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Eco-friendly and Affordable Composites Based on Waste Rubber Loaded with Polypropylene and Bagasse Fibers for Flexible Flooring Tiles

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Abstract

Synthetic polymer degradation takes decades to decompose and harms the environment. Indeed, the polymer should be reused to decrease their accumulation. In this study, devulcanized waste rubber (WR), recycled polypropylene (RPP), and bagasse fiber (BF) with different ratios were subjected to study the effect of RPP and bagasse fiber (BF) addition on composites' mechanical properties that included tensile strength (TS), Young's modulus, and elongation. The hardness, abrasion and dynamic mechanical analysis (DMA) were carried out. The experimental testing on the investigated composites showed that adding RPP and BF improved the (TS). Increment is observed for the gel fraction values, showing an adhesion between rubber, plastic and bagasse. The hardness of composites was improved with the addition of RPP and BF. The abrasion loss emphasizes the positive effect of loading RPP and BF into WR with reasonable thermal stability.

Keywords: Waste Rubber; Polypropylene; Bagasse; Recycling; Tiles.

1. Introduction

Waste accumulation increases, not annually, but daily, worldwide [1]. Various problems, such as global warming, air, soil, and water pollution, could be due to the accumulation of wastes in the environment [2]. Synthetic polymers, such as plastic, rubber, etc., are not degradable in the environment naturally [3]. These kinds of materials need severe decay procedures to decompose [4]. Rubber waste is invasive to the environment. It increased progressively, especially in tires that are used aggressively over all the world and do not decompose easily [5, 6]. For cars with more than billions of tires, tire waste is almost directly related to the excessive consumption of tires during the last decade [7]. This enormous volume of non-biodegradable waste occupies a considerable space and endangers the ecosystem [8]. Burning or utilizing tires as fuel can release dangerous gases into the atmosphere and detrimentally pollute the natural air supply [9]. Some synthetic polymers are toxic components found in tire rubber. They are extremely harmful to people [10, 11]. On the other side, plastics are used globally for different purposes, such as pipes, packages, packaging, etc. [12]. Indeed, the accumulation of non-biodegradable polymers could lead to a hazardous impact not only on the environment but also against human health [13]. Propylene monomer is polymerized to create polypropylene (PP), a synthetic polyolefin resin. PP is molded or extruded into a wide range of plastic products that require toughness, flexibility, lightweight, and thermal resistance [14]. In this context, agricultural wastes are unique in degradability rate. They may decompose spontaneously, causing fire and contaminations [15]. Sugarcane bagasse (SCB) is an agriculture waste that is produced as a byproduct in sugarcane industrial processes [16]. It has around 25% hemicellulose, 25% lignin, and 50% cellulose. SCB can be the perfect candidate for microbial processes that produce value-added products because of its widespread availability [17]. Recycling such non-degradable materials reduces their toxicity and

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negative impact against the environment. Moreover, the addition of biodegradable natural materials may contribute to decrease their toxicity [18, 19]. Furthermore, the agricultural waste is a zero-value biomass that added value when replacing virgin biosources [20]. Composite materials containing rubber and plastics widely use a virgin grade of them polymers in various applications such packaging, house hold apparatus, constriction materials and different tools [21, 22]. Otherwise, the devulcanization of waste rubber is a complicated process in which the sulfur bonds break down [23]. In this manner, interface compatibility could play an important role in waste rubber without devulcanization and polypropylene. Some kinds of plastic-rubber compatible interface materials, such as malic anhydride and succinic anhydride, could be used to increase interfacial compatibility. They could enhance the mechanical properties of these polymeric substrates. This study aims to formulate polymeric-based composites based on recycled tire waste rubber, recycled polypropylene, and bagasse fibers to produce eco-friendly, low-cost flexible flooring tiles. The mechanical, thermal, and morphological properties of the prepared composites were investigated.

2. Materials and methods

2.1. Materials

The waste tire powder was obtained from local consumed vehicle tires, Cairo, Egypt. The waste tires were cleaned and cut into small pieces. These pieces were chopped and grinded to provide a powder representing the waste tire powder. Bagasse fibers (BF) were collected from Quena Paper Industry Company, Egypt. BF were grinded to obtain fine powder. Recycled polypropylene (RPP) was delivered from Al-amal Co., Egypt. All chemicals and reagents used in this work were purchased from Loba Chem., India in analytical grade without purification. The particle size of (WTP) was determined through sieve analysis technique, and the diameter was classified as shown in Table 1 (for 100 g.), the main WTP particle size ranged between 300 and 425 microns.

