Research Article

Sustainable Biocomposites from Oyster Shell and Bamboo Charcoal Reinforced Polybutylene Succinate (PBS) for Agricultural Applications

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Abstract

Large quantities of oyster shells and bamboo waste contribute to environmental pollution, highlighting the need for sustainable valorization strategies. In this study, green biocomposites based on polybutylene succinate (PBS) reinforced with thermally treated shell waste (TSW) at loadings of 0–15 wt% (in increments of 5 wt%) and bamboo charcoal (BC) at a fixed content of 3 wt% were developed. To the best of the authors' knowledge, no previous studies have reported on hybrid PBS composites incorporating both TSW and BC fillers. The mechanical properties and fracture surface morphology of neat PBS and the developed biocomposites were investigated. The results showed that the addition of BC maintained the impact resistance while significantly improving the tensile and flexural properties, stiffness, and hardness. Although the addition of TSW enhanced stiffness, rigidity, and tensile and flexural moduli, it reduced the impact resistance and flexural strength. Overall, the impact strength, tensile strength, flexural strength, and elongation at break tended to decrease with increasing TSW content, whereas the tensile and flexural moduli increased. Overall, the PBS–BC sample was identified as the most suitable material for nursery plant pots and mulch film due to its high impact and flexural strengths, along with a relatively moderate flexural modulus among the PBS composites. The hybrid PBS–BC–TSW05 biocomposite (5 wt% TSW and 3 wt% BC) was identified as the optimal hybrid formulation because of its balanced mechanical performance.

Keywords: Agro-waste, Animal waste, Bio-polymer, Mechanical properties, Polymer composite

1 Introduction

Plastics have been extensively used in the agricultural industry, providing solutions for arid regions, increasing crop yield, reducing the need for fertilizers, pesticides, and water, and enabling effective weed control [1]. These products include films for crop protection, soil conditioning, plant nurseries, water retention, and drainage or irrigation systems. However, conventional plastics are not biodegradable and instead fragment into microplastics, polluting soil and water ecosystems, especially over the long term [2]. Thailand is the largest exporter of agricultural products in ASEAN and was ranked 15th globally in 2025, with a reported growth of 3% in the first quarter of 2025, according to the Ministry of Agriculture and Cooperatives (MOAC) [3]. This high demand for agricultural production has intensified concerns regarding plastic waste accumulation. Therefore, biodegradable alternatives for agricultural applications are urgently required to replace conventional plastics, which degrade very slowly and contribute to persistent microplastic pollution. Consequently, the use of fast-degrading biopolymers has attracted significant interest. Polybutylene succinate (PBS) is a biodegradable polymer synthesized from succinic acid or its anhydride and butanediol. It is widely used in eco-friendly packaging, films, and disposable products, as well as in agricultural and medical applications [4], [5].

The escalating accumulation of waste from food and agricultural industries presents a significant environmental challenge. Global aquaculture production reached 130.9 million tonnes in 2022 [6]. In many countries, the large volumes of waste generated by the aquaculture industry, particularly from mollusc shellfish, cause severe environmental problems along coastlines. These wastes are primarily dumped at sea or sent to landfills, leading to detrimental effects on soils, water sources, and marine ecosystems and highlighting the need for sustainable management strategies [7]. Notably, only about 10% of the total oyster mass is utilized for food consumption or ornamental pearl production, while the remaining shell waste contributes to environmental pollution [8]. However, due to its high calcium carbonate ($CaCO_3$) content, shell waste shows strong potential for value-added applications, including soil acidification mitigation, immobilization of certain toxic heavy metals, and enhancement of crop yields [9]. Wang *et al.*, [10] demonstrated that the application of calcined oyster shell powder improved soil quality and crop yield, highlighting its promising potential for agricultural utilization. Qu *et al.*, [11] showed that thermally treated oyster shell powder (TSW) at 10 and 20 wt% in low-density polyethylene (LDPE) can provide a balanced combination of mechanical, thermal, and antibacterial properties. Similarly, Chandran *et al.*, [12] reported that the incorporation of seashell fillers into poly(lactic acid) (PLA) significantly enhanced the thermal stability and impact resistance of the resulting composites.

