WOOD RESEARCH **doi.org/10.37763/wr.1336-4561/71.4.618633**
e-ISSN 2729-8906
71(4): 2026 618-633 pp.

EVALUATING THE APPLICABILITY OF PROTEINACEOUS INTERFACIAL BINDERS IN POLYLACTIC ACID-LIGNOCELLULOSIC FILLER COMPOSITES

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(RECEIVED AUGUST 2026)

ABSTRACT

This study evaluated the applicability of protein-based interfacial binders in the production of polylactic acid (PLA)-lignocellulosic filler composites. The composites were fabricated by blending PLA with radiata pine (RDP), hardwood scrub (MIX), cypress bark powder (CYP-B) powder as fillers, hydrogenated castor oil (HCO) as a dimensional stabilizer, and black soldier fly (BSF) powder, or a freeze-dried proteinaceous adhesive resin (PAR) powder formulated from BSF hydrolyzates as interfacial binders. Composites fabricated with PLA, CYP-B, PAR-1, and HCO showed superior impact/bending strengths and lower moisture absorption rate compared to RDP- or MIX-filled composites. A formulation of 50 wt% PLA, 40 wt% CYP-B, 5 wt% PAR, and 5 wt% HCO showed the best overall performance, outperforming composites using maleic anhydride as a conventional synthetic interfacial binder. Overall, PAR is a promising substitute for expensive, hazardous synthetic binders, and CYP-B is a viable waste-valorization filler with environmental and health benefits.

KEYWORDS: Polylactic acid, lignocellulosic filler, composites, protein-based interfacial binders, cypress bark.

INTRODUCTION

Fossil fuel-based plastics, when landfilled, cause the leaching of endocrine-disrupting chemicals and generate microplastics during degradation (Kabir et al. 2023; Stevens et al. 2024). Incineration, as an alternative disposal option, can cause serious environmental pollution emitting dioxins, volatile organic compounds, and greenhouse gases that are harmful to human health (Li et al. 2020). Considerable effort has been made to address these problems by

developing and commercializing bio-based plastics such as poly-lactic acid (PLA), produced by polymerizing lactic acid derived from the fermentation of plant-based feedstock, such as corn and sugarcane (Pilla et al. 2008). PLA-based plastics are environmentally friendly and non-toxic, exhibiting biocompatibility, biodegradability, thermoplasticity, and relatively high strength and elasticity, making them suitable replacements for conventional fossil fuel-based plastics in applications such as hygiene products and kitchenware (Mathew et al. 2005; You et al. 2015). On the other hand, the broader adoption of PLA is limited by its high cost, brittleness, low heat resistance, elongation at break, and restricted processability (Suryanegara et al. 2009; Cho and Cho 2016). A previous study reported that producing 1 kg of PLA can generate approximately 1.3 kg of CO_2 (Vink et al. 2010). As a potential solution, PLA-based bio-composites incorporating natural fillers have been proposed because they can lower the production costs, improve various properties, reduce CO_2 emissions, and mitigate the environmental burden through partial or complete biodegradation (Trivedi et al. 2023).

The use of wood powder as a filler in PLA-based composites offers low density, minimal equipment abrasion during secondary processing, high stiffness, sound-absorbing properties, easy availability, and relatively low cost compared to other natural fillers (Gerogiopoulos and Kontou 2015; Kariz et al. 2018; Jubinville et al. 2022; Taktak et al. 2023). Despite these advantages, blending hydrophilic wood powder with hydrophobic PLA limits interfacial adhesion between the two phases, resulting in reduced interfacial bonding, mechanical strength, and ductility (Balasuriya et al. 2001; Huang et al. 2009). Therefore, the use of interfacial binders is necessary to improve the interactions at the PLA/wood flour interface and enhance the composites properties. Maleic anhydride and silane have commonly been used as an interfacial binder in PLA/wood flour composites (Faludi et al. 2013; Du et al. 2017; Huo et al. 2026). However, the binder is an expensive synthetic compound with high reactivity that can be hazardous to human health, may cause environmental pollution during disposal, and require considerable caution in industrial processes (Lee and Wang 2006; Lazzeri et al. 2009; Faludi et al. 2013; Li et al. 2023; Pawlak et al. 2024).

Proteins contain various functional groups such as hydroxyl, thiol, carboxylate, amine, and phenol. The hydrolysis of proteins exposes a range of functional groups (polar, nonpolar, acidic, and basic) that could act as interfacial binders between hydrophobic PLA and hydrophilic wood powder. Therefore, in the present study, composites were fabricated using PLA filled with lignocellulosic powders, hydrogenated castor oil (HCO), which was previously identified as a dimensional stabilizer, and black soldier fly (*Hermetia illucens*, BSF) powder, which contains approximately 40 wt% protein, or protein-based adhesive formulated from BSF powder as interfacial binders (Wang and Shelomi 2017; Yang et al. 2025a). Composites were prepared by varying the type of filler and interfacial binder and by changing the PLA-to-filler ratio. The fabricated composites were tested for tensile strength, impact strength, bending strength, moisture absorption rate, and linear coefficient of thermal expansion. Based on the measured properties, this study evaluated the feasibility of using protein-based components as interfacial binders in PLA-based composites to determine the optimal fabrication conditions.

