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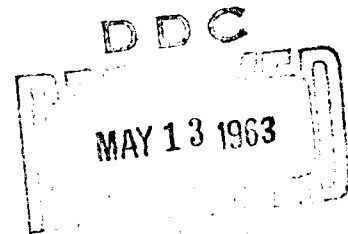
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Temperature and Rise-Time
Dependence of the Electric Strength
of Alkali Halide Crystals



(14) TR 177

Technical Report 177
Laboratory for Insulation Research
Massachusetts Institute of Technology

March, 1963

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(6) Temperature and Rise-Time Dependence of the Electric
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(15) Contract Nonr-1841(10).

(11) March, 1963

TEMPERATURE AND RISE-TIME DEPENDENCE OF THE ELECTRIC STRENGTH OF ALKALI HALIDE CRYSTALS*

by

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Abstract: Techniques were developed to extend d-c and impulse breakdown measurements to liquid-helium temperature. To avoid damage from grinding, an etching method was developed for preparing recessed alkali halide specimens.

The breakdown strength of KCl was measured from 4.2° to 425°K with d-c voltages and 1- and $10\text{-}\mu\text{sec}$ rise-time pulses. The temperature characteristic of the d-c electric strength reached a maximum of 1.16 Mv/cm at about 360°K and then dropped to 1.04 Mv/cm at 425°K . The breakdown strength was lower for d-c voltages than for pulses over the entire temperature range. For $1\text{-}\mu\text{sec}$ rise-time pulses, the maximum of the d-c temperature characteristic disappeared. Below 77°K , the breakdown strength measured with pulse and d-c voltages did not change appreciably. Changing the pulse rise time from one to ten μsec had little effect on electric strength. No thickness dependence was observed.

The electric strength of a mixed crystal (75% KCl - 25% RbCl) was measured from 77° to 375°K . The temperature characteristic differed in shape from that of pure KCl; the slope at low temperatures was smaller and the maximum shifted to about 300°K . At 375°K , the breakdown strength of the mixed crystal had dropped to 0.60 Mv/cm .

*Based on a thesis submitted in partial fulfillment of the requirements for the degree of Doctor of Philosophy in Physics at the Massachusetts Institute of Technology.

The breakdown strength of helium was measured as a function of pressure at 77°K and at room temperature. For a given pressure, the electric strength of the gas was found to increase by about a factor of 3 when cooled to 77°K.

1. Introduction

The dielectric strength of gases, liquids, and solids has been studied in the Laboratory for Insulation Research for many years. Most of the fundamental work on solids has been conducted on single crystals of the alkali halides, which are easily grown with a variety of cations and anions and allow correlations of the observed phenomena to a simple lattice structure with known defects.

The macroscopic phenomenon of breakdown can arise from a variety of molecular phenomena: impact ionization, chemical decomposition, thermal breakdown by electronic or ionic conduction, etc. The fundamental investigation of the electric strength of alkali halides by von Hippel¹⁻⁴⁾ demonstrated that the breakdown strength drops with increasing size of the ions (Fig. 1). The onset of breakdown appeared as a sudden current discontinuity (Figs. 2 and 3);⁵⁾ prior to breakdown its value is only of the order of 10^{-9} amp; hence, thermal breakdown is excluded as the primary cause of failure. In addition, von Hippel^{1, 2, 4, 6, 7)} discovered the "direction-breakdown" effect; within the

1) A. R. von Hippel, Z. Physik 75, 145 (1932).

2) A. R. von Hippel, *ibid.* 88, 352 (1939).

3) A. R. von Hippel, J. Appl. Phys. 8, 815 (1937).

4) A. R. von Hippel, Z. Elektrochem. 75, 506 (1933).

5) A. R. von Hippel, Phys. Rev. 54, 1096 (1958).

6) A. von Hippel, Z. Physik 67, 707 (1931).

7) A. von Hippel, *ibid.* 68, 309 (1931); J. W. Davisson, Phys. Rev. 70, 685 (1946); 73, 1194 (1948); M. Caspari, *ibid.* 98, 1679 (1955).

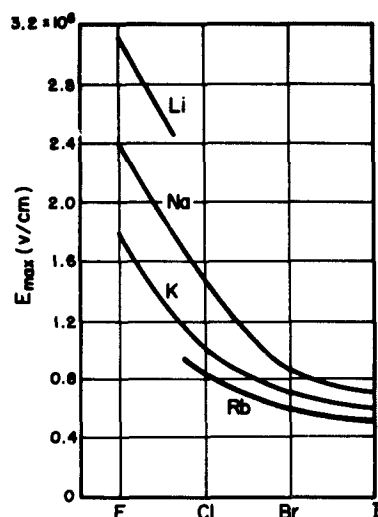


Fig. 1. Breakdown strength of alkali halide crystals. (After von Hippel. ^{1, 2})

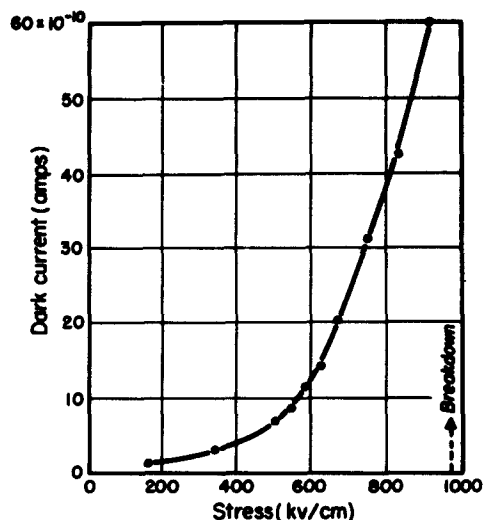


Fig. 2. Current-field-strength characteristic of the dark current in KCl. (After von Hippel. ⁵)

crystal volume as well as on the surface all breakdown paths follow preferential crystallographic directions that depend systematically on polarity, field strength, temperature, and addition agents, if secondary destruction is avoided.

The breakdown strength of NaCl rises in the lower temperature regions and the d-c breakdown-strength maximum moves to lower temperatures when some of the cations are replaced by silver ions⁸⁾ (Fig. 4). In mixed crystals at constant temperature the breakdown strength traverses a maximum as a

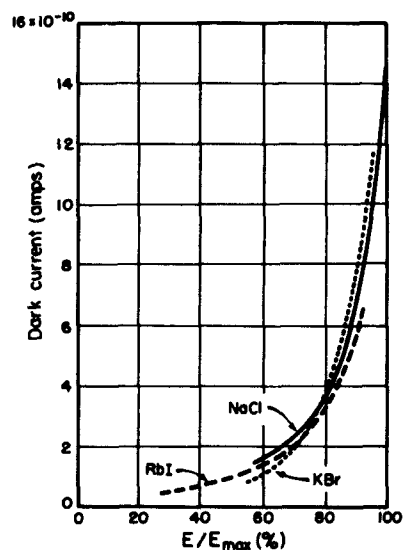


Fig. 3. Dark current in NaCl, KBr, and RbI on a reduced field scale. (After von Hippel. ⁵)

8) A. von Hippel and G. M. Lee, Phys. Rev. 59, 824 (1941).

function of composition³⁾ (Fig. 5). This behavior is very much like the dependence of ρ on composition for disordered binary alloys^{9,10)} (Fig. 6).

Earlier ideas on breakdown in solids focused on the ions as the primary charge carriers responsible for failure. But in the light of experimental facts, von Hippel concluded that electron-impact ionization

and avalanche formation were the ultimate causes of breakdown in solid dielectrics,^{1, 3, 6, 7)} and that excitation of vibrations represents the main friction barrier, in contrast to that of quantum transitions in the electronic system for gas breakdown.

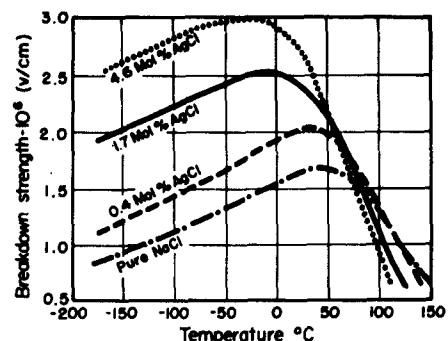


Fig. 4. Breakdown strength vs. temperature for mixed crystals of NaCl-AgCl. (After von Hippel and Lee.⁸⁾)

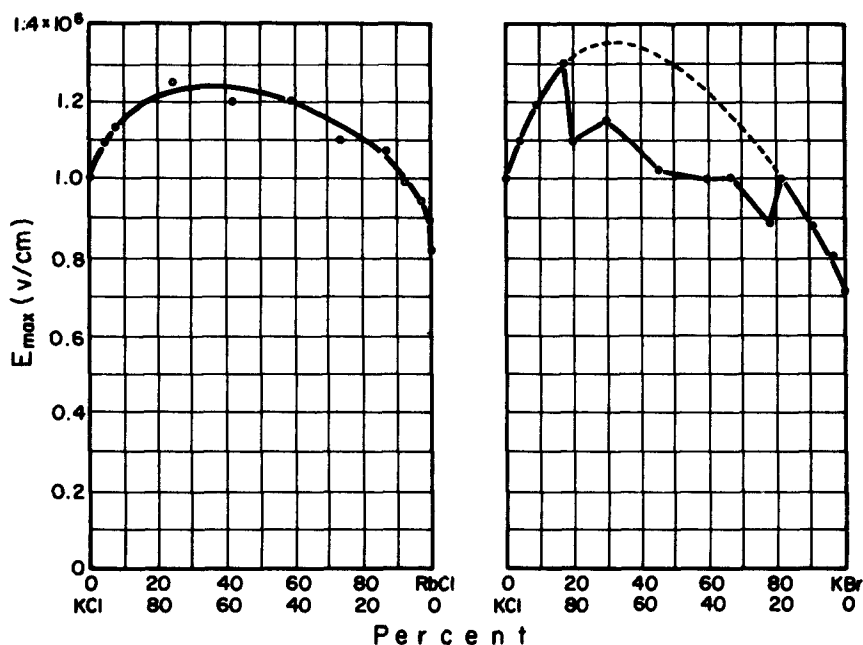


Fig. 5. Breakdown strength as a function of composition. (After von Hippel.⁸⁾)

9) C. H. Johansson and J. O. Linde, Ann. Phys. 25, 1 (1936).

