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Development of Wood Polymer Composites from Recycled Wood and Plastic Waste: Thermal and Mechanical Properties

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Abstract: The depletion of natural resources due to the aggressive industrialization in the last decades has brought considerable attention to research aimed at developing green and sustainable products using eco-friendly materials. The purpose of the current study was to develop wood polymer composites (WPCs) using recycled plastic waste (RPW) generated from university laboratories and recycled wood waste (RWW) from construction and demolition (C&D) activities by melt-blending technique. The WPCs were characterised for their mechanical and thermal properties, as well as water uptake and morphology. The SEM micrograph indicated good interaction between RWW and RPW matrix. The mechanical strength of the WPCs was found to increase from 26.59 to 34.30 MPa, with an increase of the RWW content in the matrix. The thermal stability was higher in the composite with a higher percentage of RWW in the matrix. The wettability results indicated that the composite with a higher RWW (20%) had a higher water uptake. These results suggest that the produced WPCs can be a promising environmental-friendly material, while maintaining good mechanical, thermal, and wettability properties.

Keywords: recycled plastic waste; recycled wood waste; wood polymer composites; mechanical properties; thermal properties; wettability properties

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1. Introduction

In recent years, given the limits of natural resources, the materials and manufacturing sectors have gradually shifted their focus to the development of biodegradable and sustainable materials or products [1]. Wood is a renewable and low-carbon material [2,3]. It has been widely used in different interior and exterior applications [4,5], such as decking materials [6], fencing [7], landscaping timbers [8], furniture, and automobile products [9]. However, the main drawbacks of wood are their liability to distort with the absorption of moisture and their vulnerability to degradation by microorganisms [10,11].

The formation of wood polymer composites (WPCs) has been considered a good way to minimize these troublesome inherent properties of wood. WPCs consist of reinforcing wood fibers incorporated into a continuous polymer matrix. The combination of wood and polymers has been extensively studied due to their reduced environmental impact and low cost owing to the wood fibers coming from industrial or agricultural waste. WPCs possess superior efficiency such as high specific strength, stiffness, high sustainability, lower water intake, high dimensional stability over their lifetime, and durability against environmental impacts [12–14]. The WPCs cater a wide range of products, including as decking, fencing and railing in building industries, and for manufacturing of interior and exterior parts in automotive industries. There are a few major manufacturers of WPCs in Australia, such as ModWood Technologies, Advanced Plastic Recycling (APR), and Tuff Deck—Composite Decking Melbourne.

Previously, other researchers had developed the WPCs by using virgin polymers, such as polypropylene (PP), high-density polyethylene (HDPE), low-density polyethylene (LDPE), polyethylene terephthalate (PET) etc. With the increased amount of plastic waste generated and resulting in a significant proportion in municipal solid waste (MSW), attempts have been made to recycle the post-consumer plastics in the production of WPCs in order to offsets their ecological impacts [15–17].

There are a number of published studies on the WPCs made of recycled polymers. Gulitah et al. [18] developed WPCs from different recycled polymers, such as polypropylene (PP), high-density polyethylene (HDPE), and low-density polyethylene (LDPE), mixed at different ratios with wood fibres (WF). The mechanical properties of PP:WF at 50:50 had the highest value in tensile strength (7.87 MPa), modulus of elasticity (MOE, 520.81 MPa), and modulus of rupture (MOR, 5.55 MPa) compared to HDPE-WF and LDPE-WF composites at the same composition. Novak et al. [19] studied the mechanical and water absorption properties of WPCs fabricated with recycled low-density polyethylene (rLDPE) and date palm wood (DPW) powder with concentrations ranging from 10 wt·% to 70 wt·%. The results showed that Young's modulus, flexural strength, and water absorption increased with the increase of the filler. Wicaksono et al. [20] studied the mechanical and physical aspects of PP and LDPE polymer waste for the production of WPCs. Teak wood powder was used as the filler material. The results showed that LDPE based composites resulted in better mechanical and physical properties than PP based composites. The best composition of WPC was 70% LDPE and 30% teak wood powder. Medupin et al. [21] investigated the mechanical properties of WPCs produced from the used wood and rLDPE. The composites were manufactured using compression moulding techniques. The results indicated that the water absorption rate was high in the first few hours, and the highest water absorption was observed at 60% reinforcement.

Eco-friendly and sustainable composites have been produced to save the ecosystem [9,22,23]. However, most of the research on WPCs focused on the use of either recycled plastic or recycled wood [24,25]. Research on the development of fully recycled WPCs made of both recycled plastic and recycled wood is still inadequate, which has inspired researchers to further explore in this area. Therefore, in this study, recyclable polymer waste collected from biological laboratories and wood waste collected from the construction and demolition (C&D) activities were used as the matrix and reinforcement to produce WPCs. The morphology was evaluated to understand the interaction between filler and matrix. In addition, mechanical, thermal, and wettability properties of the developed WPCs were then analysed in detail.

