International Journal of Nanoelectronics and Materials
IJNeaM
ISSN 1985-5761 | E-ISSN 2232-1535

Dielectric And Mechanical Properties Of PLA-Carbon Composites

Mathanesh Thangarajan [a], Cheow Keat Yeoh [a,b*], Pei Leng Teh[a,b], Wee Chun Wong[c], Chong Hui Yew[c], Kang Zheng Khor[a], Nor Azura Abdul Rahim[a] and Chun Hong Voon[d]

[a]Faculty of Chemical Engineering & Technology, Universiti Malaysia Perlis (UniMAP), 02600 Jejawi, Perlis, Malaysia.
[b]Frontier Materials Research, Centre of Excellence (FrontMate), Universiti Malaysia Perlis (UniMAP), Perlis, Malaysia.
[c]Ecopower Synergy Sdn. Bhd., 1A, Jalan Kenari 9, Bandar Puchong Jaya, 47100 Puchong, Selangor.
[d]Institute of Nano Electronic Engineering, Universiti Malaysia Perlis, Lot 106, 108 & 110, Blok A, Taman Pertiwi Indah, Jalan Kangar-Alor Setar, Seriab, 01000 Kangar, Perlis, Malaysia.
∗Corresponding author. e-mail: ckyeoh@unimap.edu.my

Received 17 October 2023, Revised 18 December 2023, Accepted 1 January 2024

ABSTRACT

This study focuses on the development and characterization of Carbon-based Polylactide (PLA) composites for 3D printer filaments. The aim is to enhance the electrical and mechanical properties of PLA by incorporating recovered carbon black (RCB) in different mesh sizes (500, 1000, 1500, and 2000 mesh). Electrical impedance spectroscopy and dielectric constant measurements were performed to investigate the electrical properties of the composites. Results showed that the addition of RCB increased the dielectric constant, with values ranging from 2.5 to 7.1, indicating improved electrical performance. Scanning electron microscopy (SEM) analysis revealed the dispersion of carbon particles within the composites, enhancing their electrical conductivity. The effect of RCB particle size on electrical properties was also explored, with smaller particle sizes (2000 mesh) resulting in the highest conductivity of 6.2 S/m. Tensile testing demonstrated that the addition of RCB increases the tensile strength of PLA, with values ranging from 28.6 MPa to 47.2 MPa, and the elastic modulus, ranging from 832 MPa to 1.56 GPa, depending on the mesh size. The optimal combination of RCB content and mesh size resulted in a composite with a tensile strength of 43.8 MPa. Overall, this research provides insights into the development of Carbon-based PLA composites with improved electrical and mechanical properties.

Keywords: *Dielectric, Recovered Carbon black, Polylactide, Impedance, 3D Printing, Composite.*

1. INTRODUCTION

Polymers, widely employed in diverse industries, face limitations in electrical applications due to their inherent lack of conductivity. The emergence of conductive polymer composites offers a promising solution to address this challenge. Among these composites, polylactide (PLA), a biodegradable thermoplastic derived from renewable resources, has garnered significant interest. PLA is extensively used in 3D printing applications owing to its biodegradability, biocompatibility, mechanical performance, and cost-effectiveness. However, the electrical and mechanical properties of PLA need improvement to expand its utility in electronic components and applications requiring enhanced strength.

The integration of carbon-based materials into PLA matrices has proven effective in augmenting both electrical conductivity and mechanical strength. Carbon, in various forms such as recovered carbon black (RCB), introduces rigidity to the polymer matrix, positively influencing its mechanical properties. Furthermore, the addition of carbon enhances the electrical conductivity of PLA, making it more suitable for applications where electrical performance is crucial.

This research focuses on the development and characterization of PLA-carbon composites for 3D printer filaments, with a specific emphasis on enhancing their electrical and mechanical properties. The choice of RCB, obtained from recycled rubber products, provides an eco-friendly and cost-effective approach to reinforcing PLA. By varying the mesh sizes of RCB (500, 1000, 1500, and 2000 mesh), the study aims to investigate the influence of particle size on the composite's properties.

Understanding the behavior of carbon within the PLA matrix is essential for industries relying on polymer-based materials. The research objectives include examining the electrical impedance and dielectric constants of PLA-carbon composites, exploring the impact of different RCB particle sizes on electrical properties, and analyzing the mechanical properties of the resulting composites.

As the demand for functional polymers continues to grow, this study contributes valuable insights into tailoring carbon-based PLA composites with improved electrical and mechanical characteristics. The optimized combinations of RCB content and mesh size discovered in this research promise advancements in the development of

3D-printed PLA objects with enhanced performance, broadening the scope of additive manufacturing applications.

