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Potential Use of Paddy Straw as Filler in Poly Lactic Acid/Paddy Straw Powder Biocomposite: Thermal and Mechanical Properties

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Abstract

Paddy Straw Powder (PSP) show potential as a new reinforcement based-natural fibre. A Haake internal mixer was used to incorporated paddy straw powder (PSP) into polylactic acid (PLA). Polylactic acid (PLA)/paddy straw powder biocomposite was prepared using constant rotor speed (60 rpm) for 14 minutes at 180°C. The effects of paddy straw powder content (5-20 wt %) on mechanical and thermal properties of the biocomposites were investigated. The tensile strength of the biocomposites was above 30 MPa up to 15 wt.% of PSP whereas the elongation at break was ranged between 2-3 % with the incorporation of PSP up to 15 wt.%. Modulus elasticity was increased by increasing the paddy straw powder content. Differential scanning calorimetry (DSC) results have demonstrated a minor effect of the rice straw on thermal behavior of PLA resin Thermogravimetry, analysis (TGA) demonstrated that thermal stability of PLA/PSP biocomposites is reduced by the incorporation of PSP.

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Nomenclature

PSP	Paddy Straw Powder
PLA	Polylactic Acid
SEM	Scanning Electron Microscopy
TGA	Thermogravimetry Analysis
DSC	Differential Scanning Calorimetry
Eb	Elongation at break
wt	Weight
μm	micrometer
cm	centimeter

1. Introduction

Recently, researches have shown increasingly remarked interest in biocomposite material as those materials are eco-friendly, biodegradable and renewable. In the past, biocomposites were produced by incorporating filler into conventional polymer. Most polymer composites are difficult to recycle or incur substantial cost for disposal.¹ In an effort to supplant the petroleum-based polymers and reduce waste-related environmental problems, biopolymers are the best candidate due their renewability, biodegradability and commercial viability.

Polylactic acid (PLA) is among the commodity biopolymers that are comparable in terms of properties and performances. Poly lactic acid (PLA) is produced from renewable resources that have become a useful material due to its good clarity, high strength and moderate barrier properties. PLA was frequently used for biodegradable packaging material. However, numerous tests have shown that PLA is also suitable as matrix for the embedding of fibres in composites.²⁻⁴

Paddy straw is one of the lignocellulosic filler, which is an agricultural coproduct found abundantly in Malaysia, since paddy, in Malaysia, is the first important crop in term of acreage.⁵ Paddy straw in Malaysia is presently disposed of mostly by open burning. This results in the release of various pollutants affecting the environment, weather and local communities. Yet, there are several potential uses for rice straw which can still be explored and developed to benefit Malaysia's rural economy. The lignocellulosic filler exhibits some excellent properties compared to mineral filler (e.g. calcium carbonate, talc, mica and kaolin) such as low cost, renewable, minimal health hazard, low density, less abrasion to machine, certainly biodegradability and environmentally friendly.⁶⁻¹¹

One of the most important aspects of composite manufacture is to achieve adequate adhesion between fibre and the polylactic acid matrix. In this work, we report studies on the effects of filler loading on the thermal characteristics and mechanical properties of PLA/PSP biocomposites. Scanning electron microscopy (SEM) studies were made to determine the failure mechanisms of the composites.

2. Materials and methods

Poly lactic acid or PLA type 4032D in pellet form was supplied by ADV Sdn. Bhd. The polymer has a specific gravity of 1.24 g/cm³. Paddy straws were taken from paddy fields at Mata Ayer, Perlis and ground to obtain average size of 68 μm.

Biocomposites were prepared in a Haake Polydrive with Rheomix R600/610 with the different loading of PSP (5-20 wt. %). Mixing was done at temperature 180°C and rotor speed 60 rpm for 14 min. The PLA was first charged into the mixer to start the melt mixing. After 2 min, PSP was added and the mixing was continued for another 12 min. The PLA/PSP biocomposites were compression molded using a hot press. The compression- molded procedures involved preheating for 3 min, followed by compressing for next 3 min at 180°C, and subsequent cooling under pressure for 1 min. Molded samples were then cut into dumb-bell shapes according to ASTM 638 before being analyzed.

