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A TECHNIQUE FOR STUDYING PIEZOELECTRICITY UNDER TRANSIENT HIGH STRESS CONDITIONS

by

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Technique for Studying Piezoelectricity under Transient High Stress Conditions*

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An experimental technique is described which is being successfully used to study the transient high stress piezoelectric behavior of synthetic alpha-quartz. Short, flat-faced cylinders of quartz are impacted upon each other at high velocity to produce the desired transient stress in the range from 5–70 kilobar. Precision is maintained in the alignment of the flat impact surfaces so that the Hugoniot conservation of momentum and the relationship governing the impact of flat cylinders may be used to compute the stress imparted to the target cylinders of quartz. Electric charge release data are taken in conjunction with the initial passage of the stress wave produced by impact.

INTRODUCTION

SINCE the discovery of the piezoelectric effect by Pierre and Jacques Curie in 1880, considerable application of the effect has been made utilizing a number of piezoelectric materials.[1] Most of the applications involve the use of the material in an environment of low pressure and low electric field. Within this low signal range the properties of many piezoelectric materials have been very extensively investigated. Because of the experimental difficulties involved and since little application has been found for piezoelectric materials under high stress, high field conditions, only a meager amount of experimental data is available describing the mechanical and piezoelectric properties of piezoelectric materials when they are subjected to these "high signal" conditions. Interest in high pressure research in general has been steadily increasing and considerable interest has been shown in equation of state studies of metals when they are subjected to high transient stress.[2] In conjunction with free surface velocity techniques Goranson *et al.*[3] used the piezoelectric effect in a tourmaline crystal as a quantitative measure of a Hugoniot elastic limit for steel of 15.7 kilobar.[4] Pressure measurements were also reported as high as 324 kilobar. Minshall[5] used tourmaline to indicate arrival times of the various waves but did not attempt to use the piezoelectric effect as a quantitative indication of stress. Interest in the use of alpha-quartz for a continuous record of the details of the elastic wave structure in metals was shown in discussion periods at the Technical Conference on Response of Metals to High-Velocity Deformation.[6] Neilson, Benedick, and Halpin[7] have obtained very encouraging results in using quartz for this purpose. The use of quartz under these high stress, transient conditions clearly emphasizes the necessity of investigating the properties of quartz itself under similar conditions before it can be used with confidence in this unusual environment. It is the object of this paper to describe a technique which is being successfully applied to study the piezoelectric behavior of synthetic alpha-quartz when subjected to high transient stress.

IMPACT OF PROJECTILES

In order to vary the applied stress from low stress to the high stress region in small increments, the stress produced by the impact of flat-faced projectiles fired by a gun offers many advantages. If high enough impact velocities can be achieved, stress can be produced which is as high as the lower end of the pressure range which can be conveniently achieved with the detonation of high explosives in intimate contact with a metal (say about 70 kilobar). The general procedure used in the technique reported upon here is to impact two very short flat-faced cylinders of quartz upon each other and measure the electric charge produced in one of the cylinders due to the initial transit of the stress wave produced by impact. The relationships given below may be utilized to define the stress produced by the impact.

THE HUGONIOT CONSERVATION RELATION

The conservation relations of a stress wave front were first derived by Rankine and Hugoniot and are given by Rice *et al.*[2] From Newton's law of conservation of momentum it can be shown that the following relation describes a time-independent, one-dimensional stress profile,[8]

$$\sigma=\rho_0 u_S u_P, \tag{1}$$

where σ = stress imparted by the wave front, ρ_0 = mass

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[*] This work was done under the auspices of the U. S. Atomic Energy Commission.

[1] See, for example, W. P. Mason, *Piezoelectric Crystals and Their Application to Ultrasonics* (D. Van Nostrand Company, Inc., Princeton, New Jersey, 1950); W. G. Cady, *Piezoelectricity* (McGraw-Hill Book Company, Inc., New York, 1946).

[2] See M. H. Rice, R. G. McQueen, and J. M. Walsh, "Compression of Solids by Strong Shock Waves," *Solid State Physics* (Academic Press, Inc., New York, 1958), Vol. VI.