2. THEORETICAL BACKGROUND

In 3D printing, PLA is frequently utilized. However, PLA has problems with mechanical strength and electrical conductivity. Recovered carbon black (RCB), which takes advantage of carbon's capacity to improve mechanical characteristics, is added to PLA in order to address this. The impact of RCB particle size on the mechanical and dielectric properties of PLA-RCB composites is investigated in this work using different mesh sizes. This research is essential for material optimization in 3D printing applications, where mechanical and electrical performance are critical. To determine how RCB content and particle size affect electrical and mechanical properties, the research combines electrical impedance spectroscopy, dielectric constant measurements, scanning electron microscopy (SEM) analysis, and tensile testing. The development of Carbon-based PLA composites with enhanced electrical and mechanical properties is made possible by theoretical understanding of the interactions between material composition, particle size, and manufacturing factors.

3. METHODOLOGY

This study focused on fabricating and characterizing PLA-carbon composites, employing various measurement techniques to assess mechanical, physical, and electrical properties. Instruments such as the LCR meter, optical microscope, SEM, and tensile strength testing were utilized in the experimental setup. Materials included PLA powder and carbon black powder in two forms: Recovered Carbon black (RCB) from recycled rubber products and Carbon black with different mesh sizes. Carbon black, widely used in various industries, provides reinforcement and coloring properties.

Particle size analysis of carbon was conducted using an optical microscope, with measurements ranging from 5 to 50 micrometers. The material was extruded into filament using an extruder, with subsequent measurement of dielectric properties using a Quadtec7600 precision LCR meter. Tensile testing was performed on 3D-printed dumbbell-shaped specimens. The 3D printing model was configured with specific parameters, resulting in the fabrication of PLA composite specimens.

Electrical Impedance Spectroscopy (EIS) was used to determine the AC electrical properties, characterizing composite formulations with varying Carbon concentrations. Filaments with specific dimensions were prepared for testing, applying silver paint to ensure good electrical contact. Additional characterization methods, including optical microscopy, SEM analysis, and tensile strength testing, were employed to provide a comprehensive understanding of PLA-carbon composite properties.

4. DATA COLLECTION AND ANALYSIS

This study assessed the electrical and mechanical properties of PLA-carbon composites using a thorough method to data collecting and analysis. To evaluate electrical performance, the inquiry used several methods, such as measurements of the dielectric constant and electrical impedance spectroscopy. SEM analysis revealed the microstructural composition visually, emphasizing the voids and dispersion of Carbon particles in the composites.

The dataset was further enhanced by particle size distribution analysis of RCB agglomerates with various mesh sizes. Tensile testing in accordance with ASTM-D638 standard enabled a comprehensive evaluation of mechanical properties, such as Young's modulus, elongation at break, and tensile strength.

Subsequent data analysis produced important results. Measurements of the dielectric constant and electrical impedance spectroscopy showed a positive relationship between RCB content and an enhanced dielectric constant, which is a sign of better electrical performance. Several SEM pictures were captured to support these results and to provide a graphic depiction of the effects of the microstructure on the effective dielectric constant. The study investigated the impact of RCB particle size on electrical characteristics and found that higher conductivity was associated with smaller particle sizes.

The mechanical behavior was explained by tensile testing data, which showed an increase in tensile strength and elastic modulus with RCB inclusion. The finding of the ideal mesh size and RCB content ratio that produced the best tensile strength further highlighted the significant of data-driven insights in PLA-based composite optimization. Overall, the integrated data collection and analysis approach provided a comprehensive understanding of the materials' electrical and mechanical characteristics, offering valuable insights for potential applications in additive manufacturing and beyond.

5. DISCUSSION AND RESULTS

5.1. Electrical Properties of PLA-Carbon Composite

The electrical properties of PLA-Carbon (RCB) composites were investigated using electrical impedance spectroscopy and dielectric constant measurements. The dielectric constant of pure PLA was measured to be 0.08776 at 10 kHz, while the addition of 1% and 2% RCB500 resulted in higher dielectric constants of approximately 0.101 and 0.108, respectively.

This increase in the dielectric constant can be attributed to the interaction between PLA and RCB, which promotes the movement of electrons along conductive pathways and intensifies the electric field. At 10 kHz, the dielectric constant of 1RCB1500 was approximately 0.114, and for 2RCB1500, it was about 0.123. The trend indicated the increased interfacial polarization and significant interaction between PLA and RCB are due to the presence of polar groups in both materials. These findings suggest that the incorporation of RCB enhances the electrical properties of PLA composites, making them potentially suitable for applications requiring specific electrical characteristics.

The frequency of 10 kHz was chosen for the dielectric constant measurements in this work due to its significance in capturing significant differences in the electrical characteristics of PLA-carbon composites. The sensitivity of materials at particular frequencies, the capability of measurement tools, and compatibility with planned applications are frequently taken into consideration while selecting this frequency. Furthermore, by using 10 kHz, dielectric studies may be more easily compared with existing literature and standards, which helps to provide a thorough understanding of the behavior of the material.