Tensile test, Scanning Electron microscopy, TGA and DSC were used to characterize the mechanical and thermal properties of this biocomposite. Thermogravimetric analysis (TGA) was performed to record the weight loss as function of temperature. Samples were heated from 30 °C-600 °C with the heating rate of 10 °C/min under flow of

nitrogen to prevent oxidation. The preparation and parameters of the DSC test were based on ASTM D3418-03.

3. Results and Discussion

3.1. Tensile Properties

Tensile properties are a very important parameter in terms of characterization of biocomposite. In this case, variation of tensile strength, the elongation at break and modulus elasticity of neat PLA and biocomposites are shown in Fig. 1(a)-(c).

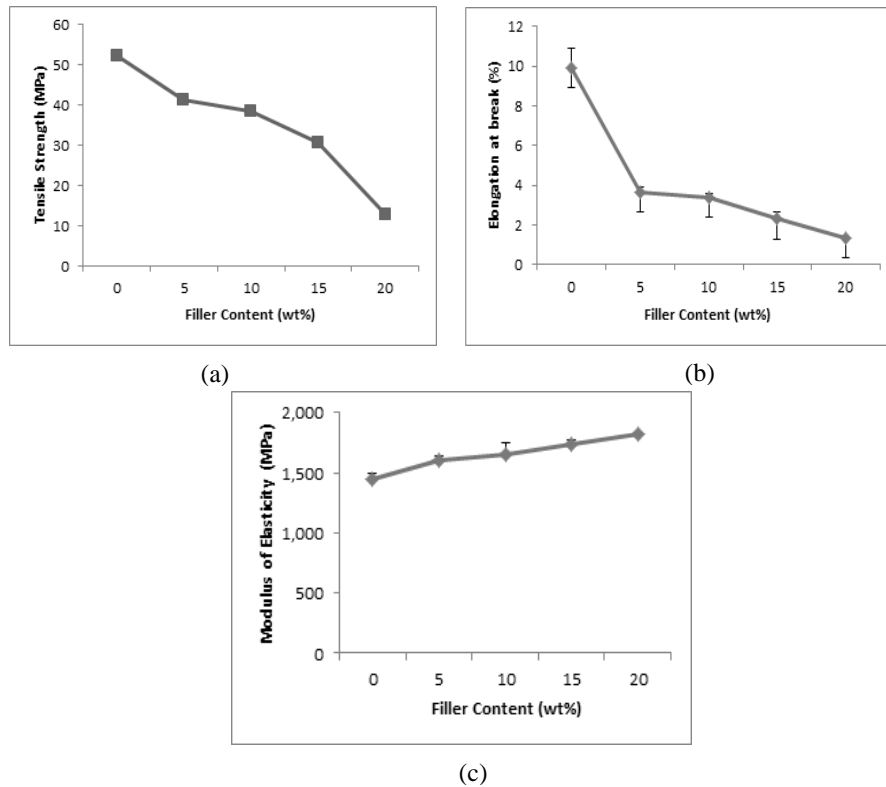


Fig. 1. (a) Tensile strength; (b) elongation at break; (c) modulus of elasticity of PLA/PSP biocomposite.

From Fig.1, neat PLA shows higher yield strength than its reinforcing filler biocomposites. The tensile strength of PLA/PSP biocomposites decreases with the increase of PSP content. This may due to dispersed phase loading increase, the effective cross sectional area of continuous phase is reduced, subsequently resulting in a decrease of tensile strength. Moreover the imperfect distribution of the filler through polymer matrix, as well as very poor adhesion between the polymer resin and filler may contribute for this finding.¹²⁻¹⁶

The above findings, related to very poor adhesion between the polymer matrix and filler, are confirmed by SEM micrographs of the fractured samples given in Fig. 2(a)-(c). It can be seen that the PSP are completely pulled out of the polymer resin and empty voids and holes are visible.

