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Characterization of Mechanical and Electrical Properties of Polystyrene Composite Reinforced by Hybrid Reinforcement Filler

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ABSTRACT

Nowadays, electrically conductive polymer plays some important roles in the modern electronic devices because the crucial importance in telecommunication and biosensors. This type of materials offers an obvious decrease in the weight of the devices and it also has an environmental compatibility. A conductive polymer composite is promising alternative materials that has several advantages in the electronic applications currently. Polyester was used as a matrix, and copper –Tin /Zink inform of particulates with Copper short wires were considered electrically conductive fillers. The study prepared two groups (A and B) using different filler materials (Cu- Tin /Zink - Polyester and Cu-Tin /Zink –Cu- short wires / Polyester) with a weight fraction percentage of up to 50%. A composite processing method was slip casting to produce rectangular and cylindrical samples; after that samples were cured at room temperature for 48 hrs. Produced samples from both mixed were tested for density, ultimate tensile strength and scan electron microscope. AC conductivity test were performed for all samples using a frequency range from 50 Hz to 50 MHz to measure the dielectrically loss factor and electrical conductivity. From the study, it has been found that filler powder almost proved better ultimate strength behavior, and the peak value was determined at 30% wt., while short wires above 40 % wt. can cause a dramatic reduction in tensile strength. The results revealed that short wires filler van improves the electrical conductivity by approximately 50%; when compared with using particulate filler only. The result from the microstructure showed that short wires with metals powder could provide a connected network for improving the conductivity in the polymer matrix.

1. Introduction

In recent decades, conductive plastic has been one of the significant interesting research subjects due to wide industrial applications, including electromagnetic shielding, antistatic protectors, and electronic sensors and adhesive materials as used as a conductor [1]. The primary goal of conductive polymer base composite research during the past years was to improve the electrical conductivity of the polymer matrix by embedding it with metallic fillers; in form even micro-scale or Nanoscale or both scales using short fibers and Carbon nanotubes. These reinforcement methods have been utilized to form a conductive network in the polymer matrix. Studies have reported that many factors could play a significant role in obtaining an acceptable combination of electrical and mechanical properties of the produced composite, such as filler size and shape and adhesion contact between matrix and fillers [2].

Two types of polymer matrix were used by [3] to improve plastic's electrical and thermal conductivity using various volume fractions that

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reached 40%. Both Polyvinyl chloride (PVC) and epoxy resin were considered a polymer matrix, while Nickle and Copper powder were metallic fillers with an average particle size of 100 µm. PVC-based compressed was prepared using hot pressing, while the casting method was considered for the epoxy base composite. It has been determined that the percolation threshold value of the electrical conductivity obtained after volume faction is more than 0.1with a significant increase in the conductivity after this critical threshold; however, below this value, the composite is an insulator. It has been argued that a layer of an infinite conductive cluster in the polymer matrix increased by increasing the metal filler packing factor and topology of the metals powder that forms the conductive phase in the plastic matrix.

Works conducted by [4, 5] have studied the effect of particle reinforcement on the mechanical properties of polymer matrix composite base materials. It has been found that the adhesion between fillers and matrix plays a significant role in improving the strength and fracture toughness. On the other hand, it has been reported that filler content is another essential factor affecting polymer-based composite materials' stiffness. They documented that when the filler content is over 30%, the fracture's toughness and tensile strength values were decreased. More importantly, the results revealed a critical value of the particle size, which is 30 nm; above this value tensile strength of the composite had been increased.

A study was conducted by [6] to investigate the effect of silica content on epoxy resin's polarization and conduction properties. A casting method was used to prepare a composite electrical polymer base composite by using Nano and micro filler sizes with weight percentage up to 65% micro with the adoption of 5% Nano filler. The study considered a different range of frequency (Low to High) from the study; it was found that the peak value of the dielectric losses factor was noticed at low frequency. After that started to decrease steadily with increasing; they argued this to fillers with high conduction properties. The author discovered that conduction and polarization values were increased by adding silica filler. The optimum electrical values were obtained at a weight ratio of 2.5 Nano and 62 % micro-scale filler content.

An intensive study was conducted by [7] to produce a polymer composite tool electrode used for the electrical discharge machine; Polyester was used as a matrix, and copper with graphite was considered electrical conductive fillers. The study prepared different samples using different process conditions and filler materials (Cu- Polyester and Cu-Graphite / Polyester) with a volume fraction percentage of up to 55%. The study found that the critical threshold of the all-mixing group was approximately above 15%; after it, the electrical conductivity has a linear proportion with the filler content. During curing cycles, cracks were seen for samples with a high volume fraction above 50%; also, it has been concluded that Nanoscale graphite improves the conductivity much better than micro-scale particle size; this was confirmed by what has been found [8]. They argue this to a conductive chain formed by adding small particles to cu powder. The study highlighted that the machining process of the composite is much better when pressing the samples after casting and has a one-week pre-cured time.