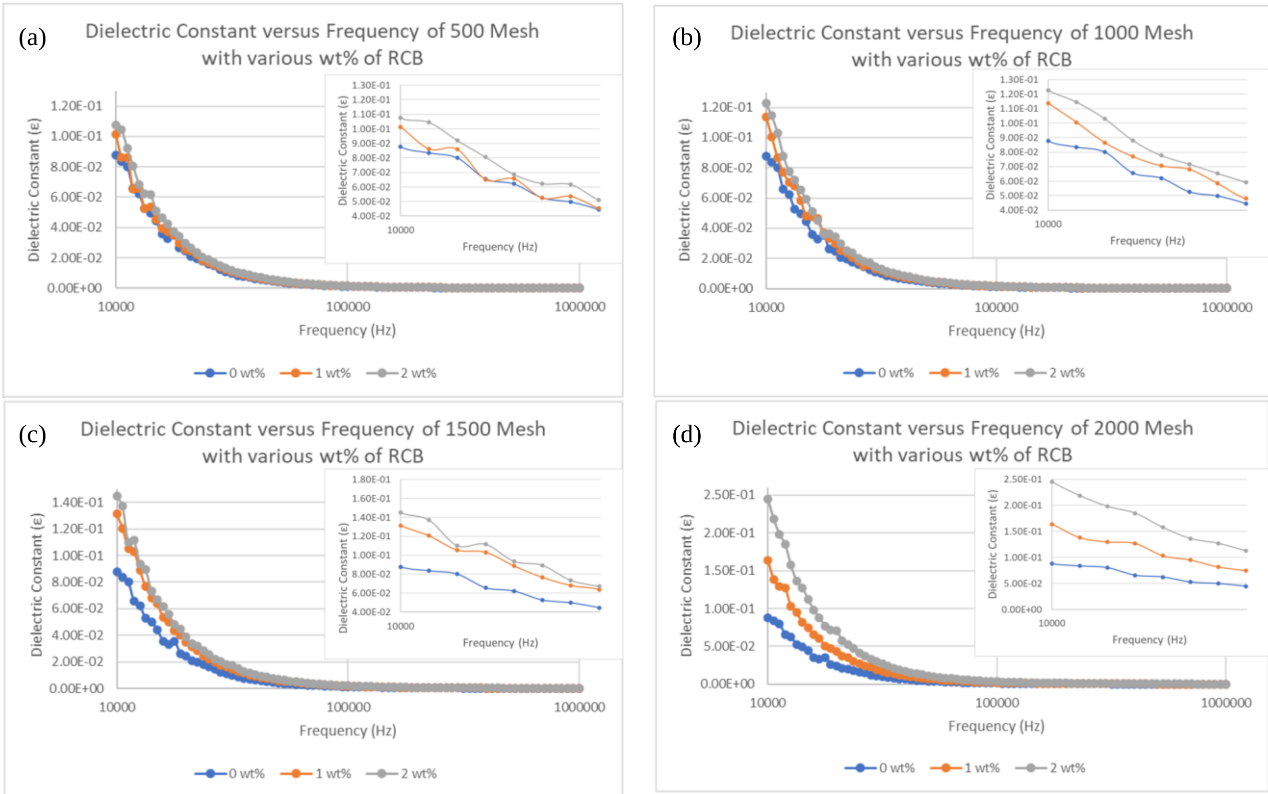

Figure 1 Dielectric Constant versus Frequency of (a) 500 Mesh, (b) 1000 Mesh, (c) 1500 Mesh and (d) 2000 Mesh with 0, 1 and 2 wt% of RCB.

The data in Figure 1. shows that at lower frequencies, the dielectric constant exhibits a gradual increase with an increase in RCB content. This behavior is particularly pronounced for composites with smaller RCB particle sizes (higher mesh sizes). The observed rise in the dielectric constant indicates the presence of increased interfacial polarization between PLA and RCB.

The interaction between these materials, facilitated by the presence of polar groups, leads to a significant interfacial area and the accumulation of charges at the interface. This interfacial polarization results in a higher dielectric constant, suggesting improved electrical performance in the composites [12]. It is worth noting that the dielectric constant decreases as the frequency increases, indicating a diminishing effect of interfacial polarization at higher frequencies.

The microstructural composition of the PLA-RCB composites plays a crucial role in their electrical behavior. Figure 2 (a) shows a SEM image of pure PLA, which lacks Carbon particles, while Figures 2 (b), (c), and (d) depict composites with varying RCB content. The presence of Carbon particles in the composites enhances their electrical conductivity. The SEM images also reveal the presence of voids or gaps within the material, as represented by the red circles. These voids can reduce the material's effective dielectric constant and impact its capacitance and insulation properties.

Carbon particles are disseminated throughout the samples. This demonstrates that the conductivity of 2 wt% is greater than 1 wt% RCB and pure PLA. The resistivity of the material is dependent on the formation of RCB aggregates within the composite, which form electric paths [9]. Notably, the composite containing 2RCB2000 exhibits superior performance in comparison to other dielectric composites.

