

Research Article

Composites from Thermoplastic Natural Rubber Reinforced Rubberwood Sawdust: Effects of Sawdust Size and Content on Thermal, Physical, and Mechanical Properties

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The aim of this work is to investigate the effects of rubberwood sawdust (RWS) size and content as well as the ratio of natural rubber (NR)/high-density polyethylene (HDPE) blend on properties of RWS reinforced thermoplastic natural rubber (TPNR) composites. The addition of RWS about 30–50 wt% improved the modulus of the rupture and tensile strength of TPNR composites blending with NR/HDPE ratios of 60/40 and 50/50. TPNR composites reinforced with RWS 80 mesh yielded better tensile strength and modulus of rupture than the composites with RWS 40 mesh. The TPNR/RWS composites with larger HDPE content gave higher tensile, flexural, and Shore hardness properties and thermal stability as well as lower water absorption. The TPNR/RWS composites with larger plastic content were therefore suggested for applications requiring high performance of thermal, physical, and mechanical properties.

1. Introduction

A thermoplastic natural rubber (TPNR) is a material blended from a natural rubber (NR) and a thermoplastic, such as polypropylene, polystyrene, and high-density polyethylene (HDPE) [1, 2]. Thus, it shows velvety surfaces and intermediate properties between the NR and the plastic, which provides flexibility in shaping, great recyclability of scrap, and low production cost [3]. Currently, the TPNR is still being developed for various applications such as in footwear, seals, hoses, automobiles, and marine engineering because the common thermoplastic machinery could be used without compounding or a vulcanization process [4]. However, due to the nature of the TPNR as a polymer, it has limitations in some physical and mechanical properties, particularly the stiffness and hardness. The addition of natural fiber (wood fiber or wood sawdust) as reinforcement can improve

the performance of TPNR in the constructional and building applications. Jamil et al. [2] revealed that tensile modulus and Shore hardness increased with rice husk loadings in the TPNR matrix, and the addition of kenaf fiber in the TPNR matrix had significantly increased the tensile, flexural, and impact properties [5]. Further, the natural fibers also offer many advantages such as low cost, high toughness, lightweight, less abrasive on processing tools, and low energy consumption in manufacturing [6, 7].

Recently, the utilization of natural fiber such as bamboo, bagasse, flax, and rubberwood as a replacement to inorganic reinforcements in the plastic composites had been extensively performed [6]. For example, Srivabut et al. [8] applied the rubberwood flour as a reinforcement in recycled polypropylene composites, and the modulus and Shore hardness of recycled polypropylene composites increased linearly with rubberwood flour loadings [6]. Vu et al. [9] revealed that

Young's modulus, flexural strength, and flexural modulus of polypropylene were improved by blending polypropylene with cellulose fibers. Li et al. [10] found that the surface modification of sisal fiber had slightly increased the tensile strength of polylactide composites. Further, Luo et al. [11] showed that using the corn stem as the reinforcement in HDPE composites achieved the highest tensile break strength and flexural strength. Petchwattana and Covavisaruch [12] reported that the increasing additions of rubberwood flour in poly (lactic acid) composites increased the tensile modulus and strength. From this information, the use of rubberwood sawdust (RWS) as a reinforcement in the TPNR composites is of great interest. Because a huge amount of the RWS waste generated from sawmills and furniture industries was greatly found in Thailand, some of the wood waste is eliminated by burning and dumping in space areas [13].

Normally, the thermal, physical, and mechanical properties of wood-polymer composites are influenced by a few factors [14], that is, the mixing and molding parameters [14, 15], wood species [14, 16], and the ratio of the material component [14, 17, 18]. Jamil et al. [2] also concluded that the major factors affecting the critical performance properties of polymer composites were wood flour loading, wood particle size, interfacial adhesion, and wood structure. The chemical composition of wood is also a factor [19]. Therefore, the development of new composite materials to response the constructional and building purposes needs a better understanding for the process of creating wood-polymer composites from the material components [14].