2. Materials and Methods

2.1. Materials

Recycled plastic waste (RPW)—centrifuge tubes, plastic tubes, and small medical containers—was collected from the Applied Sciences Laboratories (AS3.13 and AS3.14) at La Trobe University, Bendigo, Australia. They mainly consisted of mixed thermoplastic polymers, such as HDPE and LDPE. Recycled wood waste (RWW) was supplied by Hopley Recycling Pty Ltd., Bendigo, Australia. Sodium stearate and sodium hydroxide (NaOH) solutions were purchased from Bunnings, Bendigo, Australia. Hydrochloric acid (HCl) was purchased from Sigma-Aldrich Pty Ltd., Melbourne, Australia.

2.2. Cleaning and Segregating Waste Materials

2.2.1. Cleaning of Recycled Polymer Waste

The polymer waste was initially crushed into small pieces of approximately 8 mm using a polymer crusher (Dongguan Zhongli Instrument Technology Co., Ltd., Dongguan, Guangdong, China). After crushing, the plastic pieces were washed with NaOH solution (5%) for 90 min to remove excess dirt and other debris. Later, the polymer waste was washed twice using sodium stearate, followed by water. Finally, the obtained polymer

waste was dried in a vacuum oven at 45 °C for 48 h to remove excess moisture [26]. Hereafter, polymer waste is referred to as RPW.

2.2.2. Cleaning of Recycled Wood Waste

The wood waste was collected from C&D activities in various sizes, and it was sieved and cleaned using the methodology reported by Medupin et al. [21]. The sieving was conducted according to the ASTM E11 sieve method to obtain 0.05 mm particles. Afterwards, the RWW was washed with 20% NaOH solution followed by 10 M HCl solution to remove excess alkaline, dirt, and residues from the surface of RWW. Later, the RWW was washed with deionised water three times and dried in an oven for 24 h at 60 °C [21]. Hereafter, the wood waste is referred to as RWW.

2.2.3. Preparation of Wood Polymer Composites (WPCs)

The dried RPW and RWW were mixed into the various compositions, as shown in Table 1 [27,28]. The illustration of RWW reinforced RPW composites was shown in Figure 1. An internal batch mixer (model ZL-3011, Dongguan Zhongli Instrument Technology Co., Ltd., Guangdong, China) was used to compound the composites. The hopper temperature was set at 155 °C, and the spindle speed was maintained at 8 rpm. The melt-mixing process was performed for 25 min, with the spindle rotation direction changed every 15 min. Later, the obtained RPW–RWW composites with varied compositions were crushed into small pieces using a polymer crusher and processed in a microinjection moulder (Xplore instruments BV., Sittards, The Netherlands) [20] to obtain dumbbell-shaped specimens (ASTM D638 Type V) for further characterisation. The parameters used for injection moulding are presented in Table 2. The illustration of injection moulding is presented in Figure 2. The WPCs produced were referred to as RPW, RPW–RWW1, RPW–RWW2, RPW–RWW3, and RPW–RRW4. The RWW was considered as neat wood waste particles without any further treatment.

Table 1. Compositions of WPCs.