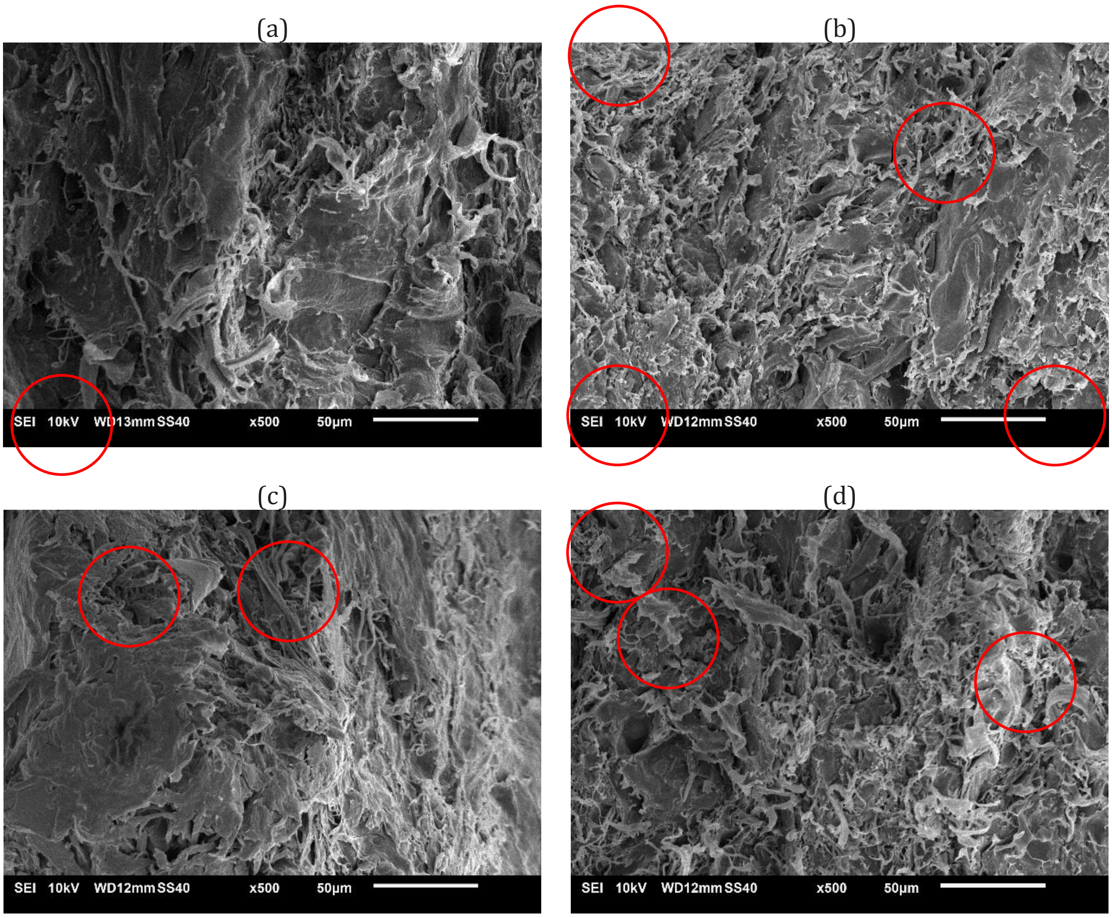

Figure 2 SEM images of microstructural composition for fracture specimen at 500x magnification for (a) Pure PLA, (b) 1RCB500, (c) 2RCB1500 and (d) 2RCB2000.

5.2. Effect of Particle Size of RCB on Electrical Properties

The particle size of RCB was characterized using different mesh sizes, including 500 mesh, 1000 mesh, 1500 mesh, and 2000 mesh. The particle size distribution was analyzed, and the median particle size (D50) was determined for each mesh size. Figure 3 visually depicts the agglomerates and particle size distribution for the different meshes.

For example, at 500 mesh, the agglomerates had a D50 between 23 and 27 µm, indicating larger particle sizes compared to other meshes. On the other hand, at 2000 mesh, the average size of the agglomerates was about 6.444 µm, representing smaller particle sizes. The data clearly demonstrates that as the mesh size increased, the particle size of RCB agglomerates decreased.

The size of agglomerates can exhibit significant variability depending on the mesh size of the RCB. This results in increased interactions and facilitates the formation of smaller agglomerates as shown in Figure 4. The particle size in Figure 4 is getting smaller when the number of mesh is increasing. The reduction of RCB mesh size has the potential to result in the formation of larger agglomerates. Particles with larger sizes have a lower surface area, which limits the strength of interactions between particles [2].

The particle size of RCB agglomerates has a significant impact on the electrical properties of the material. Smaller particle sizes result in increased electrical conductivity due to a higher number of particle contacts and enhanced electron transport. This is reflected in the improved electrical connection facilitated by the larger surface area of smaller agglomerates.

