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Cryogenic Power Transmission Technology—Cryogenic Dielectrics

Final Report
Nov. 1, 1972 — Sep. 30, 1976

July 1977

MASTER

Work Performed under Contract W7405-ENG-26

Oak Ridge National Laboratory

Energy Research and
Development Administration

Division of Electric Energy Systems

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**Final Report for the Period
Nov. 1, 1972 — Sep. 30, 1976**

**Oak Ridge National Laboratory
Oak Ridge, Tennessee**

Date Published — July 1977

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Fusion Energy Division

CRYOGENIC POWER TRANSMISSION TECHNOLOGY -
CRYOGENIC DIELECTRICS

Final Report

November 1, 1972 - September 30, 1976

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ABSTRACT

The Final Report of the Oak Ridge National Laboratory Cryogenic Power Transmission Technology Program covers the period from July 1, 1976 to September 30, 1976.

The apparatus used in the experiments is summarized and briefly described. Much of this equipment will be transferred to other ERDA-supported laboratories.

The entire body of results obtained during the course of the program on breakdown of liquid helium and on breakdown and partial discharge in thin polymer films is summarized. Recent work on ac breakdown for small gaps in supercritical helium is reported for temperatures and pressures ranging from 4.2 to 12 K and 0.3 to 1.5 MPa. Possible mechanisms for helium breakdown in various thermophysical states are considered. Two further variations on the partial discharge experiments on polyethylene films are discussed: in the first, the sample was subjected to high mechanical compressive forces to simulate winding forces in an actual cable; in the second, the sample was prepared in a glove box with particular attention to cleanliness. The results of both experiments were very similar to those of earlier measurements.

Data from related projects in the ORNL Metals and Ceramics Division on ac losses in Type II superconductors and on dispersion hardening of aluminum are summarized in Appendices A and B.

Results of these experiments are discussed in terms of their implications for superconducting power transmission line design, and conclusions are given regarding the selection of appropriate materials, configurations, and operating conditions.

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EXECUTIVE SUMMARY

The Final Report of the Oak Ridge National Laboratory Cryogenic Underground Power Transmission Technology Program summarizes all activity since the inception of the program, with emphasis on the achievements of FY 1976. Sections I through IV cover investigations into the properties of dielectrics at cryogenic temperatures. The results of related studies by the ORNL Metals and Ceramics Division on ac losses in type II superconductors and on dispersion hardening of aluminum for high-strength, low-resistivity conductor substrates are discussed in Appendices A and B, respectively.

Conclusions drawn from the experimental work on dielectrics are discussed in Section V and summarized here. In all cases, the ultimate working voltage of the insulation is determined by thermophysical helium properties and by the geometry of the electrodes, or of gaps in polymer insulation. In systems employing helium alone, failure occurs by breakdown across the gap proper or flashover on solid insulators at field strengths of up to 40 MV/m. For cellulose paper stressed with dc voltage, breakdown occurs through the pores at stresses only slightly higher than the breakdown strength of the helium impregnant alone. In wrapped polymer film systems, partial discharge (PD) is initiated by breakdown of helium in the butt gaps or in residual gaps between film layers. With ac voltage, PD inception stresses above 30 MV/m do not appear to be possible in wrapped polymer dielectrics unless the temperature is reduced to roughly 6 K. With dc, small discharges were observed sporadically only above 100 MV/m, indicating that PD should not be a problem in a dc cable.

In Section I, the Introduction, the history, goals and areas of interest of the project are outlined and a brief summary of the structure of this Final Report is given.

Section II describes experimental equipment used in the cryogenic dielectric studies. A series of unique vacuum-insulated high voltage bushings was developed for the experiments. The nearly identical 10-cm IV-1 and IV-2 bushings were successfully operated at voltages up to 130 kV dc and 45 kV ac. IV-3, a 25-cm bushing, was subjected to 55 kV before shipment to IAST at the termination of the program. Another 100-cm bushing for several hundred kV was under construction, but was not completed at program termination. Available high voltage supplies ranged from a 130 kV dc unit and an 80 kV ac transformer to a 700 kV ac resonant test set which was intended for use with the large 100-cm bushing. A 250 kV, 600 J impulse generator was also purchased. Instrumentation included chart recorders to log output signals from dc and ac voltage dividers, automatic trip circuits to shut off the high voltage on sample breakdown, and a sensitive J. G. Biddle bridge-type partial discharge detector to observe pre-breakdown current pulses in insulation. Temperatures were measured by means of carbon resistors calibrated against a commercial germanium thermometer, whose calibration is traceable to the NBS, and helium pressures were measured by either a Wallace and Tiernan Bourdon tube gauge or by a Rosemount capacitive pressure transducer with digital readout.

Several different test fixtures were developed for the various insulation systems under investigation. For breakdown tests in liquid helium, simple variable-gap electrode systems with sphere-plane,

point-plane, or ASTM geometry sufficed. Gaps could be measured and controlled to an accuracy of $\pm 10 \mu\text{m}$. For breakdown tests in pressurized supercritical helium, another small variable sphere-plane gap was installed in a pressure chamber with an epoxy top to stand off the high voltage. For tests on polymers, multiple-electrode setups were developed to allow several tests with one cooldown of the cryostat. In all these fixtures, the multiple high voltage electrodes were arranged in a circle, and could be individually selected by a rotating arm on the high voltage bushing. The flat, circular polymer samples were enclosed in a metal and epoxy pressure chamber to allow tests at elevated helium pressures. For tests in liquid helium at atmospheric pressure, a single epoxy-encapsulated high voltage electrode on a rotating arm was stepped around the surface of the film sample, with no pressure chamber required.

In Section III, experimental work on liquid and supercritical helium and on polymer dielectrics is summarized. Measurements with sphere-plane electrodes in boiling liquid helium show average and minimum breakdown strengths of respectively, 40 MV/m and 20 MV/m in millimeter gaps. With sharp-edged ASTM electrodes the average field dropped by a factor of three, while with point-plane electrodes the macroscopic breakdown fields were an order of magnitude lower than the sphere-plane values. A strong polarity effect for the point-plane geometry was also noted. The point-negative voltages were considerably lower than the point-positive values, in contrast with conventional liquid dielectrics, which show the opposite behavior.

Breakdown measurements in small ($<250\text{ }\mu\text{m}$) sphere-plane gaps were conducted with supercritical helium under pressures and temperatures ranging up to 1.5 MPa and 13 K. Most of the measurements were carried out with ac voltage. Breakdown fields in the neighborhood of 60 MV/m (peak) were obtained at 1.5 MPa and 12 K, with only a weak dependence on temperature and helium purity. At 0.3 MPa, the breakdown voltages increased strongly with falling temperature as a result of the greater density change at low pressures. Unstable behavior and considerable helium purity dependence were observed in the neighborhood of the transposed critical temperature ($\sim 5.5\text{ K}$).

Breakdown strength and partial discharge inception measurements were carried out on several polymer film samples between flat electrodes. For all materials, the polymer ac breakdown strengths in liquid helium were roughly an order of magnitude higher than the helium values. The results of impulse and dc tests were approximately twice the ac values. Films thicker than $100\text{ }\mu\text{m}$ generally failed by surface flashover to the nearest ground, typically a distance of 2-3 cm. In contrast, experiments on cellulose paper indicated a dielectric strength only slightly higher than that of the helium impregnant alone. In general the breakdown strength fell with increasing film thickness.

The partial discharge (PD) measurements were carried out with three-layer samples impregnated with supercritical helium. Small 1-mm diameter holes were placed in some of the layers to simulate butt gaps in a wrapped tape system. The PD inception fields were found to depend strongly upon the density of the supercritical helium impregnant. PD inception fields above 30 MV/m could be obtained only for temperatures

below 9 K and pressures above 1.2 MPa. Aside from the dielectric constant, which determines the voltage division between holes and film layers, the nature of the polymer used had no obvious effect on the PD performance. Aluminizing the faces of the film adjacent to the holes led to drastically lower inception voltages. The discharge magnitudes with embossed films were unusually high ($\sim 10,000$ pC).

Section IV contains a discussion of the experimental results. The reduced breakdown fields in boiling liquid helium with ASTM electrodes are consistent with geometric field enhancement at the sharp edges. However, in the more divergent fields with point-plane electrodes, the breakdown fields are higher than the values calculated from the geometric field enhancement, evidently because of space charge effects. The polarity effect is explained in terms of the different mobilities of positive and negative ions in liquid helium. Statistical analysis and probability plotting of the liquid breakdown data indicates that they follow a logarithmic extreme value distribution. A possible explanation is advanced in terms of prebreakdown charge pulses with an exponential frequency distribution. Discontinuities observed in the probability plots indicate the existence of two separate breakdown processes. Paschen plots of the supercritical helium data show agreement with the usual Paschen curve for helium at densities below 20 kg/m^3 , indicating that the classical Townsend electron avalanche process dominates breakdown at low densities. At intermediate densities, the observed peak in the breakdown voltages appears to be correlated with the ratio of viscosity to thermal diffusivity. At liquid densities, trapping of electrons in bubbles prevents avalanche processes, and the results

are consistent with Krasucki's bubble theory if the electron bubbles are assumed to nucleate breakdown. The particle theory of Kok can explain the minimum values observed with impurity concentrations far below the standard figure of 20 ppm, indicating that attempts to improve the minimum liquid breakdown strength by purification are unlikely to succeed.

Breakdown measurements on several polymer films gave results ranging from 130 to 220 MV/m with ac voltage. Impulse and dc measurements gave values roughly twice as high. Severe ripping was noticed under impulse conditions for the films with lowest mechanical strength. The rather poor flashover performance with liquid and supercritical helium suggests that stress cones will have to be designed carefully for optimum performance at minimum cost. However, it appears that dielectric strength of the polymer alone will not be a major consideration in a superconducting cable. It is rather the much lower dielectric strength of the helium filling the butt gaps and residual interlayer gaps which limits the ultimate operating stress of the cable. In porous cellulose paper impregnated with supercritical helium, the dc dielectric strength was not much greater than that of the helium alone. In the polymer samples, the ac PD inception levels were well below the breakdown values. The voltages across the 1-mm holes at inception were inferred by assuming that the voltage divides capacitively between the polymer layers and the gap in series with them. The gap voltages were comparable or somewhat lower than typical metallic-electrode Paschen curve values at an equal gap. However, a strong correlation with helium density was evident. Small discharges were seen even at positions on

the sample having no holes. Small residual gaps between polymer film layers appear to be the most likely source for this behavior. PD inception fields of about 30 MV/m were observed at 9 K and 1.5 MPa. However, there is reason for concern about whether this value could be obtained in a long cable. A few PD tests were conducted on polymers with dc voltage. Intermittent small discharges with low repetition rate were observed only above 100 MV/m, indicating that PD should not be a problem in a dc cable.

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I. INTRODUCTION

This is the Final Report of the Oak Ridge National Laboratory Cryogenic Power Transmission Technology Project. This report is also the Annual Report for FY 1976. References 1-14 are the previously issued periodic reports.

Early program activity on dielectrics in the ORNL Thermonuclear Division centered on development of reliable cryogenic high voltage experimental equipment and on measurements of breakdown voltages in slowly-boiling liquid helium. Related programs in ac losses of type II superconductors and in dispersion hardening of aluminum were also begun in the ORNL Metals and Ceramics Division.

The superconductor investigation was stopped at the end of FY 75; the results of that program are summarized in Appendix A. The investigation of dispersion-strengthened, high-purity aluminum for use as the normal metal to stabilize a superconductor was stopped in mid-FY 76; the results of this program are summarized in Appendix B. Later work on dielectrics shifted toward solid polymer films to provide additional data for superconducting cable development projects at Brookhaven National Laboratory and at Los Alamos Scientific Laboratory.

In a project such as this, which is establishing a new area of dielectric investigation involving high voltages, pressurized helium, and low temperatures, each quarter has been a new adventure with new experimental problems appearing as the old ones were solved. The evolving needs of superconducting power transmission cable development have been factored into the dielectrics investigations and the required experimental work has changed while the capabilities for carrying on

high voltage measurements at high pressures in a controlled cryogenic environment were being developed. The principal change has occurred during the past year, when the emphasis of the experimental work was turned toward an investigation of thin film dielectrics immersed in helium rather than on an investigation of helium alone and bulk solids immersed in helium. The dielectrics of interest are the polymeric thin films and papers which are candidates for use as wrapped dielectrics in the flexible ac and dc cables being developed at Brookhaven and Los Alamos, respectively. An experimental survey of partial discharge and electrical breakdown of these materials was established in cooperation with Brookhaven and Los Alamos. Other survey work on the mechanical properties and dielectric loss of these same materials was concurrently conducted by Brookhaven National Laboratory and the National Bureau of Standards in Gaithersburg, respectively. The samples tested came from Brookhaven, where partial discharge and electrical breakdown of short cylindrical samples of these same materials were being investigated.

The change in experimental emphasis motivated the development of a new apparatus for investigating high voltage phenomena in flat, multi-layer thin-film samples immersed in pressurized helium, at temperatures between 4 and 15 K. Other apparatus to measure small-gap breakdown in pressurized helium was also developed. These will be discussed in Section II.

The change also resulted in stopping the development of the large volume, high voltage cryostat, which had been under way since the inception of the project. The principal problem left in that development was the large diameter insulating disc for the vacuum bushing which must

accommodate voltages up to 700 kV ac rms between the center and edge while supporting a pressure gradient of 10 atm across the faces of the disc. A flat disc for operation at voltages up to 500 kV was being fitted for the cryostat when the work stopped, and discussions with manufacturers were under way for conical discs to go to 700 kV. The details of this cryostat design are covered fully in earlier reports, especially those for the eleventh and twelfth quarters.^{11,12}

The experimental results are discussed in Section III, which is divided into two subsections on helium alone and on thin polymer films immersed in helium. The original measurements on 4.2 K liquid helium in millimeter gaps have been extended into the supercritical region for small gaps of the order of 100 μm . These gaps are comparable to the butt gap thickness in a wrapped polymer dielectric, and are of particular interest in the analysis of the polymer partial discharge results. The measurements cover the temperature-pressure range from 4.2 K and 0.3 MPa (3 atm) to 14 K and 1.5 MPa (15 atm), for both helium breakdown and polymer partial discharge. Breakdown results on polymers in liquid and supercritical helium with dc, ac, and impulse voltages are also reported. These measurements were hampered somewhat by the unexpectedly low flashover performance in liquid and gaseous helium, so that most of the thicker polymer samples failed by flashover rather than by breakdown. However, it was possible to obtain lower limits for the breakdown strengths. It is clear from the results that the limiting factor in the high voltage standoff performance of any superconducting power transmission line design will be breakdown of the helium, as evidenced by either partial discharge in a wrapped polymer dielectric, or by spacer flashover in a liquid or supercritical helium dielectric.

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II. APPARATUS

A. Bushings

The chief problem in high voltage cryogenic work is associated with bringing the high voltage from room temperature to cryogenic temperature without flashover or corona discharge. For our purposes this was accomplished by the use of vacuum insulation between a grounded external stainless steel tube and a central high voltage tube. A typical intermediate cryostat arrangement is shown in Fig. 1. Three such intermediate voltage cryostats have been built. IV-1 and IV-2 are nearly identical and have the dimensions shown in Fig. 1. A larger cryostat, IV-3, was constructed with a vacuum bushing about two and one-half times larger in diameter to allow the use of higher voltages and larger samples. The top high voltage end of the bushing at room temperature was insulated from ground with a large glass tube. The lower high voltage end was sealed with a fiberglass-reinforced epoxy disc (G-10) manufactured by Synthane, Inc. This was sealed to stainless steel fittings with Stycast 2850 FT (blue) epoxy manufactured by Emerson and Cuming. A central tube inside the high voltage tube was used for positioning the top electrode as well as for filling the dewar with liquid helium. IV-1 and -2 were operated at voltages up to 130 kV dc and 45 kV ac.

B. Cryostats and Power Supplies

Power supplies already on hand at ORNL were sufficient for running 130-kV dc and 80-kV-rms ac experiments on thin films or for small gaps. A 250-kV, 600-J impulse generator, consisting of ten stages and purchased from Universal Voltronics, Inc., was set up for use as shown in Fig. 2.

Fig. 1.

Fig. 2.

A heat exchanger for refrigerating gaseous helium around the electrodes of IV-3 was designed and fabricated. The heat exchanger can be used to precool the cryostat to liquid nitrogen temperature without introducing nitrogen into the cryostat by circulating liquid nitrogen in the copper coils. The cryostat can then be cooled to 4.2 K by transferring helium directly. After the liquid helium boils away to below the level of the test sample chamber, liquid helium can be circulated at a controlled rate through the heat exchanger coils to maintain the temperature at any desired level greater than 4.2 K. The heat exchanger is shown (on the right side of Fig. 3) as it is being lowered into the IV-3 dewar. The IV-3 bushing (shown in the center of the figure) can then be lowered into the center of the refrigerator coil-dewar combination.

C. Instrumentation

Instrumentation during the past year has involved three areas: (1) the recording of breakdown and partial discharge voltages, (2) the measurement of temperature over the range of 4 to 15 K, and (3) the measurement of pressures from 0.1 MPa (1 atm) to 1.5 MPa (15 atm).

Measurements of dc breakdown voltages were recorded on a dc strip chart recorder with the signal coming from a voltage divider through a filter circuit. Measurements of ac voltages were done with a capacitive voltage divider. In order to record ac voltages on the same recorder, a Burr-Brown rms-to-dc convertor module was installed on the same panels as the dc network. Appropriate compensators permit calibration, accurate to 0.02 kV, of the recorder chart for either ac or dc breakdown.

Fig. 3.

A tripping circuit built into the dc supply terminated dc breakdowns. This circuit automatically removed the high voltage when the current reached 1 mA. A Schmitt trigger circuit (described on pp. 21 and 44 of Ref. 9) sensed the primary current in the ac high voltage supply and was arranged to interrupt similarly the high voltage upon breakdown. A J. G. Biddle bridge type detector (described in Ref. 2, p. 89) was used to measure partial discharge inception and extinction voltages.

For the measurement of temperature, ten Allen-Bradley 1/8-W, 100- Ω carbon resistors were calibrated against a commercially calibrated germanium (Ge) thermometer over the range of 4 to 25 K. The quoted accuracy of the Ge thermometer in the range of 5 to 20 K is 0.01 K. The calibrated points were fitted to a four-parameter equation. Tables of R vs T for each of the carbon resistors were used to convert resistance to temperature. Resistance measuring equipment was built into each of two moveable cabinets. A standard three-wire ac bridge circuit was used (as shown in Ref. 11, p. 41). A lock-in detector was used with this bridge to obtain the greatest sensitivity in partial discharge measurements. For breakdown measurements, the lock-in could not cope with the large transients at breakdown, and so a similar dc bridge was constructed using a 1.5-V mercury cell for driving voltage and a vacuum tube voltmeter as a detector. Measurements with the two bridges agreed closely.

A 200-psia Wallace and Tiernan gauge readable to 0.2 psi was used for pressures up to 1.4 MPa. A Rosemount capacitive pressure transducer with digital display readable to 0.1 psi was used for

pressures up to 1.7 MPa. Appropriate fittings and valves for connecting and regulating helium gas pressure or vacuum were incorporated in the cabinets which housed the other equipment.

A typical experimental layout for dc breakdown measurements is shown in Fig. 4, which shows most of the equipment which has previously been discussed.

D. Test Fixtures

1. Liquid Helium

Electrodes for determining the breakdown strength of liquid helium at 4.2 K and at atmospheric pressure were designed and fabricated of stainless steel in different shapes. Sets of sphere-plane and point-plane electrodes were constructed. The sphere electrode had a diameter of 38 mm, while the point electrode had a conical half-angle of 18° with an average tip radius of 230 μm . For comparison with commercial breakdown tests a set of sharp edged electrodes were made according to ASTM specifications (Fig. 5).

The electrode gap in all of these configurations could be adjusted by a micrometer screw mechanism located at the top of the high voltage bushing, and could be measured to a precision of $\pm 10 \mu\text{m}$.

2. Tests on Polymers at 4.2 K

In order to run practical tests on polymers in liquid helium, an arrangement using a flat circular sheet of dielectric to be tested on a round stainless electrode plate was designed (see Fig. 6). The upper electrode was a spring loaded hemisphere attached to a rotating arm which was fixed to the central drive rod in the high voltage lead-through. The hemispherical electrode could be manipulated to various

dc Breakdown - Experimental Layout.

Fig. 4.

ORNL DWG. 75-2545

Fig. 5.

PHOTO 4024-75

Fig. 6.

positions from the top of the high voltage bushing. Since the upper electrode exhibited excessive flashover when used (as shown in Fig. 6), the hemispherical electrode was cast in a cylindrical mold of Stycast 2850 FT epoxy. Although some partial discharge glow was still present, reproducible results for both partial discharge and breakdown could be obtained.

3. Partial Discharge Measurements in Supercritical Helium

Since the dewars for cryostats IV-1 and IV-2 were of glass and the metal dewar for IV-3 was designed for a working pressure of no greater than 0.3 MPa, a special arrangement was required for testing multiple layers of film and butt gaps at helium pressures up to 1.5 MPa and temperatures greater than 4.2 K. The test fixture which proved successful was constructed of two circular pieces that were tightly clamped together with an indium seal between them. An exploded cross-sectional view of a typical demountable test fixture is shown in Fig. 7. The top insulating epoxy disk (4) is 2.5 cm thick and is embedded with eight rounded stainless steel electrodes (1). The bottom metallic ground piece (13) holds the ground plate (10) and samples (5). A slotted central bolt (14) fits into the embedded nut (3) to prevent the fixture from bulging at the center when pressurized with helium through the plenum chamber (16). Disks of plastic foam (8) and aluminized Mylar (7) are inserted between the ground plate and plastic film specimens to ensure a snug fit in spite of any irregularities between the electrodes. A flat disk heater element, a carbon thermometer, and an electrical feed-through are incorporated in the bottom piece (13) but are not shown in Fig. 7. The assembled fixture is shown attached to the IV-1 bushing in Fig. 8.

