PHYSICAL REVIEW VOLUME 116, NUMBER 6 DECEMBER 15, 1959

X-Ray Induced Electrical Polarization in Glass

T. M. Proctor*
Research and Development Division, Corning Glass Works, Corning, New York

(Received June 22, 1959)

Electrical polarization in a lead silicate glass induced by the action of x-rays on the material is found to exist. This phenomenon is surveyed experimentally as a function of total dose (incident and absorbed), dose rate, x-ray tube potential, radiation temperature, and temperature at which the polarization is released and measured. Net surface charges of the order of 10^{-9} coulomb/cm^2 can be obtained from 3-mm thick samples irradiated at room temperature with 10^5 r of 250-kv x-rays. To the first order the build-up and decay of this condition seems to follow the normal electrical relaxation as can be theoretically predicted from the dielectric and resistivity constants of the material ($\tau=\rho\epsilon$). Measured surface charge is shown to be proportional to absorbed dose for smaller doses; however, for greater doses final equilibrium in the polarization is reached when back electrical conduction becomes as large as the forward x-ray induced displacement current. The dependence of measured surface charge upon the sample thickness has been experimentally investigated for one case.

INTRODUCTION

CHANGES in the electrical conductivity of solids exposed to strong x-radiation have been noted.[1] Gross has recently studied dielectric breakdown caused by 2-Mev electrons in borosilicate glasses[2] and electrical polarization caused by Co_{60} γ rays in Plexiglas and borosilicate glass.[3] He demonstrated that electric fields of magnitudes large enough to cause triggered electrical breakdown can be formed by the action of intense, high-energy radiation. X-ray induced electric fields in germanium have been found and investigated.[4] Similar electrical breakdown phenomena caused by Co_{60} γ rays have been shown to exist in lead silicate glasses.[5]

This research was stimulated by this last discovery. The aim was to make a survey of the variables which were thought to control x-radiation induced electrical fields in glass and to test a theory for the phenomenon.

Samples of lead silicate glass were irradiated with x-rays and the apparent charge displacement or dipole moment as seen at sample electrodes was measured as a function of total dose, dose rate, dose temperature, measuring temperature, sample thickness and minimum available x-ray wavelength limit as determined by the x-ray tube potential.

THEORY

A theoretical approach was made to this problem by Condon[6] and Culler.[7] From conservation of momentum and energy it can be shown that when high-energy radiation collides with charge centers and Compton scattering ensues, a nonspherical angular distribution of scattered electrons is produced. For Compton collisions where high energies are involved, the angular distribution is preferentially oriented in the forward direction.[8] This kind of a distribution produces charge separation and an effective dipole moment. If polarization were being formed in the manner just described, it might be expected to be controlled by (a) the kind and intensity of the incident radiation, (b) the size of the polarization field which retards and shortens the effective range of the forward going Compton electrons, and (c) the reverse conduction current as determined by the charge carrier mobility for the substance. Thus, under these assumptions, the displacement current J_D can be expressed

$$J_D=N_a er(1-kE)-\rho^{-1}E. \qquad (1)$$

The first term on the right describes the net production of Compton dipoles by a constant formation of dipoles $N_a er$ less a term $N_a erkE$ which describes the retardation of the Compton electrons as they proceed against the polarization field having been formed up to that time. This term is a first order guess at the actual retardation factor. N_a is the number of photons/cm^3 sec absorbed while er is the dipole moment formed by each absorbed photon. k is the range retardation constant. E is the electric field produced at the point. For any one irradiation er and N_a have been assumed to be constant, depending only on radiation wavelength and intensity, respectively. The second term on the right is the dipole destruction term associated with normal electrical conduction where ρ is the resistivity.

The dipole formation and destruction terms of Eq. (1) result in a final equilibrium field E_∞. Equation (1) takes the form of

$$\dot{E}+(1/\tau)E=(1/\tau)E_\infty, \qquad (2)$$

---

* Now at the National Bureau of Standards, Washington, D. C.

[1] J. F. Fowler, Proc. Roy. Soc. (London) **236**, 464, 480 (1950).

[2] Bernhard Gross, Phys. Rev. **107**, 368 (1957).

