Piezoelectric Polymer Films for Fuze Applications

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Washington, D. C. 20234

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Prepared for
U. S. Army Frankford Arsenal
Philadelphia, Pa. 19137
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This document has been prepared for the use of the U.S. Army Frankford Arsenal. Responsibility for its further use rests with that agency. The measurement results reported are derived from a statistically small number of specimen devices, and the conclusions that may be drawn are therefore tentative.

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James A. Baker, III, Under Secretary
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NATIONAL BUREAU OF STANDARDS, Ernest Ambler, Acting Director
ABSTRACT

Described is the development of polymer piezoelectric devices with sufficient electrical output in response to impact to demonstrate the feasibility of using polymer elements in place of ceramic elements in ordnance fuze systems. To satisfy this requirement, piezoelectric polymer elements have been constructed capable of providing at least 0.3 mJ into a 4-Ω load, in response to a force of 89 kN (10 tonsf). Total activities of up to 360 pC/N have been achieved from elements built up from 30 sheets of 25-μm poly(vinylidene fluoride). Test methods developed to characterize both single-sheet and total-element activities are described, as are poling procedures and details of element construction. Mathematical analyses of polymer piezoelectric output into resistive-capacitive loads are presented for static and dynamic cases. Experimental results and theoretical predictions are shown to be in agreement to within the 10% estimate of measurement accuracy.
SYMBOLS USED IN THIS REPORT

A, area

C, polymer capacitance [first defined in appendix A, (10)]

d, piezoelectric strain tensor [with elements $d_{nm}$]

d_{33}, longitudinal piezoelectric strain constant (an element of d)

D, electric displacement vector [with components $D_n$]

E, electric field vector [with components $E_n$]

E, energy delivered to L

$E_0$, energy parameter [first defined in appendix B, (39)]

$\delta$, function relating E and Y [first defined in appendix B, (40), for triangular force pulse]

F, force vector applied normal to polymer piezoid surface

$F_0$, applied force amplitude

g, piezoelectric stress tensor [with elements $g_{nm}$]

g_{33}, longitudinal piezoelectric stress constant (an element of g)

g, function relating E and Y [first defined in appendix B, (59), for haversine force pulse]

i, current

$i_G$, current generated by P [first defined in appendix A, (13)]

$L$, polymer thickness

$L$, electrical load; the subscript "L" denotes that the quantity so modified refers to the electric load; examples: $C_L$, capacitance of L; $R_L$, resistance of L; $Q_L$, charge on $C_L$; $i_L$, current through L; $V_L$, voltage across L.

P, poled active portion of polymer; the subscript "P" denotes that the quantity so modified refers to the polymer P portion as distinguished from the S portion; examples: $C_P$, capacitance of P; $R_P$, resistance of P; $Q_P$, charge on $C_P$; $i_P$, current through $R_P$. 
Q, charge
Q_o, open circuit charge [first defined in appendix B, (30)]
R, resistance
S, unpoled shunt portion of polymer; the subscript "S" denotes that the quantity so modified refers to the polymer S portion as distinguished from the P portion; examples: C_S, capacitance of S; R_S, resistance of S; Q_S, charge on C_S; i_S, current through R_S.

\( t \), time
\( t_0 \), pulse width of \( V_L \) [first defined in appendix B, (44)]
T, piezoelectric stress vector [with components \( T_{mn} \)]
V, voltage
\( V_0 \), open-circuit voltage [first defined in appendix B, (40)]
Y, the product \( \nu \tau \)
\( \beta \), dielectric impermeability tensor [with elements \( \beta_{nm} \)]
\( \epsilon \), dielectric permittivity tensor [with elements \( \epsilon_{nm} \)]
\( \zeta \), capacitive parameter [first defined in appendix A, (23)]
\( \theta \), phase shift in oscillatory component of \( V_L = V_L(t) \), for haversine force pulse.
\( \nu \), the reciprocal of the circuit decay time [first defined in appendix A, (23)]
\( \tau \), pulse half-width of applied force
\( \phi \), phase shift in oscillatory component of \( V_L = V_L(t) \), for half-sine force pulse.
Piezoelectric Polymer Films for Fuze Applications

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1. INTRODUCTION

1.1 Objectives

The central objective of this project was to develop polymer devices which would be as effective as the piezoelectric ceramic disks presently used in fuze systems and which would provide low-cost construction, mechanical flexibility, and versatility of electrical arrangement. In order to achieve this goal, it was necessary to increase the outputs of piezoid devices by developing more effective poling and stacking techniques. (The term "poling" refers to the process by which as-received polymer is rendered piezoelectrically active and is described in 2.2. A stack incorporates a sandwich of two or more individual sheets into a single structure, with leads, shields, and the like. The active polymer part of the stack is referred to as a piezoid.) The final goal was the production and delivery of polymer stacks having an electrical output comparable to that of the ceramic disks. This would enable the sponsor to set an achievable standard for piezoid devices to be employed in fuzing systems.

Objectives related to the development and delivery of polymer devices included (1) the development of methods to increase piezoelectric activity, (2) the development of methods to characterize piezoelectric activity under a number of loading conditions, (3) the application of such methods to piezoids developed in the work, and (4) the derivation and formulation of expressions relating piezoid output to applied force.

1.2 Accomplishments

This project was characterized by (1) a growth of understanding of the nature of piezoelectricity in polymers and of the practical difficulties in manipulating fairly large areas of polymer sheet, together with (2) a similar growth of understanding of the differences and similarities between the electrical properties of piezoelectric polymers and piezoelectric ceramics. This improvement in understanding was shared by the NBS staff and the project monitors of Frankford Arsenal through frequency formal and informal contacts.

The initial work statement of July 9, 1973, called for NBS to "fabricate samples and carry out joint experimental studies with the Pitman-Dunn Laboratories, Frankford Arsenal on piezo-polymer films." Shortly after experimental work was begun, it was found that the intended active areas of 30 cm² could not be effectively electroded, poled, and stacked by the techniques previously developed and used on smaller areas. With continued improvement in poling procedures, NBS was able to produce sheets with increasingly higher activity. Thus the available energy output per stack was continuously raised over the course of the project and the minimum desirable device figure of merit was raised accordingly by the sponsor. By September, 1974, the characteristics of piezo-polymer stacks delivered
to the sponsor by NBS represented greater activity from a smaller piezoid volume than had originally been considered attainable. Accordingly, the new values were taken by the sponsor as defining goals for any future polymeric devices for the intended test purposes: piezoid stacks with an active area of 30 cm² and a maximum thickness of 0.7 mm having a static piezoelectric activity of 280 pC/N and capable of delivering 2000 ergs (2.0 x 10⁻⁴ J) into a 4-Ω load in response to a force of 89 kN (10 tonsf) which has a rise time of 10 μs.

With respect to cost goals, at the time of project termination the estimated direct labor cost of producing and testing a sixteen-sheet stack had been brought below $500. This figure applies to laboratory conditions; presumably, the cost would be considerably reduced in mass production.

The development of polymer devices for ordnance fuze applications has resulted in the following specific accomplishments:

1. Development and improvement of poling techniques to achieve piezoelectric activities of 12 picocolombs per newton (pC/N) for a single sheet of poly-(vinylidene fluoride) [PVF₂] 25-micrometers (25-μm) thick, with 14 pC/N realisable on the basis of a 50% success ratio,

2. Development of stacking methodology to achieve a piezoelectric activity of 360 pC/N from a piezoid of 30 sheets of 25-μm PVF₂,

3. Delivery of stacks representative of the current level of activity, (see table 5),

4. Development of a quasi-static, or d-c, method of test to characterize piezoelectric activity of both single sheets and stacks, using a press,

5. Development of a dynamic method of test to characterize piezoelectric activity of both single sheets and stacks, using a drop-test machine,

6. Derivation of an expression relating piezoelectric and pyroelectric activity, so that measurements of either type of activity may be used to characterize the other [1]*,

7. Development of the theoretical basis for a method of test to measure the compliance of bonding layers in a stack [1], and

8. Development of theoretical analyses describing the quantitative behavior of piezoelectric polymer in response to applied force, including derivation of the relationship between electrical and mechanical parameters, and of expressions for optimizing piezoid output into a given electrical load.

*Figures in brackets indicate the literature references in section 6.
In developing piezoids with higher levels of energy output, it was necessary to electrode, pole, and test large numbers of single sheets and to assemble and test large numbers of stacks. In the course of this work test methods were developed, discarded, or modified as needed; test equipment was designed, tried, modified, and improved, and many sheets and some stacks were tested to destruction. (As one example, a stack was tested in an air gun at Harry Diamond Laboratories to investigate the ability of a new type of electrical connection to survive violent impact; as another example, several small stacks were destroyed while various bonding agents and techniques were tried in the development of a method to attach leads and glue the poled sheets together.) The understanding and improved techniques derived from this testing program made it possible to achieve the more important of the specific accomplishments listed above.

2. EXPERIMENTAL DEVELOPMENT

2.1 Background

Piezoelectricity in stretched and polarized sheets of PVF$_2$ was first reported and studied by workers at several Japanese research institutes [2, 3, 4]. Recently, very thin, uniform films of this polymer have become available. Among the advantages of this material are that it has low density, that it is available in sheets (hence devices may be fabricated with surface areas large compared to those of ceramic piezoelectrics), and that it is sufficiently flexible to permit fabrication into a range of shapes. Through the choice of sheet thickness, number of sheets, and electrical connection, a stack may be designed to have specified capacitance and to deliver a maximized electrical output into a given load, in response to an applied force.

At the inception of this project, limited experience was available with active areas as large as 30 cm$^2$; further, single-sheet piezoelectric elements had been made on what was for the most part a one-at-a-time basis, and there was no developed stacking technology. Thus, new methods and apparatus were required to: (1) extend the vacuum deposition technique to routinely apply metal-film electrodes of 30-cm$^2$ area, to apply electrodes to one side of a series of polymer elements in quick succession (i.e., without requiring separate pump-down and cycling of the vacuum chamber for each element), and to support the polymer sheet while the electrodes were being applied in such a manner as to minimize the effects from heating (especially wrinkling of the polymer sheet), (2) improve and extend poling procedures to enable the poling of several large samples at once, to utilize higher electric fields and temperatures without introducing wrinkles in the polymer, and to minimize the number of short circuits occurring through the sample during poling, (3) produce a bonded stack from a number of single-sheet elements, and (4) attach leads for parallel electrical connection of the single-sheet elements in a stack.

