Electric Breakdown of Glasses and Crystals as a Function of Temperature*

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The effect of order and disorder upon the breakdown characteristics of ionic materials has been investigated. While the dielectric strength of the alkali-halide crystals increases steeply with temperature in the lower temperature range, the trend in glasses is found to be the opposite. A comparison of the characteristics of silica glass with quartz confirms the fact that the positive temperature coefficient of the dielectric strength is produced by the periodic structure of the crystals. Also the absolute value of the breakdown strength is appreciably increased in the disordered amorphous state. The situation is explained partly by the influence of temperature and structure upon the scattering of electrons. The increase in resistivity of metals with temperature is an analogous phenomenon.

SOME time ago it was reported[1] that the dielectric strength of ionic single crystals ike KBr shows an unexpected temperature dependence (Fig. 1): The breakdown strength rises sharply in the lower temperature range, reaches a maximum just above room temperature, and falls steeply again. A reasonable explanation for the *increasing* strength of the material seemed to be offered by the fact that with rising temperature the periodicity of the structure becomes more and more disordered by lattice vibrations. In consequence, electrons passing through the lattice will be scattered with increasing probability and slowed down; a higher field strength must be applied to produce breakdown by impact ionization. The increase in resistivity of metals with temperature is an example of an analogous situation.

If the periodic order is a decisive factor for the breakdown characteristic of a material, the increase in breakdown strength should be absent in glasses, because the structure is disordered from the start. Also their strength should be higher than that of comparable crystalline materials. Our paper investigates this situation.

Apparatus and Procedure

Simultaneously and independently of our research, Austen and Hackett[2] and Austen and Whitehead[3] have studied the temperature dependence of the breakdown strength on several materials, including KBr. The specific goal of this investigation in the British Electrical and Allied Industrial Research Laboratory was a test of Froehlich's breakdown theory.[4] Though the results obtained on the same material in England and in our laboratory (see Fig. 1) show a similar trend, they are not in too good agreement. It seems, therefore, necessary to give an accurate and critical account of our method for comparison with that used by the English authors.

Figure 2 shows the latest edition of our breakdown vessel. As in earlier constructions,[1] flat test specimens are used and clamped by spring action between the two highly polished copper electrodes E_1 and E_2 of the tubular specimen holder shown at the right. The space between the slightly rounded electrode surfaces and the edge of the sample is filled under vacuum with an insulating material possessing a very
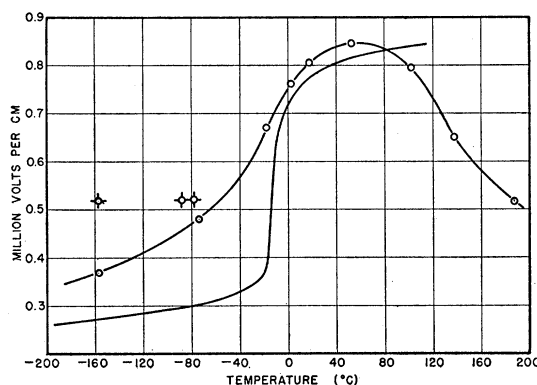
Fig. 1. The electrical breakdown strength of potassium bromide. -O- Buehl and von Hippel, -⊕- earlier data, — Austen and Whitehead.

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[1] R. C. Buehl and A. von Hippel, Phys. Rev. **56**, 941 (1939).

[2] A. E. W. Austen and W. Hackett, Nature **143**, 637 (1939).

[3] A. E. W. Austen and S. Whitehead, Proc. Roy. Soc. **A176**, 33 (1940).

[4] H. Froelich, Proc. Roy. Soc. **A160**, 230 (1937); **172**, 94 (1939); Phys. Rev. **56**, 349 (1939).

high viscosity at the test temperature. There-
upon the holder is inserted in the pressure bomb
at the left and exposed to nitrogen pressure of
between 50 and 100 atmospheres, while the
embedding material is plastic. Consequently it
forms a tight-fitting insulating ring, filling the
edges between electrodes and sample and pre-
venting edge breakdown. In addition, the gas
strengthens the insulation and suppresses corona
discharges on exposed high voltage leads.

