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Pyrolyzed Soybean Hulls as a Potential Conductive Filler in High-Density Polyethylene / Carbon–Black Composites

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Abstract

Electrically conductive polymer composites have a variety of applications in electronics and packaging. Current research focuses on environmentally friendly alternatives to carbon-based fillers, such as Carbon Black (CB), and other inorganic fillers. Biochar, i.e., pyrolyzed biomass, has emerged with great potential due to its high carbon content and thermal stability. This study investigates the feasibility of PSBH as a conductive filler in HDPE/CB composites to determine its effectiveness in enhancing electrical properties. HDPE/PSBH composites were prepared through melt mixing, and their electrical conductivity was analyzed using a four-point probe method. The influence of filler composition, dispersion, and processing parameters on conductivity was evaluated. Results indicate that while PSBH alone has limited conductivity, its combination with CB improves composite performance, offering a potential pathway toward partially bio-based conductive materials. The findings contribute to the development of sustainable polymer composites by integrating agricultural waste-derived carbon materials.

1. Introduction

Polymers are well known for being insulators and have been widely used as electrically insulating materials. Polymers are also replacing various materials due to favorable properties like being lightweight, having high chemical resistance, easy processing, low cost, and having a wide range of properties. Some typical applications of polymers are temperature-pressure sensors, the development of dielectric components, batteries, and so forth [1-3]. Reinforcement additives or fillers are incorporated into polymers to improve their inherent properties. Over the years, many developments have been made in the development of reinforcement fillers for adding properties such as thermal stability at high temperatures, electrical properties, dielectric properties, flame retardancy, and mechanical properties into polymers.

Electrically conductive composites are made using many different fillers, such as carbon Black (CB), Carbon Nanotubes (CNTs), carbon fiber, Graphene Nanoplatelets (GNPs), and metal particles [4-8]. When the resistivity of the composites drops by several orders of magnitude, the critical amount of filler is called the percolation threshold. In the case of High-Density Polyethylene (HDPE)/CB composites, 18% wt. Percolation was reported [9]. In another work, it was found that well-structuring and increased surface area decreased the threshold value from 7.5% vol to 5% vol. Traina et al. [5] report the effect of the surface area of CB particles on percolation threshold in HDPE/CB composites. The authors report a threshold as low as 1.8% vol at a very high surface area of 1353 m2/g. Evgin et al. [3] reported that Carbon Nanotubes (CNTs) of higher aspect ratios had a lower threshold than CNTs of lower aspect ratios. Liang et al. [6] also reported a 7.5% reduction from 18% vol in the percolation threshold when the aspect ratio of carbon fibers (CF) increased. The filler's higher aspect ratio increases the probability of particle-to-particle contact, leading to conductive path formation at lower filler loadings [4]. Conductive polymer composites also utilize intrinsically conductive conjugated polymers, such as polyaniline (PANI), polypyrrole (PPy), polyacetylene, and PEDOT, among others. These polymers contain conjugated π-bond structures of molecular chains, which delocalize π electrons, providing conductivity. These polymers combine the effect of metallic conductors with the benefits of polymers. However, their conductivity is much lower than that of metals. The electronic conductivity of these polymers can be altered by tuning their polymer backbone or forming composites using organic and inorganic fillers or additives [10,11].

With growing awareness of natural alternatives, natural fiber composites have attracted significant attention. The volume resistivity of natural fiber composites with various fibers, including hemp, banana, and agave fibers, and the effect of Maleic Anhydride treatment have been studied [12,13].

It was found that, for all the fibers, the volume resistivity increased with fiber content. The resistivity of these composites was in the order of 10 Ohm-cm [14]. Treatment by maleic anhydride reduced the volume resistivity slightly. It has been found that natural fiber composites have great potential in dielectric applications. Jayamol et al. [15] investigated the electrical properties of pineapple fiber-reinforced low-density polyethylene (LDPE). The author reported a percolation threshold at 15% wt. The volume resistivity improved with increasing filler loading. This was attributed to the hydrophilic nature of lignocellulosic fiber. Conductivity increased due to the presence of higher moisture content. The fibers were also subjected to chemical treatments with Benzoyl Peroxide (BPO), Silanes, peroxide, and isocyanate. It was found that untreated fibers have better conductivity than treated fibers. This is because the treatments reduce the hydrophilicity of the fibers. As moisture plays an essential role in conduction, chemical treatments reduce the conductivity of these composites. Data also showed an increase in dielectric constant when fiber loading increased from 10% to 30% [15]. A decrease in dielectric constant was observed for all treatments. The treatments increased the hydrophobicity, which reduced the orientation polarization, thus decreasing the dielectric constant in all cases [15,16]. The dielectric behavior of sisal fiber composites was also confirmed. The dielectric constant of sisal and coir fiber composites increased with fiber loading but decreased with an increase in fiber length: 1 mm length and 30% wt. Sisal fiber loading had the highest value. The volume resistivity also increased with fiber loading. The volume resistivity of 25% sisal/ 5% CB/ LDPE composite was comparable to 10% CB/LDPE. Adding a small amount of CB and 25% sisal improved the electrical and dielectric properties [14,17].

Although extensive studies exist on the effect of natural materials on the mechanical and thermal properties of polymer composites, limited research has been conducted on their impact on electrical properties [18]. Biobased fillers and fibers are highly hydrophilic, which makes processing and storage more complex. Compared to non-renewable sources of energy, biomass has low energy density. Thus, biomass can be converted into valuable products via biochemical or thermal processes [19,20]. Biochemical processes include anaerobic digestion and fermentation, among others. Thermal treatment can be classified into five types: combustion, pyrolysis, torrefaction, liquefication, and gasification. These processes are categorized based on the temperature and time required for each process. Pyrolysis is performed at temperatures between 400°C and 1200°C [20]. Pyrolysis converts biomass into a high-carbon-containing residue known as biochar. Biochar particles can be milled (ground) to reduce the particle size. Biochar is stable at temperatures of 500°C, making it easily processable. Biochar has a higher energy density and lower hydrophilicity than its biomass counterpart. These features of biochar have made it very attractive in the field of bio-based composites [21,22]. Soybeans were used as biomass for this study. Soybean hulls are composed of the following components: Cellulose (29-51%), Hemicellulose (10-25%), Lignin (1-4%), Pectin (4-8%), Protein (11-15%), and other minor extractives and minerals (trace) 23,24A significant advantage of soybean hulls is their large-scale production in the United States. In 2024, 119 million tons were estimated to be produced. Approximately 8% of the total mass of soybeans consists of soybean hulls, resulting in an estimated production of 9.52 million tons of soybean hulls in 2024 [23,25,26]. The USA produces almost 33.3% of the world's soybeans. Thus, soybean hulls are advantageous because they are inexpensive and typically discarded as waste products. For these reasons, our study focuses on integrating CB and Pyrolyzed Soybean Hulls (PSBH) into the electrical properties of HDPE composites [22].

2. Materials and Methods

2.1 Materials

HDPE in pellet form was obtained from Americhem (Cuyahoga Falls, OH). The Carbon Black (CB) VULCAN 3 (ASTM N330) was acquired from CABOT Corporation (Georgia, USA). The particle size of CB is 26-30 nm. Pyrolyzed Soybean Hulls (PSBH) used as a potential reinforcement filler to replace some amounts of Carbon Black (CB) were produced by NexGen industries (NGI, Chavies, KY). The PSBH produced was pyrolyzed at 500°C for 4 mins.

