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THE DIELECTRIC CONSTANT OF POLYETHYLENE UNDER SHOCK COMPRESSION

by

G. E. Hauver

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BALLISTIC RESEARCH LABORATORIES

REPORT NO. 1520

DECEMBER 1970

THE DIELECTRIC CONSTANT OF POLYETHYLENE UNDER SHOCK COMPRESSION

G. E. Hauver

Terminal Ballistics Laboratory

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BALLISTIC RESEARCH LABORATORIES

REPORT NO. 1520

G. E. Hauver
Aberdeen Proving Ground, Md.
December 1970

THE DIELECTRIC CONSTANT OF POLYETHYLENE UNDER SHOCK COMPRESSION

ABSTRACT

The dielectric constant of high-density polyethylene was measured under shock compression in the pressure range from 16 to 246 kilobars. A charged-capacitor technique was used, and the dielectric constant was calculated from the charging current measured during shock compression. From 36 to 246 kilobars, the dielectric constant was found to increase according to the Drude equation, assuming constant polarizability; and in this pressure range it was represented by the relationship $k - 1 = 1.47\ \rho$, where ρ is the density.

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LIST OF SYMBOLS

A	area
C	capacitance
$\underset{\sim}{D}$	electric displacement(*)
$\underset{\sim}{E}$	applied electric field (*)
$\underset{\sim}{E}_L$	local field
$\underset{\sim}{P}$	electric polarization(*)
Q	electric charge
Q_m	total change in charge measured during shock compression; (+) and (-) refer to the polarity of the applied voltage.
Q_p	charge associated with the shock-induced polarization signal; (0) refers specifically to Q_p measured with no applied voltage.
R	resistance; subscript "T" refers to the terminating resistance of a coaxial cable
U	shock velocity
V	applied voltage
V_s	signal voltage after shock arrival at the guarded electrode
X_o	initial thickness
k	relative dielectric constant of the shock compressed dielectric
k_o	relative dielectric constant at zero pressure
u	particle velocity
ϵ	permittivity
ϵ_o	permittivity of free space (8.854×10^{-12} F/m)
ρ	density of the shock-compressed dielectric
ρ_o	initial density of the dielectric

(*) subscript "o" refers to unshocked dielectric; subscript "s" refers to shocked dielectric.

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I. INTRODUCTION

The dielectric constant of a number of materials has been measured during static compression in the pressure range below thirty kilobars. References to many of the static measurements are included in a review article by Whalley[(1)*] Recently, changes in the dielectric constant have been used to detect and study pressure-induced phase transitions encountered in static measurements.[(2)] [(3)]

Relatively few measurements, however, have been performed to determine the behavior of the dielectric constant during shock compression. Graham[(4)] measured the permittivity change of sapphire under shock compression and found a linear decrease with stress to 100 kbar, although data above 60 kbar were influenced slightly by shock-induced electrical conductivity. Hawke, Mitchell, and Keeler[(5)], using a microwave technique, measured the dielectric constant of carbon tetrachloride at shock pressures of 130 and 150 kilobars. The measured values for carbon tetrachloride exceeded values predicted by the Clausius-Mossotti equation, and it was noted that the discrepancy might indicate an increase in the molar polarizability. The microwave technique was also used to measure the dielectric constant of polycrystalline aluminum oxide at a shock pressure of 375 kbar[(6)], where data were interpreted to show an increase in the dielectric constant, a decrease in molar polarizability, and negligible shock-induced electrical conductivity. Shock-induced polarization experiments on polymethyl methacrylate and polystyrene[(7)] provided values of the dielectric constants under conditions of shock compression, but the data were characterized by a large spread which may indicate problems in distinguishing relaxation effects from changes in the dielectric constant. In somewhat similar experiments[(8)], the profiles of shock-depolarization signals were analyzed to obtain values for the dielectric constant of shock-compressed

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**References are found on page 33.*

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barium titanate. Although the influence of shock compression on the dielectric constant has received little attention, there is evidence of increasing interest in this area of study.

Experiments have been performed to measure the dielectric constant of polyethylene during shock compression in the pressure range from 16 to 246 kilobars. Polyethylene was selected for several reasons. First, polyethylene has been the subject of a shock-induced polarization study, and dielectric-constant data were needed to analyze polarization signals. Second, although polyethylene produces a shock-induced polarization signal, the signal is small and it was anticipated that it would not be a major source of error in the proposed technique. Third, the dielectric coefficient of polyethylene under normal laboratory conditions is constant (approximately 2.3) from DC to optical frequencies[9]. Fourth, Keeler and Mitchell[10] have reported that polyethylene remains a reasonably good insulator to shock pressures well above 500 kilobars.

This report briefly reviews the theory that may be used to consider the effect of shock compression on dielectric behavior, describes the experimental and analytical procedures used for the measurements on polyethylene, presents the data, and includes a brief discussion of the results.

