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## The investigation of the properties of filaments fabricated from carbon biomass and LLDPE

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### Abstract

This study aims to develop composites using electrically conductive carbon and polymer polyethylene (LLDPE) to enhance electrical conductivity. Investigations have been conducted on the fabrication of electrically conductive composites and the modulus of elasticity through heat compaction using mixtures of carbon-LLDPE powders. Heat compaction is performed at temperatures ranging from 120°C to 150°C, with varying composition ratios of carbon-LLDPE, including 50:50, 60:40, and 70:30 % wt. Higher proportions of carbon and compaction temperatures are correlated with increased electrical conductivity. For instance, the C7-3LLDPE composite, compacted at 150°C, demonstrates the highest electrical current flow of 0.0018 A, whereas the C5-5LLDPE composite, compacted at 135°C, exhibits the lowest current flow at 0.0000638 A. Regarding the modulus of elasticity, the composition ratio of C7-3LLDPE, compacted at 120°C, achieves the highest value at 2686.43 [N/mm<sup>2</sup>]. Conversely, the composition ratio of C5-5LLDPE, compacted at 135°C, yields the lowest modulus of elasticity at 1530.94 [N/mm<sup>2</sup>]. Elasticity modulus testing follows the ASTM D638 standard, with a speed of 2 mm/min. It is observed that increasing the compaction temperature results in a decreased modulus of elasticity across all composition ratios. Furthermore, a higher carbon content within the composite corresponds to a higher modulus of elasticity, regardless of the compaction temperature.

### Keywords:

Carbon-LLDPE composite, electrically conductive, heat compaction, modulus of elasticity, rice husks carbon.

### 1 Introduction

This paper investigates the electrical and mechanical properties of filament materials made from biomass-derived carbon particles embedded in an LLDPE matrix. Filaments, which are slender threads or fibers, are utilized in a wide range of fields, including textile production, plastic manufacturing, and not particularly 3D printing. The main aim of this study is to develop composites that employ electrically conductive carbon and LLDPE polymer to facilitate electrical conductivity.

The carbon material derived from rice husk waste exhibits electrical conductivity and acts as a filler within a Linear Low-

Density Polyethylene (LLDPE) matrix. The rheological properties of LLDPE are advantageous for promoting flow within the gaps between conductive carbon particles and binding these particles together, ensuring a sufficient volume of interconnected carbon particles for electrical conduction. LLDPE possesses distinct rheological and melting flow characteristics that contribute to this process.

LLDPE has higher viscosity and therefore lower sensitivity to shearing during processes such as extrusion, enabling faster relaxation of stress from polymer chains. This property allows for quicker stress relief during extrusion. The rheological properties of LLDPE can be described as "rigid in shear" and "soft in extension".

LLDPE is used in various recyclable products such as bin liners, substitutes for wood, landscaping materials, floor tiles, compost bins, and shipping envelopes. This article represents the culmination of research that utilizes LLDPE as a matrix for composites, incorporating carbon powder fillers to enhance electrical conductivity.