[3] R. W. Goranson, D. Bancroft, B. L. Burton, T. Blechar, E. E. Hudson, E. F. Gittings, and S. A. Landeen, J. Appl. Phys. **26**, 1472 (1955).

[4] 1 kilobar = 982.92 atmospheres = 14 504 psi = 10^9 dynes/cm^2.

[5] S. Minshall, J. Appl. Phys. **26**, 463 (1955).

[6] Technical Conference on Response of Metals to High-Velocity Deformation held at Estes Park, Colorado, July 11–12, 1960, sponsored by Physical Metallurgy Committee, The Metallurgical Society, AIMMPE.

[7] F. W. Neilson, W. B. Benedick, and W. J. Halpin, Sandia Corporation, Albuquerque, New Mexico (private communication).

[8] Stress profile as used here refers to the relation between stress and space as viewed at a given time or the relation between stress and time as viewed at a given point.

density of the undisturbed material, u_s = propagation velocity of the wave front, u_P = particle velocity imparted by the wave front.

Stress and velocity are relative to the medium into which the stress wave is moving.

Let

$$z=\rho_0 u_s,$$

called the "acoustic impedance" by many investigators, when u_s is the elastic stress wave velocity. Then

$$\sigma=z u_P.$$

Since the stress is related to the particle velocity by the acoustic impedance characteristic of the stress profile being considered, a determination of particle velocity in a situation where the acoustic impedance is known leads immediately to the stress. It is assumed in our analysis that the stress wave propagation velocity is constant for all stress amplitudes within the elastic range and is equal to the low signal value. An experimentally determined particle velocity and the elastic acoustic impedance are sufficient for elastic stress determination. This assumption is consistent with the condition of the experiments if the stress range investigated extends from previously confirmed elastic regions into higher elastic stress regions. The assumption is also verified experimentally.

MECHANICS OF FLAT CYLINDER IMPACT

Consider now the case of a flat-faced cylinder moving at a velocity v_0 and impacting upon a stationary flat-faced target cylinder. At any time in which the surfaces of the two cylinders are in contact, the forces between the two surfaces are equal by virtue of Newton's third law. To maintain contact, the velocity of the two surfaces must also be equal. Taking the two cylinders to be of the same material, it follows that the particle velocity imparted to the stationary target by impact is

$$u_P=\tfrac{1}{2}v_0. \tag{2}$$

For an experiment meeting the requirements given in developing Eqs. (1) and (2), a measured impact velocity may be used to compute the particle velocity imparted by the impact. Knowing the particle velocity and acoustic impedance of the wave front, the stress due to impact may be computed.

In order to subject the specimen to a uniform input, it is necessary that the entire face of the specimen be impacted at the same instant. This places the requirement on any experiment which utilizes these relationships that instantaneous closure be achieved between the entire contacting surfaces of the two cylinders; that is the cylinders must be aligned at impact such that there is negligible "angle of tilt" between the two flat surfaces.

EXPECTED CHARGE RELEASE CHARACTERISTICS

Neilson and Anderson[9] have derived relations which describe the electrical characteristics of a piezoelectric medium in which a stress wave is propagating. The idealized one-dimensional situation is shown in Fig. 1. The expected behavior of the medium may be considered the same as that of two time-varying capacitors, C_1 and C_2, in series. This model is descriptive if the electric displacement D_1 is continuous across the stress wave front, that is $D_1=D_2=D_0$. As the stress wave enters the quartz, a quantity of charge $Q=PA$ is released, producing a potential V_1 on C_1. "P" here is the polarization produced due to the application of the stress and "A" is the electroded area normal to the direction of stress application. For short circuit conditions

$$V_1=V_2$$

or

$$Q_1(l-X_{(t)})/\epsilon_1=Q_2X_{(t)}/\epsilon_2,$$

where ϵ is the dielectric permittivity.