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Fig. 7.

Fig. 8.

Before assembly, the fixture is pressure tested at room temperature to 2.0 MPa. For an experimental run the sample chamber is evacuated and flushed several times with helium. Only enough liquid helium is introduced into the cryostat to cover the fixture, and gaseous helium is introduced into the sample chamber to the desired pressure. Measurements can then be made at 4.2 K. As the liquid helium boils away to a level below the test fixture, the temperature slowly rises and can be monitored with a carbon thermometer. A constant-pressure, variable-temperature run from 4.2 to 12 K requires three to six hours.

4. dc Dielectric Strength of Polymers

The demountable test fixture described in the previous section proved suitable for dc dielectric strength tests, with some limitations, on polymer films. Once a true breakdown occurred at any one electrode position, no more tests could be performed at that position. Further, since temperatures greater than 4.2 K were attained by allowing the liquid helium level to fall below the test fixture, partial discharges could occur in the cold, low pressure helium gas space surrounding the test fixture, bushing, and rotating high voltage arm.

A 30-cm-diam test fixture similar to the one (described in Section 3, above) in the IV-3 cryostat was designed to provide larger spaces between electrodes in order to reduce external discharges. A cross-sectional view of the test fixture for IV-3 is shown in Fig. 9. The top insulating plate is a 2.5-cm-thick piece of G-10, 30-cm-diam, in which the eight electrodes are embedded in epoxy. Otherwise it is quite similar to the test fixture shown in Fig. 7.

Fig. 9.

5. Breakdown Strength of Supercritical Helium

Some of the multi-layer polymer specimens contained 1- or 2-mm-diam holes filled with supercritical helium in order to simulate butt gaps. However, very little information is available on the breakdown of supercritical helium at small gaps ($\sim 100 \mu\text{m}$). A small test fixture was designed and built to measure the small-gap ($\sim 100 \mu\text{m}$) breakdown of supercritical helium at pressures between 0.3 and 1.5 MPa and temperatures between 4.2 and 12 K. A cross-sectional view of this test fixture is shown in Fig. 10. The top spherical electrode is attached to a moveable shaft that extends to the top of the high voltage bushing. A micrometer at the top of this bushing permits setting the gap between the 12.5-mm-diam sphere and the plane ground plate to an accuracy of about $5 \mu\text{m}$.

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Fig. 10.

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III. EXPERIMENTAL WORK

A. Helium Breakdown

Breakdown of liquid and cold gaseous helium is of major interest because in various cable designs under study, helium is considered either as the primary insulant or as the impregnant in a lapped polymer or paper tape insulation system. In the former case, large-gap studies up to several centimeters are necessary, while in the latter case measurements are required at gaps between 50 μm and 250 μm , corresponding to the thickness of the butt gaps between adjacent turns of tape. Breakdown measurements in liquid and cold gaseous helium have been reported by several authors.¹⁵⁻²⁴ In general the breakdown voltage has been found to depend on such factors as the temperature, pressure, and density of the helium; the presence of impurities; the size, configuration, surface finish, and material of the electrodes; and the nature and polarity of the applied voltage (ac, dc, or impulse).

1. Breakdown Experiments with Boiling Liquid Helium

a) Sphere-Plane Electrodes:

Early experiments in this program involved positive and negative polarity breakdown in slowly boiling liquid helium at atmospheric pressure, with a 3.8-cm-diam stainless steel or chrome plated brass sphere and a stainless steel grounded plane. The apparatus is described in Section II.A and shown in Fig. 1. The voltage was ramped at several rates by a motor-driven Variac on the high voltage supply. With positive polarity, no significant variation in the mean or minimum breakdown fields was observed for ramp rates between 0.08 kV/sec and 10 kV/sec. Therefore all later experiments were performed at 2 kV/sec, which was fast enough

to obtain enough values for statistical purposes, but slow enough to allow easy observation of the voltage ramp on a chart recorder. At least 60 values were taken for each gap.

Average and minimum breakdown fields vs gap are shown for the sphere positive and negative in Fig. 11. The positive field breakdowns were always slightly higher than the negative ones, in agreement with the results of other workers.²² However, both appear to be saturating at an average value near 400 kV/cm, and a minimum value of about 200 kV/cm. Probability plotting of the results (Fig. 12) indicated that the data were distributed according to the function

$$p(x) = \frac{k}{x} \left(\frac{\mu_0}{x} \right)^k \exp \left[- \left(\frac{\mu_0}{x} \right)^k \right], \quad (1)$$

where the variate x is the logarithm of the breakdown voltage and $p(x)$ is the probability of survival. Abnormalities were noted in the slopes of the plots near the 90% level.

A few ac measurements were also made at gaps of 0.25 and 0.50 mm, and the peak breakdown fields agreed closely with the dc data. Measurements at larger gaps were prevented by high helium boiloff, which was later traced to dissipation resulting from a slow leak into the cold end of the vacuum bushing.

b) ASTM Electrodes:

In order to allow a comparison with standard test data for conventional liquid dielectrics, breakdown measurements were carried out with standard ASTM electrodes,²⁵ shown in Fig. 5. The edge radius was found by shadow graph measurements to be roughly 40 μ m. Because of the helium boiloff problem mentioned above, the first measurements were conducted with

Fig. 11.

Fig. 12.

negative dc voltage. Averages of 30 breakdown voltage measurements are plotted vs gap in Fig. 13 (closed circles), using a voltage ramp rate of 2 kV/sec. The ASTM breakdown voltages were only about one-third of the sphere-plane voltages at corresponding gaps, due to the sharp edges specified in the ASTM design. However, the scatter was much lower, with standard deviations of 4 to 6% as compared to a typical value of 20% for the sphere-plane results. Cold helium vapor (open circles) was found to have one-half and liquid nitrogen (crosses) was found to have one and one-half times the boiling liquid helium ASTM breakdown strength.

ASTM breakdown results with ac voltage using a ramp rate of 1.8 kV/sec are shown in Fig. 14. The first set of readings (closed triangles) were low in comparison with the previous dc measurements (closed circles), and so the electrodes were repolished to remove pitting from previous runs. A new set of ac and dc data was then taken on the same day using a single filling of helium. The new results showed good agreement between peak ac and dc voltages but were systematically higher than the earlier readings. The scatter also increased to 8 to 10%. The increase in breakdown voltage may have resulted from the removal of some small asperity during the repolishing or from changes in the impurity content of the helium. The later measurements are comparable to the minimum acceptable value for C-10 transformer oil (square).

c) Point-Plane Electrodes:

In order to assess the effect of voltage polarity in breakdown with non-uniform fields, breakdown experiments were carried out using the asymmetric point-plane system. The stainless steel point had a half-angle of 18° and a tip radius of 166 μm . In later experiments, the tip was repolished, and the radius increased to 230 μm .

Fig. 13.

Fig. 14.

Breakdown voltages for the 230- μ m point with dc and ac voltage are shown in Fig. 15 and in Tables I-III. The points are averages of 20 breakdowns. A strong polarity effect was noted; with the point positive, the breakdown voltages were roughly twice those with negative polarity. In fact the positive voltages are comparable to the dc ASTM values (Fig. 13). The peak ac voltages corresponded to the lower negative dc voltages, as would be expected.

The dashed lines in Fig. 15 show breakdown voltages for liquid helium deliberately contaminated with air. The values are tabulated in Tables IV and V. Prior to the experiment the cryostat was cooled to liquid nitrogen temperature and the liquid helium dewar was evacuated. Air was introduced to maintain a stable pressure of 165 mm, and the dewar was then backfilled with dry helium gas to atmospheric pressure. Thus, the amount of air added could be easily reproduced in all runs. After the transfer of liquid helium, the contamination could be seen as a fine mist covering the electrodes and interior of the dewar. By converting the liquid helium and contaminant to gas volumes at STP, the equivalent impurity concentration was found to be about 1000 ppm, much higher than could be tolerated in an operational transmission line.

Even at this high impurity concentration, little effect on the breakdown voltages was seen. In fact, slightly higher average breakdown voltages were observed with contamination, although the increase was well within the scatter of the results. Furthermore, after a few breakdowns the contaminant film on the plane electrode was vaporized and condensed farther away, so that the portion of the electrodes under stress was cleaned up rapidly.

Fig. 15.

Table I. Breakdown data in technically pure liquid helium (point negative)

d (mm)	$V_{B \text{ min}}$ (kV)	$\frac{V_{B \text{ min}}}{\bar{V}_B}$	$V_{B \text{ max}}$ (kV)	$\bar{V}_B$ (kV)	$\sigma/\bar{V}_B$ (%)	No. of Readings
1.0	14.9	.66	29.2	22.7	19.8	10
1.0	9.0	.39	32.2	22.9	21.4	50
1.0	16.6	.62	36.7	26.7	17.6	20
2.0	15.7	.51	42.0	30.5	23.3	20
4.0	21.5	.61	49.6	35.3	27.2	20
6.0	22.5	.59	50.0	38.0	25.3	20
10.0	29.0	.69	53.5	42.2	22.8	10

Table II. Breakdown data in technically pure liquid helium (point positive)

d (mm)	$V_{B \text{ min}}$ (kV)	$\frac{V_{B \text{ min}}}{\bar{V}}$	$V_{B \text{ max}}$ (kV)	$\bar{V}_B$ (kV)	$\sigma/\bar{V}_B$ (%)	No. of Readings
0.5	26.7	.71	49.5	37.5	17.3	20
1.0	29.2	.67	62.0	43.3	20.8	20
2.0	35.4	.61	76.1	57.8	20.8	20
2.0	24.0	.49	67.8	49.0	25.5	20
3.0	40.0	.71	78.0	56.0	18.6	20
4.0	43.7	.74	79.5	58.8	17.5	10
6.0	36.3	.61	83.2	59.8	23.4	20

Table III. Breakdown data in technically pure liquid helium at field conditions

d (mm)	$V_{B \text{ min}}$ (kV)	$\frac{V_{B \text{ min}}}{\bar{V}_B}$	$V_{B \text{ max}}$ (kV)	$\bar{V}_B$ (kV)	Peak $\bar{V}_B$ (kV)	σ/V_B (%)	No. of Readings
1.0	8.3	.66	17.1	12.5	17.7	22.4	20
2.0	14.2	.65	28.4	22.0	31.1	15.9	20
4.0	15.7	.65	35.5	24.3	34.4	20.2	20
6.0	19.5	.70	35.1	27.7	39.2	13.7	20
10.0	20.3	.76	35.5	26.7	37.8	15.4	20

Table IV. Breakdown data in liquid helium
contaminated by air (point positive)

d (mm)	$V_{B \text{ min}}$ (kV)	$\frac{V_{B \text{ min}}}{\bar{V}_B}$	$V_{B \text{ max}}$ (kV)	$\bar{V}_B$ (kV)	$\sigma/\bar{V}_B$ (%)	No. of Readings
1.0	27.6	.60	64.0	46.3	27.2	20
2.0	30.3.	.59	66.2	51.1	17.8	20
4.0	38.9	.65	74.0	59.9	16.5	20
6.0	46.2	.70	76.0	65.7	12.8	20

Table V. Breakdown data in liquid helium
contaminated by air (point negative)

d (mm)	$V_{B \text{ min}}$ (kV)	$\frac{V_{B \text{ min}}}{\bar{V}_B}$	$V_{B \text{ max}}$ (kV)	$\bar{V}_B$ (kV)	$\sigma/\bar{V}_B$ (%)	No. of Readings
1.0	16.0	.64	31.5	25.1	16.7	20
2.0	25.5	.71	45.7	35.7	17.1	20
4.0	23.7	.54	59.5	44.1	22.5	20
6.0	26.9	.58	58.7	46.4	18.3	20

Visual observation of the electrodes with a 20-power telescope gave no evidence of prebreakdown light or liquid motion. However, at breakdown a region of diffuse boiling persisted around the point for nearly a second.

The scatter in the breakdown voltages often exceeded 20% and did not appear to be affected by contamination with air or nitrogen. Typically, the minimum breakdown voltages were between 60 and 70% of the average values. In earlier studies with the 166 μm point, a few tests were made in which 50 breakdowns were taken at constant gap for statistical analysis. Probability plots with the point positive and with a gap of 2 mm are shown in Fig. 16. As with sphere-plane electrodes, the best fit is found for a logarithmic extreme value distribution function.

2. Breakdown Experiments with Pressurized Supercritical Helium

Because of the low breakdown strengths of liquid and cold gaseous helium in comparison with polymer films, it might be expected that partial discharge in wrapped insulation begins when the field gradient in the butt gaps exceeds the helium breakdown strength. Therefore, measurements of helium breakdown in electrode gaps comparable to the polymer film butt gaps are of interest. Preliminary experiments with a sample chamber containing eight fixed gaps were mentioned in the previous quarterly report.¹⁴ These measurements were inconclusive because the gaps could not be measured precisely. A new pressurized test cell (Fig. 10) containing stainless steel sphere-plane electrodes with variable gap was fabricated. A vernier micrometer controlled the position of the high voltage sphere electrode, allowing a precision of $\pm 5 \mu\text{m}$ in the gap measurement. The pressure and temperature were

Fig. 16.

respectively monitored to ± 0.7 kPa and ± 0.05 K by a Rosemount electronic pressure gauge with digital readout and by a calibrated carbon resistor mounted inside the chamber under the plane electrode. The ac voltage was measured with a capacitive divider which drove a dc chart recorder through an electronic true rms convertor. For dc measurements, the recorder was driven directly by the divider. The voltage was ramped manually at about 2 kV/sec. Use of faster or slower ramp rates had no discernible effect. The helium fill line to the chamber contained a liquid nitrogen-cooled coil to trap pump oil and other condensable vapors. A bronze wool particle filter was added later. The sample chamber and electrodes were cleaned with trichloroethylene and alcohol, and the chamber was flushed several times with room temperature helium gas after assembly.

After precooling the cryostat with liquid nitrogen, liquid helium was transferred into the dewar until the sample chamber was partially immersed. Helium from a gas cylinder was admitted to the chamber to maintain the desired pressure during the transfer. After checking the voltage calibration with an electrostatic voltmeter, the electrodes were conditioned by repeated breakdown until average values over several shots stabilized. Ten or more breakdowns were then taken at 4.2 K and averaged. The helium in the dewar was boiled away until its level fell below the sample chamber, which then began to warm up. A heat lamp directed into the cryostat increased the warming rate, so that the range from 4.2 to 13 K could be covered in four to six hours. As the temperature rose, the pressure was carefully maintained to within 0.7 kPa of the desired value by slowly venting the chamber through a needle valve. Breakdown measurements were taken roughly every minute. Periodically the heat lamp was switched

off and measurements of breakdown voltage vs gap were taken at nearly constant temperature.

a) Technically Pure Helium:

The first measurements were taken using general purpose helium gas in the sample cell. The quoted helium purity was 99.996%, although the sample had not been spectrometrically analyzed. The same grade of helium was used in the polymer film partial discharge experiments discussed in Section III.B.

The sample chamber was evacuated at room temperature over the weekend before cooling the apparatus down. Plots of the peak ac breakdown voltage vs temperature for a 101.6- μm gap are given in Fig. 17 for pressures of 0.3, 0.9, and 1.5 MPa. A set of dc measurements for the same gap at 0.9 MPa is also plotted. The dc and ac data agreed well at 5 K, but for higher temperatures the dc measurements were about 20% lower at 8 K and 10% lower at 12 K. The breakdown voltages decreased monotonically with increasing temperature at all pressures. Below 5 K the breakdown voltages were relatively insensitive to pressure, in keeping with the low compressibility of helium in this region. However, between 6 and 12 K a strong increase in breakdown voltage with pressure was observed. At 12 K and 1.5 MPa (values appropriate for wrapped dielectric cable designs) the breakdown strength was roughly 60 MV/m.

Plots of the peak ac breakdown voltages vs gap at temperatures near 4.2, 5.5, and 8.5 K are given in Figs. 18-20. Each point is an average of five values with maximum and minimum values given by the bars. The minimum values were nearly always greater than 90% of the averages. The breakdown voltages rose roughly linearly with gap, although

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Fig. 17.

Fig. 18.

Fig. 19.

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Fig. 20.

a slight total voltage effect was indicated by the fact that the plots did not extrapolate to zero. For the 1.5 MPa measurements, the voltages at 5.5 K were actually slightly higher than those at 4.2 K. At lower pressures, the breakdown voltages decreased with increasing temperature at all gaps, as can be seen by comparison of the plots in Figs. 18-20.

When the sample chamber was opened, the electrode surfaces were found to be very finely pitted, so that the surface finish was more dulled than covered by individual asperities. The ground electrode was also covered with very fine powdered metal blasted from the electrodes by the breakdowns. However, the region between the electrodes was clean, and so this powder probably did not affect the breakdown voltages. The electrodes were thoroughly cleaned and repolished in preparation for the experiments described in the next section.

b) Highly Purified Helium:

In order to assess the effects of purity on helium breakdown, a few experiments were carried out with greater precautions for cleanliness. A supply of "Navy Grade A" helium, which had been double charcoal purified and guaranteed air and moisture free, was procured. The specified purity was also 99.996%, although conversations with the supplier²⁶ indicated that the typical purity was 99.998%. The electrodes were repolished, and the sample chamber, fill lines, and electrodes were completely flushed with trichloroethylene and alcohol. An extra in-line filter was added to the fill line. A valve was installed between the liquid nitrogen-cooled coil in the fill line and the sample chamber to prevent entry of contaminant vapors in case the nitrogen boiled away overnight. Breakdown measurements were then performed in the same fashion as described previously.

Plots of the breakdown voltage vs temperature at a pressure of 0.3 MPa are shown in Fig. 21, for gaps of 50.8, 101.6, and 152.4 μm . Comparison of the 101.6- μm measurements with the results for technically pure helium shows a clear increase in breakdown voltage with improved purity, for temperatures below about 9 K. At higher temperatures, purity does not seem to have as great an effect. In addition, the high purity results show evidence of a pronounced peak in the breakdown voltage in the region near 5.3 K. The possible implications of this will be discussed later. The scatter in the breakdown values increased strongly below 5.6 K, which is close to the transposed critical line at 0.3 MPa. Although a two-phase mixture is impossible at this pressure, helium might still be expected to exhibit more liquid-like or gas-like behavior for temperatures below or above 5.6 K, respectively.

Figure 22 shows plots of the peak breakdown voltage vs gap at various temperatures for the 0.3-MPa data. The plots show a linear gap dependence but do not extrapolate through zero gap, just as for the results with technically pure helium. The 4.2 K values showed poor reproducibility from day to day. The plot for 5.3 K was also much steeper than those for lower or higher temperatures. This is probably connected with the peak at 5.3 K noted previously.

Unfortunately, termination of the Cryoelectric Project made it necessary to discontinue these interesting experiments, and only the measurements at 0.3 MPa were completed. This pressure was chosen because it provides a greater range of density variation over temperatures from 4.2 to 12 K than higher pressures. On the other hand, pressures less than 0.22 MPa lie in the subcritical region and would have led to a

Fig. 21.

Breakdown Voltage vs Gap

Fig. 22.

two-phase system. Further helium measurements at higher pressures and under similar purity conditions would be of great interest in estimating the maximum attainable partial discharge inception voltages for wrapped tape insulation.

B. Insulating Films in Helium

1. Impulse Breakdown Strength

Impulse breakdown strength measurements were carried out on a variety of polymer films using the experimental arrangement described in Section II.D and shown in Fig. 2. Single films, immersed in slowly boiling liquid helium, were impulsed with standard 1.2/50 μ sec lightning waves (risetime 1.2 μ sec, duration at half-peak, 50 μ sec) at progressively higher levels until breakdown occurred. Ten shots were applied at each voltage before the impulse level was raised; the increments between steps were approximately 2 kV. Since the impulse generator was rated 250 kV (10 stages, 25 kV each), it was necessary to disconnect some stages for better control of the output level. The breakdown was observed on the oscilloscope display and was accompanied by a bright flash and a ticking sound. However, the only way to distinguish a bulk breakdown from a surface flashover without removing the sample was to reapply the impulse at the suspected site. In the case of breakdown, reapplication of the voltage, even at reduced levels, resulted in a considerably distorted and relatively flat output on the oscilloscope. On the other hand, a site where a flashover had occurred could withstand relatively large impulse voltages without distorting the waveform. Of course, once the film was removed from the cryostat, trackings, characteristic of surface flashover, were observed. (See Fig. 23.)

Fig. 23.

Results of the impulse measurements are shown in Table VI. With one film (Van Leer Valeron Polyethylene), impulse dielectric strengths were measured under positive and negative polarity. No significant polarity effect was observed. The thinner films (25-38 μm) showed clean breakdown sites and produced high dielectric strengths ($>400 \text{ MV/m}$). Thicker films exhibited a tendency toward surface flashover. With certain films, particularly the 100- μm -thick Rowland Polysulfone, application of the impulse voltage resulted in ripping of the film.

2. dc Measurements

a) dc Breakdown Strength of Polyethylene Immersed in Liquid Helium

The dc breakdown strength of a 38- μm -thick Cryovac D-330 SW polyethylene film immersed in liquid helium was measured. This was done primarily to assess the performance of the film under high dc fields. The results are shown in Table VII. Breakdown occurred without any visible or audible partial discharges. The average dc breakdown strength was 529 kV/mm, practically the same as the impulse breakdown strength.

b) dc Breakdown Strength Measurements Under Supercritical Helium

Conditions:

By the end of 1975 a statistically designed experimental plan evolved (see Ref. 12, Appendix A) after consultations between our group and the Los Alamos Scientific Laboratory dc cable group. The primary objective of the plan was to determine the dc breakdown strengths of various dielectric films, with voltage polarity and ramp rate as parameters under fixed helium conditions (1 MPa and 12 K). Crocker cellulose paper and a variety of polymers were candidate materials.