10) J. M. Ziman, "Electrons and Phonons," Clarendon Press, Oxford, 1960.

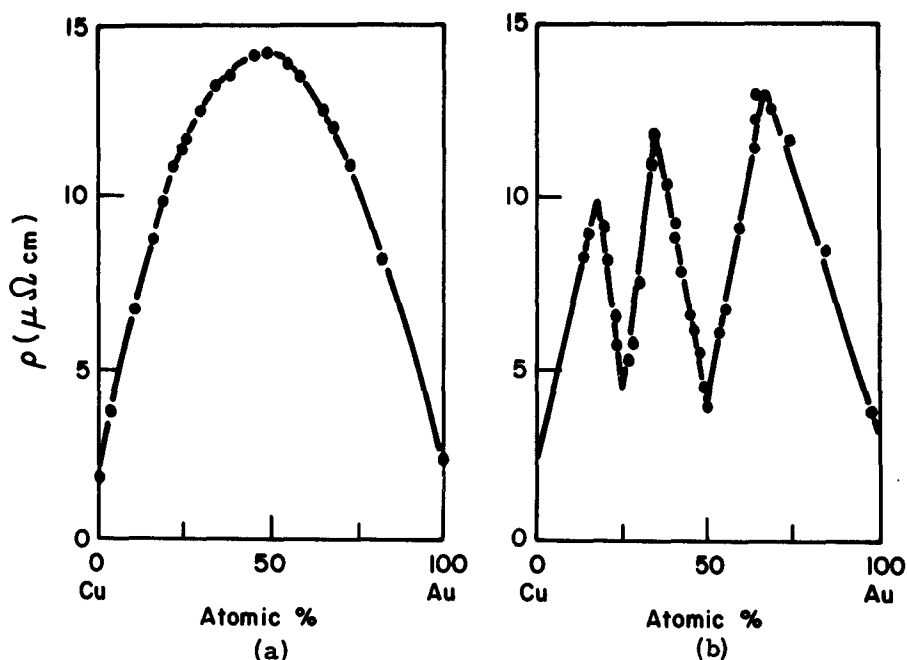


Fig. 6. Resistivity vs. composition (a) for a disordered binary alloy, (b) for a binary alloy with ordered phases. (After Johansson and Linde⁹⁾ and Ziman.¹⁰⁾)

As a qualitative description of the friction-barrier process we can think of an electron in the conduction band of a solid being accelerated by an applied electric field and at the same time losing energy through the excitation of lattice vibrations. Since a slow electron has not enough energy and a fast one not enough time to excite a lattice vibration, a plot of energy loss versus energy traverses a maximum, as does the applied field necessary to accelerate an electron of a given energy. It is this maximum field region which von Hippel^{1, 6)} recognized as the breakdown strength of the material since from here on an electron can be accelerated, produce impact ionization, avalanche formation, and destruction of the insulator. Since the number of lattice vibrations increases with temperature, electrons become more effectively scattered; thus the breakdown strength exhibits a positive temperature coefficient. The admixture of foreign ions into the crystal lattice also increases

electron scattering, thereby increasing the breakdown strength of mixed crystals.

The occurrence of a maximum in the d-c breakdown-strength temperature characteristic posed a problem. It lies at a temperature where the thermal conductivity of alkali halide single crystals is still high, probably excluding thermal breakdown as a cause. Von Hippel and Alger¹¹⁾ showed that when impulse voltages were used instead of d-c, this temperature dependence flattened out and the maximum disappeared (cf. Fig. 7). They suggested that the buildup of a cathode fall and the preparation of the cathode for electron emission is favored by time and temperature; hence, the temperature dependence might be caused by the condition of the cathode.

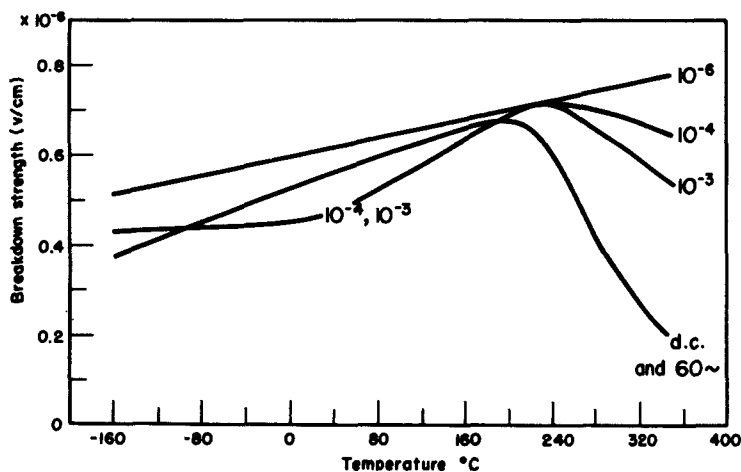


Fig. 7.

Breakdown strength of KBr vs. temperature for various rise times. (After von Hippel and Alger. 11))

The behavior of the breakdown strength of a solid at very low temperatures, where the density of lattice vibrations is very low, is of fundamental importance to our understanding of breakdown. If electron scattering by phonons is primarily responsible for retarding the accelerating action of the applied field, one might expect a low value for the breakdown strength when the lattice vibrations have been put to rest at liquid-helium temperature. If, on the other hand, the distortion of the lattice by the electron's electric field

11) A. von Hippel and R. S. Alger, Phys. Rev. 76, 127 (1949).

is decisive in exciting lattice vibrations, then the breakdown strength at 4.2°K will probably not differ appreciably from its value at liquid-nitrogen temperature. Does the maximum in the d-c breakdown characteristic disappear under rapid pulse conditions as found by von Hippel and Alger, or does it remain, as found by others?^{12, 13)} The present research was undertaken to clarify these questions and to develop improved techniques to make the liquid-helium region accessible to such measurements.

2. Experimental Techniques

A. Sample Preparation

A new method was developed for preparing cavity-type specimens free of the mechanical defects introduced by the more conventional techniques of grinding and polishing. The samples had to be properly shaped and thin enough to allow breakdown measurements in an ambient medium of low electric strength.

To provide a homogeneous field for breakdown measurements, alkali halide samples are normally prepared either as flat plates or as recessed samples (with cavities on one or both sides). The plate samples are made by cleaving blanks from a crystal boule and then reducing the thickness by grinding, lapping, and polishing. In this technique, surfaces are easily damaged by scratches and microcracks and the volume impaired by the formation of dislocations. For instance, sliding a KCl slab on a paper for a short distance with a light load proves sufficient to generate dislocations to a depth of 100 μ .¹⁴⁾ Since a sample's thickness in our case must be < 500 μ to allow measurements at reasonable voltages, a normal polishing

12) J. H. Calderwood and R. Cooper, Proc. Phys. Soc. (London) B66, 74 (1955).

13) V. D. Kuchin, Sov. Phys. - Solid State 1, 406 (1954).

14) M. Sakamoto and S. Koboyeshi, J. Phys. Soc. Japan 13, 1231 (1958).

procedure would undoubtedly affect the major part of the volume under investigation.

Plane-parallel samples with evaporated electrodes require an ambient medium of high strength to suppress surface discharges. Edge breakdown may take place unless diffuse electrodes reduce the field concentration properly. With cavity-type samples this difficulty is avoided: the high-field region is localized at the cavity bottom and the edge rendered harmless by increasing sample thickness. The cavity configuration therefore can tolerate an imbedding medium of relatively low strength.

The manufacture of fault-free samples is difficult. The standard approach of drilling, grinding, and final polishing shows little respect for the integrity of the crystal structure. To avoid this, cavities were etched in KCl with water to the desired sample thickness.

KCl plates (0.5 mm x 1 cm x 1 cm) were cleaved from an annealed crystal boule grown in this laboratory. After heating these plates for several hours at 300°C, circular (5-mm diam.) gold anode electrodes were applied by evaporation in high vacuum, after a glow discharge had pre-cleaned the surface. The sample blanks were next attached with paraffin to glass slides (anode electrode down); circular dots (1/8-inch diam.), punched out of electrical tape, were fastened to the upper surface, properly centered and the samples covered with clear lacquer. Removal of the tape left the sample water-proofed except for this circular region.

Etching proceeded by squirting water in a smooth flow from a medicine dropper. Etching time was calibrated against cavity depth in pre-experiments. When about the proper depth had been reached, the crystal was washed with anhydrous ether and dried in a stream of nitrogen. The thickness at the cavity center was premeasured by placing a tiny steel ball of known diameter

in the cavity and microscopically determining the distance between it and the gold electrode at the lower face. By calibrating the sample thickness against the amount of water applied, the efficiency of this preparation process could be kept to 90%. Sample thicknesses from 10 to 100 μ were easily attained with this method.

The washing step to avoid recrystallization on the cavity surface had to be modified somewhat in preparing samples of KCl-RbCl, because ether alone did not prevent surface fogging. Here methanol, which also acted as a chemical polish, proved effective. For the mixed crystal a different calibration was required since its solubility is appreciably greater than that of pure KCl. Figure 8 shows a sample during etching and the final cavity shape.

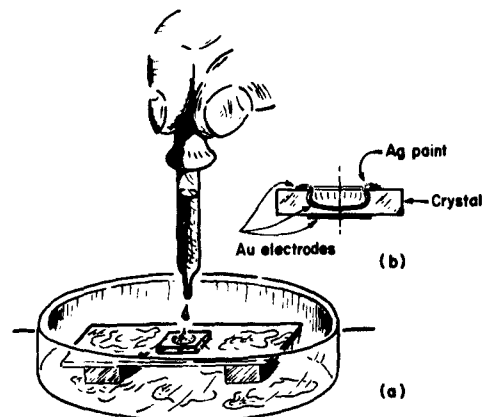


Fig. 8. (a) Sample preparation, (b) final shape of the sample.

