

Anomalous low resistance of compressed dielectric thin films

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Uniaxial compression of thin films of polymer dielectrics causes a sharp decrease in resistivity, by several orders of magnitude. This decrease is not a consequence of a high-pressure metal-dielectric phase transition.

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The electrical phenomena, which occur in dielectric films with thicknesses no greater than a few tens of microns, are of considerable interest, both for determining the mechanism for the electrical conductivity of these films and for several applications. The charge-transport mechanism in such systems may be quite different from that found for the bulk materials,¹ so that effects may occur in thin films which do not occur in thick samples.

In this letter we are reporting the observation of an abrupt decrease in the electrical resistance R of thin films of polymer dielectrics and also of (polymer insulator)–metal composites subjected to uniaxial compression. At a pressure $P = 10$ kbar, at room temperature, the lowest values of R of some of the systems studied are no higher than the values which should be exhibited under ordinary conditions by a geometrically identical metal sample (titanium, for example, with a conductivity of 10^4 mho/cm). The dependence of R on the film thickness l and the temperature T indicates, however, that this effect apparently cannot be interpreted as a consequence of the well-known insulator–metal phase transition.²

An increase in the conductivity of dielectric thin films was observed during uniaxial compression of polyethylene terephthalate (e.g., Dacron), polyimide, polypropylene, and polytetrafluoroethylene (e.g., Teflon) films ($l = 2\text{--}30\text{ }\mu\text{m}$). A similar effect was observed in the compression of polypropylene–copper, polytetrafluoroethylene–copper, and several other polymer composite systems ($l = 70\text{--}200\text{ }\mu\text{m}$). The uniaxial compression was carried out in a high-pressure Bridgman anvil, in which P could be varied from a few hundred bars to 30 kbar. After the anvil had been ground and polished with a diamond paste, the height of the microscopic irregularities on the flat surfaces of the anvil, which were in contact with the test samples, did not exceed $1\text{ }\mu\text{m}$. Figure 1 illustrates the procedure for measuring the resistance during the uniaxial compression. The resistance R was found by three independent methods: from the readings of an Shch-34 automatic ohmmeter with a dc bridge circuit, with an ac bridge operated at 1547 Hz, and from the slope of the linear I-V characteristics recorded over the voltage interval 0–10 mV. The results found by these methods agreed with 5–12%.

Figure 2 shows a plot of the resistance of a polypropylene film $l = 12\text{ }\mu\text{m}$ thick vs

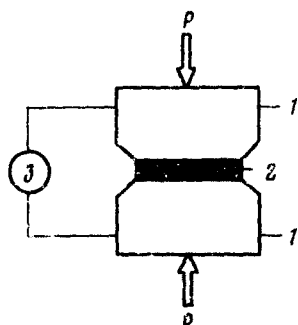


FIG. 1. Block diagram of the apparatus used to measure the resistance of the dielectric-film samples during their uniaxial compression. 1—Bridgman anvil; 2—sample; 3—measuring instrument. The arrows show the direction of compression.

the pressure. It follows from these results that as P is raised from 500 bar to a certain critical P_{cr} (13 kbar for this particular film) the value of R decreases only slightly. At $P = P_{cr}$, the resistance drops sharply, by seven orders of magnitude. At $P > P_{cr}$, there is again a slight decrease in R as the samples are compressed. These changes are reversible: When the pressure is removed, the resistance increases to its original of 500 bar.

The minimum resistance (R_{min}) achievable during the uniaxial compression of these dielectrics could not be determined because it was lower than the resistance of the leads. It can be assumed, however, that this minimum resistance satisfies $R_{min} \leq 10^{-3} \Omega$, even for those polymer systems for which reliable measurements were possible under conditions such that the ratio of l to the sample area S was 10 cm^{-1} .

Under otherwise equal conditions the critical pressure P_{cr} depends on the film material. At room temperature, for example, for a thickness $l = 20 \mu\text{m}$, the transition of the polyimide to the low- R state occurs at $P_{cr} = 10 \text{ kbar}$, while the polypropylene film of this thickness retains its high resistance ($\sim 10^8 \Omega$) over the entire pressure range studied. Furthermore, P_{cr} decreases with increasing T and with decreasing l . Interestingly, for a multilayer sample consisting of seven identical polyimide films ($l = 20 \mu\text{m}$) separated by sheets of aluminum or copper foil the critical pressure P_{cr} agrees within 15% with that found for a single-layer polyimide sample with a thickness of $20 \mu\text{m}$. On the other hand, no abrupt change is observed in the resistance of

