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Epoxy Resin/Plasticized Carbon Black Composites. Part I. Electrical and Thermal Properties and Their Applications

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The development of conductive polymer composites remains an important endeavor in light of growing energy concerns. A conducting polymer composite in the presence of plasticized carbon black (CB) and epoxy resin has been developed. Room temperature electrical conductivity, mobility carriers (μ), and number of charges (*N*) increase by increasing CB content. Electrical conductivity–temperature dependence of the composite was investigated and negative temperature coefficient of conductivity (NTC) behavior of the composites was revealed. The mechanism of the NTC effect in materials is related to the thermal expansion of the epoxy matrix and barriers height energy. The current–voltage behavior of epoxy/CB composites shows a switching effect and the mechanism of negative resistance is interpreted in details. Thermal conductivity (λ) increases with increasing filler content and the experimental data was compared with a theoretical model based on energy balance equation. POLYM. COMPOS., 00:000–000, 2007. © 2007 Society of Plastics Engineers

INTRODUCTION

Electrically conductive polymer composite (CPCs) is obtained when conductive particles such as metal powder, carbon black (CB), and carbon fiber etc. are implanted into an insulating polymer matrix [1–4]. CB is the most important reinforcing filler in the rubber industry because of its abundant source, low density, permanzent conductivity, and low cost [5–7]. To obtain high electrical conductivities for specific applications, high filler concentrations (typically ≥50 vol%) are needed in the composites. For CB-filled polymer composites, it is desirable to reduce the percolation threshold as low as possible. High CB volume fraction increases the melt viscosity, decreases the impact resistance, contaminates in clean room environment, increases the final product cost, and the mechanical properties of the composites are lost [8–12]. Recently, two promising approaches to decrease the percolation threshold in the CPCs were reported. The first is based on conductive filler filled immiscible polymer blends [13–15]. The second one is the use of nanoconducting filler loaded polymer matrix [16–18]. However, several factors determine the conducting filler distribution in the polymer matrix, such as polarity, polymer crystallinity, and melt viscosity, flexibility of polymer chains, processing, and structure of the filler. Generally, CPCs with low percolation and enhanced conductivity have found application in negative temperature coefficient (NTC) of conductivity material [19], electrical self heating [20], liquids or chemical vapor sensors [21], switching current [22], antistatic materials for electromagnetic interference shielding of electronic devices [23], corrosion inhibition, etc. [24]. Self-regulated heating ability can be obtained for CPCs with sharp NTC effect. The study of electrical and thermal conductivities of the CPCs is essential for heating devices design and allow adjusting CPCs properties and thus a wider range of application can be established [25]. With the above considerations in mind, this study was devoted to the fabrication and characterization of new composites with high electrical conductivities with low filler content for electronic application from plasticized CB implanted into an epoxy resin matrix. In this part, we

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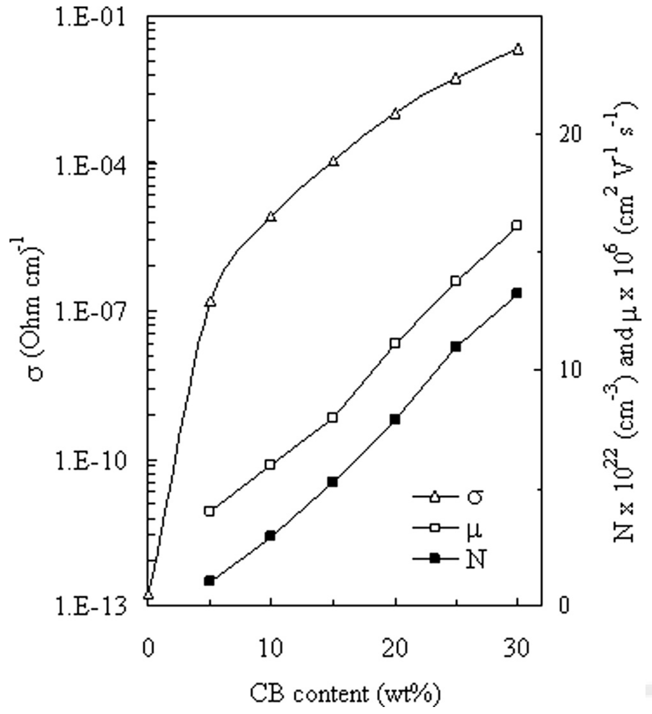

FIG. 1. Electrical conductivity at room temperature (σ), mobility carriers (μ), and density of charge carriers (N) of the epoxy resin/CB composites as a function of the volume fraction of the CB.

report the electrical and thermal properties of the composites. Their potential applicability for NTC thermistors as well as switching voltage devices, were tested for the first time. The thermal conductivity of composites was experimentally measured and compared with theoretical modeling based on energy balance equation.