2.2 Experimental Methods

2.2.1 Ball Mill Treatment

HDPE, PSBH, HDPE/PSBH Pellets, and CB were vacuum-dried for 24 hrs. The rectangular samples, measuring 12.5 cm x 12.5 cm x 1.5 mm, were first prepared by compounding HDPE, PSBH, and CB in fixed proportions by weight and then by compression molding.

2.2.2 Sample Preparation

A 764-AV Jar Mill (US Stoneware, East Palestine, OH) was used to reduce the particle size of Pyrolyzed Soybean Hulls (PSBH). The roller RPM was set at 75 RPM. The milling process lasted 24 hours. After milling, the milled PSBH was stored in airtight Ziploc bags. The particle size of milled PSBH is 1-7 µm [27-30].

2.2.3 Compounding

The compounding of the materials was performed using a Plasti-Corder Brabender Mixer (C.W. Brabender Instruments, South Hackensack, NJ) at 175°C and 20 RPM for 30 minutes. The samples were prepared to determine the variation of properties as a function of changes in operating parameters and were compounded under different operating conditions. The operating conditions used for this purpose were 5, 12.5, 20, and 27.5 RPM; 150°C, 163°C, 175°C, 187°C, 200°C; and 5, 10, 20, 40, and 60 mins, respectively.

Table 1 *Compositions of HDPE/PSBH, HDPE/CB composites*

Sample Name	Amount of HDPE (Wt. %)	Amount of PSBH (Wt. %)	Amount of CB (Wt. %)
HDPE / Unmilled PSBH (PSBH Pyrolyzed at 500°C for 4 mins)			
10% PSBH 90% HDPE	90	10	N/A
20% PSBH 80% HDPE	80	20	N/A
30% PSBH 70% HDPE	70	30	N/A
50% PSBH 50% HDPE	50	50	N/A
60% PSBH 40% HDPE	40	60	N/A
HDPE / Milled PSBH (PSBH Pyrolyzed at 500°C for 4 mins)			
10% PSBH 90% HDPE	90	10	N/A
20% PSBH 80% HDPE	80	20	N/A
30% PSBH 70% HDPE	70	30	N/A
50% PSBH 50% HDPE	50	50	N/A
60% PSBH 40% HDPE	40	60	N/A
HDPE / CB			
15% CB 85% HDPE	85	N/A	15
17% CB 83% HDPE	83	N/A	17
18% CB 82% HDPE	82	N/A	18
20% CB 80% HDPE	80	N/A	20
25% CB 75% HDPE	75	N/A	25
30% CB 70% HDPE	70	N/A	30
40% CB 60% HDPE	60	N/A	40
50% CB 50% HDPE	50	N/A	50

The samples were prepared to determine the variation of properties as a function of changes in operating parameters, and were compounded under different operating conditions. The operating conditions used for this purpose were 5, 12.5, 20, and 27.5 RPM; 150°C, 163°C, 175°C, 187°C, 200°C; and 5, 10, 20, 40, and 60 mins, respectively.

Table 2 *Compositions of HDPE/PSBH/CB composites*

Sample Name	Amount of HDPE (Wt. %)	Amount of PSBH (Wt. %)	Amount of CB (Wt. %)
HDPE / Unmilled PSBH / CB (PSBH Pyrolyzed at 500°C for 4 mins)			
5% PSBH 18% CB 77% HDPE	77	5	18
10% PSBH 18% CB 72% HDPE	72	10	18
15% PSBH 18% CB 67% HDPE	67	15	18
20% PSBH 18% CB 62% HDPE	62	20	18
25% PSBH 18% CB 57% HDPE	57	25	18
HDPE / Milled PSBH / CB (PSBH Pyrolyzed at 500°C for 4 mins)			
5% PSBH 18% CB 77% HDPE	77	5	18
10% PSBH 18% CB 72% HDPE	72	10	18
15% PSBH 18% CB 67% HDPE	67	15	18
20% PSBH 18% CB 62% HDPE	62	20	18
25% PSBH 18% CB 57% HDPE	57	25	18

2.2.4 Molding

The composites were vacuum-dried before molding. The vacuum-dried composites were compression-molded (Carver Compression Press, Wabash, IN) into rectangular samples of dimensions 12.5 cm x 12.5 cm x 1.5 mm. The molding temperature was 175°C, and the pressure was 2,100 psi (~14.5 MPa). Specimens for Dynamic Mechanical Analysis (DMA) with dimensions of 63.5 mm x 12.3 mm x 3 mm were also molded at a processing temperature of 175°C and a pressure of 2100 psi (~14.5 MPa). The mold was cooled using a water-cooling system.

The HDPE/PSBH blends were injection molded into tensile coupons using a micro-injection molding machine (DSM Micro-Injection Molding ASTM D638 Type V, Netherlands). The melting temperature was set at 175°C, the mold temperature was maintained at 65°C, and the injection pressure was kept at 0.6 MPa. The material was allowed to melt in the chamber for 5 minutes and was manually pushed using a plunger to release trapped air and allow proper melting. The material was kept in the mold for 30 seconds before being removed. Five tensile coupon specimens of each sample were made. The mold shape was according to ASTM D638 Type V [22].

2.2.5 Characterization

Mechanical properties, including breaking strain, tensile strength, and modulus, were determined through tensile testing. Tensile tests were performed by Universal Test Frame – Instron 5567 tensile testing machine (Instron, Norwood, MA). The overhead rate was 10 mm/min, and the gauge length of the sample was 7.62 mm. A 1 kN load cell was used. Five dog bone tensile testing specimens were tested for each condition to obtain a standard deviation. Thickness was measured for each of the five specimens.

The electrical properties were investigated using Kelvin's 4-point method. Voltage (10, 20, 50, 100 V) was applied across the sample using a DC power supply, and Current and Voltage were measured using Ammeter and Voltmeter, respectively. The BK Precision 3 ½ Digit Multimeter (Tektronix, 14150 SW Karl Braun Drive, OR 97077) and the Astro AI Digital Multimeter (ASTRO AI, 7423 Doig Dr, Garden Grove, CA 92841) were used to measure the current passing through the sample and the voltage across the sample. A regulated power supply model 50 (Lambda Electronics Corp., Neptune, NJ) was used as the power supply. The sample was kept between two metal plates covered with Aluminum foil, which was used to enhance surface contact. One kilogram of calibration weight was placed on the sample to ensure complete contact with the plate. The Resistivity of the sample was calculated using standard formulas. The first step is to calculate Resistance using Ohm's Law:

$$R = \frac{V}{I} \tag{1}$$

$$Resistivity = \frac{RA}{L} \tag{2}$$

In Equation 1, V represents voltage (V), and I represents current in microamps (μA) or milliamps (mA). The sample dimensions were 12.5 cm in length, 12.5 cm in width, and 1.5 mm in thickness. In the second step, the resistivity was calculated using Equation 2, where R = Resistance, A = sample length × sample width, and L = sample thickness.

