Evaluation of recycled polypropylene properties with banknote-reinforced

Ingrid Amélia dos Santos Matusinho[1] [iD], Ítalo Rocha Coura[1] [iD], Patterson Patricio de Souza[1] [iD] and Patrícia Santiago de Oliveira Patrício[1*] [iD]

[1]Departamento de Química, Centro Federal de Educação Tecnológica de Minas Gerais - CEFET-MG, Belo Horizonte, MG, Brasil

*patricia@cefetmg.br

Abstract

Banknotes often have a short lifespan and are frequently removed from circulation due to wear and tear, leading to a significant disposal challenge given the millions of banknotes discarded annually. This study evaluated the utilization of these waste banknotes, primarily composed of cotton fibers, in producing polymer composites. The mechanical and thermal properties, hydrophilicity, and degradation through accelerated aging of composites made from recycled polypropylene (PPR) and banknote residues were evaluated. The results indicated that including banknote waste increased the hydrophilicity of the composites relative to pure PPR and improved mechanical properties against weathering. Notably, composites with 10 wt% banknote content enhanced Young's modulus and tensile strength, both in weathered and non-weathered conditions. This approach not only offers a potential solution for the disposal of waste banknotes but also aligns with the broader goal of reducing ecological impact and promoting the development of sustainable composite materials.

Keywords: composite, polypropylene, banknote.

1. Introduction

The banknotes have a short useful life, with a period of use until they become unsuitable for circulation due to their dire conditions. In addition to issuing and monitoring, the issuing institutions are responsible for the activity of sanitation of the circulating medium, aiming to keep the money in circulation with good conditions of use, thus assuming also responsibility for solid waste management. This activity, in general, is regulated by law, providing principles and guidelines related to solid waste management[1].

In Brazil, the banking network collects millions of banknotes yearly, and those that are not in good circulation conditions are destroyed. Incineration is used to destroy banknotes, but this technique has become increasingly inadequate as it allows for the release of toxic waste into the environment[2]. In this context, due to the growing concern with solid waste management and others, the enormous environmental impact they generate, studies, and improvements of processes that are efficient in recovering or reusing waste[1].

Some studies have been carried out to explore different destinations for waste banknotes, such as their use as a raw material in the production of biofuels and recycling for papermaking, seeking solutions for reusing this waste[2,3]. However, it is essential to note that these alternatives usually involve multiple processing steps, often including the use of chemical reagents. Since banknotes are mainly composed of cotton and have a high cellulose content, a promising option for its destination is to use it as a filler or reinforcement in polymer composites[4]. This utilization offers a sustainable approach to managing waste and provides an opportunity to enhance the properties of the polymer[5-7]. Fillers can be added to maintain or improve the properties of functional polymers when it comes to polymer recycling[6,7]. Recycling has been used as an alternative to reduce the amount of plastic waste discarded in the environment. However, some properties of polymers are compromised when subjected to processing conditions, high temperatures, and shear mechanical recycling[8].

Given this perspective, to reduce the accumulation of waste banknotes and polypropylene in the environment, the present work aims to develop composites using recycled polypropylene as a polymer matrix and waste banknotes as a filler. The composites were characterized using Infrared Spectroscopy (FTIR), Thermogravimetric Analysis (TG), Tensile Test, Differential Scanning Calorimetry (DSC), Wetting Test, and Accelerated Weathering Test.

2. Experimental

2.1 Materials and methods

The PP-banknote composites were prepared with recycled polypropylene (PPR) supplied in granular form

by RCA *Plásticos* Company. The banknotes (BK) were provided by the Central Bank of Brazil, Department of the Circulation, with particle sizes ranging from 2.0-3.0 mm.

The PPR and the residues of BK were placed in the mixing chamber of a Homogenizer (Drais), model MH-100 of the brand MH Equipment. The banknote contents were 10, 20, and 30 wt% of the mixture, and the identification is shown in Table 1. After the polymer melting, homogenization was maintained for approximately 5 seconds, then the equipment was turned off, and the mixture obtained was removed. The same procedure was performed with the pure matrix (PPR). The materials obtained by homogenization were ground in a knife mill, Marconi, model MA 580.

2.2 Materials characterization

Infrared spectroscopy was performed using the Shimadzu IRPrestige-21 Spectrophotometer with an ATR accessory. Thirty scans were performed at room temperature in the region between 4000 and 400 cm^{-1}.