The ground electrode E_2 contains an insu-
lated center section of 3 mm diameter for current
measurements, inserted with one to two thou-
sandths of an inch clearance into a copper block
serving for shielding and heat exchange. A
spring contact connects this current electrode
through a spark plug L with a feedback current
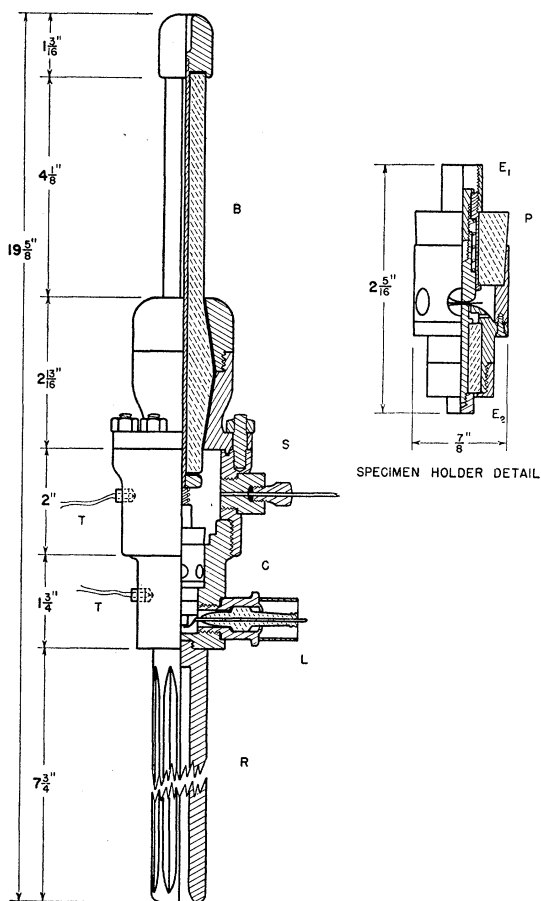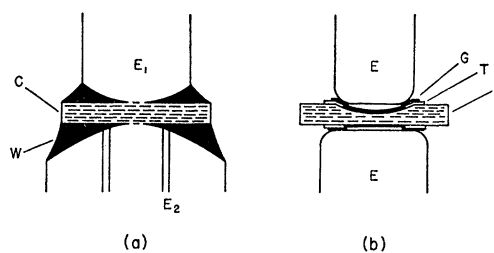amplifier similar to one previously described.[5]

Fig. 3. Electrode arrangements. E electrodes, C crystal,
W wax, G graphite, T tinfoil. Curvature of electrodes and
thickness of samples exaggerated.

An Esterline-Angus recorder in the output covers
full scale the ranges from 1×10^{-12} to 1×10^{-3}
ampere; its clock-drive is coupled to a voltage
recorder.[1] Our judgment about the reliability of a
breakdown point is based upon the current-
voltage-time records obtained, and upon a
microscopic examination of sample and elec-
trodes after breakdown. Secondary destruction
of the test piece is prevented by use of a pro-
tecting resistor (usually 100 megohm) and of an
automatic switch short-circuiting the electrodes
immediately after breakdown.

Electrode E_1 is inserted in a Pyrex block P
fitting into the copper wall of the specimen
holder. By spring contact, connection is es-
tablished with the high voltage bushing B, a
Pyrex insulator inserted in a stainless steel cup
and bolted down upon a cylinder S of the same
metal. The specimen holder fits tightly into a
copper block C screwed into the bottom of this
cylinder and carrying a copper extension rod R.
This rod, shaped for good heat transfer, pro-
vides the main temperature regulation; it is
heated electrically or immersed in a cooling
liquid. An additional temperature bath sur-
rounds the copper block and steel cylinder;
heat insulation is provided by suspending the
breakdown vessel on three Pyrex rods. The
temperature of the sample is determined by two
thermocouples T in the wall of the vessel which
have been calibrated initially against a thermo-
couple replacing the test sample.

