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LOW TEMPERATURE MEASUREMENTS OF RESISTIVITY IN LOW-DENSITY POLYETHYLENE

J. Brunson and J.R. Dennison

Abstract

Measurements of resistivity of low density polyethylene (LDPE) have been made using the standard constant voltage method to determine the temperature dependence of resistivity. Where electrons are assumed to serve as the primary charge carriers, their mobility is believed to be dependent on their probability of hopping between trapping sites treated as potential wells. We consider our measurements of this relatively simple polymeric material using temperature-dependant models of conduction mechanisms developed for amorphous solids and semi-conductors.

Introduction

High resistivity insulating polymers are ubiquitous in use, easily tailored to address specific chemical requirements, and endless in their possible applications in new technology. The prevalence of these materials in the design of spacecraft components places special emphasis on the electrical properties of the insulators, which are critical for anticipating and preventing potentially damaging spacecraft charging phenomena [1,2].

Electrical properties of insulators are significantly different from the electrical properties of conductors and semi-conductors, both experimentally and in the fundamental understanding of their behavior. The conductivity of the material, and its inverse, the resistivity $\rho = 1/\sigma$, is the relevant property for determining mobility of charge carriers and dissipation rate of accumulated charge within the material. The most promising theoretical possibilities for explaining electrical behavior in insulating polymers are concepts and hopping conductivity models that have proven successful in application to semi-conductors and amorphous solids [3,4]. These theories are well tested for semi-conductors, but remain largely unverified for insulators [5].

Low-density polyethylene is a good candidate for attempts to verify these models. It is one the most common and versatile polymers; high uniformity and high purity samples can easily be obtained for testing. Much is known about LDPE and it is relatively well characterized. LDPE is also semi-crystalline, which increases the likelihood that hopping conductivity is an appropriate model for LDPE.

Fundamental assumptions of conductivity models applied to semi-conductors include the identification of electrons or holes as the primary charge carriers. Their motion through the material is governed by availability of localized states treated as potential wells in the lattice, as illustrated in Fig. 1.

Fig. 1. Representation of carrier motion by way of hopping between potential wells. ΔH and a correspond to well depth or traped site binding energy and well separation, respectively [4,5].

The electron, or hole, moves through the material by hopping between localized states or traps. Energy is required to release the carrier from the trap and the conductivity is proportional to the probability that hopping will occur [3,4]. The resistivity is dependent on carrier mobility, which is influenced by both temperature, T, and applied electric field, E. In general,

$$\rho_{hop}(E,T) = \left[\frac{2 \cdot n(T) \cdot \nu \cdot a \cdot e}{E}\right]^{-1} \exp\left[\frac{\Delta H}{k_B \cdot T}\right] \cosh\left[\frac{\varepsilon \cdot E \cdot a}{2 \cdot k_B \cdot T}\right] \quad (1)$$

which contains terms accounting for both thermally activated conductivity and for electric field enhanced conductivity. Separation of these terms allows each behavior to be tested independently. Other parameters that appear in Equation 1 are the frequency of hops, ν, the dielectric constant, ε, the density of charge carriers, n(T), the well depth, ΔH, and well separation, a. In reality, the finite thickness of the sample introduces multiple layers of trapping sites and can significantly change the density of charge carriers, n(T). It is assumed, for simplicity, that shallow traps provide the bulk conductivity while deep traps do not contribute to charge mobility [4,5].

Probing a complex behavior such as the response of an amorphous material to a temperature change or an electric field requires multiple approaches. Polymers such as LDPE present further challenges due to the complexity of their morphology and structure. The polymer chains do not lend themselves to the simplifications of a lattice construct, and polar groups attached to the chains have significant influence on carrier mobility. These polar groups can also contribute to an overall material polarization that influences the internal electric field felt by the carriers [5]. Polarization in the presence of an applied field produces a transitory polarization current that dominates the measured initial current [15]. Inclusion of the initial polarization current results in a calculated resistivity much lower than the true resistivity of the material needed to verify conduction mechanisms. The electric field, E, dependence of resistivity in LDPE was reported in a previous paper [16].

Testing the temperature dependence of resistivity is vital because hopping conductivity is fundamentally a phonon-assisted mechanism [12]. The charge carriers are unable to transition between localized, bound states without phonon interaction. This leads to inherent temperature dependence in the probability of hopping and in the measured current due to hopping charge carriers. The temperature dependence of resistivity in LDPE was determined as follows.