The materials' thermogravimetric analyses (TGA) were carried out using DTG-60H thermal analyzer equipment, Shimadzu. The protocol used was heating from 25 °C to 800 °C, with a heating rate of 10 °C.min^{-1} and a synthetic air atmosphere. The materials were submitted to differential scanning calorimetry (DSC) using PerkinElmer DSC-6000 equipment. The samples were heated from 25 °C to 250 °C and kept at a temperature of 250 °C for 3 min, then cooled to 25 °C and heated again to 250 °C, with a heating rate of 10 °C min^{-1} and nitrogen flow of 20.00 mL.min^{-1}.

The measurements of the sample contact angle (θ) were carried out in triplicate on different regions of the samples. It used a wettability test apparatus at an ambient temperature of 24 °C. The samples were placed on the support, and then deionized water of standardized volume (10.00 μL) was dripped onto the sample's surface. The images were obtained by filming the test using the AMCap program, and the contact angle (θ) was calculated using the ImageJ program. The contact angle (θ) was given by measuring three drops deposited on each film at least twice. Later, the arithmetic mean of the measurements and the standard deviation were calculated.

The exposure test to accelerated weathering was performed in the Accelerated Weather Simulator, brand BASS, model UVV-STD-4400. Seven samples were exposed to weathering in 12-hour cycles divided into 6 hours of exposure to UV-A light at a maximum temperature of 60 ºC, without moisture, followed by 6 hours of lamps off, at a minimum temperature of 30 ºC, with forced humidity. A total of 124 cycles, equivalent to 1488 hours of material exposure to artificial weathering, was performed.

The tensile tests were carried out in the Instron universal testing machine, model EMIC 23-20, operated at room temperature (~ 23°C), using material bodies exposed to accelerated weathering and pure matrix specimens, not weathering. The materials were submitted to the test using clamps for tensile testing at a test speed of 50 mm.min^{-1}. The results were obtained from an average of 7 specimens tested for each composition of exposed materials and five pure matrixes not exposed to weathering. The tensile tests of the PPR-BK composites and PPR were carried out to evaluate the influence of fillers on the mechanical properties of materials before and after exposure to weathering.

3 Results and Discussion

In the FTIR spectrum of the recycled polypropylene and the PPR-BK composites (Figure 1), it was possible to identify the typical bands of the polymer groups at 2950, 2866, and 2840 cm^{-1} refer to the asymmetric stretching of methylene (ν_{assim}C-H_3), the symmetrical stretches (ν_{sim}CH_3) and (ν_{sim}CH_2), respectively. It can be observed two bands are associated with the deformations of the methyl group (-CH_3) in the region of 1376 cm^1 (δ_{sim}CH_3) and 1453 cm^{-1} (δ_{assim}CH_3). Low-intensity bands at 842 and 725 cm^{-1} correspond to the angular deformations of the C-H bonds (δC-H) and 1170 cm^{-1} carbon-carbon stretching (νC-C) of the polymer chain[9,10]. The 500-400 cm^{-1} bands can be attributed to stretching the iron-oxygen bond (νFe-O), corresponding to the pigments in recycled polypropylene. Black inorganic pigments applied to plastics are made of iron oxides (Fe_3O_4, (FeCO)Fe_2O_4)[11]. Furthermore, the contamination of recycled polypropylene by low-density polyethylene (LDPE) was identified in the FTIR spectrum of the PPR due to the appearance of the characteristic LDPE bands. The main bands that characterize polyethylene appear in the range of 2918 and 2840 cm^{-1} and 1463 cm^{-1}, corresponding to the C-H stretching vibrations and the vibration deformation band of the CH_2 group, respectively. Bands associated with polyethylene amorphous/crystal form are also noted at 720-730 cm^{-1}[12,13]. The analyzed material (commercial recycled PP) may contain other polymers in addition to recycled polypropylene, such as low-density polyethylene

Table 1. Identification of the composites and correlation with the banknote.

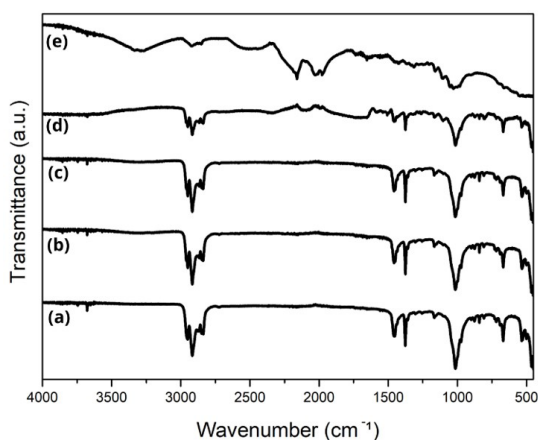| Identification	banknote (wt%)
PPR-10BK	10
PPR-20BK	20
PPRC-30BK	30

Figure 1. FTIR spectra of (a) PPR, (b) PPR-10BK, (c) PPR-20BK, (d) PPRC-30BK, and (e) banknotes.

and polystyrene, due to potential contamination during the recycling process.