[3] Bernhard Gross, J. Polymer Sci. **28**, 135 (1958). See also Phys. Rev. **110**, 337 (1958).

[4] F. I. Kolomoitsev and F. F. Kodzhespirov, J. Tech. Phys. U.S.S.R. **27**, 899 (1957) [translation: Soviet Phys. (Tech. Phys.) **2**, 823 (1957)].

[5] W. W. Shaver (private communication).

[6] E. U. Condon (private communication).

[7] V. E. Culler (private communication).

[8] C. M. Davisson and R. D. Evans, Revs. Modern Phys. **24**, 79 (1952).

where

$$\tau = \epsilon\rho/(1+N_a e r \rho k),$$

$$E_\infty = N_a e r \rho/(1+N_a e r \rho k)$$

and ϵ is the permittivity. This equation has the time solution

$$E = E_\infty[1-e^{-t/\tau}]. \qquad (3)$$

EXPERIMENTAL ARRANGEMENT

Six samples of a lead silicate glass in the form of plates, having a plane surface area of approximately 10 cm^2 and with thicknesses ranging from 1 to 6 mm, were irradiated with 250-kilovolt x-rays from a tungsten target x-ray tube. The radiation impinged perpendicular to the plane surface of the samples and the total incident radiation dose varied from 10^3 to 10^6 roentgen. The absorbed dose ranged from 30% to 85% of the incident dose, depending upon the sample thickness. The sample temperature during irradiation, designated T_r, was controlled by a thermostated temperature bath. After samples had been irradiated, they were removed from the x-rays for charge measurements.

The internal charge distributions are considered to be "frozen in" for the interval between irradiation and measurement since the relaxation time is so large ($\tau \approx \epsilon\rho > 10^5$ sec) at room temperature.

Before charge measurement irradiated samples were cleaned by an elaborate chemical sequence which included a light etch in HF. From then on they were handled with ground tongs until the experiment was completed. Electrodes of air dry silver were painted on the two plane faces. Various precautions, designed to reduce creation of extraneous static surface charge, were followed. Samples were keepered until just before measuring and the insulating edge surface between the two electrodes was kept as small as possible. By following these procedures the extraneous static surface charge was reduced to a negligible amount.

The sample, charged by irradiation to the "frozen in" charge state, was then measured for charge content. The measuring equipment for this purpose is shown schematically in Fig. 1. The release of the "frozen in" charges was brought about by plunging the sample into a specially constructed furnace which was already at a higher temperature, T_D. This furnace had a large thermal inertia and was temperature controlled by an electronic device. (Temperature gradients across the sample thickness never exceeded 5°C and usually were much less.) It contained a set of electrodes which provided support for the sample and which were well insulated electrically from the furnace and from ground. The release of the charge was measured by an electrometer acting as an ammeter. The ammeter measured the net current flowing out of the top electrode to ground. (Electrode or side A indicates the side of the sample originally nearest to the radiation, while B is the side farthest from the radiation.)

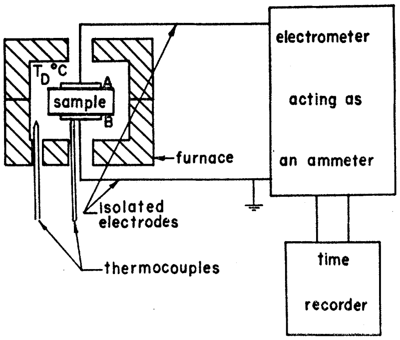

FIG. 1. A schematic showing the furnace used to release the "frozen in" charge, the ammeter (electrometer) and the associated recorder.

The current measured by the ammeter was continuously recorded with time. The temperature of the lower grounded electrode was also measured frequently during the charge release. The first part of every charge release was affected by the heating of the sample from room temperature up to T_D. This effect usually lasted about 30 seconds and the charge released during this period was measured and recorded along with the subsequent equilibrium temperature data.

As a check on the origin of the charges observed, a number of samples were put through the entire experimental procedure from being placed in the irradiation position under the x-rays to cleaning, electrode painting, and measuring the released charge. However, these samples were purposely isolated from the x-radiation by a lead shield so that the absorbed dose in the samples would be essentially zero. These samples always contained less than 10^{-12} coul/cm^2 of released charge; sometimes the released charge was as small as 10^{-14} coul/cm^2. Thus it appears that the phenomena indicated in the results are associated with the absorption of x-rays in the material.