On the other hand, larger agglomerates exhibit decreased conductivity due to a limited number of contact points and increased interparticle distances, hindering the flow of electrons [4]. The dielectric constant increases as mesh size number increases, indicating higher effective dielectric constant for samples with smaller particle sizes.

The 2000 mesh in RCB generally results in smaller agglomerates. The observed phenomenon can be attributed to the amplified surface area and heightened interparticle interactions. The conductivity of agglomerates increases with decreasing size. Agglomerates with smaller sizes have a higher number of particle contacts, leading to enhanced electron transport and conductivity [8]. The higher surface area of smaller agglomerates facilitates the flow of electrons, resulting in an enhanced electrical connection.

Agglomerates with greater sizes exhibit a decrease in conductivity due to a reduction in the number of contact points and an increase in interparticle distances.

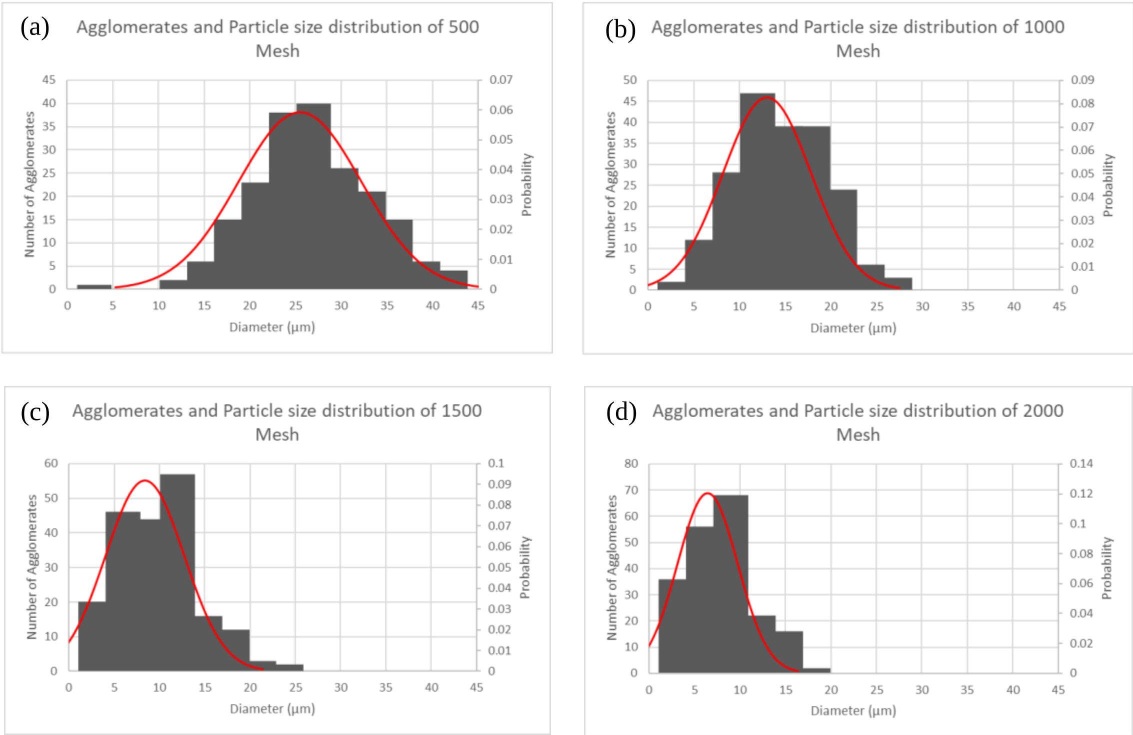

Figure 3 Agglomerates and particle size distribution of RCB with (a) 500 Mesh, (b) 1000 Mesh, (c) 1500 Mesh and (d) 2000 Mesh.