II. REVIEW OF THEORETICAL GUIDES

When a dielectric material is placed in an electric field, it develops a surface charge. The induced polarization results from the displacement of positive and negative charges in the dielectric. For a linear, isotropic dielectric, the polarization $\underset{\sim}{P}$ is proportional and parallel to the applied field $\underset{\sim}{E}$, and the electric displacement $\underset{\sim}{D}$ is defined by the relation (MKS units),

$$\underset{\sim}{D} = \varepsilon_0 \underset{\sim}{E} + \underset{\sim}{P} = \varepsilon \underset{\sim}{E},$$

where ε is the permittivity of the dielectric material and ε_o is the permittivity of free space. Then,

$$\underset{\sim}{P} = (\varepsilon - \varepsilon_o)\, \underset{\sim}{E} = (k - 1)\, \varepsilon_o\, \underset{\sim}{E},$$

where k is the relative dielectric constant. Alternately, the polarization $\underset{\sim}{P}$ is the dipole moment per unit volume, and the dipole moment per unit volume may be thought of as the additive action of N elementary dipole moments $\underset{\sim}{\bar{\mu}}$,

$$\underset{\sim}{P} = N\, \underset{\sim}{\bar{\mu}}.$$

The average dipole moment $\underset{\sim}{\bar{\mu}}$ is assumed to be proportional to the local field $\underset{\sim}{E}_L$,

$$\underset{\sim}{\bar{\mu}} = \alpha\, \underset{\sim}{E}_L,$$

where the constant of proportionality α is called the polarizability. The local field at a site in the dielectric is generally written as,

$$\underset{\sim}{E}_L = \underset{\sim}{E} + \underset{\sim}{E}_I,$$

where $\underset{\sim}{E}$ is the applied field and $\underset{\sim}{E}_I$ is the field due to polarization of the surroundings. As a model for calculations, the site is placed at the center of a cavity in the dielectric, and $\underset{\sim}{E}_I$ is evaluated as the field contribution from the charge lining the cavity walls, which is

$$\underset{\sim}{E}_I = \underset{\sim}{P}/3\, \varepsilon_o.$$

The local field becomes,

$$\underset{\sim}{E}_L = \underset{\sim}{E} + \underset{\sim}{P}/3\,\epsilon_o = \underset{\sim}{E}\,(k + 2)/3.$$

It is easily shown that,

$$\frac{N\,\alpha}{3\epsilon_o} = \frac{k - 1}{k + 2}$$

However,

$$N = N_o\,\rho/M,$$

where N_o is Avogadro's number, M is the molecular weight, and ρ is the density. Substituting for N and rearranging,

$$\frac{k - 1}{k + 2}\left(\frac{1}{\rho}\right) = \frac{N_o\alpha}{3\epsilon_o M},$$

which is the familiar Clausius-Mossotti equation. If the polarizability is constant,

$$\frac{k - 1}{k + 2}\left(\frac{1}{\rho}\right) = C_1, \qquad (1)$$

where C_1 is a constant.

The local field $\underset{\sim}{E}_L$ is actually a matter of some uncertainty. This has been noted by Royce[11], and some aspects of the uncertainty are discussed by Mott and Gurney[12]. In the study of some solids[13][14], it has been found necessary to assume the local field equal to the applied field. Assuming $\underset{\sim}{E}_L = \underset{\sim}{E}$ leads to the Drude

equation,

$$\frac{k - 1}{\rho} = \frac{N_o \alpha}{\varepsilon_o M};$$

and if the polarizability is assumed to be constant,

$$\frac{k - 1}{\rho} = C_2 \tag{2}$$

where C_2 is a constant.

Despite the uncertainties, the Clausius-Mossotti and Drude equations serve as guides for considering the effect of shock compression on dielectric behavior and interpreting experimental results. The Clausius-Mossotti equation was considered to be the safer guide by which to estimate signal sizes for the dielectric constant measurements because it predicts the more rapid change in dielectric constant with increasing density. For these estimates, the polarizability was assumed to be independent of shock compression, and the constant C_1 was evaluated at zero pressure where $k = k_o$ and $\rho = \rho_o$.

III. DIELECTRIC CONSTANT MEASUREMENTS

A. Experimental Arrangement

Polyethylene test assemblies were prepared as shown in Figure 1. Electrical leads of magnesium were embedded in a polyethylene substrate, 1.27 cm thick and 3.81 cm in diameter. The polyethylene had a density of 0.960 ± 0.002 g/cc, and is commonly referred to as high-density polyethylene. An aluminum coating, 3.50 cm in diameter, was deposited on the polyethylene substrate by vacuum evaporation, and was lightly scribed to divide the aluminized