MATERIAL AND METHODS

Raw materials used for the fabrication of bio-composites

PLA, used as a main raw material for bio-composites fabrication in this study, was produced for injection molding by TotalEnergies Corbion (Gorinchem, The Netherlands). The moisture content, melting temperature and density of PLA, which was supplied in pellet form, were 0%, 150~230℃ and 1.2~1.3 g/cm^3, respectively. Radiata pine (*Pinus radiata*; RDP) and mixed hardwood scrubs (MIX) powder as a filler were supplied by the Hansol Home Deco Co., Ltd. (Jeonju, South Korea). Fibrous cypress bark (*Chamaecyparis obtusa*, CYP-B), a waste by-product from the sawing process, was air-dried for more than one week and cut to approximately 1 cm lengths with pruning scissors. The piece was then milled using a custom high-speed grinder (2.5 HP and 22000 rpm). CYP-B, along with RDP and MIX, was screened through sieves with size opening of 0.425 mm, 0.250 mm, 0.180 mm, and 0.150 mm. Powder (0.180 mm < RDP, MIX, CYP-B < 0.150 mm) obtained by screening was used as a filler to fabricate bio-composites.

Black soldier fly (*Ptecticus tenebrifer*, BSF) powder supplied by Green Deco Co., Ltd. (Gongju, South Korea) was used as an additive to promote interfacial bonding between hydrophobic PLA and hydrophilic wood or bark powder in the bio-composites, because it is composed mainly of protein with various functional groups that promote interfacial bonding between PLA and fillers. Proteinaceous adhesive resins (PAR) prepared from BSF were also used as an interfacial binder. Flake-shaped hydrogenated castor oil (HCO), produced by the hydrogenation of castor oil, was purchased from Hebei Guanlang Biotechnology Co., Ltd. (Shijiazhuang, China) and used as a dimensional stabilizer. Maleic anhydride (98% assay; MAA) produced by Samchun Sunyak Industrial Co., Ltd. (Pyeongtaek, South Korea) was used as a control interfacial binder for BSF and PAR. Fig. 1 shows the particle size distributions of the prepared RDP, MIX, CYP-B, BSF, and PAR.

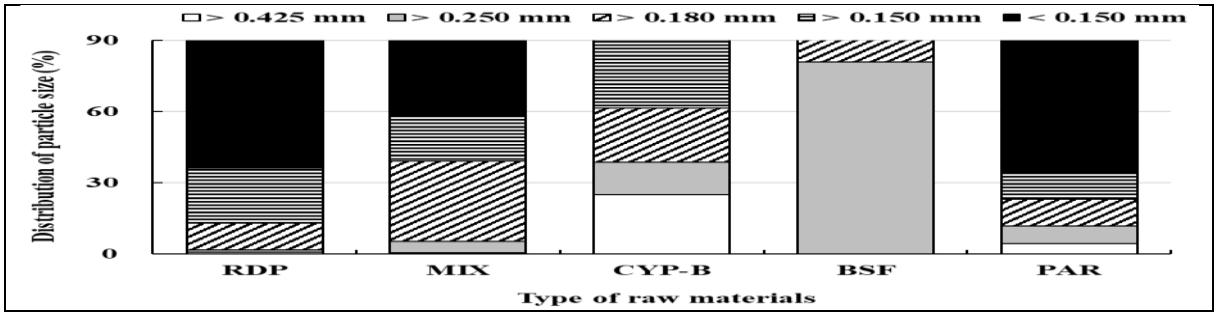

Fig. 1: Particle distribution of the raw-material powders (RDP: radiata pine; MIX: hardwood scrub; CYP-B: cypress bark; BSF: black soldier fly; PAR: proteinaceous adhesive resins) used for the fabrication of bio-composites.

Formulation of proteinaceous adhesive resin

BSF contains approximately 40 wt% protein and 15–20 wt% fat as major components, and also includes minerals such as Fe, Ca, P, and Zn (Spranghers et al. 2017). BSF powder passed through a 60-mesh sieve (< 0.25 mm) was used as a raw material for PAR preparation. The liquefaction of BSF by hydrolysis is necessary to formulate an interfacial binder. Based on

previous studies, 200 g of BSF was added slowly to 500 mL of an aqueous 5 wt% NaOH solution and reacted at 95±2.5°C for 240 min to obtain the BSF alkaline hydrolysate (BSF-AK) (Yang et al. 2014, 2018). The BSF-AK had a 38±1% solids content, a pH of 12.8, and a viscosity of 500 m·Pa·s at 25°C. The pH of BSF-AK exceeded 12. Therefore, an acidic hydrolysate of BSF (BSF-AC) was prepared as a hardener to shorten the curing time. BSF (150 g) was added slowly to 500 mL of an aqueous 5 wt% oxalic acid solution and reacted at 95±2.5°C for approximately 30 min. The BSF-AC had a solids content of 20±2% and a pH of 2.0. The hydrolysate transformed into a mud-like consistency at the end of the reaction; hence its viscosity could not be measured. Larch bark, crushed to a uniform size, was first extracted in hot water (95±2°C) for 4 h, and the extract was concentrated by evaporation to yield the hot-water extractive of larch bark (LB-HE). The LB-HE had a solids content of 38%, a viscosity greater than 1000 m·Pa·s, and a pH of 5.4. LB-HE was added to a BSF-AK/BSF-AC mixture to enhance the thermosetting property of the proteinaceous adhesive composed of alkaline and acidic hydrolysates.

The interfacial binder was formulated with 70 wt% BSF-AK, 10 wt% BSF-AC, and 20 wt% LB-HE based on previous studies (Yang et al. 2014,2018). A 5 wt% aqueous sodium borate (borax) solution was prepared. Fifteen drops of the solution were added to the protein-based interfacial binder as a preservative. The resulting liquid interfacial binder was freeze-dried (Unifreez FD-8, Daihan Scientific Co. Ltd., Wonju, South Korea) for one week to convert it to a solid form. The dried material was milled (Fritsch Pulverisette 19, Idar-Oberstein, Germany), and the powder that passed through a 60-mesh standard sieve but remained on a 100-mesh standard sieve was used as the interfacial binder (PAR).