Table VI. Lightning impulse measurements on polymer films

SAMPLE	POSITION (DEGREES)	$\hat{V}_B$ (kV)	MEAN E_B	COMMENTS
Van Leer Valeron Polyethylene (4 mil or 101.6 μm thick)	0	- 30	7.16 kV/mil or 282 kV/mm	Although breakdown punctures occurred at some positions, flashover paths could be seen over the surface of the film. The film was ripped at the 240° position.
	180	- 28		
	60	- 30		
	120	+ 30		
	240	+ 28		
	300	+ 26		
Makrofol KG Poly- carbonate (3 mil or 76 μm)	0	+ 23.7	6.67 kV/mil or 262 kV/mm	All electrode positions were interconnected by trackings. Puncture could be observed only at 60° and 120° positions, although small burned holes could be observed along the trackings
	180	+ 22.0		
	60	+ 18.0		
	120	+ 22.0		
	240	+ 20.0		
	300	+ 20.0		
Rowland Polysulfone (4 mil or 101.5 μm)	---	+ 17.5 to + 27	----	The film was badly ripped open on two separate attempts.
Cryovac 929D Polyethylene (1 mil or 25.4 μm)	0	+ 18	17.5 kV/mil or 689 kV/mm	All clean breakdowns.
	180	+ 19		
	270	+ 17		
	90	+ 16		
Cryovac D-330DW (1.5 mil or 38 μm)	0	+ 21.2	13.12 kV/mil or 516 kV/mm	Flashover paths could be seen, although the film was punctured at all four electrode positions. At 0° and at 90° flashover occurred at 21.6 and 20.6 kV respectively.
	90	+ 20.0		
	180	+ 20.0		
	270	+ 17.5		
Cryovac D-330 SW (1.5 mil or 38 μm)	0	+ 16.8	11.82 kV/mil or 465 kV/mm	Some evidence of partial discharge spread, but no major trackings. The film was punctured at all four positions.
	90	+ 16.5		
	180	+ 18.8		
	270	+ 18.8		

Table VII. dc breakdown strength measurements

SAMPLE	POSITION (DEGREES)	BREAKDOWN VOLTAGE (kV)	AVERAGE BREAKDOWN FIELD (kV/mm)
Cryovac D-330 SW polyethylene, 38 μ m thick	0	17.3	529
	90	18.2	
	180	22.6	
	240	20.0	
	300	22.7	

Preliminary experiments were performed on 75- μ m-thick Crocker cellulose paper. Two layers of this material were assembled in the specimen chamber described in Section II.D.2. The specimen chamber was installed in the IV-2 cryostat. Since there was some concern about the clearance around the specimen chamber initial measurements were made at and around 4.2 K, where the dielectric strength of the helium surrounding the specimen chamber is higher than at 12 K. Temperatures above 4.2 K were obtained by first filling the liquid helium to cover the specimen chamber and then boiling it away to a level below the chamber, thereby allowing the chamber to warm up slowly to 12 K. Measurements were made as the temperature drifted up and no attempt was made to fix the temperature constant at 12 K. The voltage was measured by a resistive potential divider with a chart recorder, calibrated against an electrostatic voltmeter. Manual and motorized (2 kV/sec) ramps were used to increase the voltage. Temperature was measured using a carbon resistance thermometer mounted inside the specimen chamber and calibrated against a commercial germanium thermometer as described in Section II.C. During the first half of the experiment the voltage polarity was positive, and during the second half it was negative. Measurements at 4.2 K were made when the liquid was in contact with the specimen chamber. Prior to measurements the bushing was conditioned to 41 kV in the case of positive polarity and 23.7 kV in the case of negative polarity.

The results of the experiments are shown in Table VIII. It can be seen that it was possible to reapply approximately the same or higher voltage even after the breakdown was supposed to have occurred. Each time the voltage tripped it was accompanied by a transient pressure and

Table VIII. Experimental observations with two layers of
75- μ m cellulose paper stressed with dc, in
helium pressurized to 1 MPa

Position	Temperature (K)	Breakdown Voltage (kV)	Comments
0°	4.2	+20.0	Manual ramp. No light visible inside dewar.
	4.2	+22.0	Higher voltage when reapplied. Manual ramp and no visible light.
	4.2	+12.7	Voltage reapplied with manual ramp. No light.
90°	4.2	+15.0	Manual ramp. No light.
		+18.5	Voltage reapplied. Manual ramp. No light.
		+14.7	Voltage reapplied. Manual ramp. No light.
		+14.5	Voltage reapplied. Manual ramp. No light.
45°	5.40	-14.8	Manual ramp. No light.
	5.45	-17.2	2 kV/sec ramp. No light. Voltage reapplied.
		-18.3	2 kV/sec ramp. No light. Voltage reapplied again.
135°	7.33	-15.4	2 kV/sec ramp. Could see glow inside the dewar.
	7.54	-14.5	2 kV/sec ramp. No light.
180°	9.1	-17.3	2 kV/sec ramp. Could see light.
		-15.5	2 kV/sec ramp. No light.

temperature rise. Examination of the specimens after removal on the following day showed clear non-carbonized breakdown punctures. Moreover, the number of punctures was the same as the number of voltage applications. The metallized Mylar surface that acted as the cathode also showed breakdown damage. In fact, the arcing caused the metallized surface to be eroded at each breakdown site. This, together with the lack of carbonization of the paper at the breakdown sites, accounts for the high levels of voltages that could be reapplied to each electrode position.

The occasional incidence of glow occurring inside the dewar revealed that the clearances in the cryostat and possibly the specimen chamber were only marginal; to achieve higher voltages another cryostat or specimen chamber was required.

The experiment with cellulose paper was therefore repeated more systematically using the IV-2 bushing in the IV-3 dewar. This allowed use of the IV-3 cooling coil for constant temperature measurements. Furthermore, the helium surrounding the sample chamber could be pressurized to 0.25 MPa absolute, to improve its dielectric strength. Two fresh layers of 75- μ m-thick cellulose paper were assembled in the specimen chamber, which was mounted on the IV-2 bushing. The metallized Mylar in the new assembly was replaced by a 25- μ m-thick aluminum foil. The bushing was then introduced into the IV-3 dewar. The specimen was precooled by circulating liquid nitrogen through the cooling coils. The sample chamber was filled with helium to a pressure of 1 MPa, and liquid helium was transferred into the dewar. The high voltage arm was lifted away from the electrodes and the bushing was conditioned to about 40 kV. The helium was then boiled off to a level below the sample chamber to allow the chamber and the specimen to warm up slowly. The temperature was monitored with a

carbon resistance thermometer mounted inside the specimen chamber. When the temperature registered approximately 11 K, the cooling coil was connected to the liquid helium tank. By adjusting the liquid helium flow through the cooling coil, the specimen temperature could be kept very nearly constant. Breakdown measurements were then made and the results are shown in Table IX.

Examination of the paper sample after the experiment showed clean breakdown sites less than 1 mm in diameter. No carbonization was visible at the breakdown site, as in the earlier experiment. Breakdown was apparently through the helium filled pores of the paper, which explains the relatively low values for the dielectric strengths.

Breakdown experiments using two layers of 75- μ m-thick Mylar were unsuccessful due to flashover problems occurring between 35 and 40 kV. However, it can be concluded from these measurements that the dc breakdown strength of 75- μ m Mylar is at least 467 MV/m.

In order to assess the effect of butt gaps on dc breakdown, three layers of 75- μ m-thick cellulose paper with 1-mm-diam holes at various positions were installed in the cryostat. This is a commonly used method for simulating butt gaps between adjacent turns in wrapped tape insulation. It must be emphasized that in an actual cable, the butt gap is a very long, narrow cavity which threads the insulation helically, rather than a compact, disc-shaped cavity. However, dielectric stresses for the two cases are expected to be similar.

The results are shown in Table X. No significant deterioration in breakdown strength was noticed at the hole positions. The average breakdown strength was 52 MV/m for the three-layer paper sample as opposed to 69 MV/m for the two-layer sample. It should be noted that the polarity and the ramp rate were different for the two cases.

Table IX. dc breakdown values for cellulose paper (2 layers, each 15- μm thick) supercritical helium at 1 MPa, at a ramp rate of 2 kV/sec (top electrode negative)

Position	Temperature (K)	Breakdown Voltage (kV)	Average Breakdown Voltage (kV) ^a	Average Dielectric Strength ^a (kV/mm)
0°	11.54	9.4	10.38 $\pm$ 1.69	69.2 $\pm$ 11.3
	11.59	6.6		
45°	11.52	8.5		
	11.42	6.5		
90°	11.74	11.7		
	11.68	4.3		
135°	11.49	12.1		
180°	11.63	10.4		
225°	11.68	12.9		
	11.74	9.4		
270°	11.68	9.6		
315°	11.84	8.4		
	11.94	4.8		

^a Only the first readings are counted toward the average.

Table X. dc breakdown values for three layers of
75- μ m-thick Crocker cellulose paper
at 1 MPa (top electrode positive)

Position (degrees)	Temperature (K)	Breakdown Voltage (kV)	Comments
135	11.92	10.2	Cathode-adjacent butt gap.
	11.92	8.5	When voltage was reapplied.
0	12.00	12.1	No butt gap.
180	12.08	12.5	No butt gap.
90	12.08	10.9	Mid-layer butt gap.
	12.06	9.1	When voltage was reapplied.
270	11.98	10.5	Mid-layer butt gap.
225	11.94	17.8	Anode-adjacent butt gap.
	12.00	5.6	When voltage was reapplied.
315	12.06	9.35	Cathode-adjacent butt gap.
	12.06	7.10	When voltage was reapplied.
45	12.02	10.6	Anode-adjacent gap. Broke down while doing PD measurements.

It was clear from the above experiments that polymer films of 75- μm thickness or more could not be broken down within the small specimen chamber that was being used. To determine whether breakdown could be realized by increasing the lateral clearances within the chamber, a larger specimen chamber with an overall diameter of 28.5 cm was constructed. (See Fig. 9.) The electrodes in this chamber were with 6.35-mm-flat and 6.35-mm-edge radius. The inside tangential clearance between the center of the high voltage electrodes and the grounded edge was approximately 5 cm. Three layers of 75- μm -thick Mylar films, with butt gaps at some electrode positions, were assembled. The experimental procedure was the same as described earlier. The high voltage arm was lifted away from the electrodes and the bushing was conditioned to 46 kV. The results of voltage applications at various electrode positions are shown in Table XI. During the experiment, when the voltage was applied to the 90° position, an arcing sound could be heard from the dewar and the bushing performance was found to have deteriorated. The cryostat was warmed to room temperature and the bushing was evacuated to an acceptable level. However, when the cryostat was cooled on a later day, a leak was found to have developed in the specimen chamber, thus preventing its pressurization to sufficient levels for a full series of measurements. A small over-pressure could, however, be maintained and the voltage was applied to all the remaining positions with results shown in Table XI. When the specimen chamber was opened, breakdown punctures were observed at most positions although they were not located directly under the electrodes. (See Fig. 24.) Apparently discharge streamers propagated along the surface of the film, and the field concentration at the tip of the streamer precipitated the breakdown.

TABLE XI. dc Breakdown Voltages for Three 75- μ m Mylar Films in Large Pressurized Sample Holder

Position (degrees)	Pressure (k/Pa)	Temperature (K)	Maximum Voltage (kV)	Comments
0	1000	4.2	35.0	No butt gap. 17.5 kV reapplied before the supply tripped again.
180	1000	12.0	33.0	No butt gap. Voltage could not be reapplied without obtaining significant current.
90	1000	12.0	46.0	Cathode-adjacent gap. Sound heard from dewar. No more than 20 kV could be reapplied. Cathode adjacent gap. Tripped at 16 kV when voltage was reapplied.
270	136	5.84	37.2	Cathode-adjacent gap. Tripped at 16 kV when voltage was reapplied.
315	113	7.25	34.8	Anode-adjacent gap. Tripped at 16 kV when voltage was reapplied.
45	110	8.02	53.7	Mid-gap. Withstood 51.5 kV at the second voltage application and 22.7 kV at the third.
135	108	9.67	30.8	Anode-adjacent gap. Withstood 43.7 kV (higher than the first) and 23.5 kV during subsequent shots.
225	107	11.16	34.0	Mid-gap. Withstood 17.4 kV when voltage was reapplied.

Fig. 24.

c) Partial Discharge Behavior Under dc Conditions:

From the dc measurements reported in Section 2, it was clear that polymers are more susceptible to flashover along the surface in a supercritical helium environment than to breakdown through the film itself. From our experience with ac measurements on polymers in helium (see Section III.B.3) it was evident that partial discharge (PD) can originate in the helium interface and that the growth and spread of these discharges with increase in voltage can precipitate a surface flashover. In order to see whether a similar situation existed under dc stresses, some PD measurements were conducted under dc conditions.

Initially the experiments were conducted on three layers of Crocker cellulose paper with butt gap holes at some of the electrode positions. However, the measurements yielded inconclusive results. The random nature of the discharge pulses, together with their low repetition rate, made them difficult to distinguish from the rather high background noise pulses. The only conclusion that could be drawn from the initial experiment was that the discharge pulses (if they were such) are of small magnitude (~ 100 pC) and their repetition rate is quite low (2-3/min). In any case it was decided to conduct a second experiment with polymer films in an area where background noise problems were less severe.

The second experiment was performed using three layers of 37- μm -thick nylon. There were two each of anode-adjacent, cathode-adjacent, and mid-dielectric butt gaps, and the remaining two positions were free of butt gaps. Prior to the PD measurements the voltage arm was lifted from the electrodes and the bushing was conditioned to about 30 kV. The high voltage arm was then connected to the 90° position (mid-layer gap of

1-mm-diam). At 7.41 kV, discharges of very low repetition rate (one in every 40-45 sec) appeared. The voltage could, however, be raised to 21.5 kV without producing discharge pulses at a higher repetition rate. When the voltage was further raised to about 25 kV, pulses of approximately 30 pC appeared at a repetition rate of around 0.4 sec. Because of possible damage to the discharge detector from flashover or breakdown the voltage was not raised beyond 25 kV. Similar results were obtained at all other electrode positions. In all cases when the voltage was higher than 9 kV, bursts of discharges of about 100 pC appeared when the voltage was raised or lowered.

It appeared from these experiments that partial discharges do not constitute a serious problem in the case of dc operation. Not only are their repetition rates and magnitudes low, but also they appear only at very high stress levels (~ 100 MV/m).

3. ac Measurements

a) Breakdown Strength and PD Characteristics of Polymers

Immersed in Liquid Helium:

Measurements of breakdown strength and PD inception levels of polymeric tapes immersed in slowly boiling liquid helium were carried out to evaluate the materials in terms of these characteristics. Multiple measurements were to be performed with one cooldown of the cryostat, using the experimental arrangement shown in Fig. 6. The electrode was spring-loaded to ensure good contact and could be lowered onto the film at any point along the circumference of a 6.35-cm-diam circle. The hemispherical brass electrode was 1.2-cm-diam. A 12-cm-diam film was placed, without wrinkles, on a 10-cm-diam stainless steel plane

with the help of two plexiglass support rings. Preliminary tests using a 125- μm -thick Mylar sample showed that poor dielectric strength and low dielectric constant of liquid helium (compared to those of the sample) caused severe partial discharge glow all around the electrode. The glow was initiated in the helium filling the rounded wedge-shaped space between the top electrode and the specimen. Raising the voltage caused the partial discharge to spread, precipitating breakdowns at points away from the interelectrode space or flashover along the surface of the film. In order to minimize this problem the top electrode was encapsulated with epoxy (Stycast 2850 FT Blue) to a diameter of 2.5 cm. The bottom surface of the epoxied electrode was carefully polished, exposing an approximately 1-mm-diam circular area of the brass electrode. Although PD glow was still present with this arrangement, reproducible results could be obtained. Breakdown damage to the epoxied electrode was minimized by using a 100-k Ω resistor in series with the voltage supply and by incorporating a Schmitt trigger relay circuit to cut off the power immediately after breakdown. Depending on the thickness of the film, four to six measurements were carried out. The number of measurements taken was determined by the distance required between adjacent positions to prevent flashover to a previously broken-down portion of the sample. Fresh electrodes were used for each sample. PD onset and extinction were measured using a Tektronics 535 oscilloscope set at 50-mV sensitivity. The breakdown voltage divider and chart recorder were calibrated with an electrostatic voltmeter. The voltage was raised manually at the rate of 500 V/sec. The breakdown puncture was sometimes difficult to identify positively with an unaided eye. Each sample, after test,

was therefore subjected to a screening test at room temperature using a pin-hole detector (Hipotronics Model E-W5). This was necessary because in the case of thick films (100 μ m or more) not all of the test positions were punctured. Surface flashover sometimes preceded breakdown. This posed a problem in our attempts to study the effect of thickness on the dielectric strength because surface flashover precluded obtaining breakdowns with thick samples.

The results of the investigations are shown in Fig. 25. The breakdown strength varied from a minimum of 125 MV/m for a 104- μ m-thick nylon (Rylsan Nylon 12B) to a maximum of 228 MV/m for a 51- μ m-thick nylon (Rylsan Nylon 11). PD inception voltage was always below 50% of the breakdown voltage. The thinner samples, as expected, were found to have higher breakdown strengths. Stacking two 37- μ m samples produced a lower strength than a single 74- μ m sample of the same material, presumably due to the presence of an added helium interface.

b) PD Measurements of Multi-layer Polymers in Supercritical Helium:

-i. Introduction:

In order to measure PD characteristics of polymers in pressurized helium, a demountable pressure vessel was developed. The details of this experimental chamber are described in Section II.D.3 and shown in Fig. 7. In order to proof-test this arrangement, PD measurements were conducted at room temperature using three layers of 50- μ m-thick Mylar with SF₆ as the impregnant. The discharge inception levels agreed closely with values calculated using published SF₆ breakdown strength data.⁴⁴

A series of measurements were then made on multi-layer polymeric insulation impregnated by helium over a range of temperatures (4.2 to 12 K) and pressures (up to 1.5 MPa). The results of these experiments are described below.

Fig. 25.

-ii. Polycarbonate Films (Makrofol KG) with Sphere-Plane Electrodes:

Three layers of 61- μ m-thick Makrofol KG polycarbonate films were used as the specimen. There were 1-mm-diam, mid-dielectric butt gap holes at all positions except one. Moreover, alternate butt gaps were connected to the central area where pressurized helium enters the chamber through radial cuts in the film. This was done to see whether there is any impregnation problem in a closed butt gap as opposed to an open one. Also, in order to make sure that cooling does not hinder impregnation, the chamber was kept under helium overpressure while cooling. The electrodes were of sphere-plane geometry (6.35-mm radius hemispherical ends against plane electrodes). In view of the small inter-electrode spacing (183 μ m) the hemispherical electrodes were considered satisfactory although some fringing of the field could be expected in the 1-mm-diam butt gap.

Measurements were first carried out at liquid helium temperature over a range of pressures (207 to 620 kPa). The results, reported elsewhere,¹³ indicated that there was no obvious pressure effect for PD inception levels in liquid helium. This is understandable in view of the fact that the compressibility of liquid helium is small compared to a gas. The change in density when the pressure is increased from 207 to 620 kPa is only about 10%. The change in inception level due to such small changes in density falls within the scatter observed in PD inception level measurements. However, a definite change was noticed in PD inception levels measured on helium in the supercritical region. (See Fig. 26.)

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Fig. 26.

The results show a definite decrease in PD inception levels with increase in temperature at constant pressure while the inception level increases with pressure for a given temperature. The butt gaps with and without the radial slots behaved similarly. The cavity stress E_g is calculated using the relation

$$E_g = \frac{\epsilon_r}{\epsilon_r \cdot g + d - g} V \dots, \quad (2)$$

where V is the PD inception voltage, ϵ_r is the relative permittivity of the film, g is the thickness of a single film (butt gap height), and d is the total thickness of the insulation. The peak voltage across the gap is

$$V_g = \sqrt{2} E_g g \dots, \quad (3)$$

and is plotted against the density-gap product in Fig. 27. The densities were computed using the NBS HEHT4 computer program.⁴²

-iii. Polyethylene Films (Van Leer Valeron)

Sphere plane electrodes — Three layers of 102- μ m-thick Van Leer Valeron, with 1-mm-diam butt gaps in the middle layer at all electrode positions except 0° , were installed in the test cell. Previous experimental procedures were used. Measurements were conducted between 4.2 and about 12 K at constant pressures of 350, 690, 1030, and 1380 kPa. The results are shown in Figs. 28-31. Generally, the discharge magnitude at inception was of the order of 1 pC, although in some cases the discharge exhibited a tendency to grow spontaneously to values in excess of 10 pC. This happened particularly when the voltage was raised slightly above the inception voltage. When such large discharges occurred, it was found that a few small discharges persisted when the voltage was

Fig. 27.

Fig. 28.

Fig. 30.

Fig. 31.

lowered to levels much lower than the original extinction voltage. The large discharge inception and extinction levels were not changed by this behavior. However, when the sample chamber was warmed and pumped out overnight, the small discharges disappeared. Usually the discharge pulses were concentrated between the 0 and 90° and the 180 and 270° portions of the ac cycle at inception. The extinction voltage was found to depend on the magnitude of the discharge and decreased as the magnitude increased. In general the extinction voltages were about 5 to 20% below the inception voltage. The position with no butt gap displayed lower inception voltages than other positions.

The voltages across the gap at inception were calculated using Eq. (2) and are plotted against the pd product to generate the Paschen plot shown in Fig. 32. Also shown in the figure are the low temperature Paschen curve for helium and the 125- μm gap data of Meats²¹ for metallic electrodes at 5.5 K. The Paschen plots agree with Meats's data up to $pd \sim 10^5 \text{ kg}\cdot\mu\text{m}/\text{m}^3$, and then begin to rise quickly. For $pd < 10^5 \text{ kg}\cdot\mu\text{m}/\text{m}^3$,

$$V_g \propto (pd)^{0.33} \quad (4)$$

Flat electrodes — In order to determine whether the above results were influenced by any non-uniformity of the electric field in the gaps, the experiment was repeated using flat electrodes in place of the hemispherical ones. The new set of electrodes were 6.3-mm diam, significantly larger than the 1-mm diam of the voids. The edges of the electrodes were rounded to a radius of 0.8 mm in accordance with ASTM D-149. Three layers of fresh Valeron samples were used. The films were arranged to study the effect of butt gap location on inception voltage.

