Journal of Applied Mechanics and Technical Physics, Vol. 38, No. 6, 1997

ELECTRICAL CONDUCTIVITY OF POLYTETRAFLUOROETHYLENE UNDER SHOCK-WAVE LOADING AND RAREFACTION

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UDC 537.312.(6+9)

Electrical conductivity is a fundamental property of substances, which is studied in solid-state physics. Electrical-conductivity measurements in the region of high temperatures and high pressures is one of a few methods that provide information on changes in the state of substances in this region [1–5].

Polytetrafluoroethylene (Teflon) is widely used as an ambient medium for specimens and gauges in studies under shock-wave loading conditions [5–7] because of its high insulating properties and also because of the proximity of its shock adiabat to the shock adiabats of graphite and high explosives (HE). In electrical measurements, data on its electrical conductivity under these conditions are required to estimate the degree of shunting of tested objects embedded in Teflon. These data are also necessary in the design of fast high-current explosive switches [8]. The electrical conductivity of Teflon under single shock-wave loading was studied in [9, 10].

The goal of the present paper is to extend the pressure range of studies to 100 GPa, to obtain data on the electrical conductivity of Teflon under double shock-wave loading and also under loading with subsequent rarefaction, to establish the role of temperature and pressure in the increase in the conductivity, and to refine the nature of the "breakdown" phenomenon observed in [9, 10].

Measurement Procedure. The geometry of the experimental setup is shown in Fig. 1a. A specimen 1 of Teflon (FP-4-60, TU No. M525-54) was located on a charge between a plate 2, which served as an electrode, and the upper electrode 3 in a Teflon ring 4. In some experiments, the specimen and the ring were manufactured as a unit part (Fig. 1c). The upper electrode was made of aluminum, copper, and graphite. In some experiments (Fig. 1b), a disk 3 of copper foil with strip leads was used as the electrode. In this case, the foil was pressed to the specimen by a disk 4 of Teflon, Plexiglas, and plastic foam.

Plane shock waves were formed in specimens by means of HE charges 5 with diameters 120 and 200 mm upon incidence of a detonation wave on plate 2 or upon impact on it by a metal plate 2–7 mm thick accelerated by explosion products. Asymmetry of the entry of the shock front into the specimen was produced by lens charges and was not worse than 0.05 μsec. The initial temperature of the specimens was (283 ± 10) K. The states of the fluoroplastic were determined by calculations of p–u diagrams using the known parameters of the charges and the D–u relations for the plate materials, given for iron in [11] and for copper and aluminum in [12]. The shock adiabat of the Teflon studied ($\rho_0 = 2.19$ g/cm^3) was constructed by the D–u relation from [12]:

$$D = 1.95 + 1.67u$$

(D and u in kilometers per second).

We used two setups which had different measurement limits and which were similar to those described in [1, 2, 5, 13]. Diagrams of measuring run Nos. 1 and 2 are shown in Fig. 1a and b. Starting of the oscillograph and recording of a signal from the entry of a shock wave into the specimen were performed using auxiliary contacts and diagrams. The measured resistances of the specimens R were calculated by the well-known formulas [5, 13] with allowance for the wave resistances of cables.

All-Union Institute of Technical Physics, Snezhinsk 456770. Translated from Prikladnaya Mekhanika i Tekhnicheskaya Fizika, Vol. 38, No. 6, pp. 16–22, November–December, 1997. Original article submitted April 5, 1996.

 0021-8944/97/3806-0826 $18.00

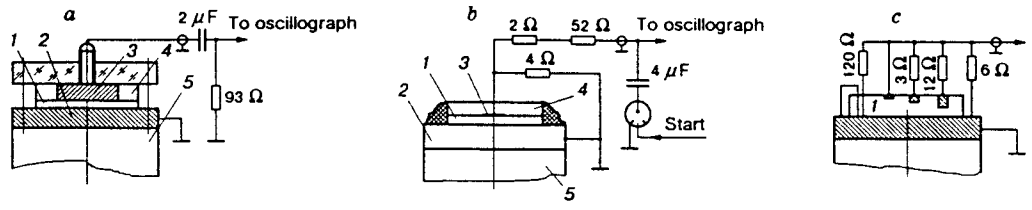

Fig. 1 Diagram of experiment: run No. 1 (a), run No. 2 (b), and run No. 21 (c), 1) specimen; 2) plate; 3) upper electrode; 4) antirarefaction ring (a) or rarefaction medium (b); 5) HE charge.

