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Mechanical Properties of Non-Toxic Polymer Based Composites for Electronic Devices

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***Abstract*: The increasing demand for smart electronics has led to research on high-performance, mechanically flexible electrically conducting polymer composites. This study examined the performance improvement of adding zinc oxide (ZnO) particles to epoxy resin polymer matrix as conductive and reinforcing filler. With a fixed shape of the composite, 0-40 wt% zinc powder filled polymer composites are mechanically mixed followed by light compression moulding process. The results showed that the addition of ZnO filler significantly decreased electrical resistivity, especially for critically loaded samples. Micro hardness device was used to examine the micro hardness, as well as Universal Machine device was used to the examination the bending strength. The results of the examination of the electrical properties are a clear improvement in the values of electrical conductivity with increasing the weight ratio of zinc oxide. The test results show increase in the mechanical strength of the polymer composite at specific filler concentrations. The tensile strength, modulus and Micro hardness of the ZnO composites containing varying wt% ZnO were improved by 14, 25 and 8%, respectively, in comparison to pure epoxy. The incorporation of ZnO as conducting fillers into an insulating polymer matrix enhances the electrical conductivity to percolation threshold. The percolation threshold value for ZnO-ER composites was found approximately at 30 vol% and addition of 40 vol% ZnO increases the conductivity to 10^{-11} ohm-cm. the incorporation of ZnO in the epoxy composite enhanced the mechanical and electrical conductivity properties of the composite Therefore, electronics industries can profit from the polymer composite by improving existing materials mechanically and electrically with polymer composites.**

***Keywords*: Polymer composite, electrical conductivity, mechanical property, reinforcing filler.**

1. Introduction

Flexible electronics have grown in prominence in recent decades as conductive polymers and organic semiconductors have improved [1]. These materials have provided the foundation for electrical devices in applications such as bending, rolling, folding, and stretching [2]. Current interest in novel conductive polymers enables high-performance, scalable electrical circuits to be produced directly onto flexible substrates, bringing up new possibilities in consumer electronics, architecture, and textiles. Zinc Oxide (ZnO) is an essential electrical and photonic substance with semiconducting and piezoelectric characteristics. When disseminated in polymeric resin, ZnO forms an electrically conducting phase, making it more cost-effective than alternative materials [3]. ZnO, is an important electronic and photonic material which possesses semiconducting and piezoelectric properties which can be used as a proper filling material to improve the electrical properties of polymer matrix [4]. ZnO can create an electrically conducting phase when dispersed in sufficient quantity in a polymeric resin [5]. Epoxy resin is the most important organic matrix for many applications, particularly in the composite industry because of its high tensile strength, high modulus, high adhesion and dimensional stability, low shrinkage, good chemical and corrosion resistance [6]. The properties of such a composite make them technologically superior or more cost effective than alternative materials [7]. Therefore, adding conductive fillers to polymer matrix materials could be pictured with specific properties to proper applications. The electrical conductivity of polymer composites is greatly affected by the concentration of the filler [8]. A conductive network can be established across the polymer matrix when a minimum volume fraction of the conductive filler in the polymer is reached, known as the conductive percolation threshold. At the percolation threshold, the polymer composite, which was initially insulating material, eventually becomes conductive. Electrical conductivity increases exponentially with the filler concentration until a plateau is reached. The percolating behavior is based on percolation theory [9]. In this work, the epoxy resin composites filled with ZnO was prepared at different percentage of loading of filler to investigate electrically and mechanically of the composite.

2. Materials and Method

In this study, epoxy resin and its corresponding hardener are used as matrix material while ZnO used as filler is micro-sized and purchased on the shelf of open market. Epoxy was chosen because it is the most extensively used thermoset polymer and has a low density (1.1 gm/cm^3). Epoxy resin and zinc oxide have qualities that make it suitable for microelectronics applications.

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A. Samples Preparation

Epoxy resin and Hardener was prepared in ratio 2:1 and mixed mechanically at a room temperature. The ZnO of different concentrations were slowly dispersed into the epoxy resin with continuous hand stirring and pour them into wooden moulder laminated with plastic for easy detachment from the moulder. The castings were left to cure at room temperature for about 24 hours after which the samples were removed from the moulds. Composites were prepared into 8 different compositions with varying ZnO content (Table 1). Eight samples of dimension measured 2cm×2cm×0.4 cm with different percentage by weight of ZnO loading with density showing in Table 1 is prepared to investigate the mechanical and electrical properties of these samples.