Table 1. Particle size distribution of WTP (100 g.).

Micron	850	600	425	300	80	<60
WTP (g)	0.5	10	35	44	05	4.6

The chemical analysis of bagasse is 45-55, 20-25, 28, 20, 5 and 2% as cellulose, hemicelluloses, pentosane, lignin, sugar and other minerals, respectively. The bagasse powder was characterized to demonstrate its particle size. Upon investigation through sieve analysis technique, it was found that the diameter of the bagasse powder was classified as shown in Table 2 (for 100 g.), the main BF particle size ranged between 425 and 850 microns.

Table 2. Particle size distribution of bagasse fiber (for 100 g.).

Micron	850	600	425	300	80	<60
BF (g)	19.3	30.0	20.5	13.0	12.8	4.01

2.2. Samples preparation

The composites were prepared according to Table 3. Waste tire rubber was subjected to a thermo-mechanical devulcanization process on a roll mill for 30 min. with a fixed distance between the 2 roll mills, producing devulcanized waste rubber (WR). The waste rubber powder was rolled and masticated with maintaining the temperature in the range of 150 ^{o}C. The friction of rubber molecules upon mechanical shearing led to elevate the matrix temperature. The shear stress was applied to cut the carbon - sulfur bonds. RPP was introduced to the devulcanized rubber on the mill for 5 min., followed by loading bagasse with continuous milling for an extra 5 min. For each sample, sulfur was used to revulcanize the used rubber. Maleic anhydride (MA), was used as a coupling agent. The composites were pressed mechanically in rectangular frames with thickness of 2 mm at 165 ^{o}C; for 15 min., forming composite sheets.

Table 3. Composition of the prepared waste rubber-based composites.

Sample	**WR (phr)**	**RPP (phr)**	**Sample**	**WR (phr)**	**RPP (phr)**	**BF (phr)**
P1	100	00				
P2	90	10	P7	90	7	3
P3	80	20	P8	80	14	6
P4	70	30	P9	70	21	9
P5	60	40	P10	60	28	12
P6	50	50	P11	50	35	15

2.3. Characterization

The mass fraction of the insoluble rubber fraction in the samples was defined according to Eq.1. Subsequently, the samples were swelled in toluene and then extracted for 72 h to get rid of any unreacted components and small fragments. The samples were dried at 80 ^{o}C in the oven and reweighed [24].

$$\boldsymbol{Gel fraction}\ (\%) = \frac{m_s}{m_i} \times 100 \qquad \textbf{Eq. 1}$$

where m_s and m_i are the weight of the dried sample after extraction and the weight of the sample before extraction, respectively.

Mechanical properties including tensile strength and Young`s modulus using determined using an Instron 34SC-5 universal tensile testing machine, UK, equipped with a load cell of 5 kN and a crosshead speed of 10 mm.min-1, according to ASTM D 882-18. The dumbbell specimens were die-cut from the casted films. The average of three parallel trials for each film was recorded. Hardness was carried out using shore A, Harteprufgerate, DIN 53505, ASTM 2240. The abrasion was tested using Taber Rotary Abrasion 5150, abrading wheel H-22, auxiliary weight 500 g load 1000 cycles.

The dynamic mechanical properties were evaluated through the dynamic mechanical analyzer DMA1 (Mettler Toledo, Switzerland). The measurements were carried out to assess the different dynamic-mechanical moduli of the tested samples. A single cantilever mode was employed. The samples were subjected to a cyclic shear with force amplitude of 0.1 N at 1 Hz frequency. Storage modulus (E'), loss (E") and loss factor (tan δ) were obtained in the temperature range of -20 to 120 ^{o}C with a heating rate of 5 ^{o}C/min.

Scanning electron microscopy (SEM) (JSM 6360LV, EOL/Noran) was used to examine the surface morphology of the prepared composites. SEM images were processed using the free software ImageJ.

TGA analysis was performed on samples using a Perkin Elmer thermogravimetric analyzer by heating the sample to 600 °C at a rate of 10 °C/min under N_2 atmosphere.

