

CONDUCTION ENHANCEMENT BY PARTICLES  
IN DIELECTRIC LIQUIDS

By

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## CHAPTER I

### INTRODUCTION

The objective of the present study is to examine the effects which a dielectrophoretic deposition of particles upon an electrode would have on the steady state current through a dielectric liquid. The study was restricted to the case of insulating solid particles in a steady field. By the term dielectric liquid, poorly conducting liquids such as organic liquids are implied. Dielectrophoresis is (1) the motion of neutral matter induced by polarization effects in a non-uniform electric field. The dielectrophoretic force upon neutral matter in an inhomogeneous field is to be distinguished from the electrophoretic force due to any electric field, uniform or non-uniform, on a charged particle.

In the past, interest in electrical conduction processes in liquid dielectrics has been confined mainly to work with liquids alone; particle effects have usually been neglected. Although there is as yet no generally accepted theory of conduction in liquid dielectrics, several facts have been experimentally established. It is generally agreed that at low fields ( $< 10 \text{ KVcm}^{-1}$ ) residual conduction currents in highly purified organic liquids are due to ionization of remaining impurities (2-4) and ionization of the solvent molecules by external radiation (4). Recently, it has been suggested that hydrocarbons have an electronic conduction mobility associated with the molecular  $\pi$ -electrons (5). Another point of general agreement is that at low fields an

electron does not spend all of its time in the liquid as a free particle but can be "trapped" for an appreciable portion of the time (2-4), perhaps by electric dipole interaction with the surrounding neutral molecules (2, 6).

Experimental evidence also indicates that current through dielectric liquids depends not only upon the liquid but upon the electrode materials used (4, 7-9), upon the electrode gap  $\bar{d}$  at least for non-concentric spherical electrodes (7)  $\bar{J}$ , and, in the DC case, upon the length of time the voltage has been applied (3, 10). When a DC voltage is applied to a dielectric medium, the current decays monotonically from an initial maximum to a steady state value which is sometimes orders of magnitude lower than the initial value. No completely satisfactory explanation of this phenomenon has been proposed. Zaky, Tropper, and House (7) have concluded from their careful studies on liquid hexane that these system parameters do not seem to influence the general shapes of the current-field curves to any extent but do have a marked influence upon the magnitude of the current for a particular field strength. They found that at low fields saturation regions were observed, and at high fields the currents increased sharply and non-linearly.

The total contribution to electrical conduction of particles of foreign material in dielectric liquids is still not fully explored. Most of the experiments on mixtures of liquids and particles have had as their purposes the study of mechanical motions of the particles in electric fields (1, 4, 10, 11-13, 14).

Kok (14) has given an extensive analysis of conduction phenomena involving conductive colloidal particles and has concluded that the dominant effect is to lower the breakdown strength of the liquid. He



attributed the breakdown effect itself to the presence of particles. His model was one in which polarized conductive particles are drawn into regions where the field is non-uniform. These large dipoles then line up head to tail to form what Kok referred to as a "pearl string." If the string bridges the gap from electrode to electrode, it will short circuit the cell either by conduction through the particles or by conduction along the surface of the particles. Kok presented descriptions of various experiments in which this phenomenon was observed and analyzed. Two points worth noting are that the largest particle size mentioned in Kok's work is 2 microns in diameter and that the majority of particle materials he mentioned could be considered excellent conductors with respect to the liquids used, e.g., gold vs water, iron vs transformer oil, and water vs transformer oil.

Kok also described the motion of these particles due to electrophoretic action, i.e., the motion in an electric field due to a net accumulated charge on the particle. The particle would then be electrophoretically attracted to an electrode where it would be discharged. This would be seen as a current pulse and is actually observed (7). Kok's conclusion was, however, that particles do not contribute significantly to the conduction in a dielectric liquid, but are a cause of dielectric breakdown.

As noted before, Pohl and coworkers (1, 11-13) have conducted separation experiments upon mixtures of solid coarse particles and dielectric liquids which were subjected to an electric field of cylindrical geometry. The solid materials were chosen to have a dielectric constant greater than the liquid. It was found that the solid material was drawn to the center electrode when a voltage was applied to the system. An

equation of motion was presented for a spherical particle immersed in a dielectric liquid which includes dielectrophoretic, viscous, and diffusional effects (15). It was further shown that at least in the D.C. case the dielectrophoretic force on a dielectric sphere in a cylindrical geometry is given by the expression

$$\vec{F} = -4\pi a^3 \epsilon_0 K_1' \cdot \frac{(K_2' - K_1') V_1^2}{(K_2' + 2K_1') r^3 (\ln \frac{r_1}{r_2})^2} \cdot \vec{r}_0 \quad (1-1)$$

where  $a$  is the radius of the spherical particle,  $\epsilon_0$  is the permittivity of free space,  $r_2$  is the radius of the outer electrode,  $r_1$  is the radius of the inner electrode,  $V_1$  is the applied voltage,  $K_1'$  the relative dielectric constant of the liquid medium,  $K_2'$  is the relative dielectric constant of the particles, and  $\vec{r}_0$  is the unit radius vector.

It has been suggested that suspended particles might contribute to conduction by means of Brownian rotation of the particles (11, 12). The mechanism pictured is that the ionic current deposits charge upon the surface of the spherical particle which then carries the charge across its diameter by means of its rotation.

Another indication that a possible effect may occur in dielectric liquids is the current increase found in glow discharge tubes when a thin layer of insulating material is placed upon the cathode (16). The explanation proposed for this effect is that a positive space charge is formed either inside the insulating layer or on its surface with the positive charge being supplied by positive ions from the gas inside the tube. This creates a large electric field which pulls the electrons through the insulating layer. The electrons are then emitted from the insulator with large kinetic energies. It would not seem that the same

mechanism would occur in poorly conducting liquids since (a) the dominant charge carriers at low and intermediate fields would appear to be ions and (b) there would be less of a voltage drop to carry the ions across the remaining viscous liquid.

To sum up the present state of understanding of the effect of particles on the conduction in dielectric liquids, the following remarks might be made. It is recognized that conductive particles present, can increase conduction (14). Properly placed insulating materials can induce increased conduction in gaseous discharge. The effect of insulating particles in liquids is a relatively unstudied area (4, 11, 12). The conduction mechanisms in dielectric liquids are fairly well understood in a qualitative sense. The dynamics of particles suspended in a dielectric liquid have also been studied (1, 11-13, 15).

The present study consisted of the examination of the effect of insulating particles upon conduction in very pure dielectric liquids. It was found convenient to use dielectrophoresis to place the solid particles against the smaller electrode in a cylindrical geometry and to compare the current flow before and after the deposition of particles. Much care was given to attainment of high purity. The particular test cell design selected permits highly effective purification of the liquid and solid particles by closed circuit recycling. Two purification methods, exhaustive sorption and continuous electrolytic ion removal, were used concurrently. It is thought that the care used in attaining high purity has been instrumental in achieving the good reproducibility observed.

## CHAPTER II

### APPARATUS, MATERIALS, AND PROCEDURES

The Glass Cell: Figure 1 is a diagram of the glass cell which held the liquid, the purification system, and the test electrode assembly. The three vertical chambers were constructed from Pyrex glass tubing which had an outside diameter of 35 mm. The longest vertical dimension was 21.6 cm. and the horizontal dimension was 14.0 cm. The glass fritted filter in the central chamber had a maximum pore size of 40-60 microns while the filter in the side chamber had a maximum pore size of 170-200 microns. The tops of the chambers were made from Pyrex 29/42 ground glass joints. The distance from the tops of the filters to the bottom of the sidearms a and b was 14.0 cm.

The two side chambers of the cell were used for the purification elements of the system while the central chamber held the test electrode system. Chamber A held an absorption bed of 6-12 mesh non-indicating silica gel which was used to chemically clean the liquid. Chamber C held the cleaning electrodes which will be described below.

The design of the cell was such as to allow the liquid to be continuously circulated as indicated by the arrows in Figure 1. A teflon covered magnetic stirring bar whose dimensions were 0.64 cm x 1.92 cm was inserted below the filter in the central chamber. The cell was then set upon a magnetic stirrer. When operating, the stirring bar drew the liquid down from the central chamber, forced it through the side arms c

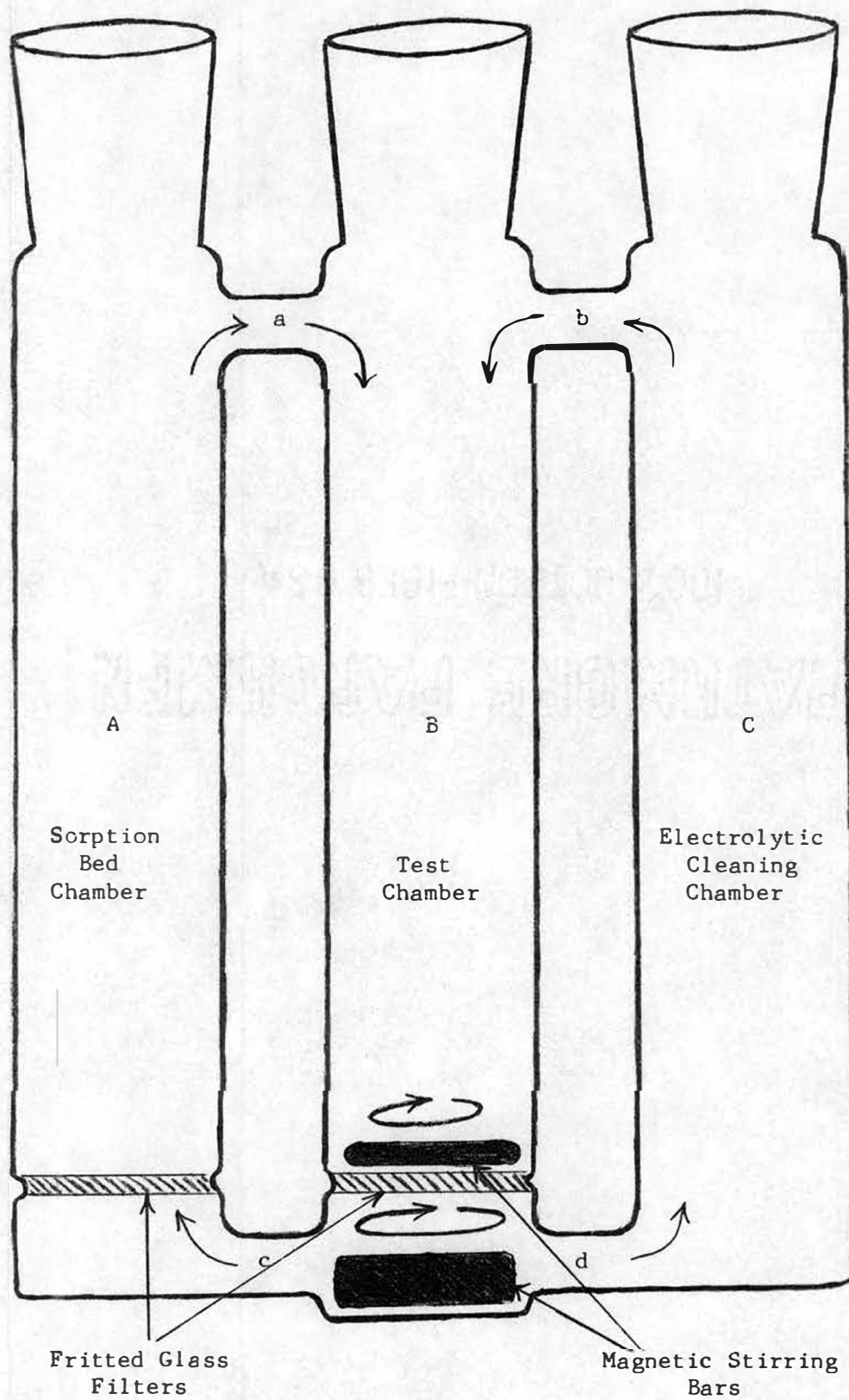


Figure 1. Test Cell

and d and up through the two side chambers. The liquid then re-entered the central chamber through side arms a and b. A continuous circulation of the liquid through the cleaning system was effected for as long as desired. A glass covered stirring bar whose dimensions were 0.41 cm x 2.44 cm was inserted in the central chamber of the cell above the filter. This stirring bar was used for stirring the powder sample up into the test region. The bar was activated when the magnetic stirrer was turned on at low speeds but could not follow the magnetic stirrer when high speeds were used and so did not interfere with the purification cycle.