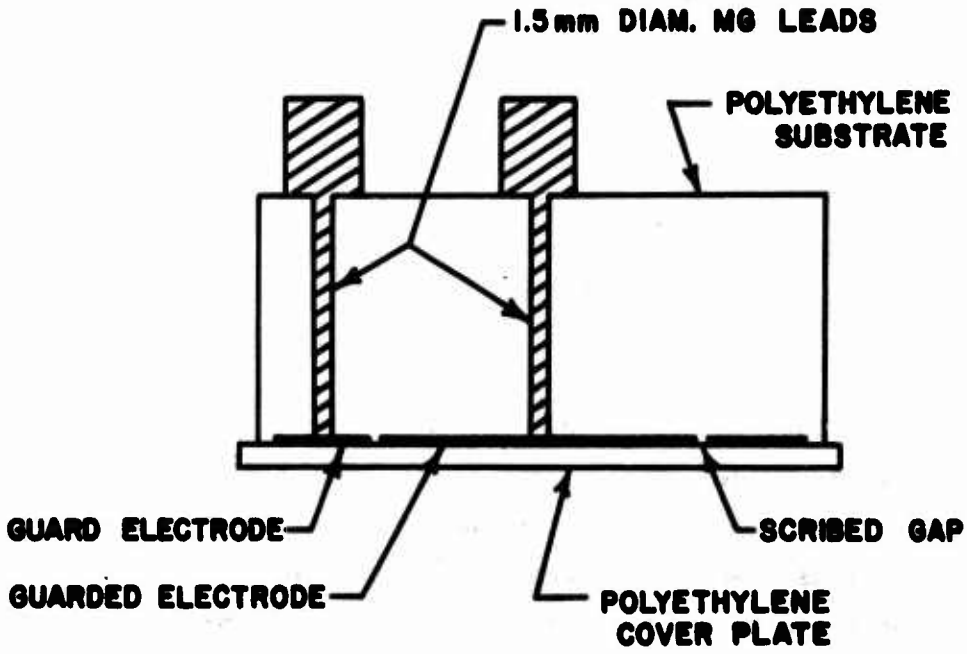

Figure 1. Polyethylene Assembly

FIG 1

surface into guard and guarded electrodes with equal areas. The natural oxide film between the aluminum coating and the magnesium leads was broken down mechanically to obtain satisfactory electrical continuity. A polyethylene cover plate, 0.1500 ± 0.0002 cm thick and 4.13 cm in diameter, was bonded to the aluminum electrode surfaces with room temperature setting epoxy. The thickness of the bond did not exceed 0.0002 cm.

The experimental arrangement for the dielectric constant measurements is shown in Figure 2. Shock pressures up to 173 kbar were produced by detonating an explosive charge in contact with the metal plate (or mismatched stack of plates) which held the polyethylene assembly. [In one test, performed at 246 kbar, the pressure was produced by the impact of an explosively accelerated plate.] The plate surface beneath the polyethylene assembly was polished, and the assembly was held against the polished surface by light spring pressure against the ends of the electrical leads. This arrangement formed a three-electrode parallel-plate capacitor, with the metal plate as the electrode at ground potential. In a comparison test, a metal film was evaporated onto the polyethylene surface which contacted the polished plate. Within the experimental uncertainty, this variation of the contact surface at the ground electrode had no influence on the measurement.

A Plexiglas II UVA specimen, 0.15 cm thick and 0.72 cm in diameter, was included on each test, as shown in Figure 2. The shock-induced polarization signal from the Plexiglas provided a measure of transit time from which the average shock velocity and pressure were established. This measurement technique is described in reference [7], although the Plexiglas II UVA Hugoniot is presently represented by the relationship $U = 2.695 \times 10^3 + 1.538\ u$, where U is the shock velocity, u is the particle velocity, and units are m/sec. This measurement locates the release adiabat of the metal plate, which