Sample	RPW (wt·%)	RWW (wt·%)
RPW	100	0
RPW-RWW1	90	10
RPW-RWW2	80	20
RPW-RWW3	70	30
RPW-RWW4	60	40
RWW	0	100

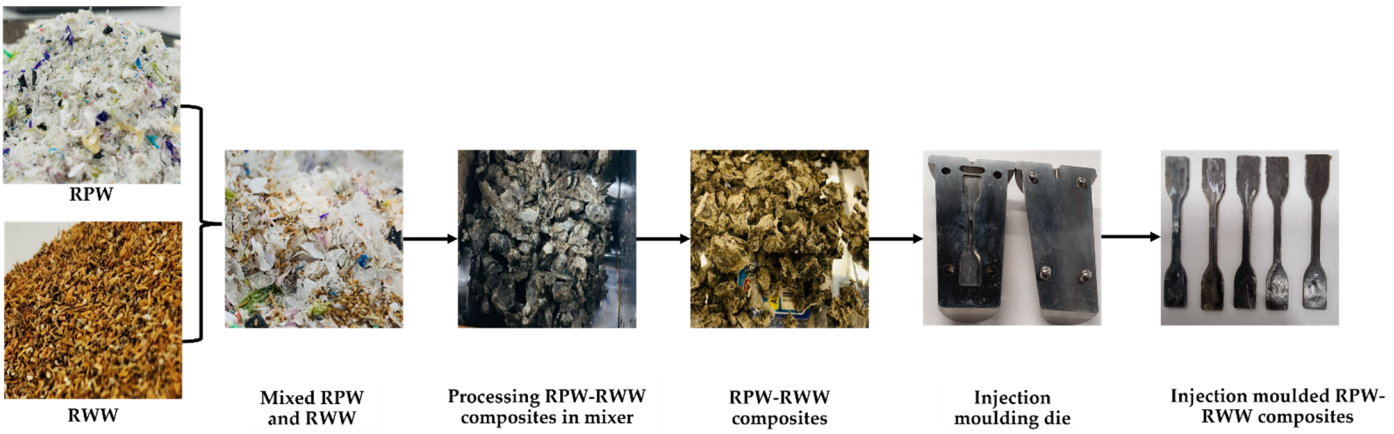

Figure 1. Illustration of the production RPW-RWW composites.

Table 2. Parameters used for injection moulding.

Parameter	Value
Mould temperature	45 °C
Melt temperature	155 °C
Pressure	8 Bar
Time step 1	2 min
Time step 2	3 min
Time step 3	3 min

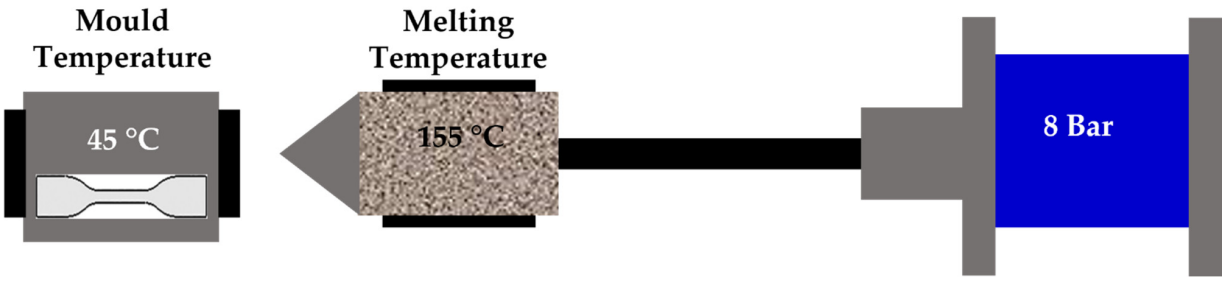

Figure 2. Illustration of injection moulding process.

2.3. Morphological Analysis

The morphology of the RPW–RWW composites was observed using a scanning electron microscope (SEM, Hitachi 3030, Tokyo, Japan). The micrographs were taken at variable pressures under voltage ranging from 10 kV to 15 kV.

2.4. Thermal Properties

The thermal properties of the samples were analysed using a thermogravimetric analyser (TGA 4000, Perkin Elmer, Waltham, MI, USA). The thermographs were obtained at thermal scan temperatures ranging from 30 °C to 800 °C at 30 °C/min under nitrogen atmosphere.

2.5. Mechanical Properties

The mechanical properties of the produced WPCs were investigated using an Instron 5980 universal testing machine (Norwood, MI, USA). Tensile tests were performed using specimens in dumbbell shapes (ASTM D638 Type IV) with a dimension of 10×15 mm^2. The tests were conducted under a 10 mm/min crosshead speed with a load of 10 kN. The stress-strain analysis was recorded from the tensile testing.

Each sample was performed in triplicate and the mean values as well as standard deviation (SD) were reported.

2.6. Hardness

The hardness of the composites was tested using a Vickers microhardness tester (Durascan 32 series G5, Kuchl, Austria). The size of the sample was according to ASTM standards (ASTM E 384) at $20 \times 20 \times 3$ mm^3. An HV 0.2 intender was used as the force on the sample for 10 s. After the indentation, the sample was analysed using a 10× camera, and the HV value was presented graphically. The test method was carried out according to the literature [29,30]. Each sample was performed in triplicate and the mean values as well as standard deviation (SD) were reported.

2.7. Water Absorption

The water absorption tests were conducted according to the ASTM D570 method. The specimens were initially weighed and immersed in water at ambient conditions. The samples were weighed at regular intervals (2, 4, 6, 8, 10, 12, 14, 16, 18, and 20 days). The percentage of absorbed water (W) in the composite was calculated according to Equation (1):

$$W = [(W_2 - W_1)/W_1] \times 100 \quad (1)$$

where W_1 and W_2 are the sample weights before and after immersion.

2.8. Water Contact Angle

The wettability of the WPCs was measured by the contact angle measurement using the sessile drop technique. A droplet was placed on the surface of each RPW–RWW composite using a micrometre syringe. The contact angle was measured by scanning the droplet profile for 20 s using an Attension Theta Flex instrument (Biolin Scientific, Gothenburg, Sweden). The size of the water droplets was maintained at about 2–2.5 µL to avoid the effect of weight [31].