Another important agro-waste is bamboo charcoal (BC), a plant-derived alternative to carbon black produced through the pyrolysis of bamboo residues under anoxic conditions [13]. BC has been widely applied in agriculture due to its multiple benefits, including nutrient enrichment, soil pH adjustment, improved soil structure, enhanced root growth, and increased microbial diversity. Moreover, the black color of BC is advantageous for horticultural applications, as it enhances light absorption, suppresses weed growth, retains soil moisture, and conserves solar heat, thereby promoting root development and reducing nutrient leaching during rainy seasons [14]. Srisuk *et al.*, [15] found that the incorporation of 2.82 wt% BC in PLA provided an optimal balance of mechanical properties and produced a dark black coloration. Similarly, Ho *et al.*, [16] reported that PLA reinforced with BC exhibited increases of 43%, 99%, and 52% in tensile strength, flexural strength, and ductility index, respectively. These improvements were attributed to the uniform dispersion, high aspect ratio, and large surface area of BC particles, with tensile strength increasing up to a BC content of 7.5 wt%. Therefore, the development of biodegradable composites incorporating such waste-derived fillers represents an eco-friendly and sustainable approach for agricultural applications. In this study, PBS was selected as the polymer matrix and reinforced with oyster shell waste and BC fillers to produce biodegradable biocomposites capable of degrading in soil. Such materials can be ploughed directly into farmland, reducing labor and disposal costs while simultaneously releasing beneficial nutrients during degradation [17]. For example, biodegradable nursery pots used for seedling cultivation can be directly planted into soil without restricting root growth, thereby eliminating post-use plastic waste and minimizing microplastic accumulation in agricultural soils [18].

Previous studies have primarily focused on the use of single-type fillers, such as oyster shell waste or BC, reinforced in polymer matrices. For example, Huang *et al.*, [19] studied eco-friendly PBS/bamboo carbon composites in which bamboo carbon was utilized as a lubricant for wear applications. Shen *et al.*, [20] investigated PBS composites reinforced with oyster shell powder using a silane coupling agent. However, the combined use of TSW and BC as hybrid fillers in biodegradable polymer matrices remains largely unexplored. Furthermore, most reported studies incorporate BC in powder form, while studies involving BC in the form of a masterbatch are still limited.

This study aims to investigate the use of TSW and BC as reinforcing fillers in a PBS matrix to develop black biodegradable composites for agricultural and packaging applications, including mulch films, plant nursery containers, and single-use plastic products. BC was produced and incorporated in the form of a masterbatch. The BC content was fixed at 3 wt%, while the TSW content was varied from 5 to 15 wt% in increments of 5 wt%. The appearance and mechanical properties of the developed biocomposites were evaluated in terms of tensile strength, flexural strength, impact resistance, and hardness.

2 Materials and Methods

2.1 *Materials*

The used biodegradable polymer is bio-based polybutylene succinate (PBS) with grade of FZ91PM, purchased by PPT MCC Biochem Co., Ltd. The PBS had a density of 1.26 g/cm^3, a melt flow index of 5 g/10 min (190 °C, 2.16 kg), and a melting point of 115 °C. Bamboo charcoal (BC) powder was obtained from Charcoal Home, containing 86% fixed carbon with a pH value of 8-10 and an average particle size of 300 mesh. Oyster shell powder from shell waste, with an average particle size of 250 mesh, was procured from Bansuan Nai Ruasom. The powder exhibited an off-white color and had a pH value of 8-10. Modified polylactic acid, Scona TPPL 5112, from BYK-Chemi GmbH was used as a dispersing and compatibilizing agent. BC powder (300 mesh) and oyster shell waste powder (250 mesh) were selected because the highest fraction of particles passed through these sieve sizes.

2.2 *Material preparation*

For the preparation of the BC masterbatch, BC powder was first mixed with the PBS matrix to produce a masterbatch (MB). Prior to mixing, the BC powder was dried at 120 °C for 48 h to remove moisture. The BC masterbatch, consisting of 30 wt% BC, 30 wt% compatibilizing agent, and 40 wt% PBS, was prepared using an internal mixer at 150 °C for 30 min at a screw speed of 65 rpm.

TSW powder was prepared by sieving oyster shell waste powder through a 250-mesh screen and calcining it in the muffle furnace (FO310, Yamato Scientific) at 900 °C for 5 hours to convert $CaCO_3$ into CaO ($CaCO_3$ + Heat → CaO + CO_2). Rosli et al. [21] confirmed that calcination at 900 °C for 5 h efficiently converts $CaCO_3$ into CaO. Figure 1 presents oyster shell waste powder before and after thermal treatment.