In addition, methods were required that were appropriate for the tasks of characterizing the piezoelectric activity of single-sheet elements, of sandwiches of elements being built into a stack, and of completed stacks.
2.2 Sample Preparation

2.2.1 Deposition of Electrodes — Aluminum electrodes approximately 0.10-μm thick were deposited on each side of the 25-μm polymer sheets using vacuum evaporation apparatus. Short lengths of the pure aluminum wire used were coiled around a resistively heated tungsten helix; the heating current was not applied until the chamber reached a pressure of 2.7 × 10⁻⁴ pascal (Pa) during pump down.

Polymer surfaces to receive electrodes were rinsed in ethyl or isopropyl alcohol before being placed in the vacuum chamber. A fixture was designed with two rollers. The polymer sheet to receive electrodes was in the form of a long strip and was initially wound on the feed roller oriented so that when unwound the intended side would face the evaporation apparatus. The take-up roller was driven by means of a mechanical link through a vacuum seal to a crank outside the vacuum chamber. The number of elements that could be produced in one run was limited in principle by the diameter of the rollers and associated clearances; however, in practice the limit was the number of evaporation that would be carried out before more aluminum wire had to be affixed to the tungsten helix.

The electrodes were deposited through a mask which consisted of four circular holes 30 cm² in area (diameter = 6.2 cm) in a 24-cm-square sheet of stainless steel. The holes were symmetrically oriented about the center of the mask with a center-to-center spacing of 7.5 cm in one direction (across the roll width) and 8.7 cm in the other (along roll length). Each of these holes had an associated slot, 3 by 12 mm, cut in the mask from the edge of the hole outward along an extension of the hole center line parallel to the intended long axis of the polymer film, with all slots on the same side of the holes. See figure 1. After one side of the polymer sheet strip had electrodes applied, the strip was rewound on the feed roller so as to have been effectively rotated through an angle of 180° (the original "beginning" end wound on the feed roller first) and turned over with former bottom side now top side. The result of the orientations so achieved was successively to evaporate electrode "tails" on opposite sides of the strip pointing in opposite directions. During poling and in subsequent use, electrical connections were made to these projections.

When the electrode thickness is less than 0.07 μm, the resistance across the 30-cm² circular face of the electrode, measured from near edge to near edge along a diameter, exceeds 2 Ω. This measurement was made in air using probe tips with a radius of curvature of 1.25 mm. The resulting activity of the poled films is several orders of magnitude less for electrode thicknesses corresponding to resistances greater than 6 Ω than for those corresponding to 2 Ω; the activity was found to be independent of electrode thickness for thicknesses greater than 0.10 μm. (At this thickness, the resistance was near 1 Ω.) If the electrode thickness were permitted to exceed 0.15 μm, the aluminum tended to flake off. An aluminum electrode thickness of 0.1 μm (1000 Å) has proven satisfactory and is recommended. Aluminum was chosen for the electrode material because it is satisfactorily conductive at reasonable thickness, because at such thicknesses it adheres well to alcohol-rinsed PVF₂, and because it is an easy material to evaporate, having a sufficiently low melting temperature not to require the use of a crucible.
2.2.2 Poling — In preparation for poling, a sandwich was made by placing a 125-μm sheet of PVF₂ on a bottom 0.32-cm sheet of polytetrafluoroethylene (PTFE), followed by the electroded polymer element to be poled, another 125-μm sheet of PVF₂, and a top 0.32-cm sheet of PTFE. Electrical connection to the top and bottom electrodes was made by using two thin strips of 12-μm-thick tin foil. This assembly was then placed between the platens of an hydraulic press. A load of approximately 30 kN was applied to press the sample and to prevent it from wrinkling during poling. The desired electrical field was applied across the sample by connecting the tin-foil leads to a high-voltage d-c power supply. The sample was heated to the desired temperature by means of heaters embedded in the platens of the press, and then cooled to room temperature by running water through cooling passages also in the platens. The field was applied to the polymer during the entire heating and cooling cycle. The PTFE served to provide electrical isolation of the sample from the hydraulic press structure. The 125-μm sheets of PVF₂ were used as liners because the surface of the insulating sheets was not smooth. The temperature was monitored by means of thermocouples inserted into the sandwich.

During the assembly of the sandwich, care was taken that the polymer element and the inner sides of the 125-μm PVF₂ were clean and free of dust in order to prevent dirt particles from being pressed into the polymer surface during poling. The thickness of the polymer would be reduced at the embedded particle, and therefore the effective electrical field would be locally high. Embedded particles are to be avoided as potential failure sites.

A regulated 10 kV d-c power supply was used to provide the poling field. The poling voltage typically ranged between 2000 and 2400 V across the 25-μm sheets, with some of the early sheets poled at 1200 V. The 2400 V potential produced a higher activity than did 2000 V, but also resulted in a higher percentage of the sheets failing. For example, the percent success-failure ratio experienced in the laboratory for poling at 2400 V and 120°C was 50-50; for sheets poled at 2000 V and 110°C the percentage of failures dropped to 10. The sheets were heated to approximately 120°C and then cooled. The temperature cycle was approximately 15 min for heating and 10 min for cooling.

2.2.3 Stack Construction — Each poled sheet had a "high side," this being the side with electrodes connected to the positive, or high, side of the d-c poling supply. As the first step in making a multi-sheet stack, two sheets at a time were bonded together, high side to high side, with a tin-foil lead providing electrical contact to the internal high electrodes. Tin-foil leads were attached to the electrode tails with silver-filled rubber paint. Bilaminate pairs were then bonded together with tin-foil leads providing contact to the internal low electrodes and extending from the opposite edge (i.e., 180° away from the edge with high-side leads). Successive high-side leads were aligned one over the other, as were low-side leads. The bonding agent was a solution of one part contact cement to five parts toluene*. Experience with stack construction showed that it was desirable during bonding to prevent entrapment of air bubbles and avoid wrinkling sheets, as both conditions degraded performance.

*Considerable experimentation was conducted with various bonding agents. Contact cement meeting Federal Specification MMM-A-130a has been satisfactory.
In the manner described the stack was built up of \( 2n \) sheets. The partially completed stack had \( n \) superposed high-side leads extending from one edge and \( n - 1 \) superposed low-side leads extending from the other edge. Two more tin-foil leads were then attached to the outside low-side electrodes. The bundle of high-side leads was bonded to form a single lead with silver-filled rubber paint and reinforced with either copper or aluminum tape. An exterior coat of silver-filled rubber was applied to the reinforced lead. Similar treatment was given to the low-side leads. Figure 2 shows a laminated stack at the bottom (A), before the outer coatings of heavy aluminum tape (providing electrical shielding and mechanical protection) were applied. B shows a single sheet with electrodes deposited on both sides; C shows a similar sheet with one foil electrode attached (which was destined to be an outer low-side lead for the stack); and D shows two sheets bonded together with the high-side lead at the left. E shows another sandwich of two bonded sheets with high-side lead. The laminated stack was made up by alternating D-and-E sandwiches.

2.3 Static Testing Procedure

Each poled sheet separately, and the various small stacks which were an intermediate stage in the fabrication of completed stacks, were tested quasi-statically on a hydraulic press.

When the piezoid sheet or stack is compressed, negative charge collects at the high-side electrode; after discharge, and upon release of the mechanical force, negative charge collects at the low-side electrode. The test procedure was as follows: With an open-circuit configuration, (see figure 3), either the piezoid by itself (with capacitance \( C \)) or in parallel electrical connection with a standard capacitor (capacitance \( C_L \)) was charged by means of compression or release of pressure (static force loading \( F_0 \)) by means of the single-pole switch show in the figure. Discharge of the charged piezoid into the preamplifier of a storage oscilloscope permitted measurement on the stored trace of the voltage developed across the capacitor(s).

The magnitude of the charge developed by the polymer is \( Q_0 \). The voltage \( V_0 \) measured when the piezoid is not connected to \( C_L \) is \( Q_0/C \).

With \( C_L \) in the piezoid circuit, the charge developed \( Q_0 \) is shared between \( C_L \) and \( C \). Under the open-circuit condition there is a common voltage across the two capacitors. In order to obtain a measure of charge output and the quasi-static capacitance of a given piezoid, two voltages, \( V_0 \) and \( V_L \) were measured:

\[
V_0 = \frac{Q_0}{C} \text{ and } V_L = \frac{Q_L}{C_L} = \left( \frac{Q_0}{C} - \frac{Q_L}{C} \right)
\]

(1)

where \( Q_L \) and \( Q_0 - Q_L \) are the equilibrium values of the charge on capacitors \( C_L \) and \( C \), respectively. From (1),

\[
C = \left( \frac{V_0}{V_L} - 1 \right)^{-1} C_L \text{, and } Q_0 = CV_0,
\]

(2)
From these two formulas and from measurements of \( V_O \) and \( V_L \), the static capacitance \( C \), the charge released \( Q_0 \), and the activity \( Q_0/F_0 \) may be calculated.

2.3.1 Summary of Single-Sheet and Small-Stack Activities — Presented are data taken from August 28, 1974, through September 14, 1974, while stack No. 7 (30 sheets; see table 5) was being constructed. Four samples in each 15 x 18-cm sheet (as obtained from the vacuum aluminizing apparatus) were poled together in parallel; then each sample in the batch of four was assigned a common batch number and a sample code letter a through d. The active areas were cut apart from each other into four separate single sheets.

