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# Physical and electrical properties of PLA-carbon composites

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**Abstract.** Polylactic acid or polylactide (PLA) is a biodegradable thermoplastic that can be produced from renewable material to create various components for industrial purposes. In 3D printing technology, PLA is used due to its good mechanical, electrical, printing properties, environmentally friendly and non-toxic properties. However, the physical properties and excellent electrical insulation properties of PLA have limited its application. In this study, with the carbon black (CB) as filler added into PLA, the lattice spacing and morphology were investigated by using X-ray diffraction (XRD) and scanning electron microscope (SEM), respectively. The physical properties of PLA-carbon composite were evaluated by using tensile test, shore D hardness test and density and voids measurement. Impedance test was conducted to investigate the electrical properties of PLA-Carbon composites. The results demonstrate that the inclusion of carbon black as filler enhances the physical properties of the PLA-carbon composites, including tensile properties, hardness, and density. The addition of carbon black also leads to improved electrical conductivity of the composites. Better enhancement toward the electrical properties of PLA-carbon composites is observed with 1wt% of carbon black in N774 grade. The N550 grade with 2wt% of carbon black shows better improvement in the physical properties of PLA-carbon composites, achieving 10.686 MPa in tensile testing, 43.330 in shore D hardness test, and a density of 1.200 g/cm$^3$ in density measurement. The findings suggest that PLA-carbon composites have the potential for enhanced performance in various industrial applications, particularly in sectors requiring improved physical and electrical properties.

**Keywords:** 3D-printing; carbon black; electrical properties; physical properties; PLA

## 1. Introduction

Polylactic acid or polylactide (PLA) can be produced from renewable materials to create a variety of components for industrial purposes. PLA has shown potential to replace existing petrochemical-based polymers in various industrial applications, and it can be easily processed using standard equipment to produce parts, fibers, or film. Its monomers are sourced from renewable resources, specifically natural organic acids (Masiuchok *et al.* 2022).

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PLA is used in 3D printing technology due to its favorable mechanical, electrical, and printing properties, as well as its environmentally friendly and non-toxic attributes (Yeoh *et al.* 2020). The physical and electrical properties of PLA are crucial for the application of 3D printing technology in producing printed parts (Madhavan Nampoothiri *et al.* 2010, Shinyama 2018). Nowadays, the use of polymeric materials has evolved from purely decorative functions to a wide range of functional applications, including fields like architecture and semiconductors (Shen *et al.* 2020).

Despite its advantages, the physical and electrical insulator properties of PLA have limited its application scope. To overcome these drawbacks, various fillers, such as carbon nanotubes (CNT), graphene (GE), expanded graphite, metal powders, etc., are added to create conductive paths for charge transport while also providing mechanical strength and durability (Jem and Tan 2020a, Tirado-Garcia *et al.* 2021, Xue *et al.* 2021). By incorporating different fillers into the PLA matrix, the physical and electrical properties of PLA can be tailored to specific requirements. Carbon black (CB), known for its high tensile strength and electrical conductivity, is an excellent choice for use as a filler material (Weaver 2017, Phua *et al.* 2016).

In this research, varying amounts of different grades of CB were added to PLA to investigate the resulting physical and electrical properties of the PLA-carbon composites. The physical properties of polymeric materials are closely related to the degree of crystallinity (Sung-Wook Lim *et al.*, 2018). Crystallinity defines the extent to which chains fold and form a lamellar structure in the material, strongly influencing its properties. The normal range of crystallinity is described as amorphous (0%) to highly crystalline (>90%). Polymers with linear chain structures and slow cooling rates tend to have higher crystallinity, as sufficient time is available for crystallization during slow cooling. Higher degrees of crystallinity in polymeric materials lead to increased strength and higher melting points (Balani 2015). The addition of fillers to PLA can affect the behavior of crystallinity and mechanical properties. The tensile strength of composites depends on the degree of crystallinity of PLA and the interfacial properties between PLA and the filler (Balani 2015).

Electrical properties are essential for engineering applications and implementations. These properties are crucial for various applications, such as low-loss cables, battery electrodes, electromagnetic shielding, electronic components like capacitors and thermistors, as well as contemporary civil and military technologies. The electrical properties of polymers play a pivotal role in engineering applications and implementations. While natural polymers tend to act as insulators due to the lack of an ionic or electronic route and the covalent nature of the polymers, this limitation has restricted their industrial applications. The electrical properties of polymers can be modified by incorporating different conducting nanoparticles into the polymer matrix (Hazarika and Karak 2021).

