



Effect of microcarbon particle size and dispersion on the electrical conductivity of LLDPE-carbon composite

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Abstract: This experimental research aimed to develop a conductive polymer composite (CPC) material for use in electromechanical devices. The composite was made by incorporating conductive microcarbon, derived from rice husks, into a Linear Low-Density Polyethylene (LLDPE) polymer matrix using hot compaction. Variations of filler composition were used, with carbon loading of 50%, 45%, and 40%, and mesh sizes of #150, #200, and #250. The experimental results showed that particle size variations did not significantly affect composite density, but higher mesh selection improved filler dispersion within the matrix, resulting in higher electrical conductivity values. The optimal conductivity value of $9.43E-04$ S/cm was achieved with a microcarbon composition of 50% loading. However, decreasing microcarbon loading had a greater impact on reducing electrical conductivity values.

Keywords: LLDPE-carbon composite, rice Husk waste carbon, electrical conductivity polymer composite, conductive carbon, composition ratio

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1. Introduction

This article discusses the engineering of composite materials fabricated from a mixture of conductive organic carbon filler and a binding matrix of LLDPE polymer. The experimental research on composite material engineering can be developed and applied for electromechanical devices known as Molded Interconnect Devices/MID. MID components are made up of both conductive and non-conductive materials, which can be utilized to simplify electronic circuits. This writing will compare the results of this research with several studies on CPC engineering previously conducted by other researchers.

Polymer composites are made by modifying the polymer's characteristics to obtain new properties needed through filler loading. LLDPE is one of the polymer products from the polymerization process of ethylene with the addition of a small amount of α -olefin (Al-Qahtani et al., 2021). LLDPE is similar to Low-Density Polyethylene/LDPE, with a characteristic of short polymer chain branching. Its impact strength and crystal structure can be modified by altering its comonomer distribution (Jørgensen et al., 2010). The unique characteristic of LLDPE polymer is its superior shear sensitivity (Ramkumar et al., 2014), and the addition of certain filler materials (such as carbon, for example) can lower the processing temperature of LLDPE. This article discusses the effect of microcarbon on the electrical conductivity value of LLDPE-carbon composites at a temperature close to the recrystallization temperature (130°C).

In general, to manufacture CPC, the filler materials are dispersed within the polymer matrix by utilizing either direct or indirect methods that capitalize on the polymer's characteristics in its liquid state (Lin et al., 2021). The filler materials and polymer chains adsorb onto each other, forming a bond, similar to the polymer adhesion phenomenon on mold walls (Napolitano, 2020). Once the adsorption on the filler surface reaches equilibrium in the liquid state, excess polymer chains will form polymer layers as the matrix (Metzger et al., 2003).

The addition of conductive carbon fillers to the polymer matrix reduces the non-ohmic phase, resulting in the formation of an ohmic channel network (Choi et al., 2019).

Theoretically, by increasing the amount of carbon filler composition, the electrical conductivity of CPC can be improved. Choi et al. (2019) reported that some polymers show high polarity compared to others. Polarity can inhibit the formation of such channels because polymers tend to have stronger polymer-polymer bonds than polymer-filler bonds. In polymers with high polarity, thicker matrix layers tend to form compared to those with lower polarity. Some types of non-polar polymers include Polypropylene/PP, Polyethylene/PE,

and Polyvinyl Chloride/PVC. According to Choi et al. (2019), a low percolation threshold can be achieved with the use of low polarity polymers, resulting in more conductive composites.

CPC with carbon fillers can be produced by utilizing various types of carbon allotropes, such as carbon nanotube, graphite, carbon black/CB, and activated carbon. Carbon nanotubes (CNTs) are characterized by their extremely small particle size, typically on the order of nanometers. When they are uniformly dispersed throughout a polymer matrix, they can create CPC with percolation thresholds as low as <0.1 loading percentage (Spitalsky et al., 2010). CNTs are further distinguished toward two types of allotropes displaying dissimilar attributes, namely Single Wall Carbon Nanotubes/SWNTs and Multi Wall Carbon Nanotubes/MWNTs. Single Wall Carbon Nanotubes (SWNTs) can form entangled fibers that are difficult to separate into individual tubes. These fibers can have a complex and intricately folded structure, which poses a challenge for achieving uniform dispersion in polymer matrices. The exfoliation process within the polymer matrix requires shear energy that is proportional to the SWNT loading in the composite. Pötschke et al. (2005) achieved a percolation threshold of 0.5- 0.75% loading in melt mixing of Polycarbonate polymer material and SWNTs. It can even be reduced to 0.25-0.5% loading with special treatment. However, Pötschke et al. (2005) only managed to lower the percolation threshold to 0.5% loading for composites with MWNT fillers. The use of CNT fillers is also prevalent in the fabrication of Carbon Fiber Reinforced Polymer (CFRP) composites, as demonstrated by Truong et al. (2018), who employed Epoxy with MWNT fillers to enhance the tensile strength of the composite. The above-mentioned types of carbon allotropes are advanced materials developed with state-of-the-art technology. In applied research in developing countries, this can sometimes be a limitation because the readiness of technology in developing countries is not yet supported.

CB and Graphite are examples of carbon allotropes that can be classified as nano particles but are readily available. Both are conductive carbons with an average particle size of about 30-50 nm. CB powder has good electrical conductivity at low material densities (below 0.5 gram/cm³). On the other hand, due to its anisometric shape, graphite powder's electrical conductivity value increases with increasing density (Marinho et al., 2012). Al-Qahtani et al. (2021) used CB powder with a density of 2.5 g/cm³ to create a CPC. Graphite powder, in contrast, is typically utilized as a filler material after undergoing further processing to produce other carbon allotropes, such as graphene. Some examples of graphite derivative products have been described in the previous paragraph, namely Graphene oxide and GNP.

Horst et al. (2020) successfully fabricated a LLDPE-CB composite for 3D filament applications (with a 15% CB loading) with an electrical conductivity value of 3.33 S/cm. The preparation was carried out using the melt mixing method with a single screw extruder machine at 20 rpm. Al-Qahtani et al. (2021), who used the same polymer material (LLDPE) and carbon allotrope (CB), did not obtain better composite results using the hot compaction molding method. When examining the distribution of fillers within the polymer matrix, Horst et al. (2020) indicated earlier that carbon accumulation occurred in certain areas with a 15% carbon loading composition. However, there is no investigation on the optimal carbon composition limit to be used. Yuan et al. (2010) found that the percolation threshold for Polyethylene/PE polymer can be achieved with a 2% loading composition of CB. In the LLDPE polymer matrix, CB accumulation points with a size of around 50 nm were formed. The presence of accumulation points in a polymer matrix indicates the level of dispersion of CB material within it. Larger particles tend to indicate a less uniform distribution of CB in the matrix, resulting in the formation of aggregates.

The study of nano-sized carbon allotropes presents an abundant and interesting avenue for obtaining comprehensive data on their use as CPC fillers. The strong Van der Waals attraction between nano particles and the increasing amount of filler composition significantly hinders uniform dispersion (Doagou-rad et al., 2018). Research indicates that there is an optimal composition range for certain allotropes that can maximize electrical conductivity (the polymer used as the matrix also has an effect) (Choi et al., 2019). Yuan et al. (2010) provide an explanation for the relationship between electrical conductivity and filler dispersion in composite matrices through their experiments. As the size of the filler aggregate in a composite material increase, there is a corresponding decrease in the determined electrical conductivity value.