Since

$$Q_1+Q_2=Q_0=PA$$

and

$$X_{(t)}=u_s t,$$

it follows that

$$Q_2=PAu_st/l \qquad \text{for} \quad \epsilon_1=\epsilon_2$$

$$i=dQ/dt=PAu_s/l. \tag{3}$$

As shown in Fig. 2, Eq. (3) predicts a current pulse constant with time during the transit of the wave. From a current versus time record obtained under short circuit conditions, several observations can then be made. If we are interested in relating an experimentally observed charge to a known applied impact stress, we may integrate the current versus time record to obtain the charge released due to the transit of this stress. If there is no tilt in the wave front, the transit time of the wave is indicated by the time from the initial indication of current to the abrupt

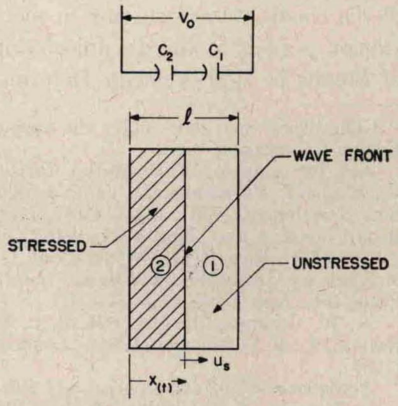

FIG. 1. Stress wave in a piezoelectric medium.

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[9] F W. Neilson and G. W. Anderson, Sandia Corporation, Albuquerque, New Mexico (private communication).

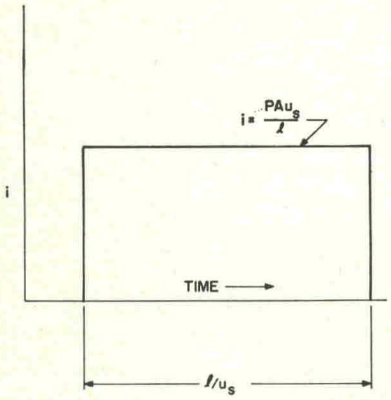

FIG. 2. Expected one-dimensional current waveform.

drop in current when the wave front exits from the specimen.

DISCUSSION OF THE EXPERIMENTAL REQUIREMENTS

We have assumed in the analysis that the wave propagation velocity is constant for all stress amplitudes within the elastic range. Some experimental verification is needed for this assumption. A stress wave front propagating from the impact surface will propagate under one-dimensional strain conditions. For the x-cut crystals[10] used here, the propagation is along the x axis with the propagation velocity given by

$$u_s=[c_{11}/\rho_0]^{\frac{1}{2}}, \tag{4}$$

where c_{11}= the one dimensional strain elastic constant for quartz in the x direction, ρ_0 and u_s as previously defined.

Bechmann[11] has published an internally consistent set of elastic constants which are the results of many observations. The values were reported to have a high degree of reproducibility. He gives $c_{11}=86.74\times10^9$ newton/m². Using the density 2.65×10^3 kg/m³ the one-dimensional strain velocity is 5.72×10^3 m/sec.

In an anisotropic solid such as quartz, it is not generally true that the particle motion is normal to the wave front and hence truly longitudinal motion alone.[12] There are certain directions of propagation called specific directions along which the particle motion is truly longitudinal. The x axis is a specific direction for quartz and the trigonal system in general[13] so no uncertainties arise in these experiments due to nonspecific particle motion.

Bridgman[14] showed that the mean linear compressibility[15] of x-cut quartz was linear up to a hydrostatic stress of 8 kilobars. The piezoelectric constant d_{11} of quartz has also been confirmed to be the low signal value up to 3.5 kilobar.[16] Our experiments are designed to begin at stress as low as 5 kilobars and then extend to higher stress levels. If linear behavior is noted in the relation between impact stress and electric charge and this extrapolates to zero within the precision of the data, a confirmation is obtained of the linearity of the combination of the piezoelectric response and the elastic constant c_{11}. For stress regions in which nonlinear behavior is observed, the stress may not be computed from the known particle velocity.

The time taken for the stress wave to propagate through the specimen is indicated on the charge release records. This time may be used to determine the wave propagation velocity. The amount of closure time complicates a precise measure of wave velocity at low impact velocities, but at an impact velocity of 2000 ft/sec our data show the wave velocity to be equal to the low signal value to within 1%.