For several decades, researchers have extensively studied electrically conductive polymer-based composites. The attractive structural and electrical properties of conductive polymers have garnered significant interest in organic composites and thermoelectric carbon nanoparticles (TE) or hybrid materials [1]. The ability of electrically conductive polymer composites to bend and stretch is a crucial characteristic for applications such as flexible wiring and electronics. One notable advancement in this field is using a 3D graphene network, which differs from conventional graphene powder and film. This 3D network serves as a filler instead of traditional powder fillers, contributing to developing Conductive Polymer Composites (CPCs) [2]. This study examines the heat dissipation characteristics of Cu-plated PEEK/carbon fiber felt (PEEK/CF-Cu) e-composite materials. The research utilizes Cu electroplating networks on a 3D PEEK/CF framework to develop efficient heat dissipation solutions for miniature electronic devices. The objective of this approach is to improve the heat dissipation capabilities of polymer-based composites, catering to the specific needs of compact electronic devices [3]. Poly (methyl methacrylate) carbon fibre composites with controlled aspect ratios are manufactured at different concentrations and heat-pressed into rectangular sheets of varying thicknesses. The orientation of carbon fibres on both the surface and within the specimen is determined, and the conductivity ( $\sigma$ ) is investigated along three different directions: X-length, Y-width, and Z-thickness [4]. This article presents a novel approach combining a double percolated structure, supercritical (scCO<sub>2</sub>) treatment, and foaming with carbon dioxide annealing to fabricate a composite foam consisting of polystyrene (PS), poly (methyl methacrylate) (PMMA), and multi-wall carbon nanotubes (MWCNTs). This innovative method yields porous composite foams with a double-percolated structure, offering enhanced electrical conductivity. The study introduces an environmentally friendly and adaptable technique for producing porous Conductive Polymer Composites (CPCs) with improved electrical properties [5]. Carbon fibre-reinforced plastics have become preferred over metal components in structural applications within sports equipment, the aerospace industry, and automotive sectors, primarily due to their favourable performance-to-weight ratio. A composite consisting of polyamide 66 (PA66) and Carbon Fibre (CF), integrated with Graphene Oxide (GO) via acyl-chloride and Carbon Nanotubes (CNTs) as hybrid fillers, is fabricated using in-situ interface polymerization [6]. This study investigates the impact of various carbon-based fillers on the performance of electrically conductive polymer mixture composites. Specifically, it focuses on comparing the effects of graphene, carbon nanotubes, and carbon blacks on PC/ABS matrices through morphological analyses and electrical and physics-mechanical characterizations. [7].

The electrical conductivity of nanocomposites demonstrates a notable enhancement upon the incorporation of 2.5 wt.% of Multi-Walled Carbon Nanotubes (MWCNT) in comparison to unfilled mixtures. Nevertheless, the dependence of conductivity on the MWCNT content diminishes beyond this threshold. Rheological characterization reveals that the normalized relaxation time increases up to 5 wt.% of nanofillers and remains relatively constant for higher concentrations of MWCNT [8]. This study details the synthesis of Poly (Monomethyl Itaconate) grafted multi-layer Graphene Oxide (PMMI-g-GO) through Atomic Transfer Radical Polymerization (ATRP) employing Activators Regenerated by Electron Transfer (ARGET). PMMI-g-GO exhibits remarkable characteristics, including high electrical conductivity [9]. Electrically conductive polymer composites are fabricated through the implementation of melt mixing techniques, utilizing polyester-based Thermoplastic Polyurethane (TPU) matrices. These matrices are combined with fillers such as Polypyrrole (PPy) or Montmorillonite/Polypyrrole (MMT/PPy), which are doped with dodecylbenzene sulfonic acid. Several composites are prepared, each containing different mass fractions of conductive additives. These composites are then compared in terms of their electrical properties, structure, and morphology. The objective of this evaluation is to determine the impact of both the content and type of conductive filler on the properties of the composites [10]. Rubber-based conductive polymer composites offer remarkable deformability and flexibility, making them highly suitable for applications in Electromagnetic Interference (EMI) shielding materials.

In this study, a comprehensive investigation is conducted to examine the properties of electrical conductivity, mechanical strength, and EMI shielding effectiveness by incorporating various carbon fillers with different dimensions individually and in combination into Isoprene Rubber (IR). These carbon fillers include zero-dimensional carbon black, one-dimensional carbon nanotubes, and two-dimensional graphene. The goal of the study is to create flexible EMI shield composites [11].

This study utilizes simultaneous electrical rheological measurements to examine the structural changes caused by shear deformations in conductive polymer composites with carbon fibre or carbon black. It is the first comprehensive investigation of the electrical properties of composites with anisotropic micro-fillers during deformation in a liquid state. The results demonstrate that composites containing carbon fiber exhibit a significant susceptibility to mechanical deformations, as evidenced by their high sensitivity in electrical conductivity [12]. Electrically conductive composites have been engineered by incorporating conductive fillers, including intrinsic conduction polymers and carbon fillers, into the matrices of insulating polymers. These insulating polymers encompass various types, such as thermoplastics, thermoset polymers, and unsaturated rubbers [13]. Incorporating a conductive carbon filler into thermoplastic resins enhances composites' electrical and thermal conductivity. This study investigated the effects of three carbon variants (carbon black, synthetic graphite particles, and carbon fibre) added to Vectra A950RX Liquid Crystal Polymer [14]. Carbon-based composites find extensive applications, particularly in bipolar plate polymer composites. This study aimed to explore the suitability of milled carbon fibre as a conductive filler in composites and to adapt the General Effective Media model for predicting the electrical conductivity of resulting polymer composites. Polymer composites were fabricated with different concentrations of conductive filler loading mixed with epoxy resins using compression moulding techniques [15]. The composite was formed by incorporating conductive microcarbon from rice husks into a Linear Low-Density Polyethylene (LLDPE) polymer matrix through hot compaction. Various filler composition combinations were utilized, with carbon loads

