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THE INFLUENCE OF TEMPERATURE ON ELECTRICAL CONDUCTIVITY OF SEMI-CONDUCTOR GRAPHITE/EPOXY COMPOSITES

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Abstract. *The wide use of polymer matrix composites requires a greater understanding of their characteristics, especially the electrical conductivity at different conditions. These materials have attractive combination of properties, they can replace with lower cost and greater efficiency metallic components. In this work, the electrical conductivity at different temperatures of an epoxy polymer matrix filled with different weight concentrations of low cost graphite powder was investigated. The goal is to analyze the electrical conductivity at temperatures ranging from 20ºC to 100ºC. Such composites combine low cost and reasonable conductivity. The tests have demonstrated that increasing graphite powder content conductivity also increases but temperature environment contributes to decrease electrical conductivity.*

Keywords: Polymer composites; Electrical properties; Semiconductors

1. INTRODUCTION

Polymer based composite materials are been used for the past decades in automotive, aerospace, construction due to their strength-weight ratio aspect, easy process and low corrosion [1]. In recent years researchers have been investigated conductive polymer composites, which the matrix is insulant, manufactured using electrically conductive fillers such as carbon based, metal powder and aluminum flakes [2,3] due to their excellent multifunctional properties when compared to the pristine polymer used [4–8]. They have been applied in various fields including light emitting diodes, batteries, electromagnetic shielding, antistatic coating or fibers, gas sensor, corrosion protection and activators [9-13]. Graphite is one type of carbon-based filler and its electrical conductivity is 104 S/cm at ambient temperature as high as carbon fibers [14,15]. Due to being layered structure of graphite, its layers are possibly expanded and exfoliated with treatment to form high aspect ratio graphite with thickness in nano-scale. Dispersion, aspect ratio, shape, orientation of conductive fillers influences the electrical and mechanical properties of conductive fillers in insulating polymer matrices. In most cases, relatively large quantities of fillers are needed to reach the critical percolation value, as the filler particle size is at micrometer scales. Too high concentration of the conductive filler could lead to materials redundancy and detrimental mechanical properties [3-6]. In case of polymer/graphite composites, a percolation threshold depends on degree of expansion and exfoliation of graphite. The more surface area and the higher aspect ratio of graphite are, the lower amount is needed to form a continuous network. One of the drawbacks of using polymer composites is temperature sensitivity of such materials [1,16]. The analysis of the temperature dependence on the electrical conductivity of a composite material is a useful procedure to understand and optimize its electrical properties.

2. MATERIAL AND METHODS

2.1 Materials

In this study, epoxy resin was selected as polymer matrix. The epoxy resin system was based on a diglycidyl ether of bisphenol A and an aliphatic amine hardener. This system has low viscosity and is processed with a maximum mix ratio to the hardener of 4:1. Commercial powder graphite powder was chosen as the conductive fillers. Epoxy resin has

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several advantages, including exceptional combination properties such as hardness, chemical resistance, high heat distortion temperature, thermal stability and weather properties [17]. Natural graphite powder was provided by Sigma Company with an average particle size of 140 Mesh (0.105 mm).

2.2 Methods

The procedure to manufacture graphite/epoxy composites consists to stir the graphite powder and the epoxy matrix, for approximate 15 min, in order to obtain 10, 20, 30, 40, 50 and 55 (wt.%) of graphite in the composite. 55% content in weight of graphite powder was the maximum content possible to manufacture graphite/epoxy composite, higher concentrations led to graphite wet powder with no polymerization from the matrix. After homogenization, the hardener was added to start the polymerization process. Then, the produced composite was pored into the mold.

A Agilent 34401A digital multimeter was used to measure through-plane electrical volume resistances of samples and silver paint as an electrode material. Molded samples were cut into 50 x 50 mm^2 specimens with thickness about 0.5 mm, then, were applied silver paint all over top and bottom of the specimen surfaces and were dried in air. Measurement apparatus consists of two acrylic plates, two rubber layers and then two layers of conductive metal, in this case we used Aluminum foil, where samples were kept under a constant pressure (2kPa).