The samples were then dried for several hours at 250°C and gold electrodes evaporated as before (Fig. 9). Since the dissolving process left an

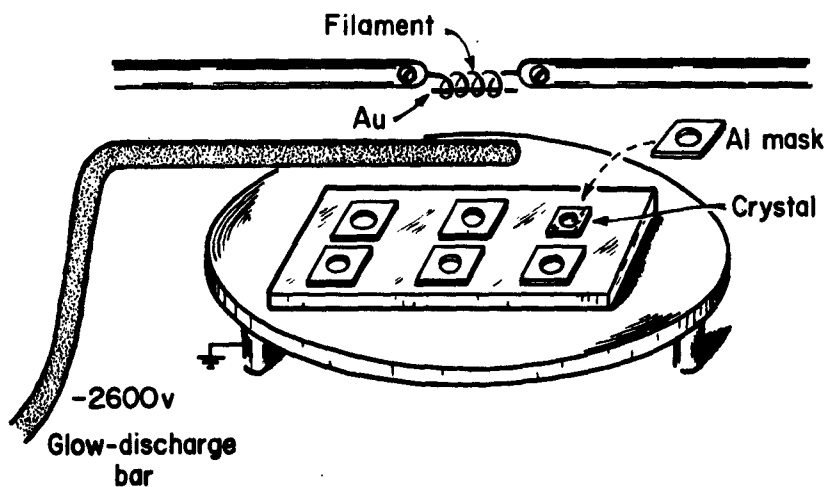


Fig. 9.

Glow discharge and evaporation setup.

undercut around the cavity rim tending to interrupt electrical contact between the gold films on the sample top and in the cavity, some silver paint or Eccobond 56C was applied at this danger zone. Contact with the sample was made outside the cavity to avoid damaging the critical area.

B. Sample Holder

Since one sample holder could not be used for measurements over the entire temperature range under investigation, modifications of a basic design were made to extend our measurements to 4.2°K . Above 77°K , pressurized nitrogen, or in some cases liquid insulators, could be used as an ambient medium to suppress surface discharges. Liquid nitrogen or helium proved unserviceable because the samples shattered during breakdown, making a

- post-mortem examination of breakdown path and sample thickness impossible.

Helium gas as imbedding medium avoided this difficulty but its electric strength is much lower than that of nitrogen. Hence the samples had to be very thin and the sample holder had to be designed for minimum helium consumption and freedom of predischarges.

The standard coaxial sample holder and pressure vessel for pulse and d-c measurements used from 150° to 425°K is shown in Fig. 10. The outer Monel shell forms a pressure vessel holding 50 atm of N_2 , the ambient medium in this temperature range. A pressure-tight seal is formed by two aluminum gaskets when the two sections are joined with hardened steel screws. The dielectric of the sample holder and pressure vessel is made of filled Teflon (Roger's Duroid 5650) to reduce shrinkage at low temperatures and extrusion under gas pressure.

The coaxial holder has an impedance of 50 ohms and is constructed of thin-walled stainless steel to minimize thermal conduction; it fits the

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upper half of the pressure vessel under spring tension. Good thermal contact is established with a thermostated copper block and removal from the conical half of the pressure bomb is facilitated.

An open-spring electrode is used for establishing contact to the cathode of the specimen, since it cannot freeze and its flexing counteracts center-conductor displacement arising from dielectric shrinkage. By passing liquid nitrogen through the copper block on which the holder base rests, the sample temperature can be adjusted between 110° and 300°K through proper flow-rate control. The nitrogen flow rate was varied by a needle valve in series with the vacuum line. Sample temperature was determined within $\pm 2^{\circ}\text{K}$ by a copper-constantan thermocouple. Heat conduction through the pressurized gas and supporting structure set a low-temperature limit of about 110°K . To reach temperatures above room temperature, a core containing an electric heater was installed in place of the cooling unit. Sample temperature could then be adjusted by Variac control of the heating current.

Neither liquid nitrogen nor liquid helium could be used as ambients at 77° and 4.2°K because samples shattered in the liquids; a gaseous ambient was required. To evaluate the feasibility of helium as an ambient

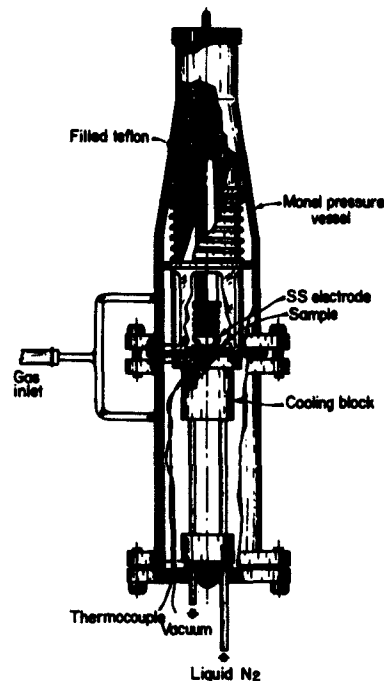


Fig. 10. Pressure vessel and sample holder.

at 77°K and below, the sample-holder and bomb assembly of Fig. 10 was modified by replacing the bottom half with a 3/8-inch thick aluminum (2024-T4) plate; the sample holder was inserted into the conical pressure vessel as before (Fig. 11). The problem of containing helium at 77°K was solved by a leak-tight seal formed by Mylar (0.005-inch thick), compressed by six screws of 304 stainless steel. Gas leakage due to shrinkage of the dielectric at the top of the pressure vessel was prevented by a tapered connector constructed of Stycast 2850GT resin and Teflon.

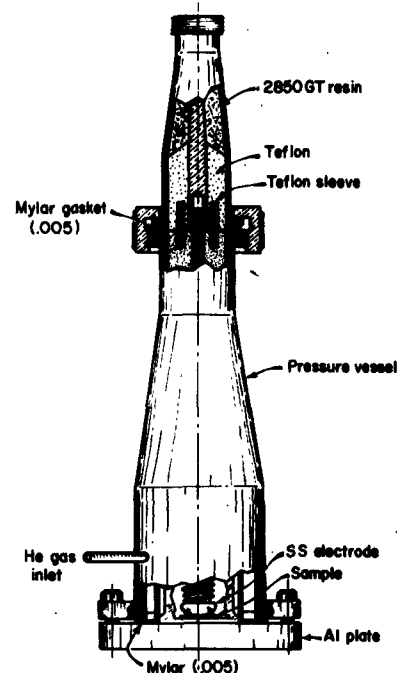


Fig. 11. Modified pressure vessel and sample holder.

After the bomb, filled with pressurized helium, proved gas-tight, the electric strength of helium as a function of pressure was determined at room temperature and at 77°K to establish its usefulness as an ambient medium below liquid-nitrogen temperature.

A gap was formed by a 0.055-cm thick slice of capillary tubing. The results of Fig. 12 show that the breakdown strength of helium at 77°K is about three times the value obtained at room temperature, illustrating a density effect. Helium at 300 psi and 77°K could therefore serve as an ambient medium at this temperature, provided the field strength in the gas space could be kept sufficiently low. This requirement was satisfied by the cavity-type specimen shape. To evaluate the possibility of using helium

vapor as an ambient at 4.2°K , its electric strength was measured by adapting the holder of Fig. 10 as indicated in Fig. 13. The external coil spring was removed, a copper block screwed on to increase the thermal mass and hold the thermocouple, and the holder attached to a tapered connector by soldering two strips of copper sheet to the holder and connector. Thus the over-all

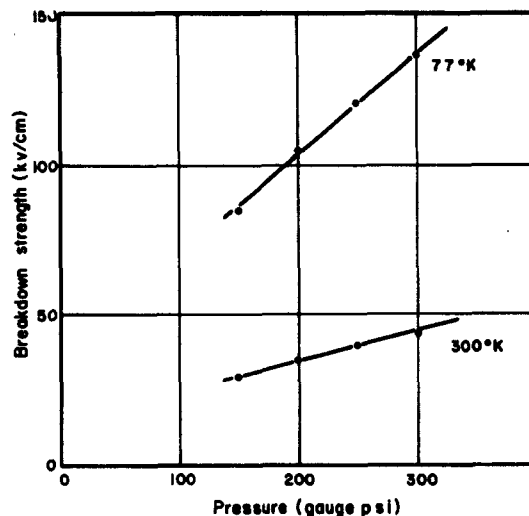


Fig. 12. Breakdown strength of helium vs. pressure at 77°K and 300°K .

diameter was trimmed to 1 inch allowing insertion of the array into the helium Dewar with enough clearance to avoid freezing-in. The breakdown strength of helium vapor at 1 atm and 4.2°K was found to be 120 kv/cm. Helium vapor would therefore serve as an acceptable ambient medium at 4°K provided the electric-field intensity was kept sufficiently low.

The breakdown experiments on KCl were carried out in helium vapor with the modified sample holder (Fig. 13) and with cavity-type samples to keep the field concentration low. The samples did not shatter as in a liquid-helium ambient. This arrangement was therefore used for pulse and d-c measurements below 77°K . By avoiding liquid-helium transfer into a separate cryostat, its consumption was minimized. Holder and cable design (cf. p. 15) was so efficient that the rate of helium evaporation during successive measurements exceeded the normal boil-off rate by less than a factor of two.

Below 77°K , sample temperature was measured with a copper-constantan thermocouple in the copper block at the sample holder base; liquid helium

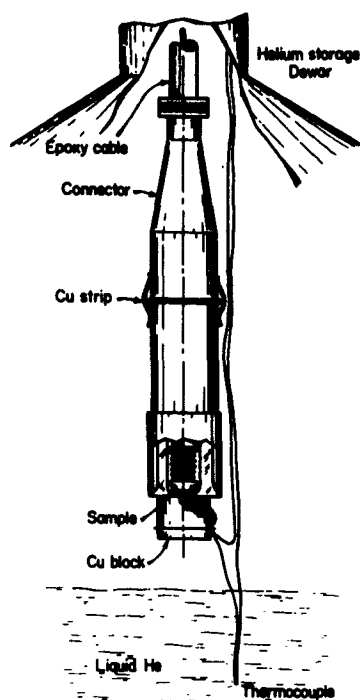


Fig. 13. Sample holder for liquid-helium temperature.