Although extensive research in the area of the plastic composites reinforcing the natural fibers has been conducted, there are just a few studies in the use of the natural fibers to reinforce TPNR composites. No prior report on TPNR composite reinforced RWS was found to focus on the effects of content and particle size as well as the blend ratio of TPNR based on the composites. Hence, the effects of filler content and size on the properties of the composites are needed to be further studied. In the present work, the effects of RWS size and content as well as the ratio of NR/HDPE blend on the thermal, physical, and mechanical properties of TPNR/RWS composites were investigated. The overall results of the current work will facilitate the development of those composites for constructional and building applications, and they will potentially replace expensive building materials like concrete, bricks, and wood lumber in the future.

2. Materials and Methods

2.1. Materials. Natural rubber used in this study was STR 5L grade from the Rubber Estate Organization (Nakhon Si Thammarat, Thailand). High-density polyethylene granules with a melt flow index of 15 g/10 min at 190°C and a density of 0.957 g/cm³ were purchased from IRPC Public Company Limited (Rayong, Thailand). Rubberwood sawdust was supplied by sawmill industry in the Songkhla Province of Thailand. Before compounding, the sawdust was sieved through a standard sieve and classified into two categories: passing 40 mesh (L) and passing 80 mesh (S), and then was dried in an oven at 120°C for 10 h.

2.2. Preparation of NR/HDPE Blends. TPNR was prepared via melt blending of NR/HDPE ratios of 60/40 (R60/P40), 50/50 (R50/P50), 40/60 (R40/P60), and 30/70 (R30/P70) in a corotating twin-screw extruder (Model CTE-D25L40 from Chareon Tut Co. Ltd., Samutprakarn, Thailand). The temperature of seven processing zones in the extruder was set in the range of 155–185°C from the feeding to the die zone, while the screw rotation speed was controlled at 60 rpm. Then, the extruded strand would pass through an air blower and was subsequently pelletized.

2.3. Preparation of TPNR/RWS Composites. TPNR/RWS composites were compounded using the same twin-screw extruder and parameters as with the preparation of the TPNR matrix. Prior to mixing, the TPNR was dry-blended with various loadings (30, 40, 50, and 60 wt%) and different sizes (L and S) of RWS (formulations in Table 1) in a mixer machine. Further, the TPNR/RWS composite pellets were molded in a compression molding machine with a temperature of 180°C and a pressure of 1000 psi for 10 min. The molding was then transferred to a cold compression set and pressed further under the pressure of 1000 psi for 8 min. Subsequently, the sample panels were processed as specimens for mechanical and physical tests, according to the American Society for Testing and Materials (ASTM) standard, such as ASTM D638-99, ASTM D790-92, ASTM D2240-91, and ASTM D570-88.

2.4. Thermal Analysis. Thermogravimetric (TG) tests were carried out using a Perkin Elmer (TGA-7, USA) thermal analyzer. The tests were conducted in the temperature range of 45–700°C under a nitrogen atmosphere at a heating rate of 10°C/min. Approximately 5–6 mg samples were heated in the sample pan. The onset temperature and thermal stability of the NR, HDPE, TPNR, and TPNR/RWS composites were determined from thermogravimetric analysis curves.

2.5. Morphological Analysis. Morphological analysis of the TPNR and the TPNR/RWS composites was carried out using a scanning electron microscope (Model FEI Quanta 400 from FEI Company, Oregon, USA). To avoid phase deformation and any damage, all specimens of the TPNR and the composites were broken in liquid nitrogen. The specimen surfaces were then coated with gold in order to eliminate electron charging during the imaging. They were imaged with magnifications of 500x at an accelerating voltage of 20 kV.

2.6. Characterizations

2.6.1. Mechanical Tests. Tensile and flexural properties were measured by using a Mechanical Universal Testing Machine (Model NRI-TS500-50 from Narin Instruments Co. Ltd., Samutprakarn, Thailand). The tensile testing of the type-IV specimens was conducted according to ASTM D638-99 with a crosshead speed of 5 mm/min. Three-point flexural testing of specimens with nominal dimensions of 4.8 mm × 13 mm × 100 mm was performed according to ASTM D790-92 with a span of 80 mm and a crosshead speed of 2 mm/min.

TABLE 1: Formulation of TPNR and TPNR/RWS composites in experiment.