In these experiments, each experimental point is obtained with a different specimen since the specimen is destroyed by the impact. Experimental scatter is introduced by the variation in properties from one specimen to another. For the case of quartz, a search[17] of the literature shows remarkable consistency in the c_{11} elastic constant as reported by many different investigators using different techniques. From these reported values, it is estimated that the c_{11} constant of our quartz specimens did not vary more than 0.5% from one specimen to another. The experimental results we have obtained indicate that no inconsistency is present that could be attributed to this cause.

Instantaneous Closure

The requirement for instantaneous closure of the entire contact surfaces is the most difficult to meet experimentally but is so important that an unambiguous experiment cannot be achieved without meeting the requirement. The flat cylinder relationships given here have been known for many years, probably originating from St. Venant in 1867.[18] There has been a noted lack of success in applying them at points close to the impact surface, however, due to difficulties in achieving instantaneous closure. The general approach that many experimenters have used to overcome this difficulty is to use rounded impact surfaces and apply other impact relationships.[19,20] The simplicity of the form of the imposed stress pulse for a flat surface impact which produces a plane stress wave normal to the axis of the speci-

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[10] For definition of crystal cuts, elastic and piezoelectric notation see Proc. Inst. Radio Engrs. **37**, 1378 (1949).

[11] R. Bechmann, Phys. Rev. **110**, 1060 (1958).

[12] For a discussion of this see, R. F. S. Hearmon, *Applied Anisotropic Elasticity* (Oxford University Press, New York, 1961), p. 79.

[13] F. E. Borgnis, Phys. Rev. **98**, 1000 (1955).

[14] P. W. Bridgman, Am. J. Sci. **X**, 483 (1925), also given in R. B. Sosman, *Properties of Silicia, Part II* (Book Department, The Chemical Catalog Company, Inc., New York, 1927), p. 430.

[15] For definition of linear compressibility see, J. F. Nye, *Physical Properties of Crystals* (Clarendon Press, Oxford, England, 1957), p. 145.

[16] J. L. Karcher, Sci. Papers Bur. Standards **18**, 257 (1922).

[17] For one such search see, R. F. S. Hearmon, Brit. J. Appl. Phys. **3**, 120 (1952).

[18] B. De Saint-Venant, Journal de Mathematiques (Journal de Liouville) 2ᵉ Series, **XII**, 237 (1867).

[19] For a review of work on the impact of long cylinders, see R. M. Davies, "Stress Waves in Solids," *Surveys in Mechanics*, edited by G. K. Batchelor and R. M. Davies (Cambridge University Press, New York, 1956), p. 68.

[20] W. Goldsmith, *Impact* (Edward Arnold, Ltd., London, 1960), p. 267.

men makes it a much more desirable system with which to study piezoelectric behavior. Because of this, the experiments are designed to satisfy the requirement of instantaneous closure.

Instantaneous from the real point of view depends upon the time scale of the experiment. The duration of the event to be observed in these experiments is the time taken for the stress wave to propagate the length of the specimen. For a given impact velocity and angular misalignment of the two impacting surfaces, the diameter of the specimen will determine the duration of closure time. To minimize closure time relative to transit time, one should make the diameter-to-length ratio d/l as small as possible. A specimen size of $\frac{1}{2}$ in. diam by $\frac{1}{4}$ in. long ($d/l=2$) was chosen for use in connecting data from the 5-kilobar region into the higher stress region. The transit time for this length specimen is about one microsecond.

Before the experiments were conducted, it was felt that if the closure time could be kept to less than 10% of transit time, the instantaneous closure assumption would be satisfied. Using the extreme precautions for alignment given later, this duration of closure time can be achieved. As a verification that this criterion is valid, the experimental data show no effect on results for closure times varying from 1 to 25% of the transit time.

Stress Profile Assumption

Having met the instantaneous closure requirement, we have confidence of achieving a well defined input into the specimen impact face. We must now consider if the stress profile for all points within the specimen may be considered one-dimensional and time-independent for times after the wave arrival. This requirement is opposed from a practical viewpoint by the instantaneous closure requirement. The $\frac{1}{2}$ in. diam by $\frac{1}{4}$ in. long specimen chosen is obviously a bounded solid and the effects of the boundedness[21] of the specimen on the stress profile at all points must be considered.