Figure 1: Oyster shell waste powder before (left) and after thermal treatment (right).

Table 1: Composition of PBS biocomposites.

Bio-composites	PBS (%)	TSW (%)	Compatibilizing agent (%)	BC MB (phr)
PBS	100	0	0	0
PBS-BC	100	0	0	10
PBS-TSW-10	87	10	3	0
PBS-BC-TSW-05	95	5	0	10
PBS-BC-TSW-10	90	10	0	10
PBS-BC-TSW-15	85	15	0	10

The diagram of BC-TSW-PBS composite preparation is shown in Figure 2. Before extrusion of biocomposites, all materials were dried at 80 °C for 5 h and then mixed according to the 6 formulations, including neat PBS, as shown in Table 1. The BC masterbatch was incorporated at 10 phr during mixing, corresponding to 3 wt% BC in the final biocomposites. For the PBS–TSW-10 composite without BC masterbatch, the compatibilizing agent was directly added during the mixing process. For the other biocomposites, the compatibilizer was introduced via a 10 wt% BC masterbatch containing 30 wt% compatibilizing agent, resulting in a final compatibilizer content of 3 wt% in the composites. Accordingly, the compatibilizing agent content was maintained at 3 wt% in all biocomposites. This concentration was selected based on the work of Kuciel et al. [22], who reported that the incorporation of 3 wt% Scona TPPL 5112PA effectively enhanced the interfacial properties of bioPET composites reinforced with mollusc shells. Each composite mixture was first hand-blended and then extruded by a co-rotating twin-screw extruder (Xinda Model RSHJ 35–40). The extrusion process was carried out at a screw speed of 285 rpm, with the five-barrel zones from the feed zone to the metering zone set to 100 °C, 140 °C, 150 °C, 150 °C, and 150 °C, respectively, and the die temperature maintained at 150 °C. Samples for mechanical testing were prepared by a compression molding process (Carver, CMG3OH-12-ASTM-X, USA) at a temperature of 170 °C. The materials were preheated for 3 min, after which the pressure was gradually increased to 5 bar and maintained for 5 min. All PBS and PBS composite samples are shown in Figure 3.

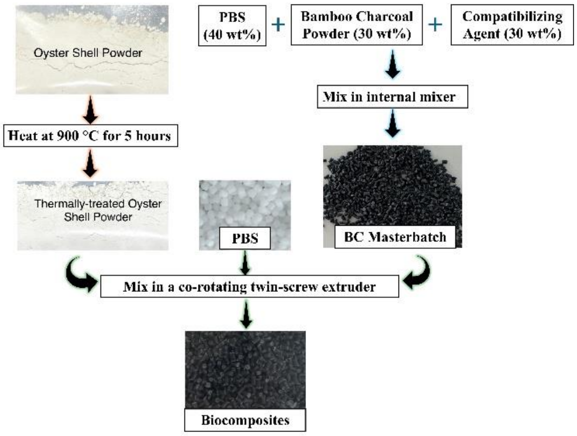

Figure 2: Diagram of BC-TSW-PBS composite preparation.

2.3 *Test methods*

2.3.1 Scanning Electron Microscopy (SEM)

The impact-fractured surfaces of neat PBS and the biocomposites were characterized using a Phenom Pharos G2 desktop field-emission scanning electron microscope (FEG-SEM, Thermo Scientific).

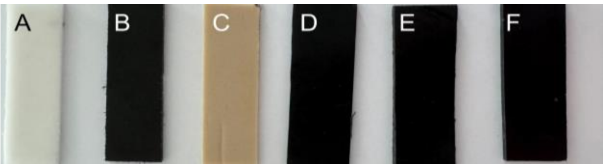

Figure 3: Photographs of A) PBS, B) PBS-BC, C) PBS-TSW10, D) PBS-BC-TSW05, E) PBS-BC-TSW10, F) PBS-BC-TSW15.

2.3.2 Impact test

The impact strength of neat PBS and the biocomposites was measured in accordance with ASTM D256 using the Izod impact test. A ZwickRoell HIT5.5P impact tester (ZwickRoell GmbH & Co. KG, Germany) was employed. The samples were compression-molded and machined into bar-shaped specimens with dimensions of 12.70 × 64.00 × 3.00 mm. All specimens were V-notched and tested using a hammer with an impact energy of 2.75 J. At least five repetitions were tested for each composition.