Sample code	RWS size (mesh)	Sample code	RWS size (mesh)	Thermoplastic natural rubber (wt%)				RWS (wt%)
				R60/P40	R50/P50	R40/P60	R30/P70	
R6P4	—	—	—	100	—	—	—	—
R6P4W30L	40	R6P4W30S	80	70	—	—	—	30
R6P4W40L	40	R6P4W40S	80	60	—	—	—	40
R6P4W50L	40	R6P4W50S	80	50	—	—	—	50
R6P4W60L	40	R6P4W60S	80	40	—	—	—	60
R5P5	—	—	—	—	100	—	—	—
R5P5W30L	40	R5P5W30S	80	—	70	—	—	30
R5P5W40L	40	R5P5W40S	80	—	60	—	—	40
R5P5W50L	40	R5P5W50S	80	—	50	—	—	50
R5P5W60L	40	R5P5W60S	80	—	40	—	—	60
R4P6	—	—	—	—	—	100	—	—
R4P6W30L	40	R4P6W30S	80	—	—	70	—	30
R4P6W40L	40	R4P6W40S	80	—	—	60	—	40
R4P6W50L	40	R4P6W50S	80	—	—	50	—	50
R4P6W60L	40	R4P6W60S	80	—	—	40	—	60
R3P7	—	—	—	—	—	—	100	—
R3P7W30L	40	R3P7W30S	80	—	—	—	70	30
R3P7W40L	40	R3P7W40S	80	—	—	—	60	40
R3P7W50L	40	R3P7W50S	80	—	—	—	50	50
R3P7W60L	40	R3P7W60S	80	—	—	—	40	60

Note: NR/HDPE ratios of 60/40 (R60/P40), 50/50 (R50/P50), 40/60 (R40/P60), and 30/70 (R30/P70). RWS: rubberwood sawdust; wt%: percent by weight.

The hardness of the composite samples was measured by using a mechanical Shore D Durometer (Model GS-702G from Teclock Corporation, Nagano, Japan), according to ASTM D2240-91. The dimensions of the specimens tested were approximately 30 mm × 30 mm × 6 mm. All mechanical tests were performed with five replications for each composite formulation at a temperature of 25°C.

2.6.2. Water Absorption Test. The measurement of water absorption was carried out according to ASTM D570-88. Prior to testing, five specimens of each formulation (10 mm × 20 mm × 6 mm) were dried in an oven at 50°C for 24 h. Their weights were then measured with a precision of 0.001 g before being immersed in water at ambient room temperature. After 24 h soaking, all specimens were removed from the water, thoroughly wiped off using tissue papers, and immediately, the weights were measured again. The results would indicate the percentages of water absorption in relation to the dry weight of the specimens.

3. Results and Discussion

3.1. Thermal Degradation Behavior of TPNR and TPNR/RWS Composites. Thermal degradation of the pure NR, pure HDPE, and TPNR was studied using thermogravimetric analysis, as shown in Figure 1 and Table 2. The pure NR displayed the lowest degradation temperature, while degradation of the pure HDPE occurred at the highest temperature. The temperature of the maximum weight loss for the NR occurred at approximately 378.1°C, while the HDPE

degraded at approximately 491.0°C. Therefore, it was clearly exhibited that degradation temperature increased with an increase of plastic contents in the NR/HDPE blends. Table 2 also illustrates the temperature at different weight losses for TPNR. For 40% weight loss, the temperatures of R6P4, R5P5, R4P6, and R3P7 are 393.8°C, 401.4°C, 460.8°C, and 464.5°C, respectively.

Thermogravimetric analysis of TPNR/RWS composites is also needed to evaluate thermal stability for determining their application and end use [20]. Figures 2(a) and 2(b) illustrate the variation of thermogravimetric curves and derivative thermogravimetric (DTG) curves, respectively, of TPNR/RWS composites affected by RWS contents and sizes. The TG curves of TPNR/RWS composites show four mass loss steps. The first step is weight loss due to the evaporation of moisture in the range of around 90 to 190°C. The second and third steps are weight loss owing to the decomposition of wood sawdust and natural rubber in the range of around 290 to 430°C. Likewise, at the third peak, the maximum weight loss rates of the NR/HDPE blend of the R60/P40 ratio reinforced by 30 wt% (case R6P4W30L) and 60 wt% (case R6P4W60L) RWS contents with 40 mesh size occur at 382.3 and 368.8, respectively. Generally, the thermal decomposition of the natural fibers occurs in the temperature range of 194–386°C, due to the degradation of cellulose, hemicelluloses, and lignin compositions [21, 22]. The last weight loss step is the decomposition of HDPE in the range of around 450 to 510°C.