With the same objective, [9] conducted experiments on polystyrene/polymethylmethacrylate polymer matrix filled with carbon black (CB) that has a volume fraction % of up to 10%. The study revealed that the initial conductivity of polymer was 10^{-12} S·cm^{-1} and a percolation threshold of 1 wt.% of CB. After that, it was seen that by increasing the CB content, a sharp increase in the conductivity value with an order of magnitude reached 10^{-3} S·cm^{-1}. Microstructure investigation showed a formation of the conductive path in the polymer matrix after CB wt.% was increased.

In contrast, the study by [10] indicated that low melting filler such as (Sn- Pb) or (Bi- Pb - Sn- Cd) could melt with the polymer matrix such as polystyrene just above the melting point of the alloy. Another method was suggested that these alloys can be melted and then extruded after solidified into short fibers to increase the conductive surface area of the filler, which forms a conductive network. Their results indicated that more than 10^{-3} $S{\cdot}cm^{-1}$ electrical conductivity value could be achieved.

In the same line of this field of study, a novel approach was proposed by [11] to enhance the electrical conductivity of the polymer matrix filled with graphene by adding a 2% weight fraction of Nanoscale gold. It has been found that a critical percolation threshold was decreased from 3 to 1weight fraction of the filler content. A research finding by [12] also points towards improving the electrical conductivity of the polystyrene embedded with clay particles had wt.% content reached up to 40%. The obtained value of the conductivity was reached 1.8 $E10^{-7}$.

Recently, an attempt was conducted by [13] to fabricate highly thermal and electrical light weight conductive polymer composite. This sort of materials has been widely utilizing in the electromagnetic shielding and antistatic protection. Polystyrene and Nano pristine graphene powder were used as a matrix and reinforcement fillers with low volume faction of fillers reached to 0.95 Vol.%. A significant increasing of the electrical conductivity was obtained and it reach to 20.5 (S/m), while the thermal conductivity reached to double when it compared with the pure polystyrene. A polystyrene base composite reinforcement by CuO (Nano-scale size) and graphene used the casting method to prepare a conductive plastic composite [14]. A good combination of electrical and mechanical properties was obtained with embedded filler that has wt. 15% and a tight and packed structure without segregation of the particles were seen in the microstructure. Above this value, the embedded filler formation of accumulation was observed. Seven orders of magnitude increased the electrical conductivity. More recently, an effort was achieved by [15] which aimed to used metals particles obtained from industrial waste that has a particle size 150 μm to enhance the electrical conductivity of polystyrene based polymer composite. The fillers contain was ranged from 10 to 40 wt%. From the study the highest electrical conductivity of the produced samples reached to 6 E-7 (S/m), however the porosity was increased by increasing the filler contain which it turns to reduce the mechanical properties of the produced samples This type of polymer composite potently uses as electrostatic dissipative materials.

It is well known that there is some limitation of using a polymer as a conductive material because it is an insulating material. However, a conductive plastic is promising composite materials which has many industrial applications nowadays. Most electrically conductive polymer composites can be filled with conductive filler in forms of particles or fibers to form a conductive path for the electrical conductivity. Compared with even using one type of conductive filler in form of fibers or powder or many conductive metals in form of particulates that dispersion in the matrix. This project describes the influence of using hybrid reinforcements filler in forms of metals particulate and short copper wires on the density tensile strength and dielectrically properties of the epoxy base composite materials. In this current investigation, two groups of polystyrene composites reinforced by hybrid reinforcement filler have been fabricated to prepare composite conductive plastic materials. Some mechanical and electrical properties have been conducted to determine the best ratio that provides a good combination of electrical and mechanical properties.].

2. Experimental procedure

2.1 Materials

For the matrix materials, polystyrene resin with a density of (1060 kg/m^3) and a melting temperature of 240°C was used. Copper, tin, and zinc are one of the metal fillers included in the powder and short fibers; all of them have an average particle size of less than (40 µm). Copper (0.5 mm) is polished for short wires. A typical property of polystyrene is shown in Table 1; additionally; the diameter and length (2 mm) of the material parameters of the base material and the conductive filler.