2.9. Statistical analysis

Statistical analyses were performed using the ANOVA method in GraphPad Prism 9.0 (GraphPad Software, Inc., San Diego, CA, USA). The investigation was carried out for three replicates (n = 3) for each data set in mechanical properties and presented as mean ± standard deviation (SD) unless otherwise stated. A significance level of the p-value of $\leq$0.05 was determined to be significant (*). Error bars in all figures represent the standard error of the mean. The analysis was performed based on the following literature [32,33].

3. Results and Discussion

3.1. Morphology

Micrographs of RWW, RPW, and RPW–RWW composites are shown in Figure 3. Figure 3a shows the cross-sectional view of the pure polymer, which was smooth and layered with even dispersion. Figure 3b shows the micrograph of 0.05 mm sieved RWW particles, which were randomly spaced with irregular sizes. In addition, the RWW can be seen in the RPW–RWW composite, indicating well dispersion in RPW. The fractured cross-sectional surfaces of the lowest and highest magnification of RPW–RWW4 composites are shown in Figure 3c,d. Figure 3c shows that part of the RWW (highlighted in a red circle) is filled with RPW, which is expected to help in increasing the strength of the composites due to the mechanical interlocking [34]. The Figure 3e–h micrographs indicate that there was no clear gap between the RWW and RPW matrix, showing a good interfacial bonding and indicating the stress transfer from the weaker matrix to the strong wood fibre, as reported by Adhikary et al. [34]. The strength and interfacial interactions determined the composite failure mode and micromechanical deformation [35]. It was evident from the micrograph that there was even dispersion of RPW with RWW, as the RWW was highlighted in Figure 3c,d. Furthermore, the good interfacial bonding between filler and matrix tended to display good physical and mechanical properties [10].

3.2. Thermal Properties

The thermographs of RPW, RWW, and RPW–RWW with different weight fractions are shown in Figure 4. The weight loss of RPW occurred in a single-step degradation process over the temperature range of 300 °C to 500 °C. The RWW decomposed between 200 °C and 600 °C, with the highest degradation rate at 350 °C [36]. According to literature reports [37–39], the degradation of wood at 220 °C to 325 °C was due to the decomposition of the hemicellulose. The weight reduction in the range of 300 °C to 400 °C was due to cellulose degradation, and lignin decomposition occurred in the wide temperature range from 200 °C to 600 °C. It was clearly observed that there were two stages of weight loss occurred for the RPW and its composites. The first stage of weight loss occurred, corresponding to the charring of the hemicellulose, cellulose, and lignin contents of the wood particles [40]. The second stage of weight reduction happened due to the breakdown of polymers [41]. From the thermographs, it can be seen that RPW showed the highest thermal stability, while the thermal stability for composites decreased with the addition of wood particles due to the low thermal stability of wood [42,43]. The degradation rate

that occurred for the composites is shown in Table 3. The obtained char residues at the end show that higher char resulted in RWW than in RPW. The char residue also acted as a thermal barrier for the composites, which helped in improving the thermal stability, as reported in the literature [44,45]. Hence, the composites with higher RWW content tended to show lower mass loss in comparison with pure polymer [36,46,47]. Similar observations were made with other types of WPCs [36,44,45,47,48].