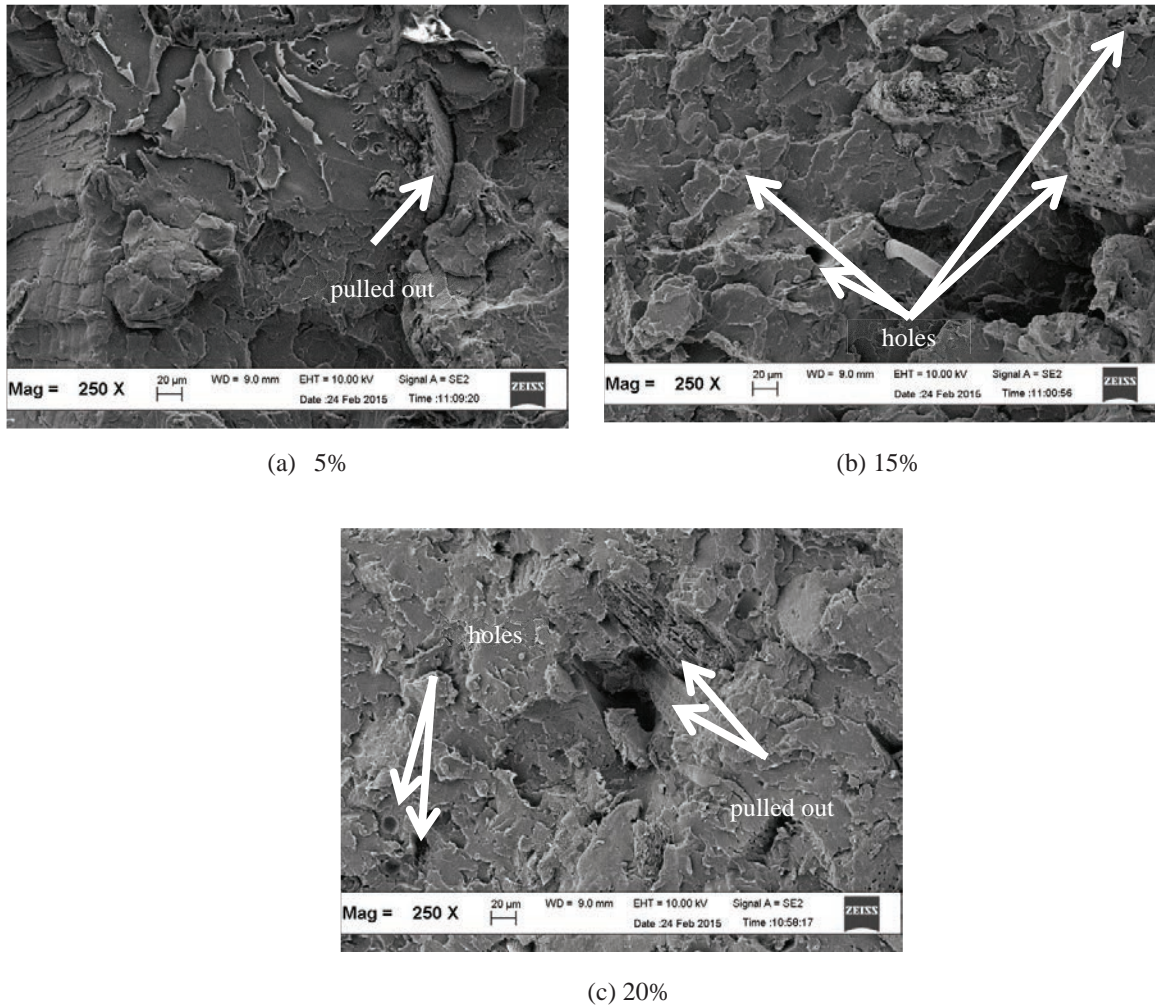


Fig. 2. SEM micrograph of biocomposites with (a) 5 wt. %; (b) 15 wt. %; (c) 20wt % of PSP.