Our method of avoiding edge breakdown
(Fig. 3a) was applied successfully between
$-80°C$ and $+150°C$ by using capacitor oil,
pyranol, beeswax, and Apiezon wax in over-
lapping ranges, but for liquid-air temperature
no solidifying material of good properties has
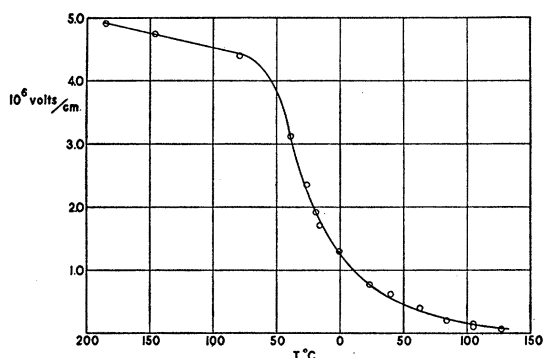been found. The highest strength resulted if

Fig. 2. High pressure breakdown vessel.

SPECIMEN HOLDER DETAIL

[5] S. Robert, Rev. Sci. Inst. **10**, 181 (1939).

FIG. 4. The breakdown strength of soda lime glass.

FIG. 5. Thermal and disruptive breakdown of glass. Current and voltage *vs.* time. The dashed line indicates a change of the current scale.

nitrogen was slowly liquefied in the breakdown vessel from its tank, and used as embedding medium under high pressure. This method was consequently adopted in the lowest temperature range. In rechecking our earlier data in the new apparatus, the liquid-air point was found lower than before (see Fig. 1), but was certainly appreciably higher than the value reported by the English authors.

Austen and coworkers tried to avoid edge effects by grinding one side of their test specimen into lens shape, painting the curvature with graphite, and providing contact over tinfoil rings to cylindrical brass electrodes (Fig. 3b). If the crystal surface was polished, a low and inconsistent breakdown strength resulted, while finely ground surfaces gave higher, reproducible values. This observation apparently indicates contact trouble between graphite and the polished crystal, which is avoided on the rough finish. But grinding of single crystals, which split easily, is likely to produce small cracks which remain unnoticed under the matt surface and create voids and field distortion. Current measurements, which would detect such disturbances, have not been made by the English authors. It may be mentioned here that the voltage should be raised very slowly to avoid possible transient effects produced by polarization or formation effects; in our tests about one hour elapses between application of the voltage and breakdown.

BREAKDOWN OF GLASSES

Figure 4 presents a typical breakdown characteristic of soda lime glass. The material came from one and the same tube and was blown into bubble 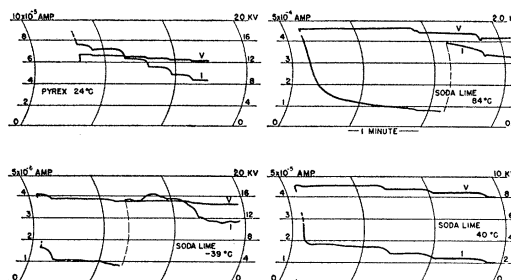windows then silver-plated in vacuum by evaporation, and tested in a thermostat. For comparison, samples were blown into thin sheets and measured in the breakdown vessel (Fig. 2) at room temperature. In the bubble windows edge effect is automatically avoided; the good agreement between the results obtained by both methods serves as a test of our apparatus.

A comparison between Fig. 1 and Fig. 4 shows that the steep positive temperature coefficient of the single crystal is in glasses replaced by a flat negative slope. Contrary to earlier observers,[6] we found no range in which the breakdown strength was independent of temperature, and our current-voltage-time characteristics seem to indicate that there exist only *two* distinct mechanisms of dielectric breakdown: At low temperatures disruptive breakdown is characterized by an abrupt jump of the current to high magnitudes without warning, while the steep decrease of the characteristic, Fig. 4, corresponds to "thermal" breakdown announced by a creeping increase of the current at constant voltage. Figure 5 gives examples of both situations: a disruptive breakdown of Pyrex at 24°C and of soda lime at −39°C, and thermal breakdown in soda lime at 40°C and 84°C. The composition of the sample, its thickness, the heat transfer, and statistics decide when the first mechanism is replaced by the second. While in soda lime glass of good ionic conductivity under homogeneous stress, a heat breakdown can be clearly observed at −30°C, a disruptive breakdown[7] may take place at +400°C if a pointed cathode is used, enhancing electron emission and impact ionization.

[6] L. Inge and A. Walther, Arch. f. Elektro. **19**, 257 (1928); **22**, 410 (1929). P. Moon and A. S. Norcross, Trans. A.I.E.E. **49**, 755 (1930).

[7] A. von Hippel, J. App. Phys. **8**, 815 (1937).