Although the use of an ill-defined bounded solid offers extreme complications for most solids, in the case of a stress-transducing solid as used here the effect of the boundedness may be determined by varying the geometry of the specimen and noting the effect on the charge release characteristics. As the wave front moves along the boundary between the quartz and the surrounding potting material, conditions of transient stability require that shear and unloading waves be generated. These waves travel inward at velocities dependent upon the particular elastic constants involved. In the case of quartz, a piezoelectric response would be expected from these unloading effects. Since the unloading effects from the lateral boundary occur at fixed velocities relative to the longitudinal velocity, the diameter-to-length ratio of a specimen is a valid parameter to use for observing the effects of the boundaries. The effects must be reserved for experimental determination.

The actual determination of the one dimensional strain piezoelectric constant must be made on a very thin specimen (large d/l) but this can be done at high impact velocity where instantaneous closure is easier to achieve if linearity has been confirmed up to that velocity. Although they are more difficult to execute, experiments can also be conducted with the charge being observed from only the center portion of a large diameter specimen. Because of the geometry of the experimental arrangement, one would not expect to see one-dimensional stress behavior.

THE EXPERIMENTAL ARRANGEMENT

To attain instantaneous closure as required by the analytical development, special experimental precautions are required. The importance of the necessity for precise alignment cannot be overemphasized. To accelerate the projectile to the desired impact velocity, an extensively modified U. S. Army 40-mm antiaircraft gun was used. To achieve the required precision of alignment, it is necessary to arrange the impact so that it occurs while the projectile is still being guided by the bore of the gun. The gun was accurately machined to a smooth bore and provisions were made to attach the specimen directly to the end of the barrel. Various impact velocities are achieved by using different amounts of T28EI propellant fired with a T104E6 electric primer. The gun as modified has the capability of achieving carefully aligned impacts for a velocity range of from 200 to 3100 ft/sec.

The o.d. of the flat-faced steel projectiles are ground 0.001 in. smaller than the bore of the barrel. The flat impact face of the projectile is ground so that it is less than 0.0002 rad out of perpendicular to the sides of the projectile. An x-cut quartz cylinder of larger diameter than the

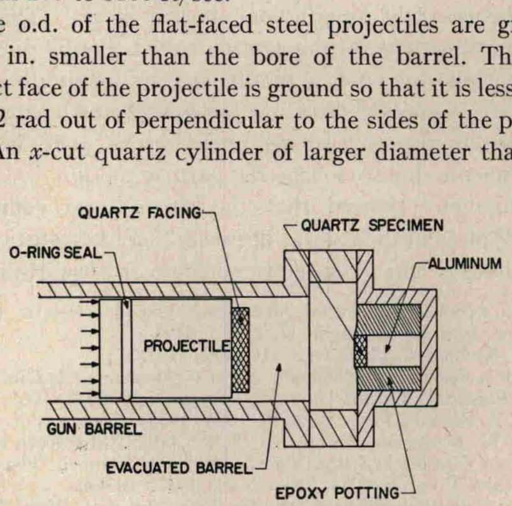

FIG. 3. Schematic of the impact.

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[21] In stress wave propagation there are two extremes of boundedness that are well defined; the solid infinite in lateral extent and the wire. In the former case there are no effects on the wave profile from lateral stress-free boundaries. In the latter case the solid is subjected to no lateral confining stresses. "Boundedness" as here used is meant to imply the relative position of the specimen between the two extremes in so far as its effects on the wave are concerned. The solid infinite in lateral extent has no boundedness, and the wire approaches infinite boundedness.

