

Mechanical and Electrical Behavior of PLA Composites with Various Carbon Black Concentrations

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ABSTRACT

Poly(lactic acid) (PLA) is a promising biodegradable polymer with low carbon footprint, yet its brittleness and absence of electrical properties limit its applicability in functional contexts. To address these limitations, a study was done by incorporate recycled carbon black (CB) a sustainable and conductive filler into the PLA matrix at loading of 0, 1, 2 and 3 wt.%. The composites were produced using melt-mixing and 3D printing Fused Deposition Modeling (FDM), thereafter examining for mechanical, electrical and biodegradation properties. The result indicated that the recycled-CB markedly enhanced tensile strength, Young's modulus, and hardness, with optimal reinforcement occurring at 2 wt.% due to efficient filler dispersion. Electrical conductivity exhibited a marked rise with the recycled CB content, where it reached a maximum at 3 wt.% due to the formation of percolation networks. Nevertheless, the increase in recycled-CB loading lowered the biodegradation rate by restricting water absorption and microbiological activity. The 2 wt% CB composites proved to be the optimal balance, providing robust mechanical properties, moderate conductivity and retained though slowed biodegradability. The finding of this study indicates that recycled CB can transform pure PLA into multifunctional material suitable for sustainable and biodegradable packaging for electrical while applying environmentally responsible engineering.

Keywords: Carbon black, PLA composites, Electrical conductivity, Biodegradable polymers, Impedance spectroscopy, 3D printing, Soil burial degradation

1. INTRODUCTION

Poly(lactic acid) (PLA) refers to a biodegradable, bio-based thermoplastic polymer which is based on renewable resources like corn starch and sugarcane material. In applications where environmental friendliness is a necessary feature, PLA is broadly used, e.g., in packaging, biomedical devices, and additive manufacturing due to good mechanical properties and ease of processing. Nonetheless, the natural brittleness and poor electrical conductivity of PLA restrict its use in high-performance and functional applications, especially in applications that need structural integrity and electrical sensitivity [1].

To overcome these restrictions, conductive fillers that tend to improve the mechanical and electrical properties of PLA have received enormous attention whereby (CB) is the most common conductive filler. Carbon black is a common frequently used additive in a polymer composite and is characterized by high surface area, good electrical conductivity, and low cost. When added to PLA, [2] CB can create conducting bridges thus enhancing electrical conductivity as well as enhancing strength of the material.

In the current research the recycled carbon black was added to the PLA at the concentrations of 0, 1, 2, and 3 wt% by weight to study the impact it has on composite strength, hardness, and electrical conductivity. Melt mixing and 3D printing were utilized to fabricate the composites and promote even filler dispersion. The characterization was done by tensile test, hardness test, impedance spectrometry, Fourier Transform Infrared Spectroscopy (FTIR), and long-term buried test in soil (30 days). This paper reports the correlation between the CB loading and the performance of composites, with the purpose of determining the optimum line of formulation towards the use of environmentally friendly, as well as multifunctional composites.

2. THEORETICAL BACKGROUND

Poly(lactic acid) (PLA) is a commonly researched biodegradable polymer which is made from renewable resources e.g. corn starch and sugarcane. It also provides green benefits including composability, biocompatibility, and reduced carbon footprint than petroleum-based polymers. But there are also limitations to PLA: as it is

brittle and poorly insulative to thermal conditions, and has low electric conductivity, thus lacking wider application in high-performance or functional formats (Zhu et al., 2021; Ghaffar et al., 2020).

To overcome such disadvantages, fillers that are conductive like CB have been added to the PLA matrix to improve its mechanical and electrical properties. CB, a low-cost, nanoscale conductive material, constitutes a conductive pathway in the polymer structure whether percolation threshold is attained. This yields a significant rise in electrical conductivity so that it suitable antistatic or low-power electronic applications.

More than electrical enhancement, CB also results in enhanced tensile strength and modulus and surface hardness, all attributable to its large surface area and filler-matrix interaction effect. Nevertheless, at higher filler loads, agglomeration of fillers may also adversely affect mechanical behavior, and this is because of the limitation of dispersion of fillers, and the existence of the stress concentration zones (7).