Poling voltage (V) and maximum temperature reached during the poling cycle (°C) are given in table 1, along with capacitance \( C \) (nF) and piezoelectric activity \( Q_0/F_0 \) (pC/N) for eleven batches. \( C \) and \( Q_0/F_0 \) are the averages of the measured values for the four samples in each numbered batch. Both quantities were obtained by measurement and calculation using (2) for applied forces up to 45 kN. The variance in the capacitance of a single sample depends on the reproducibility of the force imposed under the measurements implied by (1). Analysis of repeated measurements resulted in an error estimation \( \delta Q/Q = \delta C/C = 0.092 \) for single sheets. The data for the individual sheets are contained in table 2 (parts A and B) in the rows labeled \( n = 1 \). The variation among samples poled under identical conditions is exemplified by results from measurements on samples 34a-d, 36a-d, and 38a-d. The average values and the variation as determined by the standard deviations for these batches are:

- Batch 34 \( C = 14.1 \pm 1.1 \) nF, \( Q/F = 9.7 \pm 0.9 \) pC/N;
- Batch 36 \( C = 13.6 \pm 0.1 \) nF, \( Q/F = 9.5 \pm 0.7 \) pC/N;
- Batch 38 \( C = 13.7 \pm 0.0 \) nF, \( Q/F = 9.8 \pm 0.5 \) pC/N.

In the two parts of table 2 are shown measured values for the capacitance and piezoelectric activity for stacks having \( n = 1, 2, 4, 6, 8, 9, 10, 14, \) and 15 sheets. The entries are arranged in chart form to show from which particular sheets each stack or intermediate stage was built. For \( n > 2 \) there are three sets of entries for each stack or stage: the first set gives the measured values [capacitance (nF) on top, piezoelectric activity (pC/N) on bottom] for the particular stack of \( n \) sheets; the second set gives average sheet values, i.e., the first set values divided by \( n \); and the third set gives weighted average values based on the expected contribution of the constituents. That is, the values at the previous stage of each of the constituents were averaged with weighting factor equal to the number of sheets in the constituent stack.

The two intermediate stacks of 15 sheets were assembled into stack 7. One-thirtieth of the measured value of 360 pC/N yields \( C = 12.0 \) nF, and \( Q/F = 12.0 \) pC/N. The average values for all 30 of the single sheets are \( C = 13.6 \pm 0.5 \) nF and \( Q/F = 9.8 \pm 0.7 \) pC/N. Also, the average of the measured values per layer for the two stacks of 15 sheets is \( C = 12.9 \) nF and \( Q/F = 11.5 \) pC/N. In this example, the measured value for the piezoelectric activity per layer increases as the stack thickness (proportional to \( n \)) increases. An explanation may be that sheets located near the center of the stack undergo greater distortion than sheets near the surface when a given force is applied to the stack.
2.4 Analyses of Piezo-Polymer Electrical Outputs

2.4.1 Summary of Mathematical Development – Appendices A through E are mathematical analyses of the output from a polymer piezoid under various conditions. Appendix A is a circuit analysis of an active polymer element under dynamic loading conditions. This analysis permits calculation of energy and voltage outputs as seen by an R-C load as the piezoid experiences impact. Appendix B contains an evaluation of the formulae for the output due to triangular, half-sine, and haversine force pulses (the haversine pulse has the shape of that of the half sine squared). Appendix C constitutes an analysis of the open-circuit, static-loading voltage and charge levels developed across a piezo-polymer and the subsequent electrical output upon application of an electrical load. Appendix D is a table based on the results developed in appendices B and C and constitutes a summary of polymer piezoid outputs (voltage, energy, charge) under given conditions.

Appendix E is a discussion of the output from a piezoid sample which has non-uniform conductivity and dielectric constant. It was assumed in the analysis given in appendix A that conductivity and dielectric constant are uniform over the sample thickness. It is shown that under practical conditions no modification of the formulae developed in appendices A through C is necessary.

The theoretical conclusions show that to obtain the largest possible transfer of energy stored in the stack, either a series-or parallel-connected stack of \( n \) sheets may be constructed, and a switch arrangement should be used with a value of \( R_L C \) much smaller than the transfer time (the series stack value of \( C \) decreases as \( n^{-1} \)). For the dynamic loading arrangement, \( R_L C \) should be chosen to satisfy the relation \( R_L C = \frac{1}{2} \tau \); then in time \( \tau \) an energy transfer of approximately 40% of the energy stored in the stack, 40% of \( Q^2/2C \), will take place. [As a numerical example, consider the values \( \tau = 10 \mu s, n = 20, C = 20 \times 12 \, \text{nF}, \text{and } R_L = 20 \, \Omega \). For \( Q/F_0 = 300 \, \text{pC/N} \) (2.7 \( \mu \text{C/tonf} \)), the maximum output from \( F_0 = 89 \, \text{kN} \) (10 tonsf) would be approximately \( 0.73 \times 10^{-3} \, \text{J} \) \((7,300 \, \text{ergs})\). For the same conditions, but using \( R_L = 100 \, \text{M\Omega} \), the energy transferred would be reduced by a factor of \( 0.67 \times 10^{-6} \); while if \( R_L = 4 \Omega \), the energy transferred would be reduced to 0.5 of the maximum value.]

The theory also shows that to obtain the largest output voltage from a stack, the stack should be series connected and the quantity \( v_T \) should be \( \ll 1 \) \((v_T\) is defined as the product of the reciprocal of the circuit decay time and the pulse half-width of applied force). 

2.4.2 Energy-Release Experiments on Piezo-Polymer Stacks Connected Across an Electrical Load – Described are dynamic and static energy-release experiments on three different multilayered stacks, each with individual component sheets connected in parallel.

2.4.2.1 The voltage output of stack No. 7 was measured across various electrical loads under impact conditions. The stack was mounted on the impact table (a steel cylinder) of a drop-test machine, and the electrical output into a given load was measured and recorded during the impact sequence. A commercially
available accelerometer was attached to the upper face of the dropping carriage and had a specified sensitivity of 7.9 mV/gn* over the frequency range 5 Hz to 15 kHz. The outputs from stack and accelerometer were stored in a two-channel transient recorder and could be plotted as a function of time on an X-Y1-Y2 plotter and displayed on the screen of a monitor storage oscilloscope. A mass of 2.59 kg (corresponding to a weight of 5.69 lb), consisting of an aluminum impactor mounted on the carriage, was released from a height of 5 cm (2 in) to fall freely and impact the polymer stack. The impactor surface was a portion of a sphere with radius 23 cm (9 in). The voltage output from the calibrated accelerometer showed that the maximum force (F0) developed during impact was 23.8 kN (2.67 tonsf). The resistive load (RL) was varied between 4 Ω and 40 kΩ. The voltage was measured both with no capacitive load and with a capacitive load (CL) of 0.5 μF. Actually the transient recorder has an impedance (Rtr = 1 MΩ, Ctr = 15 pF) which is in parallel with the load; thus the net resistive load was equal to the product of RL and Rtr divided by their sum, and the net capacitive load was equal to the sum of CL and Ctr. The measured data are given in table 3. The energy delivered to the load during the force rise time τ was calculated from the data and is also given in table 3.

Table 3 shows that the voltage maxima and minima for different resistive and capacitive loads behave as predicted by the analysis of appendix B. The output pulse width to also agrees with the calculated value. However, the output voltage waveform for large ντ (small RL) was qualitatively different from that predicted for triangular and half-sine force pulses. For large RL a square wave would be the likely form expected, with the signal rising in a time much less than τ and then falling to VMIN at τ, with a fast decay time comparable to the rise time. In fact a gradual rise to VMAX at the time entered in table 3 was observed. This apparent discrepancy from predicted behavior is the result of the fact that both the triangular and half-sine waveform force pulses have a discontinuity in the time derivative of the force, dF(t)/dt, at t = 0; thus the current IG(t) would be abruptly turned on at t = 0. Because of the nature of the elastic properties of the polymer, the impacting force is better characterized by a waveform which has both zero derivative and instantaneous value of impacting force at t = 0. After t = 0 both F(t) and IG(t) should gradually rise from zero value. The voltage and energy integrals for the haversine waveform force pulse (which has these desired properties) have been evaluated in appendix B. There is agreement between the voltage pulse shapes predicted in appendix B, section 3, and those that have been observed during the drop-test evaluations of stack output.

A noticeable difference between the static and dynamic capacity of the test polymer stack was detected in the course of measurements. The capacitance of stack No. 7 was measured statically on the hydraulic press as described in 2.3 (and as also follows from the open-circuit analysis of appendix C) and found to be 0.36 μF. The stack capacitance was then measured at 1 kHz on an impedance bridge with the result of 0.25 μF. According to (31) or (51) of appendix B, the capacitance may also be calculated from the peak voltage data at large RL given in table 3:

\[ \zeta^{-1} = 1 + C_L/C = 22.5/7.5 = 3. \]

\[ g_n = 9.81 \text{ m/s}^2 \]
Since \( C_l = 0.5 \, \mu F \), \( C = (3 - 1)^{-1} \times 0.5 \, \mu F = 0.25 \, \mu F \). This result agrees with that of the bridge measurement.

The piezoid may be characterized by its frequency-independent static voltage response to unit force \( V_0/F_0 \). For stack No. 7,

\[
V_0/F_0 = (355 \, \text{pC/N})/(0.36 \, \mu \text{F}) = 1 \, \text{mV/N}. \tag{4}
\]

A comparison value for a typical piezoelectric ceramic (lead zirconium-lead titanate-4) is 0.2 mV/N.

The individual polymer sheets (average capacitance, 13.6 nF; average piezoelectric activity, 9.8 pC/N) also yield this characteristic voltage. Since \( V_0/F_0 = d_{33}/C_p \), and \( C_p \) is proportional to \( A \) (30 cm\(^2\) for the piezoid sheets), another quantity that may be used to characterize sheet performance is voltage output in a unit pressure field:

\[
V_0/A/F_0 = 3 \, \mu V/\text{Pa}. \tag{5}
\]

The difference between the effective charge output observed at low and high frequencies (between 0.35 \( \mu F \times V_0 \) and 0.25 \( \mu F \times V_0 \)) may be explained by assuming that \( C \) is frequency dependent while \( C_p \) is not (see appendix B). The dipole orientational contribution to the dielectric constant is frequency dependent according to Debye's theory of polar molecules [4]. The polarizability and relaxation frequency should be much lower for the polarized portion of the piezoid than for the unpoled portion according to the following explanation. The unpoled portion can respond in synchronism to a driving field, while the poled portion of the polymer is saturated. This situation causes the dielectric constant and the resistivity of the poled portion to be less than that of the unpoled portion [5]. The analyses in appendices A through C were carried out under the assumption that the dielectric constant and resistivity were homogeneous throughout the piezoid. In appendix E it is shown that under practical conditions even when the dielectric constant and resistivity vary through the material, the formulas are still valid.