EXPERIMENTAL

The matrix used in this study is an epoxy resin polymer based on an Epikot resin 828 and an aromatic hardener 128 made by Kayo, Tokyo, Japan. The standard curing mixture contains 75 wt% epoxy resin and 25 wt% hardener; both components are delivered in the liquid state. The conductive filler is a high abrasion furnace black (CB) with a primary particle size of about 3 μm. A glycerol is used as a plasticizer and is mixed with the epoxy resin in 8% weight quantity to reduce the viscosity of the resin and facilitate the mixing of high volume fraction of particles in the resin. The samples containing 5, 10, 15, 20, 25, and 30 parts of CB per hundred parts of CB per hundred parts of resin (composites are donated as CB5, CB10, CB15, CB20, CB25, and CB30, respectively), were prepared using the following procedures: The epoxy resin, hardener, glycerol, and CB were centrifuge mixed for 1 min at 5,000 rpm and then further sonicates for an additional 30 min at room temperature. After that the composite particles were transferred to a preheated compression Teflon mold, and then hot-pressed at 130°C under 200 KN/m^2 for 1 h to obtain composite sheets with $2 \times 1 \times 0.3$ mm^3. A JEOL-JSM 5800 scanning electron microscope (SEM) was used for the overall microstructural analysis. The specific electrical conductivity for the composites was performed by measuring the current through the sheet sample under a steady constant voltage using a digital Keithley 642 electrometer. The two sides of the samples were bended with Cu-rod during curing process to reduce the contact resistance. A Shimadzu differential scanning DSC-50 calorimeter was used to measure the thermal conductivity (λ) of the sample with a heating rate 10°C/min in an inert nitrogen atmosphere.

RESULTS AND DISCUSSION

Effect of CB on the Percolation Threshold

The electrical conductivity (σ) at room temperature of the epoxy resin/CB composites as a function of the volume fraction of the CB is shown in Fig. 1. F1 It is clear that the volume conductivity increases with increasing the concentration of CB in the composite. At low CB loadings, a large change in volume conductivity can be observed which is regulated by interparticle electron transfer because the gap distances among CB particles are small. As expected, with increasing CB content in the composites, a decrease in gap distance is observed as confirmed by SEM analysis of CB30 sample in Fig. 2. F2 This is reflected by an increase in cross linking density, which increase the conductivity in the composites. Furthermore, increasing the CB content makes CB particles more crowded and linked, leading to a rapid increase of bulk conductivity. This indicates that the conducting particles come into contact with each other or closed up enough to allow the carriers mobility hop by tunneling, thus forming continuous conducting mesh [24]. A feature of interest in

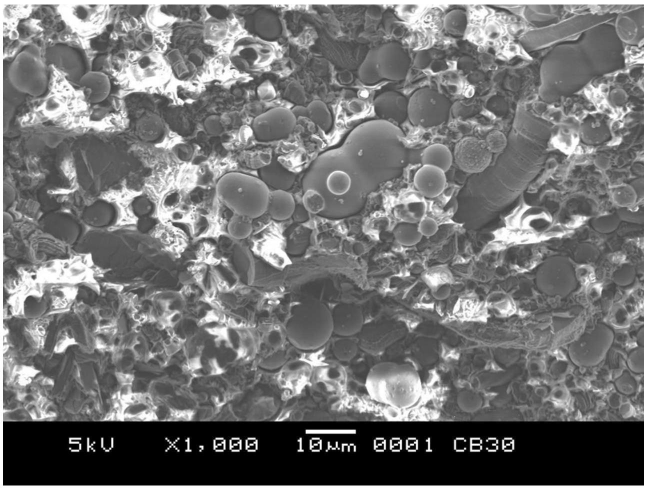

FIG. 2. Scanning electron micrographs of epoxy/CB composites containing 30 wt% CB.