The banknotes spectrum (Figure 1) shows typical cellulose bands associated with the cellulosic O-H stretching and asymmetric stretching C-H in the range of 3500 and 3200 cm^{-1} and at wavenumber at 2900 cm^{-1}, respect. The band at 1312 cm^{-1} is associated with the C-O-H (δ C-O-H) bending vibrations, and at 1052 and 1000 cm^{-1} corresponds to the stretching of the C-O-C bonds[14,15]. It was also possible to detect the presence of bands attributed to the Metal-Oxygen bonds, corresponding to the pigments in the banknote, pearl effect pigments. Among the numerous pigments used, these have a metallic appearance resulting from different thicknesses and combinations of titanium and iron oxides on mica, providing an iridescence effect[14]. The bands in the region between 600 and 420 cm^{-1} correspond mainly to the stretching of the Metal-Oxygen bonds (νTi-O, ν Fe–O) of the titanium (TiO_2) and iron (Fe_2O_3) oxides present in the pigment.

Figure 2 shows the contact angle measured between the drops of water on the surface materials at the top and the average values of the contact angles. It was observed that all samples presented a mean contact angle characteristic of hydrophilic materials. In the case of recycled polypropylene, the hydrophilicity is attributed to the presence of pores on the surface of the polymeric films and metallic oxides, as indicated by the FTIR spectra. For composites, the hydrophilicity is higher than pure PPR, which is justified by adding banknotes composed of cotton fibers. The hydroxyl groups (–OH) present in the chemical structure of the fibers promote hydrogen bonds between the cellulose of the cotton fiber and the water molecules[15]. The contact angle value of the composite with 30%wt cellulose is indeed close to that of pure PPR. This can be explained by the possible uniform dispersion of cellulose fibers within the PPR matrix, which reduces the exposure of hydrophilic cellulose on the composite's surface

TGA and DTG curves obtained for the banknotes and composites are shown in Figure 3. The banknote TGA exhibits a mass loss stage around 25-120 ºC relative to the adsorbed moisture. The thermal degradation of the banknote occurs in a temperature range between 240 and 442 ºC, presenting two stages of decomposition. According to the banknote curve, peaks of maximum loss at temperatures of 323 and 395 ºC were identified in the DTG curve. Other authors have found similar results, attributing the stages of grade degradation to the degradation of cellulose-based fibers[4]. The TGA curve of the PPR10BK composite is like the pure polymer. It can be seen through the DTG of PPR that its thermal degradation occurs in two consecutive stages of decomposition. The first stage proceeds slowly and can be evidenced by forming a broad peak in the curve, followed by another immediate step that presents a maximum peak of around 427 °C. The composites PPR-20BK and PPR-30BK present two stages of decomposition that can be better visualized in the DTG curve (Figure 3b). The incorporation of residues decreased the temperature at which the first decomposition stage occurs. In addition, it is observed that the maximum mass loss peak temperature of the composites decreases as the banknotes content increases. The residual mass visualized in the TGA curves is related to PPR contaminants and pigments in the polymer and the banknotes.