Balaed et al. (2016) utilized Typha Latifolia/TL as filler with LLDPE polymer matrix to fabricate a polymer-carbon composite. The TL filler was processed into coarse (1 and 4 mm) and fine (100 and 400 μm) powder, but it was not aimed at obtaining a CPC. Azari et al. (2021) successfully activated coconut shell charcoal to produce conductive carbon. The carbon with a particle size of mesh 200 was then added to the polymer to obtain a CPC with an optimal electrical conductivity value of 2.51E-08 S/cm. It is currently unclear whether the small particle size is the reason for the low electrical conductivity observed in the composite material. In this experimental study, LLDPE-carbon composite materials were engineered to investigate the influence of micro particle size of the carbon filler on electrical conductivity. The microcarbon particles used in this study were derived from organic waste rice husk and were obtained through pyrolysis

at 950°C without the use of chemicals. This is in contrast to the method used by Azari et al. (2021) to obtain conductive carbon. Previous studies have demonstrated that increasing the size of the filler particles can lead to a decrease in electrical conductivity. However, the optimal size range for achieving high electrical conductivity values has not yet been fully determined. This experimental study aims to investigate the effect of micro particle size of carbon filler derived from organic waste rice husk on the electrical conductivity of LLDPE-carbon composite materials.

2. Materials and methods

Microcarbon

The study utilized carbon material in powder form with mesh sizes of #150, #200, and #250, or microcarbon, as the filler. Carbon was obtained through pyrolysis of 1.5 kg of clean rice husks at 950°C, yielding 30% carbon. The rice husks were subjected to carbonization pyrolysis in an airtight furnace with a heating rate of 2°C per minute. Once the pyrolysis temperature was reached, the machine automatically held the temperature for 4 hours. Cooling occurred naturally until the furnace temperature was the same as the ambient temperature. The carbon underwent milling and sieving processes to obtain mesh sizes of #150, #200, and #250.

LLDPE plastic powder

White-colored ETILINAS LL3840UA LLDPE plastic powder with a #40 mesh size is used as the matrix. The relative density to water is 0.91-0.98 gram/cm³, with a Melt Flow Rate (MFR) of 4 gram/10 min. The polymer melting point is at 124°C, with a crystallization temperature achieved at 111°C. The recommended processing temperature from the manufacturer is between 200-300°C depending on the mold material, product wall thickness, and material weight. Impact strength of 12 KJ/m² is tested using Charpy test ISO 179, type 1, Notch A.

Hot Compaction machine

The carbon-LLDPE composite sample is molded using a hydraulic machine that can generate a maximum pressure of 200 Bar. The compaction machine is equipped with a mold that matches the geometric shape of the tensile test specimen according to ASTM D638 standard. Heating is carried out using an auxiliary heater machine with a maximum energy of 500 watts which is placed under the mold. The heating temperature value settings can be achieved automatically by installing a temperature controller that is connected to a bimetal thermocouple heat sensor.

Fabrication of composite specimens

Polymer LLDPE-micro carbon composites are designed with ratios of 50:50, 55:45, and 60:40 % weight. Mixing is done manually, and no special treatment is performed during material preparation. The compaction process is carried out at a pressure of 100 Bar. The heating process is carried out

until it reaches a temperature of 130°C, and then allowed to cool naturally until it reaches a temperature of 115°C.

Density test of specimens

Density testing is conducted following the ASTM D792 standard (Madsen & Lilholt, 2003; Pramono, Rahman et al., 2022; Pramono, Ruswanto & Indayaningsih, 2022; Saadati et al., 2019).

Microstructure test

Microstructure testing of the composite test specimens is performed using the Hitachi SU 3500 SEM machine at the Physics Laboratory of the Badan Riset dan Inovasi Nasional/BRIN, Indonesia. The imaging technique used is Secondary Electron/SE and Backscattered Electron/BSE at magnifications of 100x, 250x, 500x, 750x, and 1000x. As a complement to the test, XRD photos are taken of the polymer-microcarbon-graphite composite. The sample is tested using the TORONTECH TT-EDXPRT X-ray Fluorescence Analyzer in the same laboratory.

Electrical conductivity test

Electrical conductivity testing following the ASTM D4496 standard is conducted using the Keithley Instruments Tools, SourceMeter® Instrument 2450. The measurement is done using the four-point probe method in the Physics Laboratory of the BRIN, Indonesia. The testing process is carried out using a start current of -0.1A, a stop current of 0.1A, and a R holder resistance of 0.000937 ohm (Pramono, Rahman et al., 2022; Pramono, Ruswanto & Indayaningsih, 2022; Yang et al., 2020).

3. Results and discussion

3.1. Characteristics of rice husk micro carbon

The micro carbon obtained from the pyrolysis process of organic materials is amorphous. Previous research has confirmed the XRD test of the organic carbon microstructure to determine its crystal structure (Pramono, Ruswanto & Indayaningsih, 2022). Pramono, Rahman et al., (2022) reported that the carbon microstructure's atomic arrangement is irregular, consisting of one main crystal phase with very low intensity. The crystal structure of a hybrid composite composed of LLDPE-micro carbon-graphite with a loading composition of 50:20:30 was examined in this research. The XRD test results for the sample can be observed in Figure 1. The high intensity in the plot diagram (Figure 1) indicates the presence of graphite structure from the added graphite filler or from the rice husk micro carbon (Swarnalatha et al., 2009).

Graphite used in industry is generally a mineral, not a single element (Popova, 2017). The crystal structure of graphite is commonly observed in XRD testing at a reflection angle (2θ) of

approximately 26°, along with the allotropes CB and CNT (Horst et al., 2020). At reflection angles (2θ) of ~21° and ~24°, two types of silica phases can be found in carbon processed from rice husk organic material. In Figure 1, which shows the XRD test results for the hybrid polymer-micro carbon-graphite composite, the results are generally similar to the XRD test results for activated carbon processed from rice husk organic material (Swarnalatha et al., 2009). LLDPE as a copolymer with many short branch (Dartora et al., 2015) mostly contain Carbon with crystalline peak at 21,5°, 23,8°, 29,5°, 36,1° (Wang & Yan, 2007) shown in Figures 1.

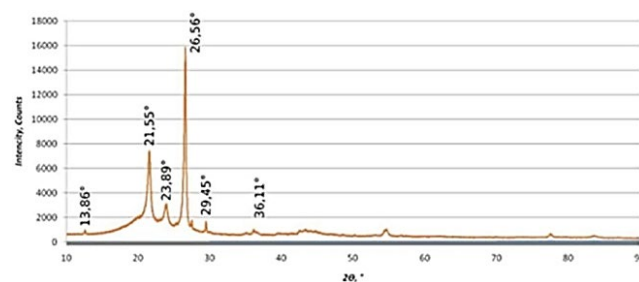
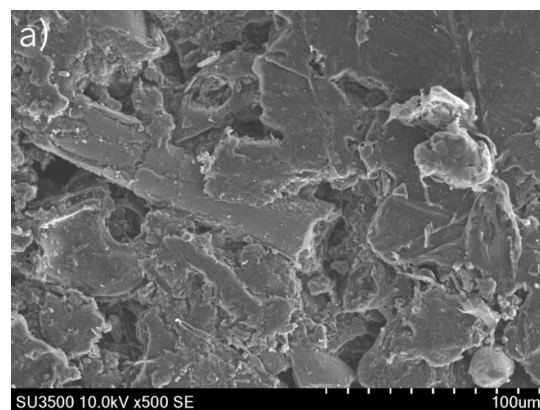


Figure 1. Diagram XRD of hybrid composite samples.