2.3.3 Flexural test

To evaluate the flexural strength and modulus, five specimens of each composition were prepared by compression molding and machined into rectangular specimens with dimensions of 12.7 × 127.0 × 3.0 mm. Flexural testing was conducted in accordance with ASTM D790 using a universal testing machine (Cometech QC-506M1-204, Taiwan) equipped with a 10 kN load cell. The support span was set to 48 mm, and the crosshead speed was maintained at 2 mm/min. At least five specimens were tested for each composition.

2.3.4 Tensile test

The tensile strength, tensile modulus, and elongation at break of each composite were evaluated in accordance with ASTM D638 using a universal testing machine (Cometech QC-506M1-204, Taiwan) equipped with a 10 kN load cell. The specimens had dimensions of 165.0 × 13.0 × 3.0 mm with a gauge length of 50 mm. The initial distance between the grips was set to 115 mm, and the crosshead speed was maintained at 2 mm/min. A minimum of five specimens were tested for each composition.

2.3.5 Hardness test

The indentation hardness of the composites was measured using a Shore D durometer (Teclock GS-720G, Japan) in accordance with ASTM D2240. A minimum of five measurements was taken for each composition.

3 Results and Discussions

3.1 *Fractured surface morphology*

The fractured surfaces of neat PBS and its biocomposites are shown in Figures 4 and 5. The surface of PBS appears relatively smooth and uniform compared with that of the biocomposites, exhibiting a homogeneous structure and a more ductile fracture surface. The addition of TSW and BC fillers led to the formation of microvoids, with the number of voids increasing as the filler loading increased due to poor adhesion between the fillers and the PBS matrix, as shown in Figure 4. With the addition of 10% TSW, the fracture morphology became noticeably rougher and showed spherical cavities associated with void formation, as highlighted by the red circle in Figure 5. Similarly, the incorporation of BC into PBS (PBS–BC sample) resulted in a roughened fracture surface. Further increases in TSW content led to higher fracture surface roughness of the biocomposites and increased agglomeration of the TSW fillers due to the high surface tension of the filler particles [23].

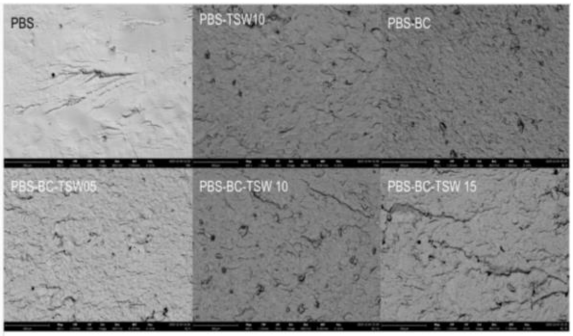

Figure 4: SEM micrographs of PBS and biocomposites with magnification 400x.

Among the TSW/BC composite samples, PBS–BC–TSW05 exhibits the least surface roughness, displaying only fine-scale features and small void formations characteristic of brittle fracture. In contrast, the PBS–BC–TSW10 sample shows greater roughness with numerous voids distributed throughout the fractured surface due to poor adhesion between fillers and matrix [24]. Moreover, PBS–BC–TSW15 exhibits a highly irregular and rough morphology, with prominent void formation visible across the fractured surface.

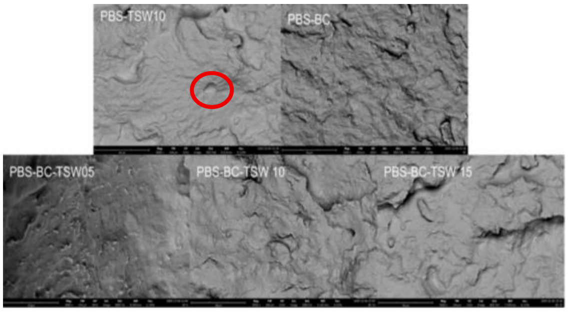

Figure 5: SEM micrographs of PBS and biocomposites with magnification 5000x.