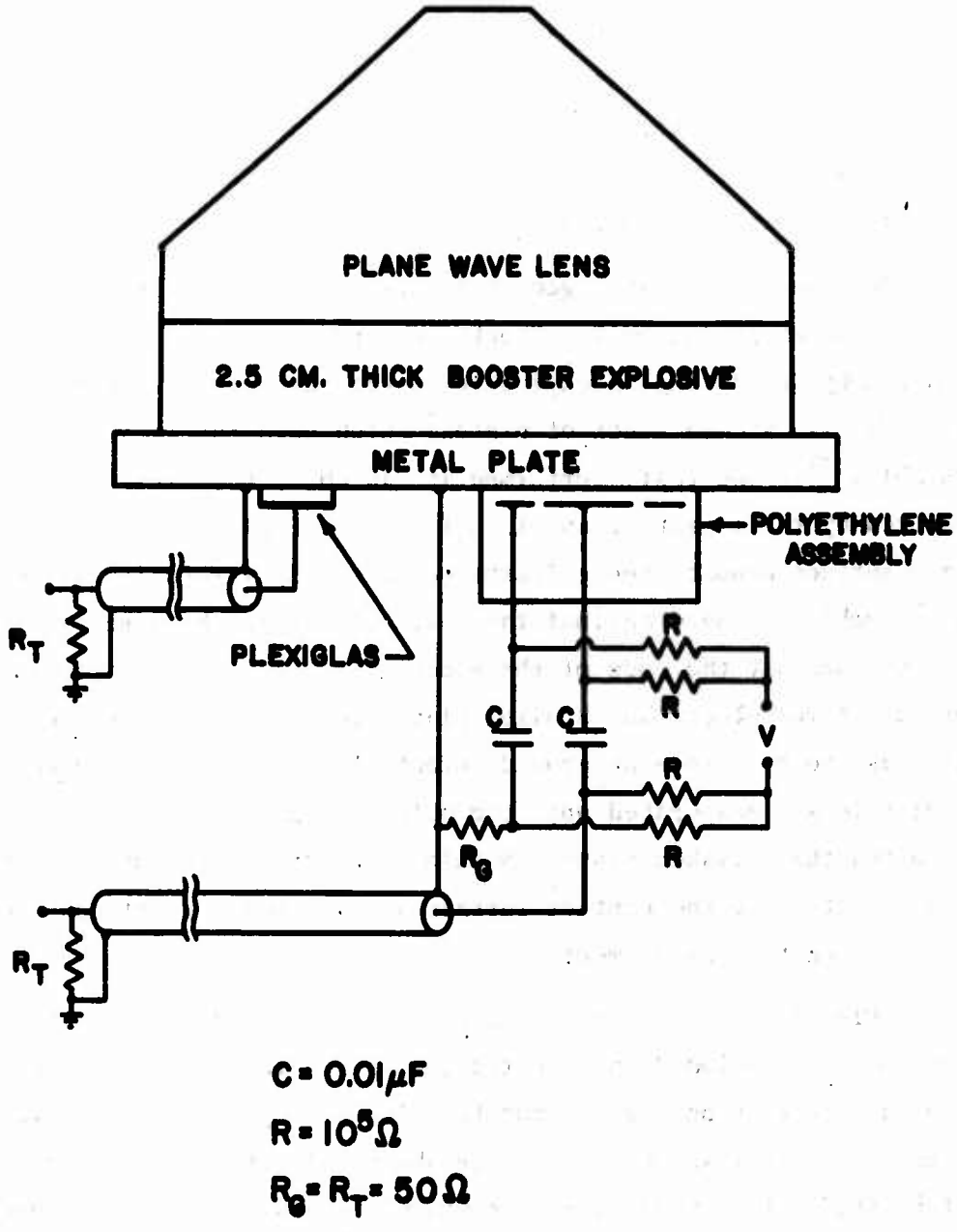

Figure 2. Experimental arrangement used to perform dielectric constant measurements on shock-compressed polyethylene.

intersects polyethylene Hugoniot and establishes the test state. However, the signal from the polyethylene assembly was found to provide an equally reliable measurement of shock transit time from which the Hugoniot state in polyethylene could be established.

B. Test and Analytical Procedure

The circuit capacitance, consisting of the polyethylene capacitor, capacitor C, and the stray capacitance, was charged to a voltage V of 500 volts. Capacitor C was approximately three orders of magnitude larger than the remaining circuit capacitance and held the applied voltage effectively constant during a test. For the present, the stray capacitance will be assumed to remain constant, although it changed a small but measurable amount in some tests.

When a shock wave enters the polyethylene capacitor from the metal plate, it compresses the dielectric and changes the dielectric constant. The usual relation between the electric displacement $\underset{\sim}{D}$, polarization $\underset{\sim}{P}$ and electric field $\underset{\sim}{E}$ are assumed to hold. Ahead of the shock front in the unshocked polyethylene,

$$\underset{\sim}{D}_o = \varepsilon_o \underset{\sim}{E}_o + \underset{\sim}{P}_o = k_o \varepsilon_o \underset{\sim}{E}_o .$$

Within the shock compressed dielectric,

$$\underset{\sim}{D}_s = \varepsilon_o \underset{\sim}{E}_s + \underset{\sim}{P}_s = k \varepsilon_o \underset{\sim}{E}_s$$

Assuming a parallel-plate configuration and no shock-induced electrical conductivity.

$$\underset{\sim}{D}_s = \underset{\sim}{D}_o = Q/A,$$

where Q is the charge on the electrodes and A is the electrode area. Then, ahead of the shock front,

$$\underset{\sim}{E}_o = Q/k_o \varepsilon_o A,$$

and within the shock-compressed polyethylene,

$$\underset{\sim}{E}_{s} = Q/k\ \varepsilon_o\ A.$$

In the tests with polyethylene, the RC time constant was in the order of one nanosecond. In this case, the load resistance may be considered a short circuit with negligible error. Then, since $V = \int \underset{\sim}{E}\, dX$,

$$V = \int_{Ut}^{X_o} \frac{Q}{k_o\ \varepsilon_o\ A}\, dX + \int_{ut}^{Ut} \frac{Q}{k\ \varepsilon_o\ A}\, dX,$$

where U is the shock velocity, u is the particle velocity, X_o is the polyethylene thickness, and t is the time measured from the entry of the shock front into the dielectric. Integrating,

$$V = \frac{Q\ (X_o - Ut)}{k_o\ \varepsilon_o\ A} + \frac{Q\ (U - u)\ t}{k\ \varepsilon_o\ A}.$$

Solving for the charge,

$$Q = \frac{V\ \varepsilon_o\ k_o\ A}{X_o + \left[(k_o/k)\ (U - u) - U\right]\ t} \qquad (3)$$

Differentiating with respect to time, the profile of current signal is given by the expression,

$$I = \frac{V\ \varepsilon_o\ k_o\ A\ \left[U - (k_o/k)\ (U - u)\right]}{\left\{X_o - \left[U - (k_o/k)\ (U - u)\right]\ t\right\}^2}. \qquad (4)$$

This expression for the current was used to calculate anticipated signal profiles, using a value for k estimated by the Clausius-Mossotti equation [Equation 2] . However, the expression for the charge [Equation 3] was used to calculate the measured dielectric constant. When t = 0 in Equation (3), the charge Q_1 on the polyethylene capacitor is,

$$Q_1 = V\, k_o\, \varepsilon_o\, A/X_o.$$

When the shock front arrives at the guarded electrode, $t = X_o/U$, and from Equation (3) the charge on the capacitor should have become,

$$Q_2 = V\, k\, \varepsilon_o\, AU/X_o\, (U - u).$$

However, this neglects any shock-induced polarization and the charge Q_p associated with it. In general, shock-induced polarization does occur and must be considered, so that,

$$Q_2 = \frac{V\, k\, \varepsilon_o\, A\, U}{X_o\, (U - u)} + Q_p .$$

In the time interval from t = 0 to $t = X_o/U$, the change in charge on the electrodes, or measured charge Q_m, is

$$Q_m = Q_2 - Q_1 = \frac{V\, \varepsilon_o\, A}{X_o} \left[\frac{k\, U}{(U - u)} - k_o \right] + Q_p .$$