3. Results and Discussion

The investigated composites were formulated using different ratios of waste rubber, waste polypropylene and bagasse fibers. The composites were visually examined and the sheets performed with a uniformly appearance with a dominant dark color of waste rubber through all samples.

Fig. 1 shows the gel fraction percentage of WR in comparison to other composite formulas. About 79% of the gel fraction associated with the network material that came from a network-forming polymerization or crosslinking procedure was represented by the devulcanized rubber waste sample (1). Furthermore, the fraction percentages have increased gradually upon elevating the content of polypropylene waste. This feature may be correlated to the computability between the mixed phases that reach a value of 97.77% for composite P6. However, in contrast to the WR, the BF addition raised the gel fraction in composite P7 to 87% and 98% in composite P11. These findings are in excellent agreement with Boopasiri, et al 2021 [25].

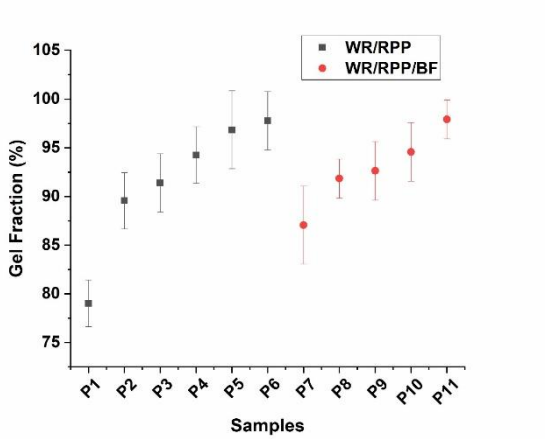

Fig. 1. The gel fraction of the waste rubber-based composites.

The tensile strength (TS) for the formulated composites is illustrated in Fig. 2. The neat WR was presented with a moderate tensile strength (5MPa). It decreases upon inserting of RPP to the rubber matrix. Initially, loading RPP showed a decrease in TS. However, tensile strength increased gradually upon increasing RPP content to reach 9.4 MPa for 50 phr of RPP. Similarly, inserting BF decreased the TS recorded as 2.2 MPa. Meanwhile, composite P10 recorded the highest value 7.31 MPa compared to all samples containing BF. In this context, loading RPP may decrease the TS of WR. It was recovered starting from loading 40 phr of RPP. The addition of BF also decreases the TS (composites P7 and P8). However, the samples P9 to P11 exhibited an improvement in TS. This is not only due to the increment of BF content but also due to the relatively high RPP percentage. These findings could be due to the compatible blending of the waste polymers and cellulosic waste together, achieving an appropriate interfacial linkage with acceptable tensile strength values [26].

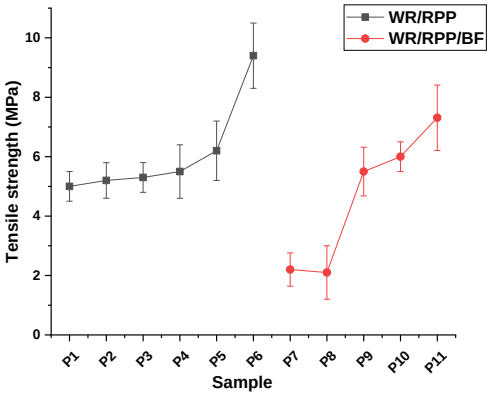

Fig. 2. Tensile strength (TS) of the formulated composites.

Fig. 3a presents Young's modulus values referring to the elasticity of the composite samples [27]. The WR sample displayed the lowest value of Young's modulus at 118 MPa. Blending RPP with

devulcanized WR led to a decrease in the elasticity. This behavior could be in accordance with tensile strength findings. Moreover, the addition of BF recorded a decrease in elasticity as well. These observations may be correlated to the compatibility between the composite components. They significantly influence the composite performance in investigations of different properties investigations. On the other hand, the elongation in mm (Fig. 3b) was observed at the highest value of 8 mm for the WR sample. However, adding RPP decreased the elongation value to 2 mm in composite P6. In addition, sample P7 observed a slightly decrease after addition of BF decreased the elongation. It is well known that Young's modulus values go in the opposite direction with elongation. The results of composites samples illustrated a good elasticity that decreased with the addition of RPP and almost disappeared with the addition of BF [28].