## 2. Materials and methods

### *2.1 Material and sample preparation*

Polylactide acid and Carbon Black from Eco Power Synergy Sdn. Bhd. were used to fabricate the samples in this research. The PLA-carbon filaments composites were extruded by Desktop Filament Extruder. After that, the PLA-carbon composites in dumbbell shape were printed by using a K7 Mini 3D Printer. The types of samples were prepared as shown in Table 1.

### *2.2 Materials characterization*

Table 1 Types of samples

| Grades of carbon black | wt% of carbon black (%) | Composites code |
|---|---|---|
| N330 | 1 | PLA-330-1 |
| | 2 | PLA-330-2 |
| N550 | 1 | PLA-550-1 |
| | 2 | PLA-550-2 |
| N660 | 1 | PLA-660-1 |
| | 2 | PLA-660-2 |
| N774 | 1 | PLA-774-1 |
| | 2 | PLA-774-2 |
| - | 0 | Pure PLA |

The lattice parameter and structural phase of the samples were characterized by using X-ray diffraction (XRD) with Bruker, USA (D2 Phaser) at scan rate of 2°/min from 10° to 60°. The samples were coated with a thin layer of platinum by using coating machine from Quorum, England (Q150R), after that the surface morphology of the samples was characterized by SEM from JEOL (JSM-6010LV). The particle size of carbon black was measured by optical microscope and analysed by using ImageJ (Kristopher Kubow 2019).

### *2.3 Tests*

Tensile tests were carried out on the tensile specimens prepared according to ASTM D 638 standard. The test was conducted at room temperature by UTM at a constant speed of 0.5 mm/min with an Instron computer-controlled testing machine. At least three specimens were tested, and the average value was taken. At the end of the tests, the tensile strength of the composites was determined. Hardness tests were carried out on the specimens according to the ASTM D2240 by using a PTC instrument model 370L (Durometer Shore D). Minimum three values were taken for a specimen, and the average value was taken for each composite. Density Measurements were conducted according to ASTM D792 precision electronic weighing machine followed the principle of Archimedes. The mass of PLA-carbon composites in air were recorded and mass of PLA-carbon composites immerse in distilled water at room temperature and conditions were recorded. The theoretical density of PLA-carbon composites was determined by rule of mixtures using below Eq. (1).

$$\rho_{th} = \rho_m * M_m + \rho_r * M_r \quad (1)$$

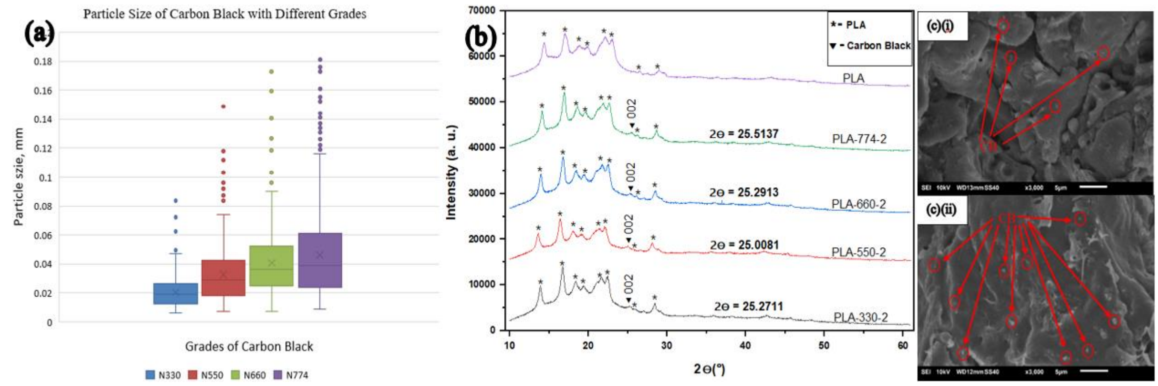
The density of PLA-carbon composites was experimentally determined by using Eq. (2).

$$\rho_{ex} = \frac{M_a}{V} \quad (2)$$

After that, the voids fraction was calculated by Eq. (3).

$$Void\ fraction = \frac{\rho_{th} - \rho_{ex}}{\rho_{th}} \quad (3)$$

Impedance tests were carried out by using HIOKI 3536 LCR meter at room temperature. The frequency measured was from 1Hz to 100 kHz. The LCR meter was connected to a data-collecting




(a) (b) (c)

Fig. 1 (a) Particle size of carbon black with different grades, (b) XRD pattern of PLA-carbon composites and (c) (i) SEM image with x3000 of PLA-330-1 and (ii) SEM image with x3000 of PLA-330-2

software on a computer, which allowed the real and imaginary parts of the impedance to be determined.