Table 1: Typical properties of polystyrene [16, 17]

Properties	Unit	Value
Density	g cm^{-3}	1.060
Glass transition temperature,	C°	95-100
Melting temperature, Tm	C°	240
Thermal linear expansion	10^{-5} k^{-1}	6.8
Heat deflection temperature at 445 kPa	C°	82

2.2 Composites preparation

The author suggests two ways to improve polymer-based electrode materials. Characteristics of the current project. The first employs metal powder, while the second uses short Conductive wires with metal powder cast into the polymer matrix. Two sets of samples are made by mixing polystyrene resin and filler in the container. The weight of the substance ranges from 20% to 50%. Tables 2 and 3 illustrate the mixture ratios; Group A was utilized for filler powder, whereas Group B was used for short and powdery wire. It is worth to mention that it has been documented that adding metals fillers in powder forms such as Zn and Sn powder could increase the polymer-based composite's electrical conductivity due to their good electrical properties, these materials could be dispersed in the matrix [18, 19]. Therefore, in this current work an assumption has been made by including these types of metals powder with weight fraction from 10 to 25% in relation to the polymer matrix.

The composite specimens were prepared using the casting method, which includes mixing the resin with the filler well for at least 20 minutes using an electric mixer to achieve uniformity of the resin with the filler materials. The mixture is then placed in an Ultrasonic device for half an hour to remove any bubbles that have formed. The hardener is combined with them once they've been extracted from the device. Methyl ethyl ketone peroxide (MEKP) is added to polystyrene at (1-1.5%) percent ratio which is used as hardener. After that the material slip into mold after they cured for 48 hr, the polishing process was used to prepare it for testing.

Table 2: For group A, percentages of matrix and reinforcing materials

No.	Polyester resin %	Reinforcement %	Details of reinforcement
1	80	20	10% Cu powder + 5% Zn powder + 5% Sn powder
2	70	30	15% Cu powder + 7.5% Zn powder + 7.5% Sn powder
3	60	40	20% Cu powder + 10% Zn powder + 10% Sn powder
4	50	50	25% Cu powder + 12.5% Zn powder + 12.5% Sn powder

Table 3: For group B, percentages of matrix and reinforcing materials

No.	Polyester resin %	Reinforcement %	Details of reinforcement
1	80	20	10% Cu powder + 5% Cu fiber + 2.5% Sn powder + 2.5% Zn powder
2	70	30	15% Cu powder + 7.5% Cu fiber + 3.75% Sn powder + 3.75% Zn powder
3	60	40	20% Cu powder + 10% Cu fiber + 5% Sn powder + 5% Zn powder
4	50	50	25% Cu powder + 12.5% Cu fibre + 6.25% Sn powder + 6.25% Zn powder

2.3.1 Test for composite density analysis

The density is defined as the mass of a substance divided by its volume. Using a sensitive scale, the mass of the materials was measured in grams, and the volume of the model was computed. The volumes of models with regular cylinder shapes were calculated using the cylindrical volume equation (1)

$$\boldsymbol{V = \pi\, r^2\, h} \qquad (1)$$

where, $\boldsymbol{V}$: cylinder volume in unit (cm^3), r : cylinder radius in unit (cm), $\boldsymbol{h}$: cylinder height in unit (cm). We'll use the density law to determine out how dense the samples are now that we've calculated their volumes and weights. In the equation below (2).

$$\boldsymbol{\rho = \frac{M}{V}} \qquad (2)$$

Where $\boldsymbol{\rho}$: density in unit (g\cm^3), M : mass in unit (g), V: volume in unit (cm^3).

2.3 Tensile test evaluation

The tensile strength of eight samples was tested at room temperature at 25°C with head speed using a universal tensile machine (10mm\min). The samples were evaluated according to ASTM 1111555. The examined sample measured (100 mm) in length and thickness (15 mm).

3. Results and discussion

Several tests have been conducted to characterize the conductive polystyrene base composite in this section. The main tests were density, young modules, microstructures inspection, and electrical properties.

3.1 Density and tensile measurements

In Figure the data of density measurements are plotted against the filler content for both reinforcement groups (A and B). It is clear that the trends of density values for powder and powder wires directly proportion with filler contents. As can be seen from the Fig.; the density values were in the range of 1.2 to 2.1 g/cm^3 , which is typically higher than the value of pure polystyrene that has a value of 0.9 g/cm^3 [20]. With filler content from 20 to 30 % it can be readily seen from the Fig. there is no noticeable changes in the density values. Whereas the noticeable rise of the density is observed after 30% concentrate which is determined by 6%.