ranging from 50% to 40% and mesh sizes including #150, #200, and #250.

The experimental results demonstrated that variations in particle size had negligible impacts on the density of the composite. Nonetheless, the utilization of finer mesh sizes enhanced the dispersion of the filler within the matrix, leading to increased levels of electrical conductivity [16].

This study aimed to manufacture carbon-Phenol Formaldehyde (PF) composites using carbonized rice husks as fillers. These composites exhibited improved electrical conductivity, increased carbon content, and reduced electrical resistance. For example, specimens consisting of 80% carbon by weight and 20% PF by weight demonstrated an electrical conductivity of 0.055 [S/cm]. In contrast, those comprising 70% carbon by weight and 30% PF by weight displayed a conductivity of 0.039 [S/cm]. Furthermore, specimens containing 60% carbon by weight and 40% PF by weight resulted in an electrical conductivity of 0.013 [S/cm] [17]. According to the evaluation of research journals, this article offers an alternative approach to engineering linear low-density polyethylene thermoplastic composites by incorporating electrically conductive carbon fillers derived from rice husks.

## 2 Experiment

### 2.1 Material Preparation

Carbonization of rice husk by pyrolysis technique using an airtight furnace at a temperature of 950°C and at a rate of 2°C/min produces carbon electrically conductively. Carbon milling with a ball milling machine and carbon sieving with a sieve machine to reach a mesh size 150, the carbon density showed  $1.3794 \pm 0.12$  [grams/cm<sup>3</sup>]. Dry Linear-Low Density Polyethylene (LLDPE) powder was obtained from the online commercial market with a mesh size of 60, nominal density according to ASTM D 1505 = 938 [kg/m<sup>3</sup>] or 0.938 [grams/cm<sup>3</sup>], melting point according to the standard ISO 3146 = 124°C, and tensile yield strength based on ASTM D 638 = 21 [MPa], and tensile break strength = 18 [MPa].

### 2.2 Sample Fabrication

Carbon and LLDPE were mixed in composition ratio 50:50, 60:40, and 70:30 % weight, indicated in sequential codes C5-5LLDPE, C6-4LLDPE, and C7-3LLDPE. Moulding is carried out in hot compaction moulds at temperature variations of 120°C, 135°C, and 150°C, and produced composite rod samples for tensile testing and electrical properties test samples, square shapes with sizes  $\pm 10 \text{ mm} \times 10 \text{ mm} \times 5 \text{ mm}$ .

### 2.3 Electric Current Testing

Testing of electrically conductive properties was carried out using the four-point probe method. Testing electrical properties with the four-point probe method is commonly used to measure a material's resistivity or electrical conductivity. The resistivity of the material ( $\rho$ ) can be calculated using the Eq. 1.

$$\rho = \frac{V}{I} \times \frac{\pi}{\ln(2)} \times t \quad (1)$$

Where  $V$  is the voltage measured,  $I$  is the current that flows and  $t$  is the thickness of the material if the measurement is made on a thin film.

The tests are to measure voltage, electric current, and electrical resistance with Keithley Instruments, the 2450 Source Meter® Instrument, at the Research Centre for Physics, National Research, and Innovation Agency (BRIN), Serpong, Indonesia. Electrical properties testing follows the ASTM D4496 standard. For comparison, some studies conduct testing of electrical properties in the same way [1, 10, 13, 14, 15, 16, 17, 18, 19, 20].