SILICA GLASS *Versus* QUARTZ

The opposing trends observed in the low temperature range of Fig. 1 and Fig. 4 lend support to our expectation that the periodic order is a decisive factor for the scattering probability of electrons in solids. For establishing this effect beyond a doubt, the same material, SiO_2, was investigated in the crystalline and the glass modification. Samples cut in different orientations were taken from the same crystal. The experiments were made difficult by the high breakdown strength of the quartz which necessitated the grinding and polishing of crystal sections down to about 0.005 cm. But the final results given in Fig. 6 confirm: The positive temperature coefficient is characteristic of the ordered state, it disappears and changes to a negative slope if the periodicity is destroyed.

Figure 6 gives more details. At $-80°C$, silica glass is about twice as strong as the quartz, while its density is about 20 percent less. The increase in breakdown strength, thanks to the amorphous state, is therefore very striking; on the other hand, it has to be expected that at some higher temperature the crystalline material will be stronger than the glass, as observed. That the crystal parallel to its axis has a slightly higher dielectric strength than when perpendicular to it, is somewhat surprising in view of the fact that the conductivity in the axis direction is a thousand times larger than in the other orientation. But this conducitivity is ionic and has therefore no direct bearing on the dielectric strength, while it may indirectly be responsible for the effect of producing field distortion by polarization. We have no evidence so far, in the case of the alkali halides,[8] that slow electrons are
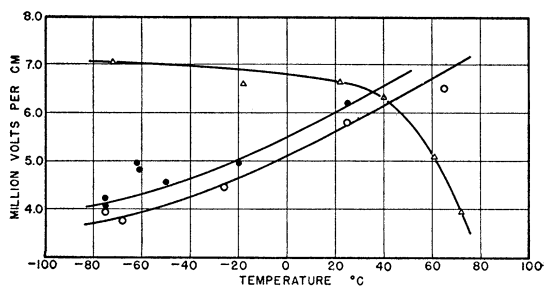
FIG. 6. The electrical breakdown strength of silica. Quartz: -●- field parallel to axis, -O- field perpendicular to axis, silica glass: -Δ-.

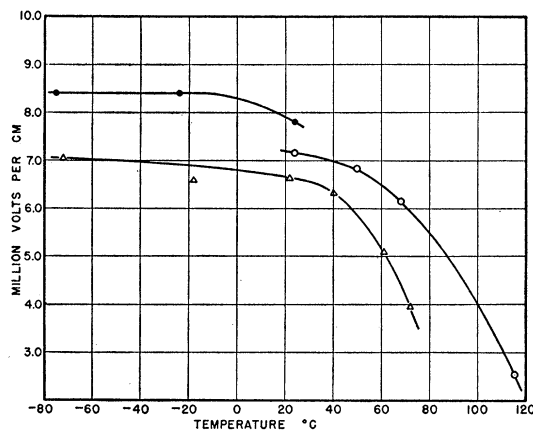[8] A. von Hippel and J. W. Davisson, Phys. Rev. **57**, 156 (1940).

FIG. 7. The electrical breakdown strength of silica glass. -●- 6×10^{-4} cm, -O- 18×10^{-4} cm, -Δ- 14×10^{-4} cm.

sensitive to the lattice direction; therefore we might expect no influence of the orientation on the strength in quartz crystals. On the other hand, we found that the optical axis in quartz is not a preferential breakdown direction; the observed path makes an angle of $41°$ with the axis. Therefore an existing effect may be small.

It is known that the properties of glasses change in wide limits with their prehistory. The breakdown strength of quartz glass is no exception to this rule. The characteristics of three samples of different thicknesses and blown at different times (Fig. 7) show the same trend, but the absolute value of the breakdown strength varies appreciably.

CONCLUSIONS

Our experiments have established that the periodic order in ionic materials has a decisive influence upon magnitude and temperature coefficient of their breakdown strength. A theoretical calculation of the temperature effect has been made by Froehlich[4] under the assumption that fast electrons are scattered by lattice vibrations. The dependence predicted is much smaller than the effect observed; in the case of silica it should be negligible, while in fact the change is of the same magnitude as in the alkali halides. We believe that Froehlich's theory is untenable for other reasons, and that the scattering effect should be calculated for slow electrons. But even then we would not expect too good an agreement, because two more physical effects enter into the picture, as is reported in the following paper.