The Electrolytic Cleaning System: Figure 2 is a diagram of the electrolytic cleaning system. The chosen geometry was again that of concentric cylinders. The two outer cylinders were of stainless steel while the central wire was 22 gauge platinum. The outside diameter of the outer electrode was 2.48 cm and its inside diameter was 2.27 cm. The outside diameter of the inner cylinder was 1.37 cm and the inside diameter was 1.13 cm. The bottom end of the platinum wire was spot welded to the outer cylinder. The center wire was clamped as shown in Figure 2 at the bottom of the assembly by means of a stainless steel screw clamp which was partially sunk into the bottom teflon spacer. It was originally intended that the spring wound in the top section of the platinum wire would keep the wire taut but the teflon plug at the top of the assembly would not hold the wire. Instead the wire was pulled down until the spring was partially jammed in the narrowed shaft running through the teflon support and then the wire was clamped at the bottom. Another hole was drilled through the teflon support as shown and a length of nichrome wire was run through it and spot welded to the inner stainless steel cylinder. The two cylinders were sunk 0.32 cm into the

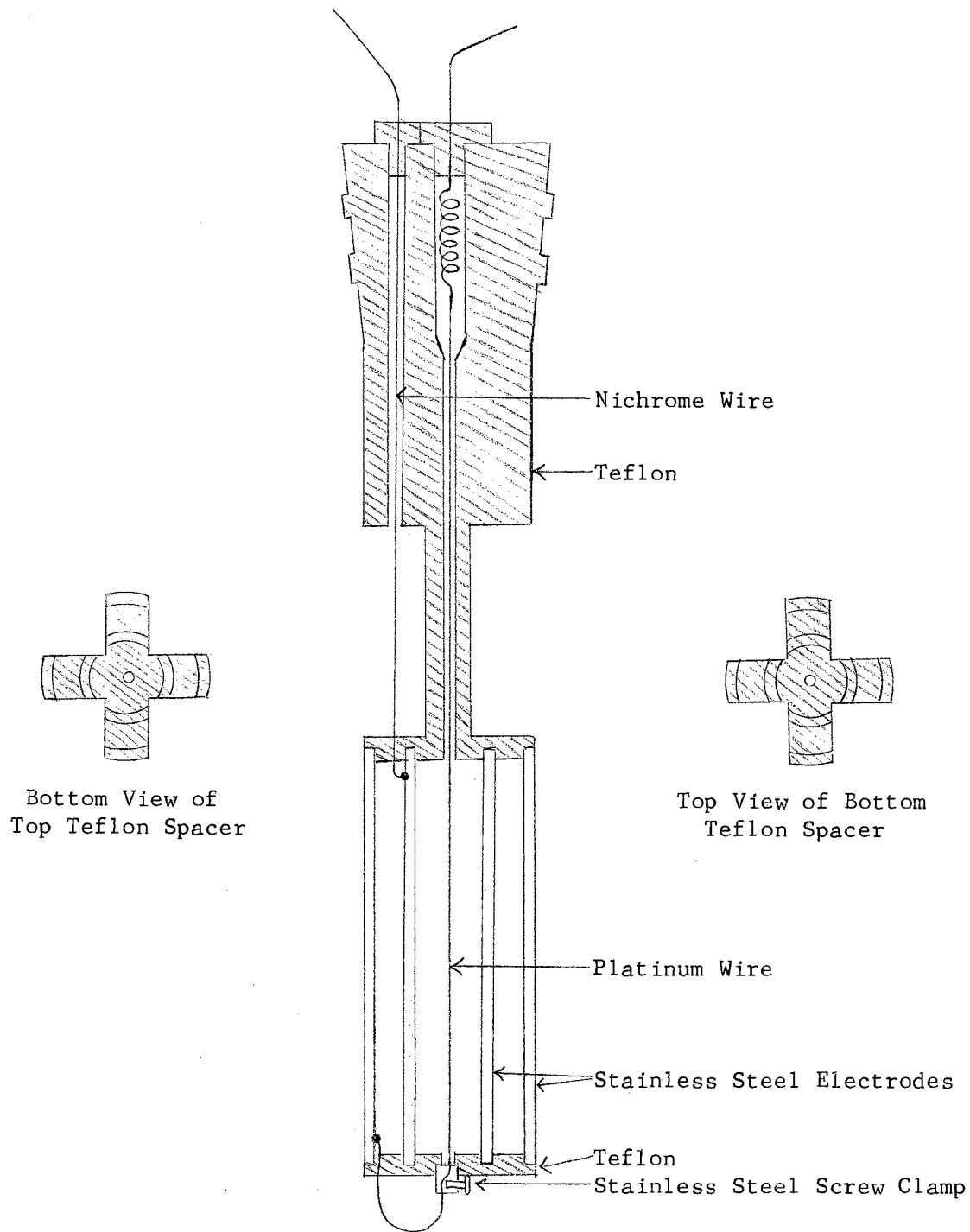


Figure 2. Axial Cross Section of Electrode Assembly for Electrolytic Cleaning and Views of Teflon Spacers

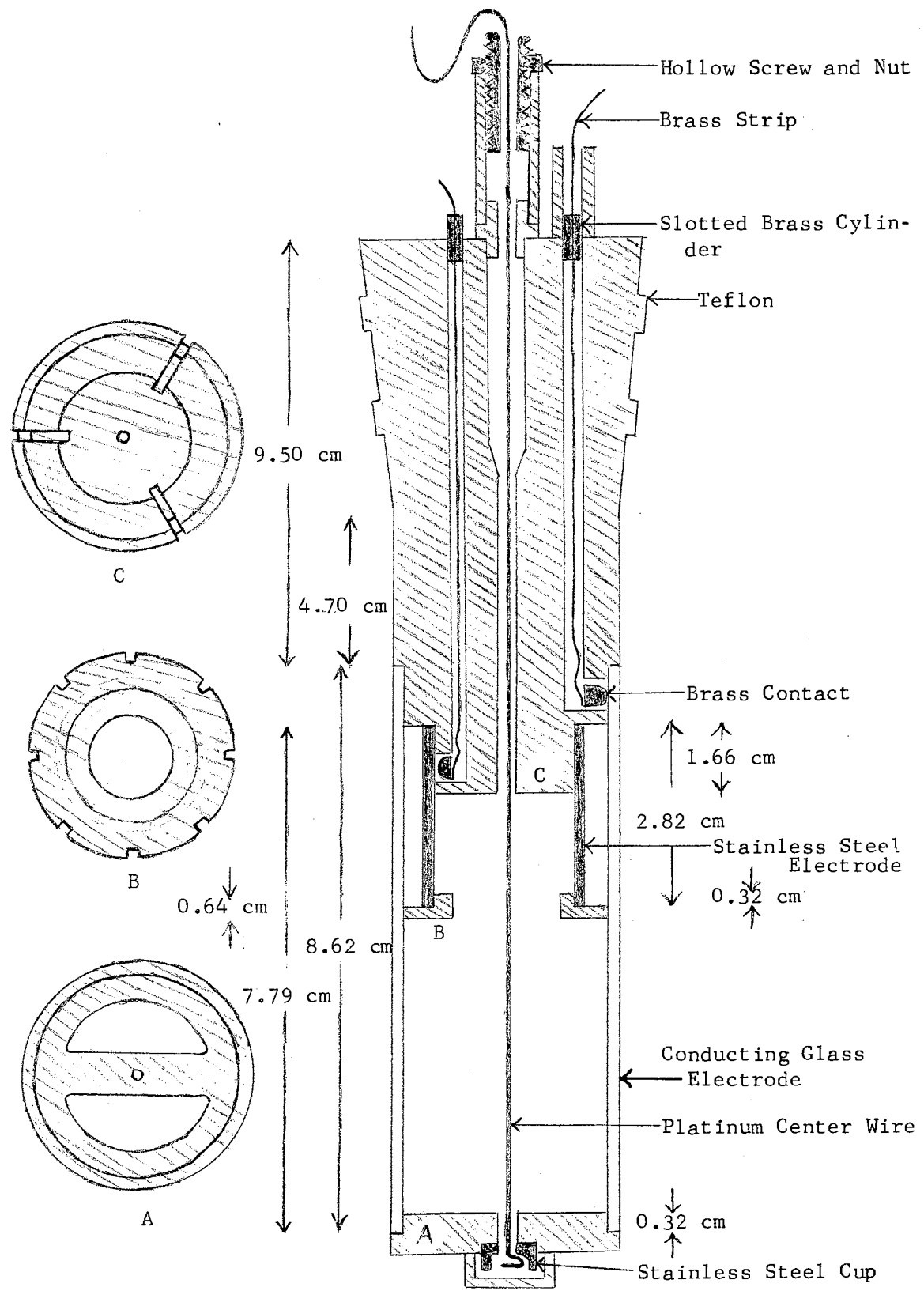
teflon spacers to insure rigidity of the assembly. The two wires were then connected to a Burgess XX30 battery whose measured voltage was 48 v.

The function of this system was to sweep ions out of the liquid as it was circulated through the chamber past the electrodes. This type of cleaning system has been described by Strier (17) who reported a considerable improvement in loss tangent characteristics of liquids when a small D.C. bias was applied to the system.

The Test Electrode Assembly: Figure 3 is a diagram of the test electrode assembly showing the details of construction and the vertical dimensions. The outer conductor was a piece of Pyrex glass tubing with a measured inside diameter of 2.16 cm. A conducting film was produced upon the inside surface of the tubing with the method described by Gomer (18). A brief description is as follows. A length of tubing of the desired diameter was first sealed at one end and then thoroughly cleaned. A second longer thin tube was then inserted deep into the first. Glass wool was inserted between the two tubes at the open end of the larger tube. The closed end of the large tube was then placed into an oven. The small tube had to be long enough to project 15 cm to 30 cm out of the oven. A few crystals of  $\text{SnCl}_2 \cdot \text{H}_2\text{O}$  were placed in the end of the small tube which was then supplied with a rubber blowing tube. The large tube was then heated to a temperature 300-400°C. The  $\text{SnCl}_2 \cdot \text{H}_2\text{O}$  crystals were then melted with a bunsen flame and the fumes blown into the large tube. A conducting coating formed upon the inside surface of the large tube. This process was repeated until the desired conductivity was reached. Figure 4 is an illustration of the method. The conducting glass produced and used in this study had a maximum resistance from side to side of  $2 \times 10^6$  ohms which was judged to be sufficiently conducting.



Figure 3. Axial Cross Section of Test Electrode  
Assembly and Views of Teflon Spacers  
Showing Slots and Holes for Circulation  
of the Liquid



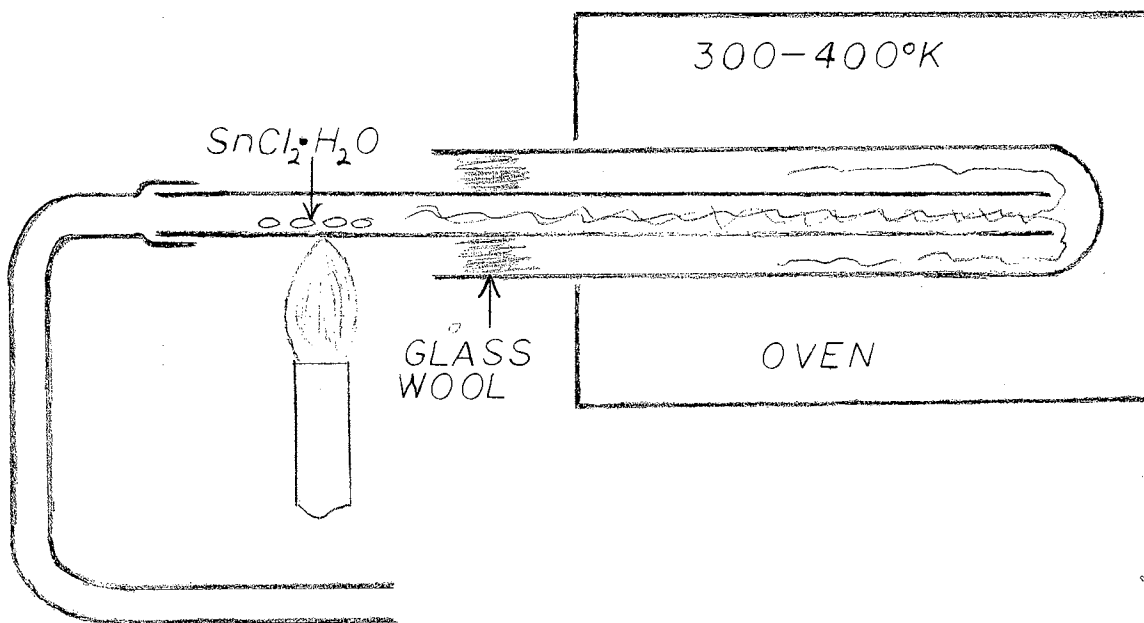


Figure 4. Illustration of Making Conducting Glass

The coated glass was extremely transparent and produced no problems in observing the behavior of the particles in the cell.

The stainless steel electrode was never used in the experiments. It was intended to provide an almost flat field between it and the glass electrode in which to study the behavior of the particles. Its inside diameter was 1.82 cm and its outside diameter was 1.98 cm.

A 0.640 cm diameter platinum wire was used as the central electrode as it was in the electrolytic cleaning assembly. The bottom end of the wire was hooked into a stainless steel cup which was countersunk into the bottom of spacer A. This in turn was covered by a teflon cap. Tension was kept upon the wire by means of the screw and nut arrangement shown in Figure 3. The wire was bent over the top of the screw shaft and was tightened by turning the screw.

Appropriate slots and holes were machined into spacers A and B and the teflon support C so that the liquid in the test region could also be

circulated through the cleaning system.

When assembled, the test cell was mounted in an aluminum chassis box whose dimensions were 20.3 cm x 43.5 cm x 7.6 cm. A sponge pad was glued to the back of the chassis box and the cell was held against this sponge backing by rubber bands. A screen door was fitted to the box so that the cell was electrically shielded. Holes were drilled into the box to admit electrical leads. The 45 v battery used for electrolytic cleaning was mounted outside the box on the side and leads were run from this battery to the electrolytic cleanup assembly through a hole in the box.

The box was then clamped to a ring stand. A transparent plastic bag was slipped over the box so that a dry nitrogen atmosphere could be provided for the cell during all experiments. The bag was tied off at points where electric leads were run through it. The box was placed upon a magnetic stirrer and positioned so that the teflon stirring bar in the cell could spin freely without much rubbing against the glass wall. An aluminum plate was inserted between the magnetic stirrer and the chassis box enclosed by the bag to help dissipate the heat generated by the operation of the magnetic stirrer. Figure 5 is a diagram of the described assembly shown without the screen door and plastic bag.