Overall, the tensile modulus of every PLA/PS biocomposites sample in this experiment is higher than the pure PLA sample (Figure 1 (c)). The increment in the modulus of elasticity of PLA/PSP biocomposites was due to the reducing of chain mobility in more filler content; which allows high stiffness in its composites.¹⁷ The presence of fillers restricted the chain mobility of the PLA, adding to the rigidity of the composites. Elongation at break (E_b) of PLA/PSP biocomposites was decreased (Figure 1(b)) with filler content and its corresponding to the inherent rigidity of the PSP particles, leading to reduction of ductility of the biocomposites. An increase modulus in natural agricultural fiber is a common since the fiber is known to act as rigid filler that increases the stiffness.¹⁸ Besides, this is because increasing of PSP loading in PLA matrix had increased the interaction between the filler which also known as filler-filler interaction.

3.2. Thermal Properties

DSC analysis was used to determine the variation in thermal properties of the PLA/PSP biocomposites. The melting and cooling data of the biocomposites are shown in Table 1. T_c of the biocomposites decreased when compared to pure PLA ($T_c = 172.03^\circ\text{C}$). This is because fibers are definitely playing an effective nucleating role to speed up the PLA crystallization process thus enhancing the crystal rate. T_m of the biocomposites decreased when

compared to pure. This was due to the lower ΔH_m and T_m listed in Table 1. The degree crystallinity has a decreasing tendency with the increase of the filler content. This may attributed to the increase in amorphosity of the materials.¹⁹

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Table 1. DSC data for PLA and biocomposite.

Samples	T_c (°C)	T_m (°C)	ΔH_m	Crystallinity (%)
Pure PLA	172.03	112.05	17.13	18.42
PLA/5PSP	170.54	93.59	35.64	38.32
PLA/10PSP	169.84	94.34	32.91	35.38
PLA/15PSP	162.43	92.26	29.34	31.54
PLA/20PSP	160.27	89.82	29.27	31.47

Thermal degradation of PLA /PSP biocomposites has been explored in terms of weight loss by TGA carried in air as shown in Table 2. The table illustrated thermal stability of PLA/PSP biocomposites is decreased by PSP that can clearly seen in Table 2. A significant drop in temperature can be seen from 5% to 20% of PSP with PLA. This result is harmony with the study of tensile properties. This is because the incorporation of PSP loading up to 20% caused a significant reduction in the interaction between PSP and PLA. Besides, this could be due to the low thermal stability of the filler which is the PSP.²⁰

Table 2. TGA data for PLA and biocomposite.

Sample	T_{max} (°C)	Total weight loss at 600 °C (%)
PLA	344.6	96.1
PLA/5PSP	287.2	93.3
PLA/10PSP	286.4	92.5
PLA/15PSP	280.6	91.2
PLA/20PSP	279.8	90.1

4. Conclusion

The mechanical and thermal properties of pure PLA and different formulation of PLA/PS biocomposites were investigated. The tensile strength and elongation at break of PLA/PSP biocomposites decreased with the increasing of filler content. However the modulus of elasticity of PLA/PSP biocomposites was increased with filler content. The study of interfacial adhesion, which is well known problem for natural fibres and synthetic polymers also shows that adhesion, needs to be improved to optimize the mechanical properties of the PLA/PSS biocomposites. The SEM micrographs show the poor interfacial interaction between PSP and PLA matrix. Crystallinity of the biocomposites reduce by addition of PSP hence make it easier for biodegradation. The total weight loss of PLA/PSP biocomposites reduced 600 °C.

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