The data in table 3 may be interpreted in terms of the haversine force pulse analysis presented in appendix B, and comparisons made between measured and theoretically predicted values. Consider as examples: { 1 } the time \( t_M \) at which \( V_L = V_{L_{\text{MAX}}} \) and the duration \( t_0 \) of \( V_L \) regarded as a pulse, { 2 } the behavior of the maximum and minimum (positive and negative) voltage peaks of \( V_L \), and { 3 } the relationship between the energy \( E \) delivered to the load and the load resistance \( R_L \).

In example { 1 }, for large values of \( R_L \) (corresponding to small values of the product \( \nu_\tau \)), the theory predicts that the time at which \( V_L \) reaches maximum value is equal to the rise time \( \tau \) of the force pulse and that \( t_0 \) equals \( 2\tau \). The measured values for \( R_L = 40 \, \text{k} \Omega \) confirm this prediction; that is, twice the time (measured from \( 0 \)) at which \( V_L \) is maximum equals \( t_0 \) (2 \times 225 = 450). For small values of \( R_L \) (corresponding to large values of \( \nu_\tau \)), the theory predicts that \( t_0 \) should be approximately equal to \( \tau \) with the \( V_L \)
maximum occurring at \( \tau /2 \). Again, the measured values for \( R_L = 4 \Omega \) confirm this prediction, with \( t_0 = \tau = 225 \), although the agreement is not as good for the time at which \( V_L^{\text{MAX}} \) occurs (2 x 100 is only approximately equal to 225).

In example {2}, the relative magnitudes of the positive and negative voltage peaks \( V_L^{\text{MAX}} \) and \( V_L^{\text{MIN}} \) agree with theoretical prediction: for large values of \( R_L \), the measured values of \( V_L^{\text{MIN}} \) tend toward zero [see (62)], and \( V_L^{\text{MAX}} \) depends on the value of \( C_L \) as given in (63) [see also (23)]. For small values of \( R_L \), the theory (60) predicts that \( V_L^{\text{MIN}} = -V_L^{\text{MAX}} \), and the measured values agree for \( R_L \) as large as 40 \( \Omega \). Further, theory predicts that the peak values exhibit no dependence on \( C_L \) for small values of \( R_L \); comparison of the measured values of \( V_L^{\text{MAX}} \) given in table 3 for two values of \( C_L \) confirms this independence \((0.14 = 0.15; 0.40 = 0.40)\).

In example {3}, the theory predicts (65) that the value of \( \nu_T \) for maximum energy transfer into the load should be approximately equal to \( \tau /R_{C} + 2.1 \). \( R_L \) therefore would be predicted to be \( \frac{225}{(0.25)(2.1)} = 430 \Omega \), which is in reasonable agreement with the measured value of 400 \( \Omega \). The normalized value, for maximum energy transferred into the load, \( E(\tau)/F_o^2 \), was computed from measurements to be 61.6 femtojoules per newton squared \( (\text{fJ/N}^2) \), which agrees well with the theoretically predicted value of 59 \( \text{fJ/N}^2 \). This value and other calculated values given in table 4 were calculated from equations derived in the haversine force pulse analysis (and also summarized in appendix D, condition 3):

\[
\begin{align*}
(V_L/F_o)^{\text{MAX}} &\approx \frac{\pi R_L C}{2\tau} \frac{V_o}{F_o} = 1.724 R_L \left( \mu \text{V/N} \right) \quad \nu_T \gg 1, \\
E(\tau)/F_o^2 &\approx \frac{R_L \pi \zeta}{8\tau} \left( \frac{V_o}{F_o} \right)^2 = 0.334 R_L \left( \text{fJ/N}^2 \right) \\
(V_L/F_o)^{\text{MAX}} &\approx \zeta V_o/F_o = 1000 \zeta \left( \mu \text{V/N} \right) \quad \nu_T \ll 1, \\
E(\tau)/F_o^2 &\approx \frac{3\sigma}{8R_L} \left( \frac{\zeta V_o}{F_o} \right)^2 = \frac{84.4}{R_L(\text{K})} \left( \text{fJ/N}^2 \right)
\end{align*}
\]

Comparison of the measured and calculated values of the various parameters given in table 4 shows good agreement between the experimental results and theoretical prediction.*

*From the general expression for \( V_L \), (56), \( V_L^{\text{MAX}} \) was calculated to occur at \( t = 0.78\tau = 175 \mu s \), and \( V_L^{\text{MAX}}/F_o \) was calculated to have the value \( (V_o/2F_o) \times (0.831) \times (1.156) = 480 \mu \text{V/N} \).
2.4.2.2 By means of mechanically charging an initially open-circuited piezoid which subsequently had its energy released when a preset stack voltage was reached, a pyrotechnic actuator (the electrical load) supplied by the sponsor was fired.

In order to test the predictions of appendix C, the circuit shown in figure 4 was designed to establish an open-circuit condition across the polymer stack. When the voltage resulting from a force applied to the piezoid rises to a predetermined level (the value used was 20 V), the Zener diode triggers the gate of the SCR, which in turn permits the energy stored in the polymer device to flow into the load. The load selected was a pyrotechnic actuator which on receiving 800 ergs \( (8 \times 10^{-5}) \) J changed its dc resistance from 3 \( \Omega \) to 2 \( \Omega \) and its capacitance from 1 \( \mu F \) to 1 \( nF \). The values given are approximate.

Stack No. 5 was used for this test with successful results. It was found that a force of 73 kN (8.2 tonsf) triggered the circuit and produced a signal of 23 V with the load being the input circuit of an oscilloscope (input impedance 1 M\( \Omega \), shunted by 18 pF). Stack No. 5 (88 sheets; see table 5) had a measured static output of 290 pC/N with a static capacitance of 1 \( \mu F \). The static voltage level was calculated according to (4) to be \( V_0/F_0 = 0.29 \text{ mV/N} \). The expected voltage level for the force (applied with the hydraulic press) was \( V_0 = 21 \text{ V} \). For these parameters \( (\xi^{-1} = 2 \text{ and } v^{-1} = \xi^{-1}R_L C = 6 \times 10^{-6}) \), the energy output to the actuator was calculated using (56) and (58) to be 1100 ergs delivered at approximately 10 V.

Note that if the actuator had been connected directly across the piezoid, a force of 73 kN with a rise time of \( \tau = 10 \mu s \) would have yielded \( V_L(\tau) \approx 6 \text{ V} \) and \( E(\tau) = 418 \text{ ergs} \). At 6 V, the threshold energy for the actuator is 1000 ergs; direct coupling therefore would not provide sufficient energy to the load.

2.4.2.3 An impact test was carried out at the air-gun installation in the Harry Diamond Laboratory in which a stack was directly impacted by a high-velocity projectile. A flat-nosed projectile with a mass of 1.38 kg was fired from a gun with a bore of 10 cm (4 in) at a polymer piezoid made from 20 sheets. The velocity at impact was determined to be 154 m/s (504 ft/s). The force sustained by the polymer stack was estimated by the air-gun test personnel on the basis of experience to be 450 kN (50 tonsf). A high-resistance probe \( (R_L = 100 \text{ M} \Omega, C_L = 3 \text{ pF}) \) was placed across the sample, and voltage output was recorded photographically from calibrated oscilloscopes which were triggered by means of a trip wire 40 \( \mu s \) before impact. Following impact, the recorded voltage rose linearly to a peak of 200 V at 0.48 \( \mu s \) after impact. According to calculation on the basis of the formulas given in appendix B, the voltage produced should be 400 V.\(^\dagger\) During the initial impact

\*\( v_L = 1.67; \xi(1.67) = 0.379 = 0.381 \), its maximum value; see appendix B.
\( ^\dagger v_L/R_L C = 0.48 \mu s/(100 \text{ M} \Omega) = 0.26 \mu F \) = 1.8 \( \times 10^{-8} \), which is much less than one; \( V_L(\tau) = Q_0/C = (233 \text{ pC/N}) (4.5 \times 10^5 \text{ N})/0.26 \mu F = 400 \text{ V} \).
duration of 0.48 $\mu$s, the energy delivered to the load was only 64 pJ because of the extreme mismatch of the 100 M\Omega load. Note that if $R_L$ had been chosen as 4 $\Omega$, then instead of 200 V, a voltage peak of 160 V and an energy output of 1.15 mJ would have been expected; while if $R_L$ had been chosen to optimize the energy output ($R_L = 1$ $\Omega$), a voltage peak of 90 V and $E^{MAX} (\tau) = 1.98$ mJ would have resulted.

3. DELIVERIES

A tabulation of the PVF$_2$ piezopolymer stacks which were delivered to Frankford Arsenal is given in table 5. The entries in the column labelled "Total Activity" were measured with an estimated sample single standard deviation of ?10% by the static open-circuit test method described in 2.3. The final column gives the average activity per sheet, the quotient of the values in the "Total Activity" column and the number of sheets in the given stack.

In all cases, the individually poled PVF$_2$ sheets had congruent 30-cm$^2$ circular aluminum electrodes vacuum deposited on both sides of the sheet. The thickness of each stack is made up of contributions from $n$ polymer sheets, $2n$ electrodes, and $n-1$ bonds. The nominal poling conditions for stack Nos. 1-6 were 95°C for the maximum temperature and 1200 V for the poling potential; to achieve more effective poling, the nominal poling conditions for stack Nos. 7-10 were 120°C for the maximum temperature and 2000 V for the poling potential.

4. SUMMARY AND RECOMMENDATIONS

4.1 Summary

A piezoelectric polymer transducer, with an active element of area 30 cm$^2$ and thickness less than 0.9 mm, has been developed that is capable of delivering into a several-ohm load an energy of 0.32 mJ in response to a 89-kN impact having a rise time of 10 $\mu$s.* In support of this development, specific poling methods and methods of stack construction were evolved.

Static and dynamic test methods have been developed to characterize the electrical output and piezoelectric activity of polymer elements, or piezoids. The static method makes use of an hydraulic press to apply force to the piezoid; in the dynamic method, the piezoid is impacted with a known force in a drop-test machine.