The morphology of the samples was studied using Scanning Electron Microscopy (SEM). JOEL SEM (Hitachi Tokyo, Model S2150) was used to study morphology. The cross-section of the samples was observed. A CT scanner was used to obtain tomographic data of the composite samples. Scans were obtained using micro-computed tomography (μCT) X-ray tomography (Skyscan 1172, Bruker, Billerica, MA, USA). For X-ray generation, an accelerating voltage of 80 kV and a current of 128 μA were used. The degrees of step rotation were set to 0.3, the average frames were set to 6, and the random movement should be set to 10. The Micro CT images were reconstructed using NRecon software. The typical acquisition time was 3 hrs [22].

3. Results and Discussion

3.1 Electrical Properties

The resistivity of HDPE composites was determined using the 4-point Kelvin Method, as described in the experimental section. The most significant percentage of errors in the resistivity values is ±9 %. The standard deviation was extremely small and is present in the figures but is not visibly distinguishable due to scale limitations. Figure 1 shows the resistivity of HDPE/CB and HDPE/PSBH (Milled and Unmilled PSBH) composites as a function of filler quantity. CB is conductive, and the resistivity of HDPE/CB decreases with increasing CB. It was found that 18% CB by weight is the percolation threshold for HDPE/CB composites, i.e., at 18% CB by weight, the resistivity drops by several orders of magnitude, and the sample becomes more conductive. The conduction threshold for HDPE/CB composites appears to be ~15.5% wt. (Fig. 1).

Although it is highly carbonized, PSBH exhibits low conductivity, and the threshold value for HDPE/PSBH composites is very high. It was found that HDPE/PSBH composites have a conduction threshold of 50% by weight for Unmilled PSBH composites and 60% by weight for Milled PSBH composites (Fig. 1). In the case of HDPE/Unmilled PSBH and HDPE/Milled PSBH, the filler loading when the first electrical reading was obtained is considered a threshold value. This difference in the conduction threshold stems from the difference in Milled and Unmilled PSBH particle sizes. At such a high loading of fillers, the large particles of unmilled PSBH quickly form a conductive path without even efforts to achieve high dispersion compared to the smaller-sized milled PSBH. Thus, a higher amount of Milled PSBH is needed than Unmilled PSBH to form a conductive path, which leads to an increase in the conduction threshold.

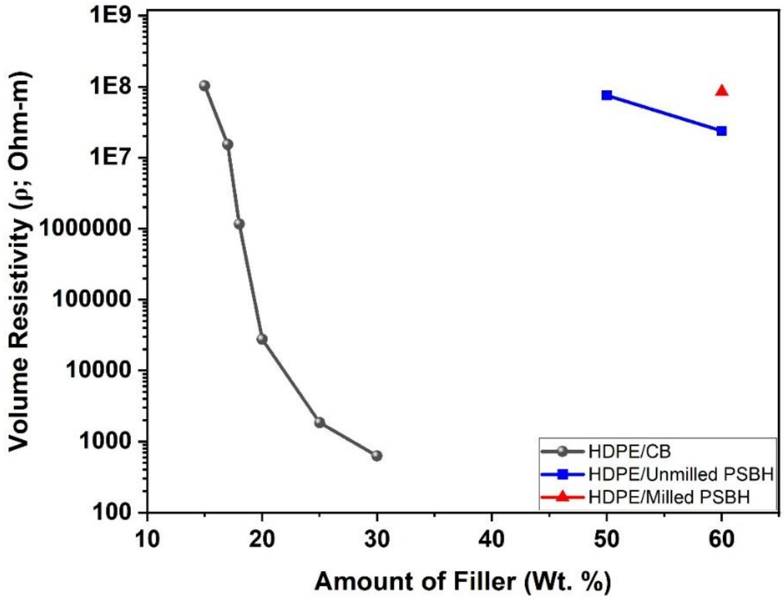

Fig. 1 *Comparison of resistivity (Ohm-m) of HDPE/CB, HDPE/Unmilled PSBH, and HDPE/Milled PSBH composites as a function of the amount of filler (CB or PSBH Wt. %). The far-left data point in each curve represents a conduction threshold*

Using 18% CB, various composites with varying amounts of PSBH (Milled and Unmilled) were prepared, and the effect of adding PSBH to 18% CB on resistivity in HDPE composites was studied in their milled and unmilled forms. Fig. 2 compares the resistivity of HDPE/PSBH/CB composites based on the particle size of PSBH (Unmilled and Milled) and HDPE/CB composites as a function of filler amount.

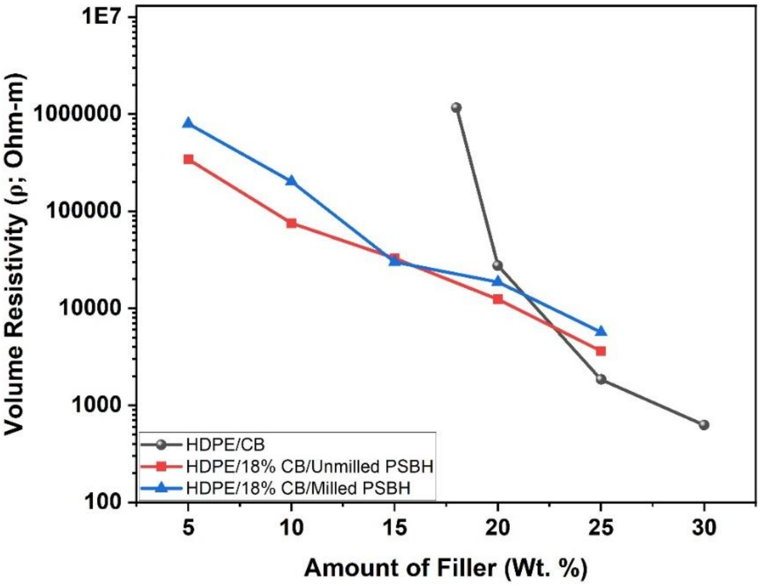

Fig. 2 *Comparison of resistivity (Ohm-m) of HDPE/CB, HDPE/18% CB/Milled PSBH, and HDPE/18% CB/Unmilled PSBH composites as a function of amount of filler (CB or PSBH Wt. %)*

In the presence of 18% CB, Unmilled PSBH composites were more conductive than milled composites. This is due to the low dispersion of Unmilled PSBH. In the milled PSBH composites, due to the small size of milled PSBH, the PSBH is well dispersed along with CB, but apparently, unable to form conductive paths well integrated with CB in comparison to the observed more efficient integration of CB with Unmilled PSBH. Here, by CB-PSBH integration, we refer to the fact that milled or unmilled, the PSBH particles are much larger than the CB particles (Unmilled PSBH >700 µm, Milled PSBH Particle Size is 1-7µm, CB particle size is 26-30nm) [27-30]. The idea is that CB particles' interaction and lining up on PSBH counterparts would increase the formation of conductive paths. The addition of PSBH, in general, improves the conductivity of HDPE/CB composites. Adding PSBH replaces a certain percentage of HDPE, increasing the ratio of CB to HDPE; thus, the sample becomes more concentrated with CB, improving the conductivity. There is also a possibility that PSBH helps with the conduction mechanism, perhaps by interacting with CB, but it is tough to confirm this hypothesis. The conductivity of HDPE/PSBH/CB composites was compared to HDPE/CB composites. Adding 20% PSBH to HDPE/18% CB provided conductivity comparable to HDPE/ 20% CB.