## 3. Result and discussions

### *3.1 Material characterizations*

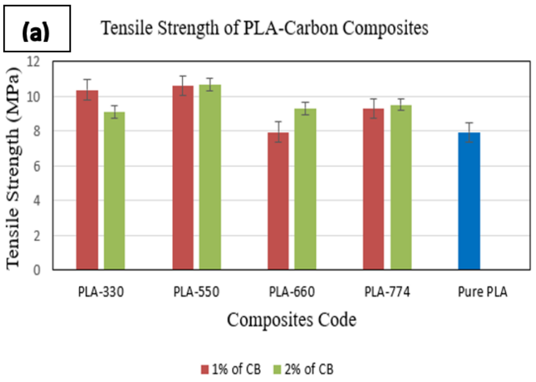
Fig. 1(a) demonstrates the particle sizes of carbon black for grade N330 as the smallest particle size, followed by N550, N660, and N774. Moving to Fig. 1(b), the XRD pattern of PLA and carbon black was determined. A distinct broad peak is evident in the range of $2\theta$=10° to 30°, corresponding to the semi-crystalline nature of pure PLA. The XRD pattern of pure PLA exhibits two main characteristic peaks at $2\theta$=16.8° and 19.5°, denoted as * (Nanaki *et al.* 2018).

Fig. 1(b) indicates that the characteristic peak positions of carbon black (CB) for different grades are observed in the range of $2\theta$=25° to 26°. Specifically, the 2θ values for N330, N550, N660, and N774 are 25.27°, 25.00°, 25.29°, and 25.51°, respectively, as indicated by ▼.

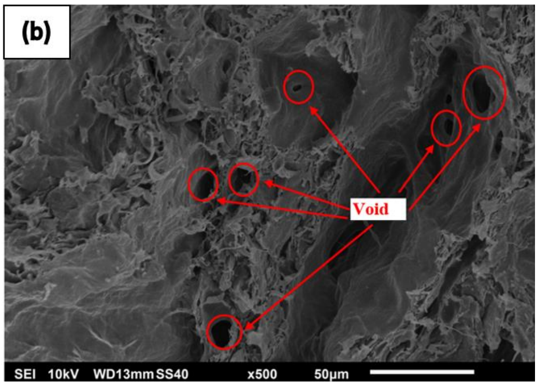
Furthermore, Fig. 1(c)(i) and (ii) show the particles of carbon black, suggesting that the dispersion of carbon black in the PLA-carbon composites is uniform for both 1wt% and 2wt% of CB. In Fig. 1(c)(ii), which has a higher composition of CB, it is clearly observed that there are more carbon black particles in the PLA matrix compared to Fig. 1(c)(i), which has a lower composition of CB.

### *3.2 Tensile strength*

Fig. 2(a) displays the tensile strength of PLA-carbon composites using different grades and compositions of carbon black. Generally, the addition of carbon black (CB) as a reinforcement within the PLA matrix enhances the tensile strength of pure PLA Guo *et al.* 2021. The tensile strength values vary depending on the grade of carbon black. The tensile strength of the PLA-carbon composite with 2wt% CB is slightly higher than that of the composite with 1wt% CB. This result aligns with Guo *et al.* (2021), who indicated that increasing the amount of carbon black




(a)




(b)

Fig. 2 (a) Tensile strength of PLA-carbon composites and (b) SEM image with 500x magnification of PLA-330

reinforcement enhances the composite's tensile strength.

In Fig. 2(a), the PLA-550-2 (10.686 MPa) shows the highest tensile strength compared to others PLA-carbon composites. This suggests that N550 grade carbon black exerts a better reinforcing effect on the PLA matrix, thus results better tensile strength occurred in PLA-550. On the contrary, Fig. 2(a) highlights that the lowest tensile strength among the PLA-carbon composites is exhibited by PLA-660-1 (7.964 MPa) compared to others. This can be attributed to the occurrence of voids and weak interlayer bonding within the PLA-660-1 composite. These voids and weak interlayer bonding, which act as stress intensifiers, significantly impact the tensile properties of the composite (Elkolali *et al.* 2022, Kumar *et al.* 2023).