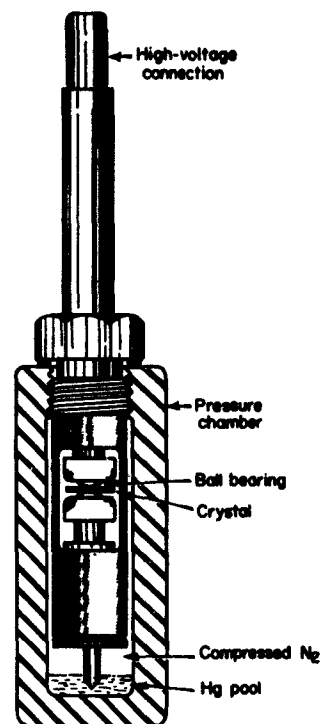


Fig. 14. Room-temperature sample holder.

served as reference bath. Since the thermocouple sensitivity below liquid-nitrogen temperature is low, a K-3 potentiometer (Leeds and Northrup) was used for the emf measurement (error $\pm 0.5^{\circ}\text{K}$).

The sample configuration and holder design effectively avoided pre-discharges in the gas medium surrounding the sample so that measurements in nitrogen at atmospheric pressure could be made at 77°K . Here slow cooling was provided by replacing the copper block in Fig. 13 with a copper cone and cooling the holder by slowly immersing the cone in liquid nitrogen. This adaptor was not required for measurements below 77°K because the temperature gradient along the neck of the helium storage vessel proved adequate for slow cooling.

At room temperature, breakdown measurements with d-c voltages were carried out with the holder of Fig. 14. The sample holder is attached to a spark plug-type nut system with the high-voltage lead passing through a long cylindrical styrene insulator. Electrical contact is made by two highly polished Monel balls, one of which is set into a lightly sprung plunger, the other in a stationary piece connected to the high-voltage lead. The lower spring-loaded electrode dips into mercury, establishing contact to ground. The outer cylindrical vessel forms a pressure chamber to hold nitrogen up to 50 atm. A pressure-tight seal is obtained by screwing the nut of the sample holder onto a Teflon gasket.

C. High-Voltage Coaxial Cable

A coaxial cable of about three feet was required to conduct the pulse and d-c voltages into the helium Dewar to the sample. Commercial cables did not qualify. A 35-inch length of RG87A/U cable (Teflon insulation) on cooling to 4.2°K showed so much shrinkage that the high-voltage insulation at the sample holder pulled away. Furthermore, the cable's center conductor was pulled back into its own insulation on repeated cycling to low temperature, thus ruining electrical contact. Teflon is excellent for low-temperature operations because it resists cracking on sudden immersion in liquid nitrogen or helium and retains some ductility at 4.2°K. However, its large thermal coefficient of expansion and its thermal memory make it unsuitable for situations demanding dimensional stability at low temperatures.

A connector and cable design compensating for the thermal effects was attempted. A cable was constructed with an outer conductor of thin-walled (0.007 inch thick) stainless steel tubing, to keep helium loss low, and with the dielectric and center conductor core of the RG87A/U cable

(cf. Fig. 15 for slip-joint design). Bonding of Teflon to itself and to steel was achieved by treating the surfaces with "Fluorobond" (Joclin Mfg. Co.) and then cementing with epoxy resins. The cable behaved satisfactorily at first but later developed electrical leakage at the Teflon-Teflon bond in the low-temperature section. It was therefore replaced by a new design.

A coaxial line was constructed with stainless steel tubing as outer conductor, copper wire as center conductor,

and silica-filled epoxy resin as the dielectric. The resin (Stycast 2850GT, Emerson and Cuming Co., Canton, Mass.), an adaptation of the "poly-glasses" developed during World War II in the Laboratory for Insulation Research,¹⁵⁾ has two outstanding mechanical properties for low-temperature applications: a thermal coefficient of expansion about equal to that of stainless steel (15×10^{-6} in/in/ $^{\circ}$ C) and a remarkable resistance to cracking when subjected to severe thermal shock. Since it adheres very well to metals, it can also be used for low-temperature metal-to-metal bonds.

The uncured resin-catalyst mixture has a serious drawback for high-voltage insulation: its high viscosity makes production of void-free castings very difficult. This problem was solved by injecting the specially prepared resin into tubing and curing under pressure.

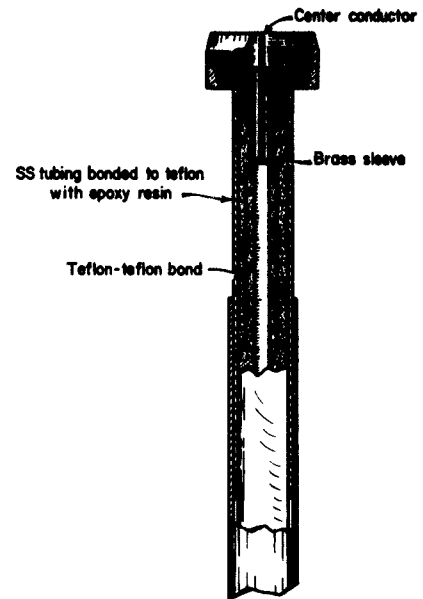


Fig. 15. Slip-joint design of Teflon cable.

15) A. von Hippel, S. M. Kingsbury, and L. G. Wesson, N. D. R. C. Report No. X, Lab. Ins. Res., Mass. Inst. Tech., October, 1945.

The cable was constructed in two sections, each ca. 16 inches long, because casting in one piece could require spacers for holding the center conductor in position, which would interfere with the casting process and introduce voids. Stainless steel tubing (Type 304, 3/8-inch o.d.) served as the outer conductor. A 3-inch piece of copper tubing equipped with a flare fitting was soldered into a hole of the stainless wall to serve as the inlet tube for the dielectric. A copper wire (0.055-inch diam.) soldered to a centering plug at the lower end of the tube was stretched taut and clamped at a centering plug at the other end under spring tension to keep it concentric at elevated temperature. The inside of the assembled cable shell was thoroughly cleaned with distilled water and then dried. A small hole at the top of the tube served as vent. A tubular oven was then slipped over the empty cable and the section heated to 100°C (Fig. 16).

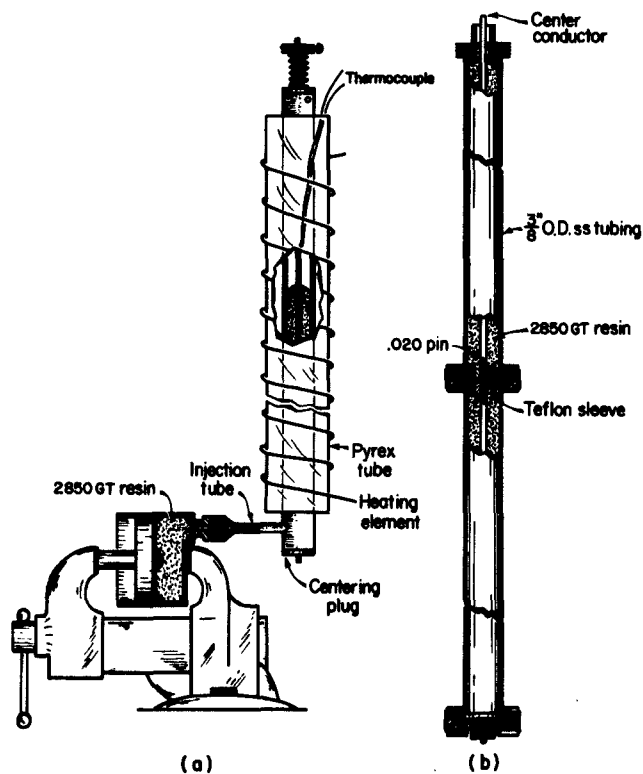


Fig. 16.

Apparatus for filling epoxy cable (a) and assembled cable (b).

The injection cylinder which was later attached to the copper tubing contained a measured quantity of resin heated to 90°C. After carefully adding catalyst No. 11 (4% by weight) to avoid bubble formation, the hot cylinder and its contents were placed in a desiccator and vacuum was applied for ten minutes to remove bubbles. With piston inserted, the cylinder was mounted in a vise, connected to the copper tube leading into the cable shell, and the resin forced in very slowly to avoid turbulence (cf. Fig. 16). This process was kept up until resin exuded at the vent. The vent was then sealed, the vise tightened, and the remainder cured under pressure for several hours at 100°C.

After curing, a length of about $1\frac{1}{2}$ inches was carefully machined off each end to remove the centering plugs and the section containing voids formed during injection. The cured resin is not machineable with the usual tools; the rough turning was therefore done with Carbaloy and the final finishing with a diamond cutting wheel. The second section was made in the same way. Circular grooves at both ends of each piece were cut with a diamond-core drill, and the connectors attached with the same resin to avoid destroying the insulator by the heat of soldering.

Both cable sections were locked together to give the desired length, with electrical continuity along the center conductor maintained by a 0.020-inch phosphor bronze pin split at both ends, fitting into holes drilled in both wires. The high-voltage insulation at the joint was formed by a Teflon sleeve fitting into the circular grooves. This design provided a path of sufficient length to avoid breakdown at the joint. Figure 16 shows the finished product.

This two-part cable proved satisfactory up to 20 kv at low temperatures and did not shrink or show hysteresis effects. Its low thermal con-

ductivity meant that liquid helium was not wasted. It proved trouble-free after many cyclings between room temperature and 4.2°K .

D. Pulse and D-C High-Voltage Equipment and Measurement Procedure

The general layout for pulse-breakdown measurements is shown in Fig. 17. The pulse generator designed by Schissler¹⁶⁾ was modified to improve accuracy. This system can produce negative pulses of peak amplitudes of 20 kv of linear rise, and rise times adjustable between 100 and $0.2\ \mu\text{sec}$. A push button triggers a generator which produces two pulses, one initiating the oscilloscope sweep, the other starting the high-voltage pulse generator after a short delay to provide a base line for the oscilloscope trace. The pulse from the generator is applied to the sample; breakdown occurs along the linearly rising voltage ramp. The pulse applied to the specimen is sampled by a voltage divider and displayed on the oscilloscope. The photographed trace shows the breakdown voltage and the time taken to reach this value (maximum error ca. $\pm 5\%$ in voltage measurement and ca. $\pm 10\%$ in time indication).