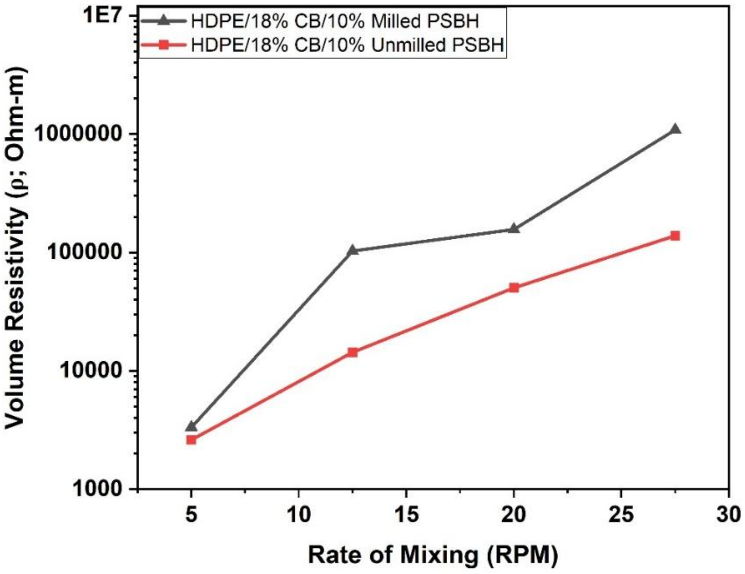

Fig. 3 *Effect of rate of mixing on resistivity (Ohm-m) of HDPE/18% CB/10% Milled PSBH and HDPE/18% CB/ 10% unmilled PSBH composites*

The effect of process parameters, Rate of Mixing (RPM), Mixing time, and Processing Temperature was also studied using HDPE/10% PSBH (Milled and Unmilled)/18% CB composite samples. Different mixing speeds (RPMs), mixing times, and processing temperatures were used, and the resistivities of the resulting composites were measured. Fig. 3 shows the resistivity variation as a function of the Rate of Mixing. The composite's resistivity increases as the mixing rate increases from 5 RPM to 27.5 RPM. When the rate of mixing is increased, the mixing becomes more rigorous. Thus, the dispersion of the fillers is improved, possibly reducing the chances of forming conductive paths and reducing CB-PSBH integration. The resistivity of Unmilled PSBH composites increases by two orders of magnitude. In comparison, milled PSBH increases by three orders of magnitude when the mixing rate increases from 5 RPM to 27.5 RPM. We think this vast difference stems from the difference in particle size of the fillers. Due to its smaller particle size, the dispersion is better for milled PSBH composites than unmilled PSBH, which leads to possible reductions in conductive path formation and CB-PSBH integration.

Fig. 4 shows the variation of resistivity as a function of mixing time. The composite's resistivity increases as the mixing time increases from 5 minutes to 1 hour. Increasing the mixing time produces the same effect as increasing the mixing rate. Longer mixing times improve the dispersion of the fillers within the polymer, which, in our case, leads to an increase in resistivity by four orders of magnitude.

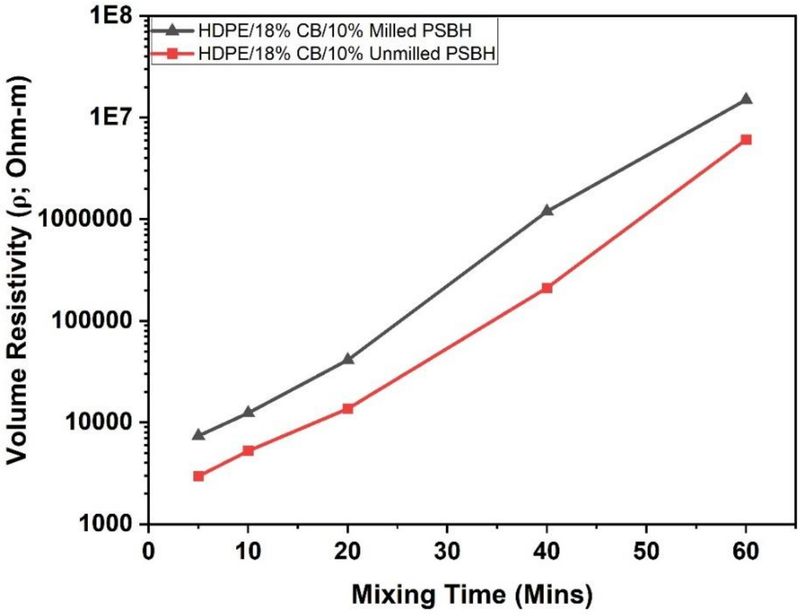

Fig. 4 *Effect of mixing time on resistivity (Ohm-m) of HDPE/18% CB/10% Milled PSBH and HDPE/18% CB/ 10% unmilled PSBH composites*

Fig. 5 shows the resistivity variation as a function of the processing temperature. As with rate or mixing and mixing time, an increasing trend is also observed with processing temperature. We think that the contributing factor is the melting viscosity. The melting temperature of HDPE is 130°C. For the processing temperature of 150°C, the melt viscosity of HDPE is higher than its melt viscosity at 200°C. With more viscous material, mixing becomes difficult, while with higher processing temperatures, the melt viscosity decreases, and the degree of dispersion improves. As stated above, improvements in dispersion cause an increase in resistivity [31].

The data clearly shows that various processing parameters affect the resistivity of composites. The conductivity mechanism in composites is governed by the size and shape of the filler particles, their interactions with each other and with the polymer, and the polymer’s material properties [32]. Unfortunately, the optimum values for the processing parameters are impossible to determine as they exhibit monotonically increasing behavior.

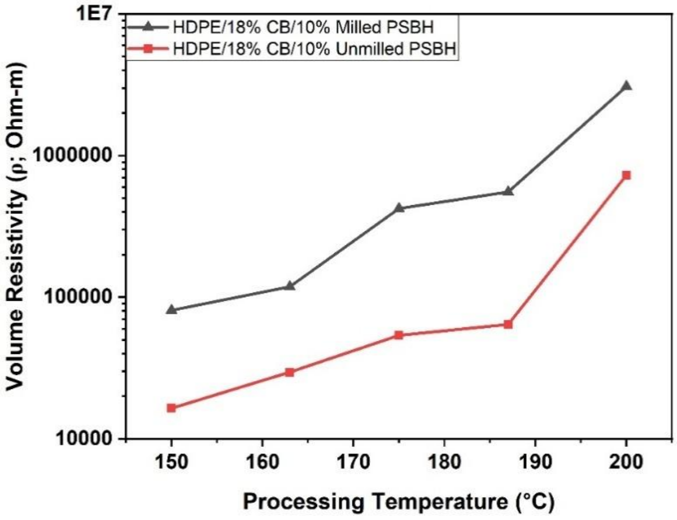

Fig. 5 *Effect of processing temperature on resistivity (Ohm-m) of HDPE/18% CB/Milled PSBH and HDPE/18% CB/ 10% unmilled PSBH composites*

3.2 Mechanical Properties

Fig. 6 illustrates the tensile strength of HDPE/PSBH composites, as well as HDPE/CB composites, produced from both milled and unmilled PSBH. In the case of HDPE/Unmilled PSBH composites, the tensile strength decreases as the filler amount increases. The maximum tensile strength measured for HDPE/milled PSBH is 50.24 MPa at 20% loading, and for HDPE/CB, it is 53.5 MPa at 25% loading, which is considerably higher than the tensile strength of neat HDPE (46.4 ± 0.4 MPa). Thus, CB and Milled PSBH can be potential reinforcement fillers.