Even though PLA is a natural biodegradable material, it is transformed by adding the non-biodegradable and inert compound CB which changes the extent on which the material is degraded in the environment. Several experiments indicated that by excluding water uptake and limiting contact between the polymer surface and microbes, CB decreases microbial and hydrolytic degradation rate [10]. This trade-off highlights the need to optimize CB content which could maintain the concept of balance between mechanical and electrical performance with the environment.

3. METHODOLOGY

This research paper examines how different amounts of recycled CB loading in the PLA composite materials impact its mechanical and electrical properties. The composites were prepared with the concentrations of CB 0, 1, 2, and 3 wt.%. The preparation of materials, manufacturing components and the mechanical, electrical, and biodegradability forms the methodology.

3.1. Materials

PLA in powder form ordered from Rimless Industry Co., Ltd. (Jilin, China), with density of 1.24 g/cm³, glass transition temperature 55–65°C, melting temperature 150–180 °C and it functions as biodegradable polymer matrix. While recycled carbon black filler, received from Eco Power Synergy (Malaysia), with density of 1.8 g/cm³, particle size 30–50 nm, was derived from end-of-life rubber and plastic trash, where it functions as thermal stability, ultraviolet (UV) resistance and potential for electrical conductivity.

PLA powder and recycled CB filler of 0, 1, 2 and 3 wt.% with various loading were premixed at 100 rpm for 3 min, then compounded via twin-screw extrusion machine at 200 °C temperature and 50-120 rpm rotation speed. The

extrudates were cooled and processed into filaments for 3D printing. The printing process was conducted at 200 °C nozzle temperature, 0.2 mm layer height and 10 mm/s speed. The specimens were fabricated as per the ASTM standard Type IV dog bone for tensile test (ASTM D638), rectangular bars for flexural test (ASTM D790) and 5 × 5 × 0.2 (mm) plates for electrical resistance measurements. For the soil burial the samples were fabricated in the size of 1 × 1 × 0.2 cm.

3.2. Mechanical Testing

Tensile tests were performed using Instron Universal Testing Machine at speed of 2.5 mm/min crosshead. ASTM D785 (Standard Test Method for Rockwell Hardness of Plastics and Electrical Insulating Materials) uses a steel ball indenter with a total load of 588 N. The depth of indentation h was determined based on the depth of indentation triangle based on

$$h = \left(\frac{base}{2}\right) \times \tan(120) \quad (1)$$

Then hardness (H) was calculated as:

$$Hardness = \frac{F}{3\pi h^2} \quad (2)$$

Where the applied load is F and indentation depth is h .

3.3. Electrical Conductivity Measurement

The electrical conductivity was intensively obtained with electrochemical impedance spectroscopy (EIS) at a constant frequency of 1 MHz. Proper electrode was made by using a conductive silver paste. The electrical conductivity (σ), measured in S/cm, was calculated following the formula in Equation (3), where R represents electrical resistance (measured in Ω), A indicates the cross-sectional area of the tested specimen (measured in cm²), and L indicates the length of the specimen (measured in cm).

$$\sigma = \frac{L}{R \times A} \quad (3)$$

3.4. Material Characterization

The filler dispersion, as well as fracture morphology, was studied through SEM with a magnification of 3000x. FTIR measurements between 4000 and 600 cm⁻¹ in the ATR mode were undertaken to measure any chemical interaction or degradation.

3.5. Biodegradable

The PLA-CB composites were evaluated in terms of their biodegradability including soil burial test in ambient conditions which lasted 30 days. The factor of degradation was measured by the loss of mass, in that, each sample was weighed before and after burial. The degree of biodegradation was shown by the percentage of weight loss calculated using Equation (4),

$$\text{weight loss} = \left(\frac{\text{Weight before} - \text{Weight after}}{\text{Weight before}} \right) \times 100\% \quad (4)$$

The pure PLA had the greatest weight loss, which inherits biodegradability of the PLA. There was a correlation between the amount of recycled CB content and weight loss whereby as the amount of recycled CB content increased the amount of weight loss decreased which is an indication of the microbial weight loss since the carbon black was inert and did not biodegrade. This decrease in mass loss indicates that the incorporation of CB prevents absorption of water and access by microbes, and this delays the degradation process. Nevertheless, this did not affect the moderate value of the biodegradability of the 2 wt.% CB composites, which is a strong point of the product and could be considered as a candidate in cases when both functionality and environmental sustainability are crucial.