### 2.4 Elasticity Modulus Testing

Elasticity modulus testing follows the ASTM D638 standard, with a 2 mm/min speed, AGS-X Testing Machine, and 10000 N



capacity. The test was conducted at the Research Center for Biomass and Bioproduct, National Research and Innovation Agency (BRIN), Cibinong, Bogor, Indonesia. Other researchers also applied the same standards to determine tensile strength [15].

## 2.5 Morphology Testing

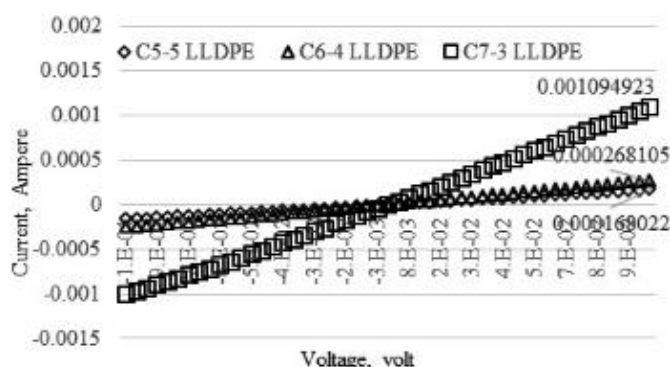
Morphology testing was conducted with FESEM-EDS at the Research Center for Biomass and Bioproduct, National Research and Innovation Agency (BRIN). Tests are carried out to ensure the formation of porosity cavities in the composite.

## 3 Results and Discussion

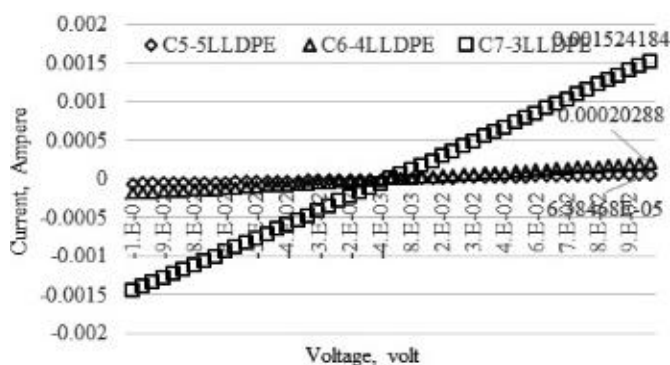
### 3.1 Electrical Properties

The electric current was measured using the four-point probe method. This method involves placing four probe needles on the surface of the material being tested: two outer needles conduct current and two inner needles measure the voltage produced. The voltage was gradually increased during testing at a rate of 0.004 volts per minute for 54 minutes. The voltage range used was -0.1 volts to 0.1 volts.

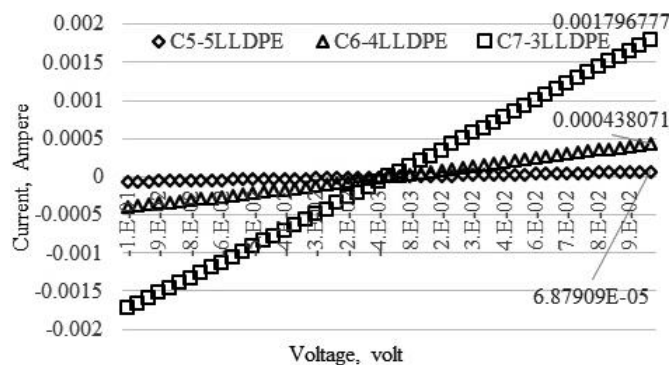
The test results showed a change in electric current from -0.001 A to 0.0011 A, as depicted in Fig. 1(a). Subsequent tests for the C7-3LLDPE sample at a compaction temperature of 135°C, shown in Fig. 1(b), resulted in a change in electric current from -0.0014 A to 0.0015 A. For samples with a compaction temperature of 150°C, as illustrated in Fig. 1(c), there was a change in current from -0.0017 A to 0.0018 A in the C7-3LLDPE samples.