The measurements were conducted at room temperature, 40ºC, 60ºC, 80ºC and 100ºC inside a thermostatic chamber to control the environment temperature. The volume resistivity, ρ_V, can be derived from the following equation [1]:

$$\rho_V = (A/t)\, R_V \tag{1}$$

where A = effective area of the measuring electrode (cm^2);
t = average thickness of the specimen (cm);
R_V = measured volume resistance, kΩ. The reciprocal of the volume resistivity is the volume conductivity.

3. RESULTS AND DISCUSSION

3.1 Eletrical Conductivity

Table 1 presents the electrical conductivity tests results, for different graphite fractions at room temperature. It can be observed that the conductivity increases as the weight fraction of graphite increases, showing a significant influence of graphite powder graphite on the polymer matrix. For very low filler contents (10–15 wt.%), the volume conductivity is close to the epoxy matrix, which is considered an isolating material [19,20] and in this case it will be the percolation threshold due to the perfect insulator a polymer and just finite graphite clusters.

A significant increase in the electrical conductivity is observed for percentages higher than 30% suggesting to a semiconductor material, resistivity higher than 10^{-8}. The higher conductivity result is displayed by the 55% content in weight of graphite powder in the epoxy matrix. Since graphite powder is randomly dispersed in the epoxy matrix, the increase of electrical conductivity demonstrates that graphite powder produces long-range connectivity with the epoxy dielectric polymer.

Table 1. Graphite/epoxy conductivity properties at room temperature

Graphite (%)	Resistivity (Ω.cm)	Conductivity (S/cm)
0%	$>1,5\text{x}10^{12}$	$<6.6\text{x}10^{-13}$
10%	$>7.6\text{x}10^{12}$	$<1.3\text{x}10^{-13}$
15%	$6.6\text{x}10^{9}$	$1.5\text{x}10^{-10}$
30 %	$2.7\text{x}10^{5}$	$3.6\text{x}10^{-6}$
40%	$4.5\ \text{x}10^{4}$	$2.2\text{x}10^{-5}$
50%	$1.6\text{x}10^{3}$	$6.2\text{x}10^{-4}$
55%	$1.1\text{x}10^{2}$	$9.1\ \text{x}10^{-4}$

The electrical conductivity results of 30% to 55% graphite/epoxy composites tested at different temperatures ranging from 20ºC to 100ºC are plotted in table 2.

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Table 2 – Electrical conductivity at different temperature (S/cm)

Graphite (%)	Temperature (ºC)				
	20	40	60	80	100
30%	$3.6x10^{-6}$	$3.4x10^{-6}$	$2.1x10^{-6}$	$1.1x10^{-6}$	$7.7x10^{-7}$
40%	$2.2x10^{-5}$	$1.3x10^{-5}$	$4.3x10^{-6}$	$1.5x10^{-6}$	$1.0x10^{-6}$
50%	$6.2x10^{-4}$	$5.4x10^{-4}$	$3.3x10^{-4}$	$2.0x10^{-4}$	$1.1x10^{-4}$
55%	$9.1 x10^{-4}$	$7.4x10^{-4}$	$7.0x10^{-4}$	$5.5x10^{-2}$	$3.0x10^{-2}$

According to table 2 it can be seen that temperature has a significant influence on the electrical conductivity of graphite/epoxy composites. As temperature increases electrical conductivity decreases in all tested formulations. For the case of 30% graphite powder content, the electrical conductivity decreases more than 10 times and the higher difference observed, at 55% graphite powder content, the decrease is more than 100 times when temperature varied from 20ºC to 100ºC.

Figure 1 presents the electrical conductivity of 30%, 40%, 50% and 55% graphite powder content measured at temperatures ranging from 20ºC to 100ºC.