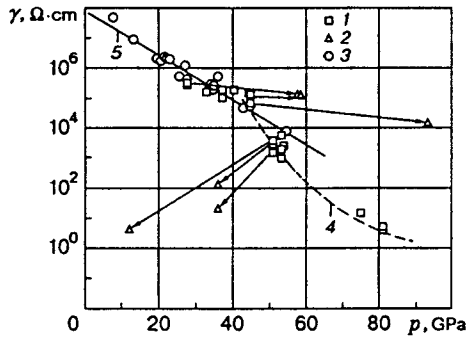

Fig. 2

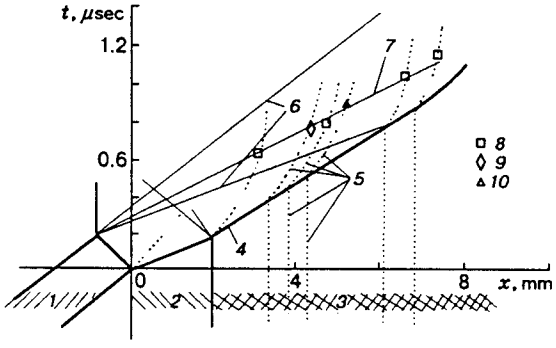

Fig. 3

Fig. 2. Resistivity of Teflon versus pressure: points 1 and 2 are the experimental data of this paper for single and double compression (rarefaction), respectively, and points 3 refer to the experiment of [10], curve 4 is the approximation of the experiment for $p \geqslant 45$ GPa and linear $\ln\gamma = f(1/T)$, and curve 5 is the approximation of the data of [10].

Fig. 3. Position of the high-electrical-conductivity front of Teflon on the x–t diagram: 1) impactor (aluminum); 2) plate (aluminum); 3) Teflon; 4) shock-wave front in Teflon; 5) position of the lower boundaries of electrodes; 6) characteristics of the rarefaction wave; 7) characteristic along which the high-conductivity front is transferred; 8) run No. 21; 9) run No. 13; 10) run No. 16.

The resistivity γ of the specimen material under shock compression was calculated from the universal dependence [13] $dR/\gamma = f(l/d)$, which takes into account the edge effects. Here d is the diameter of the upper electrode and l is the thickness of the compressed specimen at the moment the shock wave reaches the upper electrode. The validity of this dependence was checked by electrolytic modeling of the geometry of the setups using running water as an electrolyte.

Measurement Results. The results obtained are given in Table 1 and Fig. 2. It is evident that, under single compression of Teflon by a shock wave with an amplitude of 27.5 GPa, its resistivity decreases by more than 11 orders of magnitude compared to the initial value of $\sim 10^{17}$ $\Omega \cdot$ cm [14]. In the range of 27.5–45 GPa, γ changes within one order of magnitude. With increase in the shock-wave amplitude from 45 to 80 GPa, γ decreases more strongly (from $\sim 10^5$ to ~ 5 $\Omega \cdot$ cm).

Under double shock compression of Teflon in the pressure range of 58–94 GPa (behind the second shock wave), γ decreases only slightly with increase in p_2 (by only an order of magnitude).

Characteristically, in experiments at shock-wave pressures higher than 33 GPa, the resistance of the specimen additionally decreases sharply within 0.5–1 μsec after the jump with the arrival of the wave at the upper electrode. This decrease in the resistance corresponds to the decrease in the resistivity of the specimen to values of 1.0–0.1 $\Omega \cdot$ cm. Kuleshova [9] called this phenomenon the "breakdown" of Teflon.