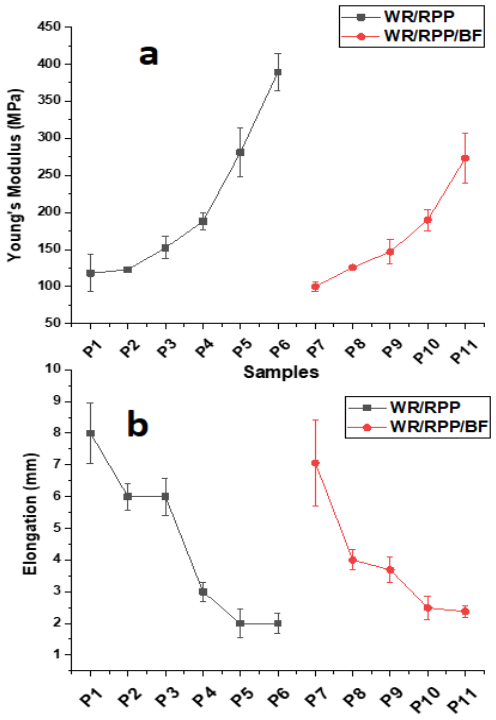

Fig. 3. (a) Young's modulus and (b) Elongation of the formulated composites.

The hardness of the formulated composites is illustrated in Fig. 4. According to the displayed data, RPP plays a role in the hardness value. Its content varies proportionally with hardness values. The sample of WR loaded with 50 phr RPP recorded 94.7N/mm^2. In contrast, the WR showed the lowest value of all samples (63.5 N/mm^2). However, addition of BF played a role in raising the hardness value. BF and RPP can enhance the hardness according to their tough structure compared to that of the elastic rubber.

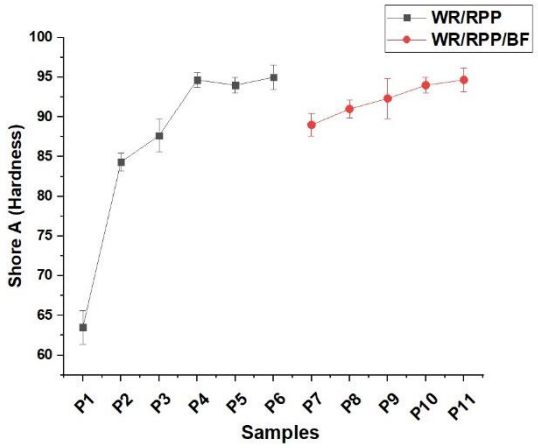

Fig. 4. Hardness of formulated composites.

The neat WR and the formulated composites abrasion results are shown in Fig. 5. Indeed, the addition of RPP led to decrease in the abrasion loss. This is due to the resistance of RPP and its solidarity compared to native WR and the composite P6 displayed the lowest abrasion loss in comparison with the WR sample. In addition, the BF plays a role in decreasing the abrasion loss. This could be due to the compatibility between the composites components, which agrees with the previous literature [29]. In this context, the abrasion loss affirmed that the RPP is supported the abrasion loss as well as the BF.

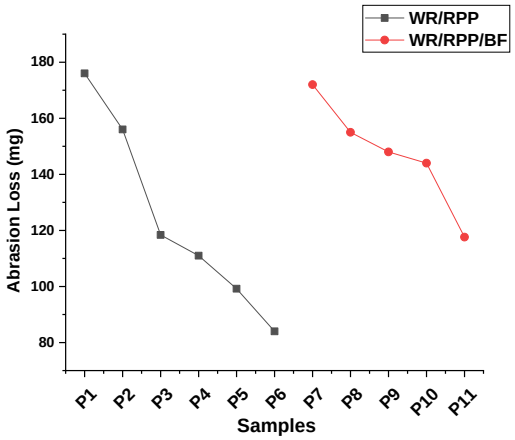

Fig. 5. The abrasion loss of the formulated composites.