3.2 *Mechanical properties*

The mechanical properties of PBS and all PBS biocomposites were examined to evaluate the influence of BC and TSW at various loadings. For impact strength, neat PBS exhibited the highest impact strength at 3.93 kJ/m², followed by the PBS–BC sample at 3.88 kJ/m² as presented in Figure 6. This slight decrease is attributed to the increased brittleness introduced by the BC powder [25]. The incorporation of TSW into the PBS matrix led to a sharp reduction in impact strength, likely due to the agglomeration of TSW particles, as observed in the SEM results, which diminished the composite's ability to absorb impact energy [26], [27]. Among all samples, PBS–TSW10 showed the lowest impact strength, which can be linked to void formation within the sample, as evidenced by the SEM micrographs. As the TSW loading increased, the impact resistance progressively decreased. However, the addition of BC to the TSW/PBS biocomposites provided a slight improvement in impact resistance. For instance, the PBS–BC–TSW10 sample exhibited a slightly higher impact strength than the PBS–TSW10 sample, indicating that the incorporation of BC can partially compensate for the reduction in toughness caused by TSW, demonstrating a synergistic hybrid effect between BC and TSW.

The flexural strength and modulus of PBS and its biocomposites are presented in Figure 7. Neat PBS exhibited the lowest flexural properties. The incorporation of fillers, both BC and TSW, enhanced the flexural strength and modulus of the PBS biocomposites. The PBS–BC–TSW05 sample showed the highest flexural strength, likely due to the synergistic reinforcing effect of BC and TSW. However, BC contributed more effectively to flexural reinforcement than TSW; therefore, increasing the TSW content resulted in a reduction in flexural strength. Similarly, Li *et al*., [25] reported that the flexural strength and modulus of the composites were enhanced with increasing BC content. In terms of flexural modulus, PBS also displayed the lowest value of 514.37 MPa, whereas the PBS–BC sample demonstrated the highest modulus of 1050.56 MPa. This improvement is attributed to the BC particles, which increased the rigidity of the biocomposites [25]. Although the addition of TSW led to a slight improvement in flexural modulus, the values remained lower than those of the PBS–BC sample. This may be due to poor TSW dispersion within the PBS matrix, as evidenced by the SEM results, which restricted its reinforcing efficiency [28]. Increasing TSW content tended to increase the stiffness of the biocomposites, leading to a gradual rise in flexural modulus due to high $CaCO_3$ content [28], [29]. There is no positive correlation between flexural strength and flexural modulus. However, Liao *et al*., [30] found a direct relationship between flexural performance and waste oyster shell powder in a cementitious composite. Therefore, according to the results, a decrease in flexural strength but an increase in flexural modulus as the concentration of TSW increased suggested that the addition of TSW may reduce the flexibility of the composite.

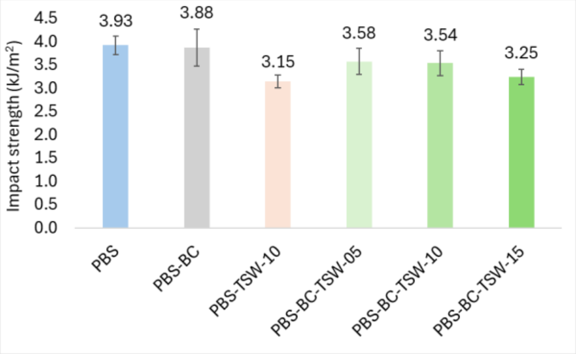

Figure 6: Impact strength of PBS and biocomposites.

Figure 8 shows that the samples with the highest tensile strength were PBS and PBS–BC, with values of 37.05 and 37.08 MPa, respectively. The addition of BC did not significantly reduce the tensile strength of PBS; however, the incorporation of TSW led to a decrease in tensile strength, likely due to TSW filler agglomeration and poor dispersion within the PBS matrix. As observed in the SEM micrographs, these agglomerates hindered effective stress transfer at the filler–matrix interface, thereby negatively affecting the mechanical properties [28], [31]. Consequently, PBS composites with increasing TSW content exhibited progressively lower tensile strength. However, the incorporation of BC into the TSW/PBS composites improved the tensile strength, indicating a positive reinforcement for tensile strength contribution from BC.

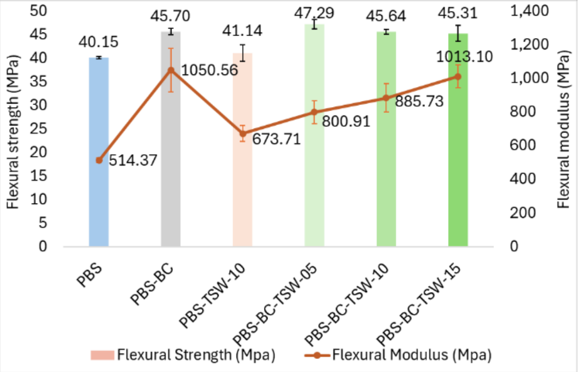

Figure 7: Flexural strength and modulus of PBS and biocomposites.

