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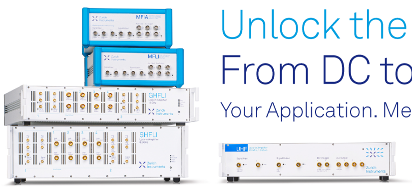

Electrical Transport Study on Low-Density Polyethelene/Carbon Black Composites at Low Temperatures

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Abstract. Carbon black/polymer composites have shown very pronounced interests in the last years due to their wide variety of applications. Many of these applications depend on enhancing the electrical conductivity of the polymer by mixing some ratios from the carbon black as filler to the polymer matrix. This paper reports on the DC electrical conductivity of low-density Polyethelene/Carbon black composites of various CB filler concentrations at low temperatures. The composites exhibit a negative temperature coefficient of resistivity (TCR) and an increase in the electrical conductivity with decreasing temperature down to 25 K. A pronounced enhancement in the electrical conductivity with increasing the CB concentration has been observed. This enhancement in the conductivity is interpreted through the percolation theory. The observed features of the electrical conductivity for LDPE/CB composites are analyzed according to the Mott hopping mechanism.

INTRODUCTION

Composite materials based on conducting additive elements such as carbon black (CB), carbon fibers and metallic powders (Ni, Al, Cu, etc.) within the insulating polymer matrices have been subject of interest of both theoretical and experimental research over the last decades, due to their wide range of applications [1-2]. At the same time, these materials hold potential interests due to their interesting fundamental physical properties [3]. These composites are typical representatives of peroclative systems. The mixture between the insulating polymers with a suitable types and concentrations of filler conducting particles leads to compounds with conductivity varied in a wide range, and at the percolation threshold the conductivity shows a pronounced change from the conductivity of the insulating polymer to the conductivity of the additive conducting element [4].

In the recent years, polymer/CB composites have been subject of extensive research, due to their high conductivity value, high stable structure and low cost production value [5]. The dielectric and electric behavior of the polymer/CB above room temperature have been reported in literature extensively [6-8]. The general consensus for these studies is that Polymer/CB composites exhibit a ‘positive temperature coefficient’ (PTC) of resistivity, and behave electrically above room temperature as semiconducting materials. There are few studies in the literature concerning the electrical behavior of conductive polymer composites in the low temperature range [9-10]. The electrical properties of CB/polymer were investigated at temperature below room temperature down to 120 K [10]. It was observed that the electrical conduction behavior is similar to semiconducting materials, and the conductivity change drastically at a percolation threshold of the CB concentration. This observation motivated our interest to carry out this experiment at lower temperatures.

This paper presents detailed data analyses on the DC electrical conductivity of the low-density Polyethelene/Carbon black composites in the temperature range 285 K – 25 K. The conductive polymer composite

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consists of Polyethelene as a matrix polymer and a carbon black as conductive component with different concentrations. The results are discussed and explained within the percolation theory and Mott's variable range hopping (VRH) process.

EXPERIMENTAL WORK

Materials: the investigated materials in this research are low-density polyethelene/carbon black composites. The concentrations of the CB in the composite as a filler are 10, 15, and 20 wt.%. The composite samples were prepared in Italy (ICTP-Napoli). The preparation of the CB composites were reported elsewhere [11]. Disks of 1 cm in diameter and 0.13 cm in thickness were cut from the molded plates of the composite for the electrical measurements.

Electrical measurements: the temperature-dependent measurements of the volume dc resistivity were performed using standard four probe method with a typical current of less than 10 mA to avoid sample heating [12]. The current and the voltage through the disk-shape samples were measured by sensitive Keithley and Philips digital multimeters. The disk samples were mounted on the sample holder of the Janis-made vacuum jacket of the cryogenic helium closed-cycle refrigerator (CCR). The current and the potential change were measured continuously as a function of temperature.

RESULTS AND DISCUSSION

The voltage versus current plotting at different temperatures for 10 wt. % CB composite is shown in figure 1. The composite displays a linear relation indicating an ohmic behavior. This ohmicity have been observed also in samples with 15 and 20 wt. % CB.

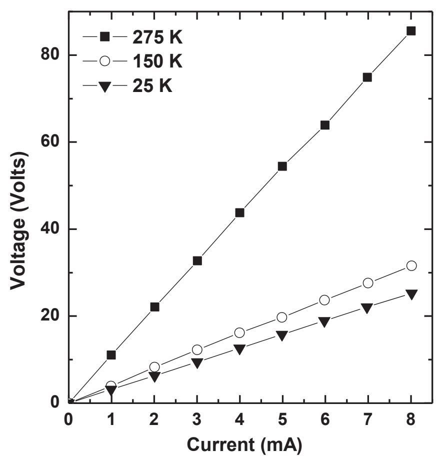

FIGURE 1. Voltage versus Current for the 10 wt.% CB composites at different temperatures.