Solving for the dielectric constant of the shock compressed material,

$$k = \frac{U - u}{U} \left[\frac{X_o\, (Q_m - Q_p)}{V\, \varepsilon_o\, A} + k_o \right] . \qquad (5)$$

In each test, the current was recorded with an oscilloscope[15] as the voltage drop across resistor R_T (see Figure 2). A test record is shown in Figure 3. This record shows a positive voltage/time signal, voltage reference lines, and a 50 MHz time reference. Each voltage/time record was measured, converted to current/time, and integrated to obtain Q_m. At each pressure, the polarization charge Q_p was obtained by conducting a test with no applied voltage. However, at the lowest test pressures (16, 20, and 36 kbar) where Q_p was the largest fraction of Q_m, it was also evaluated in another way as a check. The polarity of the applied voltage was reversed in successive tests to obtain signals of opposite polarity. The measured charges Q_m (-) and Q_m (+) were added algebraically to obtain $2\ Q_p$.

Equation (5) also requires values for the shock and particle velocities. The time duration of the signal from the polyethylene capacitor provided a reliable measurement of the shock transit time from which the shock velocity was calculated (as noted before, the Plexiglas measurement could also be used to establish the Hugoniot state in polyethylene, and served as a check). The particle velocity was obtained from the Hugoniot for the high-density polyethylene which was measured and reported[16], and which is represented by $U = 2.925 \times 10^3 + 1.570\ u$, with $\sigma_U = 0.062 \times 10^3$, where σ_U is the standard deviation of U.

Equation (5) also contains k_o, the relative dielectric constant of uncompressed polyethylene. Several specimens of the high-density polyethylene were prepared with faces plane and parallel within 0.0002 cm. The specimen faces were entirely coated with gold by vacuum evaporation, and capacitance measurements were performed in a dry-nitrogen atmosphere using a capacitance bridge[17] and a two-electrode dielectric sample holder. The diameter of the electrodes in the specimen holder exceeded the specimen diameter by four times

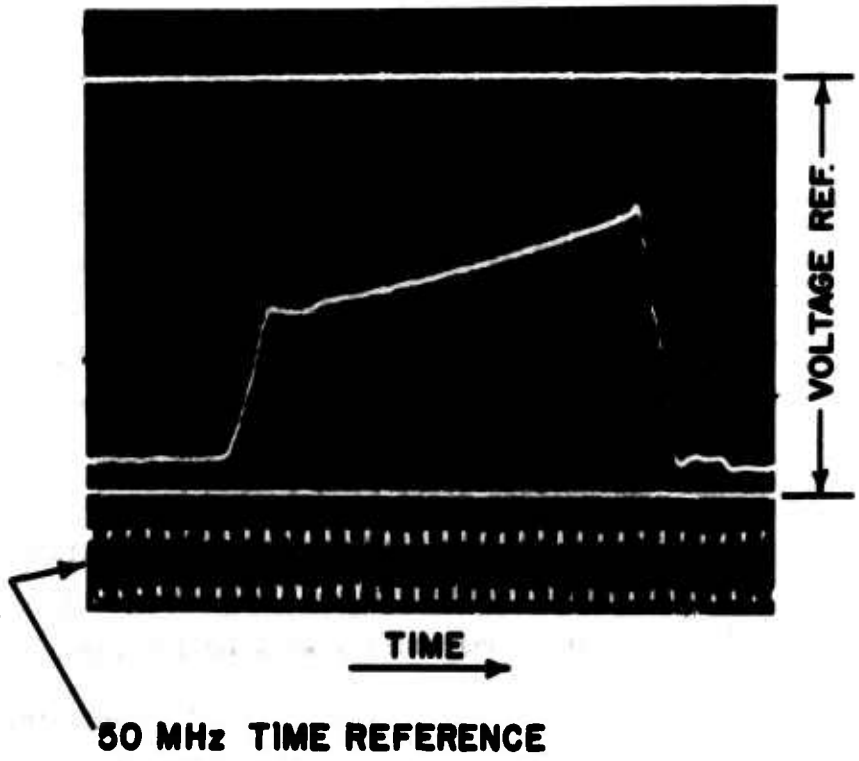

Figure 3. Signal from a shock-compressed polyethylene assembly.

the specimen thickness. The dielectric constant k_o was calculated to be 2.360 ± 0.007. The uncertainty of ± 0.007 was estimated from measurements on the different polyethylene specimens, and from additional measurements on polystyrene, and single-crystal specimens of sodium chloride and lithium fluoride which provided reference checks.

C. Corrections and Experimental Uncertainty

The evaluation of $(Q_m - Q_p)$ is subject to the least uncertainty at high pressures where Q_p is less than a few percent of Q_m. However, at low pressures where Q_p is larger than 0.15 Q_m, a significant error could occur if the applied voltage either suppressed or enhanced the shock-induced polarization signal. For this reason, Q_p was evaluated by two methods at low pressures in the hope that any influence by the applied voltage might be detected by a discrepancy. Unfortunately, an equal and opposite influence by each voltage polarity would be undetected. In this event, the charge $(Q_m - Q_p)$ would be incorrectly evaluated and would always be too large.