Microstructure of the hybrid composite sample shown in Figures 2, demonstrates irregular structures. An interphase area layered with more conductive carbon presence in the composite (Zare et al., 2022). Micro carbon fillers are visible in the SEM micrograph at 500x magnification, displaying random geometrical particles. Voids have occurred during composite processing and dominate the selected area. The hybrid composite samples demonstrate a distinction in the dispersion of graphite filler, consisting mostly of nano-particles, and carbon filler derived from rice husk, primarily in the form of micro-particles. Larger-sized particles tend to agglomerate and create voids.



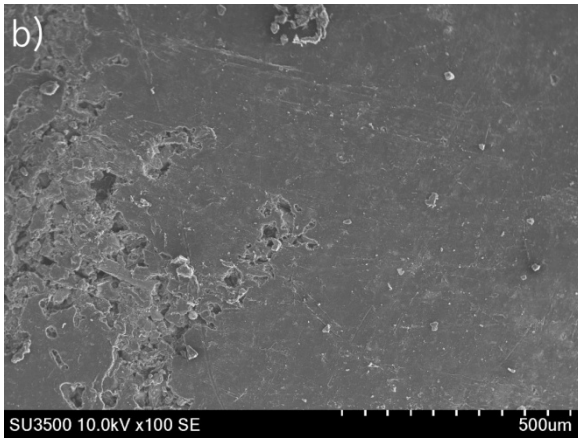


Figure 2. SEM Micrograph of hybrid composite a) 500x magnification b) 100x magnification, A distinction of filler dispersion was observed.

3.2. Density characteristics of polymer-carbon composite

The hot compaction molding process of LLDPE polymer and micro carbon composite material is quite challenging. LLDPE polymer is sensitive to temperature changes (Ramkumar et al., 2014), and carbon has a high thermal conductivity (Marinho et al., 2012). The combination of these two material characteristics reduces the hot compaction molding temperature to 130°C. In the pre-study, the hot compaction molding process was implemented at a temperature of 150°C. The outcome of this process indicated that the LLDPE polymer melted and streamed out from the partition line of the mold. Consistent maintenance of the composition ratio was not possible as the micro carbon particles remained trapped inside the mold. As a result, the LLDPE-carbon composite produced tended to exhibit a higher carbon content compared to the initial mixture during material preparation.

The compounding process was carried out after the LLDPE polymer and micro rice husk carbon materials were mixed according to the predetermined variations. The micro carbon mesh ratio was about 5 times smaller than the size of LLDPE polymer powder particles. The densities of both materials were not significantly different, with the density of micro carbon being higher than that of LLDPE polymer. The manual mixing of the two materials was conducted, and Figure 3a illustrates the mixture condition after compaction. The tiny carbon particles directly adhered to LLDPE, causing the white color of LLDPE to turn black (as illustrated in Figure 3, LLDPE polymer is shown in grey). Compression pressure was applied after filling the mixed materials layer by layer into the mold, assuming that the compression pressure maintained the distribution of LLDPE and carbon materials. As the polymer melted, it was absorbed by the micro carbon particles until equilibrium was reached, resulting in the formation of composite matrix layers as shown in Figure 3b. The carbon

nanoparticles diffused evenly during the heating process and became integrated into the matrix layers (Balberg, 2002; Napolitano, 2020).

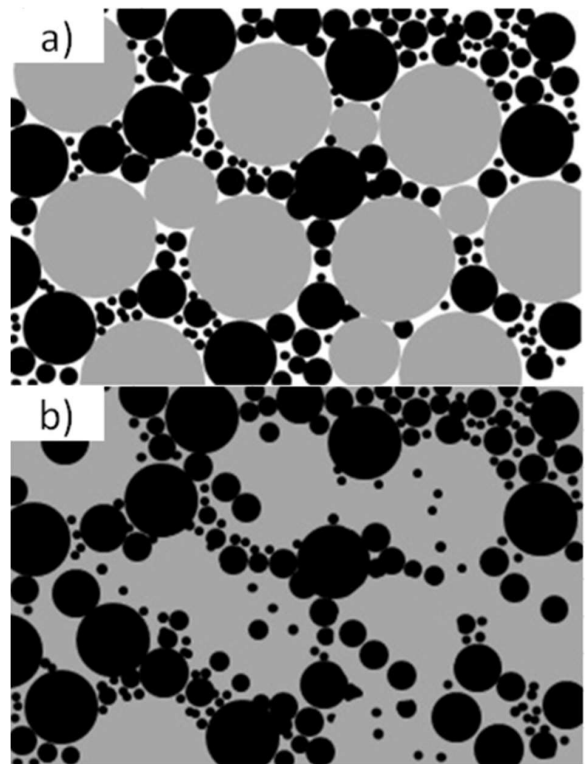


Figure 3. Illustration of the composite formation process, a) during manual mixing; b) during compounding.

The heating process to reach the compaction temperature of 130°C was achieved in 20 minutes, with the compaction pressure applied during the heating process. Pre-heating at 50 bar pressure caused the melting mixture to spread out from the mold parting line. In order to minimize material loss resulting from compaction pressure, a series of experiments were taken to identify the optimum weight of the mixture. It was discovered that a molding pressure of 100 Bar yielded an optimal mixture weight of approximately 15 grams. Once the punch dies were tightly closed on the mold, the mixture was allowed to melt completely at approximately 130°C. The polymer solution was a non-Newtonian solution (Kouzilos et al., 2015) that could not be compressed, so the density did not significantly change during melting.

Density testing was conducted on the fabricated samples to verify the consistency of the experiments, and it was necessary to compare the measured values with the theoretical calculations.

The theoretical formula for density calculation is denoted as ρ ;

$$\rho = (f_{\text{carbon}} \times \rho_{\text{carbon}}) \times (f_{\text{LLDPE}} \times \rho_{\text{LLDPE}}) \quad (1)$$

where ρ is the material density, and f is the loading fraction of the constituent material. The calculation of theoretical density mentioned earlier relied solely on the known density of the composite constituent fraction (Pramono, Rahman et al., 2022). LLDPE polymer has a density of 0.94 gram/cm³, while micro carbon has a density of 1.4 gram/cm³. The obtained densities for composite compositions of 50%, 45%, and 40% micro carbon, respectively, were 1.124 gram/cm³, 1.101 gram/cm³, and 1.078 gram/cm³. The test data from all samples using the Archimedes method are shown in Figure 4. Sample coding was only done to facilitate recording. There was a slight variance in the density values obtained from the two methods.