this figure is the minimum CB volume fraction that yields to a measurable conductivity, which is designated as the critical concentration. When the CB volume fraction reaches 9 wt%, the conductivity of the composite is found to be higher than that of the green epoxy resin, by eight orders of magnitude. The value of the critical concentration is quite low compared with the theoretically predicted value (about 17 wt%) for the CB embedded in insulating polymer matrix [25]. The lower percolation threshold value can be attributed to higher degrees of intermixing. The intermixing increases the concentration of CB in the interfacial regions or builds up the direct contact between CB particles, resulting in a lower percolation threshold. To support these findings, the mobility carriers (μ) were calculated from the following formula [9, 26]:

$$\mu = \left(\frac{\upsilon_0 e g^2}{KT}\right) \tag{1}$$

where υ_0 is the optical phonon frequency (generally $\upsilon_0 \approx 10^{-13}$ s, e is the electron charge, K is the Boltzmann constant).

The density of charge carriers (N) is given as [27]:

$$N = \frac{\sigma}{e\mu} \tag{2}$$

The variation of μ and N with respect to CB content of epoxy/CB composites are plotted in Fig. 1. It can be seen that both μ and N increase with increasing CB content within composites. This is attributed to a high degree of bridging among CB particles and intermixing with increasing CB loading level in the composites.

Temperature Dependence of the Electrical Conductivity

F3

The temperature dependent conductivity measured in the temperature range 20–140°C for epoxy/CB composites is plotted in Fig. 3a. It is found that at relatively low temperatures, the conductivity slightly decreases. At higher temperatures the conductivity decreases slightly then decreases sharply at certain temperature (namely percolation temperature), which depends on CB content in the composites. This can be explained as follows: at relatively higher temperatures, the epoxy expands more rapidly than the CB resulting in widening the gap among conductive paths. This expansion decreases the dimensions of conductive phases, resulting in a decrease in the conductivity of the composites. Particularity, thin CB particles might be broken up at several points by the expansion of epoxy particles. The decrease in the number of the CB particles decreases the conductivity significantly. At the same time the thermal fluctuations increase the potential barrier which results in less tunneling current, consequently the conductivity decreases. It is interesting to note that the percolation temperature shifts to a higher temperature with increasing CB content. This indicates that the plasticized CB improves the thermal stability of the composites as confirmed later in this article.

However, the NTC, thermistor constant (B), and activation energy, are the most important characteristics of technical interest for NTC thermistors materials. NTC is given by the following equation [2, 28]:

$$\mathrm{NTC} = \frac{\sigma_{130} - \sigma_{20}}{\sigma_{20}(T_{130} - T_{20})} \tag{3}$$

where subscripts 20 and 130 of σ indicate temperature range of measurements (i.e. 20 and 130°C).

The thermistor constant (B) is given by the following expression [3, 29]:

$$B = \frac{T_1 T_2}{T_1 - T_2} \ln\frac{\sigma_2}{\sigma_1} \tag{4}$$

where σ_1 and σ_2 being the conductivity at temperature T_1 and T_2, respectively.

The computed values of NTC and B as a function of CB volume content are plotted in Fig. 3b. It is clear that the NTC and B decrease with increasing CB content. This is attributed to the interfacial adhesion that increases with increasing the volume fraction of CB in composites. It is worthy to note that the thermistor constant and NTC values of the CB/epoxy composites are in the range of industrial NTC thermistors materials [30].

Conduction Mechanism of Conductivity

The conduction mechanism of conductivity can be understood through the evaluation of the activation and hopping energies. The activation energy (E_a) and hopping energy (E_h) have been calculated using the following relations [31]:

$$\sigma = \sigma_0 e^{\frac{E_a}{KT}} \tag{5}$$

$$\sigma\sqrt{T} = \sigma_1 \exp^{\frac{E_h}{KT}} \tag{6}$$

where σ_0 and σ_1 are the conductivity extrapolated to the temperature $= \infty$.