(a) Hot compaction 120°C



(b) Hot compaction 135°C



(c) Hot compaction 150°C

Fig. 1. Electric current and voltage of C-LLDPE composites.

The results of the electric current test indicate a direct correlation between the carbon content and the flow of electric current, with higher carbon content resulting in increased current flow. Additionally, the compaction temperature of the composite significantly influences the enhancement of electric current flow, with higher compaction temperatures leading to increased current flow, as depicted in Fig. 1. This phenomenon is believed to be attributed to the rheological behavior of the plastic flow, which melts more uniformly at higher temperatures, thereby enhancing the bonding between carbon particles. This increase in electric current flow due to elevated compaction temperatures was observed across all samples in the study.

Specifically, the sample coded as C7-3LLDPE, with a carbon weight content of 70% and LLDPE weight content of 30%, exhibited the highest electric current flow value of 0.0018 A at a compaction temperature of 150°C. Conversely, the lowest electric current flow was observed in the C5-5LLDPE sample at a compaction temperature of 135°C, measuring 0.000638 A, as illustrated in Fig. 2. The inset figure in Fig. 2 further illustrates this trend in electric current flow among composite samples, showing that higher carbon weight content and compaction temperatures result in increased current flow.

Overall, most studies focus on the electrical conductivity of polymer composites, where a higher electric current indicates higher electrical conductivity in the composite. Some previous studies have specifically addressed the electrical conductivity of carbon polymer composites [7, 11, 19, 20].

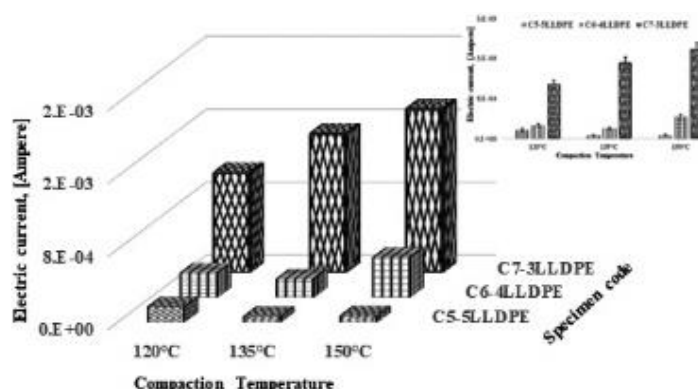


Fig. 2. Electric current vs. compaction temperature vs. sample code.

### 3.2 Modulus of Elasticity

The modulus of elasticity property of the carbon-LLDPE composite is a crucial tensile mechanical property. This information is essential for understanding the mechanical behavior of the material, particularly in the context of producing plastic cables for electricity. The modulus of elasticity, also known as Young's modulus, represents the ratio of stress to percent strain within the elastic zone of the material. In the elastic zone, the relationship between stress and strain is linear, forming a straight line on the tensile test graph, as shown in Fig. 3. Understanding this relationship is vital for ensuring the desired mechanical performance and durability of plastic cables used in electrical applications. The figure compares tensile test curves for C5-5LLDPE composites at compaction temperatures of 120°C, 135°C, and 150°C. The right triangle in the figure illustrates the comparison of elastic modulus values in the C5-5LLDPE composite. At a compaction temperature of 120°C, the composite exhibits an elastic modulus of 1756.34 [N/mm<sup>2</sup>], while compaction at 135°C results in a modulus of 1530.94 [N/mm<sup>2</sup>], and compaction at 150°C yields a modulus of 1686.24 [N/mm<sup>2</sup>].

The graph in Fig. 3 illustrates that compaction at 120°C generates the highest curve compared to other graphs, indicating the highest modulus of elasticity in all C-LLDPE composition ratios. As shown in Fig. 4, the highest modulus of elasticity recorded is 2686.43 [N/mm<sup>2</sup>], achieved by the composition ratio

of C7-3LLDPE (70% carbon weight and 30% LLDPE weight) at a compaction temperature of 120°C. Conversely, the composition ratio of C5-5LLDPE at a compaction temperature of 135°C exhibits the lowest modulus of elasticity, measured at 1530.94 [N/mm<sup>2</sup>].