Composite fabrication

RDP and MIX were dried in an oven at 105°C for 24 h to minimize the effects of moisture contained in the lignocellulosic fillers on the properties of the fabricated composites. Before specimen fabrication, PLA and HCO, along with the oven-dried RDP or MIX, were placed in weighing dishes according to the prescribed mixing ratios, and re-dried in a vacuum oven (DH WOV03030, Daihan Scientific Co. Ltd., Wonju, South Korea) at 70°C and 50 mmHg for 30 min. PAR, as an interfacial binder, was added to PLA, filler, and HCO immediately before specimen fabrication because it is susceptible to pre-curing from the heat generated during mixing and drying.

The effects of interfacial binder type and level on the strength and dimensional stability of composites were examined by fixing the filler (RDP and MIX), the contents of filler (RDP and MIX) and dimensional stabilizer (HCO) contents at 30 wt% and 5 wt%, respectively. The composites were fabricated with 62.5 wt% PLA/30 wt% filler/2.5 wt% PAR/5 wt% HCO and 60 wt% PLA/30 wt% filler/5 wt% PAR/5 wt% HCO. Composites composed of 70 wt% PLA and 30 wt% filler were also fabricated as a control to evaluate the effects of an interfacial binder.

The prepared mixtures, weighed at specified ratios, were fed into the hopper of an extruder/injection molder (Haake, MiniLab 2, Micropounder/Minijet Pro, ThermoFisher Scientific, Waltham, USA). The barrel temperature was set to 178°C because the PLA used in this study has a melting range of 173~178°C. The resulting high-viscosity mixtures were

injected into a mold maintained at 175℃ to fabricate dog-bone tensile specimens in accordance with ASTM D 638-24 (2022). Impact test specimens were prepared according to ASTM D 790-17 (2017) with dimensions of 80 mm (length)×10 mm (width)×4 mm (thickness).

Mechanical properties and dimensional stability

The mechanical properties of the composites were evaluated by tensile, flexural, and impact testing. Tensile and flexural tests were conducted using a universal testing machine (Instron 4467, Instron Corp., USA) in accordance with ASTM D638-24 (2022) and ASTM D790-23 (2023), respectively. Impact resistance was measured using an Izod pendulum impact tester (Model 892, Tinius Olsen, USA) following ASTM D256-24 (2024).

The dimensional stability was evaluated by drying the specimens in an oven at 80°C for 24 h, followed by sufficiently cooling in a desiccator. The weight of each specimen was measured, and the specimens were immersed in cold water (20°C) for 48 h, hot water (70°C) for 2 h, and in an oven at 80°C for 48 h. After immersion, the moisture that existed on the specimen surfaces was removed with paper towels, and the weight was re-measured. The water absorption rate (WAR) was calculated from these measurements to compare the dimensional stability under different manufacturing conditions.

Scanning electron microscopy (SEM) examination

The effects of interfacial binder type on the tensile behavior of composites were visually assessed by examining the fracture surfaces of composites fabricated with PLA, RDP, and each interfacial binder were examined by field-emission SEM (Supra 55VP; Carl Zeiss, Oberkochen, Germany) at an accelerating voltage of 3 kV. For microscopic observations, each specimen was mounted on an aluminum stub, and sputter-coated with a thin (~ 20 nm) layer of platinum.

Thermogravimetric analysis

Thermogravimetric analysis (TGA) was performed using a simultaneous DTA/TGA analyzer (SDT 650, TA Instruments, New Castle, USA) to evaluate the thermal stability of the raw materials and composites. Measurements were conducted under a nitrogen atmosphere from room temperature to 500°C at a heating rate of 10°C min^{-1}. Thermal behavior was assessed from the DTA and TGA curves.

Effects of filler type and its weight ratio on the properties of composites

In addition to RDP and MIX, the applicability of CYP-B was also investigated because it can be obtained at no cost from industrial waste in South Korea, and can reduce the high composites production costs caused by expensive PLA. Specimens for tensile and impact testing were prepared by mixing 60 wt% PLA 30 wt% filler (RDP, MIX or CYP-B), 5 wt% PAR as an interfacial binder, and 5 wt% HCO as a dimensional stabilizer. The composites were fabricated as described in Section 2.3. The extruder operation was halted repeatedly by excessive torque When the filler content reached ≥ 40 wt% of the total mixture. Therefore, mixtures containing 50 wt% or 40 wt% PLA with 40 wt% or 50 wt% filler and 5 wt% HCO were combined in a mixer at 5-min intervals for 4h. The heat generated during mixing provided adequate blending of the raw materials. PAR was added as described in Section 2.3.

The specimens were molded using a Teflon plate fitted with four rectangular molds measuring 200 mm × 20 mm × 5 mm (length × width × thickness) as shown in Fig. 2. The Teflon plate was placed on a steel plate sprayed with silicone oil, and a predetermined amount of mixed materials, which were adjusted to a density of 1 g/cm^3, was filled into each mold. A second steel plate was placed on top of the filled mold plate. Hot pressing was performed at 180°C under a 24 kg/cm^2 pressure for 12 min. After cooling, the specimens were demolded. Three specimens were prepared for each fabricating condition and conditioned in a chamber at 25°C and 50% RH for at least 24 h before testing the bending strength and dimensional stability.