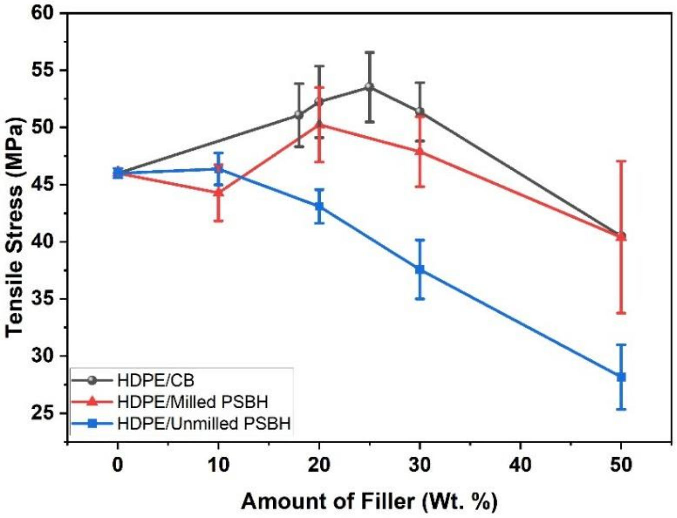

Fig. 6 *Maximum tensile stress (MPa) of HDPE/Unmilled PSBH, HDPE/Milled PSBH, and HDPE/CB composites as a function of the amount of filler (CB or PSBH Wt. %)*

We believe that particle size and filler-polymer interactions affect the tensile strength of the composites. In the case of HDPE/Milled PSBH and HDPE/CB composites, it is observed that the tensile strength of the composites increases with the addition of filler. The tensile strength reaches a maximum value and then decreases upon further addition of filler. Milled PSBH and CB have small particle sizes compared to the Unmilled PSBH. These fillers strengthen the composite when added in modest amounts and have good adhesion to the polymer when they present sufficiently separated reinforcement points that do not interact with each other. The addition of much larger filler particles such as Unmilled PSBH, however, introduces relatively weak reinforcement as they hinder polymer flow around them, causing voids and poor adhesion as well as possible Unmilled PSBH particle deterioration and failure during loading, which may lead to the creation of voids and cracks within the composite. However, the addition of too many small-sized fillers makes the resultant composite exceedingly stiff and brittle, consequently decreasing the tensile properties.

Fig. 7 shows the tensile strength results for the HDPE/ (Milled and Unmilled) PSBH/CB composites as a function of the amount of filler added. The tensile strength increased and decreased as the amount of (Milled or Unmilled) PSBH increased. For HDPE/Milled PSBH/CB, a maximum tensile stress of 57.3 MPa was obtained at 10% filler loading, and a maximum tensile strength of 56 MPa was obtained using 15% Unmilled PSBH loading, both of which were higher than the tensile strength of Neat HDPE (i.e., 46.4 ± 0.4 MPa). When too much filler is added to the composite, it becomes brittle and loses its tensile strength, as observed with the HDPE/PSBH/CB composites.

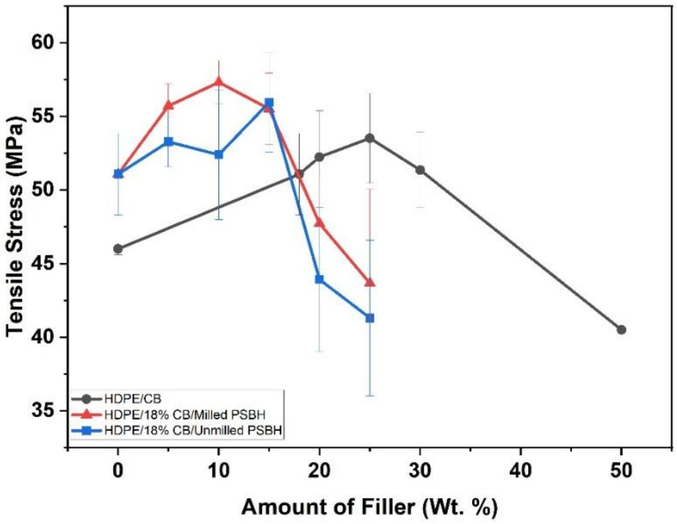

Fig. 7 *Maximum tensile stress (MPa) of HDPE/18%CB/Unmilled PSBH, HDPE/18% CB/Milled PSBH composites as a function of the amount of filler (PSBH Wt. %)*

Fig. 8 shows that strain at break for HDPE/PSBH (Milled and Unmilled) and HDPE/CB composites decreases with increasing filler content. HDPE/Milled PSBH composites show a 20% reduction in strain, while HDPE/unmilled PSBH shows a 76% reduction in strain. This is attributed to filler-polymer interactions. These interactions are more significant in HDPE/milled PSBH composites than in HDPE/Unmilled PSBH composites. Note that the addition of CB gives results closer to the addition of Milled rather than the Unmilled PSBH (Fig. 8).

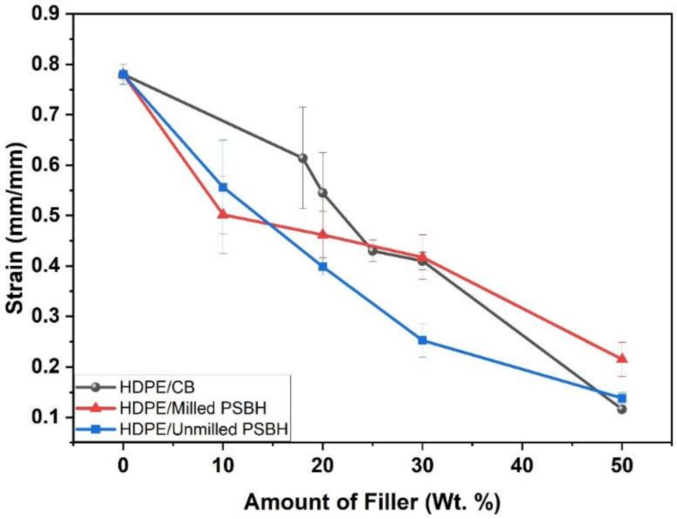

Fig. 8 *Breaking strain (mm/mm) of HDPE/Unmilled PSBH, HDPE/Milled PSBH, and HDPE/CB composites as a function of the amount of filler (Wt. %)*

Fig. 9 shows that the strain at break for HDPE/PSBH/CB composites decreases with increasing filler, and the composites with milled PSBH provide higher break strain values than those with unmilled PSBH.