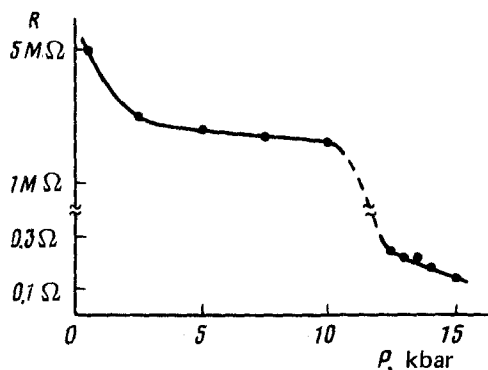


FIG. 2. Dependence of the resistance R of a $12\text{-}\mu\text{m}$ -thick polypropylene film on the pressure P .

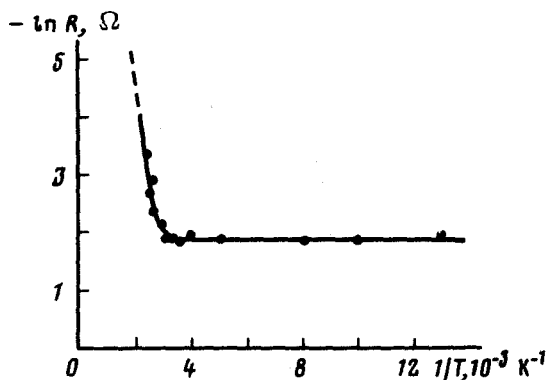


FIG. 3. Temperature dependence of the resistance of a 8- μ m-thick polyimide film at a constant pressure $P = 1$ kbar.

a polyimide film with $l = 140 \mu\text{m}$ over the pressure range studied. Similar results were found for other dielectrics.¹

When these polymer films were subjected to uniaxial compression at $P > P_{\text{cr}}$, they exhibited not only an anomalously low resistance but also several other distinctive properties. In particular, the value of R measured at a constant pressure above the critical value depends only slightly on the temperature over the range from 77 to 330–340 K; at higher temperatures the resistance falls off in the exponential manner typical of dielectrics and semiconductors (Fig. 3). The activation energy decreases with increasing P .

At room temperature and at a constant pressure, the film resistance varies in a nonlinear way with the film thickness. The experimental results show that

$$R = \frac{A}{S} \exp(l^2/l_0^2),$$

where l_0 and A are empirical parameters. For polyethylene terephthalate films with thicknesses in the range 2–10 μm , the values of l_0 and A at $P = 5$ kbar are 2.1×10^{-4} cm and $0.15 \Omega \cdot \text{cm}^2$, respectively.

The sharp decrease in the resistance of the polymer dielectric thin films under pressure is not a consequence of a change in the film thickness during the compression, since the relative change in the volume of the systems studied was no greater than 15–20% at $P \leq 40$ kbar (Ref. 3). No macroscopic defects, which might be responsible for the observed effect, were found in the samples.

It might be suggested that the abrupt change in the resistance during the compression is caused by another trivial factor: direct contact of the anvil at $P = P_{\text{cr}}$. To check this possibility, we carried out experiments with some washers as spacers. It was found that the resistance of the washer itself was not changed by the pressure. The same result was found when the washer was replaced by a disk spacer at the center of the anvil. These results mean that the microscopic irregularities on the anvil surface do not form conducting bridges in either the central or peripheral part of the

interelectrode gap. Nevertheless, if a test film is placed in the aperture of a washer, the transition to the low-resistance state will be completely reproducible. The abrupt change in the resistance is also observed for a sample consisting of a disk spacer surrounded by a ring of the test dielectric film. These results thus make it possible to rule out the direct contact of the anvil as an explanation for the effect.

We should emphasize that the sharp decrease observed in these experiments for the dielectric thin films and for the polymer composites cannot be interpreted as an insulator-metal transition, since the T dependence of R at $P > P_{cr}$ differs from that which has been established for materials exhibiting a metallic conductivity. The anomalously low resistances of these systems apparently mean that the conductivity mechanism which operates in polymer dielectrics subjected to uniaxial compression is fundamentally different from the mechanism which operates under ordinary conditions.

¹⁾When the metal-foil measurement electrodes, insulated from the anvil, did not overlap in plan view, no transition of the dielectric film to a conducting state was observed.

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