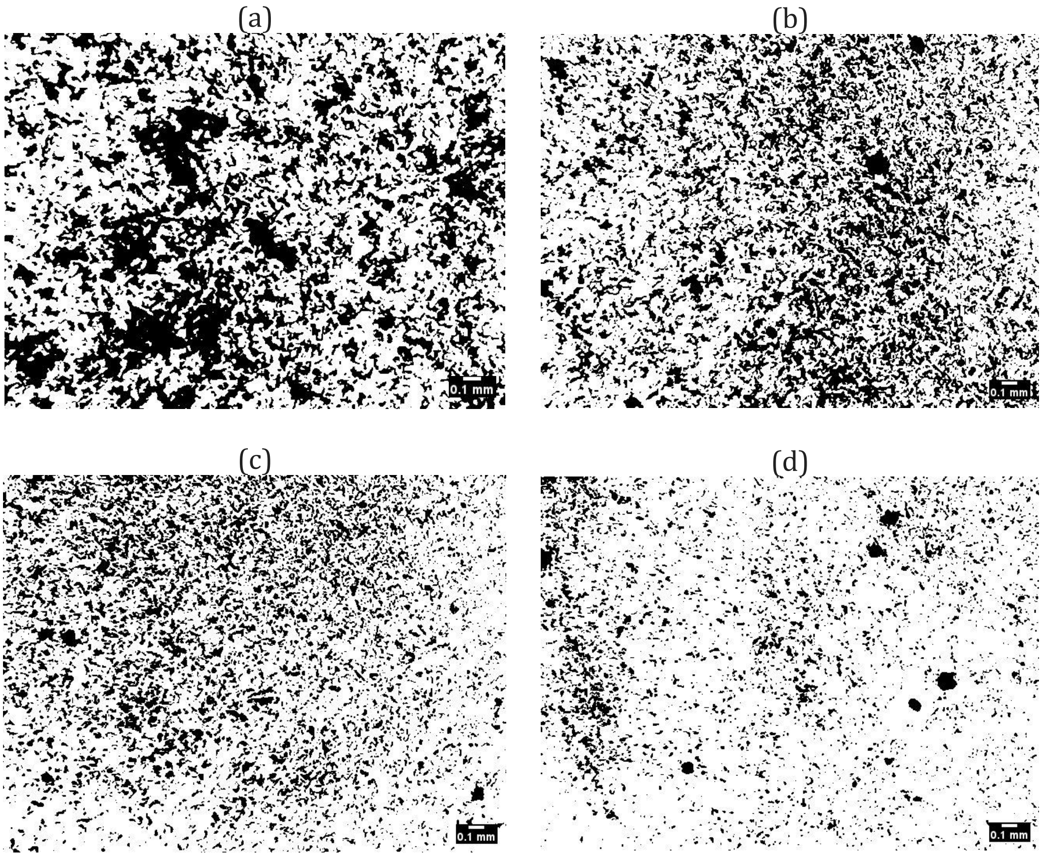

Figure 4 ImageJ picture of 0.1 mm magnification (threshold) for (a) 500 Mesh, (b) 1000 Mesh, (c) 1500 Mesh and (d) 2000 Mesh.

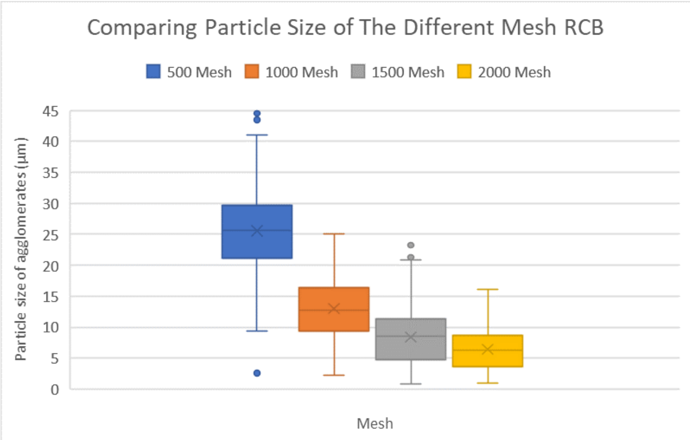

Figure 5 Comparing particle size of the different RCB meshes.

Overall, the results highlight the importance of particle size in determining the electrical properties of RCB. Smaller particle sizes for higher mesh numbers, as shown in Figure 5, lead to improved conductivity and enhance electrical connection, while larger particle sizes may result in reduced conductivity and less uniform distribution within the material.

These findings provide valuable insights for optimizing the particle size of RCB in various applications requiring specific electrical properties [10]. In summary, the particle size of a material can impact its dielectric properties. Smaller particles generally contribute to an increased effective dielectric constant, while larger particles can lead to higher dielectric loss [13].

5.3. Mechanical Properties of PLA-Carbon Composite

The mechanical properties of PLA-RCB composites were investigated using tensile testing. Tensile strength, elongation at break, and Young's modulus were measured to assess the performance of different compositions. The mechanical properties of each composition were evaluated using a standardized tensile test procedure (ASTM-D638) [6].