The Test Circuit: Figure 6 is a diagram of the test circuit. The power supply was a bank of ten Burgess U320 batteries with an individual EMF rating of 510 volts. Batteries were used in order to have a highly steady voltage source. The current through the cell was detected by means of a Keithley 610B Electrometer which has a rated grid current of  $2 \times 10^{-14}$  amperes. For current measurements of  $3 \times 10^{-12}$  to  $10^{-14}$  amps, the Keithley electrometer has a rated accuracy of  $\pm 4\%$  of full scale

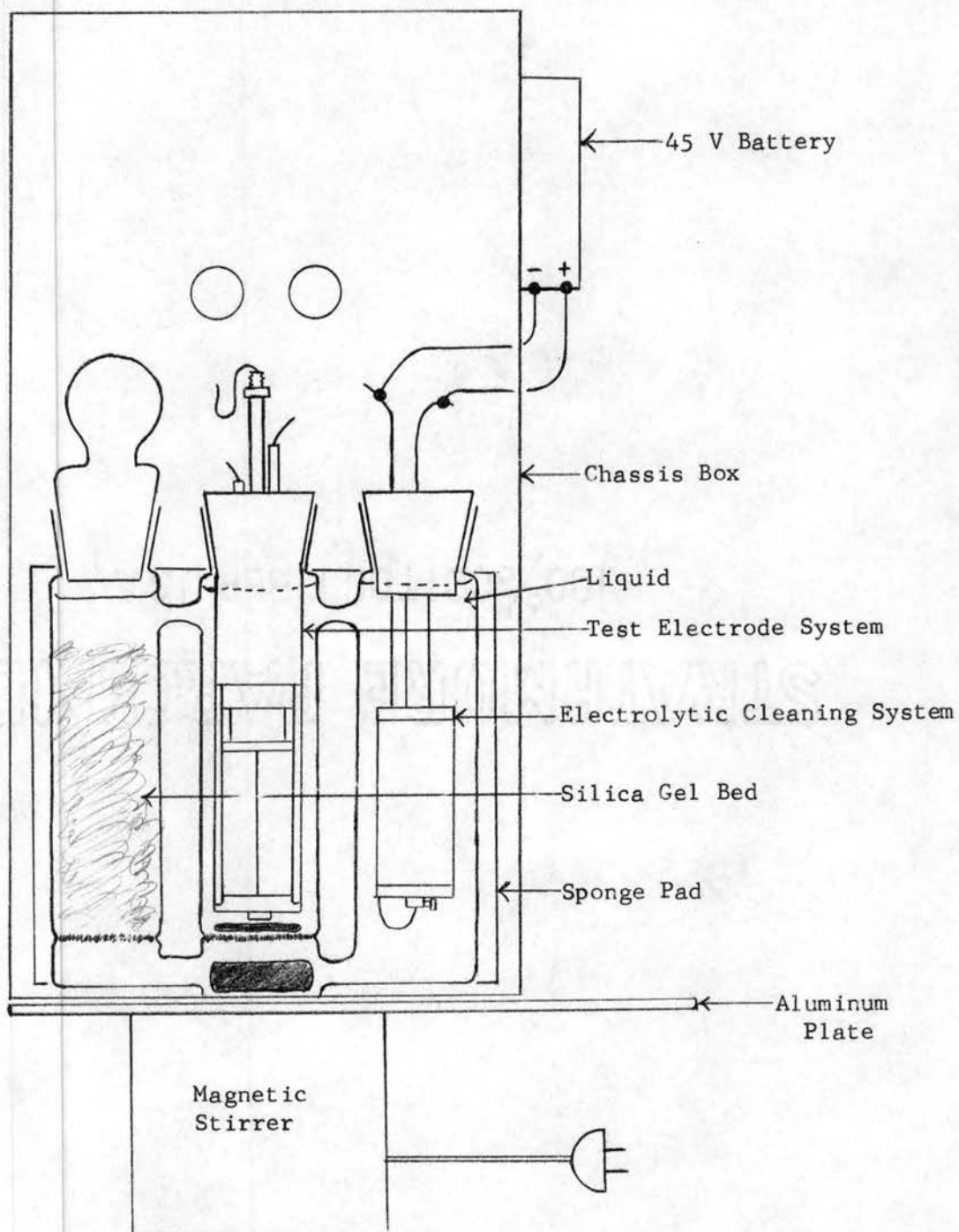


Figure 5. Illustration of Box, Cell, Plate, and Magnetic Stirrer Without Door and Plastic Bag

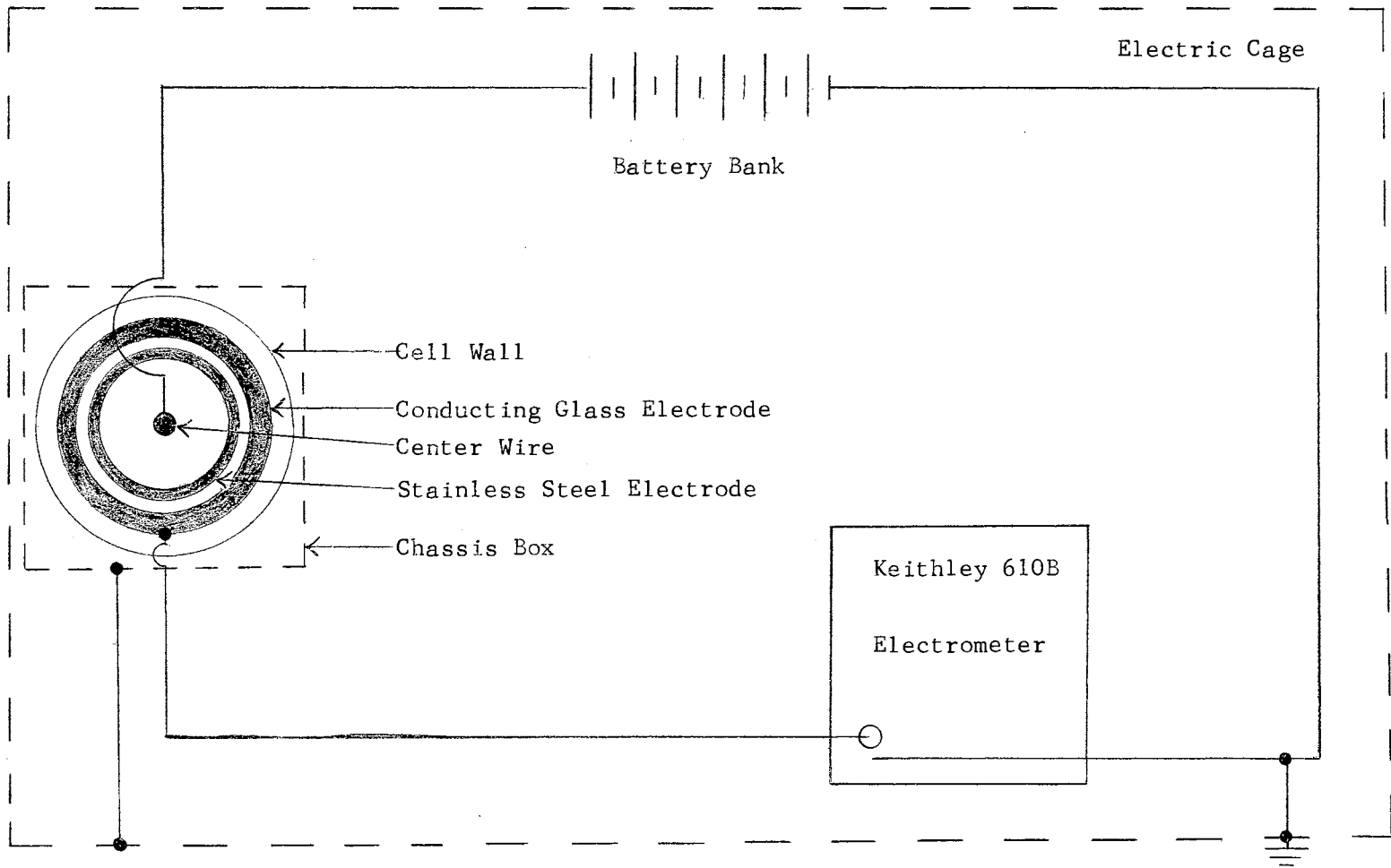


Figure 6. Test Circuit

deflection. The apparatus was placed in a cage as a safety precaution and to help shield the electrometer. The cage, low side of the electrometer, the negative side of the battery bank, the chassis box, and the case of the magnetic stirrer were all held at a common ground provided by the center screw of a 120 volt A.C. wall outlet. The voltage of each battery was measured with an Aryton Mather Static Voltmeter manufactured by the Cambridge Instrument Co. The voltmeter had a full scale deflection of 600 volts.

Materials and Procedures: The solid materials chosen for the experiments were polyvinyl chloride (PVC), sulphur, and silicon dioxide. The liquid used for the run with PVC was a mixture of carbon tetrachloride and toluene mixed in a volume ratio of about 5.5:3 so that its density was slightly less than that of the powder sample. Pure carbon tetrachloride was used for the runs with sulphur and silicon dioxide. The liquid samples were mixed from chemicals of at least reagent grade purity and were stored in a dry box when not in use. Solid silica gel was added to the stored liquids to absorb water and other polar impurities. The powder samples were stored for at least a week before use in a dessicator over anhydrous calcium sulphate.

The PVC was obtained from the Bordon Chemical Co. The sample used was purified by extracting it about 12 hours with distilled water to remove impurity salts and then by extracting with portions of reagent grade carbon tetrachloride for another 12 hours to remove organic impurities. The powder was then dried, sieved to three lots of differing sizes, and then stored. The first lot was sieved to pass 60 mesh screen and stop on 80 mesh screen, the second lot was sieved to pass 80 mesh screen and stop on 100 mesh screen, and the third lot was sieved to pass

100 mesh screen and stop on 140 mesh screen. U.S.P. grade sulphur powder was recrystallized from reagent grade carbon disulfide. Rhombic crystals were obtained. These crystals were ground, the powder sieved to pass 80 mesh screen and stop on 100 mesh screen, and this sample then stored. The silicon dioxide (Ottawa sand) was first washed with concentrated hydrochloric acid, then with distilled water, then with acetone, dried, ground, sized to pass 80 mesh screen and stop on 100 mesh screen, and then stored.

The cell was loaded by placing the powder sample into the central chamber and filling to the desired level with the liquid. In the case of the run with PVC, wads of glass wool were inserted into the two upper side arms of the cell to prevent the powder from escaping into the two side chambers. This precaution was not necessary in the other two cases since both sulphur and silicon dioxide have much higher densities than carbon tetrachloride. After loading and mounting the cell, the magnetic stirrer was turned on so that the teflon encased magnet forced the liquid through the purification cycle as shown by the arrows in Figure 1. The liquid was forced down through the central chamber and then part of it was forced up through the sorption bed where it was chemically cleaned. The other part of the liquid was forced up through the ion collector plates where residual ions were plated out as suggested by Strier's technique in measuring loss tangent characteristics in liquids (17). The liquid was tested periodically for purity by applying a 300 v battery. The stirrer was turned off when a satisfactory high level of purity was reached. When the desired purification was achieved, the liquid was electrically "conditioned" until repeatable current-voltage readings could be obtained. The process of conditioning consisted of



putting the full voltage on the system until the current had decayed to what was considered the steady state current. The time (5 to 40 hrs) required for conditioning varied from run to run. After conditioning was achieved, a current versus voltage curve was taken while decreasing the voltage at intervals in steps. The full voltage was then reapplied to the system and the stirrer was turned on at low speeds to stir the powder sample from the bottom of the chamber up into the test region. When the sample had been pulled to the center wire, another current-voltage curve was taken, while again decreasing the applied voltage in steps as before.

## CHAPTER III

### RESULTS

Polyvinyl Chloride: In the run for 60-80 mesh size PVC, .1101 grams as weighed with a Mettler balance were used. The full voltage was applied for 36 hours and then a run for the liquid was taken.

The full voltage was then applied again for another 40 hours and another current reading taken which was slightly lower than for the first run. The magnetic stirrer was then turned on until the PVC was stirred into the test region. The stirrer was turned off and current readings taken while the voltage was decreased. The full voltage was reapplied to the system for another 11 hours and a second run taken with the particles in the test region. The current-voltage curves are as shown in Figure 7.

When the PVC was stirred into the test region, the powder was first collected in a sheath around the central wire. Much of the powder was then strongly repelled to the glass electrode leaving part of the central wire bare. The PVC that remained at the wire was concentrated in two clumps, one at the top of the wire and a much smaller clump at the bottom. It is probable that the PVC was first attracted to center wire and then became charged and was repelled. The maximum sheath diameter was about 4 times the wire diameter.