The TG curves (Figure 2(a)) representing the TPNR composites with 60 wt% RWS loading (lines (B) and (D))

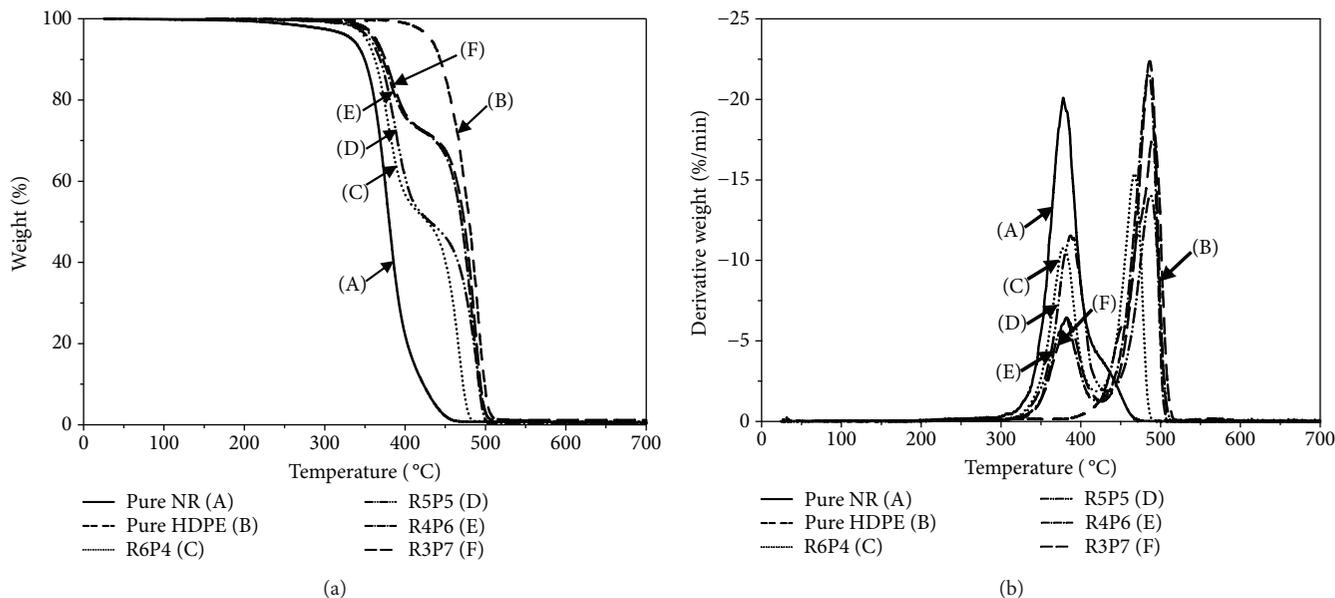


FIGURE 1: Curves of (a) TGA and (b) DTG for pure NR, pure HDPE, and NR/HDPE blends at various blend ratios.

TABLE 2: Thermal data obtained from TGA thermograms of TPNR and TPNR/RWS composites.

Formulation	Temperature at different weight loss (°C)				Temperature for maximum weight loss			
	T_{20}	T_{40}	T_{60}	T_{80}	1st peak	2nd peak	3rd peak	4th peak
Pure NR	361.8	374.9	385.6	402.7	378.1	—	—	—
Pure HDPE	456.9	473.5	485.2	493.9	491.0	—	—	—
R6P4	373.9	393.8	450.4	466.1	379.2	467.1	—	—
R5P5	381.1	401.4	466.0	485.6	387.8	487.3	—	—
R4P6	389.5	460.8	476.5	486.7	382.7	485.1	—	—
R3P7	392.4	464.5	479.7	489.1	382.1	486.3	—	—
R6P4W30L	357.7	383.1	426.8	487.9	159.2	311.3	382.3	491.3
R6P4W60L	335.1	370.6	408.2	494.8	156.5	305.2	368.8	491.9
R4P6W30L	365.5	394.4	475.2	494.5	162.1	300.8	383.7	493.5
R4P6W60L	334.1	371.0	456.3	498.6	162.8	298.4	367.9	492.6
R4P6W30S	362.8	393.1	476.4	495.0	164.4	302.3	376.4	493.9