After breakdown the sample is examined microscopically to ascertain that the breakdown spot is in the center section, and the sample thickness is determined by cleaving the sample at the breakdown hole and then measuring its edge at the hole under a Leitz metallographic microscope equipped with a filor micrometer eyepiece. The optical system is then calibrated with a

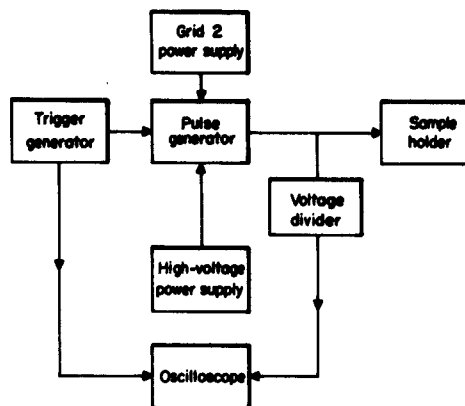


Fig. 17. Block diagram for pulse measurements.

16) L. R. Schissler, Tech. Rep. 153, Lab. Ins. Res., Mass. Inst. Tech., September, 1960.

stage micrometer slide (American Optical Co.) divided into units of $10\ \mu$. This procedure gave an error of less than $\pm 3\%$.

The accuracy of a pulse measurement obviously depends on the performance of the oscilloscope and voltage divider. A Tektronix Type 541A oscilloscope with a Type K preamplifier with a combined rise time of 12 millimicroseconds proved adequate. The ultrahigh-speed transients associated with electric breakdown did not couple to its vertical amplifier; thus the displayed wave form was not distorted and accurate breakdown-voltage measurement was possible. The pulse from the divider in the pulse generator was sampled with a high-performance Tektronix P6000 probe (attenuation 10:1). A preamplifier-sensitivity setting of 5 or 2 volts/cm proved sufficient in the voltage range used.

Since 20-kv resistors having good high-frequency characteristics apparently do not exist, a pure capacitive voltage divider was used in the pulse generator, instead of a capacitance-compensated resistive divider, to examine the high-voltage pulses applied to the specimen. A short polyethylene coaxial cable (RG11/U) served as the capacitance C_1 ; it withstood potentials up to 20 kv without developing corona. A 1000-picofarad mica capacitor for C_2 provided a voltage divider ratio of 94:1 (Fig. 20, p. 23). The response of this divider to a pulse having a rise time of ca. 5 nanosec, produced by a mercury-relay pulse generator, proved excellent. Its accuracy under operating conditions was checked by measuring pulses up to 10 kv with a Tektronix P6013 probe. The agreement was well within the accuracy limits of the oscilloscope. The oscilloscope plus divider gave an accuracy of $\pm 5\%$ for impulse-voltage measurements.

The trigger generator (Figs. 18, 19) is put into operation by a push button energizing a relay. On closing, this relay applies 550 volts through

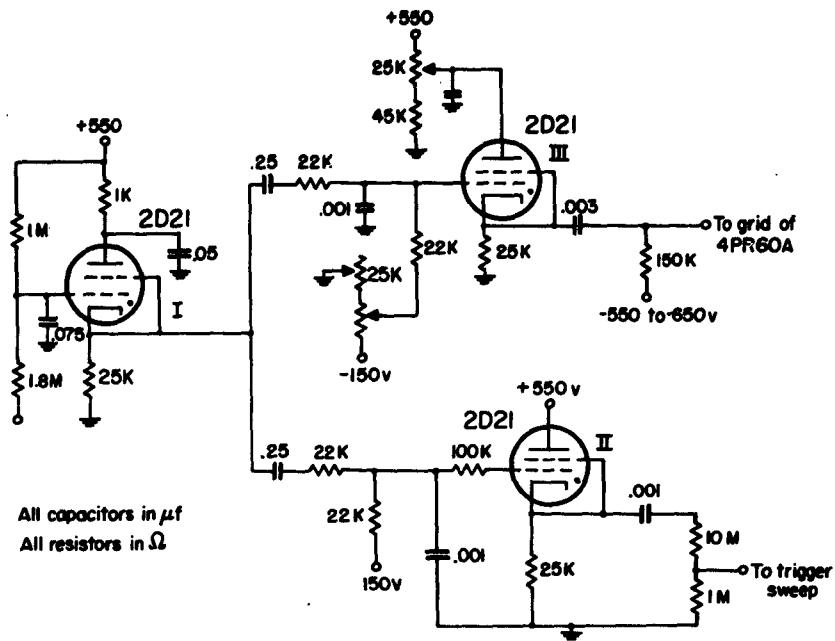


Fig. 18. Trigger-generator circuit.

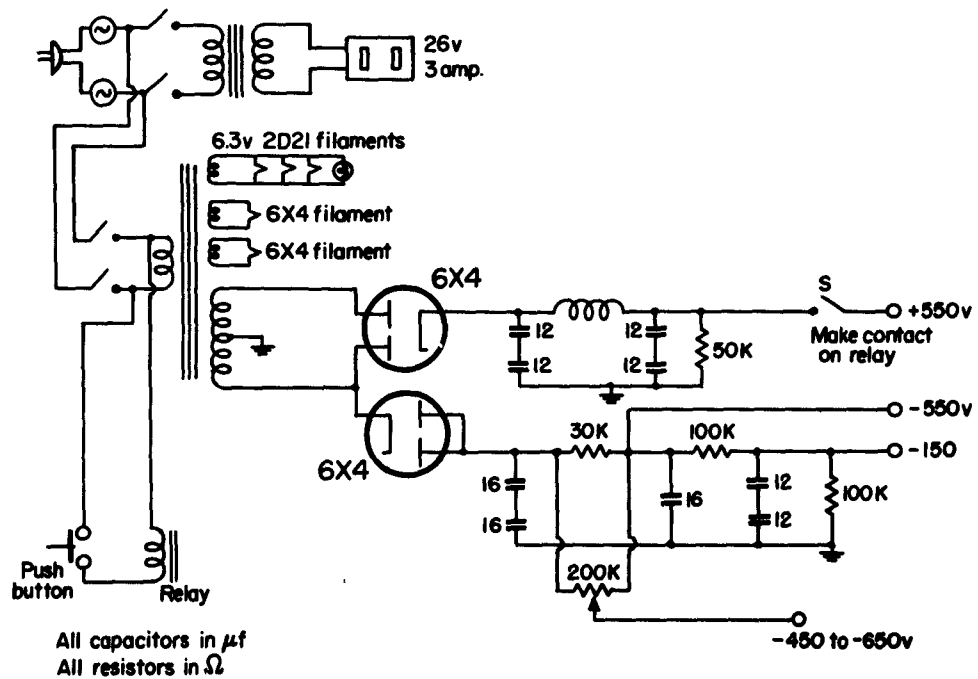


Fig. 19. Power supply of trigger generator.

contact S to the plates of three 2D21 thyratrons whose control grids are initially biased at -150 volts. Thyatron I fires after approximately 10 msec, allowing time for the relay to close and produces a rapidly rising positive pulse across its cathode resistor. This pulse is capacitatively coupled to the grids of thyratrons II and III. Thyatron II produces a pulse that activates the horizontal oscilloscope sweep and thyatron III, a pulse to trigger the tetrode of the high-voltage pulse generator. A variable time delay between the output pulses of II and III is introduced by a potentiometer adjustment of the grid bias of III. (Time constants and bias voltages are so selected that thyatron II fires at a suitable time before III to provide the base line on the oscilloscope trace.) Filament current and voltages required by the trigger generator are furnished by the power supply of Fig. 19.

The high-voltage pulse generator (Fig. 20) uses a 4PR60A tetrode as a constant current source to discharge capacitance C. This produces a linear voltage decrease across the capacitor with time; thus a negative pulse is generated at the output. After reaching its peak value it decays exponentially to zero with a time constant determined by the output circuit. The rise time and shape of the high-voltage pulse can be adjusted by varying the operating parameters of the pulse tetrode. Thus the rise time of the generator can be lengthened either by drawing less current through the tube or by increasing the capacitance C discharged. Plate current can be varied by changing the amplitude of the positive pulse firing the tetrode or by adjusting its initial grid bias. A suitable adjustment of these parameters yields a pulse that rises linearly with time. (With $C = 5 \times 10^{-10}$ farads, a rise time of 1 μ sec was obtained; $C = 2 \times 10^{-9}$ farad gave a rise time of 10 μ sec.)

The tetrode-screen grid voltage supply (Fig. 20) provides a variable potential from 0 to 3 kv and is set at 1.25 kv. A source supplying 26 volts

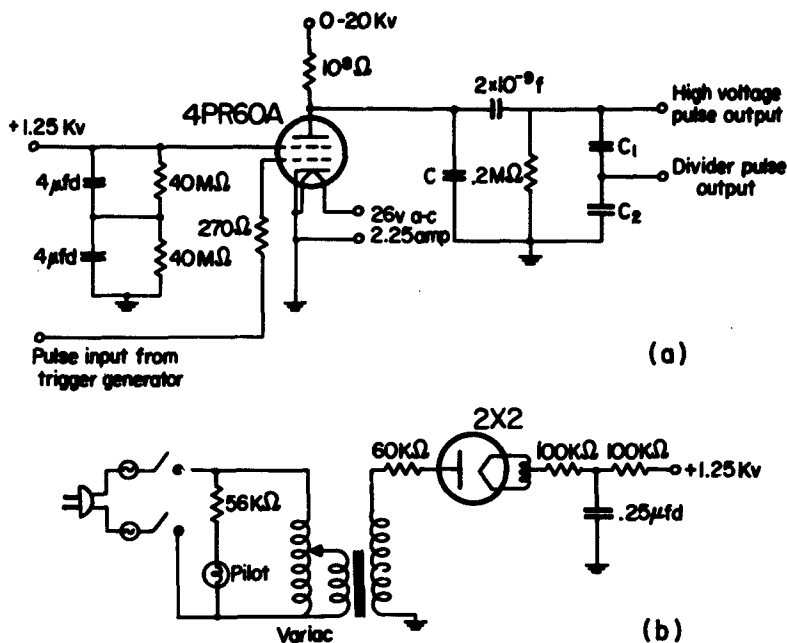


Fig. 20. (a) High-voltage pulse generator, (b) tetrode-screen grid power supply.

at 3 amp furnishes filament power to the tube. Plate voltage was applied through a high resistance from a stabilized well-filtered source which can deliver 2 milliamperes between 0 and 30 kv.