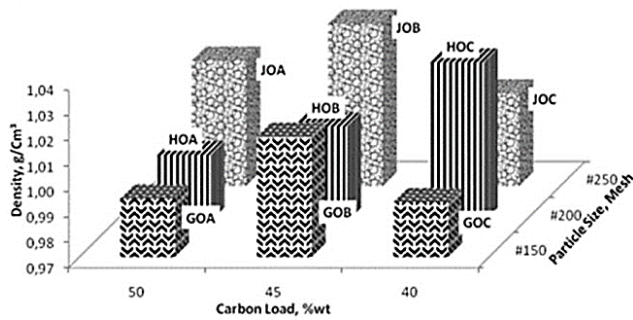


Figure 4. Average densities.

The average values for each composition from high to low carbon loading were obtained as 1 gram/cm³ ± 0.0229, 1.02 gram/cm³ ± 0.0326, and 1.01 gram/cm³ ± 0.0191. This fact indicates that the hot compaction process was consistent for all test samples.

During compaction under pre-heating pressure of 50 bars, molten material propagated through the parting line since there was no density change in the polymer melt phase. At the bottom of the mold, a thin parting line resulted in the propagation of composite with smaller particle size filler, while the punch dies gap allowed for the passage of composite with larger particle size filler (Starr et al., 2002). The density measurement results presented in Figure 3 suggest that the density tends to be lower than the average for mesh selection #150. This could happen if the particle size in mesh selection #150 (the first selection from the milling process) is relatively large on average. This hypothesis could be confirmed through SEM images of the composite sample.

Figure 5 shows the relationship between particle size variations and the density and electrical conductivity data for polymer-microcarbon mixtures with 50%, 45%, and 40% loadings at mesh sizes #150, #200, and #250. Incorporating microcarbon into the polymer mixture with a 50% loading increased the density, with the highest density achieved at the largest mesh size of #250. A similar trend was observed in the 45% loading composition, although the density decreased for

mesh size variation #200. The electrical conductivity showed a tendency to decrease with the addition of micro carbon with a smaller mesh size of #150. Madsen and Lilholt (2003) explained that voids are formed in the composite material structure due to interactions between the composite forming components. Particle size variation at mesh size #250 had a more favorable specific surface area than other variations, the easier adsorption of the polymer into micro carbon resulted in a more uniform bonding between the two materials. Ivan'kova et al. (2022) also reported similar observations when studying the CB structure, although the specific surface area was found to be inside the CB particles as nano-sized porosity.

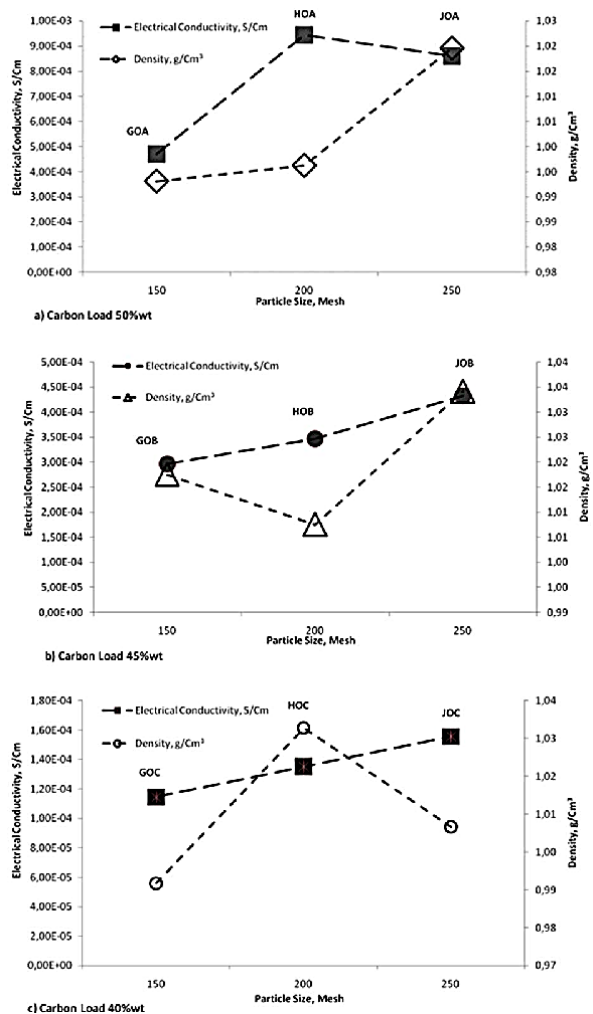


Figure 5. Effect of the carbon size mesh on densities and electrical conductivities.

The presence of voids or macro porosity within the polymer-carbon composite sample may also affect its density value. The composite sample was cut transversely to determine the presence of voids or macro porosity and presented in Table 1 (Madsen & Lilholt, 2003). The initial

pressure of 50 bars is maintained throughout the heating process of the composite mixture from room temperature to its melting point of 130°C. Material flow occurs during the compaction process due to uniform pressure applied to all mold walls. Heating is conducted from the bottom side of the mold (heater position below the mold), where the melted mixture moves upwards towards the punch dies. If air bubbles are formed during the heating process, they will not be easily released.

Table 1 indicates the presence of voids or cavities in several samples with a low unit weight (<14 grams). Annealing process at the recrystallization temperature (around 115°C for 10 minutes) was unable to help the air bubbles escape from the mold, and it is suspected that some bubbles even merged into larger ones, especially in the particle size variation with mesh size #150 selection. Low adsorption can affect the formation of voids or macro porosity within the composite (Madsen & Lilholt, 2003).

Table 1. Weight of composite specimens with mesh of 150

Sample	Weight, gram	Average, gram	Mean, gram	Hole
GOA #1	13,8	15,3	14,6	Y
GOA #2	16,8	15,3	14,6	N
GOB #1	14,6	14,85	14,6	N
GOB #2	15,1	14,85	14,6	N
GOC #1	13,4	13,65	14,6	Y
GOC #2	13,9	13,65	14,6	Y

3.3. Electrical conductivity characteristics

The results of electrical conductivity measurements using the four-point probe method are presented in Figure 6. The HOA, JOB, and JOC samples exhibited the highest electrical conductivity values for carbon loading variations of 50%, 45%, and 40%, respectively. The highest electrical conductivity values were observed for micro carbon particle sizes of mesh #200, #250, and #250 (based on sample coding), with corresponding values of 9.43E-04 S/cm, 4.32E-04 S/cm, and 1.56E-04 S/cm. The electrical conductivity of the composite was found to be strongly dependent on the amount of micro carbon particles loaded in the polymer matrix, with a decrease in electrical conductivity observed as the carbon loading decreased (Azari et al., 2021). The highest electrical conductivity value of all samples was obtained at 50% carbon loading, which was 9.43E-04 S/cm, while the lowest value was obtained at 40% carbon loading, which was 1.14E-04 S/cm. The size of micro carbon particles used as filler had an impact on increasing the electrical conductivity value of the composite. However, the increment in electrical conductivity value was not proportional to the decrease in value caused by filler loading variation.