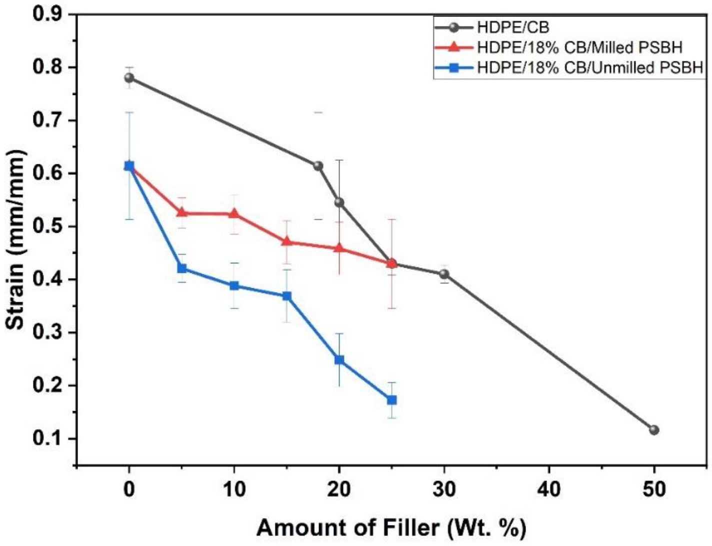

Fig. 9 *Breaking strain (mm/mm) of HDPE/18%CB/Unmilled PSBH, HDPE/18% CB/Milled PSBH composites as a function of the amount of filler (Wt. %)*

Fig. 10 shows the tensile moduli of HDPE/Unmilled PSBH, HDPE/Milled PSBH, and HDPE/CB composites as a function of filler amount. For all the composites, the modulus increased as the filler increased. HDPE/Unmilled PSBH showed an increase in modulus by 150%, while HDPE/Milled PSBH showed an increase of 250% compared to Neat HDPE. In the case of HDPE/CB, the increase was 280.5%.

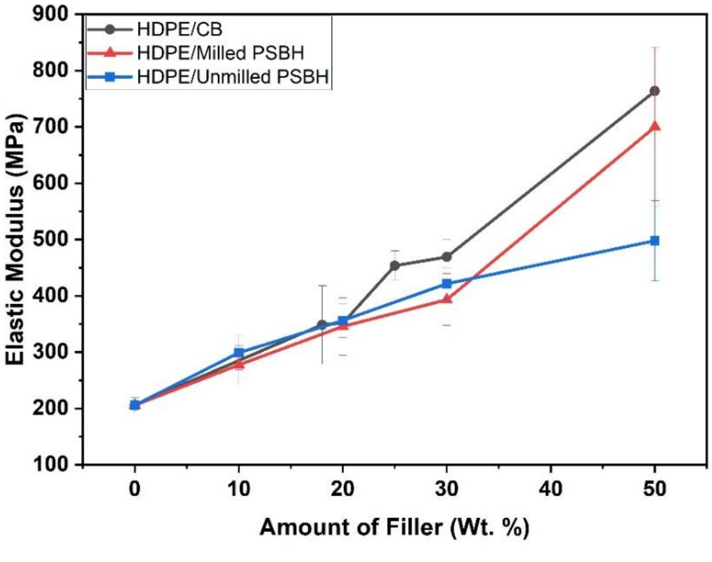

Fig. 10 *Modulus of elasticity (MPa) of HDPE/Unmilled PSBH, HDPE/Milled PSBH, and HDPE/CB composites as a function of the amount of filler (Wt. %)*

Fig. 11 shows the moduli values for HDPE/Milled PSBH/CB composites as a function of filler amount. The modulus increased as the amount of PSBH increased for all the composites. HDPE/Unmilled PSBH/CB showed higher moduli than milled PSBH. In the case of HDPE/Unmilled PSBH/CB, the modulus increased by 182.5%, and for HDPE/Milled PSBH/CB, the modulus increased by 161% compared to Neat HDPE.

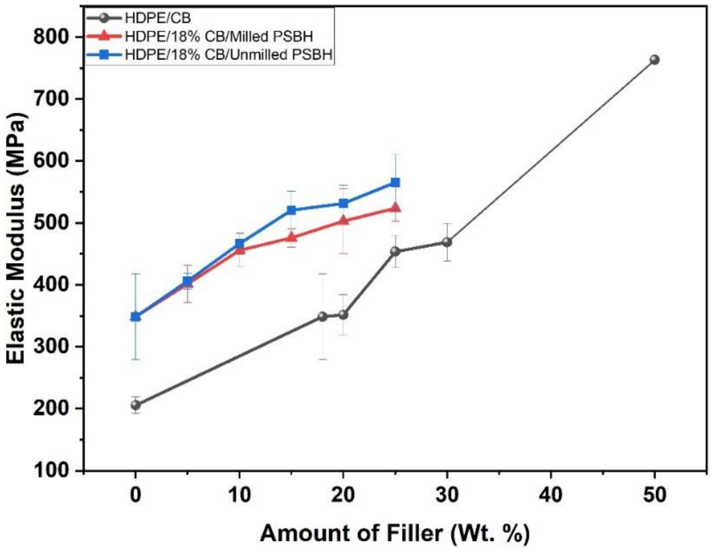

Fig. 11 *Modulus of elasticity (MPa) of HDPE/18%CB/Unmilled PSBH, HDPE/18% CB/Milled PSBH composites as a function of the amount of filler (Wt. %)*

Fig. 12 shows the SEM image of a fractured cross-section for HDPE/18% CB composite. In the image, we can see the fiber-like deformed polymer structures at the mostly homogeneous fracture surface. A particle is well embedded into the polymer, indicating good interaction. This particle is an agglomerated form of various small CB particles, which could indicate poor CB dispersion.

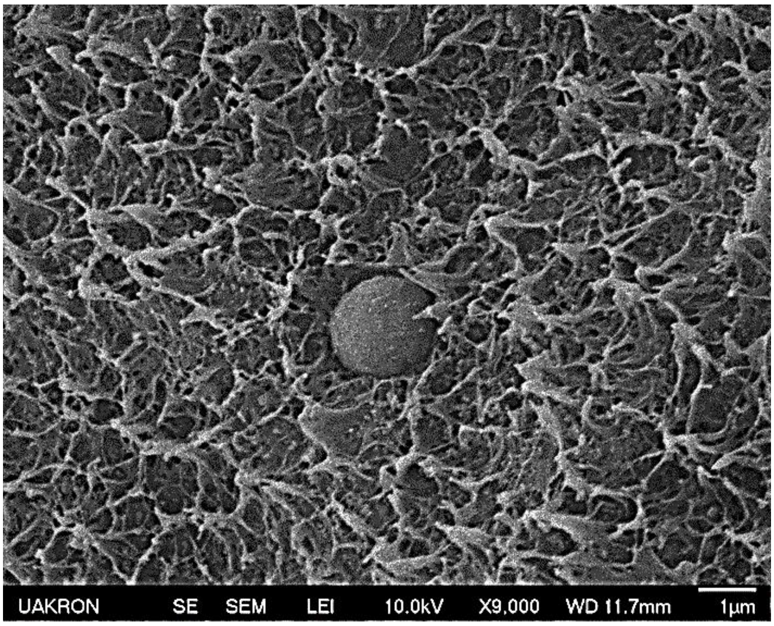

Fig. 12 *SEM image of fractured surface of HDPE/18% CB composite at X9000 magnification at 10KV applied Voltage*

Fig. 13 shows a fractured cross-section for HDPE/ 20% Unmilled PSBH composite. Unmilled PSBH particles larger than 1 micron and smaller particles are visible in the image. The existence of the smaller particles confirms that the mixing process breaks down some of the bigger particles into smaller ones. This finding is very important as this only happens due to the mixing operation and the shear force exerted by the molten polymer. In various places, voids and cracks are also seen, which suggests poor filler-polymer interactions and poor wetting of the filler particles.