In another run with 0.1099 gm of 80-100 mesh size PVC, the voltage was applied in increasing steps until the full voltage was applied to

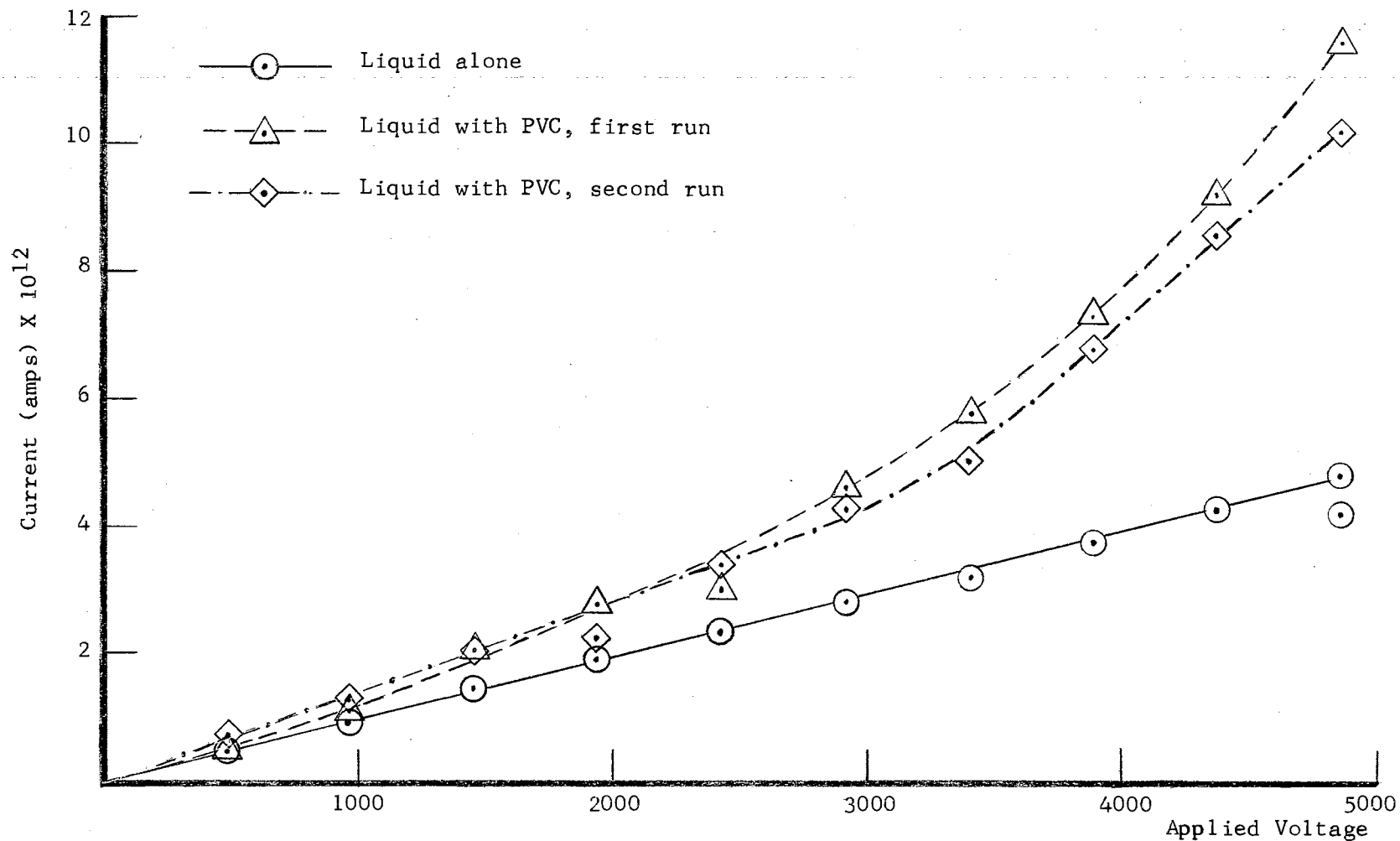


Figure 7. Current vs. Voltage Curves for Liquid Alone and for Liquid With 0.1101 gms of 60-80 Mesh PVC

the system. The coating of powder on the wire was rather uniform in this instance. The voltage was then varied up and down until the steady readings were consistent within the uncertainty of the Keithley electrometer. The voltage was then decreased and current readings recorded. The reading for the full voltage was then rechecked and was found to be slightly higher. The full voltage was left on for about 12 hrs and then the PVC was stirred into the test region with only one battery in the circuit. The full voltage was then applied to the system and after several hours readings were again taken as the voltage was decreased. After this run was completed the full voltage was again applied to the system and the current reading checked again after 1.5 hours. The batteries were then taken out of the system, the PVC shaken out of the test region and the voltage reapplied to the system. After 12 hours, the current reading was again taken. The data is shown in Figure 8.

When the PVC was stirred up into the test region, an estimated 90% of the powder collected at the central electrode and stayed there. The rest remaining in the test region was attracted to the glass electrode. When the sheath was viewed under a 10X travelling telescope, the sheath had a maximum diameter of about 4 times that of the central electrode and appeared as sketched in Figure 9. There seemed to be no preferential distribution with respect to height of the powder in the tube. Individual particles could not be distinguished but the streamers of PVC could be.

The run for 100-140 mesh size PVC was inferior in two respects; the liquid could not be purified as much as for the other two runs with PVC and the currents did not behave quite as in the two previous runs. When the full voltage was first applied to the system, the current would relax

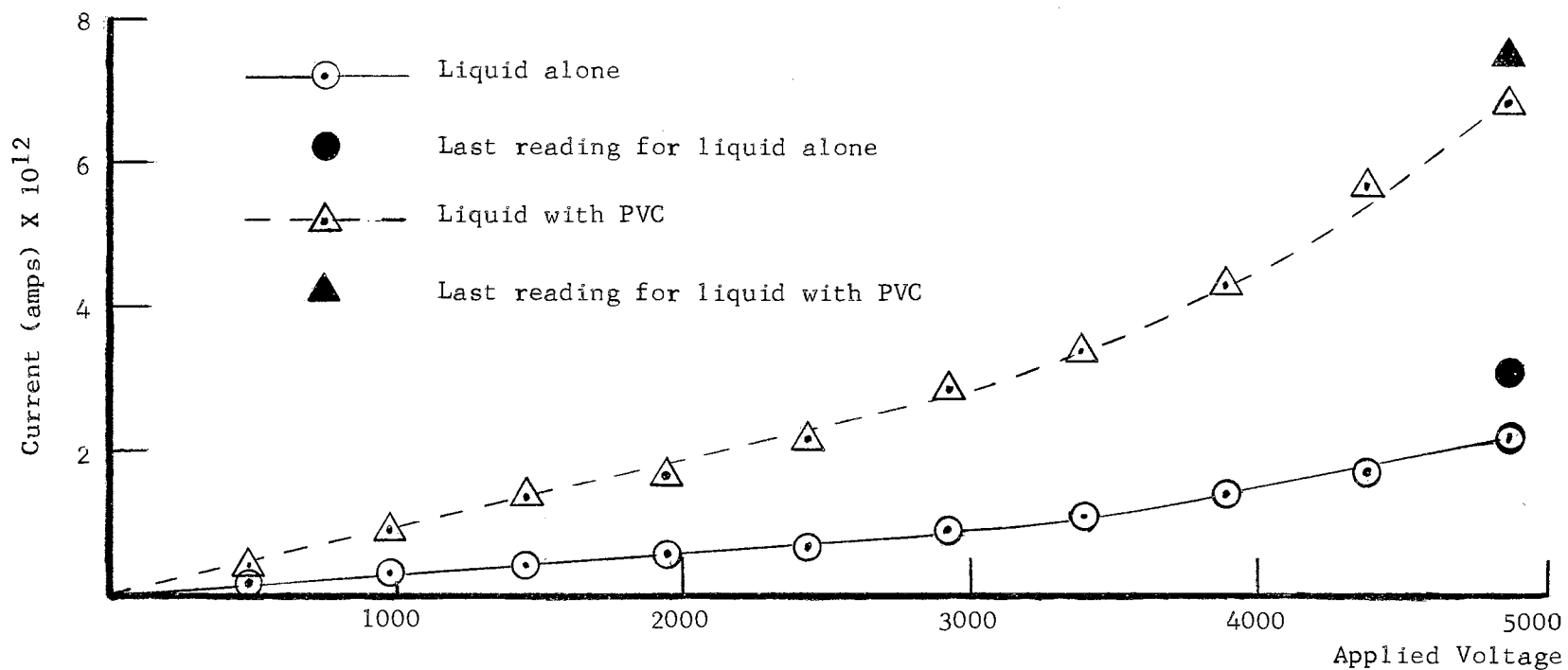


Figure 8. Current vs. Voltage Curves for Liquid Alone and for Liquid With 0.1099 gm of 80-100 Mesh PVC

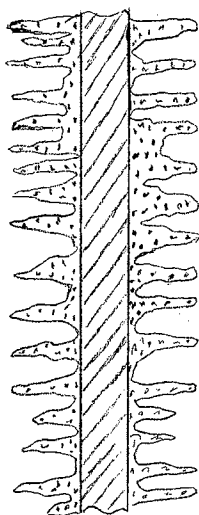


Figure 9. Sketch of Section of Center Electrode  
Showing Streamers of PVC

downward and appear to reach a steady state. However, when the run was made for the liquid alone, the current readings taken while the voltage was first decreased was smaller than the readings taken when the voltage was increased. There seemed to be a steady hysteresis upward. What was finally done was to decrease the voltage and then take readings as the voltage was increased. This was used as the data for the liquid alone. The PVC was then stirred up into the test region and readings then were taken as the voltage was decreased. This data has been graphed in Figure 10.

The lower purity in this experiment was apparently due to the PVC being of small enough particle size to partially plug the glass filter in the central chamber of the flask. This would decrease or even stop the circulation of the liquid.

The sheath formed by the PVC again appeared to be formed of streamers as noted in the prior run. This time the sheath was thicker in the center than at the top or bottom of the center wire. The center

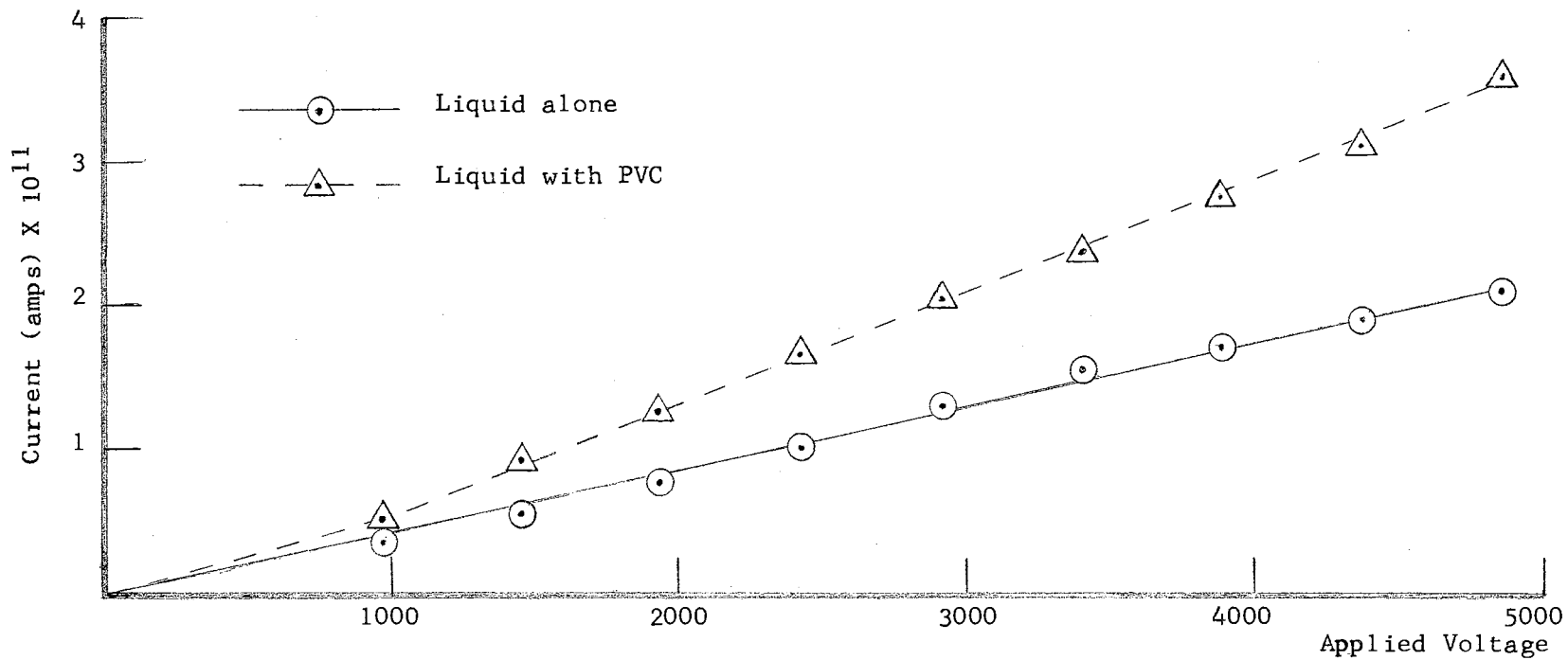


Figure 10. Current vs. Voltage Curves for Liquid Alone and for Liquid With 0.1226 gm. of 100-140 Mesh PVC

diameter was about 5.2 times as large as the center wire and the diameters at the top and bottom were about 4.5 times as large as the center wire.

The resistivity of the PVC was determined on a cast film. A sample of the purified PVC was dissolved in reagent grade cyclohexanone. The cyclohexanone was then allowed to evaporate so that a thin film of the PVC was formed. The .0241 cm thick film was air dried for 4 weeks prior to the resistance measurement. The volume resistivity of the PVC was then measured using a Keithley Model 6105 Resistivity Adaptor, a Keithley 610B electrometer, and a 300 v battery. The volume resistivity was found to be  $1.30 \times 10^{14}$  ohm-cm.

Sulphur: Reagent grade carbon tetrachloride was chosen as the liquid for this experiment. Unfortunately sulphur is slightly soluble in carbon tetrachloride. It was thought that if the liquid were first completely saturated with sulphur, then the test sample would be preserved. However, there evidently was enough of a temperature variation throughout the system to precipitate out some of the sulphur. In the cooler test region, this precipitated sulphur was pulled to the center electrode where it remained. It was decided to test the qualitative effect of sulphur particles even though quantitative results could not be taken for this particular experiment.

The original amount of sulphur powder put in the test cell was .2342 gm of 80-100 mesh size. The carbon tetrachloride had been previously saturated with sulphur by putting an amount of sulphur in a flask with the carbon tetrachloride and stirring the mixture overnight with a magnetic stirrer.



After the purification cycle, the full voltage was applied to the system for 4 days with intermittent runs to determine when a repeatable reading could be taken. As noted before, a very slight amount of sulphur precipitated out to the central electrode during this period. The repeatability was determined by how well the current for the full voltage could be repeated after the run was taken by decreasing the voltage. The second reading was slightly higher as shown in Figure 11. The sulphur in the bottom of the cell was then stirred into the test region while the full voltage was applied to the system and was pulled to the central electrode. The sheath formed by the sulphur was roughly conical in shape mainly due to the fact that the sulphur was much denser than the carbon tetrachloride. After about 4 days of full voltage application, a current-voltage run was again taken with the results as shown in Figure 11.