Voltage for d-c breakdown measurements was applied to a specimen by turning up the high-voltage power supply (Fig. 21). (The high-voltage power pack used for d-c breakdown and to supply tetrode plate voltage was the same.) The 10^{10} -ohm resistor in series with the sample limits the current to keep sample destruction on breakdown to a minimum. The 2×10^8 -ohm resistor, which damps oscillation of the kilovoltmeter, does not affect the voltage reading since the electrostatic meter has a resistance $> 10^{14}$ ohm. The voltage at which breakdown takes place is indicated by an abrupt drop of the meter reading. The absolute accuracy of the meter is $\pm 1\%$ of full scale and can be read to the same limits of error in measurement.

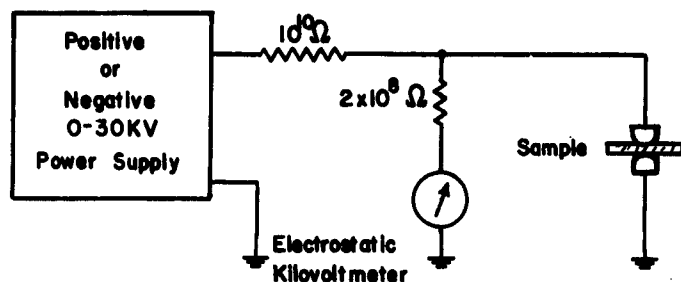


Fig. 21.

D-c measuring circuit.

3. Results

During the development of the techniques for measuring breakdown strength below 77°K , the temperature and pressure dependence of the electric strength of helium gas was determined (cf. Sec. 2, B, p. 11). The main concern of the present investigation, however, is the electric strength of alkali halide crystals as function of temperature and rise time.

A. D-C Breakdown Strength

The d-c strength of KCl single crystals in the $[100]$ direction was measured over a temperature range from 4.2° to 425°K . The open-spring electrode contact was used in all cases. Measurements on KCl at liquid-nitrogen temperature served to establish the best methods for electrode application and to check the over-all performance of the equipment. An average breakdown strength of $0.62 \pm 0.04 \text{ Mv/cm}$ was found for samples whose electrodes were deposited on surfaces not pre-cleaned with a glow discharge, while $0.74 \pm 0.05 \text{ Mv/cm}$ was obtained when a glow discharge was applied for 5 to 10 minutes prior to electrode deposition. The breakdown strength and the electrode-to-sample contact are improved by this surface cleaning; hence, no detrimental etching of the surface has occurred. All measurements were therefore carried out on pre-cleaned specimens with the cavity surface as cathode. Konorova's¹⁷⁾ measurements of pre-

17) E. A. Konorova, Bull. Acad. Sci. U.S.S.R., Phys. Ser., 24, 56 (1960).

breakdown currents in KBr with pulse voltages showed very noisy characteristics when carried out on specimens with untreated surfaces. This noise was sharply reduced when the surfaces were pre-cleaned by a gas-discharge prior to electrode application.

The results of d-c breakdown measurements are summarized in

Table 1 and in Fig. 22. The table gives temperatures, average breakdown strength, standard deviation, and a 90% confidence interval obtained from the Student "t" distribution,¹⁸⁾ which is applicable to a small sampling of a total population. The standard deviation, a measure of the scatter in experimental data, runs from 3% to a maximum of 9%.

As seen in Fig. 22, the electric strength of KCl does not change appreciably below 77°K. The breakdown strength at 4.2°K is nearly equal to the value at liquid-nitrogen temperature. The electric strength rises above 77°K at a rate of approximately 1.6 kv/cm per °K, then traverses a maximum at about 360°K and starts to drop. At 425°K it has fallen to a value of 1.04 Mv/cm, slightly lower than that at room temperature.

The d-c breakdown strength for a mixed crystal (75/25% KCl-RbCl) is given together with the data for pure KCl. The temperature characteristic for mixed crystals has a different shape. The electric strength is higher by about 30% at 77°K and by about 15% at room temperature; hence, the temperature coefficient in this region is smaller. The maximum is reached earlier

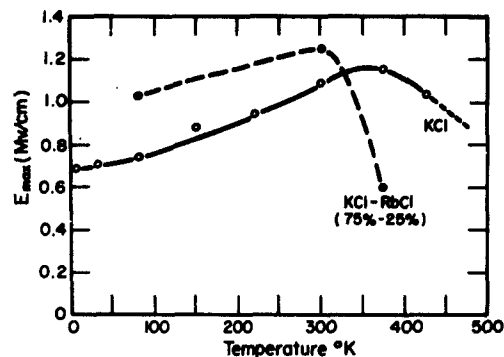


Fig. 22. D-c electric strength of KCl and 75/25% KCl-RbCl as function of temperature.

18) E. L. Crow, F. A. Davis, and M. W. Maxfield, "Statistics Manual," NAVORD Report 3369; Dover Publications, New York, 1960.

Table 1. D-c breakdown of KCl.

Temperature T°K	Average $E_{\max}$ Mv/cm	Standard deviation Mv/cm	90% confidence interval Mv/cm
4.2	0.69	0.05	0.03
30	0.71	0.05	0.03
77	0.74	0.07	0.05
150	0.88	0.04	0.03
220	0.94	0.02	0.02
300	1.09	0.07	0.04
375	1.15	0.03	0.02
425	1.04	0.05	0.04
D-c breakdown of 75/25% KCl-RbCl			
77	1.03	0.06	0.04
300	1.25	0.18	0.15
375	0.60	0.05	0.05

(at about 300°K) and the characteristic falls more steeply beyond reaching 0.60 Mv/cm at 375°K, where the electric strength of KCl is almost twice as high. This behavior agrees very well with the results on NaCl-AgCl crystals previously measured by von Hippel and Lee.⁸⁾

In Figs. 23 through 26, the d-c breakdown voltage ($V_{\max}$) and electric strength ($E_{\max}$) in KCl are plotted as function of sample thickness. Over the range covered no thickness dependence was observed. The error for d-c breakdown measurements was $< \pm 4\%$.

B. Impulse Strength

The results of impulse-strength measurements on KCl in the [100] direction for pulses of 1 and 10 μ sec rise time are shown in Fig. 27, where the

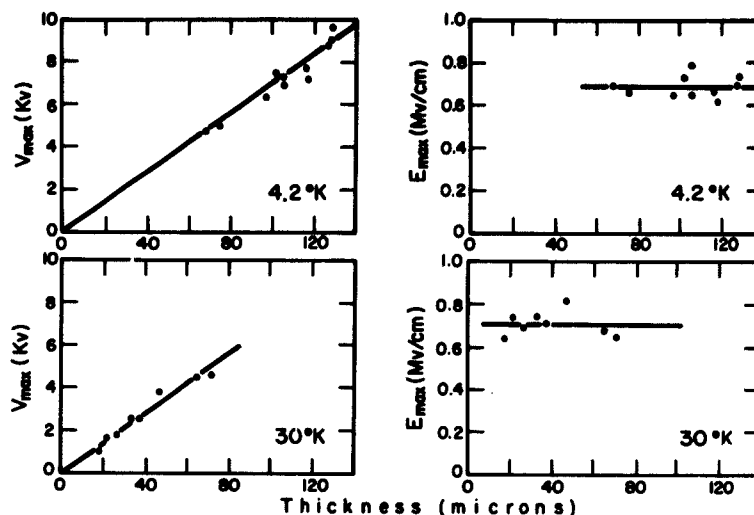


Fig. 23. V_{max} and E_{max} of KCl vs. sample thickness at 4.2° and 30°K.

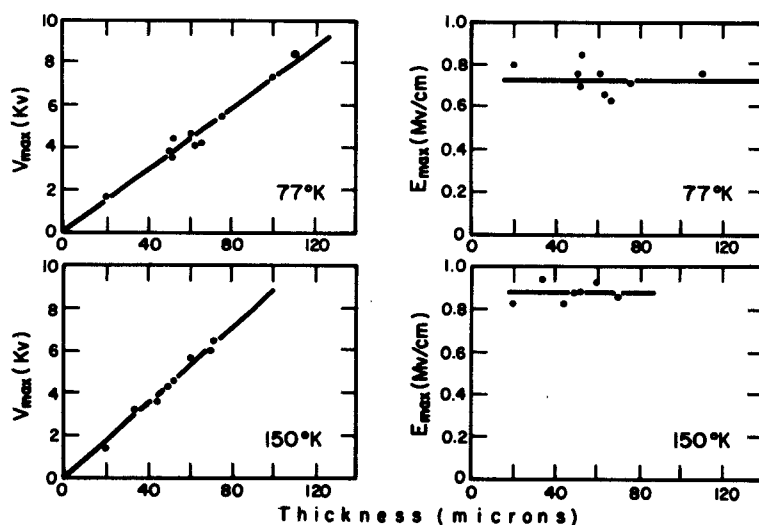


Fig. 24. V_{max} and E_{max} of KCl vs. sample thickness at 77° and 150°K.

average breakdown strength is plotted against temperature. A plot of the d-c strength is given for comparison. The particulars for impulse measurements are found in Table 2. The d-c temperature characteristic of KCl-RbCl is