In the DSC curves, Figure 4 shows the crystallization and melting temperatures for PPR, PPR-10BK, PPR-20BK, and PPR-30BK. The PPR curve shows two peaks associated with melting (peak 1 and peak 2). and two crystallization peaks (peak 3 and peak 4). The data obtained from the DSC curves (Figure S1) of the PPR and PPRbanknote materials are shown in Table 2. The peak at the highest temperature, close to 160 ºC, refers to the melting of polypropylene. The peak at temperatures ~120 ºC was associated with the melting of LDPE, present in the commercial sample of PPR, as suggested by its FTIR spectrum. In the work of McCaffrey et al.[13], similar behaviors were observed in the polypropylene-polyethylene blend. The composites PPR-10BK, PPR-20BK, and PPR-30BK showed similar T_m, around 150 ºC. The presence of the banknote reduced the melting temperature of the PPR.
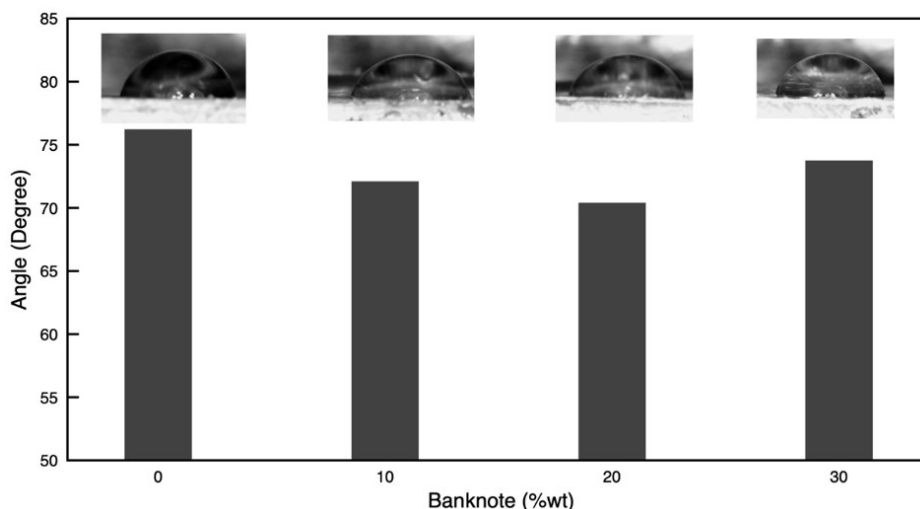
Figure 2. Bar graphic of the Contact angle PPR and Composite PPR/Banknote.

Table 2. Differential Scanning Calorimetry curves data: PPR, PPR-C10%, PPR-C20%, and PPR-C30%.

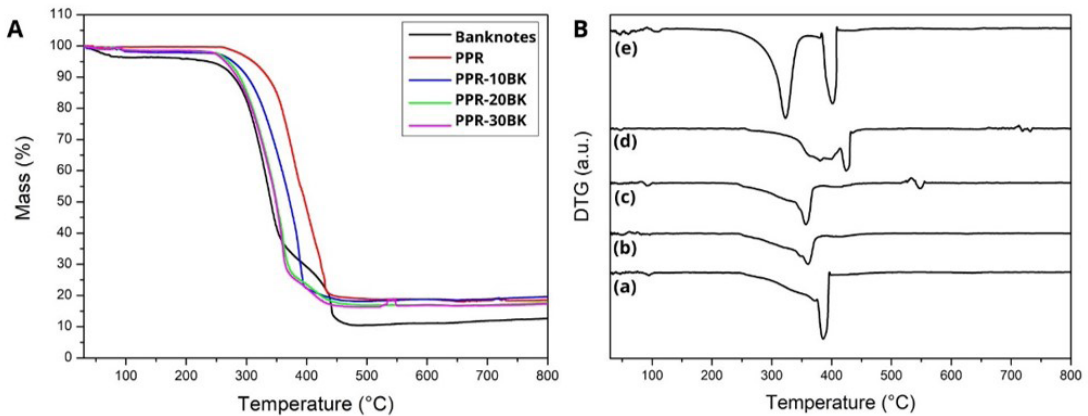| Material	DSC					
	Heating				Cooling	
	T_m (°C)	ΔH (J. g^{-1})	T_m PP (°C)	ΔH (J. g^{-1})	T_c PP (°C)	T_c (°C)
PPR	118.00	4.32	162.00	58.88	122.00	113.00
PPR-10BK	121.00	2.17	154.00	19.59	118.00	-
PPR-20BK	121.00	1.66	153.00	15.05	117.00	-
PPR-30BK	121.00	1.68	152.00	13.10	116.00	-

Figure 3. TGA (A) and DTG (B) curves of the (a) PPR, (b) PPR-10BK, (c) PPR-20BK, (d) PPR30BK, and (e) Banknotes.

In the cooling curve, the first crystallization peak was identified at 122 °C, which refers to polypropylene crystallization. A shift in the crystallization peak is observed with the addition of banknote with a decrease in T_c of 6 °C for PPR-30BK. The second exothermic peak occurs at a temperature of 113 °C and is probably related to the crystallization of LDPE. From the analysis of the enthalpy variation during the LDPE melting, it was possible to determine the amount of this polymer in the mixture. The variation of the melting enthalpy obtained by the peak area was 4.31 J.g^{-1}. Considering that for pure LDPE, the enthalpy change during melting ($\Delta H_m °$) is 140 J.g^{-1}, the ratio of values (ΔH_{peak}/LDPE) was 0.0307, corresponding to approximately 3.1%wt of mixed polyethylene to the PPR.