Fig. 32.

Two positions had no voids. The remaining six positions were divided into three pairs with voids in the top, middle, and bottom layers.

The PD inception voltages are plotted against temperature for various constant pressures in Figs. 33-37. The results agree well with the sphere-plane data under the same pressures as demonstrated by comparison with Figs. 28 and 33. A plot of average inception voltage vs temperature for a pressure of 1.22 MPa is shown in Fig. 38. The plot was made to compare the performance of electrode-adjacent voids with mid-dielectric and void-free positions. The figure shows that the inception voltages for the electrode-adjacent voids are slightly higher than those for the mid-dielectric voids, and the positions with no voids show the lowest inception voltages of all. Over the entire pressure range from 0.3 to 1.5 MPa (see Figs. 33-37), the locations with gaps adjacent to electrodes show a higher inception voltage, and the locations with no holes show the lowest inception voltages. This result will be discussed later. The spread between the highest and lowest values increases as the pressure increases and is about 30% of the average value at all pressures. Averaged values of the inception voltage in the gaps vs temperature are shown in Fig. 39. A clear temperature and pressure effect is evident, although the improvement with increase in pressure is very small beyond 1.2 MPa.

The PD magnitudes were generally a few pC at inception. The electrode-adjacent gaps exhibited a tendency for the partial discharges to grow spontaneously to several thousand pC. The discharges at those locations with no holes were generally very small (1 pC or less) and increased gradually when the voltage increased beyond the inception level.

Fig. 33.

Fig. 34.

Fig. 35.

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Fig. 36.

Fig. 37.

Fig. 38.

Fig. 39.

When the voltage was raised significantly above the initial inception level, the discharge magnitude would suddenly jump to very large values ($\sim 10,000$ pC).

The Paschen plot in Fig. 40 shows all the data points obtained at various pressures and temperatures. The peak voltage across the gap is plotted against the density-gap product. The graph also shows the low temperature Paschen curve for helium as well as the results obtained by Meats²¹ for bare metal electrodes. A detailed discussion of this graph will be made in a later section.

-iv. Polyethylene with Aluminized Gaps:

Three layers of 102- μ m-thick Valeron were assembled with six mid-dielectric gaps. Three were bare gaps and three were aluminized on the top, bottom, and sides in order to enclose the helium in a field-free conducting "pill box," as shown in Fig. 41. The outer films adjacent to the electrodes were also aluminized in order to minimize the effects of residual gaps at the electrodes. PD inception voltages for this arrangement are shown in Figs. 42 and 43 for pressures of 0.6 and 1.5 MPa. It is evident that the inception voltages for the three locations with the "pill box" were much lower than the rest. The highest inception voltages were recorded for the 315° position, which had aluminizing between all the dielectric-dielectric and dielectric-electrode interfaces but no butt gap hole. The PD magnitudes for the bare butt gap holes were always on the order of 1000 pC, whereas for the other locations they were less than 3 pC. Thus the main effect of aluminizing on the butt gap holes was to reduce the discharge magnitude considerably but at the same time to reduce the inception level.

Fig. 40.

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Fig. 41.

SAMPLE: THREE LAYERS OF 100- μ m-THICK VAN LEER

VALERON POLYETHYLENE

FLAT ELECTRODES

0° NO BUTT GAP

45°, 90°, 135° MID-DIELECTRIC BUTT GAP

180°, 225°, 270° ALUMINIZED HOLES COVERED ON TOP
AND BOTTOM BY ALUMINIZED FILM

316° FILMS ALUMINIZED ON BOTH SIDES WITH NO BUTT GAPS

PRESSURE = 0.608 MPa

Fig. 42.

Fig. 43.

-v. Embossed Polycarbonate:

Embossing improves the tapability index (defined as $1/\mu \sqrt{E_t/E_c}$), where μ is the coefficient of friction, E_t is the tensile modulus, and E_c is the compressive modulus) by effectively reducing the compressive modulus and by decreasing the interlayer friction due to reduction in contact area. However, the effect of embossing on the PD performance of a superconducting high voltage cable is not clear. In order to assess this characteristic, three layers of embossed polycarbonate films (Makrofol) were assembled in the test chamber. The basic material was 61- μm thick. With embossing, the total thickness of each film was about 100 μm . Butt gap holes 1- and 2-mm diam were cut in the top, center, or bottom layer at positions illustrated in the key in Fig. 44. (The figure shows the diameter of the hole in mm while the letter shows the position of the butt gap: B for bottom, T for top, M for middle, N for positions without holes.) The figure shows the PD inception voltages vs temperatures at three pressures. The inception voltages are slightly higher than those obtained for Valeron (shown in Fig. 38). The position or size of the hole seemed to have little effect on the inception voltages. Even the positions without butt gap holes behaved much like the positions with holes. The discharge behavior was much more unstable than those behaviors obtained with flat samples. Intermittent discharges appeared as the voltage was raised. At the inception voltages plotted in the figure, continuous discharges appeared and usually grew quickly to several thousand pC. This is not surprising in view of the multitude of inter-connected gaps between layers. When the chamber was opened extensive discharge tracking across the film surfaces was observed.

Fig. 44.

-vi. Nylon:

In order to see the effect of thinner materials on PD performance, measurements were conducted on three layers of 38- μ m-thick nylon. Of the eight electrode positions, two were provided with high voltage electrode-adjacent gaps, two with ground electrode-adjacent gaps, two with mid-dielectric gaps, and two with no gaps. All gaps were 1-mm-diam holes. Measurements were made at 0.3, 0.9, and 1.5 MPa. The results of the measurements are shown in Fig. 45. The discharge behavior, in general, was quite similar to that of polyethylene samples. As in our earlier measurements, the discharge at the non-butt gap positions was consistently low in magnitude, and the inception levels at these positions were no better than those for the butt gap positions. The total stress across the insulation at the time of PD inception was slightly higher for the thinner nylon samples when compared to the polyethylene results under identical conditions. The peak voltage across the cavity was plotted against the density-gap product to generate the Paschen plot shown in Fig. 46. As with the polyethylene and polycarbonate results (Figs. 27 and 40), considerable deviation from the Paschen plot for helium is observed. It can also be seen by comparing the three Paschen plots for the three materials that the curves do not coincide. This point will be discussed in a later section.

-vii. Clean Valeron:

In order to see whether dust or other contaminants have an influence on the PD performance of the films, three layers of 102- μ m-thick Valeron samples were assembled under very clean conditions. The sample chamber and the samples were cleaned in isopropyl alcohol using an ultrasonic

Fig. 45.

Fig. 46.

cleaner, and the entire test chamber was assembled inside a clean glove box. Of the eight positions, two were provided with high voltage electrode-adjacent gaps, two with ground electrode-adjacent gaps, and two with mid-dielectric gaps. The remaining two positions were left without any butt gap holes. The results of the measurements are shown in Figs. 47 through 49. There was no difference from the earlier measurements on Valeron in either the experimental scatter or the discharge behavior. As before, the mid-dielectric gaps showed stable discharges while the electrode-adjacent gaps exhibited a tendency to increase to large values (~ 1000 pC) with slight overvoltage. Positions without butt gap holes produced discharges of very low magnitude (< 1 pC). Comparison with earlier results (Figs. 33, 35, and 37) shows that there is almost a complete agreement for the 0.3 MPa curves, but slightly higher inception voltages are obtained with the clean samples at 0.9 and 1.5 MPa.

-viii. Specimens Under Mechanical Pressure:

One peculiarity that was observed throughout the PD measurements was that the positions without butt gaps showed PD at voltage levels either the same or lower than those for positions with butt gap holes. This suggested that residual gaps are present in the interelectrode region in spite of the flexible foam and the spring arrangement below the ground electrode. (See Fig. 7.) In an attempt to further suppress the discharges at the electrodes the apparatus was altered to compress the samples against the electrodes by clamping the stainless steel against the epoxy disc with the aid of an appropriate Bellville washer. All internal components of the specimen chamber, with the exception of

Fig. 47.

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Fig. 48.

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Fig. 49.

the spring, were unchanged. The pressure was chosen to be about 500-700 kPa since this pressure was reported to be exerted on the inner layers of the insulation in a wrapped dielectric cable. Three layers of 102- μ m-thick Valeron were used as the specimen. As before, there were two each of bottom layer, top layer, and mid-layer butt gap holes, and the remaining two positions were free of butt gaps. The results of PD measurements are shown in Fig. 50. Discharges were present at the non-butt gap positions at the same voltage levels as for the butt gap position. In general there was very little difference in either the discharge behavior or the discharge levels in the compressed samples when compared with earlier measurements.

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Fig. 50.

IV. DISCUSSION

A. Helium Breakdown

1. Electrode Geometry and Polarity Effects

The breakdown strength of saturated liquid helium has been found to be closely correlated with the electrode geometry. With sphere-plane electrodes, the average and minimum values appear to level off at 40 MV/m and 20 MV/m, respectively, for gaps greater than about 2 mm. The voltage vs gap data were best fitted by a function of the form

$$V_b = A d^n,$$

where n is between 0.7 and 0.8.⁶ The fact that no severe falloff of breakdown strength occurs as the gap is increased is encouraging for design of cryogenic electrical equipment. The average values shown in Fig. 11 agree well with those of Gerhold¹⁸ (curve G in Fig. 11), which were taken with similar electrodes. The average values of Meats²¹ or Meyerhoff²² (curve M in Fig. 11) are close to the ORNL minimum values. The measurements of Meats and of Meyerhoff were taken with larger electrodes and more rapidly boiling helium. It appears that the conditions which led to only an occasional low breakdown voltage in the ORNL apparatus occurred more or less continuously in the measurements of Meats and of Meyerhoff.

Electrodes with sharp edges or points led to considerably reduced breakdown voltages, as shown by the ASTM and point-plane results. The ASTM dc and ac breakdown voltages in liquid helium were about one-third of the sphere-plane voltages at equal gaps. Field enhancement factors for a 90° corner with edge radius ρ and an infinite plane at a distance a are plotted in Ref. 27 as a function of $(\rho + a)/\rho$. The peak edge fields

for the negative dc data on Fig. 14 were calculated assuming a 40- μm edge radius, and the resulting fields are shown as squares in Fig. 11 at gaps of 1 and 2 mm. The agreement with the sphere-plane breakdown fields is excellent. For the ASTM electrodes, the calculated enhancement factors were between 3 and 4. It can be concluded that where modest enhancement factors are involved, breakdown occurs at the same maximum surface field as for uniform field geometries.

For the point-plane experiments, the maximum surface breakdown fields at the tip of the point electrode were calculated using a computer program by W. F. Westendorp.²⁸ The results are shown in Fig. 51 for points with 166- μm and 230- μm radii. Good agreement in the surface fields vs gap is found for the two positive points; however, for the negative points, the surface fields for the 230- μm radius point were nearly twice those for the 166- μm point. This may have been due to surface asperities or damage on the smaller point. For the negative polarity results, the surface fields are about a factor of two greater than the sphere-plane values. In this case, higher surface fields are to be expected due to the small area of the point tip and to the inhibition of arc propagation by the rapid fall of the electric field with increasing distance from the point.

However, the surface fields for the positive point are about four times those of the sphere-plane configuration. This polarity effect is the opposite of that normally observed in room temperature gases. Because electrons form large bubble complexes in liquid helium, their mobilities are lower than those of the positive ions, leading to a negative space charge in the gap. It appears that this space charge shields the positive

Fig. 51.

point and inhibits breakdown. In the case of ac voltage the peak breakdown fields were close to the lower negative dc values, which indicates that space charge developing in the positive half-cycle inhibits breakdown until the polarity reverses. A small polarity effect was observed even for the sphere-plane electrode tests, as shown in Fig. 11. Due to the slightly divergent field, the breakdown pits were concentrated in a smaller area on the sphere than on the plane, and it is possible that this led to the lower fields observed for the negative sphere. It can be concluded from these experiments that the condition of the cathode plays a much greater role than that of the anode in breakdown of liquid helium.

2. Statistical Behavior

Considerable scatter is evident in the measurements, particularly for 4.2-K liquid and high density gaseous helium near the critical temperature. Probability plotting techniques indicate that the best fitting statistical model for the results in the liquid is the logarithmic extreme value function, as shown in Figs. 12 and 16. The probability density function for this model is given by Eq. (1). This distribution is strongly skewed toward lower breakdown voltages, and therefore use of the usual normal distribution model in predicting the breakdown probability for a given operation voltage from the mean and standard deviation will lead to an erroneously low estimate.

The question of the physical significance of the log extreme value behavior is of considerable interest. The fact that it is the logarithm of the breakdown voltage which is extreme value distributed suggests that the stress on the liquid produced by the applied voltage V is

proportional to $\ln V$. We use the term "stress" very loosely to signify any process which might lead to breakdown. Hence, if the voltage is squared or cubed, the stress is only doubled or tripled.

Megahed and Tropper²⁹ have found that the frequencies of prebreakdown current pulses with spherical electrodes in transformer oil are exponentially distributed. Thus, at voltage V ,

$$N(q,V) = N_0 e^{-k(V)q}, \quad (5)$$

or

$$\ln \left[N(q,V)/N_0 \right] = -k(V)q. \quad (6)$$

Here $N(q,V)$ is the number of pulses per second of charge q at applied voltage V , and N_0 is the extrapolated frequency of pulses with zero charge. Surprisingly, at a given gap N_0 was independent of the applied voltage, but rose with decreasing gap. The slope $k(V)$ fell very weakly with increasing applied voltage.

If such prebreakdown pulses existed in liquid helium, we might suppose that breakdown could occur when some critical charge pulse q_a recurred with critical frequency N_a .

The existence of a critical charge pulse leading to breakdown has been proposed as a rough criterion for streamer breakdown, as discussed by Devins and Sharbaugh.³⁰ In this case, the critical charge is the space charge in the avalanche head necessary to create a local field comparable to the applied field, so that satellite avalanches originating from electrons produced by electron-ion recombination photons can feed and extend the original ionized path.

Let V_a be the voltage at which q_a recurs at N_a . Then Eq. (6) becomes

$$\ln (N_a/N_o) = -k(V_a)q_a. \quad (7)$$

If the breakdown voltage were related by some power law to N_a/N_o ,

$$V = r(N_a/N_o)^p \quad (8)$$

then

$$1/p \ln (V/r) = -k(V)q_a \quad (9)$$

In this equation, q_a can be interpreted as the critical stress on the dielectric, and is presumed to vary with an extreme value distribution. This would lead to an extreme value distribution in the logarithms of the breakdown voltages, as observed.

Of course, in view of the fact that pre-breakdown pulses have never been observed in liquid helium with uniform field electrodes, all this is pure conjecture. However, if charge multiplication occurs in the liquid by some means (perhaps in bubbles), it is quite possible that q_a is too small for detection. Furthermore, the assumption of proportionality between $\ln V$ and the stress on the dielectric helps to explain the unexpectedly high scatter seen in the liquid breakdown voltages.

In Fig. 12, the standard deviations of the breakdown voltages were close to 20%, but those of the logarithms of the breakdown voltages were only about 5%. Equation (9) leads to an interesting interpretation of this figure. If $\ln V$ is proportional to the critical charge pulse which triggers breakdown, then Fig. 12 indicates that the average critical charge pulse for breakdown increases as the gap increases. Also, the near equality of the slopes of the curves for various gaps shows that at all gaps the critical charge corresponding to 90% survival probability

is about 0.25 "charge units" lower than that corresponding to only 10% survival probability.

The probability plots in Fig. 12 show a peculiar discontinuity near the 90% probability level. Such discontinuities are not unusual³¹ and indicate the existence of two separate breakdown processes. For example, the major portions of the curves for probabilities below 90% might result from breakdown processes in the pure liquid, while the lowest voltages could be a consequence of an occasional contaminant particle entering the gap. Gerhold³² has observed similar steps in the probability plots for oil vapor-contaminated liquid helium.

A moderate increase in field inhomogeneity, such as produced by ASTM electrodes, considerably reduces the percentage scatter, which was only 4 to 6% in the ASTM measurements. The relatively small volume of highly stressed fluid existing at the edges of these electrodes reduces the likelihood that a contaminant particle or density fluctuation will enter this region and precipitate a breakdown at a particularly low voltage. However, with point-plane electrodes the scatter is again over 20% as a result of the more random effects of space charge in the gap.

The scatter is found to decrease with reduction in helium density, as is shown by the plots (see Fig. 17) of breakdown voltage vs temperature for supercritical helium gas. It can be seen that the spread of values at 1.5 MPa is about three times as great as for 0.3 MPa. However, the percentage scatter is about the same at all densities.

3. Contamination Effects

The few experiments which have been performed with deliberate contamination by air particles have shown no particular change in the breakdown strength. However, since the contaminant film was blasted out of the high stress region by the first few breakdowns in the point-plane experiments, it seems unlikely that this contamination film participated farther in the breakdown process. Experiments by the Linde Superconducting Cable Project have indicated that if the air particles are confined, as in a long horizontal coaxial electrode geometry, such contamination can reduce the breakdown voltage considerably.³³ Gerhold^{18,32} has found that oil vapor can have a devastating effect on helium breakdown, and it is probable that some of the day-to-day variations in the ORNL breakdown voltages resulted from contamination by vacuum pump oil. This is probably the case for the experiments with technically pure supercritical helium, in which the sample chamber and fill lines were evacuated at room temperature over the weekend. Since the purity of the "Navy Grade A" helium used in later experiments was only slightly better, the improvement in the breakdown voltages at lower temperatures may have resulted mainly from thorough cleaning of the sample chamber and fill lines before the later experiments.

Theoretical studies by Kok³⁴ to be discussed later imply that a vanishingly small concentration ($\sim 10^{-15}$ at %) of nitrogen or air is sufficient to explain the minimum breakdown voltages observed for liquid helium. When nitrogen or air is deliberately added to liquid helium, it forms rather large particles which quickly settle out, or else it condenses as a film on the electrodes. Kok's ideas suggest

that any tiny ($15 \text{ \AA}$ -radius) contamination particles suspended in the helium would determine the minimum breakdown strength regardless of any additional contamination. This would explain the observed lack of contamination sensitivity in the point-plane experiments.

4. Thermophysical State

The data of Figs. 17, 21, and 22 for the breakdown voltage of supercritical helium vs temperature and pressure show that the breakdown voltage decreases as the density falls. At lower densities, the Townsend electron avalanche process is expected to govern breakdown, and plots of the breakdown voltage vs the product of the density and gap should yield the typical Paschen curve for helium. Such plots are shown in Fig. 52 for the technically pure helium data and in Fig. 53 for the later results for "Navy Grade A" helium. All plots for technically pure helium in Fig. 52 fall considerably below published Paschen curves for helium,³⁵ indicated by the solid line. Furthermore, there is no agreement between the curves taken at various pressures. This is suspected to result from pump oil contamination of the sample chamber and fill lines during the preliminary evacuation of the system over the weekend. Notice that these Paschen plots are very similar to those obtained for partial discharge in polymer films (see Fig. 55). The values (at 0.3 MPa) with "Navy Grade A" helium in Fig. 25 show much better agreement with the published Paschen curve. Considerable scatter appears for densities above 100 kg/m^3 for all gaps studied, and the breakdown voltage appears to be rising rapidly. This is the region around 5.6 K, where the transposed critical point appears for helium at 0.3 MPa. In the density region between 20 and 100 kg/m^3 , the breakdown voltage curve rises above

Fig. 52.

Fig. 53.

ORNL-DWG 76-19809

Fig. 54.

ORNL-DWG 76-19804

Fig. 55.

the Paschen curve, then flattens out and crosses it in the neighborhood of 100 kg/m^3 . For densities below 20 kg/m^3 , Paschen's curve is followed closely. Similar behavior has been observed by Meats²¹ for millimeter gaps.

5. Breakdown Theory

It is probable that no single breakdown mechanism can be advanced to explain the breakdown behavior of helium over the entire density range from dilute gas to compressed liquid. At low densities, up to perhaps 20 to 50 kg/m^3 , breakdown is strongly density-dependent and appears to follow the same Paschen plot as for room temperature helium. This indicates that the classical Townsend electron avalanche process is at work, with the well known result³⁰ that the breakdown voltage is a function only of the product of the density and gap.

As the density increases at constant gap, the breakdown voltages continue to rise and become rather erratic. Between about 60 and 100 kg/m^3 , where a liquid or liquid-like state is forming, a peak seems to appear for pressures in the range of 0.3 MPa . In this region, breakdown may be governed by a complex interplay between the thermal and transport properties of the gas.

The Townsend process treats the gas dielectric as merely a collection of scattering centers between which the electron pursues a free trajectory. However, as the density increases, the possibility of continuous interaction between the electrons and gas must be considered. In this case, the passage of the electron can alter the motion of the gas molecules considerably. Harrison et al.³⁶ have performed electron mobility studies in dense cold helium gas which indicate that at a pressure of 0.3 MPa

the electrons are already confined in "bubbles" for densities above about 15 kg/m^3 . However, they are not bound very tightly. The electron injection energy for helium is about 1 eV at the liquid density of 125 kg/m^3 , and is closely proportional to the density.³⁶ Thus, at 15 kg/m^3 the electrons are in potential wells only about 0.1 eV deep, and probably spend part of their time free and part trapped in bubbles. This might result in a reduction in the Townsend primary ionization coefficient, leading to breakdown voltages above the Paschen curve.

It is instructive to attempt to construct a "figure of merit" for helium breakdown as a function of such variables as the heat capacity C_p , viscosity η , and thermal conductivity λ , as well as the density ρ . The thermal diffusivity α is defined by

$$\alpha = \frac{\lambda}{\rho} C_p . \quad (10)$$

The reciprocal $1/\alpha$ is a measure of the time required for a temperature gradient in the gas to form as a result of a local energy input. The solid line in Fig. 20 gives the ratio $F = A(\eta/\alpha)$, where the constant was chosen to fit the 102- μm data at an intermediate temperature of 7 K. This is the simplest combination of the above properties which gives a reasonable fit to the data. This ratio is also proportional to P_r/ρ , where P_r is the Prandtl number. Note that F peaks at nearly the same temperature as the experimental points, although the peak in F is somewhat stronger. The qualitative agreement suggests that breakdown at intermediate densities is governed by the hydrodynamics of the motion of charged particles through the gas as determined by η , combined with the peak in $1/\alpha$ which makes it difficult to form a hot area anywhere in the

fluid. Contamination also plays a role, as shown by the improvement in breakdown strength noted when the gas and sample cell were cleaned more thoroughly.