Note: T_{20} (temperature for 20% weight loss).

show the weight loss 40% at 370.6–371.0°C, while the composites with 30 wt% RWS (lines (A), (C), and (E)) exhibit the weight loss 40% at 383.1–394.4°C. These results can indicate that an increase of RWS content in TPNR composites will decrease the thermal stability. Because the wood had more degradation than the HDPE, thus, the increasing addition of wood into the composites reduced the thermal stability. Likewise, stronger interfacial adhesion between the wood sawdust and the polymer matrix increased the thermal stability [13, 23]; an addition of sawdust content in the polymer composites resulted in poorer interfacial bonding between phases of the materials.

The TG curves also revealed that blend ratios of NR/HDPE affected the thermal stability of the composites. It can be observed that the TPNR composites with 30 wt% RWS loading showed the degradation at 394.4°C with 40% weight loss for the NR/HDPE blend of the R40/P60 ratio

(line (C)) and 49% weight loss at 394.4°C for the NR/HDPE blend of the R60/P40 ratio (line (A)). Normally, the lower percentage of weight loss demonstrates a larger thermal stability of the composite material [23–25]; this is due to more degradation of natural rubber than plastic occurrence. In addition, the RWS sizes slightly affected the thermal stability of TPNR/RWS composites. It can be observed that the TPNR composites based on RWS 80 mesh (line (E)) degraded at 362.8°C with 20% weight loss, while the degradation of the composites based on RWS 40 mesh (line (C)) occurred with 20% weight loss at 365.5°C. This finding is in good agreement with Ratanawilai et al. [13], who found that the particle sizes insignificantly affected the thermal stability of wood-plastic composites. In fact, the chemical constituents (e.g., cellulose, lignin, and hemicelluloses) of wood as well as their contents mainly affect the thermal stability of wood sawdust [13, 23, 25], which the particle size influences lightly.