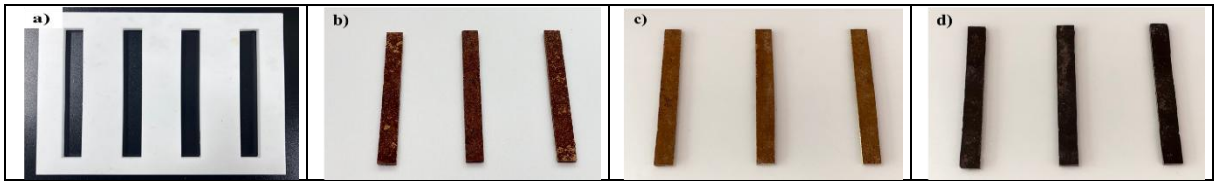

Fig. 2: Images of a) the Teflon plate used for composite fabrication and composites fabricated by blending PLA with b) radiata pine (RDP), c) hardwood scrub (MIX), and d) cypress bark (CYP-B).

The bending strength and dimensional stability [moisture absorption (MAR), length coefficient of thermal expansion (LCTE)] of the specimens were measured in accordance with ASTM D 790 (2017) and the Specifications and Quality Criteria for Wood Products designated by the National Institute of Forest Science of the Republic of Korea, respectively (NIFOS 2024). For the bending test, a load was applied at the center surface of each specimen at a crosshead speed of 10 mm/min. For the MAR, specimens previously tested for the bending strength were cut to 100 mm lengths. The initial weight (W1) of the specimens conditioned in the chamber was measured with a balance accurate to 0.01 g. The specimens were then immersed in hot water maintained at 95±2.5°C for 5 h, with a plastic mesh used to prevent contact with the aluminum-tank bottom or other specimens. After immersion, the specimens were placed in 23°C cold water for 20 min, then removed from the water tank. Moisture on the specimen surface was carefully wiped off with paper towels, and the final weight (W2) was recorded. The MAR was calculated using the following formula:

$$MAR\ (\%) = [(W2 - W1)/W1] \times 100 \quad (1)$$

For LCTE, the initial length (L0) of the specimens conditioned in the chamber was measured. The specimens were stored in a -20°C freezer (T1) for 72h, and the length (L1) was measured. The specimens were placed in an oven of 60°C (T2) for 6h, and the length (L2) was measured. LCTE was calculated as follows:

$$LCTE\ (1/°C) = (L2 - L1)/[L0 \times (T2-T1)] \quad (2)$$

Experimental design and statistical analysis

The experimental variables investigated in this study included the filler type (RDP, MIX, and CYP-B), type (BSF of PAR), particle size (BSF-1: 0.250 mm~0.425 mm; BSF-2

< 0.250 mm; PAR-1: 0.150 mm~0.250 mm; PAR-2: < 0.150 mm), loading level (2.5 wt% and 5 wt%) of interfacial binder, and weight ratio of PLA and filler used for composites fabrication (60 wt%/30 wt%, 50 wt%/40 wt%, and 40 wt%/50 wt%). The effects of these variables on the tensile strength, impact strength, bending strength, WAR, TVS, MAR, and LCTE were examined using the General Linear Model (GLM) procedure in the Minitab (Minitab LLC) programming package. A statistical test was conducted at the 95% confidence level. The effects found to be significant (p < 0.05) were further evaluated by pairwise comparisons using a Fisher's least significant difference (LSD) test at α = 0.05.

RESULTS AND DISCUSSION

Particle size distribution

Fig. 1 presents the particle-size distributions of the raw materials used in this study. RDP and MIX powders passing through an 80-mesh (0.180 mm) sieve were selected as fillers, and CYP-B in the same size range was used to minimize particle-size-related effects. BSF was separated into fractions retained between the 40- and 60-mesh sieves (BSF-1) and passing through the 60-mesh sieve (BSF-2), while PAR was separated into fractions retained between the 60- and 100-mesh sieves (PAR-1) and passing through the 100-mesh sieve (PAR-2). These fractions were used as interfacial binders to investigate the influence of particle size on strength and dimensional stability.

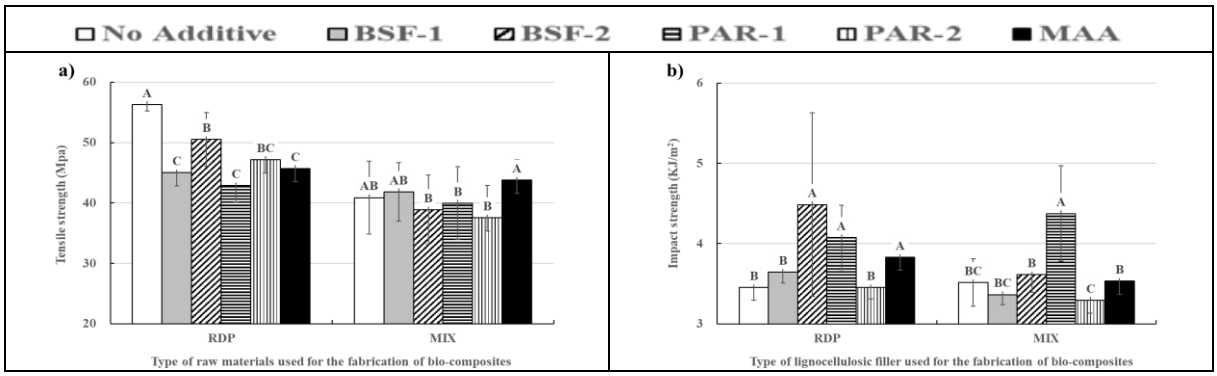

Fig. 3: Tensile (a) and impact (b) strengths of PLA/lignocellulosic composites as a function of interfacial binder type and particle size. Letters indicate significant differences (p < 0.05, LSD test).