As the temperature falls below the transposed critical point (i.e., the point where $(dP/d\rho)_T$ is a minimum and the heat capacity at constant pressure is a maximum, corresponding to the gas-liquid transition for the subcritical vapor), the breakdown voltage falls somewhat to a value which is not strongly pressure or temperature dependent. In this regime, it has been well established that electrons are trapped in bubbles in the liquid with radii of about $12 \text{ \AA}$. The electrons are confined in potential wells about 1 eV deep.³⁷ Positive helium ions, on the other hand, are bound by electrostrictive forces³⁸ in spheres of solid helium of about $7 \text{ \AA}$ radius. Consequently, the ion mobilities are rather low — about $0.02 \text{ cm}^2/\text{V-sec}$ for electrons and $0.08 \text{ cm}^2/\text{V-sec}$ for positive ions. It can easily be shown that at observed helium breakdown fields such large, slow ion complexes can attain only a small fraction of the required ionization energy for helium (24.5 eV). Furthermore, it is unlikely that the electrons can escape the bubbles under experimentally observed breakdown fields. Lau and Dahm³⁹ have calculated that the threshold field for tunneling of electrons from the bubbles is about 3 MV/cm, as opposed to observed breakdown fields of only 0.1 to 0.4 MV/cm. Unfortunately, the details of this calculation are not published. Lau and Dahm have not been successful in observing free electrons even in MV/cm fields.⁴⁰ It is therefore unlikely that breakdown in liquid helium occurs as a result of avalanches generated by free electrons moving through the liquid.

More promising explanations for breakdown in liquid helium are suggested by the particle theory of Kok³⁴ and by the bubble theory of Krasucki.⁴¹ In Kok's theory, small contaminant particles whose dielectric constant is higher than that of the fluid are drawn by polarization forces into regions of enhanced field near asperities on the electrodes. As the particles accumulate, they form a bridge across the gap, leading to breakdown. The criterion for particle accumulation is that the dielectric forces pulling particles into the field-enhanced region be greater than the diffusion forces away from the region.

The threshold field E_o is given by

$$(g^2 - 1) r^3 E_o^2 = 2 k_B T / \epsilon_o \quad (11)$$

where g is the field enhancement factor at the asperity and r is the particle radius. Typically, g is of the order of three. The above relation then gives

$$r^3 E_o^2 = \frac{k_B T}{4\epsilon_o} \quad (12)$$

We see that the breakdown strength is proportional to $r^{-3/2}$. Furthermore, the breakdown strength increases as T increases, which may also play a role in the peak in the breakdown field noted for the pressurized helium measurements in Fig. 20.

If $E_o = 20$ MV/m (the minimum value shown in Fig. 11) and $T = 4.2$ K, Eq. (12) gives

$$r = \left[\frac{(1.38 \times 10^{-23} \text{ J/K}) (4.2 \text{ K})}{4(8.85 \times 10^{-12} \text{ F/m}) (4 \times 10^{14} \text{ V}^2/\text{m}^2)} \right]^{1/3} = 16 \text{ \AA} .$$

This would correspond to a particle of solid nitrogen containing about 700 atoms. At least 3×10^5 such particles would be required to bridge

a gap of 1 mm. If the cryostat contained about 1ℓ of liquid surrounding the electrodes, the equivalent impurity concentration would then be about

$$\frac{(700 \text{ atoms/particle}) (3 \times 10^5 \text{ particles/ℓ})}{(125 \text{ g/ℓ}) (6 \times 10^{23} \text{ atoms/4 g})} \sim 10^{-17} = 10^{-15} \text{ at. \%}.$$

This is far below standard helium impurity concentrations of roughly 20 ppm. If Kok's theory is correct, any attempt to improve the minimum breakdown strength of liquid helium by purification is unlikely to succeed.

On the other hand, Krasucki's theory⁴¹ supposes that breakdown is triggered by bubbles in the liquid generated at metal particles or asperities of radius R on the electrodes. A bubble develops when the pressure at the particle surface equals zero. This occurs when the outward electrostatic pressure $\frac{1}{2}\epsilon E^2$ equals the sum of the hydrostatic pressure P_∞ and the surface tension $2\sigma/R$. The bubble then elongates until it either bridges the gap or reaches a length which corresponds to the Paschen curve minimum at the density of the vapor inside. Breakdown then takes place in the vapor by avalanche or streamer mechanisms.

For a spherical particle on a plane surface, it can be shown that the field is enhanced by a factor of 4.2 over the average field in the gap. Hence, Krasucki's criterion becomes

$$\frac{1}{2}\epsilon (4.2 E_b)^2 = P_\infty + \frac{2\sigma}{R} \quad (13)$$

or

$$E_b = \frac{0.377}{\sqrt{\epsilon}} \left[P_\infty + \frac{2\sigma}{R} \right]^{1/2} \text{ V/m.} \quad (14)$$

P_{∞} and $2\sigma/R$ are expressed in nt/m^2 , and $\epsilon = 8.85 \times 10^{-12}$ farad/m.

Liquid helium contains a ready-made source of bubble nucleation points in the bubbles surrounding electrons. Furthermore, it seems reasonable to treat an electron in a bubble as a small metallic particle. For liquid helium, the surface tension is given by⁴²

$$\sigma = 5.31 \times 10^{-4} \text{ nt/m} \left[1 - T/T_c \right]. \quad (15)$$

Letting $T = 4.2 \text{ K}$, $P = 10^5 \text{ nt/m}^2$, and using the equilibrium bubble radius of $R = 12.4 \text{ \AA}$ in (14), we have $E_b = 48.29 \text{ MV/m}$, which is close to the average breakdown field in millimeter gaps shown in Fig. 11. This suggests that Krasucki's theory is the dominant breakdown mechanism in liquid helium. However, neither the Kok nor the Krasucki theory treats such questions as the variation of breakdown field with gap, and further work is needed.

B. Polymers

1. Impulse Breakdown Strength

The impulse breakdown strengths of polymers in liquid helium are better than 250 kV/mm , almost a factor of two higher than ac breakdown strength and nearly the same as the dc strength. Because of this the dc superconducting cable will be more susceptible to damage by transients if full advantage of the dc strength is taken in the design, and protection measures are needed to avoid excessive dielectric buildup in the cable. A tendency for flashover is exhibited for thicker films ($\geq 100\mu\text{m}$) which break down at higher voltages. Severe ripping of the film was noticed with some films, particularly the $100\text{-}\mu\text{m}$ -thick Rowland polysulfone. This tape was found to have the lowest tensile strength at 4.2 K in studies at BNL.⁴³

2. dc Measurements

a) dc Breakdown Strength:

The dc breakdown strength under supercritical helium conditions could be measured only for cellulose paper. Examination of the breakdown site and the fact that the site could withstand appreciable reapplication of voltage even after breakdown suggest that the breakdown in the cellulose paper is through the helium filled pores. The breakdown strength for two layers of 75- μ m-thick cellulose paper (Crocker) impregnated by helium at 12 K and 1.0 MPa was measured to be 70 MV/m. This value is of the same order as the dielectric strength of helium under similar conditions. The polymers in helium, on the other hand, have a higher dielectric strength (>250 MV/m). Indications are that most polymeric tapes should have more than adequate dielectric strength for use in a dc cable. Also, partial discharge does not seem to be a serious problem in the case of dc if the ripple level is kept low. However, this may be troublesome during polarity reversal. It appears as though the choice of a particular polymer for use in a lapped dc superconducting cable should be made based primarily on its mechanical characteristics. Further study should be undertaken to determine the surface tracking and flashover characteristics of polymers in a helium environment.

b) Partial Discharge Under dc Conditions:

Measurements on polymers impregnated by supercritical helium under dc stresses show that discharges appear only at high stress levels (>100 MV/m). Moreover, the discharge magnitudes are generally low (100 pC or less) and their repetition rate is very small (on the order of seconds).

These characteristics continue until breakdown, suggesting that unlike the case of ac, the dc breakdown is not initiated by partial discharges. The few measurements that were made indicate that partial discharges do not constitute a problem in the case of steady dc operation. There was some indication that partial discharges could be significant during polarity reversals, as bursts of discharges were present whenever the dc voltage was varied. This suggests that the effect of ripple on partial discharges should also be evaluated.

3. ac Measurements

a) Breakdown Strength and Partial Discharge Measurements on Polymers Immersed in Liquid Helium:

Results described in Section III.B.3.a show that the average breakdown strengths for single layers of polymers range from 130 MV/m to 220 MV/m. In general the breakdown strength falls with increasing film thickness and breakdown is initiated by partial discharges in the helium interface. Appreciable PD occurs at less than 50% of the breakdown levels in polymers immersed in liquid helium. It may be pointed out that the PD measurements described in Section III.B.3.a were made using an oscilloscope in the 50-mV sensitivity range. The inception levels should thus be higher than those which would have been measured with the more sensitive Biddle detector. The investigations also revealed that for thicker films ($>100\text{ }\mu\text{m}$) the effect of PD inception in the helium interface is to initiate a surface flashover. In the presence of PD, the electric field strength for surface flashover is many orders of magnitude lower than the bulk breakdown strength of the polymers.¹⁰

b) PD Measurements on Polymers Impregnated with Supercritical Helium:

Results of PD measurements on a variety of polymers with simulated butt gap spaces, impregnated with helium in the temperature range of 4.2-12 K and pressures up to 1.5 MPa, are described in Section III.B.3.b. These results show that the PD inception voltage at a given pressure falls with an increase in helium temperature. For a given temperature the inception voltage increases with an increase in pressure (Fig. 26). The nature of the variation of the inception voltage with temperature and pressure is very similar to the variation in helium density. In other words, the fall of discharge inception voltage with temperature is steeper at lower pressures. (See Fig. 39.) The extinction voltages are generally about 90% of the inception values if the PD magnitude is less than about 5000 pC. Higher discharges resulted in lower extinction voltages. When the butt gap holes are located adjacent to the electrodes, slightly higher inception voltages are found when compared to the mid-dielectric values. Very little difference was observed when the sphere-plane electrodes were replaced by flat electrodes, indicating that the electrode edges have little effect on PD. A scatter of about 30% was observed in the inception level measurements. Increasing the pressure beyond 1.2 MPa produced only a marginal improvement. This fact is consistent with the nature of density variation of helium with pressure. For example, doubling the pressure of 8-K helium from 0.6 to 1.2 MPa almost doubles the density, but a further doubling in pressure produces only a 30% change in density.

The behavior of the discharges was different at various butt gap positions. The mid-dielectric void produced, relatively, the most stable kind of discharges. But even in this case, magnitudes of only a few pC are observed frequently, suggesting that partial breakdown along the dielectric walls of the cavity might be taking place. The discharge magnitudes at the electrode-adjacent gaps had a tendency to increase to large values ($>10,000$ pC) with slight overvoltage. This could be due to the lateral spread of the discharges.

Consistently small discharges (never more than 5 pC) were observed at positions where no butt gap holes were present. Moreover, the discharge inception levels at these positions were often lower than those at butt gap positions. This result, although puzzling at first sight, can be explained in terms of small residual gaps in the interelectrode space. Figure 54 shows schematically the situation where a small residual gap exists in series with the main gap. The thickness of the residual gap (Δg) is small compared to the thickness of the film (g) and the interelectrode distance (d). According to Maxwell's equation, the displacement vector D has the same value inside the dielectric as in the gap and since $D = \epsilon E$, the following equations are valid:

$$\epsilon_o \frac{\Delta V}{\Delta g} = \epsilon_r \epsilon_o \frac{V - V_g}{d - g} = \epsilon_o \frac{V_g}{g}, \quad (16)$$

where V is the total voltage across the electrodes, V_g is the voltage across the gap g , and ΔV is the voltage across the residual gap. It follows that the field in the residual gap is always the same as that in the main gap. Since the dielectric strength of very small gaps ($\sim \mu\text{m}$) is generally higher than that of larger gaps, it can be concluded

that whenever a residual gap exists in series with the main gap, the main gap will discharge first. Also, from the last two expressions of Eq. (16), the field across the main gap can be expressed as

$$E_g = \frac{\epsilon_r V}{\epsilon_r g + d - g} \quad (17)$$

when, as in our case, $g = d/3$,

$$E_g = \left(\frac{3}{\epsilon_r + 2} \right) \epsilon_r \frac{V}{d} \quad (18)$$

Typically for polymers $\epsilon_r \approx 2.5$.

$$E_g = 1.67 \frac{V}{d} \quad (19)$$

If we now assume that the main gap is absent (i.e., $g = 0$) and only a residual gap is present, the field in the residual gap is

$$\begin{aligned} E_{\Delta g} &= \epsilon_r \frac{V}{d} \\ &= 2.5 \frac{V}{d} \end{aligned} \quad (20)$$

Thus when a residual gap is the only gap present, the electric field in that gap is enhanced by a factor equal to the relative dielectric constant. This increased enhancement can reduce the discharge inception level, as observed. It may be noted that part of the effect due to increased field enhancement will be offset by the increased dielectric strength of very thin films of helium.

Figures 27, 40, and 45 show the Paschen plots of the results obtained for polycarbonate, polyethylène, and nylon. The peak voltage across the cavity in these figures is calculated under the assumption that the partial discharge is the result of breakdown of the cavity. To bring out certain features peculiar to the situation, the mean curves

for the three materials are replotted in Fig. 55. The figure also shows the result of Meats²¹ and an actual Paschen plot for helium obtained from Dakin.³⁵ Curves 2, 3, and 4 include data obtained at various pressures and temperatures. It can be seen that all the curves deviate from Paschen's law in the region of interest to the superconducting cable studies. This could be due to the presence of the dielectric walls around the helium gap, as in general a surface flashover is favored over breakdown in such a geometry. It may be interesting to note that each of the curves tends to increase more rapidly towards the end of its range. Closer scrutiny will reveal that the region of rising dielectric strength coincides with the region where gas-to-liquid transformation takes place (i.e., around 5.2 K). This also explains why the lower gap curves intersect the higher ones at different pd values (the liquid density is only slightly affected by pressure).

Comparison of Fig. 55 with our results in Figs. 52 and 53 for helium between metallic electrodes shows that the gap voltages are closest to the values for technically pure helium. This suggests that contamination was also a factor in the partial discharge experiments. Two alternatives are possible:

a) Both the polymer samples and the technically pure helium samples were contaminated by pump oil vapor from the fill line. Similar cleanliness conditions existed for both sets of experiments.

b) Contaminants evaporating from the polymer films themselves contaminated the sample chamber and fill lines.

In view of the fact that the partial discharge experiments with the carefully cleaned polymer sample showed no improvement in the inception voltages, it seems that the second alternative is more likely.

Two experiments were conducted to see whether the dielectric performance of the tapes could be improved. The first consisted of shielding the helium within the butt gap by metallizing all around it. (See Section III.B.3.b.iv.) The results, however, showed that although considerable decrease in the discharge magnitude could be realized, the PD inception levels with this arrangement were appreciably lower than those for non-metallized gaps. This behavior can be understood if one takes into account the reduction in effective insulation thickness due to the presence of the metallized "pill box" and the field enhancement that must occur at the sharp corners of the "pill box." Our second experiment was conducted on embossed polycarbonate films because they have a higher tapability index. (See Section III.B.3.b.v.) Again the results showed that such films are unsatisfactory in their PD performance. The discharge magnitudes were considerably higher and exhibited a tendency to grow to large values at all positions. This should be a direct consequence of the multitude of interconnected gaps present in an embossed film.

An attempt was made to eliminate the PD occurring at the non-butt gap positions by compressing the samples against the electrodes and hence eliminating the residual gaps (Section III.B.3.b.viii). Pressures comparable to those exerted on the inner layers of conventional lapped cables (500-700 kPa) were used. However, no significant difference from earlier measurements was observed.

Measurements conducted on very clean samples (Section III.B.3.b.vii) also produced no appreciable difference from the previous results.

The results of PD measurements on various polymers are replotted (ignoring the scatter in observations) in terms of pressure, keeping the temperature constant (Figs. 56-58). All the curves are mutually consistent. It can be seen that the total stress across the insulation at the time of PD inception does not exceed 30 MV/m at 9 K and 1.5 MPa. The PD inception stress (design value) for the BNL cable, under approximately the same helium conditions, is 20 MV/m.

Considering that our measurements are done on three layers of films using small area electrodes, there is some reason for concern about whether a PD inception stress of 20 MV/m can be attained on a long cable. To what extent the area and volume effects influence the PD inception level is yet to be seen. Comparison of the figures also shows that there is only a small advantage (forgetting the mechanical disadvantage) in using thinner tapes. The inception stresses are only marginally higher and that too at the low end of temperature and pressure. Another important result that is obvious from the figures is that the advantage gained in refrigeration efficiency when the operating point is shifted from 6 to 9 K is considerably offset by the disadvantage of operating at lower inception stress levels. It may also be noted that increasing the operating pressure beyond 1.2 MPa has very little advantage.

Fig. 56.

Fig. 57.

Fig. 58.

V. CONCLUSIONS

The investigations indicate that helium is a well-behaved dielectric, offering minimum breakdown strengths in the range of 20 MV/m for the liquid state in millimeter gaps, and up to 60 MV/m for supercritical gas at 12 K and 1.5 MPa in gaps of the order of 100 μm . Introduction of field inhomogeneities leads to reduction of the breakdown voltage by roughly the field enhancement factor involved. For strongly inhomogeneous fields, reduction by the full enhancement factor does not occur due to the action of space charge and the small electrode area and volume of fluid under stress. The most difficult variable to control is contamination, which seems to have unpredictable effects.

Analysis of the minimum liquid breakdown fields in terms of Kok's particle theory shows that the observed breakdown fields are consistent with an impurity concentration of only one atom for every 10^{17} helium atoms. This is due to the fact that the diffusive forces which act to break up local impurity aggregations are very small at helium temperatures. The average liquid breakdown values are consistent with Krasucki's bubble theory, if it is assumed that breakdowns are nucleated by electron bubbles. For low densities the Townsend avalanche process seems to dominate.

Breakdown strength of polymers in liquid helium, under standard lightning impulse conditions, is better than 250 MV/m. The dc breakdown strength is about the same as the impulse value and the ac strength approximately half.

The dc breakdown strength of cellulose paper impregnated by 12 K helium at 1 MPa is about 70 MV/m. The breakdown is believed to be through the helium-filled pores and hence the dielectric strength of

of helium-impregnated paper is comparable to that of helium under the same conditions. Polymers, on the other hand, exhibit much higher breakdown strength (>250 MV/m). They are, however, susceptible to surface flashover and tracking.

Partial discharges in helium-impregnated polymers, under dc stress, are of low repetition rate (few seconds) and of small magnitude (<100 pC), almost up to the breakdown value.

These results indicate that the initial selection of an appropriate polymeric material, as dielectric for a dc superconducting cable, can best be done on the basis of mechanical (tapability) considerations rather than on the basis of breakdown strength. The dc breakdown strength of most polymers should be more than adequate.

Under ac conditions, the PD inception in the butt gaps of a helium-impregnated, lapped cable will decide the operating stress of the cable. PD occurs well below the breakdown level.

The PD inception stress depends primarily on the helium density. There is very little difference among the various materials investigated. The only material-dependent property that affects the PD inception stress is the relative permittivity.

The PD inception stress is decided by the breakdown strength of the helium-filled butt gap. The breakdown strength of a cylindrical helium-filled cavity inside the dielectric falls below the Paschen's plot for helium between metal electrodes, in the pressure and temperature region of interest for a superconducting cable.

The discharge magnitudes in three-layer samples, with 1-mm-diam holes to simulate the butt gap, depend on the position of the butt gap. Electrode-adjacent gaps exhibit a tendency to increase to large values ($>10,000$ pC) while more stable discharges of lower magnitudes can be realized in mid-layer gaps. Small discharges (<5 pC) are present even when no artificial gap is provided, and the inception levels for these positions are generally lower than those for the butt gap positions. These small discharges are believed to be due to the presence of small residual gaps and should not be a problem when larger gaps are present.

PD performance could not be improved by embossing the films or by shielding the butt gaps by metallized walls.

Even on three-layer samples, it was not possible to obtain PD inception stresses greater than 30 MV/m unless the operating temperature was reduced below 9 K or the operating pressure was increased considerably above 1.5 MPa. Of the two, increasing the pressure is less effective as the improvement is only marginal beyond 1.2 MPa.

Low level discharges initiated while a cable is cold may persist for very long periods of time and thus long-term aging should be investigated at the earliest practicable time.

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Appendix A

ac LOSSES IN SUPERCONDUCTORS

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APPENDIX A: ac LOSSES IN SUPERCONDUCTORS

A. Introduction

The goal of this research has been to understand the relative importance of material parameters which control ac losses in high T_c superconductors such as Nb_3Sn . This work has been in support of the Power Transmission Project at Brookhaven National Laboratory. The design of properly optimized conductors for superconducting ac transmission requires an understanding of loss mechanisms involved. That is, the essential material parameters need to be defined in order to economically fabricate a conductor with minimum ac losses.

During FY 1975 ac loss measurements were made on Nb_3Sn samples prepared by two variations of the solid state diffusion method,¹⁻³ and with a wide range of reaction conditions. Frequency, as well as field amplitude, was used as a parameter in the loss measurements in an attempt to more clearly define the mechanisms responsible.