The computed values of E_a and E_h plotted vs. CB content of composites are shown in Fig. 3b. It can be observed that by increasing CB content, both values of E_a and E_h decrease, because samples having high conductivity have low activation and hopping energies and vice versa. Lower activation and hopping energies implies increased chain-to-chain contact area for the electron to flow, and therefore, a lower barrier height. It is worthy to note that E_a and E_h have different values. This indicates that the conduction mechanism of conductivity is controlled by hopping mechanism [1, 2].

To determine the conducting parameters, the temperature dependence of conductivity (linear portion) is fitted using the following equations [30]:

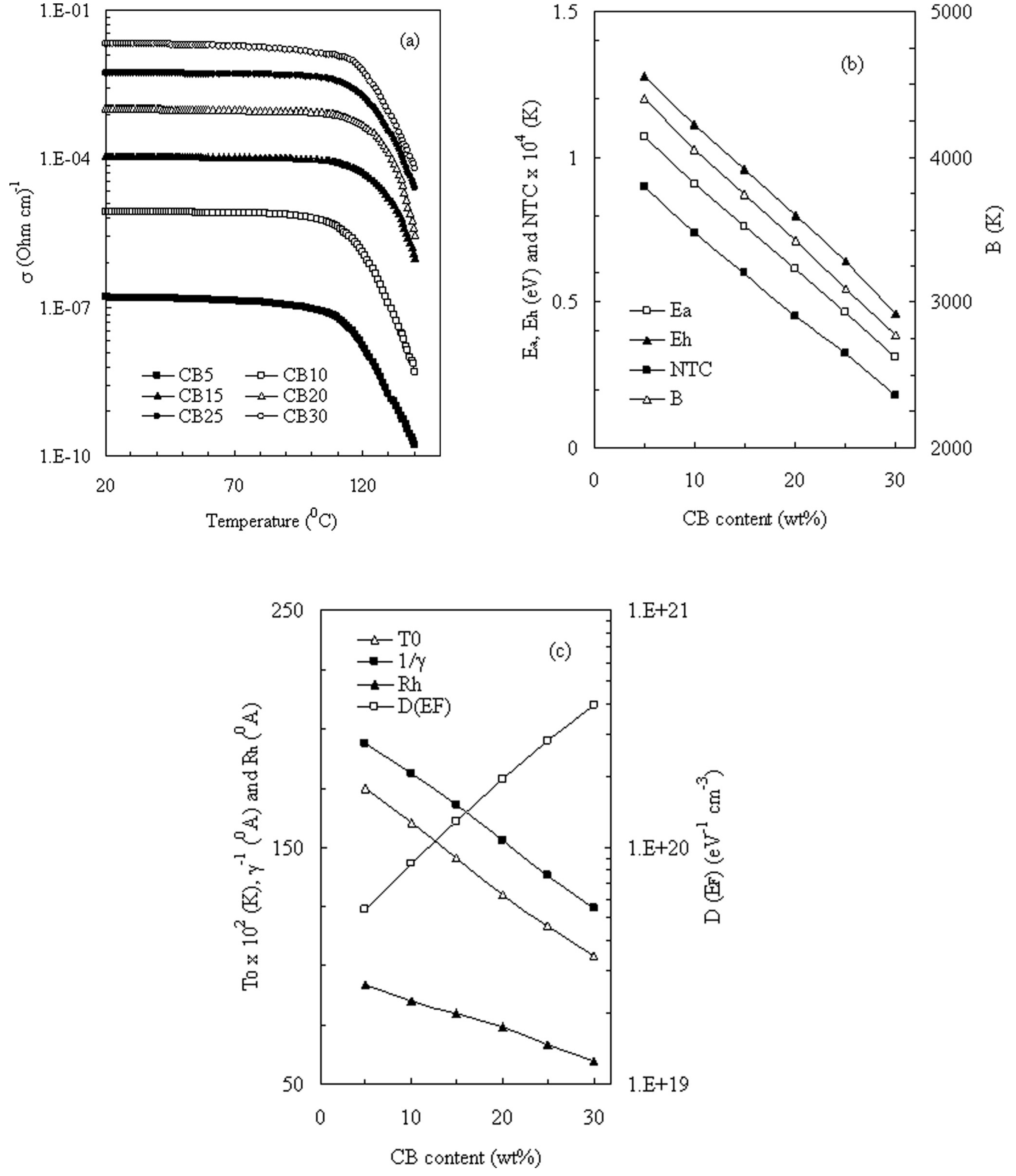

FIG. 3. (a) Temperature dependent conductivity measured in the temperature range 20–140°C for epoxy/CB composites. (b) Computed values of negative temperature coefficient of conductivity (NTC), thermistor constant (B), activation energy (E_a), and hopping energy (E_h) as a function of CB volume content. (3) Conducting parameters, temperature (T_0), charge localization length (γ^{-1}), average hopping distance (R_h), and $D(E_F)$ is the density of states at Fermi level as function of carbon content.

$$\sigma(T) = \sigma_0 \exp - \left(\frac{T_0}{T}\right)^{\frac{1}{2}} \tag{7}$$

$$T_0 = \frac{8\gamma}{D(E_\mathrm{F})ZK} \tag{8}$$

$$R_\mathrm{h} = \gamma^{-1}\sqrt{\left(\frac{T_0}{16T}\right)} \tag{9}$$

where T_0 is related to the charge localization length (γ^{-1}), Z the number of nearest neighboring chains of about 4, R_h is the average hopping distance, and $D(E_\mathrm{F})$ is the density of states at Fermi level.

The values of T_0 obtained from the linear graph are used to calculate γ^{-1}, $D(E_\mathrm{F})$, and R_h. The calculated values of T_0, γ^{-1}, $D(E_\mathrm{F})$, and R_h as a function of CB content for epoxy/CB composites are plotted in Fig. 3c. It is clear that T_0, γ^{-1}, and R_h decrease with increasing CB content, which is in agreement with the decrease of conductivity