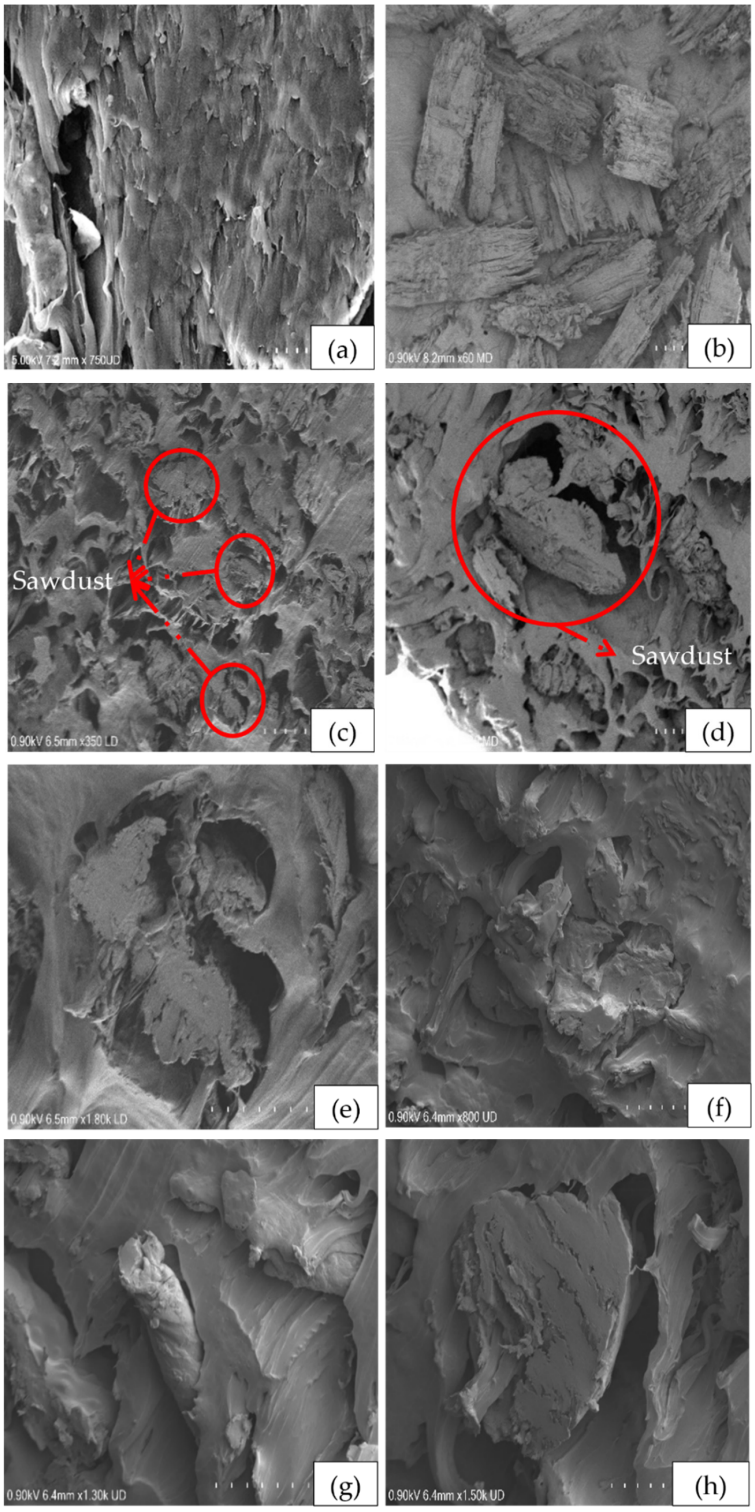

Figure 3. SEM micrographs (**a**) RPW; (**b**) RWW; (**c**) low magnification RPW-RWW4; (**d**) high magnification RPW-RWW4; (**e**–**h**) RPW-RWW4 cross-sectional images at different positions.

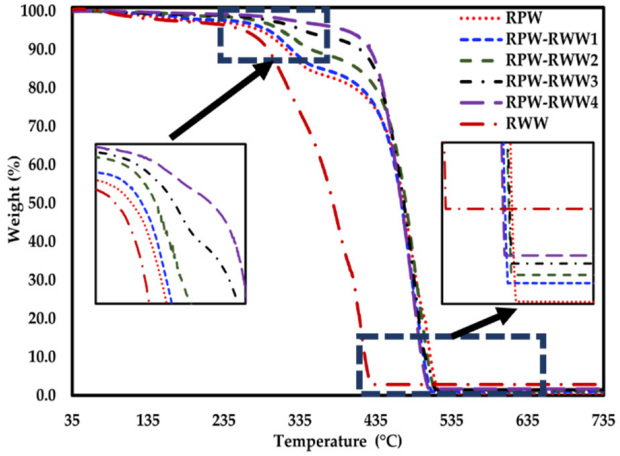

Figure 4. TGA curves of RPW-RWW composites.

Table 3. Thermal properties of RPW-RWW composites by TGA.

Composites	Onset Temperature (°C)	End Set Temperature (°C)	Maximum Decomposition Temperature (°C)	Char Residue at the End of 800 (%)
RPW	305	495	475	0
RPW-RWW1	300	505	480	1.0
RPW-RWW2	298	510	485	1.2
RPW-RWW3	295	515	490	1.5
RPW-RWW4	290	520	493	1.6
RWW	250	450	430	2.5

3.3. Mechanical Properties

The stress–strain behaviour of the RPW–RWW composites under tensile loading is shown in Figures 5 and 6 shows the ultimate tensile strength of the moulded composites. The tensile strength was expanded with an increase in RWW content in the composites. This was due to the excellent dispersion of wood particles within the polymer matrix and the efficient stress transfer from the RWW to the RPW [49]. The tensile strength of pure RPW was observed at 26.5 MPa. The tensile strength of the RPW–RWW1, RPW–RWW2, RPW–RWW3, and RPW–RWW4 was 27.81 MPa, 30.45 MPa, 32.48 MPa, and 34.30 MPa, respectively. Furthermore, Young's modulus was calculated based on the linear slope of the stress–strain curve, as shown in Table 4. RPW showed the lower Young's modulus (2.82 MPa), while RPW–RWW4 showed the highest Young's modulus (3.43 MPa). Najafi et al. [50] reported similar results with WPCs made of recycled plastics, where the stress concentration increased with an increase in the wood content.