An increase in stray capacitance was neglected in deriving Equation (5). However, the free surface of the metal plate was displaced toward the circuit during a measurement and an increase should have resulted. In an attempt to measure the increase in stray capacitance, the test configuration was duplicated, inductance was introduced, the resistance was decreased, and the circuit was set into oscillation at the resonant frequency. When the free surface around the polyethylene assembly was displaced through 1.5 mm, the frequency changed, indicating a 0.059 pF total increase in capacitance. The worst test condition was encountered at 173 kbar where the free surface of a magnesium plate moved 0.9 mm during the time interval from $t = 0$ to $t = X_o/U$. At this pressure, $(Q_m - Q_p)$ was reduced by an additional 0.47 percent to correct for the increase in stray

capacitance. No correction was introduced for the affect of stray capacitance at other pressures, but the influence was considered in the overall uncertainty of the charge measurement.

Shock-induced electrical conductivity is a potential source of error in this technique for dielectric constant measurements. If the dielectric becomes conducting, too much charge is measured, and the calculated value for the dielectric constant is erroneously high. However, the occurrence of significant conductivity should have been indicated by the signals from shock-compressed polyethylene. The guarded electrode was effectively matched to the shock impedance of the polyethylene, and with shock-induced conductivity, the signal should have decreased only to a voltage V_s when the shock front arrived at the guarded electrode. The conductivity σ may be calculated using the relationship,

$$\sigma = \left(\frac{V_s}{V - V_s}\right)\left(\frac{U - u}{U}\right)\frac{X_o}{R_T A} ,$$

where R_T is the resistance that terminates the coaxial cable (see Figure 2). Optimum conditions for detecting conductivity occurred in the plate-impact experiment at 246 kbar, where the pressure was constant in the interelectrode volume and the polarization signal was negligibly small. The signal returned very close to zero, and V_s could not have exceeded 0.2 volt. This imposed an upper limit of 1.6×10^{-5} mho/m on the shock-induced conductivity at 246 kbar, in general agreement with the results of Keeler and Mitchell[10].

Other tests were performed to establish the uncertainty in charge measurement that could be attributed to signal transmission, display, and record reduction. The display of each oscilloscope was calibrated or mapped to evaluate nonlinearity and distortion of the sweep and vertical deflections. A series of charge measurements was then performed using a standard test signal. In some tests, the

signal cable was terminated with its characteristic impedance at the oscilloscope input. In other tests, a closed system was used, and the signal was introduced into the oscilloscope through a high impedance probe from a voltage pickoff. For comparison, measurements were also performed using an oscilloscope with a low impedance input.[18] Variations in the signal line and inputs produced small changes in the signal profiles, but the charge measurements varied less than ± 0.5 percent. As a result of these tests, and considering uncertainty introduced by the stray capacitance, it was concluded that the charge could be measured within ± 1.0 percent, and this figure was used to estimate uncertainty in the dielectric constant.

The uncertainty in the dielectric constant is given by the relationship,

$$\frac{dk}{k} = \frac{dB}{B} + B \left\{ d\,k_o + \frac{1}{V\,\varepsilon_o\,A\,k} \left[Q\,d\,X_o + X_o\,d\,Q - X_o Q \left(\frac{dV}{V} + \frac{dA}{A} \right) \right] \right\},$$

where $B = (U - u)/U$, and Q is the corrected charge $\left[(Q_m - Q_p) \right.$ in Equation (5) $\left. \right]$. For B, the measured quantity is U, and u is obtained from the Hugoniot relationship $U = 2.925 \times 10^3 + 1.570\,u$. It follows that $dB = (2.925 \times 10^3/1.570\,U^2)\,dU$, where the uncertainty in U is the standard deviation. Calculated uncertainties are included in the data table and are indicated by error flags on plotted data points.

IV. RESULTS, DISCUSSIONS AND CONCLUSIONS

Measured values of the dielectric constant are listed in Table I, and data are presented in Figure 4 as $(k - 1)/\rho$ vs ρ, with individual test pressures indicated at the top. From 36 to 246 kbar, $(k - 1)/\rho$ may be represented as a constant, 1.47(2), within the experimental uncertainty. Below 36 kbar, two sets of values are presented. The upper values at 16 and 20 kbar result when the applied

Table I. Results of Dielectric Constant Measurements on Polyethylene under Shock Compression

Pressure kbar	Density (ρ) g/cc	Signal Polarity	k	$\frac{k-1}{\rho}$
0	0.960±0.002		2.360±0.007	1.417±0.011
16±2	1.098±0.010	(-)	2.789±0.053	1.630±0.064
16±2	1.098±0.010	(+)	2.779±0.052	1.621±0.064
16±2	1.098±0.010	(+)	2.672±0.050(*)	1.523±0.060(*)
20±2	1.125±0.010	(-)	2.828±0.052	1.625±0.062
20±2	1.125±0.010	(+)	2.821±0.052	1.619±0.062
20±2	1.125±0.010	(+)	2.681±0.048(*)	1.494±0.056(*)
36.5±2	1.207±0.009	(-)	2.772±0.047	1.468±0.052
36.5±2	1.207±0.009	(+)	2.770±0.047	1.467±0.052
62±3	1.301±0.008	(+)	2.929±0.047	1.483±0.045
118±3.5	1.435±0.007	(+)	3.100±0.043	1.463±0.037
173±4	1.523±0.006	(+)	3.273±0.046	1.492±0.036
246±7.5	1.609±0.005	(+)	3.346±0.043	1.458±0.032

(*) Assuming $Q_p = 0$ when the applied voltage opposes the shock-induced polarization signal.