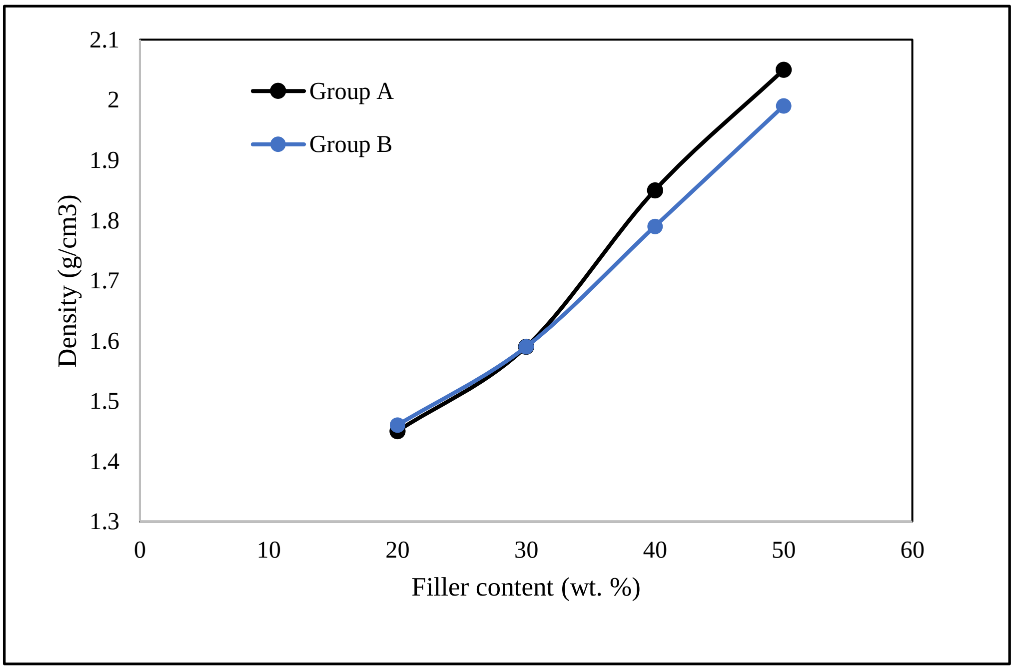

Figure 1. Density vs filler content of Polystyrene / metal powder for both groups (A and B)

Fig. 2 shows the effect of filler content wt.% on the ultimate tensile strength of Polystyrene base composites (Group A and B). It has been determined that the typical value of tensile strength of the pure polystyrene is ranged between 30-48 MPa [21, 22]. It can be noticed that from Fig., the tensile values of Group A, which include powder, only increased with increased filler content till they reached the peak value at 30 %; after that started to decrease to reach 42 MPa. This value can be considered close to the typical range of this type of polymer composite. Slight noticeable changes were seen in the tensile trend in the case of using short wires (Group B) compared with Group A. The tensile trend seemed to be a slight reduction with increasing the filler content of the short wires until they reached 50 %. A significant reduction was observed in the tensile trend, with a tensile value reaching 7 MPa. It can be argued that this behavior has been reported in previous investigations for many reasons. The first reason that could cause this is low adhesion between polymer and short wires, and it is worth mentioning that a small powder filler formed below its melting point, leading to poor interaction with the matrix and short wires. The casting method of this composite with these loading fillers might influence adhesive behaviour between matrix metals with some voids that formed during this fabrication [22].

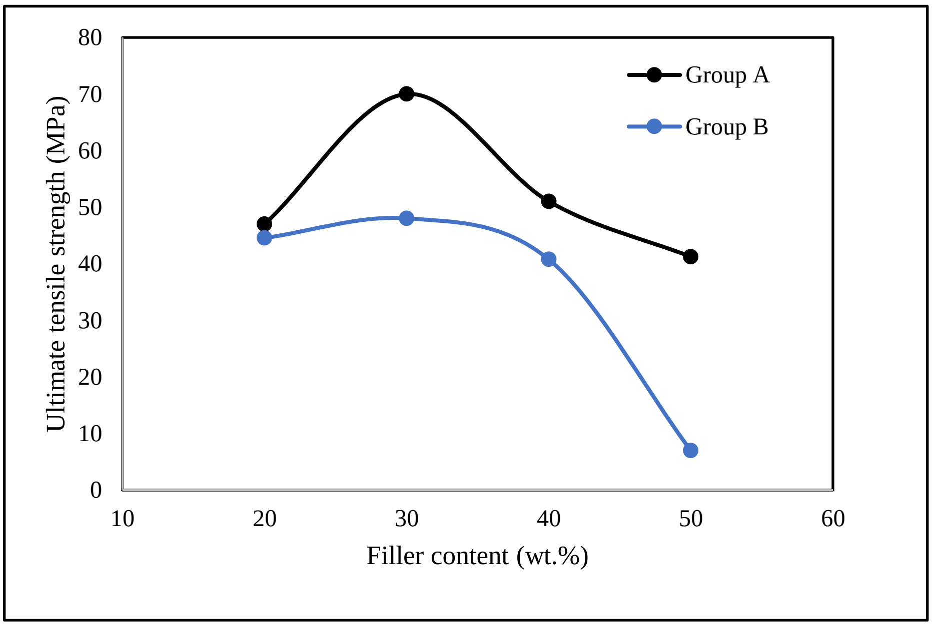

Figure 2. Ultimate tensile strength vs filler content of Polystyrene / meal powder for both groups (A and B)