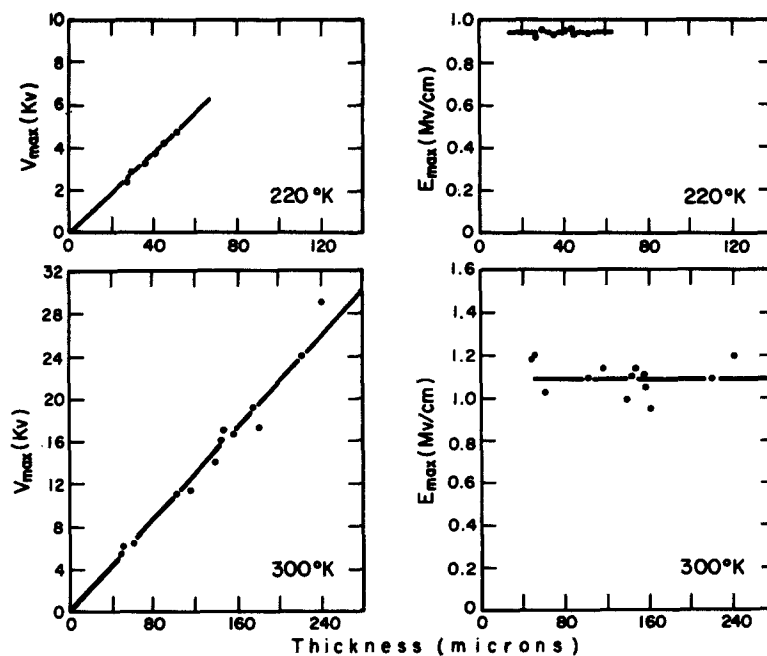


Fig. 25. $V_{\max}$ and $E_{\max}$ of KCl vs. sample thickness at 220° and 300°K.

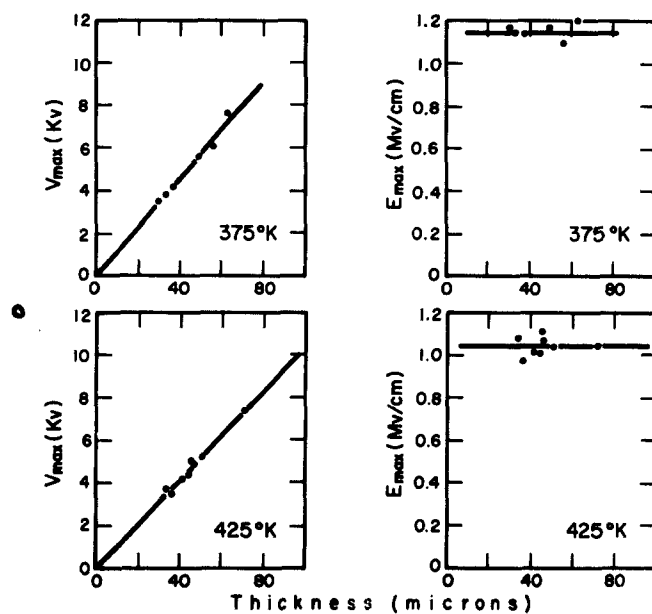


Fig. 26.

$V_{\max}$ and $E_{\max}$ vs. sample thickness at 375° and 425°K.

Table 2. 1- μ sec impulse breakdown of KCl.

T ^o K	Average breakdown strength Mv/cm	Standard deviation Mv/cm	90% confidence interval $\pm$ Mv/cm
4.2	1.15	0.12	0.08
77	1.23	0.15	0.10
150	1.27	0.17	0.11
220	1.24	0.11	0.06
300	1.37	0.12	0.08
425	1.49	0.10	0.06
10- μ sec pulse			
4.2	1.03	0.13	0.07
77	1.15	0.12	0.07
300	1.40	0.18	0.11
425	1.32	0.15	0.09
2- μ sec pulse of KCl-RbCl			
300	1.34	0.26	0.23
375	1.51	0.22	0.17

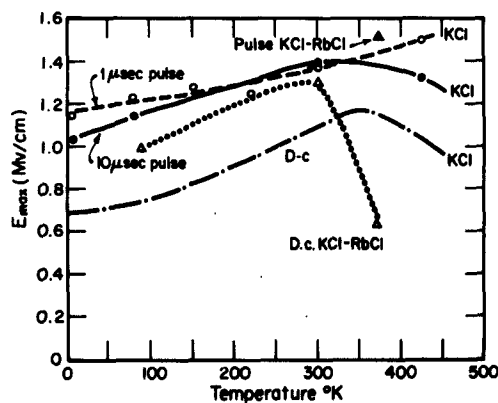


Fig. 27.

Pulse breakdown strength of KCl and 75/25% KCl-RbCl as function of temperature.

plotted in Fig. 27 together with two points for 2- μ sec rise-time pulses. The accuracy in pulse-voltage measurement was within $\pm 5\%$, so that the scatter for impulse-strength measurements was not caused by instrumentation errors. The scatter for pulse-breakdown measurements was greater than for the d-c case. The maximum standard deviation here was about 14%.

Figure 27 and Table 2 show that the impulse strength is higher than the corresponding d-c values over the whole temperature range and that the maximum disappears for 1- μ sec rise-time pulses but can still be discerned for 10- μ sec pulses. These results agree with the earlier findings of von Hippel and Alger¹¹⁾ and will be discussed later. The pulse breakdown measurements for the mixed crystal KCl-RbCl exhibit a sharp departure from the results obtained under d-c conditions. The pronounced maximum of the d-c characteristic disappears and the low value of 0.60 Mv/cm at 375°K is increased by a factor > 2 when impulses of 2- μ sec rise time are used at the same temperature.

As in the d-c case, the electric strength for 1- μ sec rise time does not change appreciably below liquid-nitrogen temperature. The temperature coefficient for 10- μ sec pulses is nearly the same as for d. c. , while for 1- μ sec pulses the temperature coefficient is smaller.

The breakdown strength of KCl versus sample thickness at different temperatures is shown in Fig. 28 for 1- μ sec and in Fig. 29 for 10- μ sec pulses. As in the d-c case, no thickness dependence was observed over the range covered.

The breakdown strength of KCl at 4.2°K and 77°K was also measured with evaporated indium cathodes instead of gold with 10- μ sec rise time, but no difference in results was discerned. Similar results were observed for 1- μ sec pulses at 77°K.

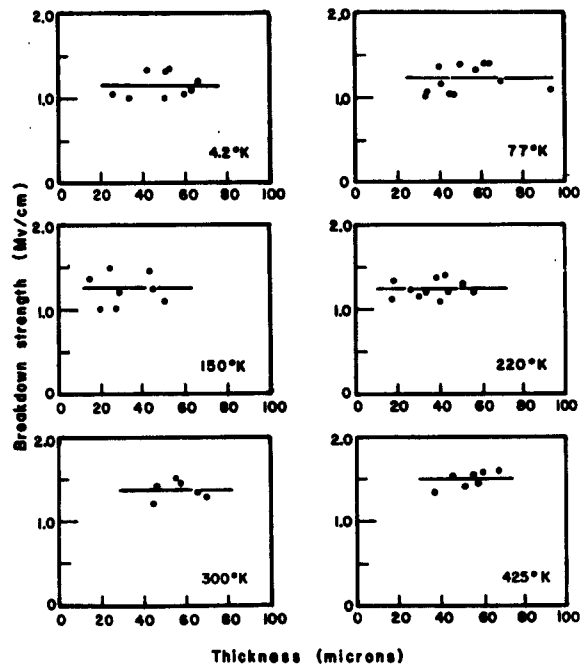


Fig. 28.

One-microsecond rise-time pulse breakdown strength of KCl versus sample thickness at different temperatures.

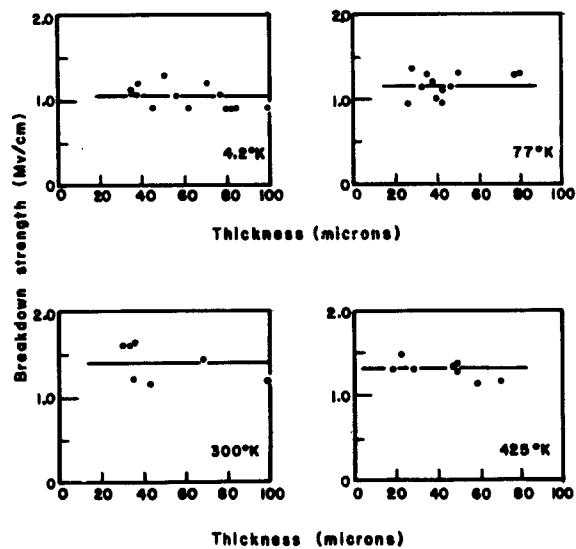


Fig. 29.

Ten-microsecond rise-time pulse breakdown strength of KCl versus sample thickness at different temperatures.

Summarizing some of our observations, we see the following:

- a) The d-c strength is smaller than the corresponding pulse strength over the entire temperature range covered.
- b) The impulse and d-c electric strengths at 4.2°K do not differ appreciably from those at 77°K.
- c) The d-c maximum vanishes when 1-μsec rise-time pulses are used for measurement.
- d) The pulse strength does not depend greatly on pulse duration.
- e) The d-c strength for KCl-RbCl is higher than for pure KCl at low temperatures but drops more sharply slightly above room temperature. Furthermore, the pulse strength of those crystals rises to 1.51 Mv/cm at 375°K while their d-c strength drops to 0.60 Mv/cm at the same temperature.

4. Discussion

The breakdown mechanism with which this research is concerned is electron-impact ionization, avalanche formation with subsequent destruction of the insulator. Von Hippel,³⁾ who discovered this effect in solids, correlated it to the vibration spectrum of the material. In his initial discussion he made the simplifying assumption that excitation of the reststrahl vibration frequency as the highest polar mode was of main importance and that the breakdown strength of an ionic crystal might be connected in a first approximation to this mode by

$$eE_{\max} \frac{a_w}{2} \sim Ch\nu_{\max},$$

where $E_{\max}$ is the breakdown strength, $a_w/2$ the interionic distance, $\nu_{\max}$ the reststrahl frequency, and e the charge of an electron. This relation is based on the assumption that a conducting electron in a polar crystal is so

strongly coupled to the lattice that it is practically in a state of constant impact. The electron was assumed to be coupled to the lattice by the field-produced distortion, which surrounds the conducting electron with a halo of countercharges. Energy transfer by vibrations results as soon as the halo structure cannot follow the electron adiabatically (polaron effect). The difference between the static and optical dielectric constants is here decisive.