The volume DC electric conductivity (inverse the resistivity ρ) was determined from the relation [12]

$$\sigma = \frac{1}{\rho} = \frac{d}{RA} \tag{1}$$

where A is the area of the disk-shaped sample and d is the average thickness of the sample. Figure 2 shows the temperature dependence of the DC resistivity (and conductivity in the insert) over the temperature range 25 – 285 K for the composites with three different carbon black concentrations. It can be seen that all composites shows a semiconducting behavior of resistivity, which is typical for conducting polymer composites [13]. The temperature coefficient of resistivity (TCR) was calculated using the formula [12]

$$TCR = \left(\frac{1}{\rho(T_1)}\right)\left(\frac{\Delta\rho}{\Delta T}\right) \tag{2}$$

where $\Delta\rho = \rho(T_2) - \rho(T_1)$, and $\Delta T = T_2 - T_1$. Table 1 shows the calculated values of the TCR for 10 wt. % CB. The composite exhibits a negative TCR from 25 K to 255 K, and above 255 K, it shows a positive TCR. The positive value for the TCR in the higher temperature range is found also in the composites of 15 and 20 wt. % CB. This results is attributed to thermal expansion occurring in the composite when the temperature increases, thus causing the gradual breakup of conducting pathways of conductive filler particles through the polymer matrix, thereby increasing the resistivity of the composite with temperature.

The insert of figure 2 shows that the electrical conductivity increases with increasing the concentration of the CB in the composite. This observed increase is due to increasing the conductive paths produced by the CB particles within the polymer matrix. The sudden change in the conductivity between 15 and 20 wt. % CB is attributed to the percolation behavior, whereas the CB concentration increased the conductivity increases slowly until a three-dimensional network is established inside the composite and a sudden transition in the conductivity from low level (conductivity of the insulating polymer) to high level (conductivity of the conducting component) occurs at a certain filler concentration [14].

TABLE 1. Calculated values of the temperature coefficient of resistivity (TCR) for 10 wt. % CB.

Temperature range	TCR
25-60	-2.60×10^{-2}
60-140	-5.03×10^{-3}
140-190	-3.83×10^{-3}
190-235	-1.80×10^{-3}
235-255	-1.07×10^{-3}
255-285	1.20×10^{-3}

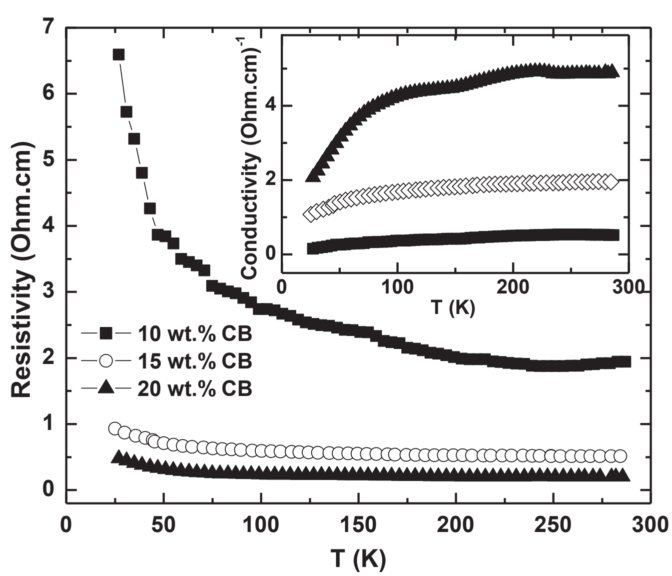

FIGURE 2. DC electrical resistivity versus temperature for Polyethelene/Carbon black composites of various CB filler concentrations.

The electric transport at low temperatures was introduced by Mott's theory [15]. According to Mott, the behavior of the dc conductivity at low temperatures is due to a variable range hopping (VRH) of the charge carriers between the neighboring sites, which can be expressed as [16]:

$$\sigma = \frac{\sigma_0}{T^{1/2}} \exp\left[-\left(\frac{T_0}{T}\right)^{1/4}\right] \tag{3}$$

where σ_0 is the material constant at infinite temperature, and T_0 is Mott's characteristic temperature. The plot of $\ln(\sigma \times T^{1/2})$ versus $T^{-1/4}$ based on eq. (3) for the composites is shown in figure 3. The figure shows that the plots based on this model were linear in the whole temperature range (20–285 K) (shown as a bold fitting line). The material constant and Mott's characteristic temperature obtained from the slope and intersect of the linear fitting for the three concentrations are given in table 2. The variation of the Mott's parameters with the CB concentrations is a result of change of density of the charge carriers in the composite, which reduce the residual resistivity at finite temperature.