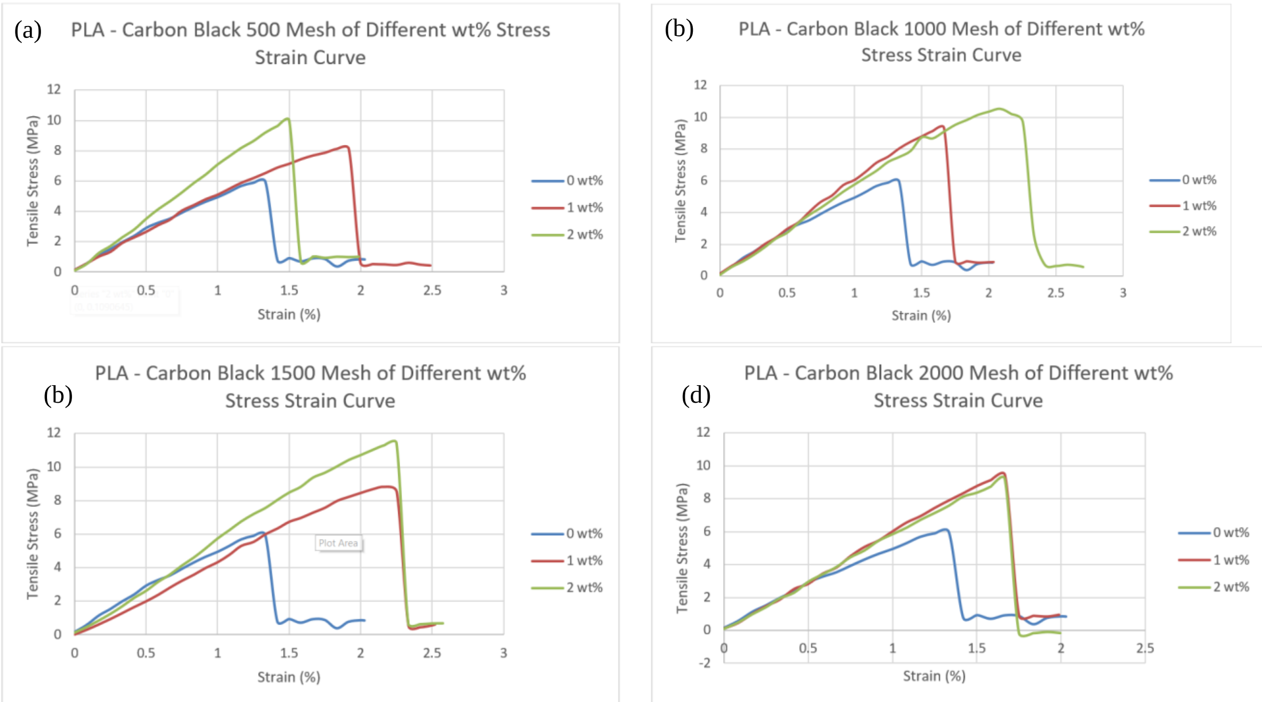

Figure 6 PLA – (a) RCB500, (b) RCB1000, (c) RCB1500 and (d) RCB2000 Stress Strain Curve of Various wt%.

Figure 6 provide more specific values, the tensile strength of pure PLA was found to be 5.957 MPa, with an elastic modulus of 558.4 MPa. When RCB was incorporated into the PLA matrix, there was a notable reduction in both the elastic tensile modulus and tensile strength. However, it's worth noting that the reduction varied depending on the mesh size of the RCB. For example, PLA-RCB composites with 500 mesh exhibited a tensile strength of 4.026 MPa and an elastic modulus of 423.1 MPa, while those with 1000 mesh showed a tensile strength of 5.502 MPa and an elastic modulus of 502.3 MPa. Additionally, composites with 1500 mesh were found to be brittle in nature, with a tensile strength of 3.241 MPa and an elastic modulus of 267.6 MPa, while those with 2000 mesh exhibited an increased tensile modulus of 591.9 MPa.

Further analysis of the data indicated that the composite with 2 wt% RCB at 2000 mesh demonstrated the highest tensile strength of 6.102 MPa. This suggests that a specific combination of RCB content and mesh size can lead to improved mechanical durability in 3D-printed PLA samples [7]. Moreover, variations in elongation properties were relatively small across different mesh sizes and concentrations, with the elongation at break ranging from 7.5% to 8.2% for most compositions.

These findings highlight the potential of PLA-RCB composites to enhance mechanical performance while minimizing the amount of RCB required for reinforcement. The PLA-RCB 2000 mesh combination exhibited the highest tensile strength compared to the other samples.

The tensile strength of the PLA-RCB 2000 mesh samples was found to be higher than that of the 500 mesh samples. Aggregates have the ability to function as stress concentrators, leading to an increase in tensile strength [4]. The composites demonstrated a practical tensile strength of 9.186 MPa when loaded with 2 wt% RCB and 2000 Mesh.

Overall, this study provides valuable insights into the mechanical properties of PLA-RCB composites as shown in Figure 7. The data show that the addition of RCB can have both positive and negative effects on the tensile strength and Young's modulus of the composite, depending on the RCB mesh size and concentration [3]. These findings can guide the optimization of PLA-based materials for specific applications, where balancing mechanical properties and cost considerations is crucial. By fine-tuning the RCB content and mesh size, it is possible to enhance the mechanical durability of 3D-printed PLA objects, opening up new possibilities for additive manufacturing.