Table 1
Sample code of epoxy composites filled with varying concentration of Zinc Oxide

Sample code	Composition
S0	Epoxy + 0vol % ZnO
S1	Epoxy + 5vol % ZnO
S2	Epoxy + 10vol % ZnO
S3	Epoxy + 15vol % ZnO
S4	Epoxy + 20vol % ZnO
S5	Epoxy + 25vol % ZnO
S6	Epoxy + 30vol % ZnO
S7	Epoxy + 35vol % ZnO
S8	Epoxy + 40vol % ZnO

3. Experimental Method

A. Electrical Conductivity Measurements

The measurements of DC conductivity in this work were carried-out using a circuit shown in Fig. 1. The figure 1 is the experimental set-up made for measuring electric current flowing through the composite from which resistance and hence conductivity is calculated. A 12 Volt battery is the source of power to the circuit. One terminal of battery is directly connected to the plate over adjustable foundation which is negative terminal and so the fixed foundation plate is cathode. The other terminal of the battery is going through the millimetre which connected in series with the circuit for current measurement. The electrical conductivity (σ) of samples were determined by passing an electric current through a specimen and then measuring the resultant voltage drop over a certain length. The electrical conductivity (S/cm) formula can be written as in Eq. (1):

$$\sigma = \frac{l}{RA} \quad (1)$$

where, R is the electrical resistance of a uniform specimen (ohms: Ω) A is the cross-sectional area of the sample (cm^2) l is the length of the specimen (cm).

1) Density and Void Contents

Density of material is an important factor for determining the properties of the composites. The density of composite depends on the respective relative proportion of matrix and reinforcement densities and volume percentage. The experimental density (ρ_e) is measured using Archmedes principle while theoretical density (ρ_t) is calculated using equation (2) [10].

$$\rho_{ct} = 1/\left\{\left(\frac{w_f}{\rho_f}\right) + \left(\frac{w_m}{\rho_m}\right)\right\} \quad (2)$$

Where, w_f and w_m are weight fractions of filler and matrix respectively and ρ_f and ρ_m are densities of filler and matrix respectively.

Where ρ *and* $\emptyset$ are the density and volume fraction, suffix c, f *and* m represents the composite, filler and matrix material. The density of neat epoxy is measured by Archimedes method are found to be 1.1g/cm^3. Later, the densities of fabricated composites are also measured by the same method.

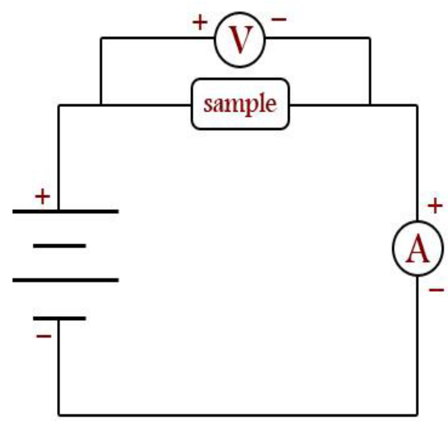

Fig. 1. A circuit used to determine the DC conductivity

B. Mechanical Characterisation

Mechanical characteristics of polymer and zinc powder filled composites are experimentally investigated.

1) Flexural tests

Flexural tests were carried out in a three-point bend arrangement in accordance with ASTM D790-86. The tests were carried out on a 10 KN servo-hydraulic testing equipment outfitted with a test data acquisition system. The machine was run in displacement control mode at a cross head speed of 2.0 mm/min. All tests were carried out at room temperature. The test samples were cut from the panels with a saw fitted with a diamond coated steel blade. For the static flexure test, nine replicate specimens were constructed from the materials. For linear elastic materials, the stress is related to the strain by the Young' modulus, E (Hook's law) [11]:

$$E = \left(\frac{Mass}{Deflecction}\right)\left(\frac{gl^3}{48I}\right) \quad (3)$$

$$I = \frac{wx^3}{12}\, m \quad (4)$$

Where, *I*, engineering bending momentum, *w*, width of samples, *x*, thickness of sample, *g*, gravity, *l*, sample length. Flexural modulus is calculated from the slope of the stress against deflection curve. Bending strength test ASTM D -790 [12]:

$$\sigma_B = \frac{3Fl}{2wx^2} \quad (5)$$

where F is the maximum load at fracture and *l* is the distance of the supports.