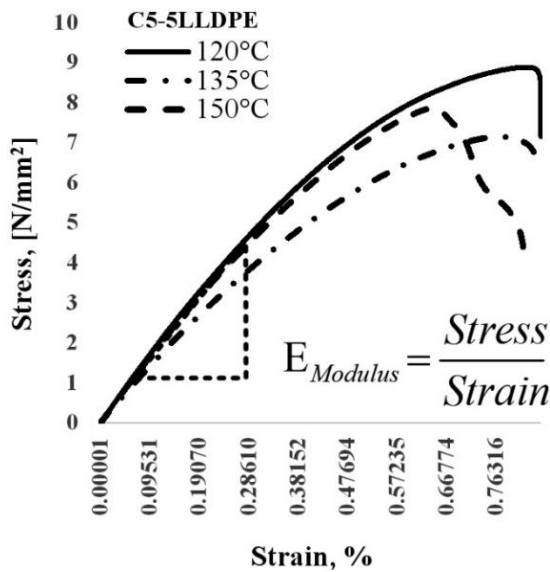


Fig. 3. Comparison of stress-strain curve.

Furthermore, it has been observed that higher compaction temperatures result in a lower modulus of elasticity in the Carbon-LLDPE composite across all composition ratios. This trend is consistent with a decrease in the composite's strength. Additionally, Fig. 4 demonstrates that increasing the carbon content within the composite enhances the modulus of elasticity, while increasing the LLDPE content does not significantly affect it. The insert image in the upper right corner of Fig. 4 illustrates this observation, along with the standard deviation.

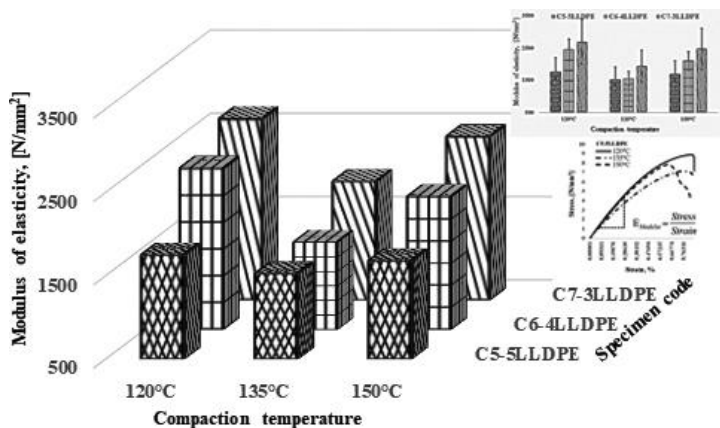


Fig. 4. Elasticity modulus vs. temperature compaction vs. sample code.

These findings can be attributed to the formation of particle attachments and clusters due to Van der Waals attraction between filler particles and their aspect ratios, as well as the high surface area of the particles. This agglomeration, combined with relatively weak interfacial interactions between carbon particles and LLDPE, can reduce load transfer efficiency, thereby decreasing the tensile strength and modulus of elasticity in the composite. The "Payne effect" refers to a phenomenon observed in filled polymer systems, where the addition of filler particles reduces the dynamic properties of the material, especially under high strain amplitudes. This effect is typically attributed to several factors, including the interaction between filler particles (such as agglomeration and deagglomeration of the filler), interactions between the filler and the polymer matrix, and the coupling of polymer chains to the filler surface.

Essentially, the Payne effect results from the complex interplay between filler particles, the polymer matrix, and the polymer-filler interface. The agglomeration of filler particles can give rise to filler networks within the polymer matrix, restricting the mobility of polymer chains and hindering the material's response to dynamic loading. Moreover, interactions between the filler and polymer matrix can influence the overall mechanical behavior of the composite material. Additionally, the attachment of polymer chains to the filler surface can modify the material's dynamic properties by altering chain mobility and stress distribution within the composite.

In conclusion, the Payne effect is crucial in the design and characterization of filled polymer composites as it has a profound impact on the material's mechanical performance and dynamic behavior.