4. RESULTS AND DISCUSSION

This section highlights the result of different testing that has been performed on the PLA-CB composites, including evaluation of mechanical performance, electrical conductivity, and biodegradability. All the composites were evaluated at various recycled CB loadings of 0, 1, 2 and 3 wt.%, and the findings are analyzed to determine the best filler concentration that coordinates strength, conductivity, and environmental sustainability.

4.1. Tensile Properties

4.1.1. Tensile Strength

The tensile strength results in Figure 4.1 show that the presence of recycled carbon black significantly impacted the mechanical properties of PLA composites. The pure PLA sample with 0 wt.% CB demonstrated the lowest tensile

strength at 8.673 MPa, indicating the polymer's brittle characteristics. A slight increase to 8.948 MPa was noted at 1 wt.% CB, although it limited statistically significant. At 2 wt.% CB, tensile strength reached 14.437 MPa, demonstrating a 66.4% enhancement, indicating efficient filler dispersion and stress transfer among the composite material. However, strength reduced marginally to 13.184 MPa at 3 wt.% CB, presumably attributed to filler agglomeration, resulting in localized stress concentrations and diminished interfacial bonding.

4.1.2. Elongation

Elongation at break as shown in Figure 1 reduced consistently with increasing recycled CB loadings which indicates the interaction between strength and flexibility. The pure PLA demonstrated the highest elongation, which is 87.97% demonstrating its inherent ductility. As the content of recycled CB loading increases the elongation is reduced to 30.57% on 1 wt.% loading while for 2 wt.% loading filler its 30.37% and 3 wt.% loading resulted 23.33% elongation at break. The firm recycled CB particles restricted the polymer chain mobility and triggered brittleness at higher loadings, supporting the negative correlation between filler concentration and ductility.

Both PLA-CB1% and PLA-CB2% composites likely resulting in similar adhesion between the recycled CB filler and PLA matrix, since well-dispersed CB at low concentration improves the bonding without affecting the polymer continuity [3]. The minimal filler content ensures that the reinforcement does not noticeably impact chain entanglement or flexibility, leading to similar elongation at break. Furthermore, elevated CB loading somewhat diminishes the crystallinity and chain mobility in PLA material, where it resulting in both composites exhibiting comparable ductility properties despite the increased filler content [4].

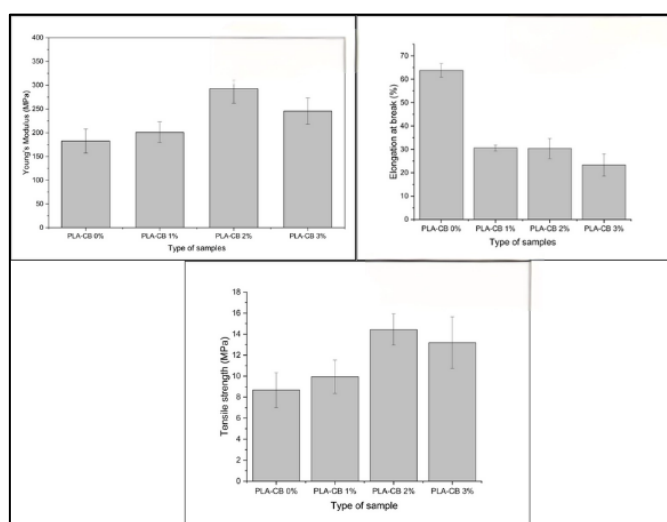


Figure1. Tensile testing of PLA–Carbon Black (CB) composites with varying filler loadings (0–3 wt.%).

Table 1 Tensile testing of PLA–Carbon Black (CB) composites with varying filler loadings (0–3 wt.%)

Sample	Tensile strength (MPa)	Elongation at break (%)	Young's Modulus (MPa)
PLA-CB 0 wt.%	8.673 ± 1.66	87.97 ± 9.24	182.43 ± 37.93
PLA-CB 1 wt.%	8.948 ± 1.58	30.57 ± 1.23	200.87 ± 27.59
PLA-CB 2 wt.%	14.437 ± 1.488	30.367 ± 4.325	292.67 ± 13.02
PLA-CB 3 wt.%	13.184 ± 2.45	23.33 ± 4.76	245.57 ± 56.37

4.1.3. Young's Modulus

As illustrated in Figure 1, the Young's modulus increases as the recycled CB loading increases up to 2 wt.% filler loading, which demonstrates the improved rigidity on the composites. The composite with recycled CB 2% has the highest modulus value, which is 292.67 MPa, where it suggests the significant improvement on interfacial bonding and efficient loading distribution. However, a reduction occurs on 3% loading at 245.57 MPa, indicating that the excessive filler created an agglomeration and micro-defects, which prevent stress transfer. Thus, 2 wt.% CB indicates the ideal filler loading on stiffness of PLA-CB composites.