The results of the tensile tests of the materials PPR-BK composites, pure PPR exposed to accelerated weathering (PPR), and recycled polypropylene not exposed to accelerated weathering (PPRS) are presented in Figure 5. It is observed from the average maximum tensile strength values that exposure to weathering provided the pure polymer (PPR) with a decrease of approximately 53% of the tensile strength compared to recycled polypropylene not exposed to weathering (PPRS). In the work of Tian et al.[16], which investigated the accelerated weathering properties of polypropylene-wood composites, the tensile strength of virgin PP showed the most significant decrease in tensile strength values for all composites were higher than those of virgin PP. The degradative processes of the polymer exposed to accelerated weathering provide the scission of the chains
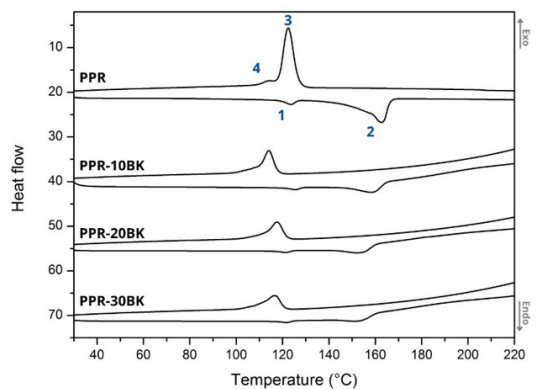

Figure 4. DSC curves of the PPR and Composites PPR/Banknote.

and, consequently, the drop in the material's mechanical properties, such as the reduction of its tensile strength.

Furthermore, an addition of the banknote in the polymer raises *Young's* modulus of the exposure weathering PPR, reaching a 55% increase for the PPR-10BK, following the 27% and 9% for PPR-20BK and PPR-30BK respect. This result indicates that the degradation of the PPR is superficial, probably caused by the breakage of a few polymeric chains, disrupting the entanglements and allowing an additional crystallization, increasing the stiffness and resistance of the polymer. The loss of mechanical properties of polymers usually results in more
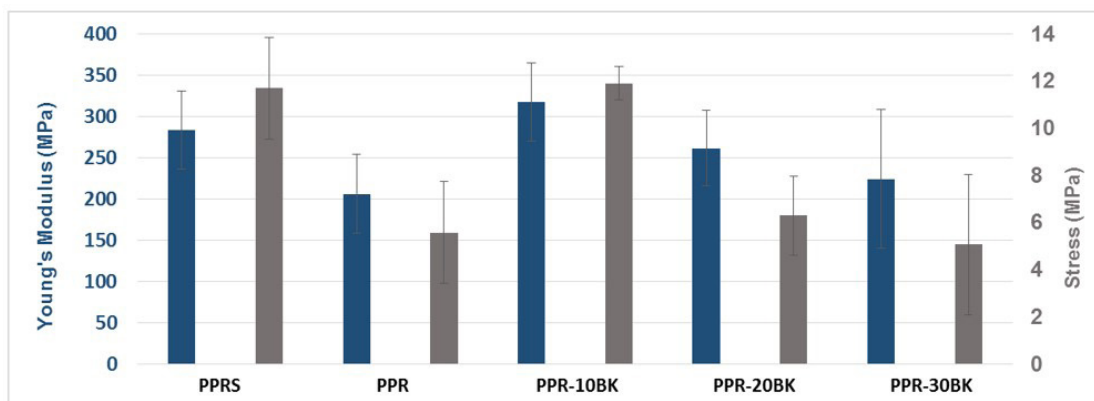
Figure 5. Bar Graph of the Young's Modulus (blue) and Maximum Tensile (grey) for PPR and Composites PPR/Banknote.

intensive degradations in the polymer matrix. Incorporating banknotes proved efficient in delaying the degradation of PPR when subjected to weathering. One of the hypotheses to explain this behavior is that the cellulose in the banknotes influences the decomposition reactions of polypropylene, deactivating free radicals. The higher filler content can compromise its dispersion in the matrix and, therefore, have a lower influence on the mechanical properties. Consequently, the dispersed phase starts as a defect, leading to early matrix rupture and compromising mechanical properties.