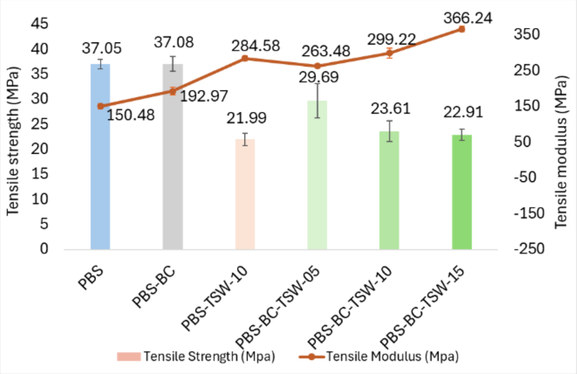

Figure 8: Tensile strength and tensile modulus of PBS and biocomposites.

The lowest tensile modulus among all samples was observed for neat PBS. The addition of fillers enhanced the tensile modulus of the biocomposites because both BC and TSW increased the rigidity of the composite due to their inherently high stiffness [15], [26]. A similar trend was observed by Benaichouba *et al*., [26], where the tensile modulus increased with increasing oyster shell particle concentration. In particular, the $CaCO_3$-rich composition of TSW contributed substantially to the improvement in tensile modulus [32]. As a result, the tensile modulus of the BC/TSW/PBS composites increased significantly with increasing TSW content.

The elongation at break of PBS and its biocomposites corresponded closely with their impact strength trends. As shown in Figure 9, neat PBS exhibited the highest elongation at break at 60.26%. The addition of BC significantly reduced the elongation to 30.03% due to the increased stiffness imparted by the BC particles, which was consistent with the findings reported by Srisuk *et al.*, [15]. When TSW was added, the elongation at break decreased further to 9.30%, as the high rigidity of TSW sharply reduced the ductility of the biocomposites [26]. Consequently, higher TSW content resulted in progressively lower elongation values. However, the incorporation of BC into the TSW/PBS composites slightly improved the elongation at break compared with the corresponding TSW-only formulations, indicating a partial compensating effect.

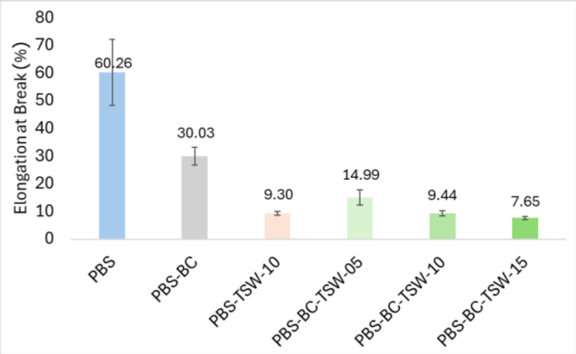

Figure 9: Elongation at break of PBS and biocomposites.

The hardness value of neat PBS was the lowest among all samples (Figure 10). The incorporation of fillers increased the hardness due to the inherently higher hardness of TSW and BC, which enhanced the stiffness of the biocomposites [28]. TSW exhibited greater effectiveness in improving hardness than BC because of its high $CaCO_3$ content [32], as $CaCO_3$ inherently possesses high hardness [23]. As the TSW loading increased, the hardness of the biocomposites also increased. However, overall, the hardness values of the biocomposites were statistically comparable.

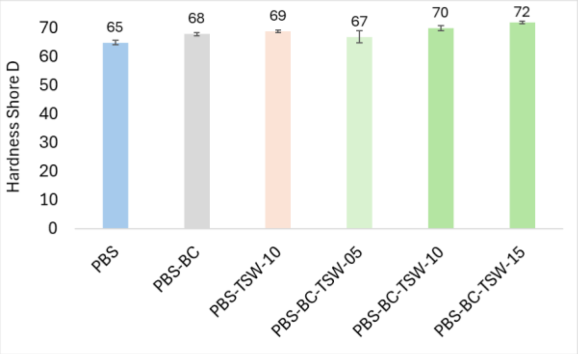

Figure 10: Hardness shore D of PBS and biocomposites.