The cell was then discharged and the sulphur shaken out of the test region. The full voltage was then applied to the system for about 15 days and a run taken again for the liquid with very little sulphur on the center wire. After this run the sulphur was again stirred into the test region where it collected at the center wire. After this collection, another current-voltage curve was taken. The data taken is as shown in Figure 12.

For Silicon Dioxide: For this experiment, 0.1769 gm of 80-100 mesh size silicon dioxide and carbon tetrachloride were used. After the purification cycle, the liquid was conditioned for about four days, then a repeatable current-voltage run was taken. The silicon dioxide particles were then stirred up into the test region and were initially pulled to the center wire. The particles gathered at the bottom of the center wire, evidently because the density of silicon dioxide is much higher

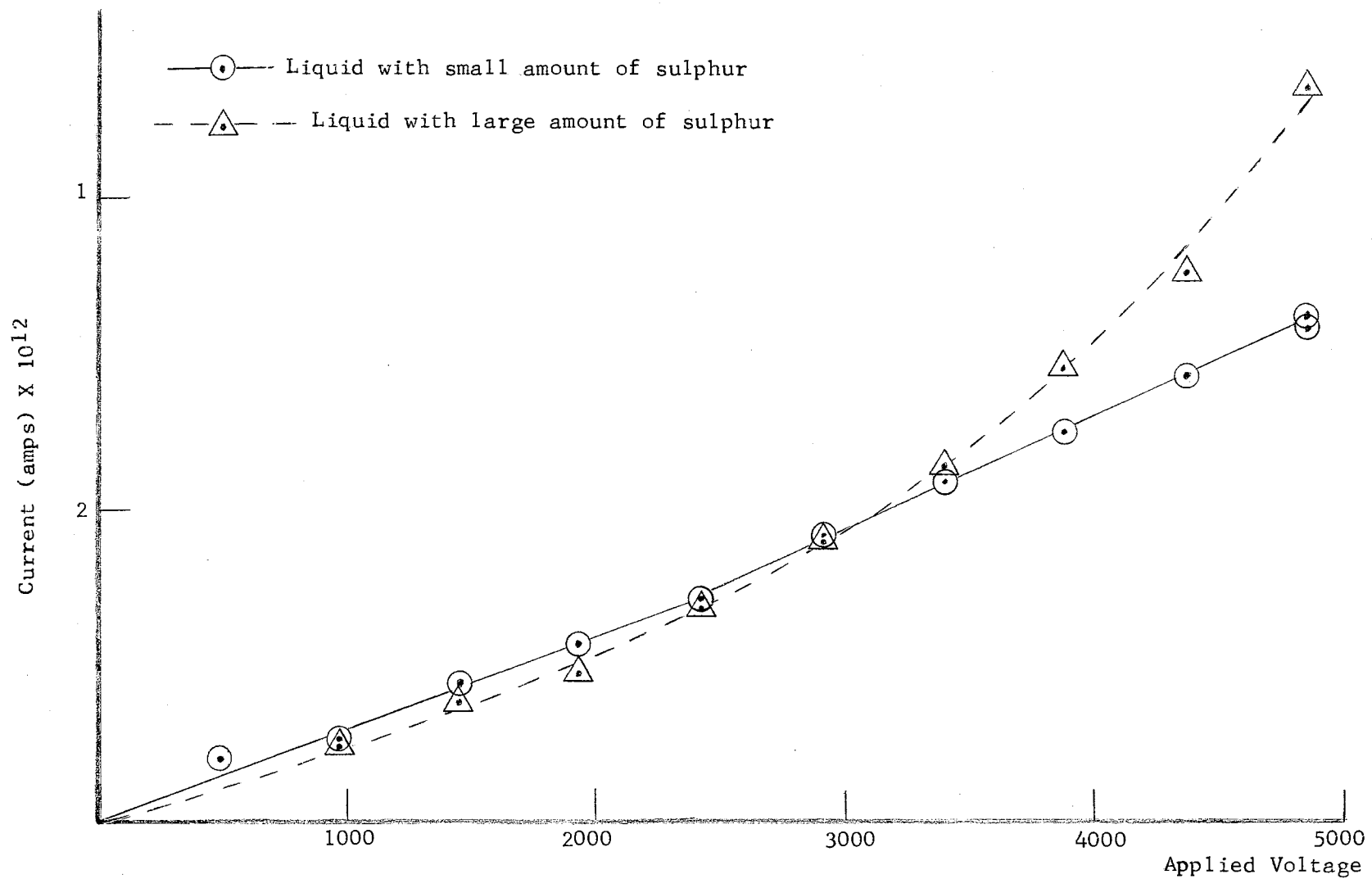


Figure 11. Current vs. Voltage Curves for Liquid and Sulphur, First Run

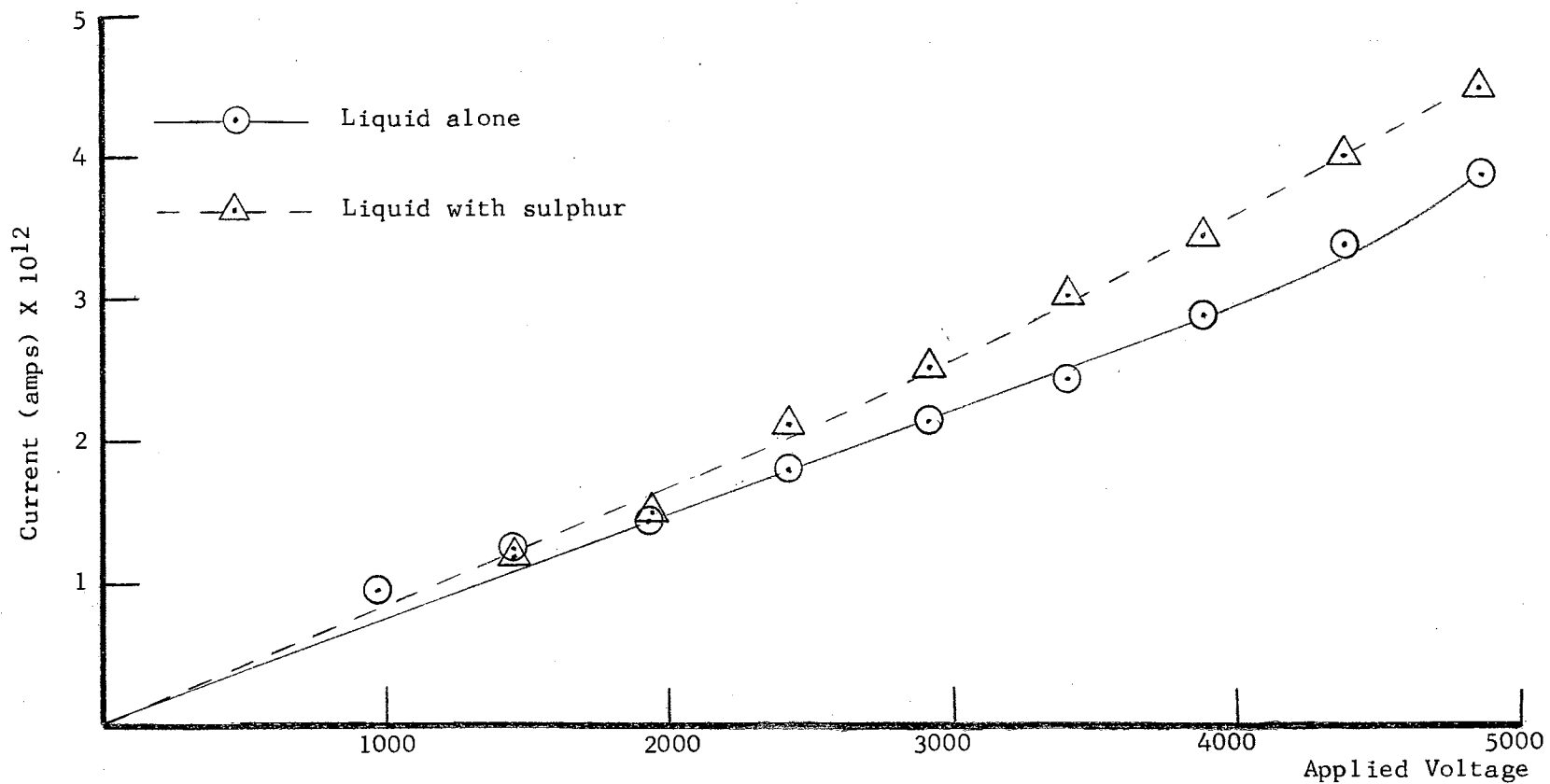


Figure 12. Current vs. Voltage Curves for Liquid and Sulphur,  
Second Run

than that of carbon tetrachloride. It appeared that only a small fraction of the powder was stirred up into the test region. Almost as soon as they were gathered, the particles began to be repelled from the center wire, evidently because of electrostatic charging. The resultant pulsing made steady state readings difficult to make. The current-voltage readings were again taken as the voltage was decreased in steps. The data taken is as shown in Figure 13.

At the end of the run the center wire was observed with a 10X travelling telescope. There were a very few single particles still clinging to the center wire. The magnetic stirrer was used again to throw the particles up into the test region. The qualitative results were the same with respect to the initial gathering of the particles at the central electrode and the subsequent repulsion of the particles to the outer glass electrode.

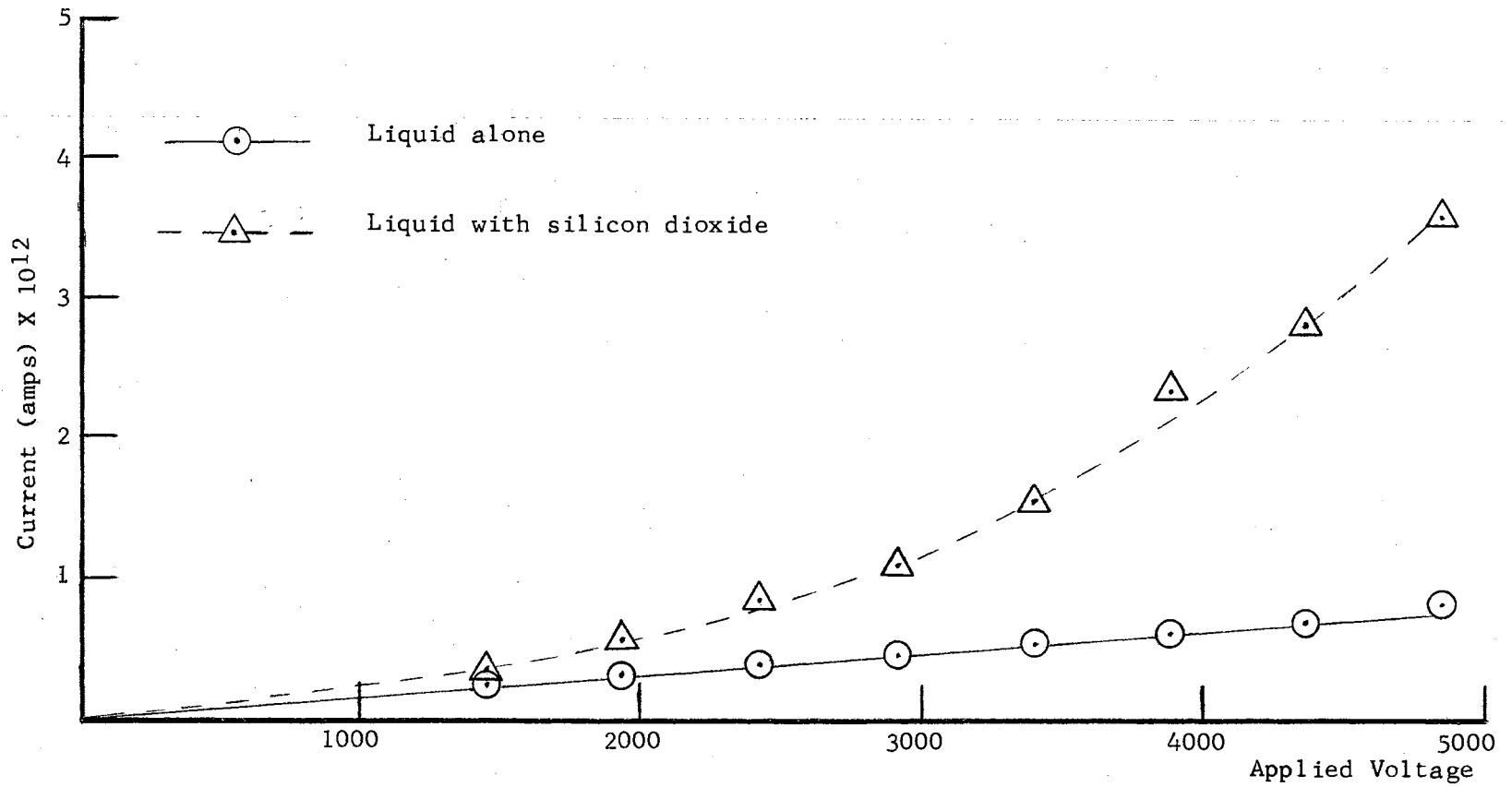


Figure 13. Current vs. Voltage Curves for Liquid Alone and Liquid With 80-100 Mesh Silicon Dioxide

## CHAPTER IV

### DISCUSSION

The most interesting feature of the experimental results is the current increase due to the presence of the particles. That the presence of insulating particles about the electrode should increase the current is, at first glance, surprising. These increases cannot be accounted for by calculating the change in ohmic resistance of the system due to the introduction of the particles into the region around the center wire. The ohmic resistance of the system with no particles is given by equation (A-8) of Appendix A as

$$R_0 = \frac{\rho_0 \ln\left(\frac{r_2}{r_1}\right)}{2\pi L} \quad (4-1)$$

where  $\rho_0$  is the resistivity of the liquid,  $L$  is the electrode length (length of center wire),  $r_2$  is the radius of the outer electrode, and  $r_1$  is the radius of the center wire. If a sheath with an outer radius of  $nr_1$  of material whose resistivity is  $\rho_1$  is placed around the center wire, the total resistance of the system is

$$R_1 = \frac{1}{2\pi L} \left[ \rho_0 \ln\left(\frac{r_2}{nr_1}\right) + \rho_1 \ln\left(\frac{nr_1}{r_1}\right) \right] \quad (4-2)$$

since the sheath is a resistance in series with the resistance of the liquid in the remaining part of the electrode system. The relative change in the resistance of the whole system  $R_1/R_0$  by the introduction of the sheath is then

$$\frac{R_1}{R_0} = \frac{\ln\left(\frac{r_2}{r_1}\right) + \frac{\rho_s}{\rho_0} \ln n}{\ln\left(\frac{r_2}{r_1}\right)} \quad (4-3)$$

Using this last equation, decreasing the resistance by a factor of 2 would correspond to expanding the diameter of the center wire by a factor of about 5.9. This clearly would not account for the increases obtained at full voltage for the runs with PVC since this calculation indicates that the sheath of PVC is more effective than a similar thickness of conductor. Similar remarks may be made with respect to the other runs with PVC and those with sulphur and silicon dioxide. Because of these considerations the current increases cannot be accounted for by ohmic calculations.