This phenomenon is influenced by various parameters, including concentration, particle size, surface characteristics, and temperature. It is important to note that the dispersion state of carbon particles within LLDPE may fluctuate during experimental studies due to the instability in polymer phase morphology [8]. For comparison, some studies also test the modulus of elasticity of composites with different fillers [21, 6, 7, 22, 23].

### 3.3 Relationship of Modulus Elasticity and Electrical Current

Studying the correlation between the modulus of elasticity and electric current aims to demonstrate the potential use of this engineering material as a medium for electrical conduction in plastic applications. Carbon-LLDPE composites designed for electrical current conduction will be subjected to tensile loads when integrated into electrical networks, simultaneously serving as pathways for electric current flow. Analysis of this relationship aims to elucidate observed trends.

In the case of the C5-5LLDPE specimen, it is observed that the modulus of elasticity tends to decrease with higher compaction temperatures; similarly, the electric current flow tends to decrease, as depicted in Fig. 5. Fig. 5 also illustrates the trend observed in the C6-4LLDPE specimen, where increasing compaction temperature leads to a decrease in modulus of elasticity and an increase in electric current flow.

This trend is also observed in the C7-3LLDPE specimen. The tendency for increased electric current flow in the C6-4LLDPE and C7-3LLDPE specimens is attributed to their higher carbon content. Conversely, the decrease in modulus of elasticity in these specimens is attributed to the formation of numerous porosity cavities at higher compaction temperatures.

Higher compaction temperatures weaken the bond between filler carbon particles and LLDPE, consequently reducing the strength of tensile load transfer and the modulus of elasticity.

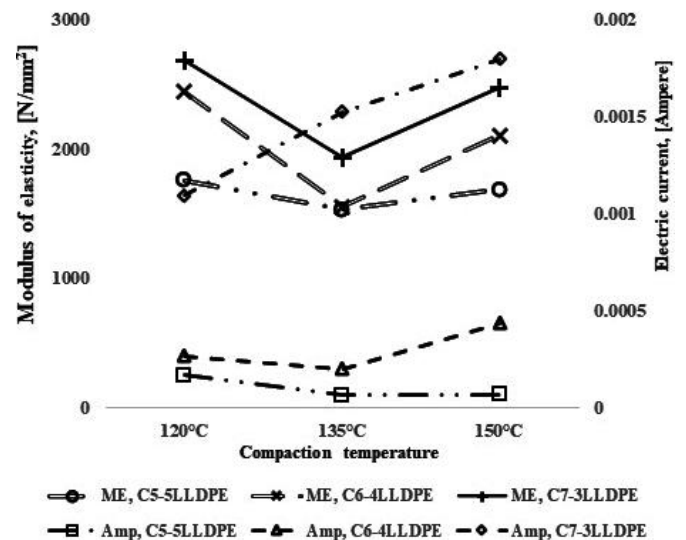


Fig. 5. The relationship of compaction temperature vs. elasticity modulus vs. electrical current.



### 3.4 Composite Morphology

Morphological analysis is performed using FESEM-EDS to determine the presence of porosity and cavities that affect both the modulus of elasticity and electrical conductivity. The morphological feature profile of the carbon-LLDPE composite is shown in Fig. 6. Suspected porosity cavities are highlighted in black, revealing prominent deep holes. Additionally, features resembling plastic melt flow, depicted in grey-white, are believed to represent the LLDPE plastic matrix within the composite. Specifically, Fig. 6(a) presents the morphological profile of the C5-5LLDPE composite compacted at 120°C. This study focuses exclusively on the morphology of the C5-5LLDPE composite, which is compacted at temperatures of 120°C, 135°C, and 150°C.