RESULTS

X-ray induced polarization is studied as a function of (1) total incident radiation dose, π_i, (2) total absorbed radiation dose, π_a, (3) radiation rate, R, (4) temperature of sample during irradiation, T_r, (5) x-ray tube potential, V_T, (6) size, i.e., sample thickness. (Charge produced per cm^2 seems to be independent of the sample area for the measured sample areas.) and (7) temperature of sample during charge release, T_D. In each case all the independent variables are kept constant except the one being studied. The state of electrical polarization is determined by the total net charge per cm^2, Q, which is released from the electrode A. This is determined by integrating the measured current as a function of time from the beginning of charge release to a time at which the remaining charge is deemed negligible. Total precision of Q (due to errors in all variables measured) is about ±50%. (The merits of Q as a measure of internal charge state are discussed in the next section.)

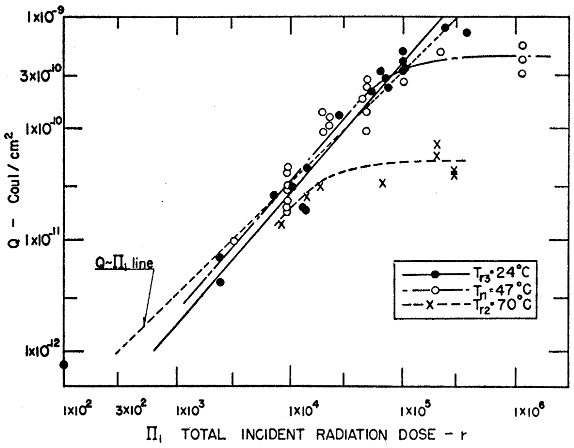

FIG. 2. Q, net charge produced on electrode-A of 3-mm thick lead glass samples as a result of being exposed to 250-kv x-rays, *vs* π_i, total incident radiation dose in roentgens. Dose rate R is constant at 300 r/min. Three different curves represent this function for three different sample irradiation temperatures T_r. The dashed line labelled $Q\sim\pi_i$ is a line of linear proportionality between Q and π_i.

In all cases measured, regardless of sample orientation between the furnace electrodes, the polarity of the charged samples is such that upon release the positive current flows from B (side farthest from radiation) to A (side nearest radiation) in the external circuit.

Figure 2 shows the experimental findings of Q, the total net released charge, *versus* the total incident dose, π_i. It is shown for three different irradiation temperatures. The other variables have the following values:

$$\pi_a=0.69\pi_i,\quad R=300\ \text{r/min},\quad V_T=250\ \text{kv},\quad T_D=95°\text{C},$$

and sample thickness L is 3 mm. These curves are characterized by the saturation of Q *vs* π_i. The saturation occurs at lower Q's for higher temperatures, T_r.

The effect of incident dose rate R on electrical polarization as indicated by Q is shown in Fig. 3. Special effort is made to keep π_a, the absorbed total dose, constant in each case by changing exposure time accordingly. The other variables are:

$$\pi_a=6900\ \text{r},\quad \pi_i=10^4\ \text{r},\quad V_T=250\ \text{kv},$$
$$T_D=95°\text{C},\quad T_r=47°\text{C},$$

and sample thickness L is 3 mm. Note the saturation in Q for higher rates.

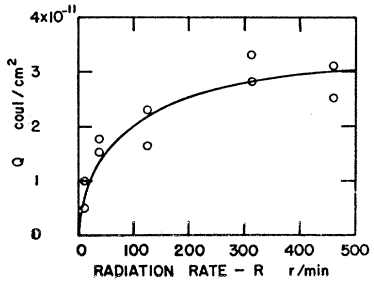

FIG. 3. Q, net charge produced on electrode-A for 3-mm thick lead glass samples as a result of being exposed to 250-kv x-rays, *vs* R, radiation rate in roentgens per min. Doses π_i are constant at 10^4 r incident while sample irradiation temperature T_r was constant at 47°C.