Table 2: Comparison of PBS-based composites.

Filler	Tensile strength (MPa)	Tensile Modulus (MPa)	Elongation (%)	Ref.
BC 3 wt%	37	193	30	Present work
BC 3 wt% TSW 5 wt%	30	263	15	Present work
Oyster shell powder 5 wt%	13	480	120	[20]
Bamboo fiber10 wt%	25	604	15	[33]
Carbonaceous biochar 5 wt%	17	390	247	[34]
Miscanthus straw biochar 5 wt%	29	150	20	[35]

Table 2 presents a comparison between the PBS biocomposites developed in the present study and other reported PBS-based biocomposites. It is clearly shown that the PBS biocomposites in the present work (PBS–BC and PBS–BC–TSW-05) demonstrate competitive mechanical properties in terms of tensile strength, tensile modulus, and elongation at break compared with other biomass-derived fillers, particularly oyster shell and biochar powders.

The PBS biocomposite containing 3 wt% BC exhibited the highest tensile strength of 37 MPa among the PBS-based biocomposites listed in Table 2. Moreover, it showed higher tensile modulus and elongation at break than PBS reinforced with 5 wt% miscanthus straw biochar. Although some previous PBS-based biocomposites showed higher tensile modulus values, those systems generally exhibited either lower tensile strength or increased brittleness.

These results indicate that BC can effectively reinforce the PBS matrix even at relatively low filler loading without severely reducing the ductility of PBS.

In addition, the hybrid PBS biocomposite containing 3 wt% BC and 5 wt% TSW maintained balanced tensile properties while still preserving acceptable tensile strength compared with other PBS-based biocomposites. It also exhibited higher tensile modulus and more balanced elongation compared with the PBS biocomposite containing only 3 wt% BC. These results highlight the potential of BC and TSW as sustainable reinforcing fillers for PBS-based biodegradable composites.

Future research should therefore focus on improving the tensile strength through the incorporation of anti-hydrolysis agents [36]. In addition, further investigations on thermal stability and degradation temperature are required, as these properties may be improved with the addition of shell waste fillers, as reported by Chandran et al. [12], and bamboo charcoal (BC), as confirmed by Srisuk et al. [37]. The thermal behavior of the developed composites should be examined in more detail, particularly in terms of crystallinity changes. Moreover, comprehensive studies on biodegradation behavior, soil burial degradation, water absorption, environmental durability, and the associated environmental impacts are still necessary.

4 Conclusions

The influence of bamboo charcoal (BC) at a fixed content of 3 wt% and thermally treated shell waste (TSW) at loadings of 5–15 wt% on the mechanical properties and surface morphology of PBS-based biocomposites was investigated. A small amount of BC incorporated into the PBS matrix significantly enhanced flexural and tensile properties as well as hardness, without reducing impact resistance. In contrast, the addition of TSW, which contains a high $CaCO_3$ content, increased the rigidity and stiffness of the composites. However, increasing TSW content led to reductions in impact strength, flexural strength, tensile strength, and elongation at break, while tensile modulus, flexural modulus, and hardness increased. The hybridization of BC and TSW improved impact resistance as well as flexural and tensile properties compared to composites containing only TSW. Overall, BC acted as a more effective reinforcement in PBS than TSW. However, the synergistic use of BC and TSW proved beneficial, suggesting that hybrid PBS–BC–TSW composites, especially PBS–BC–TSW05, offer balanced mechanical properties and suitability for plant-related product applications owing to the high base value of TSW.

The developed PBS/BC/TSW hybrid biocomposites show potential for use in agricultural applications such as mulch films, nursery pots, and other short-life biodegradable products where dark coloration and moderate mechanical performance are required. However, this study has several limitations, including long-term durability, biodegradation behavior in soil environments, and water absorption characteristics, which were not investigated. In addition, the thermal behavior and crystallization mechanisms of the composites require further clarification.

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Author Contributions

P.S.: investigation, methodology, data curation, data analysis, writing an original draft; S.M.: investigation, methodology, data curation; R.S.: conceptualization, investigation; L.T.: conceptualization, research design, data analysis, writing an original draft, writing, reviewing and editing, project administration. All authors have read and agreed to the published version of the manuscript.

Conflicts of Interest

The authors declare no conflict of interest.

Declaration of generative AI and AI-assisted technologies in the writing process

The authors utilized the ChatGPT tool to enhance the language and readability of the manuscript.

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