Experiment

Resistivity of an insulator can be found using the thin film capacitor approximation [9,10]. The most common method is the constant voltage method [11]. A thin film sample is placed between two metal electrodes, a voltage is applied across the sample, and the leakage current is measured. For highly resistive materials, this involves measurements of extremely small currents and the presence of a polarization field within the material influences the relevant time scale of the measurements. In the case of LDPE, a measurement time of one hour is sufficient to ensure that all polarization currents have ceased to contribute to the leakage current.

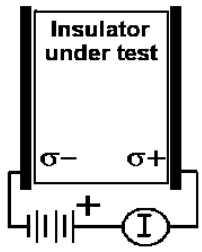

Fig. 3. Diagram of constant voltage method as an approximation of a thin film capacitor.

Two 25 μm LDPE samples of identical origin were chemically cleaned with methanol and baked at 65(±1)^{o}C under vacuum of approximately 10^{-5} torr for two days to eliminate water that may have been absorbed during processing and handling. The samples were placed in a constant voltage apparatus inside a vacuum chamber maintained at a pressure on the order of 10^{-5} torr. Using an applied voltage of 140(±1) V, which corresponds to approximately 10% of breakdown voltage, the leakage current was measured for an hour at room temperature, 23(±2)^{o}C, (see Fig.4). The resistivity was then calculated using the long-time, steady state limit and found to be 8.17(±0.08) x 10^{17} Ω-cm.

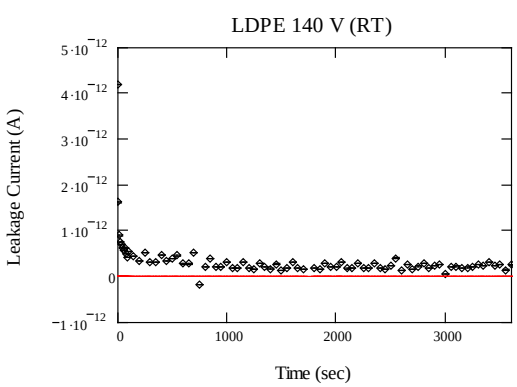

Fig. 4. Leakage current through 25 μm LDPE sample at 140(±1) V at 23(±2)^{o}C for one hour. The straight line below the data points is the base noise level of the system while all equipment is powered on but no measurements are being taken.

The chamber and samples were then cooled to -40.0(±0.5)^{o}C using liquid nitrogen and the sample that had not previously been used for the room temperature measurement was placed under an applied voltage of 140(±1) V. The lower bound of experimental temperature was chosen to avoid possible onset of ill-defined behavior due to approaching the glass transition temperature, which ranges from -60°C to -125°C in the literature, depending on the reporting source and manufacturer. Measured leakage current through such a phase transition would be unlikely to be due to hopping conductivity alone and is therefore be beyond the scope of this research.

Two samples were used in this experiment rather than one because it has been shown in previous work [13] that charging history and repeated applied voltages have an effect on the resistivity of the sample. The samples were obtained from the same manufacturer and were cut from the same allotment of material, ensuring as near to identical composition, properties, and environmental history as possible.

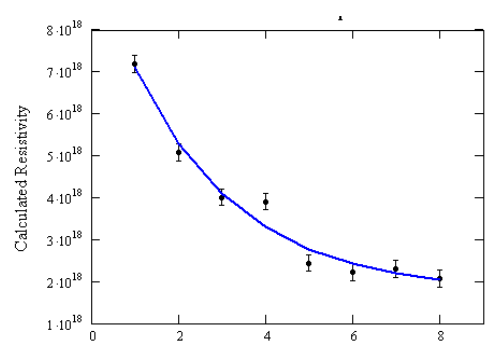

Fig. 5. Repeated measurements of resistivity of LDPE under the same applied voltage.

Once temperature equilibrium was reached, the chamber and samples were then allowed to return to room

temperature without the aid of internal or external heating and the leakage current was monitored throughout the warming period. A typical warming period without intervening heating lasts approximately twenty hours, corresponding to an average warming rate of approximately 3°C per hour. The collected data are shown in Fig. 6.