TABLE 1

Experiment No.	Material	Dimensions of the specimen under the electrode $l_0 \times d$, mm	p_1, GPa	γ_1, $\Omega \cdot$ cm	p_2, GPa	γ_2, $\Omega \cdot$ cm
1	Cu/Cu (6.0)	1.59 × 31.1	27.5	$3.0 \cdot 10^5$	57.7	$1.2 \cdot 10^5$
2	Cu/graphite(6.0)	1.73 × 30.1	27.5	$3.4 \cdot 10^5$	—	—
3	Cu/Cu (0.05)	2.00 × 30.0	33.1	$1.7 \cdot 10^5$	—	—
4	Al/graphite (5.0)	1.97 × 30.0	37.2	$1.8 \cdot 10^5$	—	—
5	Al/Al (0.12)	1.98 × 35.0	37.2	$1.1 \cdot 10^5$	—	—
6	Al/Al (0.13)	2.00 × 35.0	40.3	$1.9 \cdot 10^5$	—	—
7	Al/Al (0.1)	2.00 × 35.0	44.5	$1.35 \cdot 10^5$	—	—
8	Al/Al (6.0)	2.00 × 30.0	44.6	$1.2 \cdot 10^5$	58.7	$1.1 \cdot 10^5$
9	Al/Cu (6.0)	2.00 × 30.0	44.6	$7.0 \cdot 10^4$	93.2	$1.3 \cdot 10^4$
10	Al/Cu (0.03)	0.98 × 20.0	51.0	$1.6 \cdot 10^3$	36.0	$1.9 \cdot 10^1$
11	Al/Cu (0.03)	0.96 × 20.0	51.0	$2.8 \cdot 10^3$	36.0	$1.2 \cdot 10^2$
12	Al/Cu (0.05)	0.98 × 20.0	51.0	$3.7 \cdot 10^3$	12.0	$3.8 \cdot 10^0$
13	Al/graphite (6.0)	1.60 × 26.0	53.8	$2.7 \cdot 10^3$	—	—
14	The same	1.61 × 28.0	53.0	$1.9 \cdot 10^3$	—	—
15	»	1.60 × 34.5	53.0	$6.0 \cdot 10^3$	—	—
16	Al/Al (0.12)	2.17 × 35.0	53.0	$1.0 \cdot 10^3$	—	—
17	Fe/graphite (6.0)	1.50 × 10.0	75.2	$1.4 \cdot 10^1$	—	—
18	The same	1.50 × 10.0	75.2	$1.5 \cdot 10^1$	—	—
19	»	1.64 × 3.0	81.0	$4.8 \cdot 10^0$	—	—
20	»	1.50 × 10.0	81.0	$3.9 \cdot 10^0$	—	—
21	Al/graphite	1.88 × 6	53.0	—	—	—
	(3 electrodes)	4.08 × 6	—	—	—	—
		4.80 × 6	—	—	—	—

Note. In the second column, the plate material is indicated ahead of the stroke and the material and thickness of the electrode in millimeters are indicated behind the stroke, p_1 and p_2 are the pressures behind the 1st shock front and behind the front of the reflected shock wave (rarefaction wave), and γ_1 and γ_2 is the resistivity under the 1st and 2nd loading.

To refine the nature of this phenomenon, we performed run No. 21 (Fig. 1c), in which three graphite electrodes with diameter 6 mm were cut in a Teflon specimen at the same distance from the axis of the charge and at different distances from the plate. In this experiment ($p_1 = 53$ GPa), we detected a sequential sharp decrease in the resistance of the specimen under the electrodes, which was associated with the perturbation that moved behind the shock-wave front in the rarefaction wave propagating from the rear side of the impactor plate. Computer simulation of the loading of the specimen under conditions of run No. 21 shows that the front of the high-conductivity zone propagates along the characteristics of the rarefaction wave on which the pressure is 30 GPa. The plotting of the moments of breakdown on the x–t diagram for other runs (Fig. 3) supports the aforesaid. Experiments performed on charges with the thickness of the impactor plates differing by two times showed a unique relationship between the time of "breakdown" and the time interval between the arrival of the shock front and the rarefaction wave at the upper electrode.

To obtain an additional confirmation of the relationship between the "breakdown" of Teflon and its rarefaction, we performed run Nos. 10–12 in which rarefaction of loaded specimens was realized from the side of the upper foil electrode into PMMA and polystyrene foam with a density of 0.35 g/cm^3. Figure 2 and Table 1 show that, with rarefaction to a pressure of 36 GPa, the resistivity of the specimen decreases to 120 and 19 $\Omega \cdot$ cm, and, for rarefaction to a pressure of 12 GPa, it decreases to 3.8 $\Omega \cdot$ cm.

Discussion of the Results. Figure 2 shows, in addition to our results, the experimental points and the straight line that approximates them from [10]. It is evident that, at pressures of 27.5–45 GPa, the data show satisfactory agreement within the spread of experimental points. However, at high pressures, our curve

TABLE 2

Experiment No.	ρ_{00}, g/cm^3	D, km/sec	$\bar{D}$, km/sec	$\bar{u}$, km/sec	$\bar{p}$, GPa	V, g/cm^3
22	1.670	6.98	7.07	3.575	43.5	0.2874
23	1.770	7.15				
24	1.670	8.49	8.47	4.405	64.2	0.2790
25	1.770	8.46				