of CB/epoxy composites. On the other hand, it is seen that the density of states at Fermi level increases with increasing CB content in composites. This means that the CB acts as a carrier's reservoir within resin matrix. The values of the density of states at the Fermi level of the composites are reasonable for localized states [22].

Applicability of Composites as Switching Voltage and Current

Switching voltage is one of the important electrical engineering factors to judge the quality of NTC composites with high conductivity. The current–voltage characteristics for epoxy/CB composites are plotted in Fig. 4. It is seen that at low applied voltage, the current increases linearly with applied voltage. This reflects that the conduction is related to the electron tunneling across insulating paths separating adjacent particles. As the applied voltage increases, the cold-emission leads to the production of additional conducting paths, which is responsible for the nonlinear intrinsic conduction. The rate of increase then falls until an ultimate current is reached, beyond which the current falls with increasing voltage (this voltage is namely switching voltage). This negative regime is characterized by apparently random fluctuations in the current [31]. The negative-resistance regime is attributed to the charge storage on the CB particles. The higher the applied voltage was, the more the Joule heat produced (i.e. the bulk temperature of the samples increase). Too much Joule-heat may affect the electrical conductive chains leading to an increase of the distance between CB particles. Furthermore the charge carriers can build up on a particle where there is a high barrier for charge flow in the field direction (i.e. large separation to the next particle in the conduction network that acts as a dead-end [18]. Moreover, the trapped charge will produce an enhanced local field that will create potential barriers in adjacent conduction paths. This result suggests that the epoxy/CB composites can be used as a switching voltage and current in microelectronic devices.