2) Hardness tests

The hardness tests involve the use of a static diamond tip under a specific load, over a tested material and over a specific period of time, which forms an indent after removal of the load. This indent is microscopic and in a Vickers hardness test, the shape resembles a pyramid-square shaped impression. Vickers hardness number (VHS) of materials is obtained by dividing the applied force *L,* in Kgf, by the surface of the pyramidal depression yielding the relationship ASTM E-92 -82 [13]:

$$HV_S = (1.8544)\frac{L}{S^2} \quad (6)$$

Where, S is the average length of diagonals in mm. The procedure is suited to a wide range of metals and alloys due to the form and hardness of the indenter. Depending on the test load, a range of 1 to 120 Kgf is selected.

4. Results and Discussion

A. Electrical Conductivity

The variation in DC conductivity of polymer composites with different loading of ZnO is presented in Table 2. It is clear that the polymer composites synthesized is substantially improved by the addition of ZnO. The conductivity of composites arises from a gradual formation of conducting network within the polymer matrix by the insertion of ZnO particles, which results in higher conductivity of composites than the epoxy resin. The maximum conductivity is obtained for epoxy resin with 30-40 wt% ZnO, the region termed percolation threshold.

Figure 2 shows that there is a positive synergistic effect produced conducting path in ZnO and epoxy resin-based polymer composite. Hence, it shows that addition of the electrically conductive ZnO allows for pathways to form with the epoxy resin which results in enhanced electrical conductivity. Metallic oxide particles, especially ZnO particles having considerable semiconducting properties can impart good electrical properties to the insulating polymer.

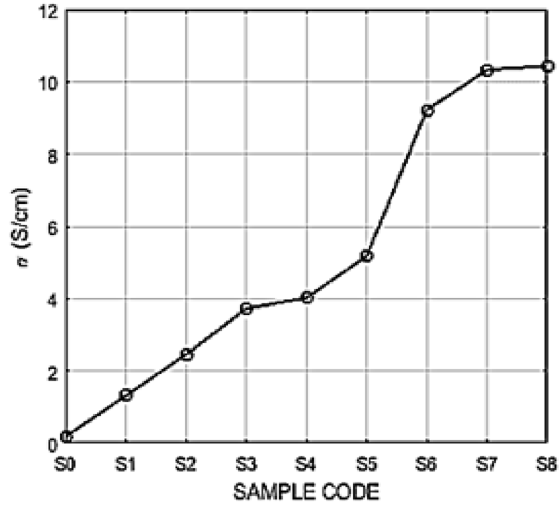

Fig. 1(a)

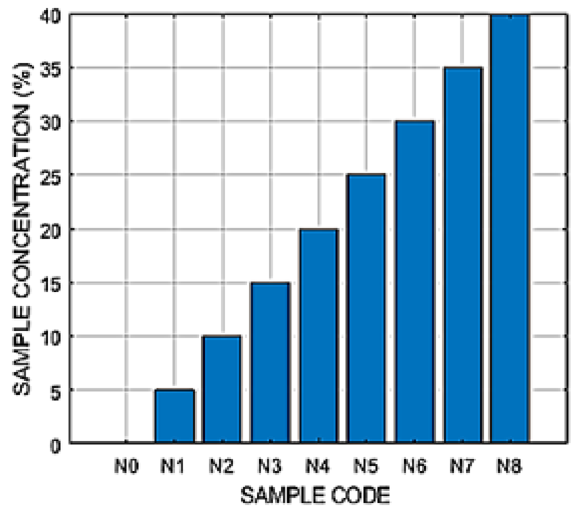

Fig. 1(b)

Fig. 2. (a) Electrical Conductivity of the composite against percentage by weight of filler, (b) Increase in concentration of the filler and sample code

B. Density of the Composite

Fig. 3 depicts experimental and theoretical densities of epoxy- ZnO for different volume fraction of ZnO. It is seen from Fig. 3 that the densities increase with the filler contents.