Effect of interfacial binder type

Fig. 3 presents the tensile and impact strengths of composites prepared from 60 wt% PLA, 30 wt% RDP or MIX, 5 wt% HCO as a dimensional stabilizer, and 5 wt% interfacial binder based on a previous study (Yang et al. 2025b). The composites made from PLA and RDP without an interfacial binder (PLA/RDP) showed the highest tensile strength, which was attributed to the additional 5 wt% PLA in the composites compared with the interfacial binder-containing composites. Among the PLA/RDP composites, those fabricated with BSF-2 (<0.250 mm) (PLA/RDP/BSF-2) showed a higher tensile strength than those fabricated using BSF-1, PAR-1, or MAA, which was included as a control (PLA/RDP/BSF-1, PAR-1 or MAA).

FE-SEM observations of the fracture surfaces obtained after tensile testing revealed more fiber-breakage in PLA/RDP/BSF-2 than in PLA/RDP, PLA/RDP/PAR-2 or MAA (Fig. 4). The tensile strength was similar in the composites fabricated with PLA/MIX and those with interfacial binders, indicating that the interfacial binders primarily affect interfacial bonding in PLA/RDP.

For the impact strength, PLA/RDP fabricated with BSF-2, PAR-1, or MAA showed higher values than those fabricated with BSF-1 or PAR-2, while PLA/MIX showed the highest impact strength when fabricated with PAR-1 (Fig. 3). These results suggest that PAR-1 is the most effective interfacial binder for improving the impact strength in composites fabricated with either RDP or MIX. All composites produced in this study satisfied the minimum impact strength (≥ 3 kJ/m²) of the standard of wood–plastic composite specified by the National Institutes of Forest Science of the Republic of Korea.

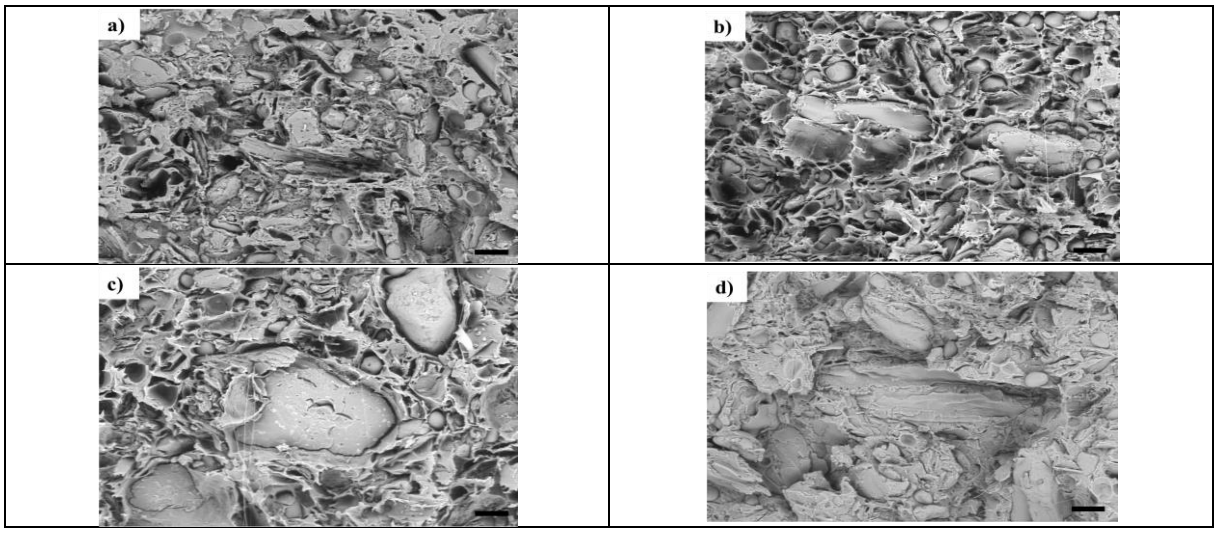

Fig. 4: SEM images of the fracture surfaces from tensile-tested composites: a) 70 wt% polylactic acid (PLA)+30 wt% radiata pine powder (RDP), b) 60 wt% PLA+30 wt% RDP+5 wt% hydrogenated castor oil (HCO)+5 wt% black soldier fly powder (BSF-2 < 0.250 mm), c) 60 wt% PLA+30 wt% RDP+5 wt% HCO+5 wt% proteinaceous adhesive resin powder (PAR-2 < 0.150 mm), and d) 60 wt% PLA+30 wt% RDP+5 wt% HCO+5 wt% maleic anhydride (Scale bar = 20 μm).

Overall, PLA-based composites fabricated with RDP showed higher tensile strength than those with MIX, and the type and particle size of the interfacial binder had little effect on the tensile strength. By contrast, the impact strength increased in PLA/RDP fabricated with BSF-2 and PAR-1, and in PLA/MIX fabricated with PAR-1, suggesting that PAR-1 is the most effective interfacial binder for improving the impact strength.

Effect of interfacial binder level

Fig. 5 shows the effects of interfacial binder level on the tensile strength of composites fabricated with PLA and either RDP or MIX. Increasing the interfacial binder from 2.5 wt% to 5 wt% caused a decrease in tensile strength for PLA/RDP containing BSF-1 and MAA, but no significant change was observed for PLA/RDP containing BSF-2, PAR-1, or PAR-2. In PLA/MIX, the tensile strength decreased as the level of BSF-2 and PAR-1 increased, while

the other interfacial binders had no effect. The decreases in tensile strength are likely due to the concomitant decrease in PLA level from 67.5 wt% to 65 wt% as the binder level was increased. Therefore, BSF-2, PAR-1, and PAR-2 appear to be suitable interfacial binders for maintaining tensile strength in PLA/RDP, whereas BSF-1, PAR-2, and MAA are more appropriate for PLA/MIX.