Fig. 6 shows the dynamic mechanical analysis (DMA) of devulcanized WR and its composites with RPP. Upon increasing the temperature from -20 oC, the storage and loss moduli of devulcanized rubber and its composites showed decrements in these values as shown in Fig. 6 (a and b). These dynamic properties passed through various regions upon elevating the temperatures. A glassy stage possessing high storage (E') and loss (E") moduli. Such behavior can be referred to as the limited mobility of the elastomeric chains. After that, a transition region displaying a

modulus decline can be observed. It indicates the transfer from a glassy state to a rubbery one. The following elastomeric rubbery area showed low moduli values. It points out to the variation in dissipated energy with high mobility of the devulcanized rubber chains [30]. Loading RPP in the rubber domain increased E' and E'' values, showing more rigid polymeric chains than sole waste rubber. The introduction of BF decreased the stiffness of elastomeric chains with these samples' relevant segmental motion and stress relaxation properties [31]. BF interacted well with WR and RPP. It is noticed in Fig. 6c that WR/RPP 50:50 composite showed the lowest values of tan δ values when compared to the other samples. This feature suggests a fine distribution of these cellulosic fibers in the elastomeric matrix.

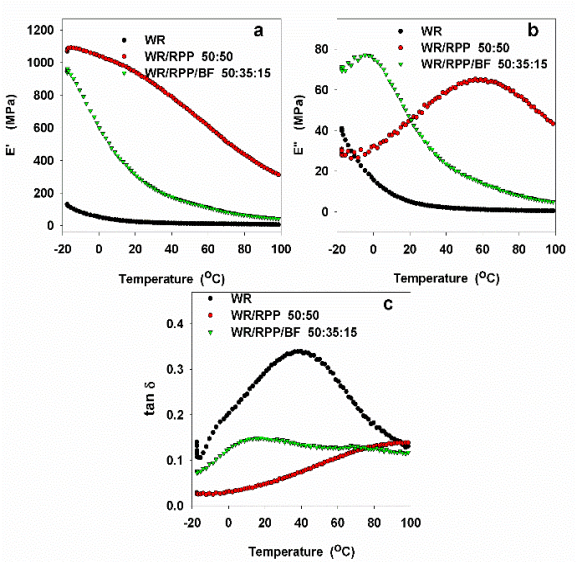

Fig. 6. Dynamic mechanical analysis for (a) storage (b) loss moduli (c) tan delta vs. temperature of waste rubber-based treated composites.

TGA technique was performed to monitor the changes in the thermal stability of the prepared samples. Fig. 7a represents the thermal decomposition of WR samples beside that of vulcanized rubber (VR) which was plotted for comparison with devulcanized waste rubber sample P1. Below 285 °C, a low weight loss % is observed. There is a notable decrease in weight, taking place around 305 °C. Hereby, the organic fragments in tested samples degrade at elevated temperatures. Upon comparing WR with the other composites loaded with RPP and BF, it can be noticed that composite P6 exhibits a higher thermal stability than WR. This behavior elucidates the interaction between the elastomeric chains of WR and those of RPP. It assumes an interfacial adhesion between the present polymeric materials with improved thermal stability [32]. Further weight loss arises at higher temperatures as a result of liberating unstable chemical components. Above 420 °C, the degradation of amorphous polypropylene and rubber chains occurs. When BF was loaded, a slight decrease in the thermal stability arises due to the existence of these cellulosic fibers. Cellulosic substrates decompose through several steps. Hemicellulose decomposes at less than 330 ºC. Lignin shows thermal degradation above 500 ºC. Fig. 7b illustrates the derivative thermograms of the investigated composites as a function of temperature. The peaks in the differential curves coincide with mass conversion. WR shows various peaks with a wide thermal decomposition range due to the multicomponent structure in waste rubber. It comprises inorganic fillers alongside carbon black. RPP elucidated a shifted peak from that of WR. BF did not show significant changes after being loaded to WR/RPP composite. Furthermore, a breakdown in C-C bonds takes place as well as the presence of hollow cellulose and volatiles in bagasse. Thermal decomposition continues while raising the temperature till bagasse is transformed into char.

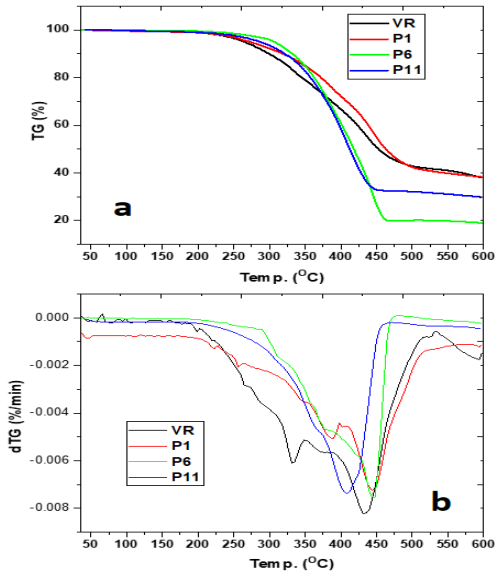

Fig. 7. (a) TGA and (b) dTG thermograms of waste rubber-based composites.