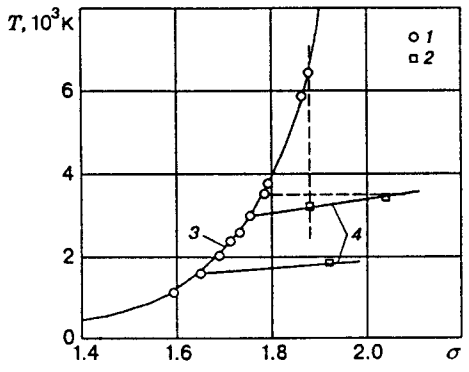

Fig. 4

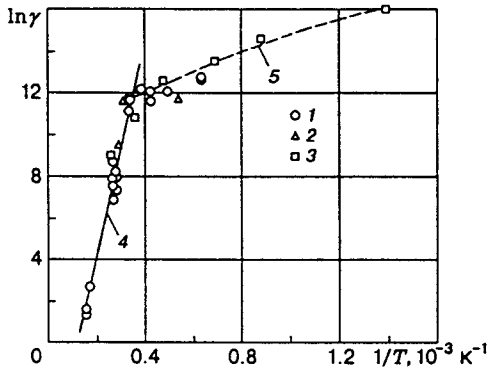

Fig. 5

Fig. 4. T–σ diagram of the states of Teflon under single and double compression: 1 and 2 are the calculated points of single and double compression, 3 and 4 are the curves of state under single and double compression, and the dashed curves refer to T = const and σ = const for comparison of single and double compression.

Fig. 5. Logarithm of the resistivity of Teflon versus the inverse temperature: points 1 and 2 refer to the experiment of the present work, single and double compression, respectively, points 3 refer to the experiment of [10], and points 4 and 5 are approximations of the experimental data for $T < 3000$ K and $T > 3000$ K, respectively.

of $\log \gamma = f(p)$ is steeper.

Since the electrical conductivity of solids is determined primarily by two independent factors temperature and pressure (compression), it is of interest to establish their role in the variation of the conductivity.

To obtain data on the thermal components of the equation of state for Teflon, we performed four experiments on electrocontact recording of the shock-wave velocity on a 3-mm base in a porous material (particle size 0.05 × 0.15 × 0.15 mm and average density of the specimens of 1.72 g/cm^3). Using the reflection method of [15], we obtained two points of the shock adiabat of porous Teflon (Table 2), which allowed us, using the procedure described in [15], to find the value of the Gruneisen coefficient $\Gamma = 1.43$ for a specific volume of ≈ 0.28 cm^3/g.

Then, assuming that Γ = const, we obtained the Mie–Gruneisen equation of state with the cold and thermal pressure components p_{cold} and p_{therm} and energy components E_{cold} and E_{therm} [15]. This equation allowed us to calculate the experimental parameters (see Table 1) of single compression of Teflon (Fig. 4), including its temperature (under the assumption of constant heat capacity $c = 1050$ J/(kg·K) [14]). Using this equation of state, we also obtained adiabats of the second compression realized in the experiments and the temperature values for double compression (Fig. 4).

The experimental values of the resistivity of Teflon under single and double loading were compared using the T–σ diagram (Fig. 4) for the same compression ($\sigma_1 = \sigma_2 = 1.878$) and the same temperature

TABLE 3

Compression	σ	T, K	p, GPa	γ, $\Omega \cdot$ cm	Compression	σ	T, K	p, GPa	γ, $\Omega \cdot$ cm
Single	1.878	6440	81.0	4.4	Double	1.878	3200	59.5	$1 \cdot 10^5$
Single	1.776	3440	48.6	$2.2 \cdot 10^4$	Double	2.040	3440	93.2	$1.3 \cdot 10^4$

$T_1 = T_2 = 3440$ K (Table 3). Evidently, at constant temperature, the resistivity changes only slightly with increase in σ, and at constant σ, it decreases strongly with a rise in temperature. Hence it follows that the effect of temperature on γ is predominant under shock-wave loading.

The curve of $\ln\gamma = f(1/T)$ (Fig. 5) is of interest. Note that the experimental points for double compression of Teflon fit the curve in the coordinates $\ln\gamma$, $1/T$ obtained for single loading. This is additional confirmation of the dramatic impact of temperature on the variation in resistivity of the material under shock-wave loading. It can be seen from Fig. 5 that, at $T > 3000$, the dependence is linear and corresponds to

$$\gamma = \gamma_0 \exp(E_a/kT),$$

where $E_a = 4.6$ eV is the activation energy of conduction, and $\gamma_0 = 1.2 \cdot 10^{-3}$ $\Omega \cdot$cm is the resistivity that corresponds to the so-called [16] minimal conduction. The value of γ_0 that is obtained is typical of many amorphous and liquid semiconductors [16]. For $T < 3000$, the dependence $\ln\gamma = f(1/T)$ is significantly weaker and corresponds to the activation energy of conduction 0.1–0.3 eV. In solid state physics [16], this character of the dependence $\ln\gamma = f(1/T)$ is usually associated with the change of the type of conduction at the point of inflection.