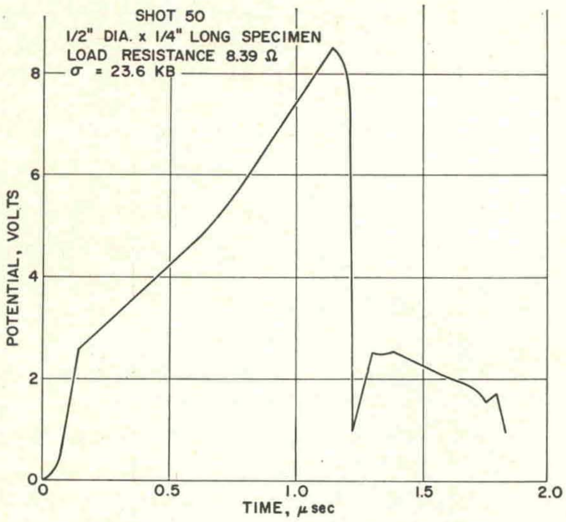

FIG. 4. Experimental current waveform for $D/l=2$.

quartz specimen is cemented to the impact face of the projectile so that an impact of quartz upon quartz is achieved. The dimensions of the quartz facing are chosen large enough to prevent the arrival of wave reflections at the specimen during the time of interest of the experiments. Figure 3 illustrates the impact end of the barrel showing projectile and specimen.

Since the impact occurs while the projectile is in the bore of the gun, it is necessary to measure the impact velocity in the bore of the gun. This is accomplished by three velocity stations 0.1000 ft apart with the last station 2 in. from the impact surface. The measurement is made by means of 0.010-in diam electrically charged pins which protrude through the side of the barrel. The projectile electrically grounds each pin during its passage, thereby discharging R–C circuits of short time constant characteristics. These signals are used to actuate two Hewlett–Packard 524C electronic counters with a least count of ±0.1 μsec. By using three stations and two counters, the acceleration of the projectile can be measured and corrections made for the slight change in impact velocity from the last station to the impact surface. An error analysis of the system shows that the impact velocity obtained is accurate to a precision of at least 0.5%.

The quartz specimens used are short right circular cylinders of various diameters and lengths. The specimens used presently are x cut with the specimen oriented so that the x axis is aligned with the axis of the impacting projectile. With this arrangement, the stress wave produced by the impact travels along the x axis. The positive and negative electrical orientation of the x axis relative to the impact face is noted for all experiments. Polishing the specimen to a plate glass finish makes it possible to observe any visible flaws and to attain extremely flat surfaces.

The quartz specimen is placed upon an electrode of 7075 aluminum of the same diameter as the specimen and of a length long relative to the specimen. Aluminum was chosen to minimize acoustic mismatch and an epoxy potting was cast around the specimen and rear electrode for dielectric strength. This precaution is necessary due to the high electric fields developed as the stress wave propagates through the specimen. As shown in Fig. 3, the specimen and potting are placed in a Lucite specimen holder. This holder serves as a mold for the potting and as a means of attaching the specimen to the end of the barrel. The flat surface of the holder mates with a flat end surface of the barrel which is accurately perpendicular to the bore. The specimen holder is precisely machined after the specimen is assembled so that the flat mating surface is parallel to the impact face of the quartz within 0.0005 rad. With more elaborate care this can be held to 0.0002 rad.

Aluminum foil 0.001 in. thick was placed on the impact surface to serve as the front electrode. Both surfaces of the quartz used as the projectile facing were grounded with aluminum foil to prevent any of the charge developed on the projectile facing from being introduced into the circuit measuring the charge released from the quartz specimen.

When the specimen is attached to the end of the barrel, the bore of the gun is completely closed at both ends. To prevent air shock formation ahead of the projectile, pressure buildup as the projectile closes on the specimen, and the premature discharge of the various charged pins in the barrel, the barrel of the gun is evacuated to a pressure of approximately 10^{-2}mm Hg.

INSTRUMENTATION

The charge produced by a given impact is measured by means of a low resistance current-viewing resistor. The potential drop across this resistor as a function of time is observed on Tektronix 545 oscilloscopes using type L preamplifiers. The oscilloscopes and preamplifiers are calibrated frequently and precise time and voltage calibration traces are recorded on each record. Care is taken to position the traces on the same portion of the face of the oscilloscope with the pulse being recorded on the center 4 cm of the oscilloscope face. This is done to minimize errors caused by the nonlinearity of the oscilloscope face. The signal is transmitted through coaxial, low loss cable, RG-8/u. The traces are recorded on Polaroid type 47 film. The resistance of the current-viewing resistor is measured frequently to a precision of 0.2%.