3.2 Composite electrical analysis

One of the most critical factors that can be used to determine the proper electrical ties of the materials is a dielectric tangent loss factor (tan δ). It can be defined as "the amount of electrical energy dissipated because of differences in the material's electrical conduction. According to the results of Fig. 3, the dielectric tangent loss factor decreased as the frequency increased. One of the important points of the polystyrene base composite can be seen is that tan δ values this can be argued to the principle of the classical physics of the electrical materials is that factor face a reduction in its value when the frequency increases because an external electrical filed that generated in the matrix [23]. In Fig. 4, similar behaviour was seen. The only differences were observed in that group B has almost higher values than group A, particularly in the low-frequency range (from 50 to 1E6Hz). Group A has a value of 0.5. In contrast, in group B the value of tan δ reached 1.7.

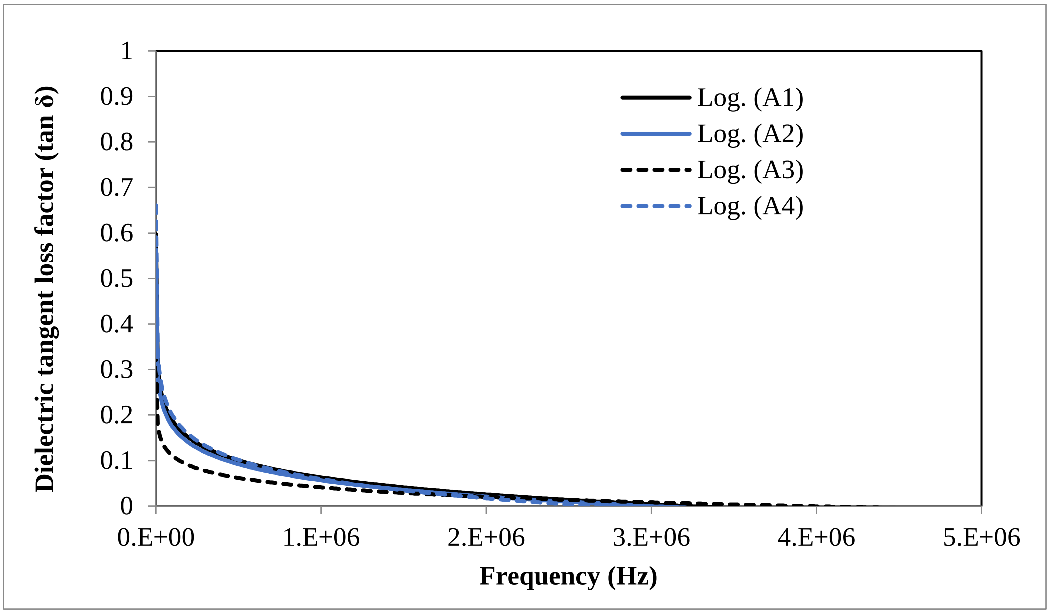

Figure 3. Dielectric tangent loss factor (tan δ) vs. Frequency for Polystyrene / meal powder (Group A) at various filler content