Theoretical analyses, in particular applying to both test-method situations, have been formulated to predict the electrical output of a stack under various conditions. The theoretical predictions are in good agreement with the experimental measurements that have been made, especially for the haversine-force pulse model of the drop-test situation.

*The values cited refer to Stack No. 7, which was slightly thicker than the 0.7-mm value specified. Stack No. 8 met all the requirements given in 1.2, with a thickness of 0.68 mm and an output under the specified conditions and into the specified load of 0.24 mJ.
4.2 Recommendations

Theoretical [6] and experimental studies [7] in our laboratory and elsewhere indicate that most of the activity of a polymer film occurs in very thin layers near each electrode and that the bulk of the material is relatively inert. A conclusion that requires further experimental verification is that higher activity should be achieved with the use of thinner sheets, since less "inert" material would be present. The activity of two 9-μm PVF₂ samples was measured in our laboratory and found to be approximately twice that of two 30-μm samples said to be poled with the same field strength according to the supplier. If more empirical evidence becomes available supporting the use of thin polymer sheets or films to achieve high piezoelectric activities, techniques of depositing very thin polymer films on suitable substrate material should be considered.

Other means of improving per-sheet activity, for example, multiple poling cycles, the use of high-frequency electrical poling pulses, and treatment such as stretching prior to electrical poling, require investigation.

The methods of test need to be further developed to improve the precision of results. The use of a commercially available force transducer for calibration is recommended.

Results of measurements on completed stacks show that the activity of the stack as a whole tends to be greater than the sum of the activities of the individual sheets. This phenomenon should be investigated, as it may hold clues to improved stack design leading to higher per-stack activities.
Output from Piezoid Sample Under Dynamic Loading Conditions

Voltage and charge measurements have been conducted during static- and dynamic-loading experiments on single polymer sheets and on stacks of active and inactive polymer sheets electrically connected in both series and parallel.

Consideration of the results of these tests has led to the equivalent circuit shown in figure 2. The polymer is modeled as consisting of two uniform parts: An active part in series with an inactive region. The active part is understood to coincide with the poled portion, is denoted by the letter "P", and has a thickness $\ell_p$. The inactive, unpoled portion may be regarded as a shunt with respect to P, is denoted by the letter "S", and has a thickness $\ell_s$. Each part is modeled separately as a parallel R-C circuit with a charge $Q$ on $C$ and a current $i$ in $R$. Theoretical and experimental results for a poled sample show that the surface regions are strongly poled, while the bulk of the interior is less effectively poled. Figure 3 shows the polymer element with a series load of resistance $R_L$ (with current $i_L$) and capacitance $C_L$ (with charge $Q_L$).

Consider a piezoelectric material. A pair of equivalent piezoelectric equations relate electric field $E$, electric displacement $D$, and stress $T$.[8]

$$E_n = \beta^T_{nm} D_n - g_{nm} T_m$$

and

$$D_n = \varepsilon^T_{nm} E_m + d_{nm} T_m,$$

expressed in Einstein summation convention.

In this convention, $\beta$ is the dielectric impermeability tensor (elements $\beta_{nm}$), and $g$ is the piezoelectric stress tensor (elements $g_{nm}$), which are related to the dielectric permittivity $\varepsilon$ and the piezoelectric strain $d$, respectively by:

$$\beta = \varepsilon^{-1} \quad \text{and} \quad g = \varepsilon^{-1} d.$$  

If stress $T$ is applied uniformly to the piezoid (in a direction perpendicular to the plane of the polymer (i.e., in the negative-3 direction), a voltage $V$ develops across its thickness $\ell$ and charge $Q$ appears at the surface of area $A$. It is assumed that in a polymer the polarization is induced predominantly parallel or antiparallel to the poling field direction. After poling when a stress in any direction is applied to the poled material, the voltage along directions other than the poling direction are negligible. This implies that the dielectric tensor elements $\varepsilon_{3i}$, $\varepsilon_{i3}$, $\beta_{3i}$, $\beta_{i3}$ are zero when $i \neq 3$, and that the $g_{nm}$ and $d_{nm}$ tensors have non-zero elements only in the row $n = 3$.

Note that

$$C_p = \varepsilon_p A / \ell_p \quad C_S = \varepsilon_S A / \ell_S, \quad C^{-1} = C_p^{-1} + C_S^{-1}, \quad \text{and} \quad Q = CV.$$  

(10)
Integration of (7) under the conditions $D = 0$ inside the piezoid (the open circuit condition), $g \neq 0$ only across the poled portion, and the applied force $F = AT$, yields:

$$
V = -\int_0^L E dx_3 = \frac{\varepsilon_p}{A} g F = \frac{d_{33}}{C_p} F \quad \text{and} \quad Q = \frac{C}{C_p} d_{33} F. \quad (11)
$$

The quantity $d_{33}$ is taken as the longitudinal piezoelectric constant of a poled polymer.

For short-circuit conditions, $E$ is zero inside the piezoid, and it is convenient to use equation (8). As a stress is imposed, a short-circuit current equal to the displacement current is generated in the piezoid. This current is:

$$
i = A \frac{dD}{dt} = d_{33} \frac{dF}{dt} = d_{33} \dot{F}. \quad (12)
$$

The sign of the force for compression is opposite to that for tension, and the sign of the resulting charge is also opposite for compression to that for tension.

During the application of a time-dependent force to the polymer, the active portion of the piezoid (see figures 5 and 6) acts as a generator of current $i_G$. As a convention,

$$
i_G(t) = d_{33} \dot{F}(t). \quad (13)
$$

The current, $i = i(t)$, flowing in the external circuit is predominantly determined by the electrical load. Under dynamic loading $[C_L \neq 0, R_L \neq \infty]$, some of the generated power is lost internally to $R_p$ and $R_S$.

Application of Kirchoff's laws to the circuits in figures 5 and 6 yields:

$$
i_L R_L + i_p R_p + i_S R_S = 0, \quad (14)
$$

$$
i = i_G + i_p + \dot{Q}_p = i_S + Q_S = i_L + \dot{Q}_L, \quad \text{and} \quad (15)
$$

$$
0 = i_L R_L - Q_L C_L^{-1} = i_S R_S - Q_S C_S^{-1} = i_p R_p - Q_p C_p^{-1}. \quad (16)
$$

Substituting in (14) and (15) for $i_p$, $i_S$, and $i_L$ in terms of $Q_p$, $Q_S$, and $Q_L$ as given in (16 yields:

$$
Q_L C_L^{-1} + Q_p C_p^{-1} + Q_S C_S^{-1} = 0, \quad (17)
$$

$$
\dot{Q}_S + (R_S C_S)^{-1} Q_S = \dot{Q}_L + (R_L C_L)^{-1} Q_L, \quad \text{and} \quad (18)
$$

$$
\dot{Q}_p + (R_p C_p)^{-1} Q_p = \dot{Q}_L + (R_L C_L)^{-1} Q_L - i_G. \quad (19)
$$
The time derivative of (17) added to the product of (17) and the quantity \((R_pC_p)^{-1}\) give:

\[
0 = C_L^{-1}[\dot{Q}_L + (R_pC_p)^{-1}Q] + C_p^{-1}[\dot{Q}_p + (R_pC_p)^{-1}Q_S].
\]

(20)

In solving (20), (18), and (19) it is convenient to assume that

\[
R_pC_p = R_SC_S.\]

(21)

This relation holds if the active and inactive portions have the same conductivity and dielectric constant, since \(C\) and \(R\) are inversely and directly proportional to thickness, respectively. Substituting first (19) into (20) and then (18) with \((R_SC_S)^{-1}\) replaced by \((R_pC_p)^{-1}\) into (20) gives the result:

\[
\dot{Q}_L + \nu Q_L = \zeta \frac{C_L}{C_p} i_G,
\]

where

\[
\nu = \zeta\left(\frac{1}{R_LC} + \frac{1}{R_pC_p}\right), \quad \zeta = (1 + \frac{C_L}{C})^{-1}, \text{ and } C^{-1} = C_p^{-1} + C_S^{-1}.
\]

(22)

The solution of (22) is given by

\[
Q_L(t) = \frac{C_L}{C_p} \int_0^t dt' \exp(\nu t' - \nu t) i_G(t').
\]

(24)

From (16) and (13),

\[
i_L(t) = (R_LC_L)^{-1}Q_L(t) = (R_pC_p)^{-1} \varepsilon_{33} \int_0^t dt' \exp(\nu t' - \nu t) F(t').
\]

(25)

The voltage across the load is

\[
V_L(t) = R_i_L(t),
\]

(26)

while the energy delivered thereto is given by

\[
E(t) = \int_0^t dt' R_L i_L^2(t').
\]

(27)

For stacks consisting of \(n\) piezoid sheets electrically connected in series or parallel \((k\) indicates \(k\text{th sheet})\):

\[
0 = i_R L - Q_L C_L^{-1} = i_{S_k} R_{S_k}^{-1} - Q_S C_S^{-1} = i_{P_k} R_{P_k} - Q_p C_p^{-1}.
\]

(28)

For the parallel arrangement,

\[
i_R L + i_P R_p + i_{S_k} R_{S_k} = 0,
\]

(29)

*See appendix E.
\( i = i_L + \dot{Q}_L = \sum_{k=1}^{n} i_k \) \hspace{1cm} (30)

Where

\( i_k = i_{G_k} + i_{P_k} + \dot{Q}_{P_k} = \dot{i}_{S_k} + \dot{Q}_{S_k} \).

For the series arrangement,

\[ i_L R_L + \sum_{k=1}^{n} (i_{P_k} R_{P_k} + i_{S_k} R_{S_k}) = 0, \quad \text{and} \]

\[ i = i_L + \dot{Q}_L = i_{G_k} + i_{P_k} + \dot{Q}_{P_k} = \dot{i}_{S_k} = \dot{Q}_{S_k}. \quad (32) \]

If the sheets have identical electrical characteristics, the subscript \( k \) may be dropped and the summations may be replaced by a multiplicative factor, \( n \).