Changing the size of the micro carbon particles by choosing mesh sizes of #150, #200, and #250 led to only minor variations

in electrical conductivity values. The obtained values still fell within the same range (10 to the power of minus 4). Overall, variations in filler particle size resulted in an increase in electrical conductivity values. In their study, Horst et al. (2020) employed a filler loading of 15% with different variations of the polymer matrix. Using filler particle sizes ranging from 2 to 10 nm and three types of carbon allotropes (CB, GNP, and SWCNT), they obtained electrical conductivity values that were 10,000 times higher than those found in our study, which used an LLDPE polymer matrix. Al-Qahtani et al. (2021) found a decrease in electrical conductivity values with CB filler above 5% loading. The average size of the CB particles used was 30 nm, larger than the filler particle size in the research by Horst et al. (2020). The difference in particle size between the two studies can be approximated as a threefold increase, with the smaller particles being roughly one-third the size of those used in the other study. The resulting electrical conductivity values were markedly dissimilar, with conductivity being a million times greater when smaller particle sizes were used.

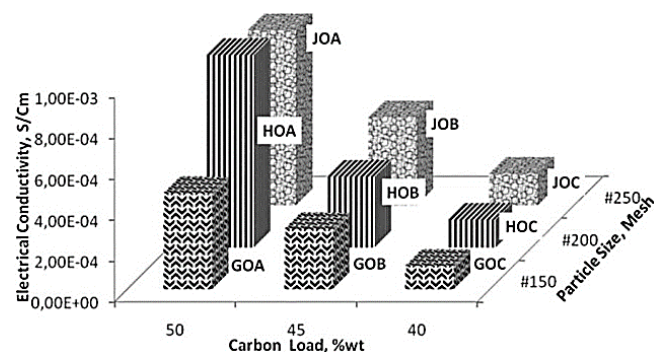


Figure 6. Electrical conductivities of composites.

Percolation theory predicts percolation threshold by measuring resistance of composites. Balberg (2002) tested the classic percolation law by studying the number of conductive channels formed between particles in a composite. However, the experiment revealed inconsistent electrical conductivity values for the same composite composition with different types of CB fillers, including low structure CB/LSCB and high structure CB/HSCB. The classic percolation formula, R as the resistance value of the sample, can be approximated by equation (2),

$$R \propto (m - mc\check{d})^t \tag{2}$$

where m represents the loading of the filler material used, and $mc\check{d}$ is the loading of filler at the percolation threshold obtained in the experiment (Ren et al., 2014). The notation t is used to indicate the exponential value in the experiment. Dibutyl Phthalate solution was used to test the CB material, with the LSCB material having a DBP value of 43 cm³/100g, and

the HSCB material having a DBP value of 350 cm³/100g. Increasing the DBP value or CB structure led to a lower percolation threshold.

The fabrication process of LLDPE-carbon polymer composites varied among the three studies discussed in the previous paragraph. Horst et al. (2020) used melt mixing in a single-screw extruder machine and a 3D printer to create composite filament samples. This technique is frequently used to manufacture polymer nanocomposites because it yields consistent filler dispersion. Oberoi et al. (2019) examined the difference between melt mixing and compression molding for composite fabrication. Results showed that melt mixing (combined with 3D printing) produced samples with 7.9% higher electrical conductivity than compression molding. The three mixtures used by Oberoi et al. (2019) were LLDPE polymer (92%), GNP (6%), and Polypyrrole (2%), with an optimal electrical conductivity value of 3.00E-04 S/cm.

Balberg (2002) has addressed the process control in molding, which can be associated with different exponential values t of the CB types investigated. If no molding treatment is applied to LSCB materials with particles that are almost spherical in shape, it becomes more challenging to form conductive channels. It can be verified by using variation in manufacturing techniques to fabricate composites. The melt mixing process produces higher electrical conductivity values than compression methods. It seems that the exponential value t can also be associated with the rheology phenomenon of composite compounds during the mixing process (Ren et al., 2014). The CB structure phenomenon in HDPE polymer materials modifies the rheological properties of the polymer when HSCB materials are used, leading to stronger interactions between the polymer-filler and filler-filler, which impact the viscoelastic properties of composite compounds, in contrast to the utilization of LSCB materials.

The investigation of conductive filler channels or networks after annealing revealed that the network remained stable during the relaxation process when using HSCB materials. Strong filler-filler interactions support the formation of conductive networks, and the larger specific surface area can absorb more polymer chains to form stiffer bonds (Ren et al., 2014).

HSCB materials are generally considered to have a better structure due to better porosity (larger specific surface area), which can absorb more polymer chains. This denser bonding is believed to improve electron transfer (conductive channels) in composite materials (Hwang et al., 2007). Based on the electrical conductivity data presented in this article, it can be concluded that the micro carbon structure remains unchanged despite variations in particle size. The milling process used to obtain micro carbon in mesh sizes #150, #200, and #250 is believed to improve particle size without modifying the micro carbon structure.

3.4. Micro carbon dispersion characteristics

The characteristics of polymer-carbon composites are highly dependent by the allotrope used as filler. CNTs are the most conductive allotrope, even surpassing the electrical conductivity of copper. CNTs are available in bundles comprising nano sized CNTs with diverse diameters, lengths, and asymmetry, therefore the advantages of using CNTs as fillers represent only a small portion of their exceptional properties. In order to facilitate the dispersion of CNTs throughout the polymer matrix, it is crucial to prevent the formation of agglomerates, which tend to occur in high loading of CNT (Spitalsky et al., 2010). In contrast to CNTs, the observed micro carbon dispersion pattern in HOA samples is quite homogeneous, with thin polymer layers (Figure 7) (Hwang et al., 2007; Ren et al., 2014). In HOA samples with a 50% loading of micro carbon, the electrical conductivity value is the most optimal among all samples.

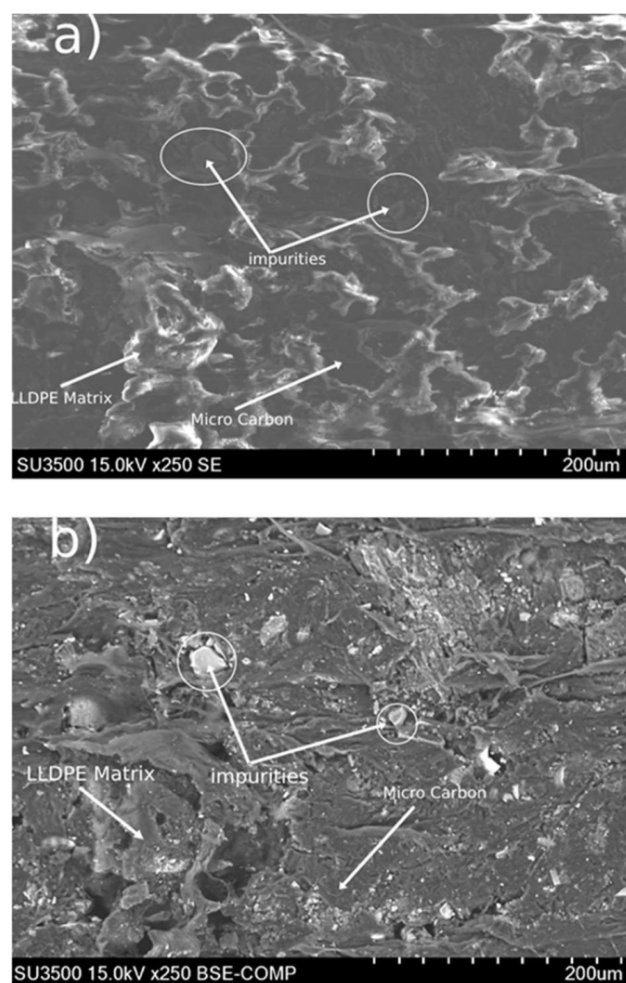


Figure 7. SEM Images, (a) Scattered electrons; (b) backscattered electrons.