The large (by 11 orders of magnitude) increase in the conductivity of Teflon under loading by a shock-wave pressure of 27.5 GPa can be explained, as in the case of ionic crystals [17], by the transition of the specimen to a semiconducting state with donor and acceptor levels in the forbidden zone that are formed by defects generated by the shock wave. Thermal ionization of these levels ensures electron conduction, which increases with increase in temperature. The low activation energy of conductivity in the loading range of 27.5–45 GPa ($T < 3000$ K) indicates saturation of the dependence of the rate of increase in defect concentration with increase in the shock-wave amplitude.

The sudden change in the slope of the curve of $\ln\gamma = f(1/T)$ at $T > 3000$ K and the sufficiently high activation energy (4.6 eV) indicate transition to intrinsic conduction with thermal transfer of charge carriers over the forbidden zone [16].

In connection with the aforesaid, the hypothesis on the destruction (depolymerization) of Teflon at pressures higher than 45 GPa appears to make sense. Destruction under shock-wave compression conditions should be accompanied by the occurrence of a large number of uncompensated carbon anions and a sudden increase in the number of charge carriers.

If this hypothesis is true, the observed "breakdown" phenomenon of Teflon can be associated with rarefaction of the depolymerized material. The occurrence of the "breakdown" is explained only from a certain pressure threshold — the boundary of the destruction region. Lower values of the threshold loading pressure (33 GPa in the present work, 37.5 GPa in [9], and 35 GPa in [10]) for which "breakdown" occurs during rarefaction may indicate the existence of the range of 33–45 GPa of the metastable polymeric state of Teflon, which breaks by depolymerization only during rarefaction. The significant increase in conductivity during "breakdown" should then be associated with a sudden increase in the number of charge carriers or with an increase in their mobility during rarefaction of the depolymerized material.

Note the validity of use of the word "breakdown" in quotation marks in [9], because the observed phenomenon is not an electrical or thermal breakdown of a dielectric, which occurs in a strong electric field via the propagation of an electron avalanche [18].

Approximation of our experimental points by a straight line in the coordinates $\ln\gamma, 1/T$ leads to the necessity of approximating these points in the coordinates $\ln\gamma, p$ by a nonlinear curve with a downward

convexity in the range of 45–80 GPa and with a point of inflection at $p \cong 45$ GPa (see Fig. 2).

Conclusions. (1) During single shock-wave loading of Teflon with a wave amplitude of 27.5 GPa, its resistivity decreases from the initial resistance by 11 orders of magnitude and is about $3 \cdot 10^5$ $\Omega \cdot$ cm. With increase in pressure to 45 GPa, the resistivity decreases additionally by one order of magnitude. With increase in pressure from 45 to 81 GPa, the resistivity of Teflon increases significantly from $\sim 10^5$ to 5 $\Omega \cdot$ cm. Under double shock-wave loading of Teflon (in the first wave, to 27.5–45 GPa, and, in the second wave, to 58–94 GPa), its resistivity decreases relatively slightly, by an order of magnitude (from $\sim 10^5$ to $\sim 10^4$ $\Omega \cdot$ cm) with increase in pressure.

(2) The decrease in the resistivity of Teflon with increase in pressure under both single and double shock-wave loading is associated exclusively with a rise in temperature (and not with an increase in pressure).

The curve of the logarithm of the resistivity versus the inverse temperature is linear at $T > 3000$ K and corresponds to an activation energy of 4.6 eV. At $T < 3000$ K, the activation energy of conduction is 0.1–0.3 eV. It is suggested that there is a relationship between the point of inflection near 45 GPa ($T \simeq 3000$ K) in the curve considered and destruction (depolymerization) of the material.

(3) The sudden increase in the conductivity of Teflon observed previously and in the present work some time after shock-wave loading (which is called "berakdown" in [9]) is associated with rarefaction of the specimen and occurs only when the pressure threshold of 33 GPa is exceeded. This occurs in rarefaction waves that propagate from both the rear and the leading surface of the specimen. At $p_1 = 53$ GPa, the front of the high-conductivity zone propagates in the rarefaction wave along the characteristic that corresponds to a rarefaction pressure of 30 GPa. The resistivity in this case reaches ~ 4 $\Omega \cdot$ cm. It is suggested that this phenomena is associated with the rarefaction of Teflon from the region of its destruction (depolymerization) or the region of metastability of the polymeric state.

The authors are grateful to A. T. Sapozhnikova and A. V. Pershin for computer simulation of the conditions of run No. 21.

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