If the current is assumed to be space charge limited, then two cases present themselves for examination, (a) when the sheath surface is smooth and (b) when the surface is rough as is the case in this study. When the current is space charge limited, the conductivity is a function of the electric field. The current-voltage curves obtained experimentally for the liquids with no particles in the test region appear to be sectionally ohmic or concave upwards indicating that the conductivities of the liquids used were constant or an increasing function of the electric field. This implies that the highest conductivity regions of the liquids

occurred at the surface of the center wire where the electric field was greatest. Given this situation then, if a smooth sheath of insulating high dielectric solid is placed around the center electrode, the total resistance of the system should increase since the major decrease of the electric field would be found across the region occupied by the sheath, leaving the field in the liquid lower than before. This implies that the liquid in the remaining part of the system will be subjected to less of an electric field when the sheath is present and its total resistance will be at least equal to, if not greater than, its resistance when the sheath is not present.

A possibility for current increase may occur when the sheath is composed of particles. The increase is visualized as arising from the enhancement of the electric field just outside the surface of a particle due to the differing dielectric constants of the solid and liquid (14, 15). Consider the situation as shown in Figure 14 and Figure 15. The particles are stirred into the test region and are dielectrophoretically drawn to the center wire as described by equation (1-1). Since the dielectric constant of the solid is greater than that of the liquid, the electric field in the liquid is greater around the polar regions of the particles. If the liquid is very pure, i.e., only traces of impurities are present and almost all of the foreign ions have been removed, then a possible charge production enhancement effect may be present if most of the remaining carriers are ions created by field dissociation of impurity or solvent molecules in the regions of most intense electric field. An approximate expression for the relative current increase due to this mechanism is shown in Appendix C to be



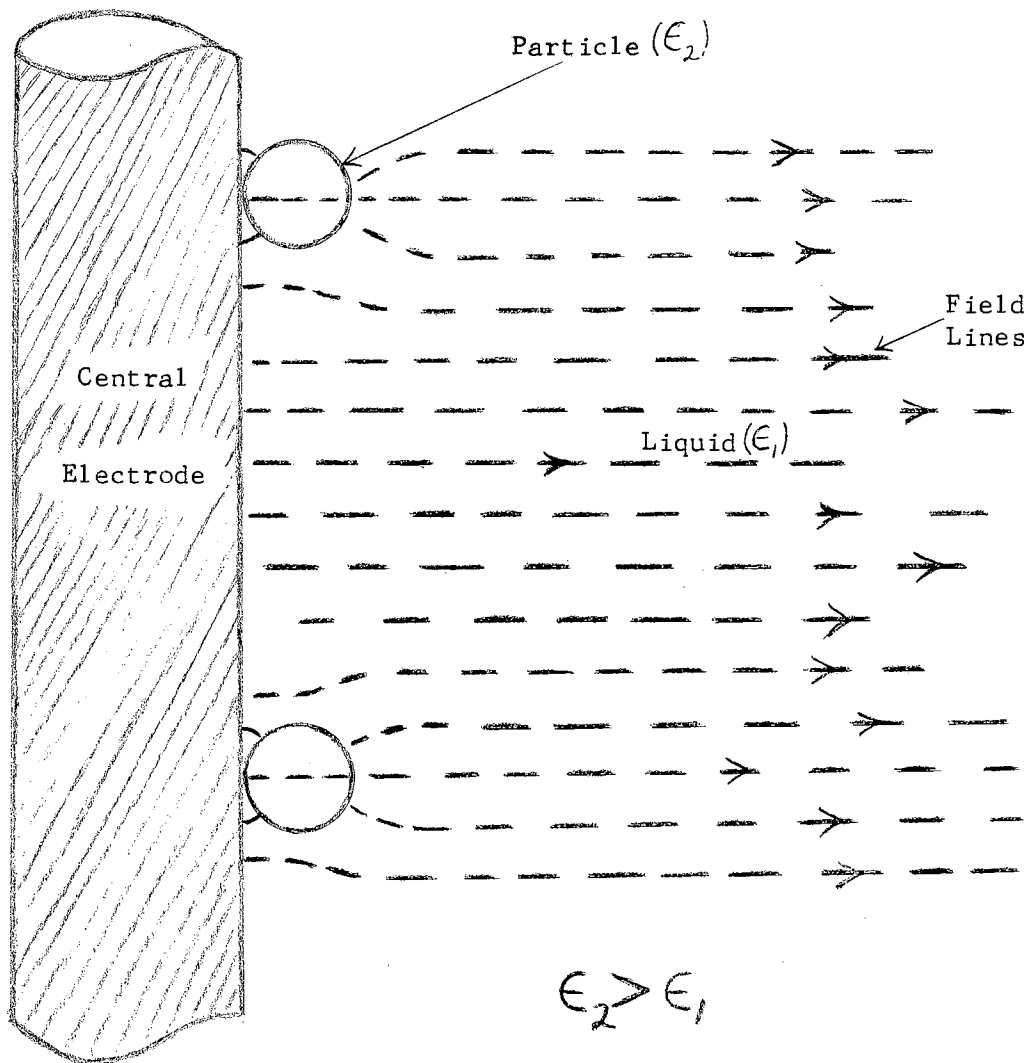


Figure 14. Illustration of Electric Field Enhancement at Surfaces of Particles at Center Electrode (Side View)

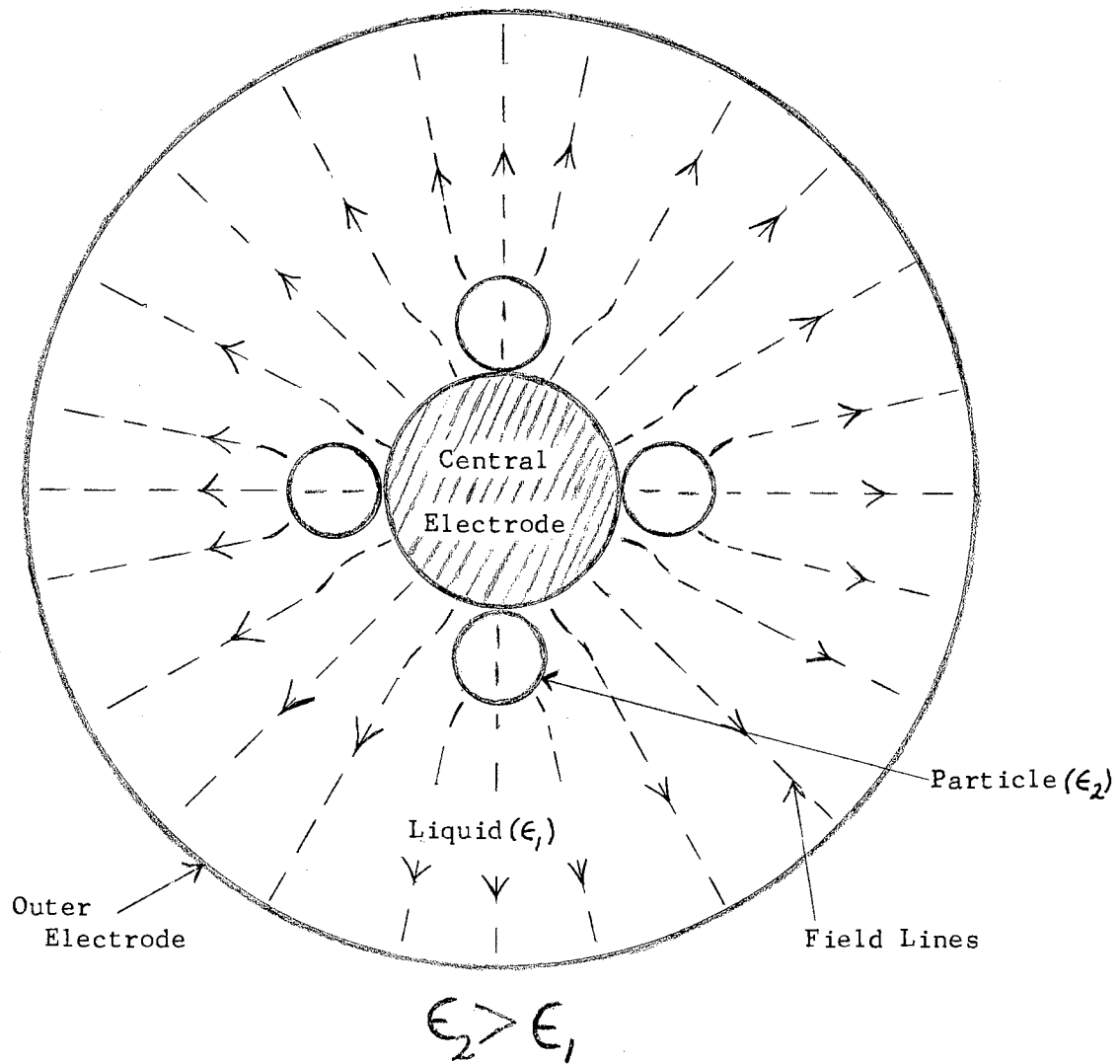


Figure 15. Illustration of Electric Field Enhancement at Surfaces of Particles at Center Electrode (Top View)

$$\frac{I_p}{I_0} = \left\{ 1 + \frac{3Mf \left[ \left( \frac{3\epsilon_2}{2\epsilon_1 + \epsilon_2} \right)^{3/2} - 1 \right]}{16\pi d L n(n-1) r_1^2} \left[ 1 + \frac{E_1^{3/2}}{\alpha (n^{3/2} - 1)} \right] \right\} \\ \times \left\{ 1 + \frac{f'(\sigma_2 - \bar{\sigma}_1) \ln n}{\bar{\sigma}_1 \ln n + \frac{\bar{\sigma}_1'}{\bar{\sigma}_1} \left[ f'(\sigma_2 - \bar{\sigma}_1) + \bar{\sigma}_1' \right] \ln \left( \frac{r_2}{nr_1} \right)} \right\} \quad (C-6)$$

where  $M$  is the mass of the powder sample,  $d$  is the density of the solid,  $\epsilon_2$  and  $\epsilon_1$  are the dielectric constants of the solid and liquid respectively,  $f$  is the fraction effective area of a single particle,  $r_1$  is the radius of the center wire,  $nr_1$  is the outer radius of the sheath,  $r_2$  is the inner radius of the outer electrode,  $L$  is the length of the electrodes,  $E_1$  is the electric field just beyond the surface of the center wire,  $\alpha$  is a voltage dependent factor derived in Appendix B,  $f'$  is the average cross-sectional fraction as particles in the sheath,  $\sigma_2$  is the conductivity of the solid,  $\bar{\sigma}_1$  is the space averaged conductivity of liquid in the sheath, and  $\bar{\sigma}_1'$  is the space averaged conductivity of the liquid in the rest of the electrode system.

One of the properties predicted is the variation of  $I_p/I_0$  as the applied voltage is varied. There are four quantities in equation (C-6) which are voltage dependent,  $\bar{\sigma}_1$ ,  $\bar{\sigma}_1'$ ,  $E_1$ , and  $\alpha \cdot E_1$ , and  $\alpha$  should monotonically increase with applied voltage. The exact

dependence of these quantities is not known since equation (B-5a) does not appear to be integrable in closed form. If, however,  $\bar{\sigma}_1$ ,  $\bar{\sigma}_1'$ , and  $E_1^{3/2}/\alpha$  change very slightly with voltage, then  $I_p/I_0$  should remain rather constant over a considerable voltage range if all other variables are kept constant. For these experiments the values of  $I_p/I_0$  are as shown in Figure 16. Three of the runs appear to display this property over most of the voltage range. The runs referred to are those for 80-100 mesh PVC, 100-140 mesh PVC, and the second run for sulphur. The relative current increase obtained in the silicon dioxide run is not shown in Figure 16 since the mass of the powder sample on the center wire steadily decreased during the run. The curve for  $I_p/I_0$  therefore cannot really be compared with the curves for the other runs.

A closer comparison between experiment and theory at the present seems unjustified. That there is a current enhancement when solid particles are deposited in the regions of highest field is beyond doubt, even when an excellent insulator such as sulphur is used. The particular mechanism involved has yet to be completely determined. It would appear that these experiments lend support to the proposed mechanism of increased charge production at the surfaces of the dielectric particles.