The material's surface topography can be observed using the SEM. Fig. 8 displays the morphology of WR and its composites with RPP and/or BF. Generally, a mechanical force has improved the surface roughness and deteriorated the rubber powder's surface. However, a chemical action raised the surface activity groups [33]. WR was shown in Fig. 8a with a low magnification. It is observed that a basically surface morphology appeared as elastomers with irregular shapes. The high magnification SEM image (Fig. 8b) illustrated an aggregated external facade with irregular bulks expressing the surface morphology of the samples [34]. The composite P6 is displayed in low magnification (8c) and high magnification (8d) with a

1:1 ratio of rubber and RPP. The low-magnification image demonstrates a rough surface. Moreover, the high magnification image displays two phases of a matrix with such compatibility. The high-magnification image process in red marked the rubber matrix, and the gray part relates to RPP. In addition, the green color processed image observed a plastic matrix in a ground with black color. The green parts refer to plastic materials (RPP) that appear as embedded parts into the elastomeric matrix. Moreover, the black space in the processed image is referred to as the rubber matrix. It is almost covered with RPP. These observations affirmed a convenient interaction between two polymers. They emphasized the good mechanical properties of composite P6. Additionally, the composite P11 image with low magnification (Fig. 8e) illustrated an irregular surface terrain with some slits. Otherwise, the high magnification image (Fig. 8f) showed a surface similar to that of composite P6 with fibers structure overall the surface that due to the presence of BF. The processed image in red showed the major matrix (rubber) is spreading across the surface with some gray parts. They denotes to the presence of fibers and RPP. In the following image, RPP appears in green. BF are shown in a clear view as longitudinal fibers covering most of the surface. The dark area revealed that the elastomeric matrix is well embedded with RPP and BF. These observations defined the nature of dispersion and emphasized the interaction between WR, RPP and BF as well.

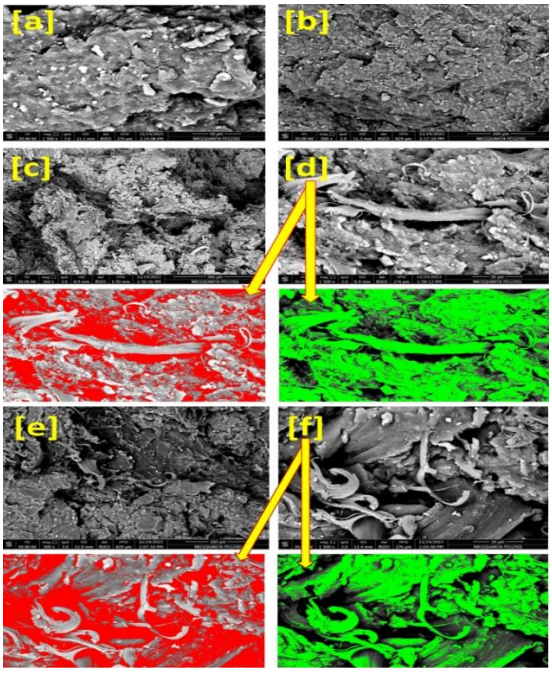

Fig. 8. SEM images of: (a), (b) WR; (c), (d) WR/RPP 50:50; (e), (f) WR/RPP/BF 50:35:15.

4. Conclusion

Synthetic polymers accumulation excessively affects the environment according to the decomposition rate. In this manner, reusing such polymers is considered as a tool to decrease their hazardous effect. This study formulated the composites based on all waste polymers using WR, RPP, and BF. Mechanical properties were observed that the tensile strength, Young`s modulus and elongation properties were not enhanced scientifically. The hardness of composites was improved with the addition of RPP and BF. The abrasion loss emphasizes the positive effect of loading RPP and BF into WR with reasonable thermal stability. Finally, the finding observed that the composite produced containing WR, RPP and BF could be used as a tile in the floor construction applications according to the hardness and abrasion loss properties.

5. Conflicts of interest

There are no conflicts to declare.

6. Funding

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