabe, R. P. "A Survey Of Particle Size Methods." <u>Ceramic Age</u>, L (September, 1947), 155-157, 190
- Vol. X. New York: Longmans, Green and Company, 1930
- Lüner, D. F. Insulation of Electrical Apparatus. New York: McGraw-Hill Book Company, Inc., 1941

- Moon, P. H. and Norcross, A. S. "Three Regions of Dielectric Breakdown." <u>Transactions of the American Institute of Electrical Engineers</u>, XLIX (April, 1930), 755-765
- Marphy, E. J. and Morgan, S. O. "The Dielectric Properties of Insulating Materials." Bell System Technical Journal, XVI (October, 1937), 493-512
- Operating Instructions for Type 821-A Impedance Measuring Circuit. General Radio Company, Cambridge, Massachusetts.
- Pauling, L. "The Rotational Motion of Molecules in Crystals." Physical Review, XXXVI (August, 1930), 430-443
- Peek, F. W. Dielectric Phenomena In High Voltage Engineering. New York: McGraw-Hill Book Company, Inc., 1915
- Hemo, S. and Whinery, J. R. Fields and Waves in Hodern Radio. New York: John Wiley and Sons, Inc., 1949
- Rice, F. O. and Teller, E. The Structure of Matter. New York: John Wiley and Sons, Inc., 1949
- Sawyer, C. B. and Tower, C. H. "Rochelle Salt as a Dielectric." Physical Review, XLV (May, 1934), 269-273
- Schwaiger, A. and Soronson, R. W. Theory of Dielectrics. New York: John Wiley and Sons, Inc., 1932
- Sidgwick, N. V. Some Physical Properties of the Covalent Link in Chemistry. Ithaca, New York: Cornell University Press, 1933
- Skilling, H. H. Fundamentals of Electric Waves. New York: John Wiley and Sons, Inc., 1948
- Smyth, C. P. and Hitchcock, C. S. "Dipole Rotation in Grystalline Solids." Journal of the American Chemical Society, LIV (December, 1932), 4631-4647
- Valasek, J. "Infrared Absorption by Rochelle Salt Crystals." Physical Review, XLV (May, 1934), 654-655
- Wagner, K. W. "The Physical Nature of the Electrical Breakdown of Solid Dielectrics." Transactions of the American Institute of Electrical Engineers, XLI (April, 1922), 208-299

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- Mnitehead, J. B. Lectures on Dielectric Theory and Insultion. New York: McGraw-Hill Book Company, Inc., 1927
- Wul, B. M. and Goldman, I. M., Translated by Hausner, H. H. "Dielectric Hysterisis in Barium Titanite." <u>Comptes Rendus</u> (<u>Doklady</u>) <u>de l'Academie</u> <u>des Sciences de l'U.R.S.S., LI (January, 1946), 21-23</u>

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