The total loss per surface area per cycle was found to have the form $W_T(h_o, \nu) = W_o(h_o) + \nu F(h_o)$ where $W_o(h_o)$ is identified as a hysteretic term (i.e., frequency independent), ν is the frequency and h_o the ac field amplitude. Hysteretic losses (W_o) in the Nb_3Sn specimens were strongly dependent on the degree of surface roughness on a scale of $\sim 1 \mu$, while the non-hysteretic losses showed only a mild dependence on the topology of the surface. This report will detail the sample preparation and characterization, the ac measurements, the results of these measurements and their interpretation. A discussion of the extraction of eddy current losses in the pickup coil is presented in Appendix AA and an analysis of the ac experimental method is given in Appendix AB.

B. Sample Preparation and Characterization

Nb_3Sn samples were prepared in the form of layers on 3 mm diam Nb rod ~ 30 mm in length. As detailed in ORNL-TM-4824 (ORNL Cryogenic Power Transmission Technology Quarterly Report, July 1-Sept. 30, 1974.), 48 different samples were prepared by the solid-state diffusion ("bronze") technique with bronze compositions of 8 wt % Sn, 11 wt % Sn, and 13 wt % Sn reacted at temperatures from 700°C to 850°C for times up to 336 hr. In addition, several samples were prepared by copper plating a Nb rod, coating with tin at 350°C , and subsequently reacting to form Nb_3Sn . After final reaction heat treatment the bronze layer was removed by a HNO_3 solution in the case of the co-deformed samples and by boiling HCl for the tin-coated samples.

The superconducting-normal transition temperatures, T_c , of the samples were measured by an inductive technique which also served as a check on the continuity of the Nb_3Sn layer. There was a general trend for higher onset temperatures and sharper transitions in samples reacted for longer times at higher temperatures. The dc magnetization measurements and estimates of J_c by two ac susceptibility techniques^{4,5} were also made on selected samples. All the samples showed nearly complete flux exclusion up to fields of ~ 2 kOe to the resolution of dc magnetization. The flux penetration measurements gave estimates for J_c of $\sim 2 \times 10^7$ A/cm² in the field range ~ 1000 Oe.

Scanning electron microscopy was used to characterize the sample surfaces. The surface geometry will be discussed along with the results of the ac loss measurements.

C. ac Loss Measurements

Since the ac loss measurements are central to this work, the technique will be discussed in some detail here and in the Appendices.

The measurement of ac losses was done electronically in a manner similar to that developed by Sekula⁶ in which a tuned lock-in amplifier is used to examine that component of the voltage, induced in a pick-up coil wound around the sample, in phase with and at the same frequency as the applied ac magnetic field. In our method, however, no steady dc magnetic field is present and the ac field oscillates over the range $-h_0 \leq h \leq +h_0$ while at the same time h_0 itself is slowly varied. The mean power loss per unit cylindrical surface area is given by:

$$\bar{p} = \frac{1}{2\pi R\tau} \int_0^\tau h \frac{d\phi_s}{dt} dt \quad (1)$$

where R is the radius of the sample, h is the applied ac magnetic field of period τ and ϕ_s is the flux contained within the sample. For sinusoidal $h(t)$, this can be rewritten as an energy loss per unit surface area per cycle in the following way,

$$W = 0.127 \frac{\langle h \rangle \langle v \rangle}{NRv} \quad (2)$$

where $\langle h \rangle$ is the rms field in Oersteds, $\langle v \rangle$ is the rms voltage as registered by the lock-in amplifier in volts, N is the number of turns in the pick-up coil, v is the frequency of the applied magnetic field in Hertz and R is the specimen radius in cm. Using these units for the measurable quantities yields a W that is in units of Joule $\text{cm}^{-2} \text{ cycle}^{-1}$.

The output of the lock-in amplifier drives the Y-axis of an X-Y recorder while the X-axis is driven by a voltage proportional to the rms current flowing in the field producing solenoid. In this manner, a sweep of h at a fixed frequency produces a response $v(h)$ and only the product of these two quantities is needed via Eq. (2) to determine the surface loss per cycle.

The solenoid used to produce the ac field is a copper coil wound from Litz wire and is situated directly in the helium bath. Copper windings were chosen after initial work with superconducting wires, both single and multifilament, indicated complications due to intrinsic losses in these materials. Test runs on dummy micarta samples revealed spurious losses which were eliminated by removing all extraneous metal (e.g., solder joints) from the vicinity of the sample and pick-up coil.

Compensation of the unwanted signal due to changes of flux between the pick-up coil and the sample was achieved by means of a coil of a few turns located in the fringing field of the copper solenoid and connected in opposition to the sample coil. Complete cancellation of total signal was achieved in fields typically of a few Oersted by mechanically adjusting the location of the sample pick-up and compensating coils within the bore of the solenoids. This condition was checked after each field sweep to insure that compensation was maintained throughout the data gathering period. This method of compensation used in conjunction with a lock-in amplifier having an input impedance of $\sim 10^7 \Omega$ avoided the possibility of unwanted phase shifts being introduced electronically while at the same time maintaining the sample and its pick-up coil

within the uniform field region of the solenoid. The reference phase of the lock-in was established by nulling the signal induced in the compensator coil (i.e., set to examine any signal in phase with the applied ac magnetic field). This procedure was repeated before any field sweep was taken following a change of operating frequency.

The sensitivity of this method is ideally limited by the ability of the lock-in amplifier to extract the signal of interest from the attendant noise. It should be noted, however, the $\langle v \rangle$ may contain contributions from sources external to the superconductor and, therefore, an uncritical use of Eq. (2) may give rise to an erroneous value for the loss per area per cycle, W . Our apparatus was able to reliably detect losses of the order 10^{-9} Joule cm^{-2} cycle $^{-1}$.

The ac loss measurements were made on a number of selected Nb_3Sn samples. However, our detailed analysis concentrated on two extreme cases--a "low" and a "high" loss sample. The other samples had intermediate loss values and exhibited no anomalous behavior. The "low" loss sample was prepared by co-deformation of Nb in a Cu-10 wt % Sn alloy and reaction for 42 hours at 800°C. The "high" loss sample was prepared by the tin coating process described above with a reaction heat treatment of 48 hours at 800°C. The similar final reaction treatment produced similar onset T_c values for the "low" and "high" loss sample of 17.83K and 17.78K, respectively, although the "high" loss sample exhibited a broader transition width ($\sim 2.5\text{K}$) compared to the "low" loss sample ($\sim 0.3\text{K}$).

To help validate the measuring technique and elucidate the loss mechanisms, ac losses were also measured on a Mo-25 at % Re alloy sample and on a Nb single crystal sample.

D. Results

First, results on ac losses in Mo-25 at % Re are presented to give evidence for the validity of the measuring technique and illustrate a material in which the ac losses are overwhelmingly hysteretic in nature. Then a discussion of the eddy losses from the copper pick-up coil is given with the method used to extract it from the losses in the superconductor. Finally, the loss data on the Nb_3Sn and Nb samples are presented.

1. Mo-25 Re

Flux enters the bulk of Mo-25 Re at a relatively low field ($H_{C1} \approx 450$ Oe) and the losses above this value appear to be purely hysteretic in nature. The criterion used throughout this paper is to assume that any loss per cycle which is independent of frequency is, in fact, hysteretic. At high field amplitudes such as, for example, 1456 Oe, the induced signal $\langle v \rangle$ varies linearly with frequency yielding $\langle v \rangle = 5.22 \times 10^{-4} v$. This, in turn, produces a W_0 of 1.68×10^{-3} Joule cm^{-2} cycle $^{-1}$ constant over the frequency range examined (40 to 100 Hz). While there was no obvious dependence of W on frequency at any field levels, it should be noted that at lower field amplitudes there is some scatter in the results due to variations in the previous magnetic history of the sample. The losses from hysteresis loops obtained by dc magnetization and the ac losses are in good agreement over much of

the mixed state region examined. Fig. 1 is a B versus H (dc) trace in which the external field was varied between ± 756 Oe. The total loop was symmetric and so for simplicity only half of it is shown in the figure. Fig. 2 shows the surface loss per cycle determined as a function of peak magnetic field for both ac and dc conditions. The dc data shown for four field values are in agreement at the lower two values where the induced voltage waveform taken directly from the pick-up coil can be shown to qualitatively yield B versus H responses as shown in Fig. 1. For peak fields in excess of 1200 Oe, the losses determined via the ac technique increase linearly with the value of the peak field. This is revealed as a saturation of the induced voltage signal with field at a fixed frequency. It is interesting to note that this is the behavior predicted for such losses by the Bean model when $H_{\text{peak}} > H^*$ (the so-called filling field).

The ac losses in Mo-25 Re at low fields depend critically on the magnetic history of the sample. Having cycled the Mo-25 Re well into the mixed state ($h_0 \sim 1400$ Oe) and subsequently measured the losses at low fields, one finds that the loss at a given field value can be as much as a factor of twenty greater than in the virgin sample.⁷ Fig. 3 shows the normalized difference, $(W_2 - W_1)/W_2$ referred to as the loss enhancement factor, as a function of peak ac field, W_1 being the loss in the virgin sample and W_2 being the loss at the same field and frequency but after a field excursion well into the mixed state. A rapid drop in this quantity begins at about 600 Oe, that field value at which the dc magnetization indicates $dM/dH \approx 0$ (not at H_{C1}). This would seem to

Figure 1

Figure 2

Figure 3

indicate that in bulk hysteretic materials one must achieve a field amplitude at which a certain threshold amount of flux penetrates the sample and not simply H_{fp} , the field of first penetration, before the losses become independent of previous magnetic history.

2. Eddy Losses in Copper Pick-Up Coil

The loss response of the high-loss Nb_3Sn sample was examined in detail with two pick-up coils wound from copper wire of different diameter in separate experiments. By so doing we were able to extract the eddy-loss contribution of the pick-up coil from the total measured loss.

The rms voltage signals for the two cases can be represented as follows:

$$\langle \epsilon_1 \rangle = S_1 + A_1$$

$$\text{and } \langle \epsilon_2 \rangle = S_2 + A_2$$

where S_1 and S_2 are the voltage signals from the superconductor while A_1 and A_2 are those arising from within the pick-up coils themselves.

$\langle \epsilon_2 \rangle$ can be rewritten, normalizing it with respect to $\langle \epsilon_1 \rangle$ as follows:

$$\langle \epsilon_2 \rangle = \frac{N_2}{N_1} S_1 + \frac{N_2}{N_1} \left(\frac{a_2}{a_1} \right)^3 A_1$$

where $N_{1,2}$ refers to the number of turns on the respective coils and $a_{1,2}$ to the respective radii of the wires used. Thus:

$$S_1 = \frac{\left(\frac{a_2}{a_1} \right)^3 \langle \epsilon_1 \rangle - \frac{N_1}{N_2} \langle \epsilon_2 \rangle}{\left(\frac{a_2}{a_1} \right)^3 - 1} \quad \text{and} \quad A_1 = \frac{\frac{N_1}{N_2} \langle \epsilon_2 \rangle - \langle \epsilon_1 \rangle}{\left(\frac{a_2}{a_1} \right)^3 - 1}$$

In our cases,

$N_1 = 325$, $N_2 = 151$, $a_1 = 2.5 \times 10^{-3}$ cm and $a_2 = 6.4 \times 10^{-3}$ cm, and therefore, $S_1 = 1.068 \langle \epsilon_1 \rangle - 0.147 \langle \epsilon_2 \rangle$ for a given h_o and v . Examining this at 165 Oe and 200 Hz we find $\langle \epsilon_1 \rangle \approx 4.0 \mu V$ and $\langle \epsilon_2 \rangle \approx 7.5 \mu V$ yielding $S_1 \approx 3.2 \mu V$ and $A_1 \approx 0.8 \mu V$. This value for A_1 compares favorably with our approximative first principles calculation of $\sim 0.4 \mu V$ -- see Appendix AA.

Next, the data of the two runs were examined to determine the loss per unit area per cycle associated with the copper voltage signal as a function of field amplitude h_o at a fixed frequency of 200 Hz. The results of this are shown in Fig. 4. It is to be noted that W_{Cu} displays the expected h_o^2 dependence associated with eddy behavior. Fig. 5 shows the copper signal at 500 Hz versus that at 200 Hz for the same field values. The slope is about 6.10 and that expected on the basis of eddy behavior is 6.25. These results taken together provide confirmation of the existence of eddy-current voltages in the copper pick-up coil which in turn give rise to an apparent W . In the subsequent reduction of our data we have taken the experimentally determined relations for

$\langle \epsilon_{Cu} \rangle$ and W_{Cu} , i.e.

$$\langle \epsilon_{Cu} \rangle = 1.81 \times 10^{-15} \frac{NR}{\rho} a^3 v^2 h_o \text{ volts}$$

$$\text{and } W_{Cu} = 1.67 \times 10^{-16} h_o^2 v \text{ Joule cm}^{-2} \text{ cycle}^{-1}.$$

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Figure 4

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Figure 5

3. Nb and Nb₃Sn

The ac loss behavior of the niobium single crystal and the low-loss Nb₃Sn sample was markedly different from that of the Mo-25 Re and could be represented by $W_T(h_o, \nu) = W_o(h_o) + \nu F(h_o)$ where $W_o(h_o)$ is the hysteretic loss per area per cycle and $F(h_o)$ represents the field dependence of the frequency dependent contribution. Fig. 6 is a dc magnetization curve of the Nb single crystal while Fig. 7a reveals the total loss per area per cycle of the niobium as a function of frequency for various values of the amplitudes of the ac field. Two things should be noted: (1) the linear dependence of the loss per cycle on frequency, and (2) the fact that the losses decrease to very small values at zero frequency (i.e., $W_o \leq 5 \times 10^{-9}$ Joules cm⁻² cycle⁻¹). This is to be compared with the work of Rocher and Septfonds⁸ who found a slight linear dependence of the loss per cycle on frequency in niobium and also a hysteretic loss in fields as low as 90 Oe. Their method of measurement was a calorimetric one and so avoided any problems associated with the loss behavior of pick-up coils. The low-loss Nb₃Sn exhibits a similar response as shown in Fig. 7b but with the distinction that in this case a measurable hysteretic contribution (finite W as $\nu \rightarrow 0$) is in evidence which increases rapidly with field amplitude.

Fig. 8 shows the hysteretic loss per cycle W_o in the low and high-loss Nb₃Sn. The latter reveals an h^3 dependence over the field range investigated. The hysteretic loss per cycle in the low-loss Nb₃Sn is at least two orders of magnitude less than for the high-loss material at fields $\lesssim 1$ kOe.

Figure 6

Figure 7

ORNL-DWG 75-8062

Figure 8

Fig. 9 shows $F(h_0)$ versus h_0^2 for the two Nb_3Sn samples and the single crystal niobium. The low-loss Nb_3Sn reveals a departure from h_0^2 only at the highest fields tested while the high-loss Nb_3Sn appears to depart from h_0^2 behavior at even the lowest fields examined. $F(h_0)$ for our single crystal niobium shows a departure from h_0^2 dependence occurring at a field value intermediate between the two Nb_3Sn samples.

Losses measured in the single crystal Nb and Nb_3Sn show very little dependence on previous magnetic cycling in fields up to 1400 Oe. Thus there should be little ambiguity in the magnitude of such losses between various experiments in contrast to the results on Mo-25 Re and other such alloys.

E. Discussion

Losses in all the samples we examined (including the Mo-25 Re) begin to appear at very low fields and a word concerning this aspect of the response is in order here. There are two factors working toward finite losses in small fields and both arise from the departure of the sample surface from ideal planar geometry. First, the demagnetization factor D associated with pseudo-ellipsoidal protuberances leads to the onset of the mixed state within these regions at a field value of $(1-D)H_{C1}$ where H_{C1} is the bulk lower critical field.⁹⁻¹¹ Second, there is the phenomenon of field enhancement in the vicinity of these surface irregularities. Melville¹² has discussed this effect in analogy to the electric field enhancement known to occur from studies on field emission and electrical breakdown. If γ is the enhancement factor of a magnetic field applied parallel to the overall surface near a projection, then the field amplitude

ORNL-DWG 75-8064

Figure 9

h_o need only take on the value $(1-D)H_{C1}/\gamma$ in order that fluxoids enter regions of surface irregularity. Plausible values of D and γ lead one to expect such establishment in applied field amplitudes significantly reduced from H_{C1} .

1. Mo-25 Re

Various models for hysteretic losses predict a dependence on magnetic field of h_o^n where $n \geq 3$ (Ref. 29-32). Our results on Mo-25 Re indeed display a cubic dependence on h_o at the lowest fields providing that the sample has not undergone excursions to high fields with subsequent flux trapping. This type of loss dependence on h_o is typical of hysteretic losses and is predicted by the Bean model for low fields (i.e., $h_o \ll H^*$, the "filling field"). The extremely rapid increase of W with h_o over the range from 600 to 900 Oe makes it very difficult to compare results between various researchers on such alloy superconductors (aside from complications due to different magnetic histories). To fit our data in this relatively small field region to $W \propto h_o^n$ would yield a value of n between 10 and 12.

While there is reasonable agreement between ac and dc determined losses in Mo-25% Re at fields where such losses are not influenced by previous history, we do not have an explanation for the observed differences measured for the losses at fields in excess of 1 kOe. Earlier work conducted by Linford and Rhodes¹³ on hysteretic losses determined via ac and dc methods reveals much the same degree of agreement as we find in this work. An inspection of the waveform of the signal induced in the compensated pick-up coil at these fields does indeed indicate a

greater degree of flux exclusion in increasing field which in turn gives rise to a larger ac loop and a greater loss than the corresponding hysteresis loop taken from the dc magnetization studies. The additional losses may be due to bulk viscous effects in the Mo-25 % Re as these fields are well in excess of H_{C1} , but more detailed work needs to be done to clarify this, particularly since the ac losses here appear to be independent of frequency, at least over the narrow range (40 to 100 Hz) which could be achieved in this work at these higher fields. As we have often seen, measurements need to be taken over a significant range of frequencies to avoid misinterpreting possible scatter in data.

Irie *et al*¹⁴ have examined the field and frequency behavior of ac losses in Nb-50 Ta at field amplitudes up to $H_{C1} \sim 60$ Oe at 4.2K and for $0.5 \lesssim \nu \lesssim 18$ kHz. This system can be expected to behave similarly to our Mo-25 Re, both materials exhibiting relatively low pinning. Irie *et al* find losses per cycle varying at h_0^3 in fields less than H_{C1} with an extreme increase in the dependence of W_0 on h_0 occurring near H_{C1} . In addition, while they do find losses per cycle varying with frequency, these are seen to occur only at frequencies $\gtrsim 500$ Hz. For frequencies $\lesssim 500$ Hz only hysteretic losses can be resolved from their data. Our data taken up to 500 Hz are consistent with these results. The field and frequency response of both materials are qualitatively similar and may be representative of superconductors where the dominant loss mechanism is hysteretic.

2. Nb and Nb₃Sn - Hysteretic Losses

Fig. 10 consists of six scanning electron micrographs taken of the Nb and two Nb₃Sn specimens. For each sample, two micrographs are presented, one perpendicular to the specimen surface and one viewing its edge profile. The high-loss Nb₃Sn (Figs. 10a and b) has a rough surface structure on the scale of a micron. The low-loss Nb₃Sn (Figs. 10c and d) and the single crystal niobium (Figs. 10e and f) have much smoother surfaces than the high-loss Nb₃Sn with the low-loss Nb₃Sn showing somewhat less structure on a scale of the order of a micron than that of the single crystal Nb. The ridges and depressions in both the low-loss Nb₃Sn and the single crystal Nb are predominantly aligned in a direction parallel to the axis of the cylindrical sample and therefore parallel to the applied magnetic field.

Hysteretic losses per cycle, if present in our single crystal Nb sample, must be less than the order of 5×10^{-9} Joules cm^{-2} cycle^{-1} in fields approaching H_{C1} (see Fig. 7a). This is compatible with the extremely smooth surface of the Nb specimen as seen in the micrographs (Figs. 10e and f) and the correspondingly small values of γ and D .

The hysteretic loss per cycle of the high-loss Nb₃Sn has an h_o^3 dependence over the field range investigated (Fig. 8) for which the Bean model predicts a critical current density of:

$$J_c = \frac{5 \times 10^{-7}}{12\pi^2} \left(\frac{h_o^3}{W_o} \right) \text{ A cm}^{-2}$$

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Figure 10

which for our case yields a value of $J_c = 6.6 \times 10^6 \text{ amp.cm}^{-2}$ for $165 \leq h_o \leq 1324 \text{ Oe}$. While the Bean model can be used to calculate a uniform J_c whenever $W_o \propto h_o^3$ it is not apparent that such a J_c should be associated with the actual coarse structure seen at the surface of our high-loss Nb_3Sn sample. More to the point in this case is the prediction of Melville¹² for losses occurring in surface irregularities. Melville has shown that the hysteretic loss per cycle per unit surface area is related to the field amplitude by $W_o \propto h_o^{u+2}$ where u is characteristic of the shape of the surface irregularities. In our case, a scanning electron micrograph taken of the surface at $10^4\times$ magnification reveals the contours of the irregularities to be best approximated by a series of straight line segments which can in turn be represented by some average slope. For such a case, the parameter u (called w in Melville's paper) assumes the value of unity and $W_o \propto h_o^3$. Melville's estimated magnitude of loss for $u = 1$ is about $(0.2 - 6) \times 10^{-7} \text{ Joules cm}^{-2} \text{ cycle}^{-1}$ at a field of $\sim 1000 \text{ Oe}$. This is in good agreement with our measured value of $6.5 \times 10^{-7} \text{ Joules cm}^{-2} \text{ cycle}^{-1}$ at this field.

The low-loss Nb_3Sn has a much smoother surface on a scale of the order of $\sim 1 \mu\text{m}$ and thus correspondingly reduced enhancement and demagnetization factors. The result is a higher degree of flux exclusion from the surface in the lowest fields, and due to a very large J_c , a high degree of flux exclusion from the bulk even in fields somewhat in excess of H_{c1} , yielding lower losses per cycle but also an extremely strong dependence of W_o on field amplitude once flux enters. This would seem to indicate that at higher fields the hysteretic losses per area per cycle of the low and high loss samples of Nb_3Sn may become comparable.

as one might expect once the loss mechanism becomes characterized by the bulk response of the superconductor. In general, the hysteretic loss contribution appears strongly correlated to the physical nature of the surface in the Nb and Nb₃Sn cases.