Figure 7 shows the microstructure of the HOA sample captured using two different imaging methods. The Scattered Electron (SE) method produces a high-contrast image by applying high charging energy (15kV). In Figure 6a the presence of both micro carbon and LLDPE polymer were represented by dark and light color contrasts, respectively (Zhao et al., 2015). The micro carbon channels are formed between radiant polymer matrixes, enclosed within the white circle. SEM images reveal thick polymer layers on the surface of the micro carbon and its aggregates, overlapping the grain boundaries. The interlocking polymer layers indicate the formation of a network. Balberg (2002) emphasized that the geometry of the filler determines the percolation threshold of the carbon- polymer composite. The utilization of CB as a filler has displayed variations of investigated CB grade's impact on percolation. In this study, micro carbon possesses comparable potential, given that a higher number of aggregates does not lead to a reduction in the electrical conductivity value of the HOA sample.

The particle size of micro carbon for the HOA sample was mesh #200, with a maximum grain size of approximately 74 μm . The micro carbon obtained from the milling process was irregular and non- uniform in shape. The micro carbon occupying the matrix layers was not always intact particles but also aggregates of smaller particles and even agglomerates (Yuan et al., 2010). The presence of nanocarbon aggregates is a commonly observed phenomenon in carbon nano-allotropes, including in CPC produced by incorporating 15% loading of GNP, SWCNT, and CB through melt mixing fabrication method (Horst et al., 2020). Jin-hua et al. (2012) reported that aggregates with a diameter of 500 nm was observed when using 2% loading MWCNT fillers, with an average filler particle size of 30-40 nm. When using CB fillers with the same composition, (Yuan et al., 2010) reported aggregates with a diameter of around 50 nm. The CPC was fabricated using the melt mixing method on a single screw extrusion machine.

Utilizing the Backscattered Electron/BSE method (Figure 7b) it was observed that impurities were present in the micro carbon, which appeared brighter as carbon has a generally lighter atomic number than the impurities found in micro carbon produced from rice husk waste. The primary impurity in micro carbon obtained from rice husk waste is believed to be silica (indicated by the arrow) (Maksum et al., 2019). Impurities with particle sizes of approximately 5-10 μm can be observed in the image, and their distribution within the carbon environment appears to be uniform. The presence of impurities in both the micro carbon and polymer matrix can create channels that enhance the composite's conductivity, enabling electron hopping between micro carbon particles (Zhang et al., 2007). HOA sample with mesh #200 showed the highest electrical conductivity among three micro carbon

loading variations. It is suspected that a thin layer of polymer matrix and excellent dispersion of micro carbon were achieved in the HOA sample. Uniform dispersion and optimal electrical conductivity can be obtained by achieving a proportion between the matrix and filler through a 50% loading of micro carbon. It can be inferred from Figure 8 that if calculated as a volume fraction, the polymer matrix constitutes 60% of the sample volume.

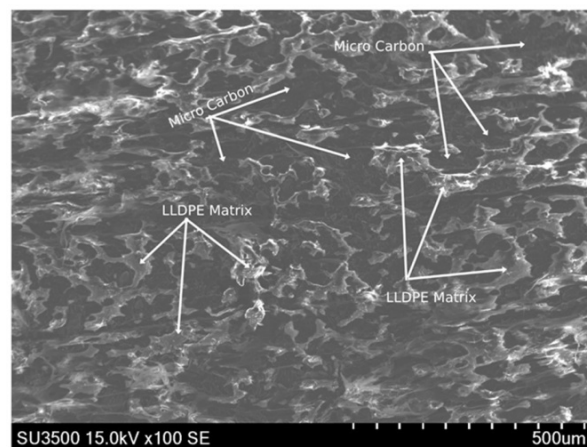


Figure 8. SEM image of HOA sample.

Xia et al. (2004) aimed to improve filler dispersion using mechanochemical pulverization when fabricating a PP-CNT polymer composite. The experiment resulted in a reduction in CNT length from 1-10 μm to 0.4-0.5 μm , increased adhesion between the polymer and CNT, and improved filler dispersion. Figure 8 shows that the formation of micro carbon agglomeration can be reduced, likely because of the larger size of the filler particles, which enhances the adhesion between the polymer matrix and micro carbon. In contrast, when CNT fillers are used, additional measures are needed to increase the adhesion between the composite-forming materials due to the smaller particle size of the CNT fillers. Ikeda et al. (2004) added that CNT purification can also affect CNT adhesion with polymer materials. Jin-hua et al. (2012) confirmed the presence of 0.5 μm -sized aggregates in the 2% loading of CNT filler. The melt-mixing technique with an extrusion machine can improve interfacial bonding between fillers. In contrast to CNT fillers, the use of micro carbon fillers does not exhibit any drawbacks. In fact, aggregate formation can be well- controlled even at very high micro carbon filler content (50% loading), indicating its superior performance over CNT fillers in this study.

Micro carbon particles with a particle size of mesh #150 and #250 were observed for their dispersion in samples with 45% micro carbon loading. The conductive networks in the GOB sample (Figure 9a) were clearly visible at 100x magnification, where the micro carbon aggregates were surrounded by

LLDPE polymer layers with a thickness of approximately 5 μm . This resulted in the formation of a tunneling effect that could enhance the electrical conductivity value (Zhang et al., 2007). In contrast, the micro carbon in the JOB sample (Figure 9b) was uniformly dispersed within the LLDPE polymer matrix, with the matrix layers surrounding the micro carbon aggregates that had formed. Assuming that the micro carbon loading was consistent and proportional in both samples, the dispersion in the JOB sample was more homogeneous. This uniform dispersion led to a composite with better electrical conductivity values compared to the GOB sample. Electrical conductivity measurements showed an increase in the electrical conductivity value for both observed samples: GOB (2.97×10^{-4} S/cm) and JOB (4.32×10^{-4} S/cm).

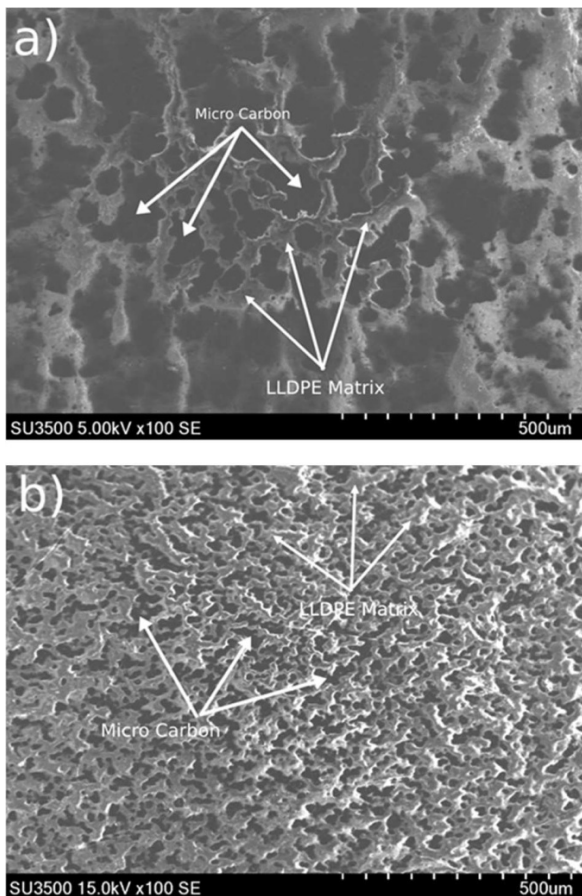


Figure 9. SEM image of sample, (a) GOB and (b) JOB.