TABLE 2. Values of $\boldsymbol{T_0}$ and $\boldsymbol{\sigma_0}$ for the CB composites.

wt.% of CB	T_0 [K]	σ_o [Ω^{-1}cm^{-1}K$^{1/2}$]
10	2.022×10^4	157.53
15	6.640×10^3	295.58
20	1.1034×10^4	1039.9

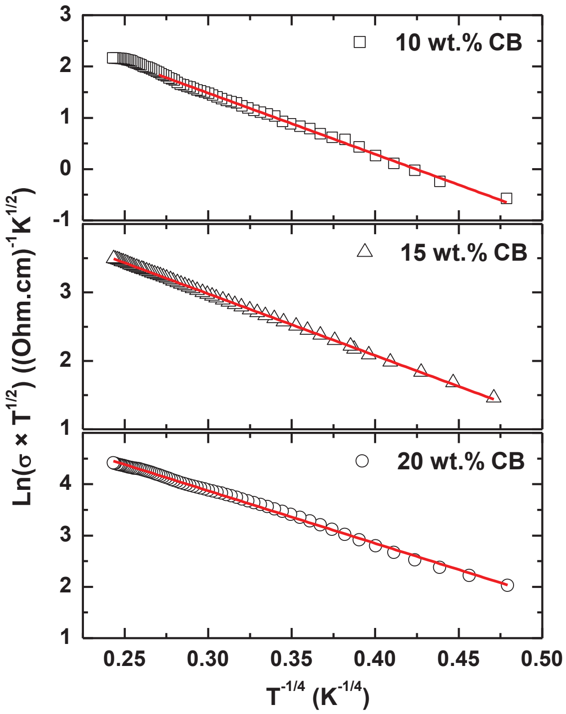

FIGURE 3. $\ln(\sigma \times T^{1/2})$ versus $T^{-1/4}$ for Polyethelene/Carbon black composites of various CB filler concentrations.The bold straight line is a linear fitting line (see the text).

The density of states at the Fermi level and the decay constant are expressed as [16]

$$N(E_F) = 5.55416 \times 10^{10} {\sigma_0}^3 T_0^{1/2} \text{eV}^{-1}\text{cm}^{-3} \quad (4)$$

$$\alpha = 64.303 \sigma_0 T_0^{1/2} \text{cm}^{-3} \quad (5)$$

The values of $N(E_F)$ and α for each composite sample are given in table 3. The density of states at Fermi level and the decay constant are clearly influenced with the CB concentration in the composite.

TABLE 3. Calculated values of the density of states at Fermi level and the decay constant.

wt. % of CB	$N(E_F)[\mathrm{eV^{-1}cm^{-3}}]$	$\alpha[\mathrm{cm^{-1}}]$
10	8.87×10^{25}	4.139×10^{12}
15	6.32×10^{25}	8.379×10^{11}
20	7.61×10^{27}	8.147×10^{12}

The hopping distance between the neighboring sites R and the mean hopping energy W are given by the expressions [16]:

$$R = \left(\frac{9}{8\pi\alpha k_B T N(E_F)}\right)^{1/4} \tag{6}$$

$$W = \left(\frac{3}{4\pi R^3 N(E_F)}\right) \tag{7}$$

The calculated values for $\boldsymbol{R}$ and $\boldsymbol{W}$ are given in table 4. According the Mott's hopping model, the necessary conditions of the validity of the model are $\boldsymbol{\alpha R \gg 1}$ and $\boldsymbol{W \gg k_B T}$. It appears from the results shown in table 4 that it fulfil these conditions where the calculated values for $\boldsymbol{\alpha R}$ are much larger than one and the values for the mean hopping energy are much larger than $\boldsymbol{k_B T}$ value ($\boldsymbol{k_B T = 8.62 \times 10^{-3}}$ for $\boldsymbol{T = 100}$ K).

TABLE 4. Calculated values of the hopping distance and the mean hopping energy at 100 K.

wt. % of CB	$R[\mathrm{cm}]$	$W[\mathrm{eV}]$	αR
10	8.87×10^{25}	4.139×10^{12}	2401.75
15	6.32×10^{25}	8.379×10^{11}	788.6
20	7.61×10^{27}	8.147×10^{12}	1310.85

SUMMARY

The resistance of the CB/LDPE composites exhibit an ohmic behavior, and its resistivity decreases with increasing the carbon black concentration. The electrical resistivity of the composites showed a semiconducting behavior at low temperatures with a negative value of TCR. The observed increase of the electrical conductivity with increasing the CB concentration was attributed to the percolation behavior. At low temperatures, the hopping conductivity based on Mott's variable range hopping (VRH) model was considered as the dominating charge transport mechanism.

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