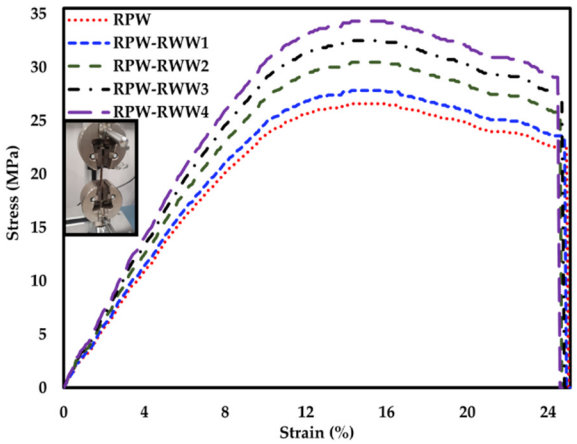

Figure 5. Stress-strain curves of the RPW-RWW composites.

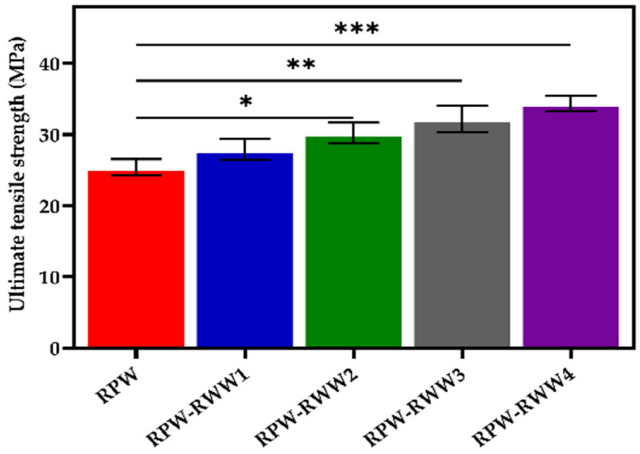

Figure 6. Ultimate tensile strength of RPW-RWW composites (n = 3, * $p \leq 0.05$, ** $p \leq 0.01$, *** $p \leq 0.001$).

Table 4. Mechanical properties of the RPW-RWW composites.

Sample Name	Young's Modulus (MPa)	Ultimate Tensile Strength (MPa)
RPW	2.82	26.59
RPW-RWW1	2.87	27.81
RPW-RWW2	3.06	30.45
RPW-RWW3	3.32	32.48
RPW-RWW4	3.43	34.30

3.4. Hardness

Figure 7 shows the optical micrographs of Vickers microhardness indentation on composites and their hardness values. The RPW had a hardness of 9.21 HV. Whereas the composites had a hardness of 12.08 HV (RPW-RWW1), 14.73 HV (RPW-RWW2), 16.64 HV (RPW-RWW3), and 19.72 HV (RPW-RWW4), respectively. In general, the Vickers hardness values of the RPW–RWW composites increased with the increasing RWW content. There was a significant influence of RWW content on hardness values. The obtained results were similar to reports that demonstrated the hardness behaviour of typical thermoplastic composites containing lignocellulosic fillers [51,52] where the hardness value of WPCs generally increased with an increase in filler.