Then for the parallel case,

\[ (1 + \frac{C_L}{nC}) \dot{Q}_L + \left( \frac{1}{nR_L C} + \frac{1}{R_P C_P} \right) Q_L = \frac{C_L}{C_P} i_G. \quad (33) \]

For the series case,

\[ (1 + \frac{nC_L}{C}) \dot{Q}_L + \left( \frac{n}{nR_L C} + \frac{1}{R_P C_P} \right) Q_L = \frac{C_L}{C_P} n i_G. \quad (34) \]

Thus in (23) through (27) all symbols remain the same except that for the parallel case

\[ C \rightarrow nC, \quad (35) \]

while for the series case

\[ C \rightarrow n^{-1}C \text{ and } i_G \rightarrow n i_G. \quad (36) \]

Note that the polymer resistance term occurs only in the combination \( R_P C_P \) and that the value of this product does not change whether the arrangement is series or parallel.
APPENDIX B


1. Triangular Force Pulse

The triangular force pulse is characterized by:

\[
F(t) = \begin{cases} 
(t/\tau)F_0, & 0 \leq t \leq \tau \\
(2 - t/\tau)F_0, & \tau \leq t \leq 2\tau 
\end{cases}
\]  

Then

\[
V_L(t) = \zeta V_0 (\nu_t)^{-1} \begin{cases} 
(1 - e^{-\nu t}), & 0 \leq t \leq \tau \\
(2e^{\nu t} - 1 - e^{-\nu t})e^{-\nu t}, & \tau \leq t \leq 2\tau \\
-(e^{\nu t} - 1)^2 e^{-\nu t}, & 2\tau \leq t
\end{cases}
\]  

Also

\[
E(\infty) = E_o \delta(\nu t), \quad E_o = \zeta CV_o^2 (1 + \frac{R_c}{R_p C_p})^{-1},
\]  

where

\[
\delta(Y) = \frac{2}{Y} - \frac{1}{Y^2} (3 - e^{-Y})(1 - e^{-Y}), \quad Y = \nu t, \quad \text{and} \quad V_o = \frac{d_{33} F_0}{C_p}.
\]  

Since Rp is greater than \(10^{12} \Omega\), \(R_L \ll R_p\); consequently, \(\nu^{-1} \approx R_L(C + C_L)\), and

\[
E(\infty) = (C + C_L)^{-1}Q_o^2 \delta(Y) \text{ where}
\]

\[
Y = \frac{\tau}{R_L(C + C_L)} \quad \text{and} \quad Q_o = \frac{Cd_{33} F_0}{C_p} = CV_o.
\]  

The voltage has its maximum at time \(t = \tau\), where \(V_L(\tau) = Y^{-1}(1 - e^{-Y})\zeta V_o\); this expression is a monotonic decreasing function of \(Y\). Then,

\[
V_L(\tau) = \begin{cases} 
\frac{Q_o}{C + C_L} = \zeta V_o, & Y << 1 \\
\frac{R_L Q_o}{\tau} = \frac{R_i C}{V_o}, & Y >> 1
\end{cases}
\]
The minimum value of the voltage occurs at time $t = 2\tau$ where $V_L(2\tau) = -(1 - e^{-Y})V_L(\tau)$. This expression has its least values for $Y$ small and large,

$$
V_L(2\tau) = \begin{cases} 
-YV_L(\tau), & Y \ll 1 \\
-V_L(\tau), & Y \gg 1 
\end{cases}
$$

(43)

and its largest value at $Y = 1.2565$, where

$$
V_L(2\tau) = \frac{-0.407 Q_o}{C + C_L} = -0.407\xi V_o = -0.715 V_L(\tau).
$$

The voltage has value zero for $t_o$ given by

$$
t_o = \nu^{-1} \ln(2e^Y - 1), \quad \tau \leq t_o \leq 2\tau;
t_o > \tau, \quad Y \gg 1; \quad t_o \leq 2\tau, \quad Y \ll 1.
$$

(44)

The energy has a maximum when the load resistance is chosen to be an appropriate value for a given rise time and polymer capacitance. That is, $\delta(Y)$ reaches its maximum 0.381 at $Y = 1.893$. Then

$$
E^{\text{MAX}}(\infty) = \frac{0.381 Q_o^2}{C + C_L} = 0.381\xi CV_o^2.
$$

(45)

Note that $\delta(Y = 0.25) = 0.139; \delta(Y \leq 0.25) = \frac{2}{3} Y - \frac{1}{2} Y^2; \delta(Y = 4) = 0.317; \text{ and } \delta(Y \geq 4) = \frac{2}{Y} - \frac{3}{Y^2}$. When $Y \gg 1$ (very small $R_L$),

$$
E(\infty) = \frac{2 R_L}{\tau} Q_o^2.
$$

(46)

Calculation of the energy delivered to the load during time $t = \tau$ and $t = 2\tau$ yields

$$
E(\tau) = \frac{1}{2} E(\infty) \text{ and } E(2\tau) = E_0 \delta_2(\nu\tau),
$$

(47)

where

$$
\delta_2(Y) = \delta(Y) - \frac{1}{2Y^2} (1 - e^{-Y})^4.
$$

For the values $\tau = 10^{-5}s$, $R_L = 4\Omega$, and $C = 0.3\, \mu F$, $Y \approx 8$ and the approximation of (46) is valid. With the expression of (41) or (70) for the effective charge $Q_0$ developed on the capacitor, (46) and (47) yield $E(\tau) \approx R_L Q_o^2 / \tau$. 

Under optimum conditions, as given in (45), $E^\text{MAX}$ increases linearly with the number of sheets in a stack, $n$, whether the sheets are connected in series or parallel. For $Y \gg 1$, the energy is independent of $n$ for series connection; for parallel connection, the energy varies as $n^2$. However for $Y \ll 1$ (i.e., very large $R_L$),

$$E(\infty) = \frac{2\pi}{3R_L} \frac{Q_0^2}{(C + C_L)^2} = \frac{2\pi}{3R_L} (\zeta V_0)^2,$$

and the remarks applying to series and parallel stacks are reversed.

2. Half-Sine Force Pulse

For the half-sine pulse given by

$$F(t) = F_0 \sin \frac{\pi t}{2\tau}, \quad 0 \leq t \leq 2\tau,$$

$$V_L(t) = \zeta V_0 \cos \phi \begin{cases} \sin \left(\frac{\pi t}{2\tau} + \phi\right) - e^{-\nu t} \sin \phi, & 0 \leq t \leq 2\tau \\ -(1 + e^{2\nu \tau}) e^{-\nu t} \sin \phi, & 2\tau < t \end{cases}$$

where

$$\tan \phi = \frac{2}{\pi} \nu \tau.$$ 

For continuous sinusoidal force the first equation of (50) applies with $0 \leq t < \infty$.

Note that if $\nu \tau \ll 1$, the voltage reaches its maximum at $t = \tau$. Then

$$V_L(\tau) \approx \zeta V_0$$

and the minimum occurs at $t = 2\tau$, where

$$V_L(2\tau) \approx -\frac{4Y}{\pi} V_L(\tau).$$

(As a numerical example, consider the following: a frequency of 100 Hz corresponds to $\tau = 2.5$ ms; a stack of $n = 4$ sheets in series corresponds to a value of $C \approx 0.0025 \mu F$; for $R_L = 10$ MΩ, $\nu \tau \approx \tau / R_L C \approx 0.10$.)

If in addition $C_L \ll C$ and if the stack is made up of sheets connected electrically in parallel, $V_L(\tau)$ is unaffected, while $V_L(2\tau)$ depends on $n^{-1}$; but for the series connection, $V_L(\tau)$ varies as $n$, and $V_L(2\tau)$ depends on $n^2$. 

On the other hand, if $\nu \tau >> 1$, the voltage amplitude is reduced considerably. The voltage minimum occurs at $t = 2\tau$, and the voltage maximum occurs at $t$ on the order of $\nu^{-1} \approx RL/C \ll \tau$, since $\phi \approx \frac{\pi}{2} - \frac{\pi}{2\nu \tau}$:

$$V_L^{MAX} \approx \frac{\pi R_L C}{2\tau} V_0 = -V_L(2\tau).$$

(As a numerical example, consider the following: a frequency of 1 kHz corresponds to $\tau = 0.25$ ms; a stack of $n = 4$ sheets in parallel corresponds to a value of $C \approx 0.40 \mu F$; for $R_L = 100$ $\Omega$, $\nu \tau \approx \tau/RLC = 57.$)

If a stack consists of $n$ sheets electrically in parallel, $V_L^{MAX}$ is increased $n$-fold times; for the series connection, $V_L^{MAX}$ is unaffected.

3. Evaluation of the Output for Haversine Waveform Force Pulse

For the haversine pulse given by

$$F(t) = \frac{1}{2} F_0 \left(1 - \cos \frac{\pi t}{\tau}\right) = F_0 \sin^2 \frac{\pi t}{\tau}, \quad 0 \leq t \leq 2\tau,$$

$$V_L(t) = \frac{1}{2} \zeta V_0 \sin \left\{ \begin{array}{ll}
\sin \left(\frac{\pi t}{\tau} - \theta\right) + e^{-\nu t} \sin \theta, & 0 \leq t \leq 2\tau \\
-(e^{2\nu t} - 1) e^{-\nu t} \sin \theta, & 2\tau \leq t
\end{array} \right.$$  

where $\cot \theta = \frac{\nu \tau}{\pi} = \frac{1}{2} \tan \phi$.  

The energy is calculated as

$$E(\infty) = 2E(\tau) = \frac{1}{4} E_0 g(\nu \tau, \nu \tau/\pi)$$

$$E(2\tau) = \frac{1}{8} E_0 g(2\nu \tau, \nu \tau/\pi)$$

where

$$g(a,b) = (1 + b^2)^{-1} [a + (1 + b^2)^{-1}(1 - e^{-2a})].$$

Here for $\nu \tau >> 1$ the voltage rises slowly to a maximum at $t = \frac{1}{2} \tau$, falls gradually to zero at $t = \tau$, and then reaches a minimum at $t = \frac{3\tau}{2}$. Then since $R_L/Rp << 1$,

$$V_L^{MAX} \approx \frac{\pi R_L C}{2\tau} V_0 = -V_L^{MIN}. $$
This behavior contrasts with that of the two force pulses discussed previously. For example, compare this temporal behavior to that described by (38) and (50) for which the voltage rises very quickly in times on the order of \( v^{-1} \ll \tau \) [see also (43)].