The data from the Polaroid pictures are enlarged by a factor of about 5 by using a Telereader, a commercial optical data projector, in conjunction with a Moseley X–Y plotter. The charge release is obtained by mechanical and electronic integration of the area under the current versus time record for the initial transit of the stress wave through the specimen. Using the precautions as given above, the estimate of the experimental precision of the reduced charge data is ±3%.

TYPICAL RESULTS

Typical current versus time records are shown in Figs. 4 and 5 for two different specimen geometries. It is seen that the exit of the wave front from the specimen is marked by a definite drop in the current. Although the current does not drop completely to zero in all cases, the record is well defined if one considers only the time corresponding to the initial wave transit. Times after initial transit are not well defined since slight acoustic impedance mismatches at the aluminum electrode quartz interface cause wave reflections of unknown amplitude. The amplitude of this reflected wave is also dependent on the stress for stress greater than the dynamic yield point of the aluminum. Also complicating the later times are unloading waves from the boundary which are still present in the crystal after the exit of the initial wave front.

The charge released due to the initial transit of the wave is taken as the integral of the current versus time record during the indicated transit time.

The variation in waveform with specimen geometry is quite typical and behaves in a predictable fashion. This is explained by an added piezoelectric response from the unloading waves propagating inward from the lateral boundary. Electrical fringing effects would also be expected to be present for certain geometries. As shown by the $d/l=10$ waveform in Fig. 5, the waveforms approach the ideal square current pulse for the less bounded solids.

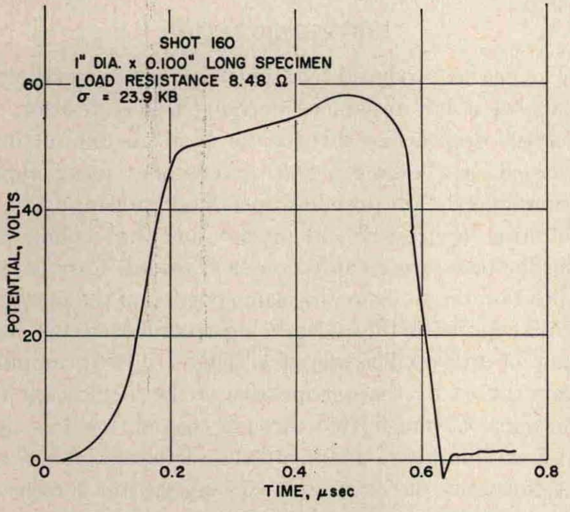

FIG. 5. Experimental current waveform for $D/l=10$.

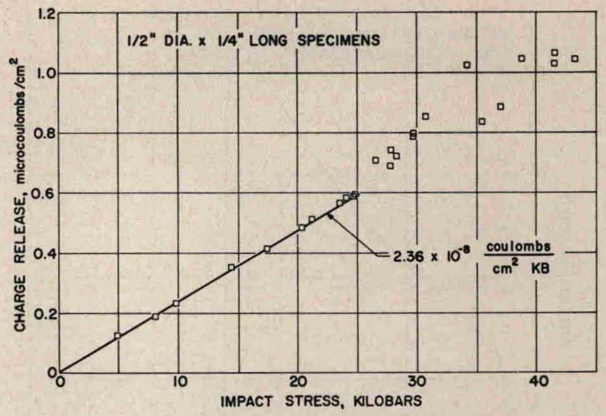

FIG. 6. Experimental piezoelectric relationship for $D/l=2$.

The piezoelectric behavior is illustrated in Fig. 6 by results published previously.[22,23] The experimental points shown in the linear range are all within ±2% of the linear least squares fit to the data. This is within the ±3% estimate of experimental precision for the charge data. A definite linear region is observed which extrapolates to zero within the experimental precision of the data. The slope of the line is an apparent piezoelectric constant which is found to be geometry dependent. The apparent constant approaches the low signal one-dimensional strain behavior for the large diameter-to-length specimens. Anomalies are also noticed in specimens oriented for negative electrical signal. A complete report on the results of the investigation is to be published later.

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[22] R. A. Graham, J. Appl. Phys. **32**, 555 (1961).

[23] R. A. Graham, Bull. Am. Phys. Soc. **5**, 511 (1960).

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