Table 2
Electrical conductivity σ(S/cm), measured and theoretical densities of the ZnO composites

Sample code	Wt. %	σ (S/cm) $\times 10^{-12}$	Density (g/cm^3)		Void Content
			Theo	Expt.	
S0	0	0.1820	1.10	1.13	0.03
S1	5	1.312	1.17	1.19	0.02
S2	10	2.451	1.20	1.24	0.04
S3	15	3.723	1.26	1.28	0.02
S4	20	4.024	1.29	1.33	0.04
S5	25	5.171	1.34	1.39	0.05
S6	30	9.216	1.38	1.45	0.07
S7	35	10.326	1.47	1.51	0.04
S8	40	10.431	1.53	1.63	0.10

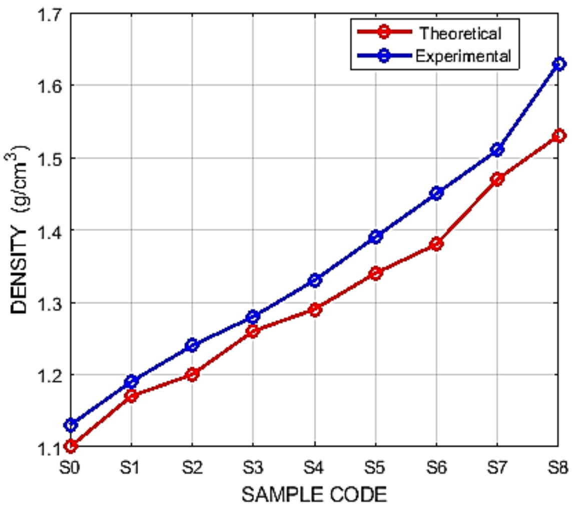

Fig. 3. Experimental and theoretical densities of epoxy- ZnO against percentage by weight of filler

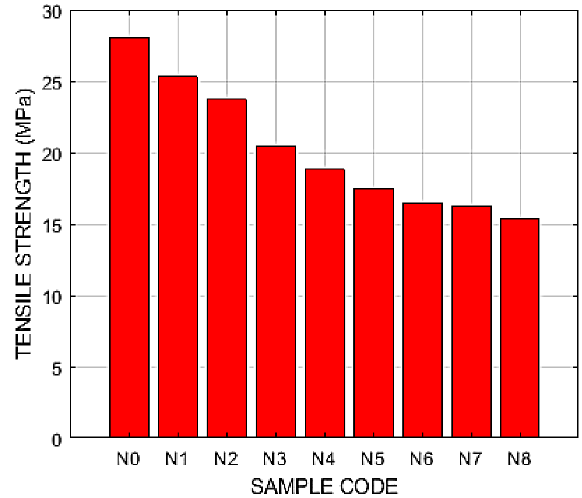

Fig. 4. Mechanical measurements of tensile strength in epoxy resin/ZnO composites

C. Mechanical Measurements of Epoxy Resin/ZnO Composites

The tensile strength, modulus and Micro hardness properties of neat epoxy and composites with different ZnO loadings are presented in Table 3. The effect of different content in the composites is observed on mechanical properties. The same behavior was observed in ZnO composites whereby increasing filler content lead to decrease in the mechanical parameters but hardness increased. Moreover, the composites contain lower values of these properties at lower content than those contain higher ZnO content. This decreasing trend may be due to the low bonding force between the composites. As a result, the sample lost its elasticity and became a more brittle especially for large amounts of ZnO. This means that the poor interfacial compatibility between composites caused low mechanical properties. This decrease was expected due to the cracks and number of pull-out holes that showed in the content void as revealed by difference in experimental and theoretical density calculated. The hardness values of ZnO composites is highest at 0.269 GPa due to the high crosslinking density of ZnO composites in higher content of the composite.

Tensile strengths of the composite specimens were evaluated and the test results for the ZnO composites are presented in Tables 3 This is illustrated in Figure 4 which shows a gradual drop in tensile strength values with increase in filler content. The tensile strength of pure epoxy resin is measure as 50 MPa, this decreases by about 5 % to 55.06 MPa with the addition of 35 vol% of ZnO in the epoxy resin.