If the current output from the sample due to the formation of Q is measured as a function of time, a decaying function is obtained. This curve is not characterized exactly by one relaxation time τ, but in a chosen range the deviation of this measured curve from $e^{-t/\tau}$ is small. Figure 4 is a plot of τ's obtained in this way from relaxation data *vs* inverse temperature. The τ's reported here are determined from the slope of the current *vs* time curves at a time lying between 100 and 200 seconds after the charge release experiment was started. Thus they are roughly comparable. On Fig. 4 the solid line is a calculated line of $\tau=\epsilon\rho$ where ϵ is the 60 cps value for this glass and ρ has been experimentally measured. In general the measured τ's seem to be about 50% larger than the calculated $\tau_{60\ \text{cps}}$.

Some effort has been made to determine the effect of sample thickness on Q. The effect on Q of sample thickness expressed in terms of μL is shown in Fig. 5. μ is the linear absorption coefficient for these x-rays.

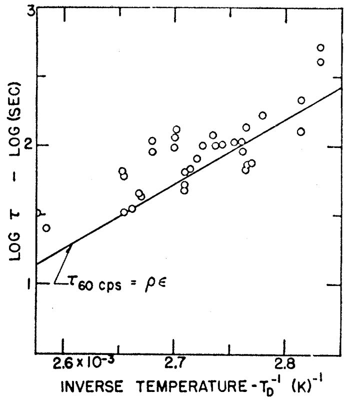

FIG. 4. Log of the relaxation time τ as obtained from current data associated with the relaxation of x-ray induced polarization *vs* inverse release temperature T_D^{-1}. Circles are data points while the solid line represents the predicted $\tau_{60\ \text{cps}}$ function.

The other variables are controlled to

$$\pi_i=10^4\ \text{r},\quad T_r=47°\text{C},\quad T_D=95°\text{C},$$
$$R=300\ \text{r/min},\quad V_T=250\ \text{kv},$$

and π_a is in accordance with sample thickness. The original data are marked with small x's while the means of these groups of data points are indicated by the large circles with the limits being the average deviation. In general each data point has an uncertainty of ±50%. Thus these data are not very conclusive.

The change in Q with changing tube potential V_T was investigated. Results are characterized by a sharp increase in Q as V_T is increased past the critical voltage of the K_α line of tungsten. This is followed by a leveling off of Q values as V_T is increased up to 250 kv. Changing of the x-ray tube potential does not change the effective photon wavelength in a continuous manner, and the results are probably determined by the spectral response of the tube.

Charge release experiments conducted at different temperatures, T_D, indicate that Q is not greatly dependent on T_D. For these measurements:

$$\pi_i = 4.7\times 10^4 \text{ r},\quad \pi_a = 3.2\times 10^4 \text{ r},\quad V_T = 250 \text{ kv},$$
$$T_r = 47^\circ\text{C},\quad R = 300 \text{ r/min},$$

and L is 3 mm. In the range for T_D of 80°C to 120°C, Q is constant to within a factor of 2 while relaxation time and resistivity of the glass change by a factor of about 50. Such variation as has been observed is possibly related to the difficulties of observation on the long relaxation time measurements.

DISCUSSION

The scatter of the charge data Q around the curves of Figs. 2, 3, and 5 is in general within $\pm 50\%$ of the curve's value at that point. (This error is composed of errors in all variables.) This scatter can be explained by the uncertainties in the measured charge and in the total radiation doses, π_i and π_a, for any particular sample. The precision is tolerable considering that this is a survey over a number of decades in Q and in π_i.

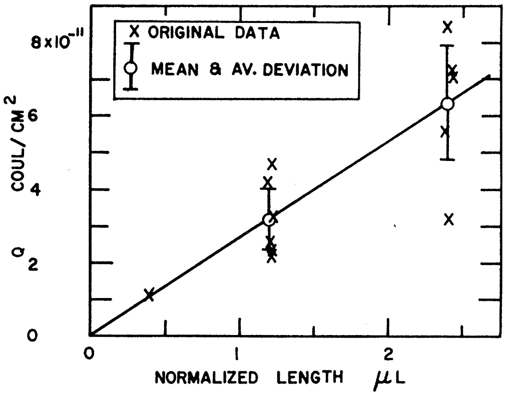

FIG. 5. Q, net charge produced on electrode-A for lead glass samples by 10^4 incident roentgens of 250-kv x-rays, *vs* μL, normalized sample thickness. Radiation temperature T_r was constant at 47°C while radiation rate R was kept at 300 r/min. Experimental data is shown by x's. Mean values and average deviations are shown for two clusters of data.