The improvement of interfacial adhesion between the PLA matrix and recycled CB is an important component which helps to enhance the stiffness of the PLA-CB composites. Effective bonding promotes optimal stress transfer from the matrix to the filler, hence enhancing stiffness to the material [5]. The improved modulus in PLA-CB2% indicates that the CB particles are well-dispersed, hence reinforcing the matrix better when comparing to 1 wt.% filler loading. At 2 wt.% CB, the filler content is ideal for mechanical reinforcement, enhancing stiffness without inducing agglomeration.

Additionally, CB-induced chain immobilization and a slight increase in crystallinity enhance stiffness. The inflexible filler limits the movement of polymer chains, whereas enhanced packing at higher CB concentrations enhances load-bearing capability [6]. While 1 wt.% CB offers marginal enhancement, the enhanced matrix-filler interaction at 2 wt.% resulting in a significant rise in Young's modulus, however excessive loading by 3 wt.% loading induced agglomeration and diminished the performance.

4.2. Hardness

Hardness testing of the PLA-CB composite was done with 588-N load is demonstrated in Figure 2, showing a significant relationship between the recycled CB filler and the impact resistance. The pure PLA and PLA-CB1% samples both demonstrated at 162.08 MPa, which is a minimal improvement at low filler content.

As the loading of CB increases to 2 wt.%, a notable increase in hardness to 196.57 MPa was observed. This enhancement can be rationalized by improved dispersion of the recycled carbon black particles, potentially leading to a more effective stress transfer mechanism between the particles and the PLA matrix, thereby improving mechanism between the filler particles and the PLA matrix, thereby improving the mechanical properties [7]. The research indicates that a semi-continuous network of fillers can significantly limit the chain mobility of polymers, thus enhancing hardness and resistance to deformation [8]. This trend of the hardness of the PLA-CB composite continuous with 3% CB loading level, where the hardness further increases to 212.40 MPa, attributed to a denser packing of CB particles, which facilitates between load transfer and reinforces the structure of the composite [9].

The implications of these findings advocate for careful consideration of filler concentration and dispersion quality when developing PLA composites designed for applications that require high surface hardness and mechanical stability. The results highlight the necessity for effective mixing strategies to optimize filler distribution and maximize hardness contributions in the composite material.

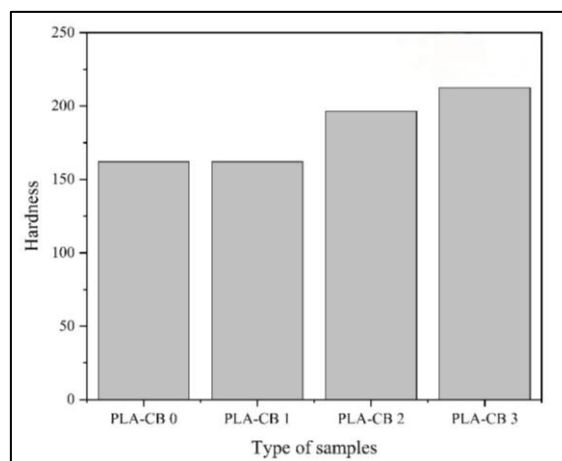


Figure 2. Variation of hardness with carbon black content in PLA-CB composites.

4.3. Electrical Conductivity

The electrical conductivity of PLA-CB composites testing using the Electrical Impedance Spectroscopy (EIS) at 1 MHz, showed a clear rise with the increase of recycled CB loading into the PLA composite as shown in Figure 3 and Figure 4. The pure PLA displayed conductivity attributed to its insulating properties. At 1 wt.% CB, the conductivity remained minimal, where it shows the percolation threshold is not achieved. Whereas, on PLA-CB 2 wt.% a significant increase was noticed, this indicates the initial establishment of conductive networks. The maximum conductivity output 0.1092 S/cm was indicated on 3 wt.% CB loading composite due to the formation of continuous conductive channels.