Figure 10a shows a 250x magnification SEM image illustrating that aggregates formed in the GOB sample can reach up to 100 μm . The filler used passed through a #150 mesh sieving, with a maximum particle size of 100 μm . The micro carbon dispersion appeared to have a low structure, predominantly with aggregates smaller than 100 μm . The matrix layers also appeared thick (Figure 10a) with some impurities present

(Figure 10b). In the GOB sample, no dominant quantity of impurities was observed, and they tended to be in the form of flakes and large (as shown in Figure 10a).

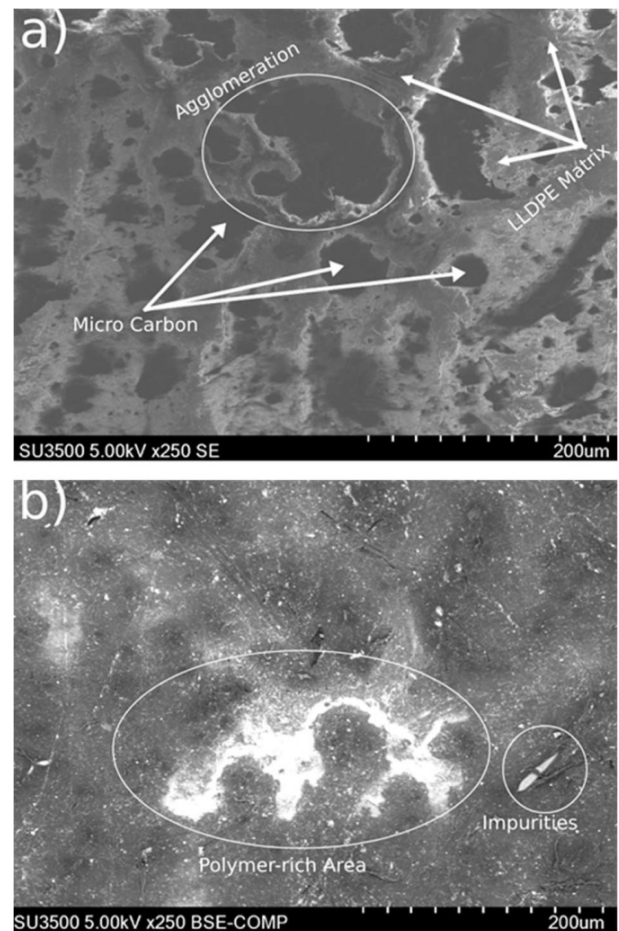


Figure 10. SEM image of GOB sample, (a) SE method; (b) BSE method.

Increasing the micro carbon composition from 45% to 50% loading in the same particle size variation (GOA sample) resulted in a 50% increase in electrical conductivity value. It is hypothesized that in the same #150 mesh particle size variation, the particle geometry factor inhibits the increase of the composite's electrical conductivity value (Balberg, 2002).

Whereas nanocarbon fillers can form conductive networks by occupying positions on comonomer chains (Banerjee & Dutta, 2019), polymer adsorption to micro carbon is mainly influenced by the porosity of micro carbon. Ren et al. (2014) stated that the porosity level of CB fillers is directly proportional to the specific surface area of the CB structure. Higher porosity levels in HSCB enhance conductivity more efficiently than in LSCB. Upon further examination of the GOB sample (Figure 11a), it can be observed that micro carbon agglomeration grains are bounded by thick polymer matrices.

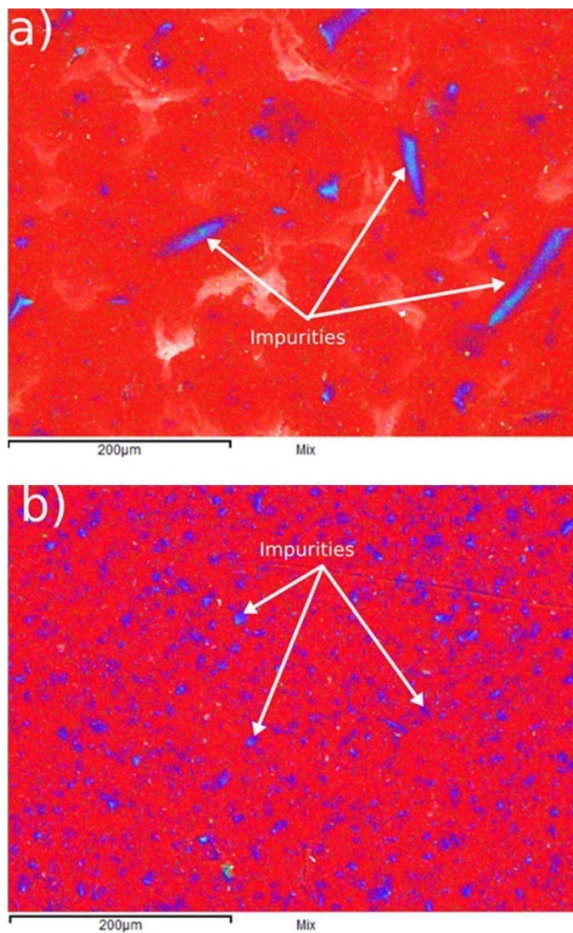


Figure 11. SEM EDX image of impurities.
(a) GOB sample; (b) JOB sample

The equilibrium formation of the polymer as a matrix, with slight variations in thickness, is elucidated in Figure 9. The thickness of the polymer matrix layers tends to be similar for both GOB and JOB samples. Assuming that the bond between micro carbon in GOB and JOB samples is uniform, the dark areas of micro carbon may be comprised of single micro carbon grains or agglomerations, possibly resulting from differences in particle size that form clusters. Agglomeration with high electrical resistance has the potential to decrease the absolute conductivity value measured in four-point probe tests. The higher mesh (JOB sample) yielded particle size uniformity among the GOB and JOB samples. As a result, there was a 50% increase in electrical conductivity, indicating a more uniform dispersion of the filler in the polymer matrix. The presence of small dark areas (Figure 9b) in both samples may be attributed to individual micro carbon particles or micro carbon aggregates, with the latter being more likely due to

their potential to form, suggesting variability in particle size within the mesh scale. The smaller particle size may lead to reduced inter-particle porosity when micro carbon clusters are present. The SEM EDX image in Figure 11b reveals a homogeneous distribution of impurities, particularly silica, in the micro carbon structure. These impurities are latent elements of rice husk organic material that has been processed into micro carbon (Maksum et al., 2019; Swarnalatha et al., 2009;). In the GOB sample, the silica latent impurities from micro carbon are large, as they are found inside micro carbon particles of mesh #150.