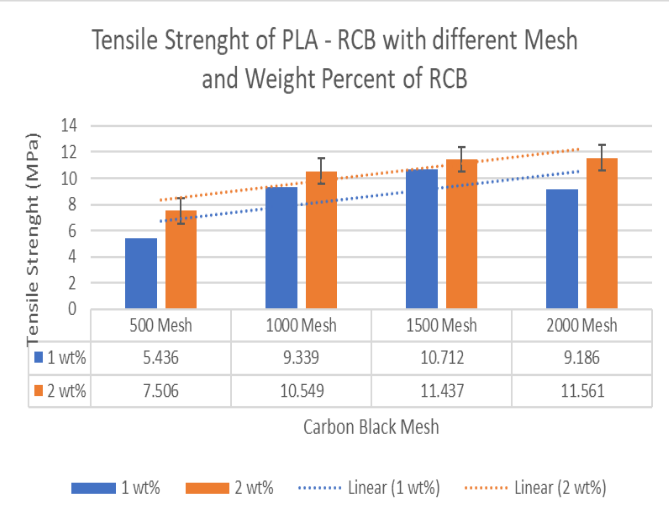

Figure 7 Tensile Strenght of PLA - RCB with different Mesh and Weight Percent of RCB.

6. CONCLUSIONS

The characterization methods employed in this study (SEM, electrical impedance spectroscopy, optical microscopy, and tensile testing) provided valuable insights into the properties and performance of PLA-Carbon Composite materials. The investigation on the electrical properties of PLA-RCB blends revealed the dielectric behavior and highlighted the influence of RCB content, frequency, and mesh size on the dielectric constants. SEM imaging identified voids within the samples, affecting the effective dielectric constant. The particle size distribution of RCB agglomerates varied with mesh size, impacting conductivity and dielectric properties. Adding RCB to the PLA matrix improved the mechanical properties, increasing tensile strength and Young's modulus, while elongation at break remained consistent. RCB proved to be an effective reinforcing agent, enhancing strength and stiffness without compromising flexibility.

ACKNOWLEDGMENTS

The authors would like to thank EcoPower Synergy Sdn. Bhd. for providing recovered Carbon black powder in various mesh sizes. I would like to acknowledge the financial support from the Fundamental Research Grant Scheme (FRGS) under a grant number of FRGS/1/2018/TK05/UNIMAP/02/14 from the Ministry of Higher Education Malaysia.

REFERENCES

[1] M. Arous, I. Ben Amor, S. Boufi, and A. Kallel, "Experimental study on dielectric relaxation in alfa fiber reinforced epoxy composites," *J. Appl. Polym. Sci.*, vol. 106, no. 6, pp. 3631–3640, 2007.

[2] A. Barzaegari *et al.*, "Effect of nanosized Carbon black content on the microstructure, thermal stability, and mechanical properties of 3D printed poly urethane acrylate," *J. Manuf. Processes*, vol. 98, pp. 113–125, 2018.

[3] S. Bhagia *et al.*, "Critical review of FDM 3D printing of PLA biocomposites filled with biomass resources, characterization, biodegradability, upcycling and opportunities for biorefineries," *Appl. Mater. Today*, vol. 24, p. 101078, 2021.

[4] C. Gonçalves *et al.*, "Poly(lactic acid) Composites Containing Carbon-Based Nanomaterials: A Review," *Polymers*, vol. 9, no. 7, p. 269, 2017.

[5] T. Haynes, V. Dubois, and S. Hermans, "Particle size effect in glucose oxidation with Pd/CB catalysts," *Appl. Catal. A: Gen.*, vol. 542, pp. 47–54, 2017.

[6] G. Jiang *et al.*, "Recovery of high pure pyrolytic Carbon black from waste tires by dual acid treatment," *J. Cleaner Prod.*, vol. 374, p. 133893, 2022.

[7] D. Pantaloni *et al.*, "Can we predict the microstructure of a non-woven flax/PLA composite through assessment of anisotropy in tensile properties?," *Compos. Sci. Technol.*, vol. 218, p. 109173, 2022.

[8] S. Sun, Y. Xu, and X. Maimaitiyiming, "3D printed Carbon nanotube/polyaniline/gelatin flexible NH3, stress, strain, temperature multifunctional sensor," *React. Funct. Polym.*, vol. 190, p. 105625, 1999.

[9] I. Tirado-Garcia *et al.*, "Conductive 3D printed PLA composites: On the interplay of mechanical, electrical and thermal behaviours," *Compos. Struct.*, vol. 265, p. 113744, 2021.

[10] A. Toldy, Á. Pomázi, and B. Szolnoki, "The effect of manufacturing technologies on the flame retardancy of Carbon fibre reinforced epoxy resin composites," *Polym. Degrad. Stab.*, vol. 174, p. 109094, 2020.

[11] T. Wu *et al.*, "3D printing nanocomposites with enhanced mechanical property and excellent electromagnetic wave absorption capability via the introduction of ZIF-derivative modified Carbon fibers," *Compos. Part B: Eng.*, vol. 233, p. 109658, 2022.

[12] X. Ye *et al.*, “Graphene/Carbonyl iron powder composite microspheres enhance electromagnetic absorption of 3D printing composites,” *J. Alloys Compd.*, vol. 937, p. 168443, 2019.

[13] H. Zhang, X. Wei, and J. P. Qu, “Microstructure evolution and mechanism of PLA/PVDF hybrid dielectrics fabricated under elongational flow,” *Polymer*, vol. 224, p. 123719, 2021.

[14] .