Theoretically, smaller particle sizes generally result in higher tensile strength for composites (Savetlana *et al.* 2017). According to the previous results, among these four grades of carbon black, the order of particle size is N330 < N550 < N660 < N774. However, Fig. 2(a) clearly shows that the tensile strength of PLA-330 is lower than that of PLA-550. This phenomenon may be attributed to the presence of voids within the PLA-330 specimen, which become weak points and consequently decrease the tensile strength. Fig. 2(b) illustrates an SEM image of the PLA-330 grade at x500 magnification, wherein voids are observed and marked with red circles.

### *3.3 Hardness*

As shown in Table 2, the hardness values of PLA-carbon composites are generally higher than those of pure PLA. The results indicate that the addition of carbon black (CB) to the PLA matrix enhances the hardness of the composites, which is consistent with findings from other researchers (Ansari and Kamil 2022, Espinel-Rubio and Feo-Ardila 2022).

As depicted in Table 2, as the carbon black composition increases from 1wt% to 2wt%, the Shore D hardness value of PLA-carbon composites tends to increase. This suggests that as the CB composition increases, the hardness of the resulting composites also increases. These results align with the research conducted by Rahmalina *et al.* (2019), where an increase in carbon black content as a filler within the composites leads to an increase in composite hardness. The relatively good adhesion between the matrix and reinforcement materials contributes to the increased hardness of the composites.

Table 2 Shore D hardness of PLA-carbon composites.

| Percentage of Carbon Black (%) | Composite code | Hardness (Shore D) | | | Min. | Max. | Mean | StDev |
|---|---|---|---|---|---|---|---|---|
| | | 1 | 2 | 3 | | | | |
| 1% | N330 | 39.00 | 40.00 | 39.00 | 39.00 | 40.00 | 39.33 | 0.58 |
| | N550 | 42.00 | 41.00 | 41.50 | 41.00 | 42.00 | 41.50 | 0.50 |
| | N660 | 42.00 | 39.00 | 40.00 | 39.00 | 42.00 | 40.33 | 1.53 |
| | N774 | 41.00 | 40.00 | 39.00 | 39.00 | 41.00 | 40.00 | 1.00 |
| 2% | N330 | 42.50 | 42.50 | 42.00 | 42.00 | 42.50 | 42.33 | 0.29 |
| | N550 | 44.00 | 43.00 | 43.00 | 43.00 | 44.00 | 43.33 | 0.58 |
| | N660 | 42.00 | 43.00 | 41.00 | 41.00 | 43.00 | 42.00 | 1.00 |
| | N774 | 41.00 | 41.50 | 42.00 | 41.00 | 42.00 | 41.50 | 0.50 |
| Pure PLA | | 36.00 | 35.50 | 35.00 | 35.00 | 36.00 | 35.50 | 0.50 |

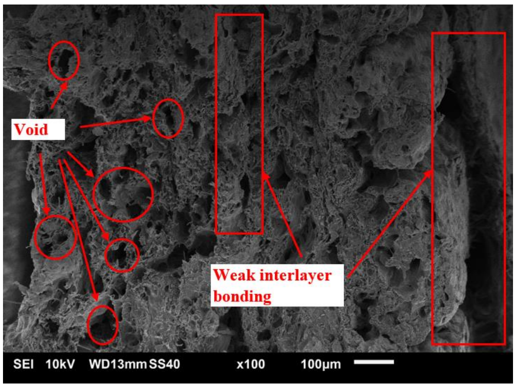



Fig. 3 SEM image of PLA-330 at x100 magnification.

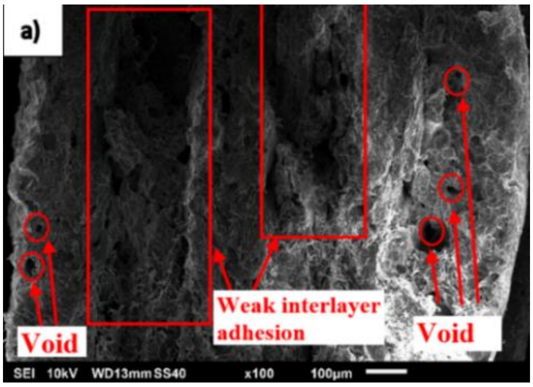
Generally, Table 2 demonstrates that the hardness values of PLA-carbon composites follow the theoretical expectations, with PLA-550-2 (43.330) having higher hardness values than PLA-660 (42.000) and PLA-774 (41.500). Theoretically, smaller particle sizes tend to yield better mechanical properties for composites (Savetlana *et al.* 2017). The order of particle sizes for CB grades is N330<N550<N660<N774. However, the hardness value of PLA-330 is lower than that of PLA-550. This may be attributed to the presence of voids and weak interlayer bonding within the printed structures (Hasanzadeh *et al.*, 2023). This is further supported by the SEM result in Fig. 3, which displays an SEM image of PLA-330-2 at x100 magnification, revealing voids and weak interlayer adhesion marked by red boxes.