How well does Q as measured represent the internal charge concentration ρ? ρ will be a function of both time, t, and distance, x, through the sample. Assuming that Eq. (1) represents the situation and that the partial differential equation describing ρ is solvable by separation of variables, the time solution for ρ, i.e., $\rho(t)$, will be proportional to $Q(t)$. If the complete behavior of $\rho(x,t)$ were known, i.e., the way it forms and decays, $Q(t)$ would be determined. However, $Q(t)$ data is insufficient to reveal $\rho(x,t)$ directly. Considering that Q is much easier to measure than ρ, one approach would be to assume a model of formation and decay from which $\rho(x,t)$ could be developed and then to test this model by comparing predicted $Q(t)$ with measured $Q(t)$ for different sample thicknesses.

A specific electrical polarity of the samples with respect to the original photon direction is always noticed, i.e., current always flows into the electrode which was nearest the radiation. Depending upon the type of relaxation mechanism assigned, e.g., charges being neutralized at the electrodes or dipoles relaxing internally, either direction of relaxation current could be explained. In fact two currents of different directions are noticed; a stronger one occurring first is the one discussed in this paper while the second one is much weaker and has a much longer time constant than the first. The second one in general is small enough to be close to the limit of resolution of the measurements and can be considered nonexistent for the purposes of this paper.

Next examine Fig. 2 in the region below saturation of Q with π_i. Within the experimental limit of error the amount of charge Q out of A is directly proportional to the incident dose π_i. The solid curves have been drawn so as to fit the data. However, the $Q\sim\pi_i$ line as indicated is within the limits of error and could be selected for the below saturation curve. Notice when each curve saturates to a final value of charge, Q_∞, that

$$Q_\infty|_{T_{r1}=47^\circ\text{C}} \approx 10 Q_\infty|_{T_{r2}=70^\circ\text{C}},$$

where the subscripts on Q_∞ indicate the two different irradiation temperatures. This might be expected if we consider Eq. (3) to be conductivity controlled because temperatures T_{r1} and T_{r2} are chosen in such a way that

$$10\sigma|_{T_{r1}} = \sigma|_{T_{r2}}.$$

σ is the experimental dc conductivity of this glass at these specific temperatures and in the absence of any radiation.

When the $(1-e^{-1})$ points from the data curves of Fig. 2 are compared, the determined τ's are

$$\tau(T_{r1}) \approx 1.4\times 10^4 \text{ sec},\quad \tau(T_{r2}) \approx 4.0\times 10^3 \text{ sec}.$$

If one had selected the $Q\sim\pi_i$ line as the unsaturated portion of each data curve, then

$$\tau(T_{r1}) \approx 10\tau(T_{r2}).$$

The presence of ionizing radiation or even a past history of radiation might be expected to affect the magnitude of the electrical relaxation time. There is some evidence[9,10] that one might expect τ to decrease in substances which are under strong radiation. In many solids including glass ρ is decreased. The product $\rho\epsilon$ would decrease if ϵ were essentially constant. Thus the conditions of any measurement of τ must be understood. Data points of Fig. 2 are determined by the total net released charge, Q. In this case Q, which is assumed to be proportional to the radiation induced polarization, has been built up while the sample was under radiation. Therefore, the τ's determined from

---

[9] S. V. Svechnikov, J. Tech. Phys. U.S.S.R. **27**, 2492 (1957) [translation: Soviet Phys. (Tech. Phys.) **2**, 2320, 2328 (1957)].

[10] W. C. Roentgen, Ann. Physik **64**, 1 (1921).

Fig. 2 are in effect like electrical relaxation times measured under the conditions of radiation. This is in contrast to the τ's predicted from the $\rho\epsilon_{60\text{ cps}}$ line of Fig. 4 which indicates the simple electrical relaxation time in the absence of radiation. The τ's measured from Fig. 2 are approximately four times longer than the $\rho\epsilon_{60\text{ cps}}$ line would predict.