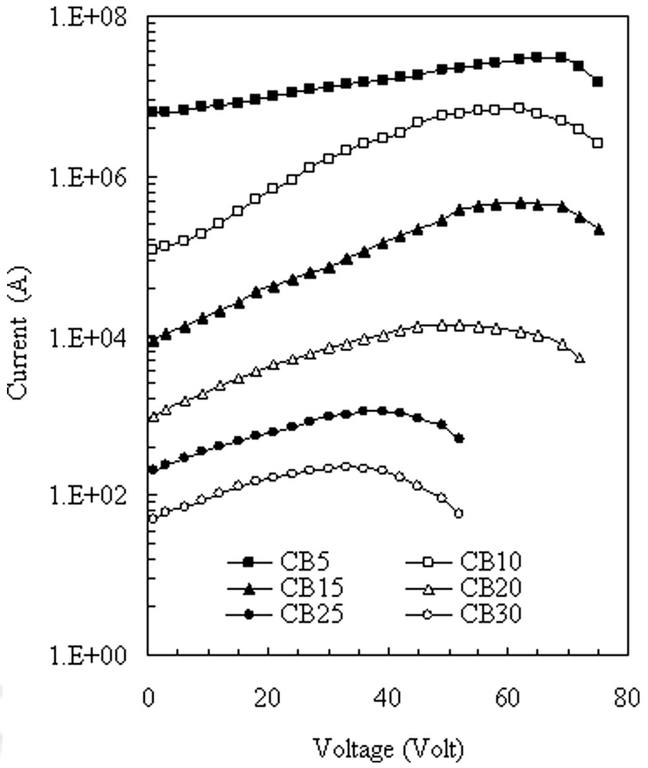

FIG. 4. Current–voltage characteristics for epoxy/CB composites.

Thermal Stability Under Applied Voltage

Figure 5 shows the response of the ultimate temperature on time under certain applied voltage ($V = 30$ V) for CB15 and CB30 samples. It is seen that the samples show Joule heating which causes the bulk temperature to increase with time. The ultimate temperature of the CB30 sample suddenly increases in an applied voltage, and returns immediately to the initial temperature when it is transferred into voltage off at high CB loading level. However, it is observed that CB30 sample gives rise to much higher temperature at the same applied voltage than CB15 sample. This indicates that the response of temperature of CB30 sample to applied voltage was higher than that of CB15 one. This may be due to the higher interface adhesion of CB30 sample. This means that the inclusion of CB particles into resin matrix improves their thermal stability, whereas the initial current decays with time to a steady value.

Modeling for Computed Thermal Conductivity and Heat Transfer

To determine theoretically the thermal conductivity (λ) and the amount of heat transfer by radiation and convec-

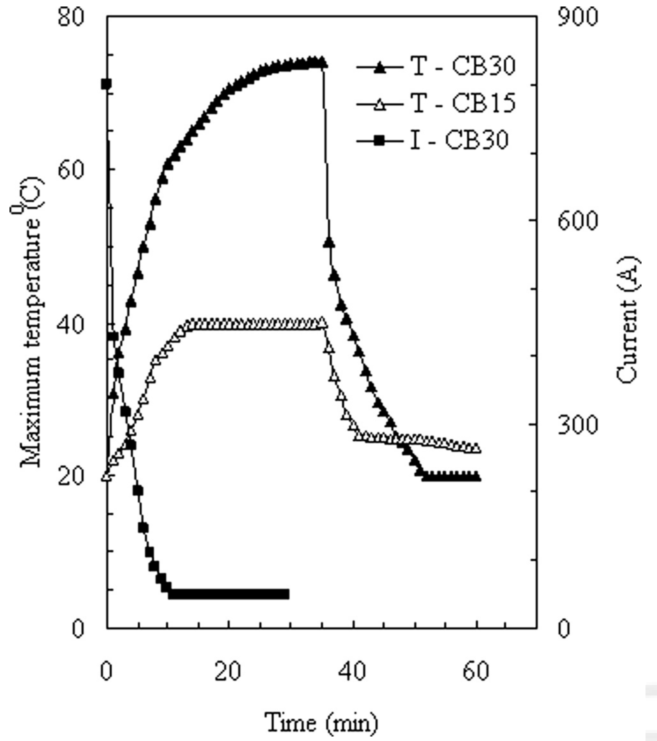

FIG. 5. Response of the ultimate temperature on time under certain applied voltage ($V = 30$ V) for CB15 and CB30 samples.

tion in the composites, energy balance equation has been used [13]. In the steady state zone (see Fig. 5), the power dissipated in the sample is the sum of heat flows exchanged by radiation and convection according to the following equation [13, 14]:

$$VI = S_e(h_c + h_r)(T_b - T_i) \tag{10}$$

where V is the voltage, I is the current, S_e is the total exchange surface, T_b and T_i is the bulk and air temperature, respectively, and h_c and h_r are the heat exchange coefficients by convection and radiation, respectively. h_c is given by the following expression [13]:

$$h_c = \frac{0.55\lambda(G_r P_r)}{L} \tag{11}$$

where L length of the sample, G_r is the Grasshof number, and P_r is the Prandtl number.

h_r is given by the following equation [14]:

$$h_r = 4\alpha\psi T_m^4 \tag{12}$$

where $\alpha = 0.9$, $\psi = 5.67 \times 10^{-8}$ W m^{-2} K^{-4}, and T_m is the maximum temperature.