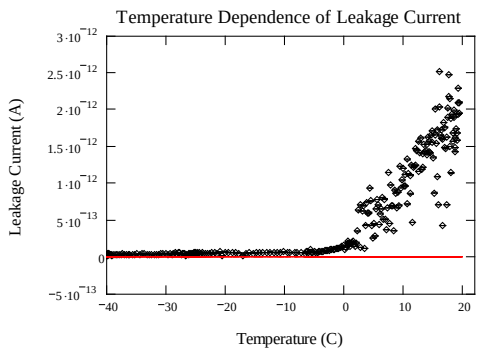

Fig. 6. Leakage current through 25 μm LDPE sample from -40.0(±0.5)°C to 20(±0.5)°C. The straight line seen below the collected data is the base noise level of the system while all equipment is powered on but no measurements are being taken.

At low temperatures, the current approaches the instrumentation limit of the Keithley 616 picoammeter and the leakage current is barely distinguishable from the base noise level of the system. The base noise level alone corresponds to a resistivity of 6 (± 1) x 10^{19} Ω-cm. The current increases slowly until the temperature reaches approximately -5°C where it rises suddenly, corresponding to an order of magnitude drop in resistivity as T increased to room temperature.

Comparing the calculated resistivity at regular intervals throughout the temperature range with the calculated resistivity at room temperature reveals that the resistivity is indeed dependent on temperature, as seen in Fig. 7.

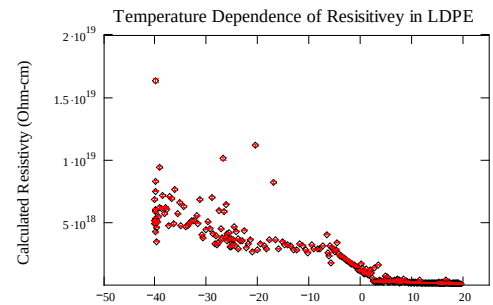

Fig. 7. Calculated resistivity as a function of temperature in 25 μm LDPE from -40.0(±0.5)°C to 20(±0.5)°C.

In the low temperature range, the resistivity is significantly increased. This behavior theoretically corresponds to lower carrier mobility and a decrease in hopping conductivity probability.

Data Analysis

To confirm hopping conductivity as an appropriate model for charge transport in LDPE, the resistivity must be inversely proportional to temperature in a range where it is energetically favorable for an electron to hop to nearest neighbor states. The temperature dependence must transition to a $T^{-1/4}$ in the low temperature limit, corresponding to the theoretical onset of variable-range hopping where it is energetically favorable for the electron to hop to lower energy states beyond the nearest neighbor states [14]. Using Eq. 1, the resistivity at should be proportional to an exponential with powers of T^{-1} and $T^{-1/4,}$ according to the temperature range.

Plotting the natural log of the calculated resistivity as a function of temperature, two regions with distinct behavior are observed. Below -(±0.5)°C, the resistivity follows the $T^{-1/4}$ dependence expected for variable range hopping (shown in red in Fig. 8). Above -5(±0.5)°C, the resistivity is

inversely proportional to T, (shown in blue in Fig. 8).

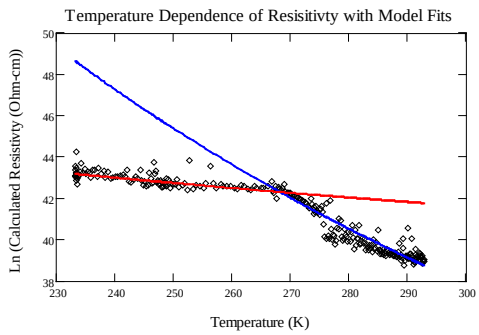

Fig. 8. Temperature dependence of the natural log of calculated resistivity in 25 μm LDPE with linear regression fits corresponding to regions of $T^{-1/4}$ (shown in red) and 1/T (shown in blue) dependence. Temperatures are in Kelvin.

The point at which the low temperature behavior transitions must correspond to a physical, morphological phase transition within the material, but it is unknown what that transition is.

Unexpectedly, the calculated resistivity upon returning to room temperature is an order of magnitude lower than the initial resistivity calculated using the first sample at room temperature. Since both samples came from the same manufacturer and source sheet of LDPE, it is more likely that the process of cooling and heating influenced the resistivity. It is also possible that the duration of exposure to the electric field is responsible for the discrepancy.

Conclusions

The resistivity of LDPE shows two regimes of temperature dependence. At low range temperatures, a $T^{-1/4}$ dependence is observed that could suggest variable range hopping as a mechanism. At higher temperatures, the resistivity is inversely proportional to temperature; behavior that is also consistent with hopping conductivity as a model of phonon-assisted charge transport. However, more research is needed to eliminate mechanisms that behave similarly and result in similar temperature dependence.

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