#### Summary and Conclusions

The particular experiments performed for this study were designed to explore the effect a deposition of solid insulating particles into regions of highest electric field would have upon the current in a dielectric liquid. The test cell was designed such that current-voltage curves could be taken for liquid alone in the system and then after the particles had been introduced into the system and dielectrophoretically

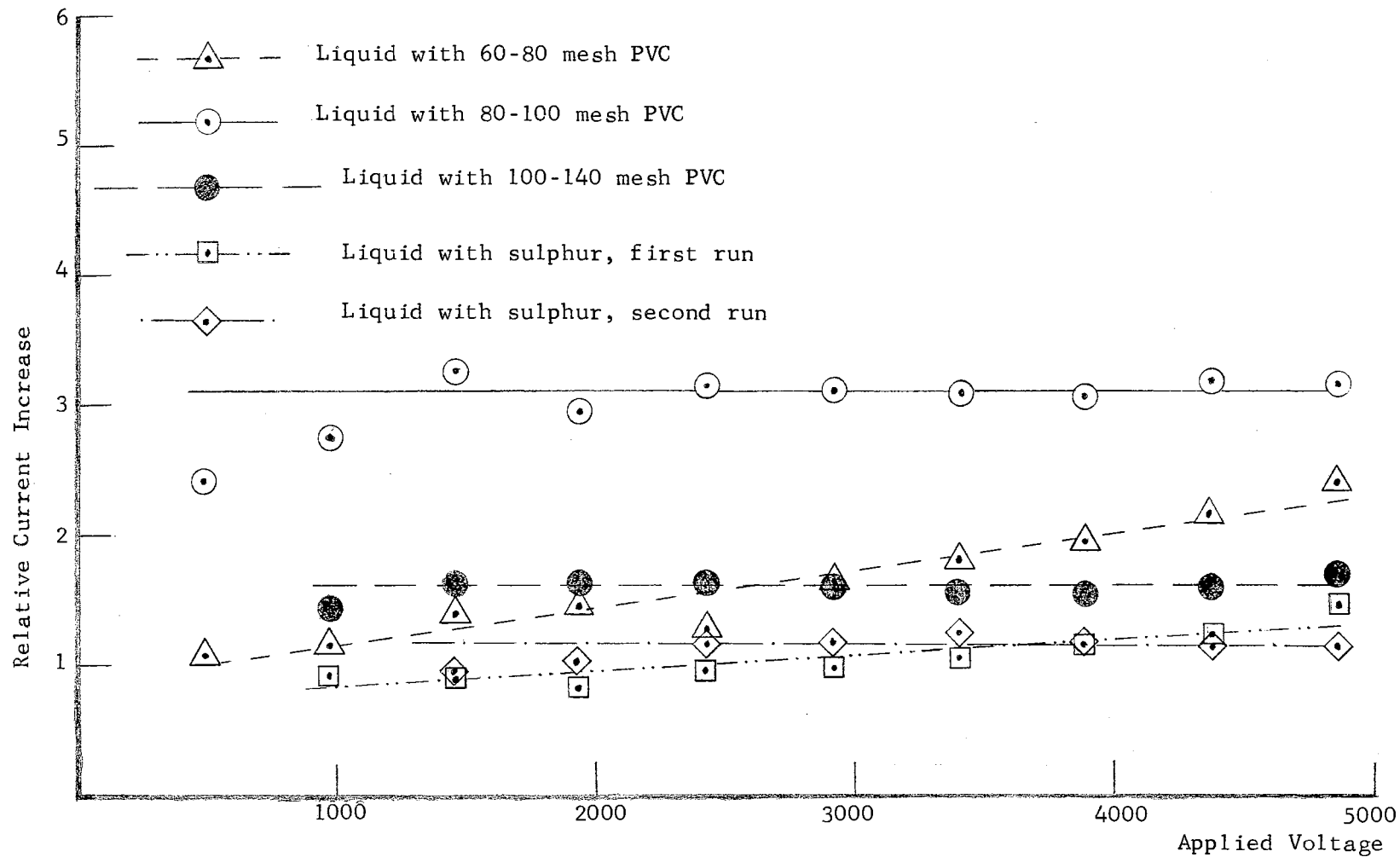


Figure 16. Relative Current Increase vs. Applied Voltage

deposited in the region of highest field. The experiments were performed upon polyvinyl chloride in a mixture of toluene and carbon tetrachloride, sulphur in carbon tetrachloride, and silicon dioxide in carbon tetrachloride.

In all cases a current increase was observed which could not be accounted for by consideration of the ohmic conductivities of the materials used. A mechanism for increased production of charge carriers is suggested as being responsible for the current increases. The theoretical model proposes that the geometry of the particles enhances the electric field in the liquid just outside of the particles because of the differing dielectric constants of the liquid and solid. Assuming that the charge carriers are produced preferentially in the regions of highest field strength by field dissociation of impurity or solvent molecules, the increase in field strength at the particle surfaces would produce a corresponding increase in charge carriers. A quantitative comparison between the experimental data and predictions based upon this model supports this mechanism.

#### Suggested Extensions

Some of the experimental extensions that would be helpful in understanding these current increases are a more thorough determination of the dependence upon the dielectric constant and conductivities of the materials used. This would involve tests of a much wider variety of materials than used in this study. A determination of the dependence upon mass also seems important in view of the results with the silicon dioxide run. One other extension which might prove fruitful would be to test with different particle geometries, e.g., ellipsoids, splinters,

etc., although this might prove to be impractical. The effect of different electrode geometries might also be tested, e.g., concentric hemispheres. AC techniques would be helpful in avoiding the sometimes severe charging effects and might sharply reduce the time needed to perform a particular experiment.

A much more detailed calculation based upon the suggested model needs to be carried out since the derivation of equation (C-6) involved numerous approximations.

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A P P E N D I X E S

## Appendix A

### OHMIC RESISTIVITY AND CONDUCTIVITY FOR A CYLINDRICAL ELECTRODE SYSTEM

An expression will be derived for the ohmic resistivity and conductivity of a medium with a concentric cylindrical electrode geometry and in terms of the applied voltage, the current through the medium and the electrode dimensions.

The system considered is composed of two concentric cylindrical electrodes, each having a length  $L$  along the common  $Z$ -axis. The outside diameter of the inner electrode is  $r_1$  and the inside diameter of the outer electrode is  $r_2$  (Figure 17). The medium between the two electrodes is here assumed to be homogeneous and isotropic.

When a voltage drop is applied across the region separating the two electrodes an electric field is established and, hence, a current. The electric field  $\vec{E}$  and current density  $\vec{j}$  are related by

$$\vec{j} = \sigma \vec{E}$$

where  $\sigma$  is the conductivity of the medium and  $\sigma = \sigma(\vec{E})$ . Since  $\rho$ , the resistivity, is the reciprocal of the conductivity,

$$\rho \vec{j} = \vec{E} \tag{A-1}$$

Now  $\vec{j}$  and  $\vec{E}$  are functions of position and, since the system has cylindrical symmetry, the magnitudes of  $\vec{j}$  and  $\vec{E}$  are functions of

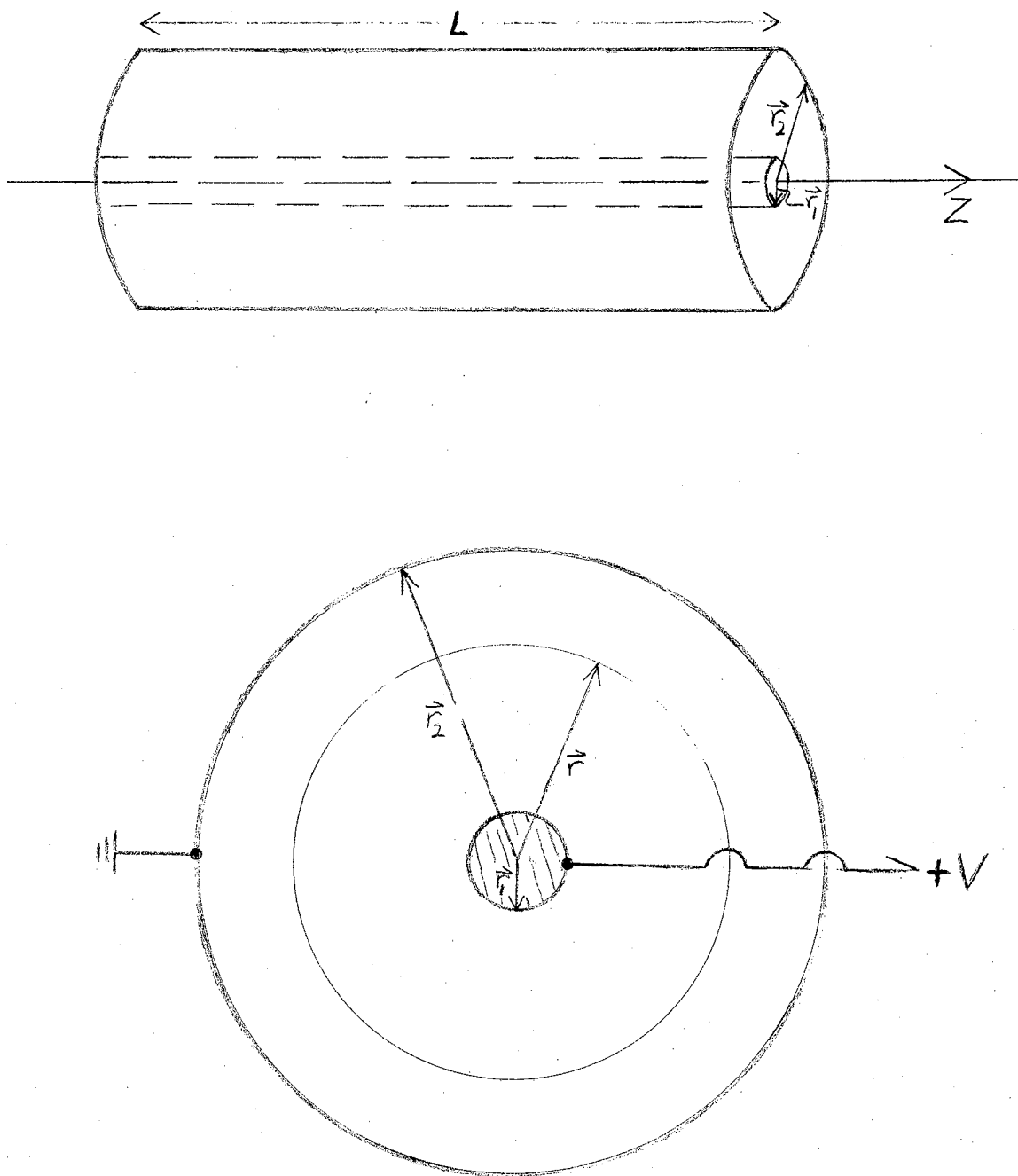


Figure 17. Illustration of Cylindrical Electrode Geometry

the radial distance  $r$  away from the common axis. If the total current is  $I$ , then the current density over the cylindrical surface with radius  $r$  is

$$\vec{j}(\vec{r}) = \frac{I \hat{r}}{2\pi rL} \quad (\text{A-2})$$

where  $2\pi rL$  is the total area of the surface and  $\hat{r}$  is the unit radial vector. Ohm's Law then takes the form

$$\frac{I \hat{r}}{2\pi rL} \rho = \vec{E}. \quad (\text{A-3})$$

The line integral from inner to outer electrode is then

$$\int_{r_1}^{r_2} \frac{I \hat{r} \rho}{2\pi rL} \cdot d\vec{r} = \int_{r_1}^{r_2} \vec{E} \cdot d\vec{r}. \quad (\text{A-4})$$

Since  $d\vec{r} = \hat{r} dr$  and if  $\rho$  is a constant, i.e., an intrinsic property of the medium, then the left-hand side of (A-4) becomes

$$\int_{r_1}^{r_2} \frac{I \hat{r} \rho}{2\pi rL} d\vec{r} = \frac{I \rho}{2\pi L} \int_{r_1}^{r_2} \frac{\hat{r} \cdot \hat{r} dr}{r} = \frac{I \rho}{2\pi L} \ln\left(\frac{r_2}{r_1}\right). \quad (\text{A-5})$$

For the right-hand side of (A-4),

$$\int_{r_1}^{r_2} \vec{E} \cdot d\vec{r} = \int_{r_1}^{r_2} (-\nabla V) \hat{r} dr = -\int_{r_1}^{r_2} dV = V(r_2) - V(r_1) = \Delta V \quad (\text{A-6})$$

where  $\Delta V$  is the total voltage drop across the distance separating the electrodes.

(A-4), (A-5), and (A-6) then give

$$\rho = \frac{2\pi L}{\ln\left(\frac{r_2}{r_1}\right)} \frac{\Delta V}{I} \quad (\text{A-7})$$

and defining the resistance of the system to be  $\Delta V/I \equiv R$ ,

$$\rho = \frac{2\pi L}{\ln\left(\frac{r_2}{r_1}\right)} R. \quad (\text{A-8})$$

Since the resistivity is the reciprocal of the conductivity,

$$\sigma = \frac{\ln\left(\frac{r_2}{r_1}\right)}{2\pi LR}. \quad (\text{A-9})$$

## Appendix B

### SPACE CHARGE LIMITED CURRENT AND CONDUCTIVITY FOR CYLINDRICAL GEOMETRY

In this appendix, expressions for the space charge limited current and conductivity of a medium with concentric cylindrical electrode geometry are derived. The principal condition assumed here is that the charge density in the medium is non-zero. It will be assumed that the carriers are of a single species with the same charge and that the mean distance  $l_0$  between collisions is the same throughout the medium. It is also assumed that a single carrier will lose all of its excess kinetic energy in a collision.