In the below saturation region of the data of Fig. 2, Q can be described as

$$Q\approx 3.0\times 10^{-15}\pi_i,$$

by assuming the $Q\sim\pi_i$ line for this region. Equation (2) predicts a linear relation between E, the field and $N_a t$, the absorbed radiation, in the region where $t\ll\tau$. If E and Q have the same time dependency and the absorbed radiation is proportional to the incident radiation, the linear relation above is expected for the region of $t\ll\tau$.

Figure 3 demonstrates how Q saturates at high rates of radiation dosage while at lower rates Q is diminished. At high rates of dosage dipoles formed by the charging phenomenon have little chance to relax during the experiment; at low rates the size of Q is determined not only by the radiation dose but by the number of dipoles which relax. This can be seen from Eq. (3). Assuming that $E(t)$ is described by $Q(t)$ and that $\pi_i=Rt=\alpha N_a t$ where t is the exposure time and α is a constant relating incident dose rate R to absorbed dose rate N_a, then

$$E=E_\infty[1-e^{-\pi_i/R\tau}]. \tag{4}$$

But this has the form of Fig. 3 because at

(1) large dose rates, i.e., $R\gg\pi_i/\tau$;

$E=er\pi_i/\epsilon\alpha$ which is a constant for constant π_i,

(2) $R=\pi_i/\tau$; $E=(er\pi_i/\epsilon\alpha)(1-e^{-1})$,

and (3) small dose rates, i.e., $R\ll\pi_i/\tau$;

$$E=(er\tau/\epsilon\alpha)R.$$

By using the $(1-e^{-1})$ point to determine τ, $\tau=6.3\times 10^3$ sec. This τ should compare with the τ's of Fig. 2 as they are determined in essentially the same way. This τ is only 20% longer than the τ computed from the $\rho\epsilon_{60\text{ cps}}$ line.

Figure 4 indicates that the τ's measured from 100 to 200 seconds after the start of a relaxation experiment are about 50% larger than the $\rho\epsilon_{60\text{ cps}}$ line. The τ's shown in Fig. 4 are different from either the calculated $\rho\epsilon_{60\text{ cps}}$ line or from the measured τ's of Figs. 2 and 3. τ's of Fig. 4 are measured from the actual shapes of the current decay curves which are performed in the absence of radiation. However, these τ's may be affected by the existence of a past history of radiation. Even if past radiation history is unimportant, τ's of Fig. 4 might be expected to be longer than the $\rho\epsilon_{60\text{ cps}}$ predicted ones. As the length of the experiment is changed from 60 cycles/sec to 100 seconds/experiment the experimental ϵ's measured for many glasses may increase by twofold.[11]

Examination of Fig. 5 indicates a linear relationship between Q and sample thickness μL. One would expect the charge Q eventually to saturate for samples of large thickness because of the attenuation of radiation with thickness.

CONCLUSION

X-ray induced polarization has been found to exist in a lead silicate glass. The predictions of Condon and Culler that there is a time constant for the formation of x-ray induced dipole space charge and that this constant is essentially the normal electrical relaxation constant is approximately true. Variations in this relaxation time due to the existence of ionizing radiation of 250-kv x-rays appear to be slight. If the charge-total dose relationship is examined, where $t\ll\tau$, the net charge appearing on one electrode is proportional to the incident number of photons on that surface. The value of the net charge on the electrode develops to $(1-e^{-1})$ of its final value in approximately one time constant $\epsilon\rho$. Variation of rate of x-ray dose appears to adjust Q in a consistent manner. In the region of the variables studied the build-up of Q is essentially conductivity controlled and is apparently unaffected by the retardation factor k.

ACKNOWLEDGMENTS

The author wishes to thank Dr. W. H. Armistead, Director of Research and Development Division of Corning Glass Works, for his permission to publish this work. He also thanks Dr. P. M. Sutton for his extremely helpful suggestions and criticisms and Dr. V. E. Culler and Dr. E. U. Condon for their enlightening discussions.

---

[11] Volger, Stevels, and Van Amerongen, Philips Research Repts. 8, 452 (1953).