3.5. Relationship between particle size and micro carbon weight for electrical conductivity

He and Tjong (2013) experimentally investigated percolation theory using electrical conductivity constants. A formulation derived from equation 2 to predict electrical conductivity value thus,

$$\sigma_m = \sigma_0(m - m_{c0})^t, m \geq m_{c0} \quad (3)$$

The electrical conductivity of conductive composites can be calculated with predictability, whereby σ_0 can be obtained through sample tests once the percolation threshold is achieved. Zare et al. (2022) suggest that this equation represents only a few factors that influence electrical conductivity, to be specific the filler composition and the initial percolation threshold. In polymer-micro carbon composites, the filler particle size proportion can be predicted, but for nano-sized filler particles, Zare et al. (2022) introduced the influence of interphase and tunneling (conductive channels) area. Both areas have the potential to change the electrical conductivity patterns of conductive composite samples.

Several regions indicating the interaction of composite components can be observed in HOA samples with the most favorable electrical conductivity compared to all samples. Figure 12 shows uniformly dispersed small particles, visible with contrasting colors, including carbon particles and natural impurities. The matrix layers formed in HOA samples are typically thin, although they form a network. These particles exist within the polymer matrix, creating an interphase region where the filler acts as the nucleus of crystal structures formed during the solidification process. Zare et al. (2022) suggest that the electrical conductivity of conductive composite samples can be improved with a thick interphase region. When this area is linked to the tunneling area, it can increase the density and proportionally enhance electron flow within the composite.

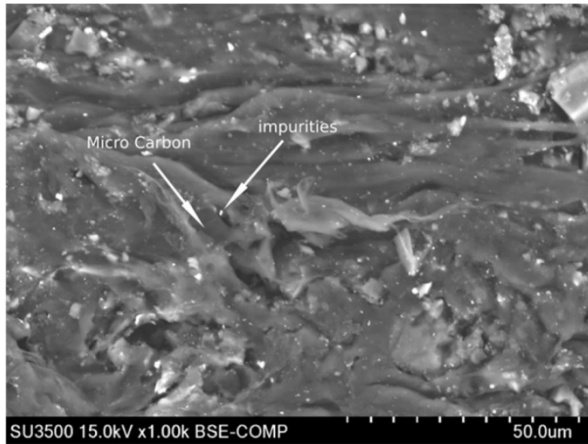


Figure 12. SEM image of HOA sample.

To comprehend the distribution pattern of nano and micro-sized fillers within a polymer matrix, one can make use of the analogy of placing balls in a volume of l^3 . Two distinct types of balls, i.e., non-conductive polymer and conductive carbon material ball, are inserted into a specific volume, l^3 . If the composite is formed by two components having uniform sizes, a higher percentage of conductive balls can improve the electrical conductivity within that volume. There is no significant effect from the difference in ball sizes. If the diameters of both the balls are d and d' respectively (when value of n , $n \approx \infty$) a conductive ball with a size much smaller (e.g., nano) compared to the matrix will be obtained. In a volume having a significant difference in particle sizes, the conductive ball will occupy the empty space between the larger non-conductive balls. The conductive ball is referred to as filler that fills the matrix gaps of non-conductive balls (Cipriani & van Ginkel, 2020; Chiarini & Nitzschner, 2021).

Referring to equations 2 and 3, the filler's loading percentage is denoted by the symbol m . If the particle sizes can be categorized into several types, each size of the filler particles can be denoted specifically, such as ma , mb , mc , ..., and so on. In the case of micro carbon used as filler for this study, the particle size variation is only categorized using mesh scale. An analysis using a particle size analyzer to determine the particle size distribution of micro carbon for each mesh variation has not been conducted yet. The manual sieving process is limited to controlling the mesh size by allowing many particles smaller than the mesh scale to pass through. If the lattice position during the composite solidification process is formed through the formation of nuclei, then the position is randomly occupied by filler particles ma , mb , mc , ..., and so on, either as single particles or particle clusters (Chiarini & Nitzschner, 2021). Particle clusters may occur due to the interaction between fillers in the composite, such as van der Waals bonding.

Marinho et al. (2012) reported that CB materials tend to exhibit high electrical conductivity in powder form at low

densities. Their study revealed an improvement in the electrical conductivity of carbon allotropes by increasing material density. CB materials are particularly affected by the porosity of the CB material, as noted by Ren et al. (2014). Density affects electrical conductivity in composites via improved filler dispersion in the interphase area and tunneling (Zare et al., 2022), contrasting Marinho et al. (2012) findings that density is influenced by compaction pressure in bulk carbon materials. The alignment of density improvement is more acceptable due to variations in filler structure, such as CB materials, compared to variations in compaction pressure or process pressure. Differences in the variation of filler particle sizes in the same mesh scale are thought to have a similar effect to the filler structure. Different particle sizes were found to have different specific surface areas, although the level of improvement obtained in this study was not significant compared to the variation of structures present in CB materials (Ren et al., 2014).

Micro carbon loading as filler affects the electrical conductivity of conductive composites. There was no reduction in electrical conductivity value observed upon the inclusion of filler composition up to 50% loading (Azari et al., 2021). In hybrid composites with a combination of CB and CNT fillers, the addition of CNT fillers can improve the contact between CB particles, thus increasing the electrical conductivity value (Burmistrov et al., 2016). Figure 11 also explains how interactions between micro carbon and smaller-sized micro carbon can occur, forming a conducting area. Meanwhile, small-sized single micro carbon particles that form nuclei in the polymer matrix can improve the density of the interphase area. It is not possible to confirm variations in the microstructure at specific mesh variables, as any observed disparities among the three mesh variables are solely attributed to variations in the distribution of particle sizes. Moreover, the origin of micro carbon is acquired from an identical origin and is solely affected by uncomplicated ball milling mechanical processing.

4. Conclusion

An experiment using micro carbon derived from rice husk organic waste yielded different results compared to the use of nano carbon fillers. Despite having a high carbon composition of up to 50% loading, the electrical conductivity values remained relatively high. Reduction in the amount of micro carbon in the experiment showed an inverse relationship with the decrease in electrical conductivity values.

The use of micro carbon particles with mesh selections of #150, #200, and #250 resulted in a general increase in density and electrical conductivity values. The dispersion of micro carbon particles within the matrix was observed through SEM imaging, showing that particle size improved the distribution

of micro carbon particles within the matrix. SEM imaging also revealed that micro carbon aggregates within the LLDPE polymer matrix tended to be larger in the #150 mesh size variation. The particle size variations used in the experiment did not have a significant effect on the increase in electrical conductivity values unless combined with a balance between matrix composition and filler. This balance is influenced by the amount of micro carbon particles in relation to the loading percentage of the polymer.

The most optimal electrical conductivity value was obtained in a sample with a micro carbon composition of 50% loading and a filler particle size of mesh #200. Meanwhile, the lowest electrical conductivity value was obtained in a sample with a micro carbon composition of 40% loading and a filler particle size selection of mesh #150. More intensive experiments can be conducted for further research, with a larger variation in particle size. The specific surface area of micro carbon plays a significant role in improving polymer adsorption into micro carbon, thus enhancing filler dispersion within the composite matrix. Improving micro carbon dispersion within the polymer matrix greatly affects the formation of conductive channels, which can increase the electrical conductivity value of the polymer-micro carbon composite material.

Conflict of interest

The authors have no conflict of interest to declare.

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