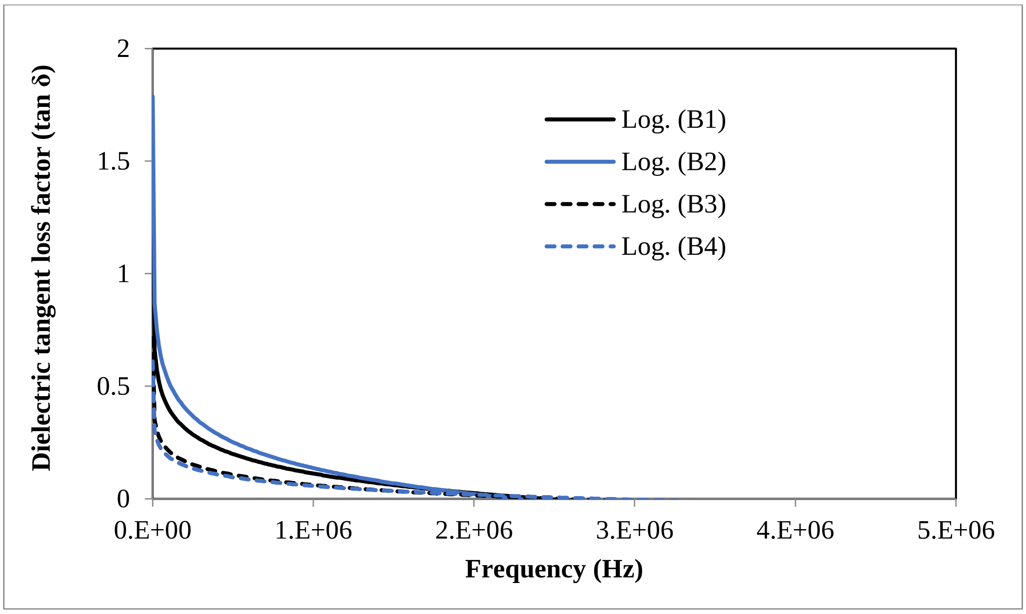

Figure 4. Dielectric tangent loss factor (tan δ) vs. Frequency for Polystyrene / meal powder (Group B) at various filler content

The results of the electrical conductivity for both groups (A and B) were shown in Fig. 5 and Fig. 6, respectively. The general trends of both Fig.s have inverse proportion when compared with the tan δ plots at different frequency ranges because the poor electrically conductive materials are dielectric. This is quite normal as fundamentally explained according to the hopping mechanism when an electrical field was applied to the conductive materials [24]. It is worth mentioning that the typical conductivity value of the polystyrene base matrix is 10E-14 (S /cm); however, by adding an electrically conductive filler in the group, the conductivity was enhanced and reached the value of 2E-4 (S /cm). Moreover, the peak value of the electrical conductivity was 3E-4 (S /cm) when using copper wires group B.

The effect of conductive filler content for both groups on the polystyrene base composite's electrical conductivity was investigated, and the results were presented in Fig. 7. According to the results shown in Fig. 7, conductive filler was added at 20 to 50 wt.%; it has been seen that increasing the filler conductivity of the polymer base composite can be enhanced. The most significant point of the polystyrene base conducting polymer is that more than 13 orders of magnitude can improve its electrical conductivity by adding both fillers powder and short wires.

A rapid enhancement of the electrical conductivity in the case of using short wires of copper (Group B) reaches approximately 50%. Another most significant point has been noted in the case of using short wires with only 30% wt. Content; the best value of conductivity can have obtained without needing to increase the filler. In contrast to using powder only, this is an interesting point. The moderate value of the filler content can get both acceptable combinations between mechanical and electrical properties.

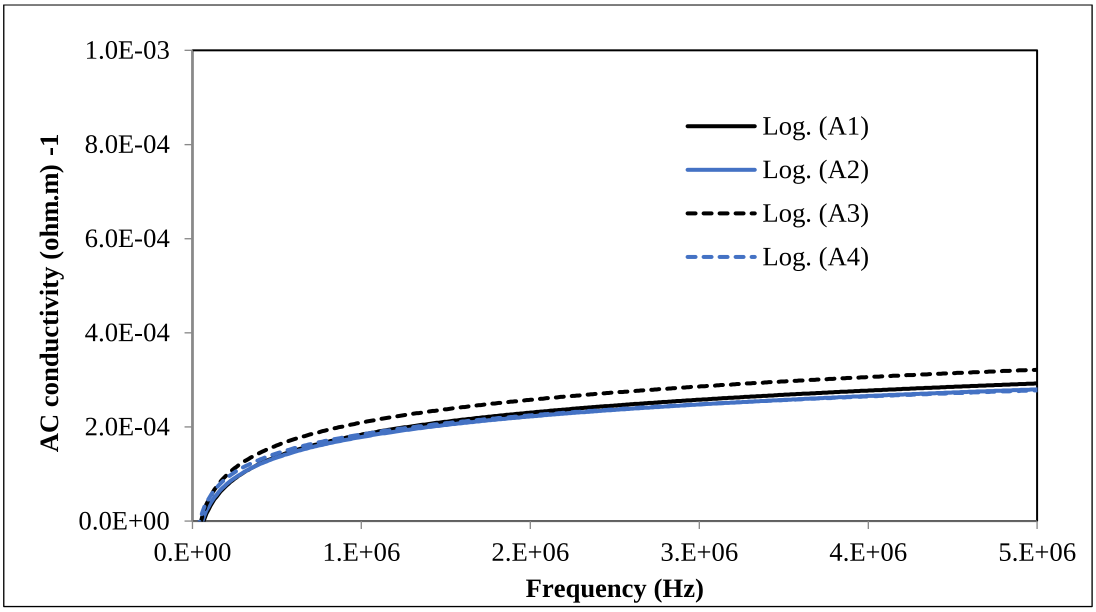

Figure 5. Ac conductivity vs. Frequency for Polystyrene / meal powder (Group A) at various filler content