Fig. 5 shows the results of the hardness values of pure epoxy resin reinforced with zinc oxide. Zinc oxide in the contents mixed with the matrix increases the hardness apparently. With the maximum enhance examined for concentrations of 40% aluminum. The enhancement in hardness may due to increased surface area of filler in the matrix. On the other hand, as can be suggested from the hardness test, the elastic behavior of the matrix consistently varies with the addition of zinc oxide. An increase in the concentration of zinc oxide particles increases the ability of matrix to absorb hardness and thereby decreasing the tensile and flexural strengths (Fig. 4) but increasing the modulus as shown in fig. 5.

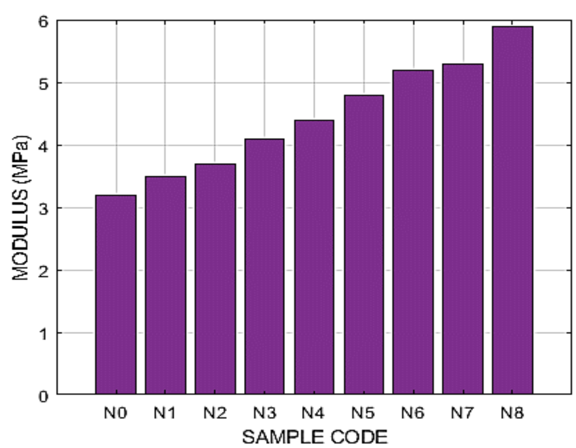

Fig. 5. Mechanical measurements of Young modulus in epoxy resin/ZnO composites

Table 3
Mechanical parameters of epoxy resin/ZnO (with different wt% of ZnO) composites

S.No.	Sample code	Samples concentration %	Tensile strength (MPa)	Modulus (MPa)	Micro-Hardness (GPa)
1	S0	0	28.10	3.2	0.085
2	S1	5	25.31	3.5	0.092
3	S2	10	23.74	3.7	0.105
4	S3	15	20.45	4.1	0.139
5	S4	20	18.84	4.4	0.182
6	S5	25	17.48	4.8	0.198
7	S6	30	16.50	5.2	0.234
8	S7	35	16.26	5.3	0.255
9	S8	40	15.35	5.9	0.269

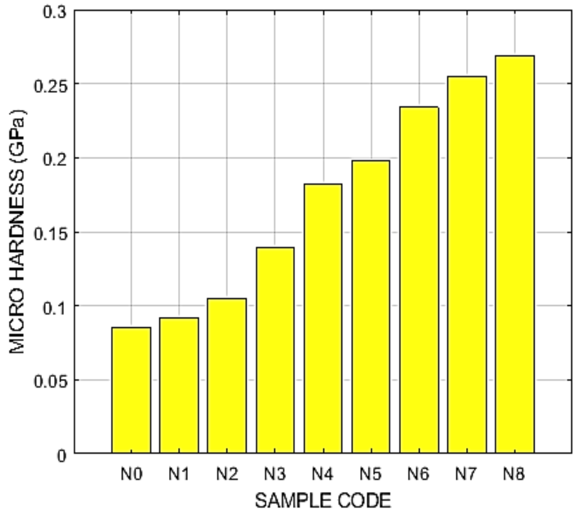

Fig. 6. Mechanical measurements of micro hardness in epoxy resin/ZnO composites

5. Conclusion

Epoxy resin composites filled with ZnO was successfully prepared at different percentage of loading of filler and therefore electrically and mechanically investigated. The results of the study's overall mechanical tests (Figure 4, 5 and 6) showed that adding ZnO particles at certain filler concentrations might greatly increase the epoxy resin polymer matrix's overall mechanical strength. The tensile strength, modulus and Micro hardness of the ZnO composites containing varying wt% ZnO were improved by 14, 25 and 8%, respectively, in comparison to pure epoxy. The incorporation of ZnO as conducting fillers into an insulating polymer matrix enhances the electrical conductivity to percolation threshold. The percolation threshold value for ZnO-ER composites was found approximately at 30 vol% and addition of 40 vol% ZnO increases the conductivity to 10^{-11} ohm-cm. In summary, the incorporation of ZnO in the epoxy composite enhanced the mechanical and electrical conductivity properties of the composite. The spatially confined forced network provides a simple route to construct a continuous and compact conducting filler network in Epoxy/ZnO composites and results in super high electrical conductivity yet mechanical flexible composites with lower filler concentrations. This approach will significantly prompt the applications of conductive polymer composites in the areas of flexible electronics products.

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