The result from FTIR stated in Figure 5 verifies the absence of substantial chemical integration between the PLA matrix and recycled CB filler, showing that the improvement of conductivity resulted from physical dispersion between the matrix and reinforcing filler rather than chemical modifications.

The measured conductivity values ranging from 0.03 to 0.10 S/cm of the PLA-CB composites, indicate that the composites are semiconductive material, which supporting their suitability for electrostatic discharge protection, antistatic applications or moderate electrical shielding. The CB functions as an efficient filler that improves the electrical performance of PLA without modifying its structure. The composites have semi-conductive properties, indicating the viability of recycled CB of electrically functioning, sustainable polymer materials.

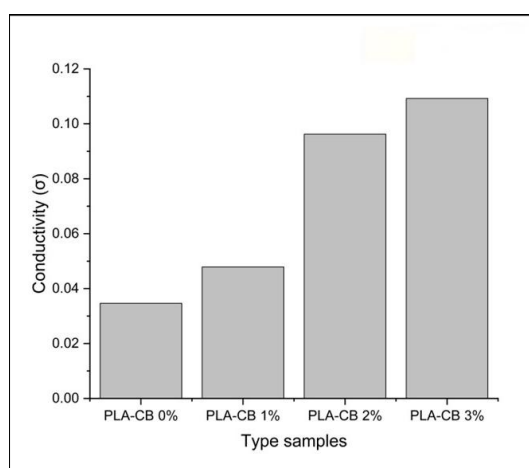


Figure 3. Electrical conductivity of PLA-CB composites measured at 1 MHz.

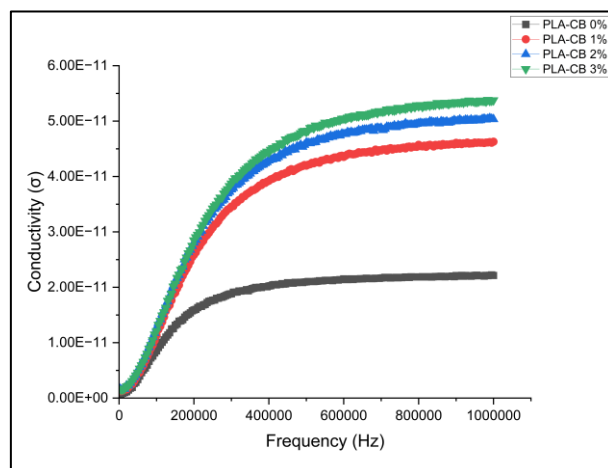


Figure 4. Electrical conductivity of PLA-CB composites as a function of frequency.

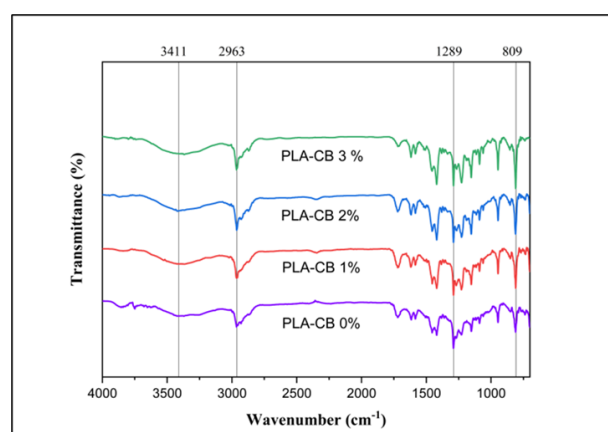


Figure 4. The Fourier-Transform Infrared (FTIR) of PLA-CB composites.

The FTIR spectra of pure PLA and PLA-CB composites (1, 2 and 3 wt.%) exhibit consistent characteristics peaks at 3411 cm^{-1} (O-H stretching), 2663 cm^{-1} (C-H stretching), 1289 cm^{-1} (C-O or C-O-C stretching) and 809 cm^{-1} (C-H), which no emergence of new peaks significant changes seen. This stability verifies the lack of chemical interactions between the PLA and recycled CB, proving that CB functions solely as physical filler. The minor decrease in the transmittance intensity with increasing in recycled CB loading is due to the intrinsic light-absorbing characteristics of carbon black rather than chemical alteration. The finding is supported by [10], [11] affirming that the inclusion of untreated CB does not modify the chemical structure of PLA but rather improves its mechanical properties via physical reinforcement and enhanced dispersion within the matrix.