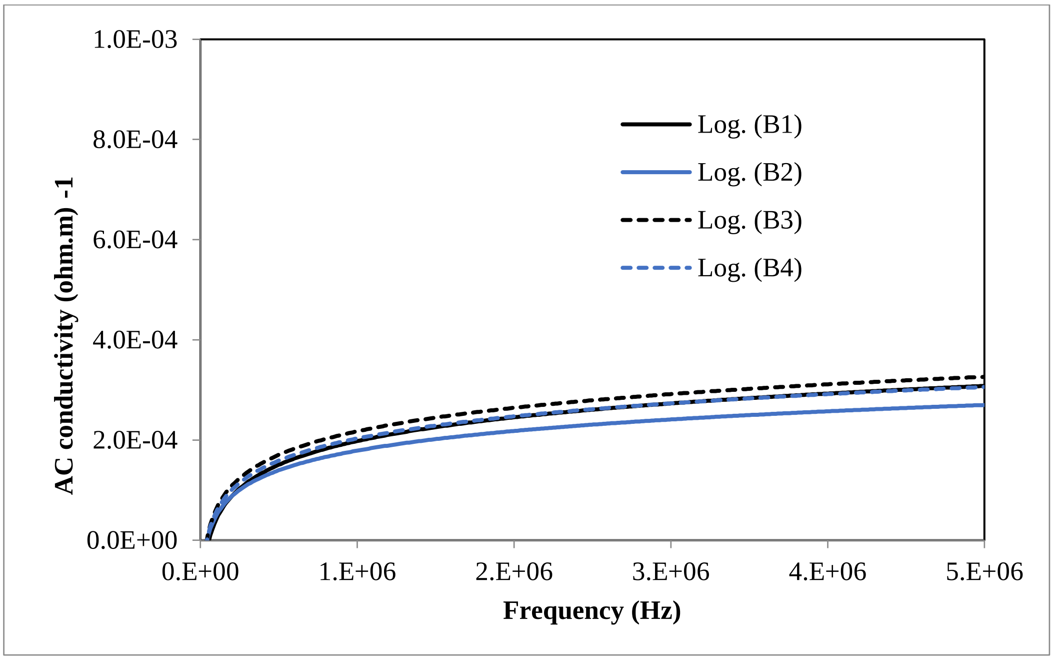

Figure 6. Ac conductivity vs. Frequency for Polystyrene / meal powder (Group B) at various filler content

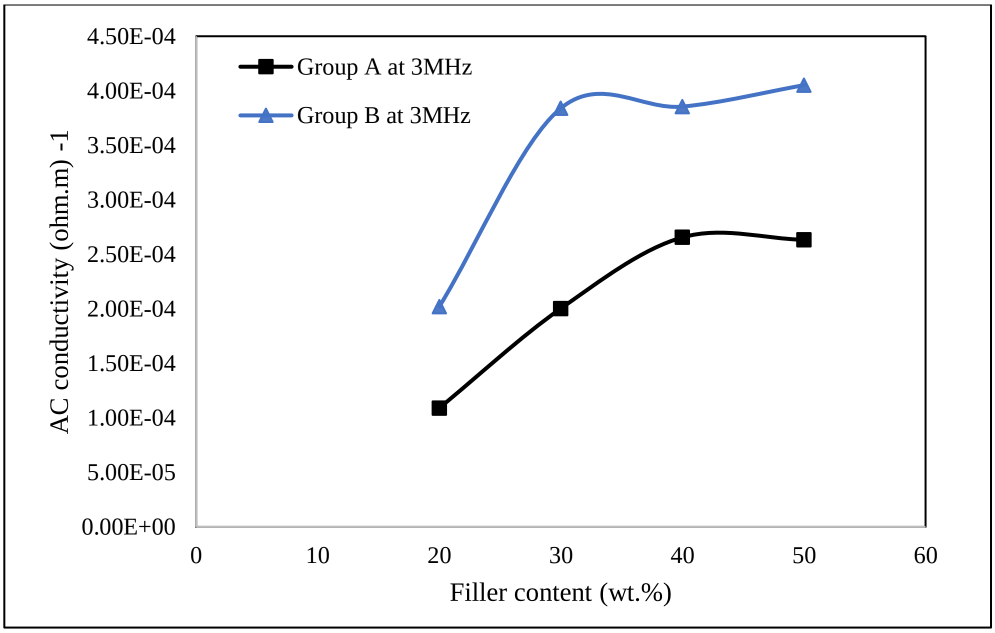

Figure 7. Plot of Ac conductivity vs. filler content for both reinforcements type A and B groups at 3MHz frequency value