The average maximum kinetic energy that a carrier will acquire before a collision is then

$$\frac{1}{2} m v_{max}^2 = q E l_0$$

where  $m$  is the mass of the carrier with charge  $q$ .  $E$  is the electric field and has been assumed to vary insignificantly between collisions. Therefore, the average velocity,  $v_{max}/2$ , between collisions will be

$$\bar{v} = \sqrt{\frac{q E l_0}{2 m}}$$

For constant current,

$$\frac{d\xi}{dt} = 0$$

where  $\xi$  is the charge density of the carriers. This together with the continuity equation for charge implies that

$$j = \xi v \quad (\text{B-1})$$

where  $j$  is the current crossing unit area. Using as  $v$ , the average velocity between collisions,

$$j = \xi \sqrt{\frac{q E l_0}{2m}}$$

Assuming two concentric cylindrical electrodes each having length  $L$ ,

$$j(r) = \frac{I}{2\pi r L}$$

where  $I$  is the total current. Solving for  $\xi$ ,

$$\xi = \left( \frac{I}{2\pi L r} \right) \sqrt{\frac{2m}{q E l_0}} \quad (\text{B-2})$$

Poisson's equation gives

$$-\nabla^2 V = \frac{\xi}{\epsilon} + \frac{q \delta(r-r_1)}{\epsilon},$$



where  $q$  is  $-\int_{r_1}^{r_2} \epsilon 2\pi L r dr$  and is the negative charge needed to neutralize the positive space charge in the liquid. It is assumed here that this charge is located on the surface of the center wire. If the radius of the inner cylinder bounding the region of integration is chosen to be  $r_1 + \eta$ , then the term with the delta function gives no contribution and may be ignored if  $\eta \neq 0$ . Poisson's equation for the region of interest  $r_1 + \eta$  to  $r_2$ , where  $\eta \ll r_1, r_2$  then becomes simply

$$-\nabla^2 V = \frac{\epsilon}{\epsilon} \cdot \quad (\text{B-1 a})$$

This together with (A-2) implies

$$-\frac{d^2V}{dr^2} - \frac{1}{r} \frac{dV}{dr} = \frac{1}{\epsilon} \left( \frac{I}{2\pi L r} \right) \sqrt{\frac{2m}{qE l_0}}.$$

Since

$$E = -\frac{dV}{dr},$$

$$\frac{dE}{dr} + \frac{1}{r} E = \frac{1}{\epsilon} \left( \frac{I}{2\pi L r} \right) \sqrt{\frac{2m}{qE l_0}}.$$

Rearrangement yields

$$\frac{E^{1/2} dE}{\frac{I}{2\pi \epsilon L} \sqrt{\frac{2m}{q l_0}} - E^{3/2}} = \frac{dr}{r}$$

Integrating from the radius  $r_1$  of the inner electrode to a cylindrical surface between the electrodes,

$$\ln\left(\frac{I}{2\pi\epsilon L} \sqrt{\frac{2m}{q l_0}} - E^{3/2}\right) \Big|_{E=E(r_1)}^{E=E(r)} = -\frac{3}{2} \ln r \Big|_{r=r_1}^{r=r}$$

Solving for the current gives

$$I = 2\pi L \epsilon \sqrt{\frac{q l_0}{2m}} \left[ \frac{(rE)^{3/2} - (r_1 E_1)^{3/2}}{r^{3/2} - r_1^{3/2}} \right] \quad (\text{B-3})$$

If now the conductivity of the medium is defined as

$$j = \sigma E$$

then

$$\begin{aligned} \sigma &= \frac{j}{E} \\ &= \sqrt{\frac{q l_0}{2m}} \frac{\epsilon}{rE} \frac{(rE)^{3/2} - (r_1 E_1)^{3/2}}{r^{3/2} - r_1^{3/2}} \quad (\text{B-4}) \end{aligned}$$

The numerator of the factor in braces in (B-3) deserves some comment. For the case in which there is no net charge density between the electrodes, Laplace's equation may be solved to yield

$$rE = r_1 E_1.$$

$[(rE)^{3/2} - (r_1 E_1)^{3/2}]$  is then a measure of the distortion of the electric field away from its geometric value due to the presence of the unbalanced space charge density. If the field actually is the one required for ohmic behavior, i.e., the geometric field, the current given by (B-3) and the conductivity given by (B-4) would both be zero. For current as described by (B-3) to appear then

$$\frac{r_1 E_1}{rE} < 1.$$

Since the total current passing through a cylindrical surface of radius  $r$  must be constant despite the value of  $r$ , then

$$\frac{(rE)^{3/2} - (r_1 E_1)^{3/2}}{r^{3/2} - r_1^{3/2}} = \text{constant} = \alpha \quad (\text{B-5})$$

and

$$\begin{aligned} rE &= \left[ \alpha r^{3/2} - r_1^{3/2} (\alpha - E_1^{3/2}) \right]^{2/3} \\ &= \left[ \alpha r^{3/2} - \beta \right]^{2/3} \end{aligned} \quad (\text{B-5a})$$

where

$$\beta = r_1^{3/2} (\alpha - E_1^{3/2})$$

is constant.  $\alpha$  and  $\beta$ , however, are functions of the applied voltage.

The conductivity (B-4) may be now written as

$$\sigma = \sqrt{\frac{q \cdot d_0}{2m}} \frac{E \alpha}{[\alpha r^{3/2} - \beta]^{2/3}} \quad (\text{B-6})$$

From (B-5), it is seen that the space charge limited conductivity has its maximum value at the surface of the inner electrode and its minimum value at the surface of the outer electrode.

## Appendix C

### RELATIVE CURRENT INCREASE DUE TO INSULATING PARTICLES

An expression will be derived in the appendix for the relative current increase through a dielectric liquid when particles of higher dielectric content than that of the liquid are introduced into the region of the center wire of a concentric cylindrical electrode system. For simplicity it will be assumed that the particles are spherical and have the same diameter,  $a$ .

It is assumed that the particles have been attracted to the center electrode and have formed pearl strings which are evenly distributed over the surface of the center wire. It is assumed further that the same number  $k$  of particles are in each string. Let  $f$  be the average current density in the liquid that would occur over a cylindrical surface of radius  $ka + r_1$  if the spherical shape of the particles had no effect. Here,  $r_1$  is as usual the radius of the center electrode. Let  $f'$  be the current density in the liquid occurring at the tip of a string when the geometry of the particles is accounted for. Since, in general, the current density is defined to be the conductivity  $\sigma$  times the electric field  $E$ ,

$$\frac{f'}{f} = \frac{\sigma(E') \cdot E'}{\sigma(E) \cdot E}.$$

Because of the electrode geometry and the assumptions made about charge carrier production, the conductivity must be space-charge-limited such as the expression given by equation (B-6) of Appendix B. Using this expression

$$f' = \frac{\sqrt{\frac{q_0}{2m}} \frac{\epsilon}{rE'}}{\sqrt{\frac{q_0}{2m}} \frac{\epsilon}{rE}} \frac{(rE')^{3/2} - (r_1E_1)^{3/2}}{r^{3/2} - r_1^{3/2}} \frac{E'}{E} f.$$

Simplifying,

$$f' = \frac{\left(\frac{E'}{E}\right)^{3/2} - \left(\frac{r_1E_1}{rE}\right)^{3/2}}{1 - \left(\frac{r_1E_1}{rE}\right)^{3/2}} f.$$

The total current  $i'$  emanating from the string will then be  $f'$  times an effective surface area

$$i' = A_{\text{eff}} f' = 2\pi \left(\frac{a}{2}\right)^2 f f' = \frac{\pi a^2}{2} f f'$$

where  $f$  is dimensionless and has a value between 0 and 1, and represents the fraction (judged to be about  $\frac{1}{2}$ ) of the area in the most effective field.

The number of strings present will be

$$\frac{M}{d \frac{4}{3} \pi a^3} \frac{1}{k}$$

where  $M$  is the total mass of the powder sample and  $d$  is the density of the solid.

The total current  $i_p$  emanating from all the strings is then

$$i_p = \frac{3M}{4\pi d a^3} \frac{1}{k} \cdot \frac{\pi a^3}{2} f j' = \frac{3Mf}{8da k j'}$$

The total current  $I_p$  for the system will then be

$$I_p = i_p + \left[ 2\pi(ka + r_1)L - \frac{3Mf}{8dak} \right] j$$

where  $L$  is the length of the electrode system.

Then,

$$I_p = 2\pi(ka + r_1)Lj + i_p - \frac{3Mf}{8dak} j$$

The total current for the case when no account is taken of the particle geometry, i.e., where  $j$  is the current density over all the surface of the sheath, is

$$I = 2\pi(ka + r_1)Lj$$

Therefore,

$$\begin{aligned} \frac{I_p}{I} &= 1 + \frac{i_p - \frac{3Mf}{8dak} f}{2\pi(ka+r_1)Lf} \\ &= \frac{\frac{3Mf}{8dak} \left[ \frac{\left(\frac{E'}{E}\right)^{3/2} - \left(\frac{\epsilon_1 E_1}{\epsilon_1 E}\right)^{3/2}}{1 - \left(\frac{\epsilon_1 E_1}{\epsilon_1 E}\right)^{3/2}} \right] f - \frac{3Mf}{8dak} f}{2\pi(ka+r_1)Lf} + 1 \\ &= 1 + \frac{3Mf \left[ \frac{\left(\frac{E'}{E}\right)^{3/2} - 1}{1 - \left(\frac{\epsilon_1 E_1}{\epsilon_1 E}\right)^{3/2}} \right]}{16\pi dka(ka+r_1)L} \quad (C-1) \end{aligned}$$

If now  $E'/E$  is approximated by the solution of the problem of the neutral dielectric sphere in a uniform field and letting  $ka+r_1 = nr_1$ ,

$$\frac{I_p}{I} = 1 + \frac{3Mf \left[ \frac{\frac{3\epsilon_2}{2\epsilon_1 + \epsilon_2} - 1}{1 - \left(\frac{\epsilon_1 E_1}{\epsilon_1 E}\right)^{3/2}} \right]}{16\pi d(n-1)nr_1^2 L \left[ 1 - \left(\frac{\epsilon_1 E_1}{\epsilon_1 E}\right)^{3/2} \right]}$$

where  $\epsilon_1$  and  $\epsilon_2$  are the dielectric constants of the liquid and solid respectively.

Using equation (B-5) of Appendix B, it is easily shown that

$$1 - \left(\frac{\epsilon_1 E_1}{\epsilon_1 E}\right)^{3/2} = \frac{\alpha(n^{3/2} - 1)}{\alpha(n^{3/2} - 1) + E_1^{3/2}}$$



so that

$$\begin{aligned} \frac{I_p}{I} &= 1 + \frac{3Mf \left[ \left( \frac{3\epsilon_2}{2\epsilon_1 + \epsilon_2} \right)^{3/2} - 1 \right] \left[ \alpha(n^{3/2} - 1) + E_1^{3/2} \right]}{16\pi dL(n-1)n r_1^2 \alpha(n^{3/2} - 1)} \\ &= 1 + \frac{3Mf \left[ \left( \frac{3\epsilon_2}{2\epsilon_1 + \epsilon_2} \right)^{3/2} - 1 \right] \left[ 1 + \frac{E_1^{3/2}}{\alpha(n^{3/2} - 1)} \right]}{16\pi dL(n-1)n r_1^2} \end{aligned} \quad (C-2)$$

Now the simplifying approximation is made that the conductivity of the sheath may be represented by the ohmic expression  $f'\sigma_2 + (1-f')\bar{\sigma}_1$  where  $\sigma_2$  is the conductivity of the solid,  $\bar{\sigma}_1$  is the space averaged conductivity of the liquid in the sheath and  $f'$  is an average cross sectional fraction of sheath as particles and  $(1-f')$  is the average cross section fraction as liquid.

The relation between  $I$  and  $I_0$ , the current with no particles in the system, can be calculated using the relation

$$\frac{I_0}{I} = \frac{R}{R_0} \quad (C-3)$$

since total voltage applied to the whole system is the same.  $R$  and  $R_0$  are the effective resistances corresponding respectively to  $I$  and  $I_0$ . Using equations (4-1) and (4-2)

$$R = \frac{1}{2\pi L} \left( \frac{\ln \frac{nr_1}{a}}{\bar{\sigma}_1} + \frac{\ln \frac{r_2}{nr_1}}{\bar{\sigma}_1'} \right) \quad (C-4)$$

where  $\bar{\sigma}_1'$  is the spaced averaged resistivity for the region between  $r = nr_1$  and  $r = r_2$ . Both  $\bar{\sigma}_1$  and  $\bar{\sigma}_1'$  will be functions of the total applied voltage. Similarly,

$$R = \frac{1}{2\pi L} \left( \frac{\ln \frac{nr_1}{r_1}}{f'\sigma_2 + (1-f')\bar{\sigma}_1} + \frac{\ln \frac{r_2}{nr_1}}{\bar{\sigma}_1'} \right). \quad (C-5)$$

Now

$$\frac{I_p/I_0}{I/I_0} = \frac{I_p}{I_0}$$

and using (C-2), (C-4), (C-5), and simplifying,

$$\begin{aligned} \frac{I_p}{I_0} = & \left\{ 1 + \frac{3Mf \left[ \left( \frac{3\epsilon_2}{2\epsilon_1 + \epsilon_2} \right)^{3/2} - 1 \right]}{16\pi dL n(n-1)r_1^2} \left[ 1 + \frac{E_1^{3/2}}{\alpha(n^{3/2}-1)} \right] \right\} \\ & \times \left\{ 1 + \frac{f'(\sigma_2 - \bar{\sigma}_1) \ln n}{\bar{\sigma}_1 \ln n + \frac{\bar{\sigma}_1'}{\sigma_1} [f'(\sigma_2 - \bar{\sigma}_1) + \bar{\sigma}_1] \ln \frac{r_2}{nr_1}} \right\} \quad (C-6) \end{aligned}$$

VITA

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