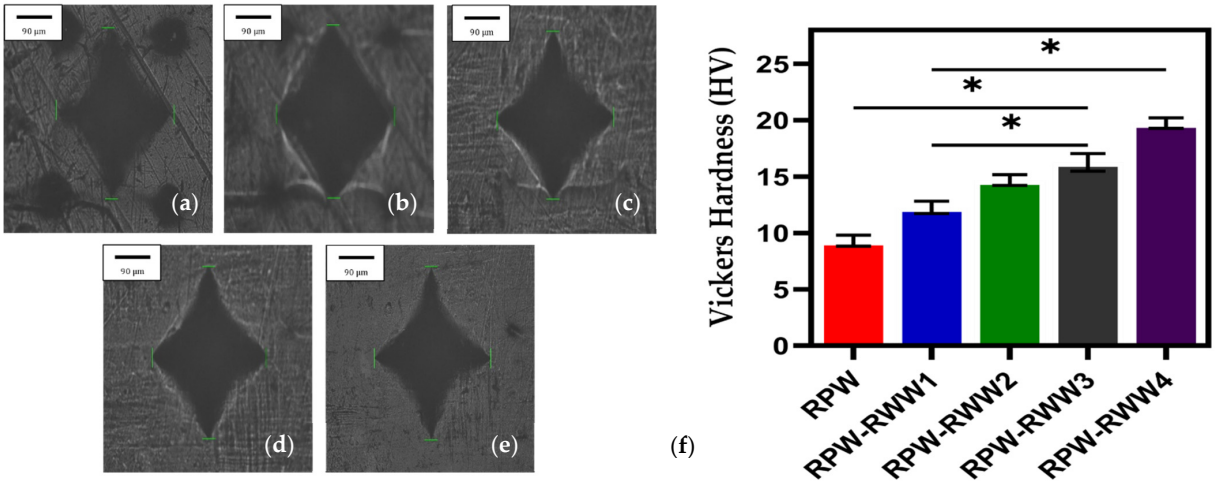

Figure 7. Optical micrograph images of Vickers microhardness indentation on (**a**) RPW; (**b**) RPW-RWW1; (**c**) RPW-RWW2; (**d**) RPW-RWW3; (**e**) RPW-RWW4; (**f**) Vickers hardness number of RPW-RWW composites (n = 3, * $p \leq 0.05$).

3.5. Water Absorption

The water absorption curves of RPW–RWW composites are shown in Figure 8a. Figure 8b shows the images of WPCs immersed in water. It can be observed that the water absorption rate increased rapidly in the first few days and it slowed down after 8 days [52–54]. The water absorption process continued with the prolonged immersion until the specimen reached saturation after 20 days. The percentages of water absorption of RPW, RPW–RWW1, RPW–RWW2, RPW–RWW3, and RPW–RWW4 were 1.2%, 4.4%, 8.5%, 15.1%, and 19.9%, respectively, when the specimen was in an equilibrium state of water absorption (allowing the water absorption to change in time range and period of immersion). RPW showed the lowest water absorption percentage, as it is hydrophobic and thus absorbed less water [55]. The incorporation of RWW into RPW displayed that RPW played a significant role in the water absorption due to wood is lignocellulose material and can absorb more water due to the polar group hydroxyl. Furthermore, the obtained results was associated with the findings from the literature [10,56,57].

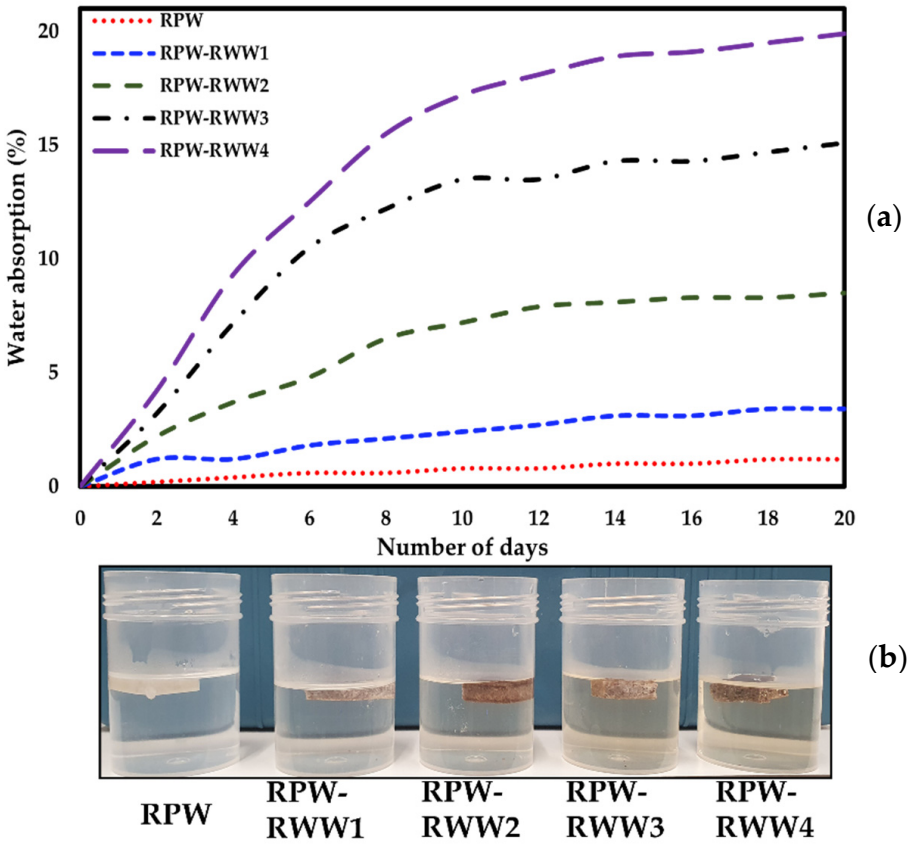

Figure 8. (**a**) Water absorption curves of RPW-RWW composites; (**b**) immersed RPW-RWW composites.