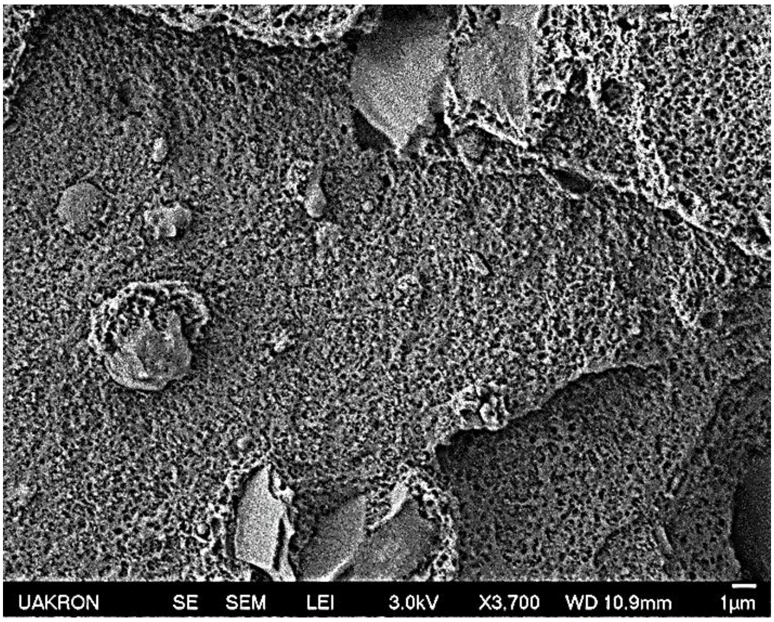

Fig. 13 *SEM image of fractured surface of HDPE/20% Unmilled PSBH composite at X3700 magnification at 3KV applied voltage*

Fig. 14 shows a fractured cross-section for HDPE/18% CB/10% Unmilled PSBH composite. We can see large, unmilled PSBH particles and smaller particles. CB particles are nanosized and are not visible in this image, but we can see a few clusters of particles that may be CB. This image confirms the poor consolidation of unmilled PSBH into the composite.

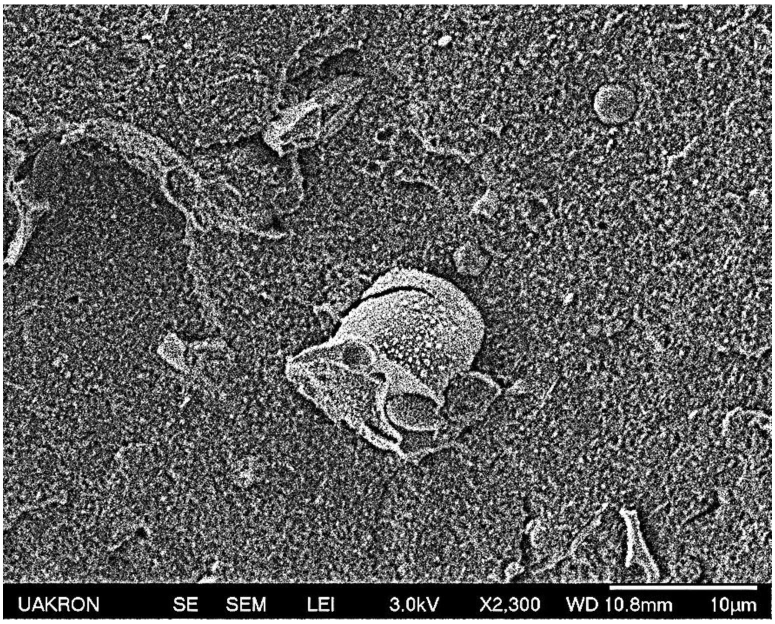

Fig. 14 *SEM image of fractured surface of HDPE/18% CB/10% Unmilled PSBH composite at X2300 magnification at 3KV applied voltage*

Fig. 15 shows a fractured cross-section for the HDPE/18% CB/Milled PSBH composite. The image shows good dispersion and distribution of PBSH. CB cannot be seen since it is nanosized and homogeneous with HDPE. This image confirms the dispersion of PSBH and thus confirms the hypothesis that better dispersion of milled PSBH causes the resistivity of the HDPE/18% CB/Milled PSBH to be lower than HDPE/18%CB/unmilled composites.

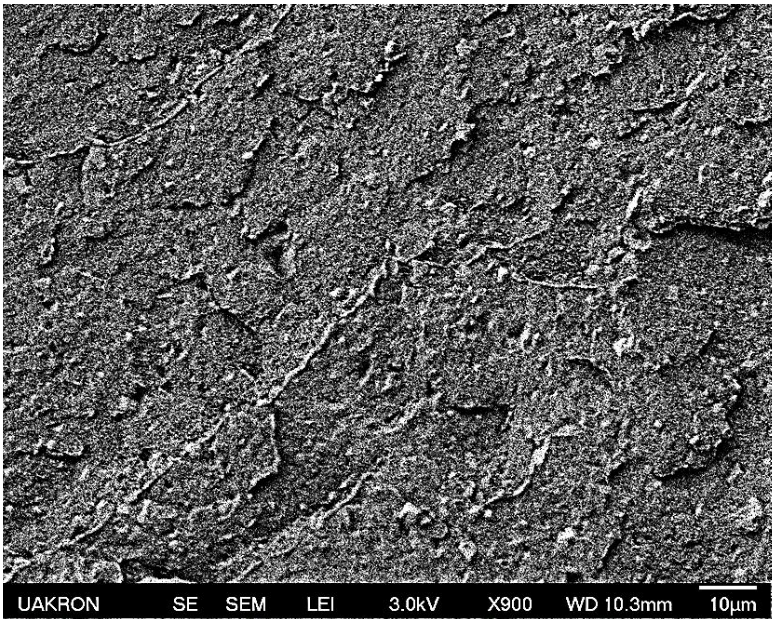

Fig. 15 *SEM image of fractured surface of HDPE/20% CB composite at X900 magnification at 3KV applied voltage*

These SEM images also confirm the effect of PSBH and CB particle size. Cracks and voids were seen in the fracture images of composites with unmilled PSBH, suggesting poor filler-polymer interactions. It was also found that the mixing process causes the particle aggregates to break into smaller particles. This was observed in HDPE/unmilled PSBH composites.

Fig. 16 and Fig. 17 show micro CT X-ray topographical images of HDPE / 18% CB / 10% Unmilled PSBH and HDPE / 18% CB / 10% Milled PSBH samples compounded at various processing conditions. The effect of processing parameters on the composite structure was studied using X-ray tomography. In Figures 16 and 17, a and b samples were compounded at different melting temperatures, i.e., 150°C and 200°C at 20 RPM for 30 mins; c and d were compounded at different mixing times of 5 mins and 60 mins at 175°C and 20 RPM, e and f were compounded at 175°C for 30 mins at 5 and 27.5 RPM.