Returning to the haversine pulse, when \( vt >> 1 \) (very small \( R_L \)),

\[
E(2\tau) \simeq E(\infty) \simeq \frac{R_L}{4\tau} (\pi Q_0^2). \tag{61}
\]

On the other hand, for \( vt << 1 \), the voltage behaves as in the previous cases, the maximum occurring at \( t = \tau \) where

\[
V_L^{\text{MAX}} \simeq \xi V_0\tag{62}
\]

and the zero occurs almost simultaneously with the minimum at \( t = 2\tau \) where

\[
V_L^{\text{MIN}} = -\nu \tau \xi V_0 \approx 0.\tag{63}
\]

Also if \( vt << 1 \) (very large \( R_L \)),

\[
E(2\tau) \simeq E(\infty) \simeq \frac{3\tau}{4R_L} \frac{Q_0^2}{(C + C_L)^2} = \frac{3\tau}{4R_L} (\xi V_0)^2. \tag{64}
\]

Now the energy, \( E(\infty) \) is largest when \( vt \) is chosen so as to maximize the function \( g(vt, vt/\pi) \). This occurs at \( vt = 2.106 \) when \( g = 1.922 \). Then (with \( R_L/Rp \approx 0 \))

\[
E^{\text{MAX}}(\infty) = 0.481 \frac{Q_0^2}{C + C_L} = 0.481 \xi CV_0^2. \tag{65}
\]

Because of the close identification between (42) through (48), (52) through (54), and (60) through (65), the remarks in sections 2 and 3 herein about series- and parallel-electrically connected stacks apply to all three pulse waveforms. Note that the inequalities (44) satisfied by \( t_0 \), the instant when the voltage null occurs for the triangular pulse, apply also for the half-sine and haversine pulses. Also, it can be proved in general that if the force pulse is symmetric about \( \tau \) [i.e., if \( F(t) = F(2\tau - t) \), \( 0 \leq t \leq 2\tau \) and \( F(t) = 0 \) for all other \( t \)], then the identity \( E(\infty) = 2E(\tau) \) holds.
APPENDIX C
Piezoid Output Produced by Open-Circuit Charging
Followed by Application of an Electrical Load

The polymer stack may be electrically charged under open-circuit conditions by applying pressure with an hydraulic press or by impacting the stack with a moving mass. The developed charge and energy may then be dumped into a load by means of a switch. This procedure results in an optimal charge and energy transfer from the polymer piezoid to the load. The process of charging under open-switch conditions followed by switching in a load is equivalent to turning on a force infinitely rapidly under closed-circuit conditions. This process may be described by the relation

\[ F(t) = \lim_{\mu \to \infty} (1 - e^{-\mu t})F_0, \quad t \geq 0. \]  

(66)

If at \( t = 0^+ \), i.e., at a time instantaneously greater than \( t = 0 \), the switch across the load is closed (after application of pressure or after impact), then the voltage level across the load is

\[ V_L(t) = \zeta V_0 e^{-\nu t}, \]  

(67)

the charge which flows off the polymer is given from (15), (22), and (25) of appendix B by

\[ Q(t) = \int_0^t dt' i(t') = Q_0(1 + \frac{R_L C}{R_P C_P})^{-1}[1 - \zeta(1 - \frac{R_L C_L}{R_P C_P})e^{-\nu t}], \]  

(68)

and the energy delivered to the load is

\[ E(t) = \frac{1}{2} (1 - e^{-2\nu t})E_0 \]  

(69)

For times long compared to \( \nu^{-1} \approx R_L C \), and using the usual approximations \( C_L \ll C, R_L \ll R_P \),

\[ Q \approx Q_0, \quad E \approx \frac{1}{2} CV_0^2 = \frac{Q_0^2}{2C}. \]  

(70)

This \( Q \) is the charge built up on the polymer stack. \( Q \) is independent of \( n \) for series electrical connection and proportional to \( n \) for parallel connection. The quantity \( E \) is the energy stored in the stack and is proportional to \( n \) for both series and parallel connections. For times of the order of tens of microseconds and capacitances of the order of tenths of microfarads, \( R_L \) should be somewhat less than 100 \( \Omega \) to have full energy transfer to the load.
However, if $R_L$ is large (certainly if $R_L > 10$ kΩ, and $t < 1.0$ ms),

$$Q(t) = \zeta C_L V_o + \frac{\zeta^2 t}{R_L} V_o$$ and $E(t) = \frac{t}{R_L} (\zeta V_o)^2$. \hspace{1cm} (71)

Note that if $C_L/C < \nu t < 1$, the second term in the expression for $Q(t)$ in (71) dominates; however if $C_L/C > \nu t$, the first term is larger than the second. If $C_L/C << 1$, $Q$ and $E$ are independent of $n$ for the parallel electrical connection, while for the series electrical connection, $Q$ is proportional to $n$, and $E$ is proportional to $n^2$. 
APPENDIX D

Expressions for Piezoid Output Into Load

<table>
<thead>
<tr>
<th>Condition 1</th>
<th>Condition 2</th>
<th>Condition 3</th>
<th>Condition 4</th>
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<tr>
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<td>Energy</td>
<td>Voltage</td>
<td>Energy</td>
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<td>$E(t)$</td>
<td>$V_{L_{\text{MAX}}}$</td>
<td>$E(t)$</td>
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<tr>
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<td>$E(t)$</td>
<td>$V_{L_{\text{MIN}}}$</td>
<td>$E(t)$</td>
</tr>
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\[
\begin{align*}
\nu t << 1 & : \quad cV_0 \quad - \nu tcV_0 \quad \frac{\pi}{3R_L}(cV_0)^2 \quad cV_0 \quad - \nu tcV_0 \quad \frac{\pi}{2R_L}(cV_0)^2 \quad cV_0 \quad - \nu tcV_0 \quad \frac{\pi}{8R_L}(cV_0)^2 \\
\nu t \gg 1 & : \quad \frac{\nu t R_L Q_0}{\pi} \quad \frac{\nu t R_L Q_0}{\pi} \quad (2\nu t)^{-1} \frac{\nu t R_L Q_0}{\pi} \quad \frac{\nu t R_L Q_0}{\pi} \quad (2\nu t)^{-1} \frac{\nu t R_L Q_0}{\pi} \quad \frac{\nu t R_L Q_0}{\pi} \quad \frac{\nu t R_L Q_0}{\pi} \quad \frac{\nu t R_L Q_0}{\pi} \\
(\nu t)^* & : \quad 0.449 cV_0 \quad -0.381 cV_0 \quad 0.190 cV_0^2 \quad 0.483 cV_0 \quad -0.513 cV_0 \quad 0.263 cV_0^2 \quad 0.480 cV_0 \quad -0.408 cV_0 \quad 0.240 cV_0^2
\end{align*}
\]

Conditions:

1. When stack is subjected to triangular force pulse; $(\nu t)^* = 1.89$ yields maximum.
2. When stack is subjected to half-sine force pulse; $(\nu t)^* = 1.73$ yields maximum.
3. When stack is subjected to haversine pulse; $(\nu t)^* = 2.11$ yields maximum.
4. After open-circuit storage of charge an electrical load is switched in at $\tau = 0$; when $(\nu t)^* \gg 1$, $C_L = 0$ yields maximum.
APPENDIX E

Output from Piezoid Sample Having Non-Uniform Conductivity and Dielectric Constant

Both theoretical and experimental investigations show that the dielectric constant and conductivity of the poled portion of a piezoelectric material are different from the unpoled bulk portion.

In appendix A, the condition $R_S C_S = R_P C_P$, was assumed for convenience of exposition. However, an equation for $Q_L$ can be found by substituting for $Q + (R_P C_P)^{-1} Q_P$ from (19) into (17). Then the resulting equation may be combined with (18) to determine an expression for $Q_S$ in terms of $Q_L$, $Q_L$, and $i_G$. Finally, this expression and its time derivative (the expression for $Q_S$ in terms of $Q_L$, $Q_L$, and $d i_G/d t$) may be substituted in (18) and there results:

$$
[C^{-1} + C_L^{-1}] \ddot{Q}_L + [(C^{-1} (R_L C_L)^{-1} + (C_L^{-1} + C_S^{-1}) (R_P C_P)^{-1} + 
(C_L^{-1} + C_P^{-1}) (R_S C_S)^{-1}] \dot{Q}_L + [(R_P^{-1} R_L^{-1} + (R_S R_L)^{-1} + (R_P R_S)^{-1}) \times 
(C_P C_S C_L)^{-1} Q_L = C_P^{-1} [d i_G/d t + (R_P C_P)^{-1} i_G].
$$

(72)

This equation is easily solved; however the solution is lengthy, and therefore only the expression for $R_L \ll R_P$ and $R_S$, which holds in all practical situations, is given here.

$$
i_L = (R_L C_L)^{-1} Q_L = \int_0^t dt' i_G(t') \{(R_L C_P)^{-1} \xi \exp[\xi (R_L C_L)^{-1} (t' - t)] + 
+ (C_P C)^{-1} [(R_P C_P)^{-1} - (R_S C_S)^{-1}] \exp[(R_P^{-1} + R_S^{-1}) (C_P + C_S)^{-1} (t' - t)]\},
\quad R_L C_L \ll R_P C_P.
$$

(73)

Only if $\tau \gg R_P C_P \approx 10^4 s \gg R_L C$, does the second term in (73) give a non-negligible contribution. Therefore retention of only the first term in (73), which would result in equation (25), yields a useful formula even when the condition $R_S C_S = R_P C_P$ does not obtain.
6. REFERENCES


<table>
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<th>Batch No.</th>
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TABLE 2A
CAPACITANCE AND PIEZOELECTRIC ACTIVITY OF COMPONENTS
FOR FIRST INTERMEDIATE STACK OF FIFTEEN SHEETS

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*The table is arranged in chart form to show how a fifteen-sheet stack was assembled. This stack was itself one of two intermediate stacks comprising Stack No. 7, as described in the text. For n = 1, the values of two parameters are given: the upper number is the measured sheet capacitance (nF) and the lower, the measured sheet piezoelectric activity (pC/N). For each intermediate stack, the values of six parameters are given as follows:

- measured stack capacitance
- calculated average sheet capacitance = measured stack capacitance / n
- calculated average sheet activity based on calculated average sheet capacitance of component stacks, weighted by the number of sheets in each component
- measured stack piezoelectric activity
- calculated average sheet activity = measured stack activity / n
- calculated average sheet activity based on calculated average sheet activity of component stacks, weighted by the number of sheets in each component.
### TABLE 2B

CAPACITANCE AND PIEZOELECTRIC ACTIVITY OF COMPONENTS
FOR SECOND INTERMEDIATE STACK OF FIFTEEN SHEETS

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- Calculated average sheet activity = measured stack activity / n
- Calculated average sheet capacitance of component stacks, weighted by the number of sheets in each component
- Calculated average sheet activity of component stacks, weighted by the number of sheets in each component.
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<th>0.400</th>
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<td>7.4</td>
<td>7.5</td>
</tr>
<tr>
<td>$V_L^{\text{MAX}}$ [C_L = 15 pF]** (V)</td>
<td>0.15</td>
<td>0.40</td>
<td>1.33</td>
<td>9.6</td>
<td>19.4</td>
<td>22.5</td>
</tr>
<tr>
<td>$V_L^{\text{MIN}}$ (V)</td>
<td>-0.15</td>
<td>-0.40</td>
<td>-1.33</td>
<td>-8.4</td>
<td>-4.0</td>
<td>0.0</td>
</tr>
<tr>
<td>$V_L^{\text{MAX}} / F_0$ (μV)</td>
<td>8.0</td>
<td>21.3</td>
<td>70.7</td>
<td>511</td>
<td>1030</td>
<td>1200</td>
</tr>
<tr>
<td>$t_M$ (μs)***</td>
<td>100</td>
<td>125</td>
<td>125</td>
<td>150</td>
<td>200</td>
<td>225</td>
</tr>
<tr>
<td>$t_0$ (μs)</td>
<td>225</td>
<td>225</td>
<td>250</td>
<td>275</td>
<td>375</td>
<td>450</td>
</tr>
<tr>
<td>$E(\tau) / F_0^2$ (fJ)</td>
<td>1.2</td>
<td>3.4</td>
<td>10.5</td>
<td>61.6</td>
<td>19.9</td>
<td>2.7</td>
</tr>
</tbody>
</table>

* The table is explained in the text.

** This C_L is the capacitance contribution of the transient recorder and is taken as the C_L for all measurements except as noted.

*** $t_M$ = time at which $V_L = V_L^{\text{MAX}}$. 
<table>
<thead>
<tr>
<th>$R_L$ (k$\Omega$)</th>
<th>0.004</th>
<th>0.010</th>
<th>0.040</th>
<th>0.400</th>
<th>4.0</th>
<th>40.0</th>
</tr>
</thead>
<tbody>
<tr>
<td>$Y = \nu t = \tau/R_L C$</td>
<td>225</td>
<td>90</td>
<td>22.5</td>
<td>2</td>
<td>0.225</td>
<td>0.023</td>
</tr>
<tr>
<td>$V_L^M$ ($\mu V$)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Measured*</td>
<td>8.0</td>
<td>21.3</td>
<td>70.7</td>
<td>511</td>
<td>1030</td>
<td>1200</td>
</tr>
<tr>
<td>Calculated**</td>
<td>6.9</td>
<td>17</td>
<td>69</td>
<td>480†</td>
<td>900†</td>
<td>1000</td>
</tr>
<tr>
<td>$\frac{L(t)}{F_0}$ ($\mu J$)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Measured</td>
<td>1.2</td>
<td>3.4</td>
<td>10.5</td>
<td>61.6</td>
<td>19.9</td>
<td>2.7</td>
</tr>
<tr>
<td>Calculated</td>
<td>1.3</td>
<td>3.3</td>
<td>13.4</td>
<td>59†</td>
<td>21</td>
<td>2.1</td>
</tr>
</tbody>
</table>

*Measured values are those of table 3.

**Calculated values are based on the conditions $\tau = 225 \mu s$, $C = 0.25 \mu F$, and $V_0/F_0 = 1 \text{ mV/N}$.

†Value calculated for $L^M$ with $\nu t = 2.1 (R_L = 430 \Omega)$.

‡Value calculated from (56), with $t = \tau$. 
### Table 5

**Deliveries of Piezoid Stacks to Frankford Arsenal in 1974**

<table>
<thead>
<tr>
<th>Delivery Date</th>
<th>Stack Number</th>
<th>Sheet Thickness (μm)</th>
<th>Number of Sheets</th>
<th>Stack Thickness (mm)</th>
<th>Static Capacitance (μF)</th>
<th>Total Activity (pC/N)</th>
<th>Average Sheet Activity (pC/N)</th>
</tr>
</thead>
<tbody>
<tr>
<td>February 14</td>
<td>1</td>
<td>12</td>
<td>10</td>
<td>0.14</td>
<td></td>
<td>30</td>
<td>3</td>
</tr>
<tr>
<td>February 14</td>
<td>2</td>
<td>12</td>
<td>10</td>
<td>0.14</td>
<td></td>
<td>30</td>
<td>3</td>
</tr>
<tr>
<td>February 14</td>
<td>3</td>
<td>12</td>
<td>10</td>
<td>0.14</td>
<td></td>
<td>30</td>
<td>3</td>
</tr>
<tr>
<td>March 18</td>
<td>4</td>
<td>12</td>
<td>10</td>
<td>0.14</td>
<td></td>
<td>30</td>
<td>3</td>
</tr>
<tr>
<td>May 6</td>
<td>5</td>
<td>25</td>
<td>88</td>
<td>2.42</td>
<td>1.00</td>
<td>290</td>
<td>3.3</td>
</tr>
<tr>
<td>June 7</td>
<td>6</td>
<td>25</td>
<td>100</td>
<td>2.69</td>
<td>1.25</td>
<td>300</td>
<td>3</td>
</tr>
<tr>
<td>September 20</td>
<td>7</td>
<td>25</td>
<td>30</td>
<td>0.88</td>
<td>0.36</td>
<td>360</td>
<td>12</td>
</tr>
<tr>
<td>September 27</td>
<td>8</td>
<td>25</td>
<td>24</td>
<td>0.68</td>
<td>0.31</td>
<td>270</td>
<td>11</td>
</tr>
<tr>
<td>November 18</td>
<td>9</td>
<td>25</td>
<td>20</td>
<td>0.60</td>
<td>0.23</td>
<td>215</td>
<td>11</td>
</tr>
<tr>
<td>November 18</td>
<td>5-1Δ</td>
<td>25</td>
<td>42</td>
<td>1.13</td>
<td>0.56</td>
<td>145</td>
<td>3.4</td>
</tr>
<tr>
<td>November 18</td>
<td>5-2Δ</td>
<td>25</td>
<td>46</td>
<td>1.24</td>
<td>0.55</td>
<td>145</td>
<td>3.2</td>
</tr>
<tr>
<td>November 18</td>
<td>6-1Δ</td>
<td>25</td>
<td>56</td>
<td>1.52</td>
<td>0.68</td>
<td>150</td>
<td>2.6</td>
</tr>
<tr>
<td>November 18</td>
<td>6-2Δ</td>
<td>25</td>
<td>44</td>
<td>1.19</td>
<td>0.50</td>
<td>115</td>
<td>2.7</td>
</tr>
<tr>
<td>December 10</td>
<td>10</td>
<td>25</td>
<td>20</td>
<td>0.58</td>
<td>0.22</td>
<td>235</td>
<td>12</td>
</tr>
</tbody>
</table>

ΔStacks Nos. 5 and 6 were returned to NBS for repair because the tin foil leads had broken off during static testing after delivery. Each of the two stacks was split approximately in half and new leads were attached to the resulting four stacks.
Figure 1: Mask and resulting electrodes on PVF₂ film. On the left is the 24-cm-square stainless-steel mask used in the vacuum deposition of aluminum electrodes onto the polymer sheet material. On the right are four electroded and poled active elements. For each piezoid, the tail seen at the bottom forms part of the upper electrode and the tail seen at the top forms part of the lower electrode (hidden by the upper electrode in the photograph).
Figure 2: Stack construction. At the bottom (A) is a completed stack. The other stages shown are (B) a single-sheet piezoid element, (C) a single sheet with one foil electrode lead attached, (D) two sheets bonded together with the high-side lead between the sheets (at left) and two low-side leads attached to the sheet outer surfaces (at right), and (E) two sheets bonded together with an interior high-side lead.
Figure 3: Circuit used in static-testing procedure. Storage oscilloscope vertical-axis inputs are marked 0; P is the piezoid to which force $F_0$ is applied; and $C_L$ is the standard capacitor, as described in the text.
**Figure 4:** Circuit used to connect polymer device to load. "Low" and "high" refer to polarities established at the time of poling; "high" corresponds to positive.

For compression tests, the double-pole, double-throw switch is in the position shown, and the low side becomes charged positively with respect to the high side. When the voltage sensed by the Zener diode reaches the Zener reference voltage, the polymer device discharges through diode CR2 into the load. For tension tests, the switch is thrown to position 2, as the device polarities are reversed.
Figure 5: Polymer element equivalent circuit.

Figure 6: Polymer element connected to electrical load.
### 16. ABSTRACT

Described is the development of polymer piezoelectric devices with sufficient electrical output in response to impact to demonstrate the feasibility of using polymer elements in place of ceramic elements in ordnance fuze systems. To satisfy this requirement, piezoelectric polymer elements have been constructed capable of providing at least 0.3 mJ into a 4-Ω load, in response to a force of 89 kN (10 tonsf). Total activities of up to 360 pC/N have been achieved from elements built up from 30 sheets of 25-μm poly(vinylidene fluoride). Test methods developed to characterize both single-sheet and total-element activities are described, as are poling procedures and details of element construction. Mathematical analyses of polymer piezoelectric output into resistive-capacitive loads are presented for static and dynamic cases. Experimental results and theoretical predictions are shown to be in agreement to within the 10% estimate of measurement accuracy.

### 17. KEY WORDS

Calibration; electrical energy; electrical output; piezoelectric; piezoelectric output; polymer; poly(vinylidene fluoride); stack construction; theory; voltage.

### 19. SECURITY CLASS (THIS REPORT)

UNCLASSIFIED