3.6. Water Contact Angle

Water contact angles of the RPW–RWW composites were measured to investigate the wetting properties of the composites. The water contact angle measurement was carried out according to Sdrobiş et al. [58] and Wang et al. [59]. Figure 9 shows the sessile drop photographs and the contact angle's mean value. According to the literature, a contact angle below 90° indicates a good wetting surface for any liquid [33,60–62]. RPW had a higher water contact angle of about 80.94°, followed by the composites with RWW. In general, with the addition of RWW, the water contact angle decreased due to the hydrophilic nature of wood, as shown in Figure 6. As a result, the average contact angle of the composites with RWW decreased from 79.35° to 39.22°. Lazrak et al. [43] reported the drastic decrease in the contact angle was associated with the composites surface roughness.

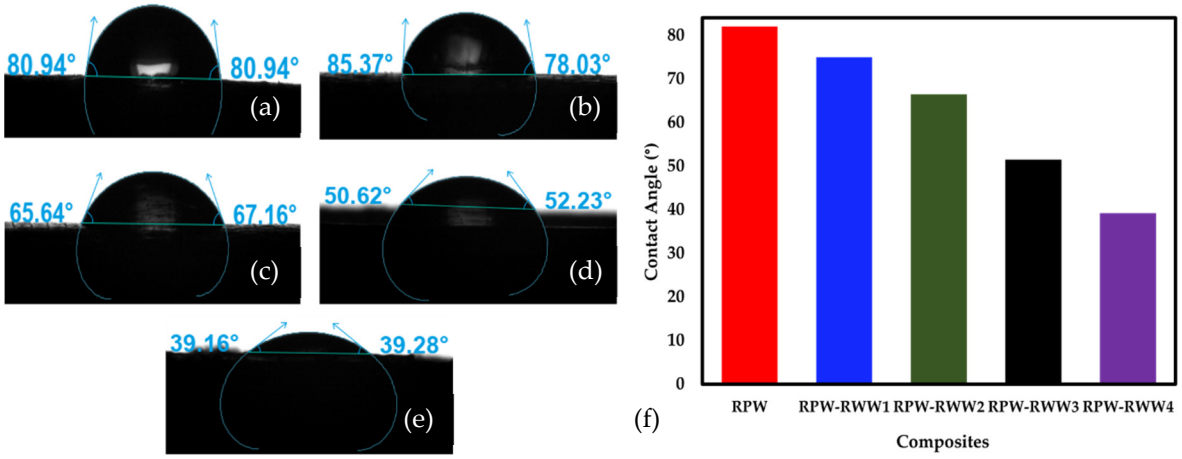

Figure 9. Contact angle and sessile drop images of (**a**) RPW; (**b**) RPW-RWW1; (**c**) RPW-RWW2; (**d**) RPW-RWW3; (**e**) RPW-RWW4; (**f**) Contact angle values of RPW-RWW composites.

4. Conclusions

In this study, the WPCs were successfully fabricated via a melt-mixing process. The RPW–RWW composites were characterised for morphology, mechanical and thermal properties, water absorption, and water contact angle. The SEM micrographs showed homogenous dispersion of RWW in the RPW matrix. The results from mechanical testing revealed that RPW–RWW4 exhibited the highest tensile strength of 34.30 MPa, while RPW showed the lowest (26.59 MPa). RPW–RWW4 showed the highest hardness value of about 19.72 HV and RPW had the lowest hardness value of 7.21 HV. The thermal properties increased, and the degradation rate decreased with the increase of wood waste content, and a very minimal amount of residual char remained. The water absorption (%) increased with increasing wood waste content, as wood is hydrophilic and can absorb more water. The RPW–RWW4 absorbed water up to 20%, while RPW absorbed little water (about 2% during the immersion time). A further study on the recycled WPCs is necessary to analyse the immiscibility, crystallinity, and rheological properties, which would help to evaluate interfacial adhesion between plastic waste and wood waste for different products such as fencing, railing, and decking.

Author Contributions: Conceptualisation: I.K. and S.G.N.; methodology: S.G.N.; validation: S.G.N., I.K. and A.B.K.; formal analysis: S.G.N., I.K. and A.B.K.; investigation: S.G.N. and I.K.; resources: I.K.; data curation: S.G.N.; writing—original draft preparation: S.G.N.; writing—review and editing: I.K., W.K. (Win Kong), W.K. (Wei Kong) and A.B.K.; supervision: I.K.; All authors have read and agreed to the published version of the manuscript.

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