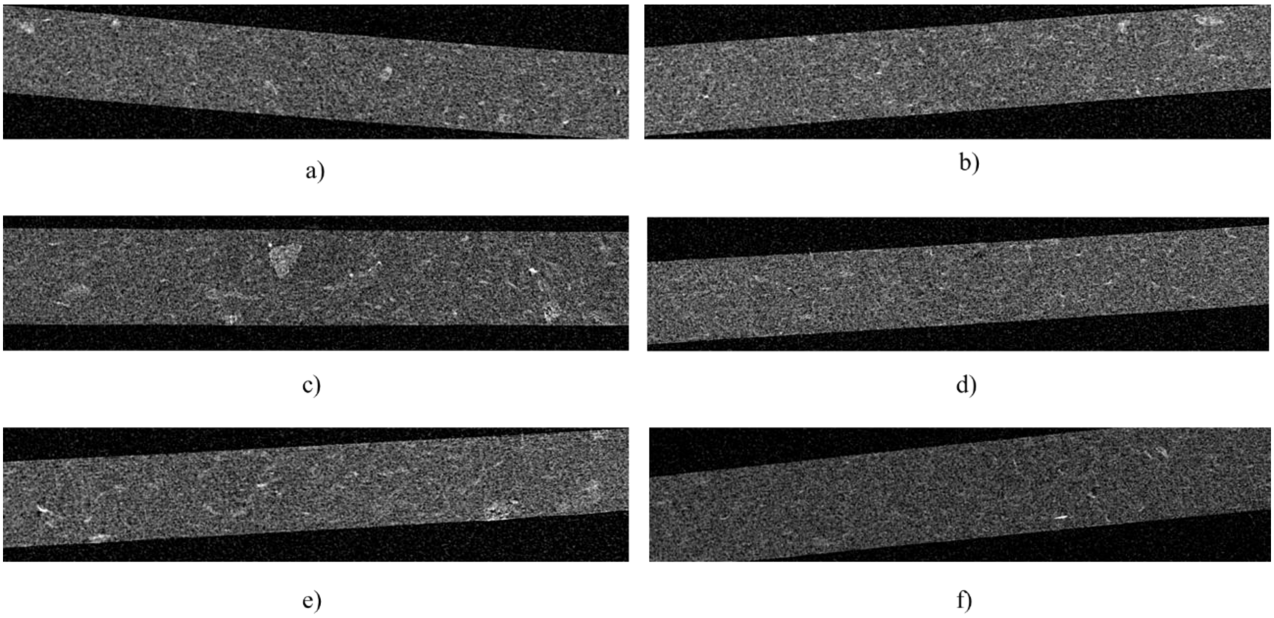

Fig. 16 *Micro CT X-ray tomography images of HDPE / 18% CB / 10% Unmilled PSBH samples (a) Melting temperature 150°C (b) Melting temperature 200°C (c) Mixing time 5 mins (d) Mixing time 60 mins (e) Mixing speed 5 RPM (f) Mixing speed 27.5 RPM*

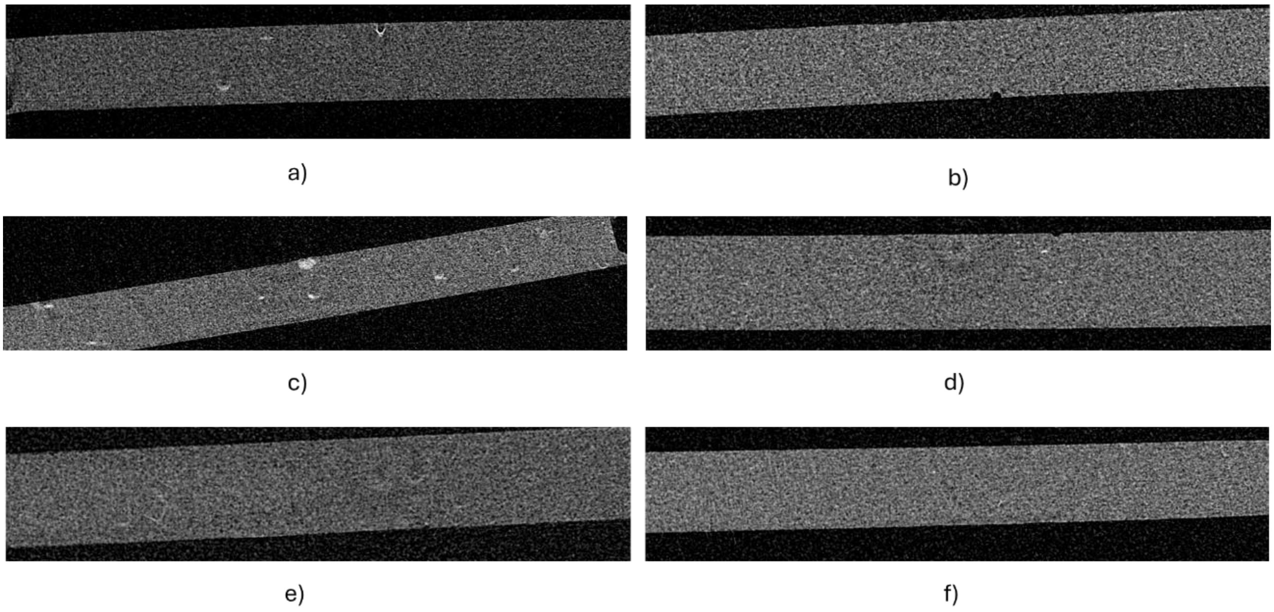

Fig. 17 *Micro CT X-ray tomography images of HDPE / 18% CB / 10% Milled PSBH samples. (a) Melting temperature 150°C (b) Melting temperature 200°C (c) Mixing time 5 mins (d) Mixing time 60 mins (e) Mixing speed 5 RPM (f) Mixing speed 27.5 RPM*

The scan images show an increase in dispersion as the melting temperature increases from 150°C to 200°C. They also show a significant reduction in agglomerated particles and increasing dispersion as the mixing time increases from 5 to 60 minutes. A similar trend was observed as the mixing rate increased from 5 to 27.5 RPM. These trends are analogous to the electrical conductivity results. As the dispersion of fillers improved, the integration of CB and PSBH increased, which led to drastic changes in electrical conductivity, as seen in Fig. 3, Fig. 4, and Fig. 5. It is clear from the images that Carbon Black (CB) and PSBH integrate well together.

4. Conclusion

This study investigated the effects of PSBH as a conductive filler in HDPE/CB composites, focusing on electrical conductivity, mechanical behavior, and processing parameters. The electrical percolation threshold for HDPE/CB was found to be 18% wt. Incorporating PSBH improved composite conductivity, with unmilled PSBH exhibiting better conductive performance due to enhanced CB integration. In contrast, milled PSBH resulted in superior mechanical properties, demonstrating a trade-off between conductivity and mechanical strength. The effect of processing parameters such as mixing speed, mixing time, and processing temperature was also evaluated, revealing that optimized processing conditions play a crucial role in filler dispersion and composite performance. These findings highlight PSBH's potential as a bio-based conductive filler, thereby contributing to the development of sustainable polymer composites. Future work should investigate advanced dispersion techniques and evaluate their long-term durability for real-world applications.

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Conflict of Interest

Authors declare that there is no conflict of interests regarding the publication of the paper.

Author Contribution

The authors confirm their contributions to the paper as follows: ***study conception and design****: Udayan Jayant Dabke and Erol Sancaktar;* ***data collection****: Udayan Jayant Dabke;* ***analysis and interpretation of results****: Udayan Jayant Dabke and Erol Sancaktar;* ***draft manuscript preparation****: Udayan Jayant Dabke and Erol Sancaktar. All authors reviewed the results and approved the final manuscript version.*

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