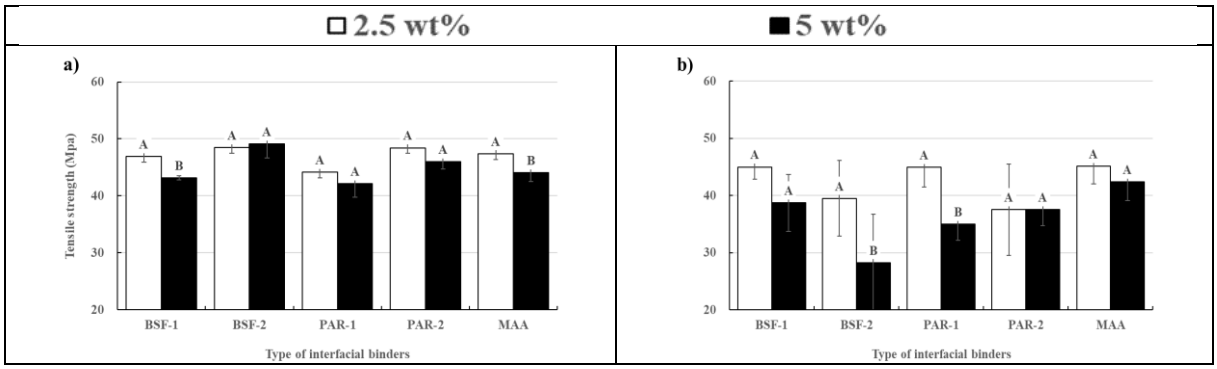

Fig. 5: Effect of binder type, particle size, and loading level (2.5 vs. 5.0 wt%) on the tensile strength of (a) RDP-filled and (b) MIX-filled PLA composites. Letters indicate significant differences ($p < 0.05$, LSD test)

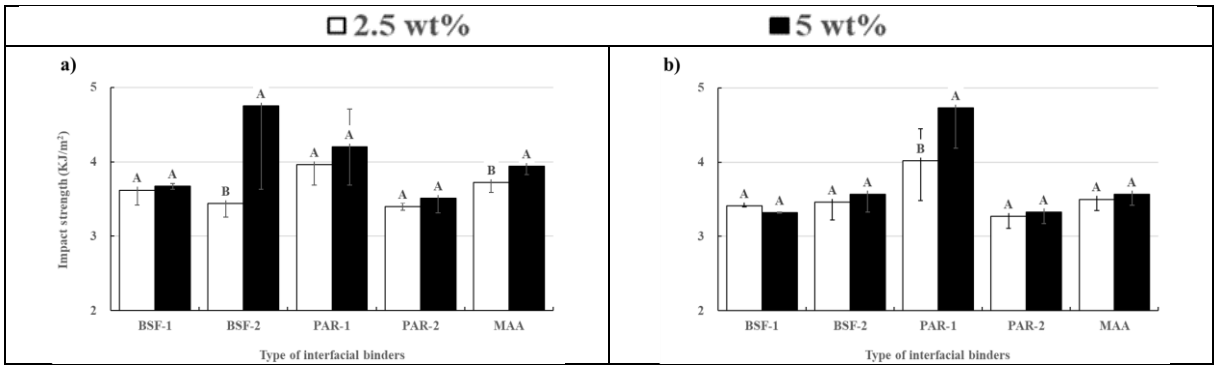

Fig. 6: Effect of binder type, particle size, and loading level (2.5 vs. 5.0 wt%) on the impact strength of (a) RDP-filled and (b) MIX-filled PLA composites. Letters indicate significant differences ($p < 0.05$, LSD test)

In contrast to the tensile results, the impact strength increased with higher interfacial binder levels for BSF-2 and MAA in PLA/RDP and for PAR-1 in PLA/MIX (Fig. 6). Increasing the level of the other binders did not affect the impact strength. Therefore, increasing the interfacial binder level enhances interfacial bonding between PLA and the fillers (RDP or MIX). Accordingly, a 5 wt% binder level is recommended as the optimal condition for the fabrication of PLA-based composites using lignocellulosic fillers.

Moisture absorption rate

Fig. 7 presents the moisture-absorption rate (MAR) measured after immersing the composites in cold water (20°C) for 48 h, in hot water (70°C) for 2 h, and after storage in an oven at 80°C for 48 h. Regardless of the immersion and storage conditions, negligible differences in MAR were observed between RDP and MIX. Under cold-water immersion, all

composites exhibited MAR values below 2%, with the lowest value observed for composites fabricated without an interfacial binder. This result was attributed to the higher PLA level (65 wt%) in those composites. Composites fabricated with PAR-1 and PAR-2 as binders showed a higher MAR than those with MAA as binders for PLA/RDP and PLA/MIX. These results were attributed to the reduced molecular weight of PAR, which was formulated via the alkaline and acid hydrolysis of BSF, increasing the surface area available for water contact and promoting higher MAR.

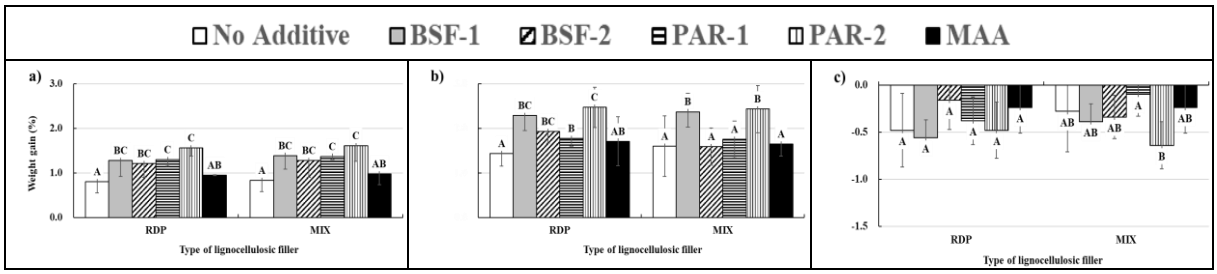

Fig. 7: Moisture absorption (weight gain) of WPC samples under different exposure conditions: (a) 20 °C water / 48 h, (b) 70 °C water / 2 h, and (c) 80 °C oven / 48 h. Letters indicate significant differences ($p < 0.05$, LSD test).