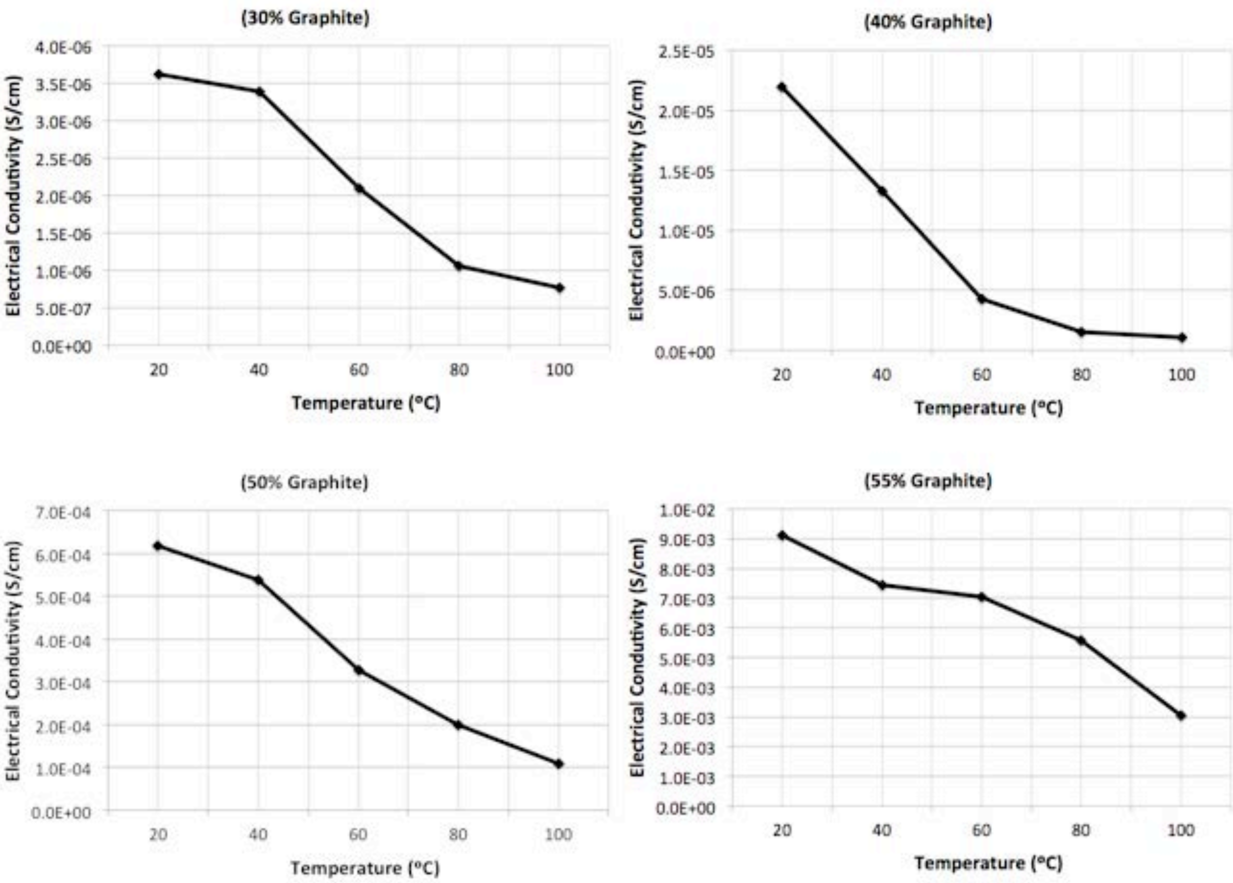

Figure 1 - Electrical conductivity at different temperature for 30%, 40%, 50% and 55% graphite powder content.

According to figure 1 it can be seen that the decrease in the electrical conductivity of graphite/epoxy composites with 30%, 40%, 50% and 55% graphite content is almost linear, suggesting that higher temperatures led to lower electrical conductivity. Measurements at temperatures higher than 100ºC were not possible due to composite thermal degradation. Glass transition temperature of the epoxy matrix is 60ºC [21].

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3.2 Scanning Electron Microscopy

Figure 2 shows the surface SEM micrograph of natural graphite/epoxy composites with 30% (a), 40% (b), 50%(c) and 55% (d) of epoxy resin.