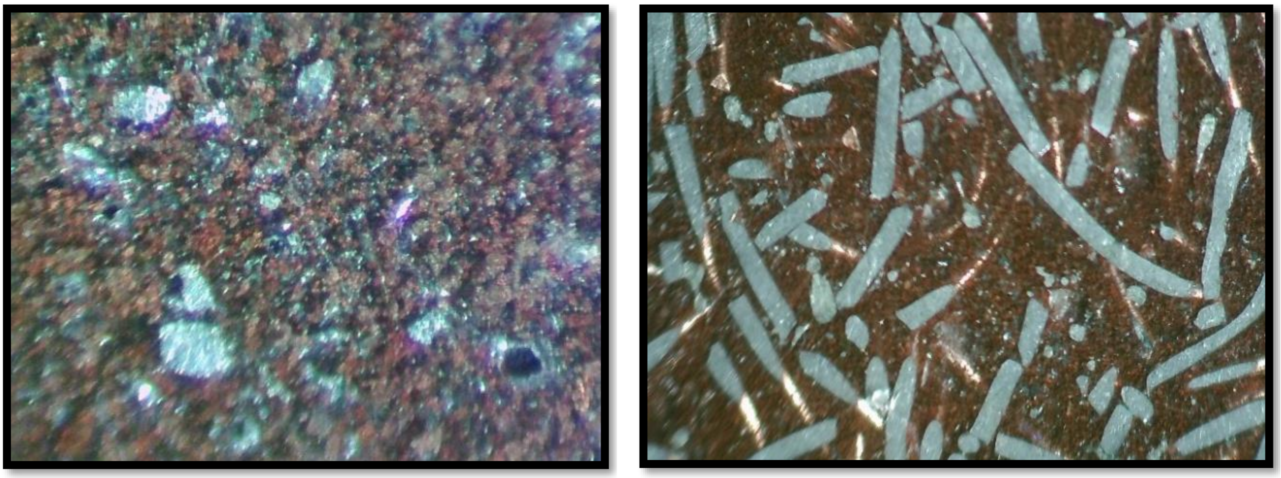

Figure 8. Micro Structure of a polystyrene composite; Left Powder reinforcement; Right short wires with particulate both images have % Wt. content by optical microscopy

The composite microstructure with filler content % is presented in Fig. 8 for both mixed groups (A and B). Clearly, the left image with powder reinforcement showed a sort of uniform distribution for the metal's filler with some voids (a black spot in the image). The bright with luster particles represent the metals fillers, while the matt brown color of the phases refers to the polymer matrix. On the other hand, the right image shows the short wires with metals powder and their spatial arrangement in the polystyrene matrix, again, the light with luster phase represents the metals fillers phase. The poor mechanical properties of the short wires with high content of 50 % wt. can be produced. Because it can cause more brittleness, this type of non-homogenous distribution of the short wires could lead to lower interaction between the matrix and metals phase. However, this kind of structure has enhanced the electrical properties, as shown in the result section.

4. Conclusions

The results obtained from this study with the investigated two reinforcements, metals powder and short wires with particulate powder, allow us to draw some essential points for summarizing the main findings.

1. The density has a linear portion with a filler content of both mixed groups (A and B). A noticeable increase in density is observed after 30% concentration, which is determined by 6%. The density values of the samples have a filler content of more than 30 wt.% has increased the density by 90 % to the pure polymer.
2. Overall trends of the ultimate tensile strength showed a reduction of tensile values after 30% wt. of filler. Using filler powder almost proved better ultimate strength behaviour. The peak value was determined at 30% wt. for Group A. However, a dramatic reduction has been noticed by increasing the short wires for wt.% above 40.
3. This study has found that generally, the electrical conductivity can be enhanced by more than 13 orders of magnitude by adding both fillers powder and short wires compared with the typical value of polystyrene base polymer.
4. The second major finding was that short wires filler van improves the electrical conductivity by approximately 50% compared with using particulate filler only.
5. The close relationship between electrical-mechanical properties and morphological structure of composite shows using short wires with only 30% wt. content; can provide the best conductivity value without increasing filler content above this value. Therefore, dispersion of short wires and metal particulate at this ratio in the matrix is crucial for obtaining a reasonable distribution of fillers that powerfully creates a good combination of mechanical and electrical properties.
6. Increasing the number of layers of the composite material leads to weak adhesive force between layers that leads to decrease in most mechanical properties

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