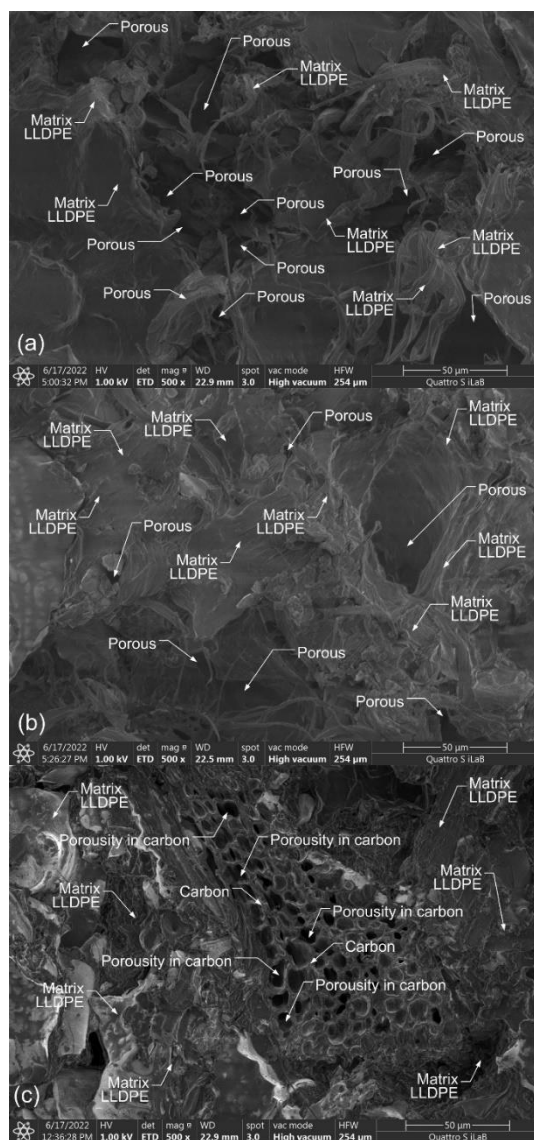


Fig. 6. Morphology of C5-5LLDPE composite.

Fig. 6(b) displays prominent features that resemble the flow of melted plastic, which can be attributed to the LLDPE matrix. The black areas in the figure are believed to be porous cavities. Additionally, Fig. 6(c) shows the porosity within the carbon filler. This morphology represents the composite of C5-5LLDPE compacted at 150°C.

The honeycomb-like features are assumed to be made of carbon and their geometric pattern represents the porosity present in the carbon material.

Morphological tests conducted with FESEM can reveal various features, including porosity, the flow of plastic in the LLDPE matrix, and porosity within the carbon filler of the composite studied here. These observations can be attributed to various factors that influence the mixture's morphology, such as mixing temperature, shear rate, composition ratio, differences in melt

viscosity, interfacial stress of the material, and the amount and type of additives incorporated, among others [8].

In this study, no additives were utilized. Porosity and cavities may have formed due to the porous nature of the carbon filler material and the entrapment of air during the compaction process in the mould. Additionally, the rheological properties of the LLDPE matrix play a role in the formation of porosity cavities within the composite. For comparison, other studies on polymer composites also investigate morphological phenomena related to electrical conductivity and tensile mechanical properties [10, 24, 1, 25, 23].

### 4 Conclusion

In the composite, there is a positive correlation between the carbon content and compaction temperature with the flow of electric current. Specifically, the compaction temperature has a significant influence on the increase in electric current flow. For example, the sample labeled C7-3LLDPE, which consists of 70% carbon weight and 30% LLDPE weight, exhibits the highest electric current flow, measured at 0.0018 A, when compacted at 150°C. On the other hand, the lowest electric current flow is observed in the C5-5LLDPE sample at a compaction temperature of 135°C, registering at 0.0000638 A.

Regarding the modulus of elasticity, the highest value of 2686.43 [N/mm<sup>2</sup>] is achieved by the C7-3LLDPE composite at a compaction temperature of 120°C. Conversely, the lowest modulus of elasticity of 1530.94 [N/mm<sup>2</sup>] is recorded for the C5-5LLDPE composite at a compaction temperature of 135°C. It is worth noting that an increase in compaction temperature generally leads to a decrease in the modulus of elasticity across all composition ratios.

Furthermore, a higher carbon content within the composite corresponds to a higher modulus of elasticity at all compaction temperatures. The tendency for increased electric current flow in the C6-4LLDPE and C7-3LLDPE specimens is primarily attributed to their higher carbon content. Conversely, the decrease in modulus of elasticity in these specimens stems from the formation of numerous porosity cavities, particularly at elevated compaction temperatures.

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