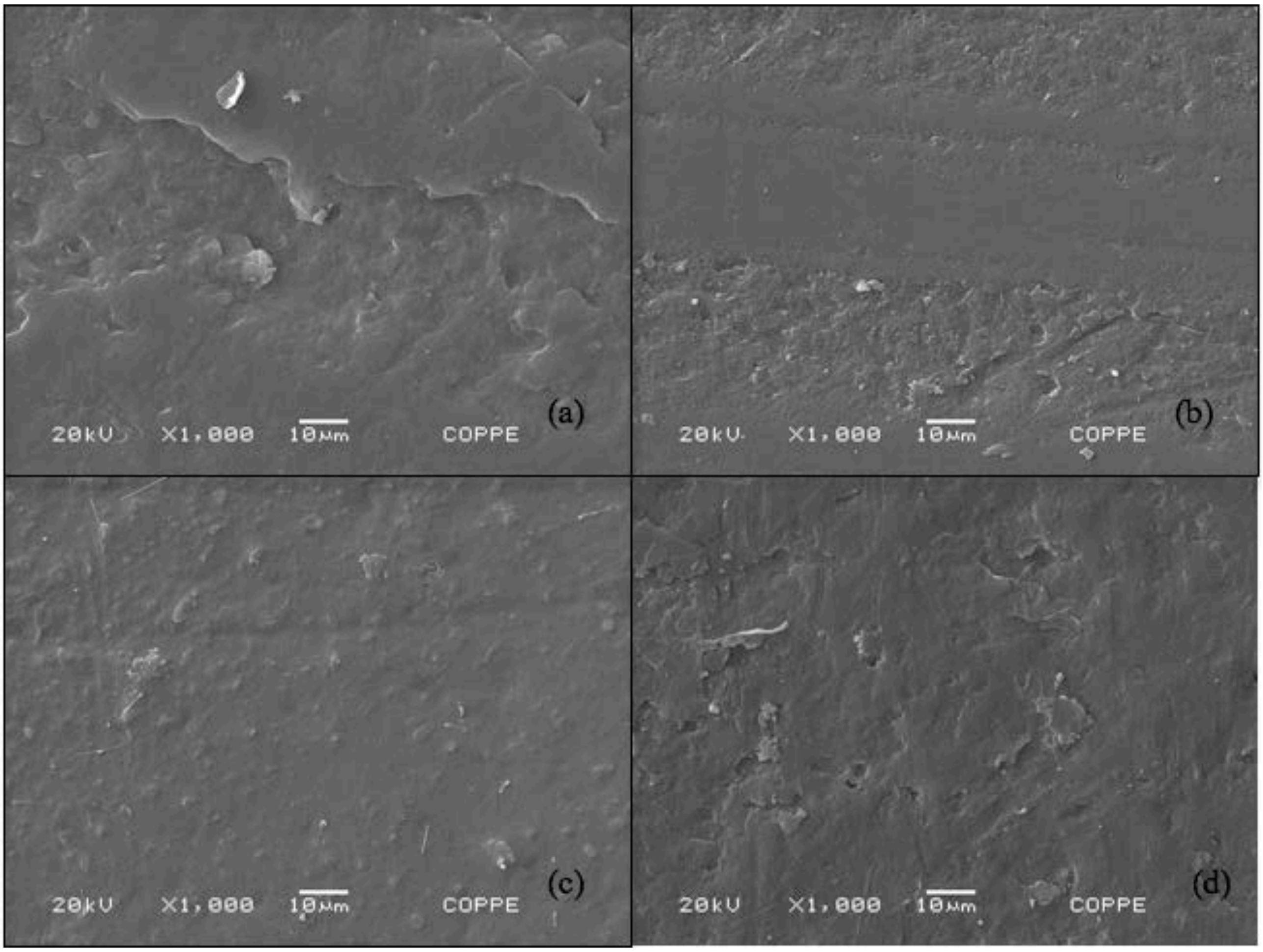

Figure 2- Scanning electron micrograph of sample with 30% (a), 40% (b), 50% (c) and 55% (d) of graphite powder content. Magnification: 1,000x

4. CONCLUSIONS

The electrical conductivity of epoxy polymer matrix is improved by the addition of commercial graphite powder. Since, below the percolation threshold we have no conductivity, 10% and 15% of graphite powder content are considered dielectric like plain epoxy polymer matrix. Composites with 30% of graphite powder and above enter the semiconductor classification, which is excellent from the economical point of view since graphite powder is very cheap compared to others carbon based materials like carbon fibers, carbon black and carbon nanotubes. Temperature severe influences the electrical conductivity of graphite/epoxy composites decrease the results in some cases 100 times.

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