The difference between the mechanical properties found after the analysis of the materials was statistically evaluated by the F (variance) and T (mean) tests with a confidence level of 95%. The results obtained for the tensile strength and Young's modulus values are presented in the work's Table S1 (Supplementary Material). It can be seen in Figure 5 that the composites with 20% and 30 wt% of banknotes showed slightly higher values of tensile strength when compared to the PPR. However, according to the average (T-test) analysis, the PPR-20BK and PPR-30BK composites appear statistically equivalent to the PPR. Therefore, it is considered that these composites, in terms of tensile strength, are similar.

Finally, the mechanical properties of the PPR-10BK composite were compared with those of the pure polymer before weathering. Statistical analysis revealed that they are statistically equivalent with 95% confidence. This result indicates that the composite, even after exposure to weathering, was similar to the unexposed pure polymer (PPR) in terms of its tensile strength. Regarding statistical analysis, the T-test confirmed that the means of Young's modulus values are statistically similar between PPR and PPR10BK and distinct from PPR and PPR-10BK with 95% confidence. This outcome suggests that the composite, regarding Young's modulus, remained similar to the unexposed polymer. All variances were equivalent, indicating that the variation in the results of the analyses performed was identical between the materials.

4 Conclusion

In this work, the batch homogenization technique effectively incorporated waste banknotes into recycled polypropylene for composite production. This method facilitated the development of materials without needing prior treatment of the banknote waste or additional reagents. Using polymeric composites presents an effective strategy to reduce the accumulation of discarded banknotes and polymers, addressing sustainability requirements and reducing environmental waste.

The FTIR analysis revealed the presence of distinct functional groups in the PPR and the waste banknotes. In the spectra of the BK, bands associated with cellulose at 1312 cm^1 (δ_{C-O-H}) and at 1052 and 1000 cm^{-1} (ν_{C-O-C} bonds), as well as to additives such as metallic pigments, at 600 and 420 cm^{-1}, correspond mainly to the stretching of the Metal-Oxygen bonds. In the spectrum of recycled polypropylene, typical polypropylene bands were observed, along with signs of contamination by low-density polyethylene (LDPE). These observations were confirmed by an exothermic transition related to the crystallization of LDPE in the DSC curves. Moreover, the results from the DSC analysis demonstrated that the inclusion of waste banknotes led to a reduction in the melting temperature as the banknote content increased. Thermogravimetric analysis revealed a decrease in the thermal stability of the composites with an increased concentration of banknote waste compared to pure recycled polypropylene. Nevertheless, incorporating these wastes conferred stability against degradation from weathering, resulting in materials with enhanced mechanical resistance, particularly in terms of Young's modulus and tensile strength. The addition of 10 wt% of banknote waste significantly increased Young's modulus of the composite compared to the pure matrix, suggesting the potential for application of these materials in outdoor environments and the manufacturing of durable items.

5 Author's Contribution

- **Conceptualization** – Patrícia Santiago de Oliveira Patrício; Patterson Patricio de Souza

- **Data curation** – Ingrid Amélia dos Santos Matusinho; Ítalo Rocha Coura.

- **Formal analysis** – Ingrid Amélia dos Santos Matusinho; Ítalo Rocha Coura.

• **Funding acquisition** – Patrícia Santiago de Oliveira Patrício.

• **Investigation** – Patrícia Santiago de Oliveira Patrício; Ingrid Amélia dos Santos Matusinho; Ítalo Rocha Coura.

• **Methodology** – Ingrid Amélia dos Santos Matusinho; Ítalo Rocha Coura.

• **Project administration** – Patrícia Santiago de Oliveira Patrício.

• **Resources** – Patrícia Santiago de Oliveira Patrício; Patterson Patricio de Souza

• **Software** – NA.

• **Supervision** – Patrícia Santiago de Oliveira Patrício.

• **Validation** – NA.

• **Visualization** – NA.

• **Writing – original draft** – Patrícia Santiago de Oliveira Patrício; Ingrid Amélia dos Santos Matusinho; Ítalo Rocha Coura.

• **Writing – review & editing** – Patrícia Santiago de Oliveira Patrício, Patterson Patricio de Souza.

6 Acknowledgements

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Supplementary Material

Supplementary material accompanies this paper.

Table S1. Statistical Comparison of Mechanical Properties in PPR/BK Composites

Figure S1. DSC curves (a) PPR-30BK, (b) PPR-20BK, (c) PPR-10BK, (d) PPR-30BK.

This material is available as part of the online article from https://doi.org/10.1590/0104-1428.20240044