### *3.4 Density measurement*

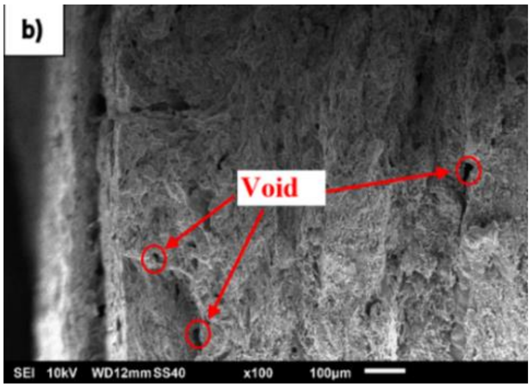
Table 3 presents the measured density of both PLA and PLA-carbon composites. The disparity between experimental and theoretical densities indicates the presence of voids and pores within the fabricated composite samples, significantly impacting their material properties. In Table 3, PLA-carbon composites display a higher density than pure PLA samples due to improved adherence of carbon black to the PLA matrix. This enhanced adherence results in a higher density compared to

Table 3 Theoretical density and experimental density of pure PLA and PLA-Carbon composites

| Composite | Theoretical Density (g cm$^{-3}$) | | Experiment Density (g cm$^{-3}$) | | Void Fraction | |
|---|---|---|---|---|---|---|
| | 1wt% | 2wt% | 1wt% | 2wt% | 1wt% | 2wt% |
| PLA-330 | 1.2476 | 1.2451 | 1.1486 | 1.1395 | 0.0793 | 0.0848 |
| PLA-550 | 1.2457 | 1.2415 | 1.2000 | 1.2000 | 0.0367 | 0.0334 |
| PLA-660 | 1.2472 | 1.2444 | 1.1282 | 1.1324 | 0.0954 | 0.0901 |
| PLA-774 | 1.2476 | 1.2451 | 1.1833 | 1.1695 | 0.0515 | 0.0607 |
| Pure PLA | 1.2500 | | 1.1017 | | 0.1186 | |




(a)




(b)

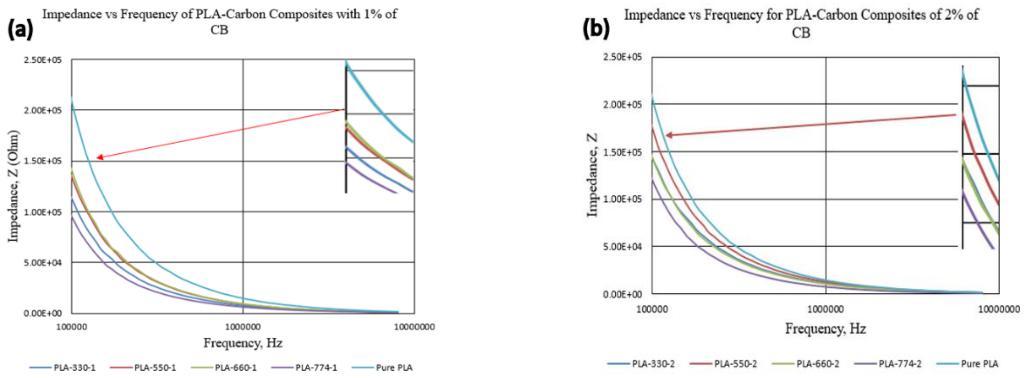
Fig. 4 SEM image with (a) 100x magnification of pure PLA and (b) With 100x magnification of PLA-carbon composites

pure PLA samples. The reduction in air gaps and the achievement of better interlayer bonding between layers of PLA-carbon composites contribute to a lower voids fraction recorded for these composites compared to pure PLA samples (Suresha *et al.* 2022).

In Fig. 4, it is evident that pure PLA samples contain more voids compared to PLA-carbon composites. Fig. 4(b) highlights weak interlayer bonding observed in pure PLA samples (marked in the red box) when compared to the PLA-carbon composites in Fig. 4(a). This may be attributed to PLA cooling too quickly during the printing process, as PLA has a rapid cooling and solidification rate compared to other materials. Excessive cooling from the print fan or its close proximity to the print can cause the layers to cool too rapidly, resulting in weak interlayer bonding (Lee and Liu 2019).