3. Nb and Nb₃Sn - Non-Hysteretic Losses

The frequency dependent losses per surface area per cycle are characterized by $\nu F(h_0)$ reflecting the fact that in all cases any variation of W_T with frequency was found to be a linear one. Fig. 9 shows $F(h_0)$ versus h_0^2 . It was found that the initial variation with h_0 was quadratic up to a certain field h_0^* . The low-loss Nb₃Sn has an h_0^* of about 1200 Oe while the value associated with the single crystal Nb is about 930 Oe. The high-loss Nb₃Sn departs from an h_0^2 variation at very low fields making it difficult to assign a value of h_0^* . As can be seen in Fig. 9 the magnitudes of $F(h_0)$ for the three samples at a given applied field are only slightly different (about a factor of 5 at 1200 Oe). There appears, however, to be a stronger correlation of h_0^* with surface condition although the reason for this is not understood at present.

Normal excitations scattering near the specimen surface would yield a $W \propto \nu$ and, assuming a layer of fixed depth from which these "electrons" originate, a dependence for h_0 of $W \propto h_0^2$. In fact, the Nb and low-loss Nb₃Sn showing only slight departure from ideal geometry on a scale of the order of a micron display an initial quadratic field dependence up to 900 Oe and 1200 Oe, respectively. In this regard, it is of interest to note that Campbell technique¹⁵ measurements of flux gradients in some

superconductors reveal an absence of any gradient within a region located at the surface. Tunneling experiments¹⁶ have also indicated the possibility of normal material within a coherence length of the surface.

The possibility also exists that the frequency dependent term arises from fluxoid motion. The degree of surface smoothness of the Nb and low-loss Nb₃Sn specimens would seem to mitigate against this, especially in the case of the Nb from which $H_{C1} \approx 1275$ Oe. Kramer,¹⁷ however, has postulated that in the process of vortex formation there is a stage at which nascent vortices first broaden to form a normal layer at the surface from which Abrikosov vortices subsequently nucleate.

Thus, there are in fact a number of independent studies which have suggested the possible existence of a normal-like region in the immediate vicinity of the surface. Certainly ac losses, in those superconductors for which such processes are confined to the neighborhood of the surface, should be expected to be extremely sensitive to such a layer.

Table I illustrates the contributions of W_o and $W(v) \equiv vF(h_o)$ to the total loss per area per cycle occurring in the listed superconductors at a frequency of 60 Hz and a field amplitude of 745 Oe. It is to be noted in passing that if a detailed frequency dependence were not made, and if no correction were made for losses in the copper pick-up coil, the loss value assigned as being totally hysteretic would be an overestimate of the actual W_o . For example, at $v = 60$ Hz and $h_o = 890$ Oe,

$$\frac{W_o}{W_o + W(\text{Cu}) + W(v)} \approx 0.272 \text{ and not unity. This points out}$$

the need for examination of the frequency response of ac losses and careful analysis of whatever experimental method is used in order to properly identify the contributions to the total apparent loss and to assign correct values to their magnitudes.

F. Summary and Conclusions

Losses per surface area per cycle in all materials examined were of the form $W_T(h_o, \nu) = W_o(h_o) + \nu F(h_o)$. Extensive examination of the frequency dependence of the losses was necessary in order to correctly identify the hysteretic loss contribution $W_o(h_o)$. Elimination of the eddy signal induced in the copper pick-up coil allowed a determination of any frequency dependent loss per cycle occurring in the superconductor.

The Mo-25 Re, used primarily to provide a method of checking our method of ac loss determination against the dc case, revealed no resolvable frequency dependent contribution to W . Its response may be representative of that associated with weakly pinned superconductors in which flux can penetrate well into the bulk sample during a field cycle. In this case, the large hysteretic losses mask any frequency dependent contribution.

Nb_3Sn prepared by the solid state diffusion technique (low-loss sample) exhibited only small hysteretic losses ($W_o \leq 7 \times 10^{-9}$ Joules cm^{-2} cycle $^{-1}$) for fields below about 950 Oe. The small hysteretic loss per cycle for fields well in excess of H_{c1} appears to be the result of an extremely large critical current density ($J_c \approx 2 \times 10^7$ amp cm^{-2} at ~ 1000 Oe) associated with this specimen with consequent little flux penetration. A small but measurable frequency dependent term was observed at all fields for which $F(h_o) \propto h_o^2$ up to about 1200 Oe.

Single crystal Nb showed small hysteretic losses, ($W_o \lesssim 5 \times 10^{-9}$ Joules cm^{-2} cycle $^{-1}$) for fields up to about H_{C1} (1275 Oe). A frequency dependent term somewhat larger than that of the low-loss Nb_3Sn was observed, however, having $F(h_o) \propto h_o^2$ up to about 900 Oe and then varying more strongly with h_o for higher fields.

The "high-loss" Nb_3Sn sample had both large hysteretic and non-hysteretic loss contributions. A hysteretic loss per area per cycle varying as h_o^3 was observed across the field range examined (100-1300 Oe) and a frequency dependent term was revealed whose $F(h_o)$ departed from quadratic dependence at fields of the order of 100-200 Oe. The hysteretic term appeared to be that predicted by Melville¹² for rectilinear surface irregularities.

While the magnitude of the hysteretic losses appears to be strongly correlated with the degree of surface roughness on a scale of the order of a micron, the same is not the case for the frequency dependent contribution to the total loss per area per cycle.

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Appendix AA

Eddy Signal from the Pickup Coil

The experimental method of ac loss determination in which a pick-up coil is wound on the sample and in which an ac magnetic field is produced at the site of the sample by means of an external solenoid, merits a careful analysis of the observed induced voltage signal. For pure sinusoidal applied magnetic fields, the mean loss per cycle is determined by the component of the induced voltage having the same frequency as and being in phase with the applied magnetic field.

The method used in our experiments allows an isolation and examination of this component by means of lock-in techniques using phase sensitive detection. Nevertheless, there will be a contribution to this component of the induced voltage due to eddy-currents set up in the pick-up coil itself. The following is a calculation of the order of magnitude of such signals expected to appear in general and in our experiments in particular.

Consider a single layer coil of N turns wound on a sample of radius R , the wire itself being of radius a . The peak induced voltage ϵ_0 in phase with the applied field is given by $\epsilon_0 = 8\pi^2 N R a \nu \mu'' h_0$ where ν is the frequency of $h(t)$, μ'' is the imaginary part of the permeability of the copper under application of $h(t)$ and h_0 is the magnitude of $h(t)$. Here we have assumed that $a \ll R$. In general, $b_0 = (\mu' - i\mu'')h_0$ where the real and imaginary part of the permeability are taken to be those given by Landau and Lifshitz¹⁸ for an infinite cylinder of radius a situated in a transverse ac magnetic field. Our geometry approximates this case quite well since $R \gg a$.

In the limit of $\delta \gg a$ where δ is the skin depth (a condition also well met in our case) one can write:

$$\mu' = \mu_0 \left[1 - \frac{1}{24\pi} \left(\frac{a}{\delta} \right)^4 \right] \approx \mu_0$$

and
$$\mu'' = \frac{\mu_0}{8\pi} \left(\frac{a}{\delta} \right)^2 \text{ where } \mu_0 = 4\pi \times 10^{-7}$$

Substituting for the skin depth its usual expression and converting from MKS units to those of experimental convenience, we arrive at the form for the RMS value of the eddy signal, $\langle \epsilon \rangle = 8.77 \times 10^{-16} \frac{NR}{\rho} a^3 \nu^2 h_0$ volts where R and a are in cm, ρ in $\Omega \cdot \text{cm}$, h_0 in Oersted and ν in Hz. For the actual parameters occurring in our experiments this yields a value of $\langle \epsilon \rangle \approx 0.4 \mu\text{v}$ at a field amplitude of 165 Oe and a frequency of 200 Hz.

Appendix AB

Analysis of the Experimental Method

Consider the applied ac magnetic field to be represented by $h(t) = h_0 e^{i\omega t}$. We can then represent the response of the induction as follows: $b_0 = \mu'_1 h_0$ in the helium space interior to the copper windings of the coils and $b_0 = (\mu' - i\mu'')h_0$ within the windings themselves. If no flux penetrates the superconductor (a good approximation in fields of a few Oe where the compensation condition is set) then the compensated signal is given by:

$\epsilon = N_c \frac{d\phi}{dt} P$ where N_c , N_p are the number of turns of the compensator and pick-up coils respectively, and ϕ_c , ϕ_p refer to the flux threading the compensator and pick-up coils respectively, both within their enclosed areas and within the actual windings. In terms of the peak induced signal ϵ_0 , we have:

$$\epsilon_0 = 2\pi d (N_p R_p - N_c R_c \eta) \mu'' \omega h_0 + i\pi \{ 2N_p [R_s \Delta R \mu'_1 + R_p d \mu'] - \eta N_c R_c (R_c \mu'_1 + 2d \mu') \} \omega h_0$$

where R_p , R_c , R_s are the radii of the pick-up coil, compensating coil and sample respectively, ΔR is the mean difference between R_p and R_s , d is the effective thickness of the wire assumed to be the same for both pick-up and compensator windings, ω is the angular frequency of the magnetic field, η is the fractional value of the field at the location of the compensating coil relative to the field value at the location of the sample. The compensation of the induced signal in phase with $\frac{dh}{dt}$ is achieved by mechanically adjusting the former on which the coils are wound as described in the text since obviously $\eta = \eta(x)$. Proper compensation then means that the imaginary term in the above expression for

ϵ_0 is zero which yields for $\Delta R \ll R_s$, $\eta(x) = \frac{2N_p R_s}{N_c R_c} \left(\frac{\Delta R + d}{R_c + 2d} \right)$ independent

of h_o . At the low frequencies examined here it is well justified to take $\mu'_1 \approx \mu' \approx \mu_o$. Even at proper compensation, however, there will be a non-zero real part to ϵ_o and inserting the above expression for η yields:

$$\epsilon_o = 2\pi N_p R_s d\mu''\omega h_o \left(\frac{R_c - 2\Delta R}{R_c + 2d} \right)$$

where replacing the expression in the parentheses by unity introduces an error of about 5% for our geometry. This signal cannot be compensated out and in fact represents the loss occurring in the copper windings--a problem which has been examined in the text and analyzed in Appendix A.

It is worthwhile pointing out that the signal from the compensator alone which is used to set the lock-in phase to accept only voltage signals in phase with $h(t)$ is shifted from ideality by only $\sim 1.5^\circ$ of arc at 200 Hz due to the eddy component in the compensator.

It can also be shown that any channel overloading which might occur within the lock-in amplifier due to the coherent signal in quadrature with the one of interest will give rise to at most a small pseudo-hysteretic type term--its W being frequency independent and varying as h_o^2 .

Finally, any magnetoresistance that a pick-up coil material might possess can be shown to lead to a weaker than h_o^2 dependence of W_{coil} . The mean resistivity can be written as:

$$\bar{\rho}(h_o) = \rho_o (1 + \alpha h_o^2)$$

where ρ_o is the resistivity in the absence of any applied magnetic field. For $\alpha h_o^2 \ll 1$, $W_{coil} \cong 1.63 \times 10^{-16} \frac{a^3 h_o^2}{\rho_o} (1 - \alpha h_o^2) \nu$ Joule cm^{-2} cycle $^{-1}$ as long as δ (skin depth) $\gg a$.

Appendix B

PRECIPITATION HARDENING AND RESISTIVITY IN DILUTE
ALUMINUM-GOLD ALLOYS OF VERY HIGH PURITY

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PRECIPITATION HARDENING AND RESISTIVITY IN DILUTE
ALUMINUM-GOLD ALLOYS OF VERY HIGH PURITY

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The use of aluminum as an electrical conductor has gained much momentum in recent years. Breakthroughs into applications that hitherto were the bastions of copper have been and apparently will continue to be achieved.¹

Several currently developing technologies envision utilizing the fascinating phenomena of superconductivity. These include, for example, high current underground power transmission lines, energy storage devices and large, high-field magnets. The current-carrying elements in these electrical systems will undoubtedly be constituted of some form of composite.² These may be wires, tapes, or tubes depending on the application and designer, but in all cases will at the very least consist of a superconducting material (Nb, Nb₃Sn, NbTi, etc.) joined to or embedded within a normal conducting metal (Cu, Al, etc.). The normal metal is required mainly for stabilization purposes at rated current and to provide an alternate current path should the superconductor temporarily go normal. The normal metal may also need to function as a heat sink, provide a barrier between vacuum and liquid-helium-filled channels, mechanically support the superconductor (which may be brittle) assembly and carry external forces.

High-purity aluminum is being considered as a candidate material to serve this stabilizing function. If it could also carry load and

mechanically support the superconductor assembly certain design simplifications and weight savings might be realized. In these functions aluminum is, of course, in competition with copper which is stronger at low temperatures. But in high magnetic fields where magneto-resistance becomes important, aluminum is apparently superior.³ Both aluminum and copper have excellent conductivity at low temperatures (T approaching 4.2K); however, it is possible⁴ to produce a zone-refined aluminum with a residual resistance ratio (RRR)* of 40,000 which surpasses that currently available with copper.

A major disadvantage in using high-purity aluminum is its low yield strength - at this point defined loosely as the stress at which plastic flow sets in. A second drawback is the relatively more difficult task required to bond aluminum, as opposed to copper, to other materials.

This manuscript is a final report which describes experimental work begun in December 1972 and supported by the U. S. Atomic Energy Commission's Division of Applied Technology, now Energy Research and Development Administration's Electric Energy Systems Branch. The work was administered through ERDA's underground superconducting power transmission systems project. Its aim was to discover feasible ways to strengthen aluminum while maintaining a reasonably high RRR. More specifically, the possibility of manipulating the dispersant in a dilute precipitation hardenable aluminum alloy whose solid solubility is extremely limited, was explored. The solute element chosen for addition to aluminum was gold.

*RRR is defined as the ratio of the resistance at ambient temperature to that at liquid helium temperature (4.2K).

Statement of Problem, Hypothesis, and Rationale

The stabilizing element in a superconductor must have a very low resistivity at the operating temperature of the system. Thus Taylor⁵ estimated that for a Nb/Al tubular conductor composite in a superconducting ac power transmission cable, a RRR of 2500 would be required for the aluminum. A more sophisticated, flexible superconducting coaxial cable designed at Brookhaven National Laboratory⁶ specified a 5000-RRR aluminum backing for a Nb₃Sn tape. Boom, McIntosh, Peterson, and Young⁷ chose a 1000-RRR aluminum to stabilize the composite conductor in their proposed, large superconducting energy storage device. From these examples, it would seem that an aluminum with RRR in the range of 1000 to 10,000 is called for.

As alluded to before, a serious drawback to using high-purity aluminum is its low yield strength and the ease with which it can be deformed. Deformation produces dislocations in the material which raises its resistivity substantially,⁴ i.e., lowering RRR. Reed⁸ has assembled data on the deformation properties of high-purity aluminum and dilute alloys. According to Arp, Kasen, and Reed,⁴ tensile tests on 1900 RRR aluminum performed at 4.2K indicated a yield stress at 0.2% offset (σ_{YS}) of approximately 27.6 MPa (4000 psi). At ambient temperature, σ_{YS} was even smaller. Baylis⁹ suggested that for design purposes a σ_{YS} of 44.9 MPa (6500 psi) should be considered as necessary in aluminum-stabilized tubular superconducting cables. In the superconducting energy storage device noted earlier,⁷ a σ_{YS} of 34.5 MPa was sought. Strengths greater than these would be accepted without reservation. To attain acceptable strength levels, use of a slightly prestrained high-purity aluminum was

proposed.⁹ But because this prestrain introduces defects which raise the material's resistance, its fault-current-carrying capabilities were reduced to at best only a marginally acceptable level.⁹ It seemed highly desirable then to search for a method to strengthen aluminum to $\sigma_{YS} = 45$ MPa while holding a RRR of 2500.

The major sources of resistivity in the metals pertinent to this problem are (1) scattering due to lattice vibrations; (2) scattering by crystal defects, including point, line and planar defects (vacancies, dislocations and grain boundaries as example); and (3) scattering by dissolved solute atoms either purposely added or present as unavoidable impurities. As temperature approaches absolute zero the resistivity from lattice vibrations diminishes rapidly and tends toward zero whereas that due to crystal defects and solute atoms is relatively temperature independent. Thus, at very low temperatures, i.e., temperatures where a superconducting device would operate, solute atoms and crystal defects are the important residual resistivity contributors. The use of well-annealed metal would help to minimize the crystal defect resistivity contribution. Thus, RRR could and often has been regarded as an indicator of impurity level in specially purified metals.

Not all solute atoms contribute equally, however, so that in aluminum, transition element solutes seem to increase the residual resistivity 5 to 10 times more (per atom added) than non-transition solutes do.¹⁰ In applications where low resistivity is essential, it is imperative to reduce the level of transition element atoms. This can be accomplished by special purification techniques such as zone-refining.

There are numerous procedures whereby a metals mechanical strength or hardness can be improved. Some of the more established strengthening mechanisms include (1) work or strain hardening, (2) solute strengthening, (3) precipitation hardening, (4) quench hardening. Unfortunately each of these (and also other less well known hardening processes) require the build-up of either the above mentioned lattice defects and/or the solute atoms both of which cause the resistivity to increase. Thus strain hardening comes about from the multiplication and accumulation of dislocations. Solute hardening and precipitation hardening require the addition of alloying elements and quench hardening involves the agglomeration of excess point defect concentrations.

It seems then to be generally true that those factors which give rise to strengthening metals also increase their resistivity. Hence, we find ourselves in a "trade-off" or compromise situation when attempting to produce a high-strength, low-resistivity aluminum.

The case of precipitation hardening deserves a further look for the following reason: Some alloying elements are extremely insoluble in aluminum, e.g., they prefer to be gathered together (like atoms are attracted to one another) and exist as second-phase particles embedded in a reasonably pure matrix. These insoluble particles would also be expected to raise the resistivity of the alloy but in a way that probably depends upon the mean free path between particles. This contribution is expected to be much smaller than that due to an equivalent amount of impurity atoms completely dissolved in the aluminum. Added to this is the possibility that such second-phase particles might be effective strengthening agents by blocking the passage of dislocations through the

material. All this suggests that the controlled addition of a specific, low solubility solute to an already very high-purity aluminum might offer the possibility of precipitation hardening to achieve suitable strength while maintaining an acceptable RRR level.

Few non-transition element solutes have the required low solid solubility. But past studies^{11,12} of dilute aluminum-gold alloys suggested that gold may have an extremely low solid solubility in zone-refined aluminum. This was ascertained from recrystallization studies¹¹ where grain boundaries were observed to move at very fast rates through a 40-ppm alloy in which the gold was completely precipitated in an aluminum matrix. Similar observations in a 3-ppm alloy with all the gold in solid solution, on the other hand, revealed the boundary migration rates to be somewhat slower. Interpretation of these results on the basis of the solute-drag theory of grain boundary migration led to the conclusion that the aluminum matrix in the precipitated 40 ppm alloy necessarily contained less than 3 ppm gold and probably as little as 1 ppm at 373 K (100°C). Unpublished data from the earlier work is shown in Fig. 1 where the resistivity at 4.2 K is plotted against dissolved gold concentration. The impurity resistivity contribution of dissolved gold in aluminum was estimated from the slope of this line to be $1.4 \times 10^{-3} \mu\Omega\text{-cm/ppm}$. So, when 1 ppm of gold is added to a pure aluminum with an RRR of 13,000, an RRR = 7500 would be anticipated if no other hindering factors were present.

Heimendahl¹² has reported a remarkable age hardening in somewhat more concentrated aluminum-gold alloys. In an alloy containing 270 at. ppm (0.2 wt %) gold, for example, a dispersion of gold in the form of Al_2Au precipitates was generated by heat treatment to increase the room

Fig. 1. Electrical resistivity as a function of alloy concentration for aluminum-gold alloys.

temperature yield stress by a factor of three. Less concentrated alloys would still exhibit precipitation but the degree of hardening would probably be smaller.

The purpose of the present study was to characterize the precipitation hardening behavior of dilute aluminum-gold alloys with an important objective being to "map out" the yield strength (hardness)/RRR relationships for various dispersion states. The unique aspect of this work was the preparation of these alloys from ultrapurity starting materials. Thus, it was demonstrated that on a laboratory scale, superpure aluminum alloys could be melted, cast, fabricated, and tested with no apparent contamination.

EXPERIMENTAL

Starting Materials

Specially prepared zone-refined aluminum specified to have a residual resistivity ratio equal to $13,000 \pm 1000$ was purchased from Cominco Products, Inc., Spokane, Washington, and used as the starting stock to produce the alloys investigated in this study. (This RRR represents an equivalent total impurity level of less than 1 at. ppm.) The alloying addition was high-purity gold stated by the suppliers to be 99.999% pure. Two lots were purchased in the form of 1.6 mm (1/16 in.) diameter wire. These were provided by the Leytess Metal and Chemical Corporation, New York, and the Materials Research Corporation, Orangeburg, New York.

RRR Measurements

The standard four probe d-c technique was used to measure resistivity. It involves passing a known current through a wire specimen 2.8 mm in diameter by 50 mm long, measuring the voltage drop across the specimen and calculating the resistance from Ohm's law. Figure 2 shows a close-up view of a typical test specimen. The current leads (A in the bottom view)

Fig. 2. Close-up of RFR specimen with the current, A, and voltage, B, leads attached. Top view shows specimen secured at the end of probe.

made of zone-refined aluminum were fusion welded with a tungsten arc under a helium atmosphere to the wire specimen. Voltage leads (B in the bottom view) made of 0.107 mm diameter high-purity copper wire were very carefully taped to the specimens so as to prevent accidental deformation of the material. The top view of Fig. 2 shows a properly supported specimen nestled at the end of the probe awaiting insertion into the measuring baths. For each specimen the resistance was ascertained at two temperatures: 273 K in an ice-water bath and 4.2 K in a liquid helium cryostat. RRR was calculated as the ratio of these two resistance values. Duplicate measurements on the same specimen were repeatable to within $\pm 1\%$ for $50 < \text{RRR} < 1000$. For wires with $\text{RRR} > 5000$ the measurements are probably reproducible to $\pm 10\%$.