4.4. Biodegradable

The biodegradability of PLA-CB composites is dependent upon the content of carbon black introduced during material fabrication. Soil burial test in this study showed that samples containing 1 wt.% CB exhibited the highest weight loss (2.75%) indicating a notable degradation response over 30 days [12-13]. This finding is consistent with literature suggesting that an optimal loading of carbon-based fillers can enhance biodegradability while maintaining material integrity. For instance, [9] reported

that maximum degradation occurred in PLA composites loaded with 1 wt.% CB, while degradation decreases with higher loadings due to the densification of the matrix [14].

The reductions in weight loss observed in samples with increased CB content, particularly the 3 wt.% level yielding only 0.36% degradation, can be attributed to the hydrophobic nature of the composite [15]. This hydrophobicity limits water absorption, a crucial factor for biodegradation, as bacteria and microorganisms require moisture to effectively colonize and break down organic materials [16]. Studies indicate that hydrophilic additives can facilitate moisture uptake and enhance microbial activity, thereby accelerating the degradation process.

Moreover, research has emphasized the relationship between hydrophobicity and microbial accessibility, stating that a denser matrix composition renders microbial colonization and moisture uptake less feasible. The significant reduction in degradation rates as carbon black content increases underscores the composite's resistance to biodegradation, correlating with observations that a low moisture absorption rate results in slower degradation in PLA and similar composites [17-18].

The degradation kinetics of the PLA-CB composites support the premise that carefully optimizing carbon black content

can lead to tailored biodegradability. As mentioned by [19] that the presence of additives in polymer matrices plays a crucial role in determining both mechanical properties and decomposition rates. Specifically, increasing the filler content beyond the optimal threshold decreased the

composites' susceptibility to biodegradation, emphasizing the fine balance needed to leverage the beneficial properties of filler additives [20].

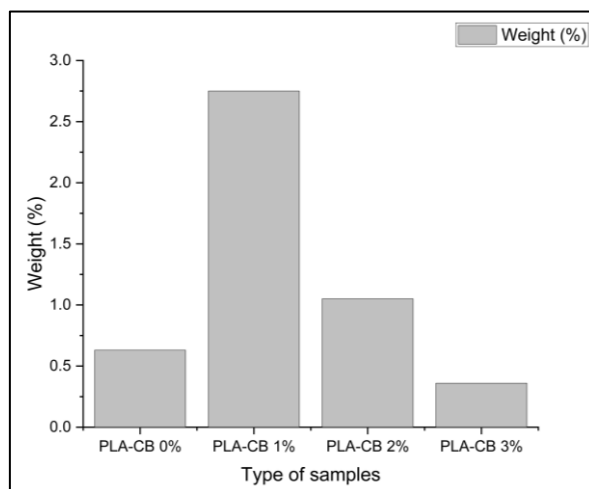


Figure 5. The biodegradable of PLA-CB 0%, PLA-CB 1%, PLA-CB 2%, and PLA-CB 3%.

5. CONCLUSION

This research demonstrated the levels of mechanical and electrical properties boost of PLA composites through the addition of recycled CB in its different proportions (0, 1, 2 and 3 wt.%). The findings showed that the presence of the CB increased the tensile strength, Young's modulus, and the Rockwell hardness to the level of optimal loading of 2 wt.% before effects began to reduce mechanical performance at higher loadings. Electrical conductivity improved significantly with the loading of the CB so that the composite with 3 wt.% of CB was the most conductive as the continuous conductive paths were created. FTIR studies showed that the PLA matrix did not undergo any significant chemical modifications, which means that changes were mainly physical.

Biodegradability however decreased with the rising concentration of CB content as indicated by decreasing weight loss in the soil burial test. It indicates a dilemma between an increase in functional properties and the preservation of the environment. The PLA-CB 2% was the best overall performing composition out of all tested as it had enhanced mechanical strength and acceptable conductivity as well as biodegradation. These results raise the possibilities of the PLA-CB composites, especially with 2% proportion of filler loading to be used on sustainable packaging, biodegradable electronic and sustainable engineering products.

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