Our experimental results of breakdown measurements on KCl in the lower temperature range are consistent with this picture. At 4.2°K the breakdown strength is not lowered appreciably by the quieting of thermal agitation. The halo effect remains and leads to multiphonon energy transfer when acceleration of electrons is attempted. Disordering of the lattice by thermal agitation or ion substitution increases electron scattering but does not change the breakdown strength by orders of magnitude.

The quantitative theory of this breakdown mechanism which places the breakdown strength in relation to the maximum of the vibration barrier and predicts that the acceleration of relatively slow electrons is decisive has not yet been carried through adequately. The difficulty is that perturbation theory does not suffice. Callen,¹⁹⁾ Gross,²⁰⁾ and Nettel²¹⁾ treated certain aspects of the problem.

Our research was concerned with two main problems: what happens to the electric strength at liquid-helium temperature, and how does the maximum of the d-c breakdown characteristic change under impulse con-

19) H. B. Callen, Phys. Rev. 76, 1394 (1949).

20) E. P. Gross, Tech. Rep. 55, Lab. Ins. Res., Mass. Inst. Tech., December, 1952.

21) S. J. Nettel, Phys. Rev. 121, 425 (1961).

ditions? The breakdown strength of KCl did not change appreciably at 4.2°K even though the phonon density had been reduced by a factor of 10^7 from that of 77°K. In addition, we observed a time effect: The impulse strength of KCl was much greater than the corresponding d-c values over the whole temperature range covered, in contrast to the results of Calderwood, Cooper, Grosshart et al.^{12, 22)} but in agreement with the earlier results of von Hippel and Alger.¹¹⁾

One possible explanation for this increase in electric strength with faster rise time is the existence of a partial transparency near the maximum of the vibrational friction barrier: Avalanches of a critical size might develop at a field strength lower than that of the barrier maximum if enough statistical chance is given. Passage through the friction barrier near its peak would be favored by time; the d-c strength could thus be lowered. Other possible contributions to the lowering of the d-c breakdown strength might arise from space-charge effects or preconditioning of the cathode for field emission. Field emission of electrons into the dielectric initially raises the field strength toward the anode but, as the electrons fall into traps, the field gradient at the cathode is lowered and electron injection slowed down. All such effects would increase the breakdown strength for pulses of short rise time.

Our conclusion that the maximum in the breakdown characteristic for KCl observed for d. c. vanishes for 1-μsec pulses confirms the results of von Hippel and Alger. In KCl-RbCl the maximum traversed for d-c shifts to lower temperature, and the characteristic falls more sharply. In this case again the maximum disappears under impulse conditions.

Since the breakdown current in alkali halides is about 10^{-7} amp at

22) R. Cooper, D. T. Grosshart, and A. A. Wallace, Proc. Phys. Soc. (London) B70, 169 (1957).

425°K, Joule effects are not likely to be a primary cause of the maximum in the d-c temperature characteristic. However, the ionic conductivity arising from cation migration increases exponentially with temperature, and an increase in ionic conduction could introduce field distortion in the specimen by space-charge formation. The importance of this factor was recognized by Cooper et al.²³⁾ The field at the positive electrode can be raised by an anode fall, and field emission at the cathode assisted, if the cations are not discharged. Thus field distortion can account for the maximum and the subsequent fall in d-c breakdown strength at higher temperatures. For fast-rising impulse voltages, field distortion is not given an opportunity to develop, and the maximum vanishes. The results for mixed crystals of KCl-RbCl are consistent with this mechanism. Here the impulse electric strength of KCl-RbCl is more than twice as high as the corresponding d-c value at 375°K. Since the maximum in KCl disappears with 1-μsec rise-time pulses but still shows some traces for 10-μsec pulses, the time constant of this effect is in the microsecond range.

Our results showing that the impulse strength of KCl is higher than its d-c strength and that the d-c maximum in the temperature characteristic disappears with impulses contradict those of Calderwood, Grosshart, and Wallace,^{12, 22)} who found the d-c and impulse-strength characteristics identical. Since they applied a series of flat-topped pulses of increasing amplitude until breakdown, their pulse method was in effect equivalent to d-c measurement. The time lags of several microseconds which they observed were not statistical time lags at all but actually equivalent to the temporal effects associated with our fast-rising pulses.

The scatter in pulse-breakdown-strength measurements according to

23) R. Cooper, R. M. Huggin, and W. A. Smith, Proc. Phys. Soc. (London) 76, 817 (1960).

our results is wider than in the d-c case and greater than the errors of pulse measurement. This has also been observed by Vorob'ev²⁴⁾ and Kuchin.¹³⁾ Such increased scatter might be expected from the same causes which also raise the impulse-breakdown strength, since they involve an additional statistical spread. The presence of different distributions of dislocations and stresses in different samples must have some influence on the breakdown strength. Although some information on this subject has been reported previously,²⁵⁾ this situation is not clear yet.

As mentioned earlier, the quantitative theory for breakdown in ionic crystals is still far from satisfactory. Callen¹⁹⁾ derived the breakdown strength of alkali halides by considering electron-phonon collisions. The electron was considered to interact with lattice vibrations through the polarization field set up by longitudinal vibrations of the optical modes. From the rate at which an electron loses energy to the vibrational modes he evaluated the relaxation time and determined the rate at which an electron gains energy from the external field. Equating these expressions gave the electric field necessary to accelerate an electron as a function of its energy. Using von Hippel's low-energy criterion, he then identified the maximum in the electric field versus electron energy plot as the breakdown strength for a given temperature. The breakdown-strength calculations of this theory for KCl are compared in Fig. 30 with our experimental results for 1- μ sec rise-time pulses, where space-charge effects are minimized. The polaron effect, not considered in this theory, may account for the fact that the experimental breakdown strength is greater than predicted, especial-

24) G. A. Vorob'ev, Sov. Phys. - JETP 3, 225 (1956).

25) R. Cooper and A. A. Wallace, Proc. Phys. Soc. (London) B69, 1287 (1956).

ly at low temperatures.

We found that at low temperatures the electric strength of KCl-RbCl was higher than for KCl, because foreign atoms increase electronic scattering. The slope in the temperature characteristic for mixed crystals, however, was less than for pure KCl. This slope change can be understood by the

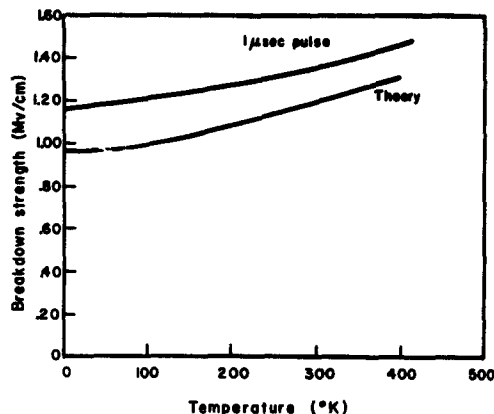


Fig. 30. Experimental breakdown strength of KCl compared with Callen's¹⁹⁾ theory.

following approximation: If we consider A the rate at which the external field supplies energy to an electron and B that at which it loses it to lattice vibrations, the breakdown strength can very roughly be determined from the equilibrium condition

$$A = B.$$

When the applied field is such that $A > B$, electrons can be accelerated to impact ionization, and the insulator breaks down. Let the rate at which energy is supplied to the electron in KCl be

$$A = \frac{e^2 \tau}{m} E_{\max}^2,$$

where τ is the relaxation time for electrons. Setting this expression equal to the rate of energy loss to the lattice, we obtain, for the breakdown strength $E_{\max}$,

$$E_{\max}^2 = \frac{Bm}{e^2 \tau}.$$

Actually A and B are functions of electron energy, but we shall ignore this in our attempt to explain the difference in slopes. In KCl-RbCl, electron

acceleration is impaired by the addition of foreign ions. If we interpret $1/\tau$ as the probability per unit time for scattering by lattice vibrations, then let us assume that $1/\tau + 1/\tau_0$ is the scattering probability for the mixed crystal. $1/\tau_0$ depends only on the concentration of foreign ions and not on temperature. For the mixed crystal,

$$E_{o(max)}^2 = \frac{Bm}{e^2} \left(\frac{1}{\tau} + \frac{1}{\tau_0} \right).$$

We finally obtain a ratio between temperature coefficients of

$$\frac{\frac{d}{dt}(E_{max})}{\frac{d}{dt}(E_{o(max)})} = \frac{\left(\tau^{-1} + \tau_0^{-1} \right)^{1/2}}{\left(\tau^{-1} \right)^{1/2}},$$

if we assume B to be independent of temperature and identical for both materials. This crude approach predicts that the temperature coefficient of electric strength for pure KCl is greater than for the mixed crystal, as observed.

A more detailed study of prebreakdown currents is required to ascertain the role played by space-charge buildup near the maximum of the d-c breakdown-temperature characteristic. Study of prebreakdown currents as function of temperature with voltage pulses of different rise time will probably tell more about trapping and field-emission effects. Konorova²⁶⁾ has already carried out some research in this direction with 100- and 1000- μ sec rise-time pulses. Measurements should also be extended to very short rise times and to higher temperatures.

26) E. A. Konorova and L. A. Sorokina, Bull. Acad. Sci. U. S. S. R., Phys. Ser., 22, 403 (1958).

Acknowledgments

The author wishes to express his gratitude to Professor A. R. von Hippel for suggesting the problem and for many helpful discussions.

Thanks are due to Janis Kalnajs for advice on chemical problems; to D. A. Powers, J. A. van Raalte, and C. E. Woodward for discussions pertaining to electronic instrumentation.

The author is very grateful for the financial aid provided by the Xerox Corporation and Woodrow Wilson Fellowships.

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