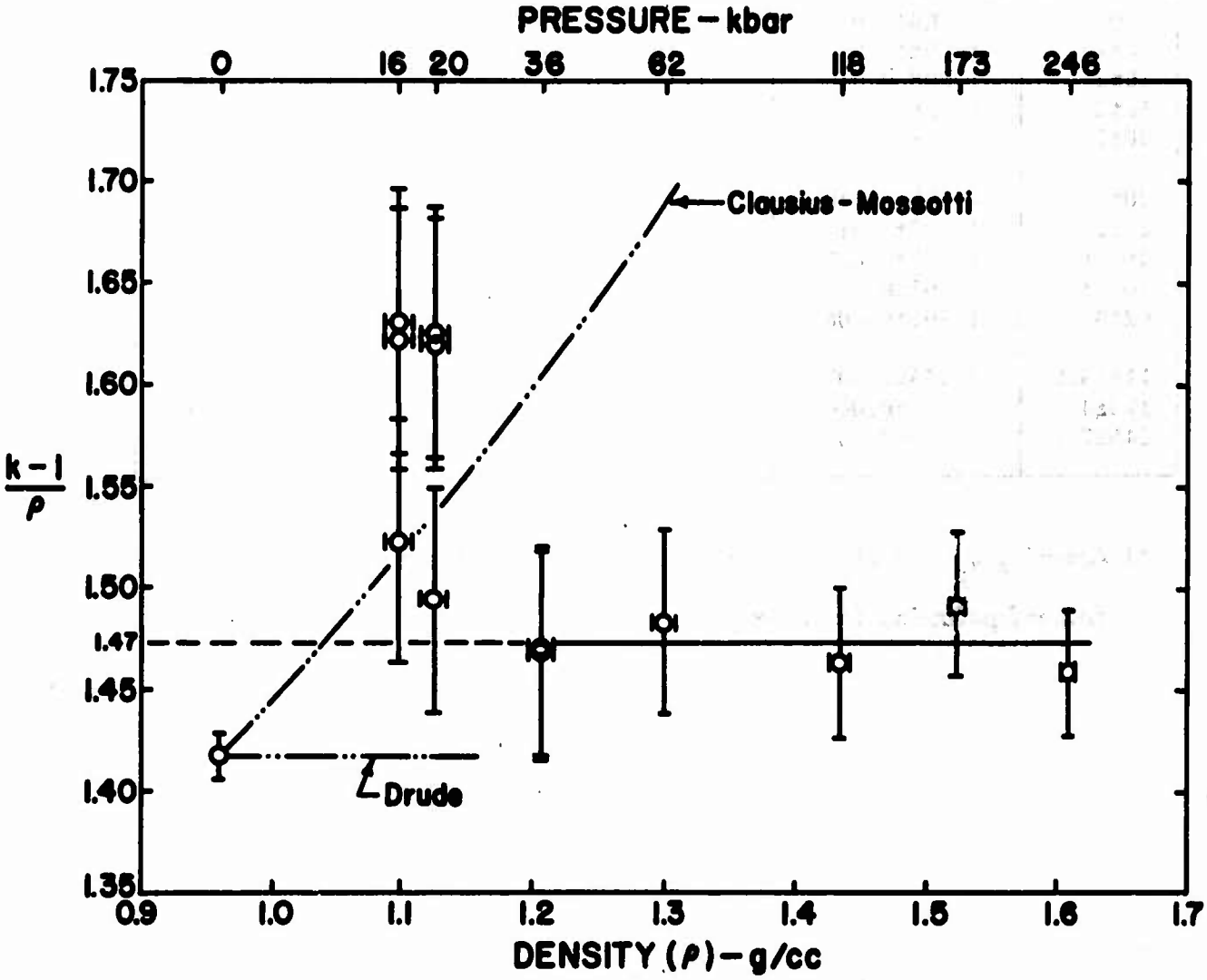

Figure 4. Experimental results, presented as $(k-1)/\rho$ vs density (ρ) of the shock compressed polyethylene.

voltage is assumed to have no influence on the shock-induced polarization signal. The lower values result from assuming a voltage effect.

The applied voltage was assumed to have no influence on Q_p at 36 kbar or at higher pressures. At 36 kbar where Q_p was evaluated by two methods, $\left[Q_m(+) + Q_m(-)\right]/2$ was within 0.14 percent of Q_p (0) and gave no indication of a voltage effect (such close agreement was undoubtedly fortuitous. Another factor to consider is the ratio Q_p/Q_m. At 36 kbar, $Q_p/Q_m \approx 0.15$. This ratio decreases to 0.05 at 62 kbar, to 0.02 at 173 kbar, and becomes negligible at 246 kilobars. An influence by the applied voltage should follow this decreasing ratio. Since $(k-1)/\rho$ remains unchanged as Q_p/Q_m decreases, any voltage effect in the pressure range from 36 to 246 kbar is assumed to be negligibly small. The emphasis placed on the constant trend of the $(k-1)/\rho$ representation may be unwarranted, but the trend does suggest physical significance since it represents dielectric behavior predicted by the Drude equation.

