Electrical Resistivity of a Polyamide–Epoxide Copolymer

This article is the third in a series of investigations into the electrical volume resistivity as a function of temperature of various types of polymers. Previously, we have reported on a crosslinked amorphous polymer[1] (a resole type phenolic) and a semiflexible linear polymer[2] (a polyamide quinoxaline). Presented herein are the results for a flexible crosslinked polymer (a polyamide–epoxide copolymer).

This copolymer consisted of 60% epoxide (ERL-2795, Bakelite Corp.). The epoxide contains 10% of a reactive diluent, butylglycidyl ether, and has an epoxide value of 0.54/100 g. Polymerization was conducted by mixing the epoxide with 40% of an amine terminated aliphatic polyamide (Versamid 125, General Mills Corp.), which is a condensation product of a dimerized linoleic unsaturated fatty acid and an excess of an alkyl polyamide having an amine value of 290–320.[3] This mixture was poured into a cylindrical electrode[4] and was polymerized directly to the electrodes. Polymerization was conducted for 24 h at 170°C. Measurements of the temperature dependence of the resistivity were made by monitoring the resistivity of the completely polymerized copolymer as it was slowly cooled from 170°C.

Shown in Figure 1 is a semilog plot of resistivity vs. the reciprocal of the absolute temperature for this copolymer. At large values of reciprocal temperature (i.e., low temperature), this Arrhenius plot is linear. At 55°C, however, the curve changes slope, gradually curving more and more at high temperature. As is well established, the initial change in slope corresponds to the glass transition temperature T_g of the copolymer. The value found here, 55°C, agrees reasonably well with that found by others,[3] about 60°C. Normally, a polyepoxide of this type will exhibit a glass transition temperature somewhere in the range 110–160°C, depending on the amine hardener employed. In the present case, the amine-terminated aliphatic polyamide has lowered the glass transition temperature significantly through the mechanism of internal plasticization.

The curvature of the Arrhenius plot at temperatures higher than the glass transition has previously been noted in plasticized systems.[4,5] In this case, we have internal plasticization in which aliphatic polyamide reacts with the epoxide molecule and the aliphatic chains separate the rigid benzene rings of the epoxide molecule.[3] Thus, changes in temperature produce progressive structural changes in the short range order of this polymer in addition to the free volume increase resulting from thermal expansion. At temperatures less than the glass transition, the only effect is the thermal expansion, and the Arrhenius plot is linear.

From the slope of the Arrhenius plot at temperatures less than T_g, we find an activation energy for conduction of $E_c = 57$ kcal/mol. This relatively high value is consistent with the magnitude of the resistivity at room temperature, $\rho_{25°C} = 2 \times 10^{16}\ \Omega \cdot \text{cm}$. We have previously shown that the room temperature resistivity and the activation energy of polymers are not independent. A plot of log $\rho_{25°C}$ vs. E_c for various polymers was shown to be linear. Two somewhat different plots were found. One plot was found to correlate data for unplasticized polymers[6] and the other for externally plasticized polymers.[5] For any given activation energy, the resistivity for externally plasticized polymers is lower (by as much as 5 decades) than for unplasticized polymers. The data for this polymer fit the relationship for externally plasticized polymers. This is the first crosslinked polymer to fit the above relationship, and it demonstrates that the effect of internal plasticization is similar to that of external plasticization and is more important than the effect of crosslinking, in this case.

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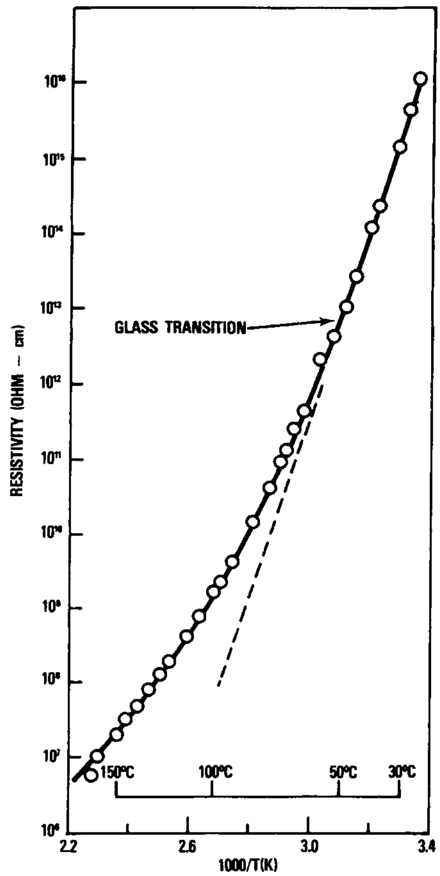

Fig. 1. Resistivity of a polyamide–epoxide copolymer as a function of temperature.

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