Microhardness

In this work Vicker's microhardness measurements were used to probe the strength of the alloys. Though much maligned, the hardness test because of its simplicity remains nevertheless a useful tool for correlating strength properties of materials. Important correlations with yield strength and flow stress in metals have been demonstrated in several studies.^{13,14} Our measurements were performed using a Leitz microhardness tester under applied loads of either 15, 25, 50, or 100 g. Initially tests were made on the cylindrical surfaces of wire samples 2.8 mm in diameter and 25 mm long. Poor reproducibility and measuring difficulties led us to repeat many measurements on flat, parallel-sided surfaces. These samples were machined from the wires and had a thickness of 2 mm. Unless otherwise stated the quoted microhardness numbers (VHN) are the average of between 5 and 10 measurements on the same specimen.

Alloy Preparation

Before any alloys were mixed, the zone-refined aluminum starting stock was subjected to a RRR study to characterize its purity and check whether the supplier's "assay" was accurate. Two batches, both supposed to have a RRR of $13,000 \pm 1000$ according to the supplier were tested. Table I summarizes the results. Batch I was left-over stock from a previous study. Batch II was the material from which the majority of the present alloys were produced. The table illustrates how sensitive the RRR of Batch II material is to machining and thermal history, i.e., heat treatment and cooling rate. At best the RRR was 30% lower than that specified by Cominco. The nature of the quality control one might expect from one lot to another is also indicated in the table. It should be noted in this connection that later tests on Batch II material taken from a different portion of the as-received stock when melted, cast and fabricated, yielded an RRR of 15,000.

Initially 280 g of a master alloy containing nominally 1.1 wt % gold in zone-refined aluminum were prepared. A suitable quantity of gold wire was placed with saw-cut chunks of aluminum in an ultrahigh purity graphite crucible. The charge was melted together by induction heating under a quartz-enclosed vacuum of better than 1.33×10^{-3} Pa (1×10^{-5} torr) in the apparatus pictured in Fig. 3.

Care must be taken at all stages in the preparation of these aluminum alloys to prevent accidental contamination. Therefore, after sawing, the aluminum was carefully cleaned by chemical polishing in a solution of 70% by volume of phosphoric acid, 25% sulfuric acid and 5% nitric acid heated to between 333 and 353 K. Previous experience had shown that a 600-sec

Fig. 3. Photograph of the melting and zone-leveling facilities used for preparing laboratory melts of controlled purity aluminum alloys.

TABLE I

Residual Resistivity Ratio of Cominco Zone-Refined Aluminum*

Temperature (K)	Condition or Heat Treatment		RRR
	Time (ksec)	Remarks	
883	3.6	Furnace cooled	9,772
883	260	Furnace cooled	10,450
800	3.6	Furnace cooled Batch II	10,300
		Furnace cooled Batch I	~ 30,000
883	3.6	Quenched into ice-brine solution	2,800
		As-received centerless ground from ingot	6,165

*Except where noted, all specimens were from Batch II stock.

cleaning operation would remove any surface contamination due to the sawing. After chemical polishing, the aluminum was rinsed in distilled water, dried in air and stored in argon-filled plastic bags placed in the crucible.

The graphite crucibles were manufactured by United Carbon Products, Bay City, Michigan, and treated so as to have a very low ash content, < 10 ppm, thus minimizing contamination of the molten aluminum.

Several steps were taken to ensure macroscopic and microscopic homogeneity. First, the alloy was brought slowly to the molten state assuring adequate degassing. Second, a molten zone was passed along the length of the finger-shaped ingot alternately in both horizontal directions. This process, known as zone leveling was repeated 24 times. Finally, after directional solidification, the ends of the alloy ingot were cropped off; it was then replaced in the graphite boat, sealed off in quartz under argon (Fig. 4) and diffusion annealed for 1210 ksec in the solid state at 873 K. Chemical analysis and microstructural examination verified that homogenization of the master alloy had occurred.

Four alloys containing nominally 0.2, 0.16, 0.1, and 0.05 wt % gold in zone-refined aluminum were produced by mixing appropriate quantities of the master alloy with additional starting stock. The melting, casting, and homogenization procedures were the same as before. Two finger-shaped ingots approximately 127 mm long by 25 mm in diameter of each composition were made. In addition, zone-refined aluminum with no alloying addition was also subjected to these identical procedures for the purpose of producing stock from which base-line information could be gathered and to check whether our experimental technique would lead to contamination.

Fig. 4. Photograph of master alloy prior to homogenization anneal.

One continually recurring problem encountered early in this work involved the strong adherence between portions of the graphite boat and the solidified aluminum ingot. In many cases the graphite boat had to be cut or ground away and the aluminum alloy "scalped" with a file. After such scalping the billet was always heavily pickled with the aforementioned chemical polishing solution. At the suggestion of United Carbon Products, it was decided to test crucibles with a pyrolytic carbon coating placed on the standard crucible. The pyrolytic-coated crucibles proved to be vastly superior and very little adherence occurred when these were used. Ingots melted and solidified in these coated crucibles developed a grey film that could be removed by pickling. RRR tests confirmed that no significant contamination resulted. The pyrolytic carbon coated crucibles were also found to couple with the induction heating coil in a considerably more efficient manner. About 20 to 30% less power was required to do the melting and zone-leveling procedures.

Specimens for chemical analysis were removed from three of the alloys. Three types of analytical techniques were tried in an effort to obtain consistent results for the gold content. The analyses displayed in Table II are the best estimates based upon x ray fluorescence, neutron activation, and spark source mass spectrographic analyses. They agreed well with the material balances based on the starting weights of the initial charges.

Alloy Fabrication

The billets of the alloys and the pure aluminum were fabricated at room temperature by rod-rolling to a modified round cross section having a final diameter of 3.0 mm. Prior to rolling, the grooved rolls were carefully cleaned. All rolling was done without lubrication. The rolling reductions per pass were chosen so as to minimize flash formation. When

TABLE II

Gold Analysis on Al-Au Alloys

Alloy Billet Number	Gold (wt %)
55	0.12
56	0.11
60	0.26
61	0.24
64	0.07
65	0.06
72	0

flash or any other defects were noted, rolling was halted, the defective material removed and the rods cleaned by chemical polishing to prevent contamination. Finally, the rods were swaged to produce wire 2.8 mm in diameter.

Heat Treatment

All specimens were chemically polished prior to heat treatment. Each specimen was solution heat treated (SHT) in a molten salt bath for 57.6 ksec at 873 K. The salt bath consisted of a mixture of 40% sodium nitrite (NaNO_2) and 60% potassium nitrate (KNO_3). After SHT the specimens were rapidly quenched (Q) into an iced-brine solution of 10% sodium chloride (NaCl).

Aging was carried out on SHTQ specimens for times up to 3600 ksec at temperatures of either 371, 460, 513, 573, or 623 K. The aging media were boiling heptane (371 K), boiling decahydronaphthalene (460 K) or molten salt. After aging the specimens were water-quenched to room temperature.

Transmission Electron Microscopy (TEM)

All microstructural studies were carried out by transmission electron microscopic examination of thin foils using a Siemens-Elmskop 1A microscope operating at 125 KV. Specimens were prepared from the 2.8 mm diameter rods fabricated, SHTQ, and aged. Discs approximately 0.5 mm thick were carefully cut with a diamond saw and hand ground on emory paper to remove 0.1 mm from each face. This gave a section suitable for thinning by the dimpling process.¹⁵ Two different electrolytes were used for thinning:

- (1) 8% perchloric acid (by volume); 12% distilled water; 70% ethanol; and 10% ethylene glycol monobutyl ether
- or
- (2) 6% perchloric acid; 58% methanol; 35% ethylene glycol monobutyl ether; and 1% solvent X (ref. 16).

Both solutions were held at room temperature during thinning. After dimpling, further polishing in either

- (1) 95% ethanol (by volume); 5% perchloric acid
- at a temperature between 263 and 273 K or
- (2) 49% methanol (by volume); 49% nitric acid; 2% hydrochloric acid
- at a temperature of 258 K, were utilized to produce a pinhole in the foil. Most of the specimens that were examined were prepared in the second dimpling solution and the second polishing solution.

Unfortunately, a black film of gold always formed on the foils during these procedures. This hindered examination and prevented quantitative analysis of the alloys' microstructure. This film has been a problem in previous studies^{17,18} of aluminum-gold alloys. It was sometimes possible to improve the thin foils by dipping them in a stripping solution to remove the black film from the area to be examined. The stripping solution contained:

75% water (by volume)
8.75% phosphoric acid
13.75% sulfuric acid
2.5% nitric acid

The solution was used at a temperature of 343 K. The time required was of the order of a few seconds. In cases where this stripping procedure proved fruitless the foils were electropolished again, this time in a solution containing

67% methanol (by volume)
33% nitric acid

at a temperature maintained at 273 K. While observing the pinhole in the TEM specimen through an optical microscope, the cell current was gradually increased until polishing commenced and the black film could be seen floating away. This procedure sacrificed viewing area but in a few cases enough area was left for examination in the electron microscope.

RESULTS

One of the initial findings of this work was a demonstration that the laboratory procedures developed for melting, casting, fabricating and testing did not introduce any unwanted contamination into aluminum. This was verified by subjecting Batch I zone-refined aluminum to melting and casting procedures identical to those of the alloys and swaging the ingot to 2.5 mm diameter rod. Then RRR specimens were prepared, annealed for 3.6 ksec in an air furnace at temperatures between 673 and 883 K and furnace cooled. The results are displayed in Fig. 5 where RRR is plotted versus annealing temperature. As-swaged material has a relatively low RRR value presumably due to the dislocations introduced during swaging. The strong increase in RRR between 673 and 798 K was interpreted as being due to the annealing phenomena of recrystallization and grain growth which

Fig. 5. RRR as a function of isochronal annealing temperature for Batch I zone-refined aluminum.

eliminated excess dislocations and grain boundary area. The figure also suggests the possibility of a maximum at 798 K; the cause of the slight decrease in RRR at the higher annealing temperatures is not known. These data qualitatively exhibited the same trends as that reported for high-purity copper by Carapella, Bess, and Laseur.¹⁹ (The RRR of their copper was at least an order of magnitude lower than our aluminum.) The important aspect of our results is that the RRR can be retained at a very high level in spite of the various handling procedures we employed.

In Table III the RRR and VHN data are presented for each alloy in the as-fabricated (after swaging) condition and in the SHTQ state. Where more than one test was performed an average value is listed. For both of these "defect" conditions the addition of gold increases microhardness. The effect is much larger in the as-fabricated condition even to the point of doubling the microhardness by the addition of 0.26 wt % gold. Alloying also makes a tremendous change in RRR but here these two "defect" states yield nearly comparable results. Apparently the deformation-induced defects are relatively more effective hardening agents than the quenching-induced ones while both affect RRR about equally.

After SHTQ each alloy was aged. Figures 6 through 10 demonstrate the RRR aging response for the temperature range 371 to 623 K and aging times up to 3600 ksec of the four alloys and zone-refined aluminum. Quite clearly aging causes the RRR to increase only slowly at the low temperatures but rather more strongly at the higher ones. Increases of the order of 20 times were noted in some cases.

Figures 11 through 14 portray in capsule form the pertinent microhardness results. One curious phenomenon that was noted was a variation

TABLE III
Microhardnesses and Residual Resistivity Ratios of
Aluminum and Aluminum-Gold Alloys

Condition Property	As-Swaged		SHTQ	
	RRR	VHN	RRR	VHN
Alloy*				
Zone-refined Aluminum	3637(2)	20.3	3715(4)	17.5(5)
Al + 0.05% Au	116.5(4)	34.9(2)	138	20.6(3)
Al + 0.12% Au	92.7(4)	36.7(2)	96(5)	21.3(7)
Al + 0.16% Au			57.5(2)	
Al + 0.26% Au	74(4)	40.7(2)	61(5)	21.7(8)

*Alloy percentages are by weight.

The number in parentheses indicate the number of tests averaged.

Fig. 6. RRR aging response of Al + 0.26 wt % Au alloy at a series of temperatures.

Fig. 7. RRR aging response of Al + 0.16 wt % Au alloy.

Fig. 8. RRR aging response of Al + 0.12 wt % Au alloy at a series of temperatures.

Fig. 9. RRR aging response of Al + 0.05 wt % Au alloy at several temperatures.

Fig. 10. RRR aging response of zone-refined aluminum at several temperatures.

Fig. 11. Effect of load on the VHN of an aluminum-gold alloy.

Fig. 12. VHN aging response for Al + 0.26 wt % Au at a series of temperatures.

Fig. 13. Effect of gold content on the VHN aging response of aluminum-gold alloys at 513K.

Fig. 14. Effect of gold content on the VHN aging response of aluminum-gold alloys at 573K.

of the measured VHN with the load applied to the hardness penetrator. In Fig. 11, VHN is plotted versus load for a wide range of conditions from as-fabricated to SHTQ to overaged. The result was qualitatively always the same in that VHN decreased with increasing load. Such behavior and its explanation have been discussed by Bückle.²⁰ This finding requires that realistic hardness comparisons can only be made when the tests were carried out under the same load, or when a correction factor could be estimated and applied to the data.

A complete set of hardness data at all aging temperatures was recorded for the 0.2 wt % alloy only and Fig. 12 shows those aging responses as a function of time. A hardness maximum was evident for each aging temperature except 623 K. As expected the maximum hardness shifted to longer times the lower was the aging temperature. A complexity was seen in the 460 and 371 K data in that the hardness went through a minimum before climbing to a maximum.

It should be recalled that the most drastic changes in RRR occurred during aging at the higher temperatures in the 0.26 and 0.12 wt % alloys. Figures 13 and 14 contrast the hardness response during aging at 513 and 573 K for these alloys. Zone-refined aluminum data are included for comparison. The more concentrated alloy produced the higher hardness as anticipated.

The primary goal of this work was to try to obtain a strengthened aluminum with reasonable RRR values. The maximum strength that could be produced would be expected for aging treatments giving rise to the peaks in the hardness curves. In Table IV values of RRR are listed along with the corresponding maximum hardness values for the two most promising

TABLE IV

RRR of Al-Au Alloys Aged to Maximum Hardness

Alloy	Aging Temperature K	RRR	Maximum VHN
Al + 0.12 wt % Au	513	260	22.2
	573	240	21.2
Al + 0.26 wt % Au	371	85	24.0
	460	165	22.9
	513	310	25.5
	573	110	24.5
	623	62	21.7

alloys. The RRR values are disappointingly low. The best condition was for the 0.26 wt % alloy aged at 513 K. Here an RRR of 310 and a maximum VHN of 25.5 were achieved.

The highest RRR values, unfortunately, occurred when the alloys were in the overaged condition, i.e., well past the hardness maximum. This effect is demonstrated graphically in Fig. 15 for both the 0.26 and 0.12 wt % alloys aged at 513 K. The major rise in RRR takes place for aging times where the microhardness is undergoing a steady decrease.

Finally, in Fig. 16 we "map out" RRR versus microhardness for all alloys and conditions where direct comparisons could be made. This plot illustrates the range of values that were obtained in our experiments.

In spite of the extreme difficulty encountered in preparing thin foils, various quenched and aged conditions of the 0.12 wt % gold alloy were examined in the electron microscope. The interpretation of these

Fig. 15. A comparison of the RRR and VHN aging responses of the 0.26 and 0.12 wt % gold alloys at 513K.

Fig. 16. RRR as a function of VHN for the 0.26 (circles) and 0.12 (triangles) wt % gold alloys and zone-refined aluminum (diamonds).

microstructures was sometimes ambiguous. Nevertheless, a sequence that characterizes what we believe to be the evolution of microstructure upon aging at 573 K is shown in Figs. 17 through 20. In the SHTQ condition heavily jogged dislocations and numerous dislocation loops were apparent. These are considered to result because of the severe quenching procedure. The dislocations persisted during short aging times (Fig. 17) but at longer times they vanished. Coincidentally, however, these were replaced by very fine precipitates (Fig. 18). While the first clear indication of precipitates was noted after about 3.6 ksec of aging it was not until 36 (Fig. 18), 360 (Fig. 19), and 3600 ksec (Fig. 20) that the precipitates were readily discernible. The precipitates, presumed to be Al_2Au , were in the form of thin discs with truncated surfaces parallel to a specific set of crystal planes thought to be $\{001\}$ planes. As aging progressed from 36 ksec to 3600 ksec, the precipitate density clearly decreased while the average size increased (compare Figs. 18, 19, and 20). This process is called coalescence. A few dislocation lines were still present even after the long aging times but these were usually attached to the precipitate particles. It was not clear whether these were some quenched-in dislocations or new ones generated during the precipitation process.

DISCUSSION

The SHTQ microhardness values obtained for the supersaturated alloys in this study were about 30% higher than those found by Heimendahl¹² in a study of precipitation in Al-Au alloys. This may have been caused by differences in specimen size, severity of quench or purity of starting material. It is known that quenching stresses can increase the vacancy concentration and generate excess dislocations that would increase hardness. The TEM observation of heavily jogged dislocations

Fig. 17. Transmission electron micrograph of Al + 0.12 wt % Au alloy SHTQ and aged 0.18 ksec at 573K. 111500x.

Fig. 18. Transmission electron micrograph of Al + 0.12 wt % Au alloy SHTQ and aged 36 ksec at 573K. 63700x.

Fig. 20. Transmission electron micrograph of Al + 0.12 wt % Au alloy SHTQ and aged 3600 ksec at 573K. 56000x.

and quenched-in loops would support this contention. Also the fact that zone-refined aluminum when quenched, had a higher hardness which could be subsequently annealed out, would indicate our quenching conditions were quite severe.

A rationalization of our aging results is as follows: After solution heat treatment and quench the alloys retain the bulk of their gold content in solid solution. In addition there are quenching defects (dislocation loops and vacancies) present. This "defect" state gives rise to a very low RRR and moderate VHN. Upon aging the gold atoms precipitate out of solution forming Al_2Au clusters that initially are finely dispersed and fully coherent with an aluminum matrix which is depleted in gold.^{17,18} At later stages of aging the clusters become identifiable precipitates and are only semi-coherent with the matrix. Simultaneous with that action, the quenched-in dislocation loops disappear. It is not clear whether they anneal out separately in a conventional manner or if they become nucleation sites for the gold-rich clusters and become integrated with the precipitates. Finally, coalescence of the precipitates takes place, i.e., large precipitates grow at the expense of small ones.

Figure 21 describes in schematic terms how these various individual factors might contribute to the overall microhardness. The formation of a finely dispersed coherent precipitate gives rise to a substantial hardness increase which is lost when the precipitate becomes semicoherent and coalesces. Matrix depletion of solute and defect cluster annealing both cause softening. At the various aging temperatures the rates of each of these component processes will be different relative to each other. The net effect could give the aging curves shown in Fig. 12.

Fig. 21. Schematic representation of the components of the micro-hardness aging response of precipitation hardenable Al-Au alloys.

During aging for the most part, there is an increase in RRR. This increase is not very substantial, however, until the precipitates undergo coalescence, i.e., aging times 360 to 3600 ksec. There is probably a strong contribution of the mean particle spacing to RRR. It is for this reason the highest RRR was < 3000 even though the matrix was very pure in the overaged condition. But to achieve this value much coalescence had to occur. Comparing the TEM results with the VHN data revealed that the maximum hardening set in when the precipitates were still too small to be seen. This implies finely dispersed particles. While this condition produces effective hardening the relatively small mean particle spacing must also contribute heavily to a low RRR. Thus greatest increases in RRR will not happen until precipitate coarsening processes are well underway.

While microhardness is a relatively easy-to-measure strength probe, its connection to yield strength and flow strength properties is not straightforward and at best only empirical. The final table, Table V, is a summary of the strength and RRR properties of the 0.26 wt % gold alloy for various conditions. The VHN was converted to flow stress (at 8% strain) according to

$$\sigma_{FS} = \frac{VHN}{2.4} \quad (1)$$

This is an empirical equation found by Abson et al.¹⁴ to be valid for aluminum. These workers also noted that the σ_{FS}/σ_{YS} ratio was about 1.5 and this factor was used to estimate σ_{YS} in Table V. A corresponding calculation for well-annealed aluminum (VHN = 15) gave a σ_{YS} value of 40.9 MPa. Thus, the best RRR values were obtained for strengths about 30% higher than those of zone-refined aluminum.

TABLE V

Estimates of Strength Properties of Aluminum + 0.26 wt % Gold Alloy

Condition	As-Fabricated	SHTQ	Maximum Hardness			Over-Aged	
			573K	513K	460K	573K	513K
RRR	74	61	110	310	165	1187	989
VHN	40.7	21.7	24.5	25.5	22.9	19.4	20.0
σ_{FS} (MPa)	166	88.7	100	104	93.6	79.3	81.7
σ_{YS} (MPa)	111	59.1	66.7	69.3	62.4	52.9	54.5

While these RRR values are adequate for the tubular Nb/Al conductor,^{5,9} the strength levels fall short of those sought. On the other hand, these alloys do appear to meet the design requirements of the stabilizing component of a superconducting energy storage device.⁷

SUMMARY AND CONCLUSIONS

1. It was demonstrated that zone-refined aluminum (RRR = 13,000) could be melted, cast, fabricated, heat treated, and tested without encountering measureable contamination.
2. Precipitation was monitored by micro-hardness and RRR in four aluminum-gold alloys.
3. RRR was a sensitive probe of the coalescence (coarsening) phase of precipitation in these alloys.
4. RRRs of the order of 1000 to 2500 were obtained but only after overaging to a point where the yield strength was 30% higher than the unalloyed zone-refined aluminum.

5. Under the conditions of maximum hardness where the yield strength was estimated to be 100% higher than pure aluminum the RRR of the alloys was between 100 and 300.

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