If the applied voltage is assumed to have no influence on Q_p at pressures below 36 kbar, $(k-1)/\rho$ increases abruptly. The polarization charge Q_p was also measured by two methods at 16 and 20 kbar, and was examined for a discrepancy. At 20 kbar, the measurements indicated that $\left[Q_m(+) + Q_m(-)\right]/2 > Q_p$ (0). However, the discrepancy could be explained by oppositely directed errors of 0.4 percent in the measurements of $Q_m(+)$ and $Q_m(-)$, which is within the experimental uncertainty. At 16 kbar, measurements again indicated that $\left[Q_m(+) + Q_m(-)\right]/2 > Q_p(0)$. At this pressure, the discrepancy could be explained by oppositely directed errors of 0.5 percent in the measurements of $Q_m(+)$ and $Q_m(-)$. In evaluation tests, it was possible to measure the charge of a standard signal within ± 0.5 percent; and since it should be possible to neglect changes in stray capacitance when comparing the measurements from similar experiments, it is marginally possible that a discrepancy actually exists in the

two measurements of Q_p at 16 kilobars. Furthermore, it had been anticipated that $\left[Q_m(+) + Q_m(-)\right]/2$ should exceed Q_p (0) if the applied voltage influenced the polarization signal. The extent of any influence could not be determined. Therefore, it was assumed as a limit that Q_p becomes zero when the applied voltage opposes the polarity of the polarization signal. This limit results in the lower values at 16 and 20 kbar in Figure 4. The lower values are appealing because they are in agreement with the constant that represents the data at higher pressures, and can also be reached from zero pressure using the Clausius-Mossotti equation.

Signal profiles were calculated at 16 kbar in an attempt to evaluate the two values for the dielectric constant. One profile was calculated by Equation (4) using Q_m and the lower value of k. A second profile was calculated by Equation (4) using $(Q_m - Q_p)$ and the higher value of k, and was added algebraically to a polarization signal profile calculated by the Allison theory[19] using Q_p and the higher value of k. Unfortunately, both calculated profiles closely followed the profile of the test signal at 16 kilobars. The lower value of k produced a profile that was slightly too flat, while the higher value produced a profile that was slightly too steep. This result suggested the need for an intermediate value of k, which is not unreasonable since it is unlikely that the applied voltage actually reduced Q_p to zero (if, indeed, a voltage effect exists).

In another attempt at evaluation, the two values of k were used to calculate the profile of a shock-induced polarization signal at 16 kilobars. In these calculations, the higher value produced the profile in better agreement with the measured polarization signal. However, the result was not considered to be conclusive evidence that the higher value of k is correct.

The measurements at 16 and 20 kbar do not permit a particular value of k to be established with confidence, so two values have been included in Table I and Figure 4. These values are intended to serve

as limits, and k may lie at either limit, or in between. Despite this uncertainty, some change in dielectric behavior undoubtedly occurs at low pressures. The constant that represents $(k-1)/\rho$ at high pressures is approximately four percent above the zero-pressure value, so $(k-1)/\rho$ must increase during small compressions. Gibbs and Jarman[20] measured the dielectric constant of polyethylene during static compression to approximately three kilobars, and found only one percent deviation from change predicted by the Clausius-Mossotti equation, assuming constant polarizability. Under shock compression, it also seems likely that the dielectric constant increases at low pressures according to the Clausius-Mossotti prediction. The pressure at which the behavior changes cannot be established by these measurements. It is possible that the change is associated with a low-pressure transition. Hugoniot measurements provide no definite indication of a transition between 16 and 246 kilobars. Also, static measurements by Bridgman[21] gave no indication of a transition in the pressure range below 39 kilobars. However, Van Valkenburg and Powers[22] found evidence of a low-pressure transition during static compression of polyethylene between diamond anvils. The transition was observed optically, was accompanied by an irreversible decrease in thickness, and post-test analysis suggested the presence of a triclinic cell. However, no transition pressure was reported. Cleron, Coston, and Drickamer[23], reporting on apparatus for NMR studies at high pressures, presented data on polyethylene in the pressure range below 25 kilobars. The width of the ^1H resonance line as a function of pressure showed no measurable change until a pressure of five kilobars. A rapid increase occurred above five kbar and leveled off near 22 kilobars. The sudden increase in line-width above five kbar was not attributed to a phase transition, but it was noted that the motion of the polyethylene chains suddenly became severely restricted.

Between 36 and 246 kbar, the dielectric constant of polyethylene varies according to the Drude equation, where $k-1 = 1.47\ \rho$. This

suggests that the polarizability is constant, and that the local field is given by the applied field. Calculations by Brodsky and Burstein[13] and Gill and Bloembergen[14] have shown that the effective field for electronic polarization is the macroscopic applied field when the electrons are delocalized. Royce[11], in considering dielectric constant data for polymethyl methacrylate obtained from shock-induced polarization experiments,[7] suggested that electrons may become delocalized as a result of shock compression. This suggestion would also provide an appropriate explanation for the dielectric behavior of shock compressed polyethylene.

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13. ABSTRACT

The dielectric constant of high-density polyethylene was measured under shock compression in the pressure range from 16 to 246 kilobars. A charged-capacitor technique was used, and the dielectric constant was calculated from the charging current measured during shock compression. From 36 to 246 kilobars, the dielectric constant was found to increase according to the Drude equation, assuming constant polarizability; and in this pressure range it was represented by the relationship $k - 1 = 1.47\rho$, where ρ is the density.

rho

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