Under hot-water immersion, composites fabricated without an interfacial binder or with MAA exhibited lower MAR regardless of filler type (Fig. 7). In addition, PLA/RDP and PLA/MIX fabricated with PAR-1 showed a lower MAR than those fabricated with PAR-2, indicating that the particle size of PAR influences moisture uptake in hot water. For the oven-stored specimens, the weight of PLA/RDP was unaffected by the presence, type, and particle size of interfacial binder. In contrast, the weight of PLA/MIX fabricated with PAR-1 was lower than that with PAR-2 because of less moisture absorption of PAR-1. These results suggest that the finer PAR-2 particles, which have a larger specific surface area, absorb more moisture during composite processing and exhibit a higher MAR. On the other hand, PAR-1 showed a relatively lower MAR, making it a more suitable interfacial binder for PLA/RDP and PLA/MIX composites.

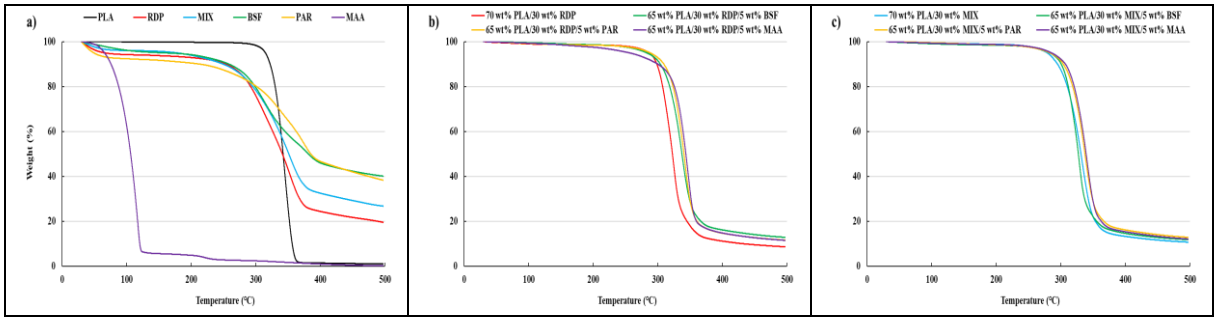

Fig. 8: TGA curves showing weight loss (%) versus temperature for a) raw materials (PLA, RDP, MIX, BSF, PAR and MAA), b) PLA+RDP-based composites, and c) PLA+MIX-based composites fabricated with BSF, PAR, and MAA as interfacial binders.

Thermal properties

Fig. 8 shows the thermogravimetric analysis (TGA) results of the raw materials and

composites. PLA began to degrade at approximately 310°C, whereas RDP, MIX, and CYP-B exhibited increased weight loss above 250°C, primarily due to lignin decomposition. In contrast, MAA showed weight loss below 100°C, indicating lower thermal stability.

The incorporation of RDP or MIX reduced the onset temperature of thermal degradation of PLA. However, the addition of interfacial binders partially mitigated this effect by increasing the degradation onset temperature, likely through improved interfacial interactions that restricted heat transfer (Meng et al. 2011). Because the binder content was limited to 5 wt%, the improvement was modest. Nevertheless, BSF and PAR exhibited higher thermal stability than RDP and MIX, suggesting their potential to enhance the thermal stability of the resulting composites.

Applicability of cypress bark as a filler

Fig. 9 shows the tensile and impact strengths of composites fabricated by mixing 60 wt% PLA, 30 wt% RDP, MIX or cypress bark powder (CYP-B) as a filler, 5 wt% PAR-1 as an interfacial-binder, and 5 wt% HCO as a dimensional stabilizer. The tensile strengths of PLA/RDP and PLA/CYP-B did not differ significantly ($p = 0.93$), but both were significantly higher than that of PLA/MIX/PAR-1/HCO ($p < 0.01$). PLA/CYP-B exhibited higher impact strength than PLA/RDP or PLA/MIX ($p < 0.01$), with no significant difference between the latter two ($p = 0.75$). These results support the potential of CYP-B as an alternative filler to wood powder.

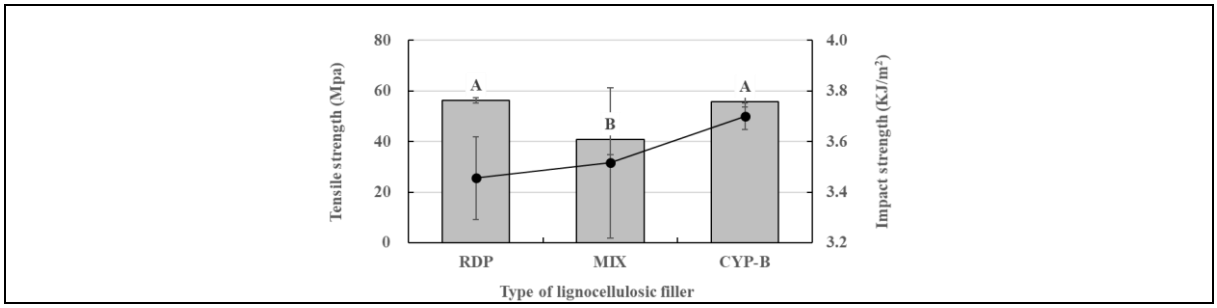

Fig. 9: Tensile and impact strength of composites fabricated with 60 wt% polylactic acid, 30 wt% filler [radiata pine (RDP), hardwood scrub (MIX) or cypress bark (CYP-B)], 5 wt% hydrogenated castor oil as a dimensional stabilizer, and 5 wt% proteinaceous adhesive resin powder as an interfacial binder. The different capital letters above the columns show significant differences in tensile strength at p = 0.05 (least significant difference test).