The thermal conductivity can be computed from the heat equation in the form [13]:

$$\frac{d^2T}{dx^2} + \frac{IV}{V_s\lambda} = 0 \tag{13}$$

Integration of *Eq*. 21 leads to the expression of temperature gradient between the centre and the surface of the sample and thermal conductivity is given by [13, 14]:

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$$\lambda = \frac{IVd}{v_s T} \tag{14}$$

where d and v_s are the thickness and the volume of the sample, respectively.

The experimental and calculated values of λ of the epoxy composites vs. CB content are plotted in Fig. 6. The experimental thermal conductivity values of the composites are quite close to the theoretical values. It is noted that the thermal conductivity increases with increasing CB content, due to the fact that the particles are closer to each other. Furthermore, the increase in thermal conductivity with increasing CB content may be due to the increase of the crystallinity of the composites. The calculated dependence of h_c and h_r of CB volume content for epoxy/CB composites is illustrated in Fig. 6. It is seen that the h_c and h_r increase with increasing CB content. This can be explained that at high CB loading level, the heat transfer from particle to particle is easier because of thinner epoxy layers between the CB particles. At the same time, the microstructure formation having local filler content, results in more dense filler packing inside the epoxy structure and in the improvement of heat transfer conditions [14].

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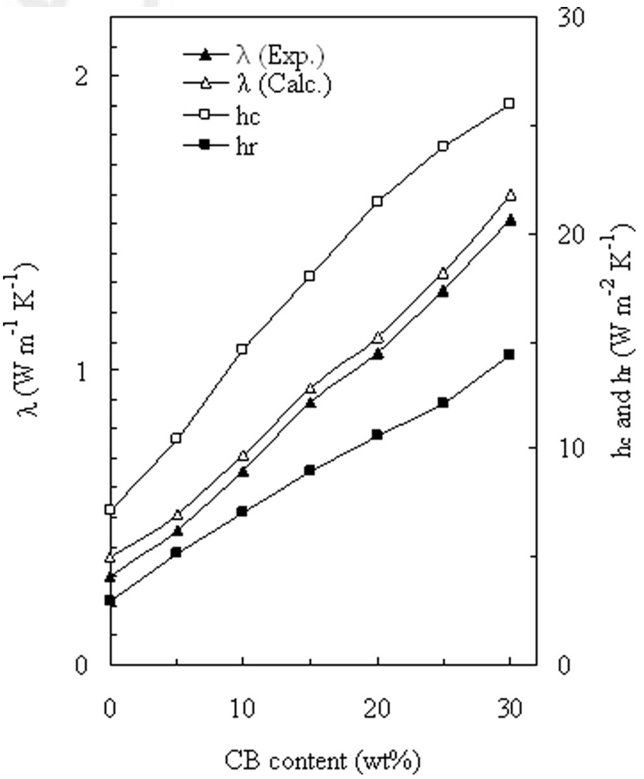

FIG. 6. Experimental λ(Exp) and calculated λ(Calc) thermal conductivity, heat exchange coefficients by convection h_c, and radiation h_r, of the epoxy composites vs. the CB content.

CONCLUSIONS

The results of this research clearly show the following.

1. Room temperature conductivity, mobility carriers, and number of charges of the plasticized CB/epoxy composites increase by increasing CB content.
2. Electrical conductivity temperature dependence shows a NTC thermistor and is noted that the samples having lower resistivity, have lower activation energy and vice versa.
3. The current–voltage behavior shows a switching effect, which makes the proposed composites a potential candidate to a new switching voltage and current in electronic devices.
4. The thermal conductivity increases with increasing CB content in composites and the experimental data are in good agreement with the theoretical model.

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AQ1: Kindly note that the total equations given in this article are 14. Please confirm whether the Eq. 21 given here is justified.

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