In Table 3, among the composites with the same carbon grade, no significant difference in density is observed between the 1wt% and 2wt% compositions. For example, both PLA-550-1 (1.2000 g/cm$^3$) and PLA-550-2 (1.2000 g/cm$^3$) have the same density despite having different compositions. This suggests that the variation in carbon content within the studied range (1wt% and 2wt%) did not significantly impact the density.

The presence of voids creates weak points within the material, causing stress to concentrate in those areas during tensile loading (Elkolali *et al.* 2022). This stress concentration can lead to premature failure or reduced load-bearing capacity, resulting in the lower tensile strength observed in the PLA-660-1 sample. Table 3 highlights the significant observation that PLA-660-1




(a) (b)

Fig. 5 (a) Impedance plot of PLA-carbon composites with 1wt% of CB and (b) Impedance plot of PLA-carbon composites with 2wt% of CB

demonstrates the highest voids fraction among the PLA-carbon composites. This finding aligns with the tensile testing results, as PLA-660-1 exhibits the lowest tensile strength among the PLA-carbon composites. Conversely, the PLA-550 composites, which exhibit the highest tensile strength among the composites, also have the lowest voids fraction.

### *3.5 Impedance*

The conductivity characteristics of the material are determined by its impedance, which acts as a barrier to electron movement (Chakraborty *et al.* 2020). In Figs. 5(a) and (b), the impedance of pure PLA at 100 kHz surpasses that of PLA-carbon composites, indicating that pure PLA impedes more AC current compared to PLA-carbon composites.

The relationship between the carbon black content and the impedance of carbon black composites shows that higher carbon black content results in a better conductive network for carbon black composites, leading to lower impedance (Ceregatti *et al.* 2017). In Fig. 5(b), lower impedance is observed for PLA-330-2 and PLA-550-2 compared to PLA-330-1 and PLA-550-1, as shown in Fig. 5(b). This suggests that a better conductive network is formed within PLA-330-2 and PLA-550-2 relative to PLA-330-1 and PLA-550-1. Increasing carbon black content reduces the impedance of the composites and enhances their electrical conductivity (Ceregatti *et al.* 2017).

As depicted in Fig. 5(a), the impedance of PLA-330-1 is smaller than that of PLA-660-1, followed by PLA-774-1. The conductivity of the carbon black network follows the sequence: PLA-330-1>PLA-550-1>PLA-774-1. These results align with the theory that smaller particle sizes of carbon black typically result in a larger surface area for contact and interaction with the polymer matrix, facilitating the formation of a more effective conductive network (Macías-García *et al.* 2020). However, in Fig. 5(b), the impedance of PLA-774-2 at 100 kHz is lower than that of PLA-330-2, PLA-550-2, and PLA-660-2, indicating that PLA-774-2 exhibits better electrical conductivity than the other three. This discrepancy may be attributed to the porosity, surface groups, and structure of the carbon black in the PLA-carbon composites (Macías-García *et al.* 2020).

## 4. Conclusions

This study investigates the impact of adding carbon black to the PLA matrix on composite properties. The findings demonstrate enhanced tensile strength in PLA-carbon composites compared to pure PLA, with PLA-550-2 displaying the highest values (10.686 MPa) for both 1wt% and 2wt% carbon black compositions. However, PLA-660-1 (7.964 MPa) exhibited lower tensile strength, attributed to voids. Notably, the particle size order of carbon black grades (N330<N550<N660<N774) didn't directly correlate with tensile strength, suggesting other factors like interlayer bonding influenced it.

Hardness values were generally higher in PLA-carbon composites, particularly with smaller carbon black particles. PLA-550-2 (43.330) had the highest hardness, while PLA-330-1 (39.330) had lowest values, possibly due to voids and weak interlayer bonding. PLA-carbon composites also exhibited higher densities due to improved carbon black adhesion, with 1wt% and 2wt% compositions showing similar density.

The addition of carbon black led to improved electrical conductivity and lower impedance in PLA. The carbon black network's conductivity sequence was N330>N550>N774, except for N774. Overall, composites with 2wt% carbon black displayed better physical properties, albeit slightly higher impedance than 1wt%. Smaller particle size contributed to improved properties, while voids and weak interlayer bonding affected various composite aspects.

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