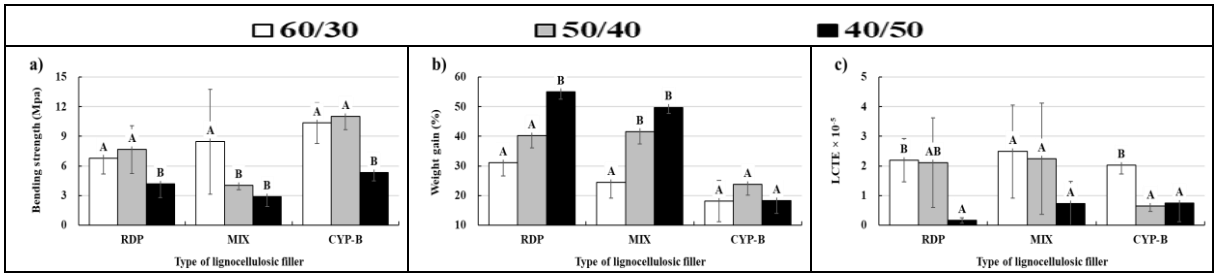

Fig. 10: Flexural strength (a), moisture absorption (b), and LCTE (c) of composites at various PLA-to-filler weight ratios. Letters indicate significant differences (p < 0.05, LSD test).

Based on these results, the levels of PAR and HCO were adjusted to 5 wt% each. The PLA level (60 wt%, 50 wt%, and 40 wt%), and the filler level (30 wt%, 40 wt%, and 50 wt%) were varied to fabricate composites and evaluate their properties (Fig. 10). The bending strengths of PLA/RDP and PLA/CYP-B did not differ between 60 wt%/30 wt% and 50 wt%/40 wt%, but decreased at 40 wt%/50 wt%. In PLA/MIX, the bending strength was higher at 60 wt%/30 wt% than 50 wt%/40 wt% and 40 wt%/50 wt%. These differences were attributed to the filler morphology. For example, MIX contains a larger proportion of square-shaped particles, whereas CYP-B and RDP contain more fibrous and rectangular particles with higher aspect ratios, which likely contribute to the higher bending strength (Yang et al. 2025b).

The MAR of PLA/RDP and PLA/MIX increased as the proportion of RDP or MIX in the mixture increased (Fig. 9). By contrast, PLA/CYP-B exhibited a lower MAR than PLA/RDP and PLA/MIX, which was attributed to the superior water resistance of bark powder compared to wood powder (Kain et al. 2018; Zhao et al. 2024). The ratio of PLA and CYP-B did not affect the MAR of PLA/CYP-B. PLA/RDP and PLA/CYP-B fabricated with 50 wt%/40 wt% and 40 wt%/50 wt% showed lower LCTE values than those fabricated with 60 wt%/30 wt%. an analysis of the results for different PLA and filler ratios showed that provided the best properties was 50 wt% PLA, 40 wt% CYP-B, 5 wt% PAR-1, and 5 wt% HCO.

CONCLUSIONS

(1) The tensile and impact strengths were higher in the composites containing RDP than those containing MIX. The application of BSF-2 and PAR-1 as an interfacial-binder had little effect on tensile strength but substantially increased the impact strength. FE-SEM observations of the fracture surfaces of the tensile specimens corroborated these findings. *(2)* When the level of interfacial binder was increased from 2.5 wt% to 5 wt%, the impact strength of the composites increased. In particular, the effect was pronounced for BSF-2 in PLA/RDP and for PAR-1 in PLA/MIX. *(3)* No significant differences in MAR were observed between PLA/RDP and PLA/MIX after cold- and hot-water immersion and oven storage were found. By contrast, MAR increased when a finer PAR (smaller particle size) was applied as an interfacial binder. *(4)* The tensile strengths of PLA/RDP/PAR-1/HCO and PLA/CYP-B/PAR-1/HCO did not differ, but both exceeded that of PLA/MIX/PAR-1/HCO. PLA/CYP-B/PAR-1/HCO showed higher impact strength than PLA/RDP/PAR-1/HCO and PLA/MIX/PAR-1/HCO, which did not differ significantly from each other. *(5)* Bending strength, MAR, and LCTE of PLA/CYP-B/PAR-1/HCO exceeded those of PLA-1/RDP/PAR-1/HCO and PLA-1/MIX/PAR-1/HCO. The composites containing 50 wt% PLA and 40 wt% CYP-B showed the best overall performance.

Overall, the optimal condition was identified as 50 wt% PLA, 40 wt% CYP-B (filler), 5 wt% PAR-1 (interfacial binder), and 5 wt% HCO (dimensional stabilizer). Composites fabricated under the above conditions exhibited superior bending strength, lower MAR, and improved LCTE compared with those fabricated with MAA, the conventional synthetic interfacial binder for wood-plastic composites. Thus, the non-toxic protein-based PAR-1 might replace the costly, toxic, and environmentally-hazardous MAA.

ACKNOWLEDGMENTS

This research was funded by the Korea Forest Service (Korea Forestry Promotion Institute) through R & D program for Forest Science Technology (Project No. RS-2023-KF002481).

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