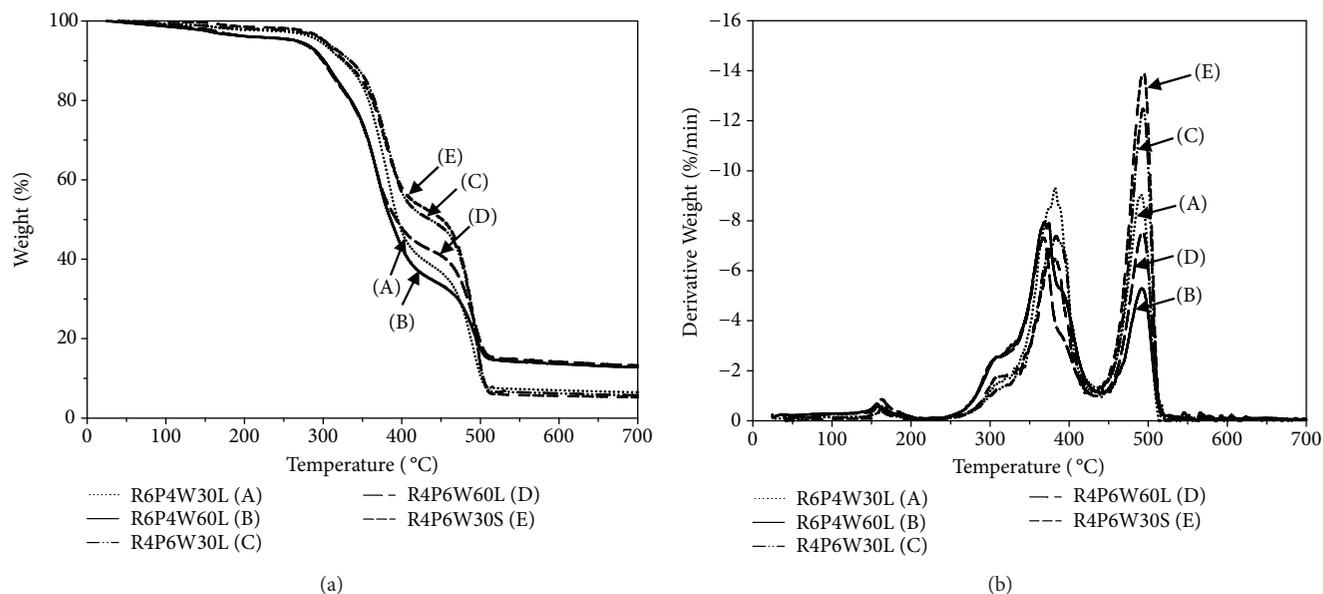


FIGURE 2: Curves of (a) TGA and (b) DTG for TPNR composites containing different RWS contents and sizes.

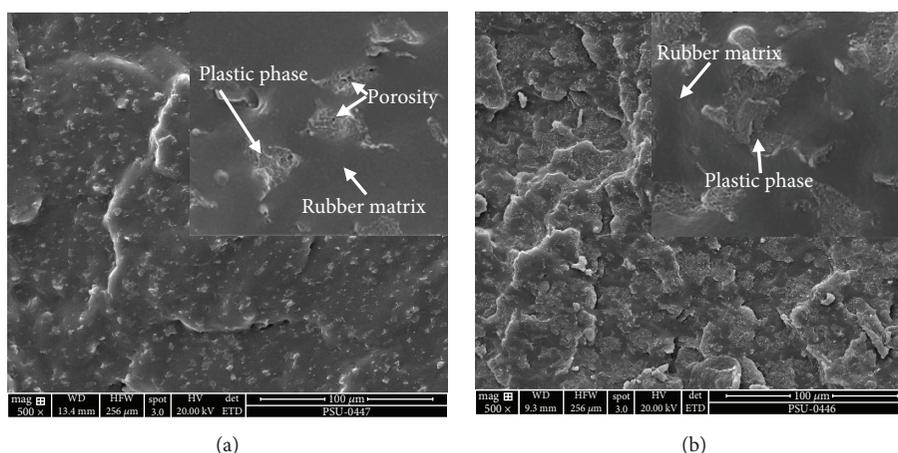


FIGURE 3: SEM images of TPNR blending with NR/HDPE ratios of (a) R60/P40 and (b) R30/P70.

3.2. Morphological Analysis of TPNR/RWS Composites. The morphology of the NR/HDPE blends reveals some interesting realities as shown in Figure 3. In NR/HDPE blends of R60/P40 and R30/P70 ratios, SEM images of both systems showed that plastic could not be melted as a continuous phase in the rubber matrix. Further, more some porosities were also found in the phase of the plastic (Figure 3(a)). In addition, the fractured surfaces of the composites from NR/HDPE blends of R60/P40 and R30/P70 ratios reinforced with RWS 40 mesh and 80 mesh are shown in Figures 4 and 5, respectively. It can be seen that the TPNR/RWS composites with blend ratios of higher rubber content (Figures 4(a), 4(b), 5(a), and 5(b)) show smoother surfaces and fewer voids. Besides, SEM micrographs were also exhibited that the RWS did not completely attach to the polymer matrix due to poor compatibility between the wood and the polymer matrix [26]. Ashori [26] said that plastic and wood did not merge well. Especially, for the TPNR/RWS composites with the

NR/HDPE blend of the R30/P70 ratio, the morphology presented many voids and poor interfacial adhesion, indicating that the interaction between the polymer matrix and wood was weak [2]. Efficacy of interfacial adhesion significantly affected the mechanical and physical properties of wood-polymer composites.

The TPNR composites containing RWS sizes of 40 mesh (Figures 4(c) and 4(d)) showed more porosities and larger gaps between the wood sawdust and the TPNR matrix than the composites with RWS sizes of 80 mesh (Figures 5(c) and 5(d)). This is likely caused by agglomeration and entanglement of bigger wood sawdust [15]. Furthermore, the TPNR composites with 60 wt% RWS (Figures 4(b), 4(d), 5(b), and 5(d)) exhibited more voids than the composites with 30 wt% RWS (Figures 4(a), 4(c), 5(a), and 5(c)) because the addition of RWS content in the polymer composites increased the dispersing difficulty of the wood particles and their tendency to form strong agglomeration [23].

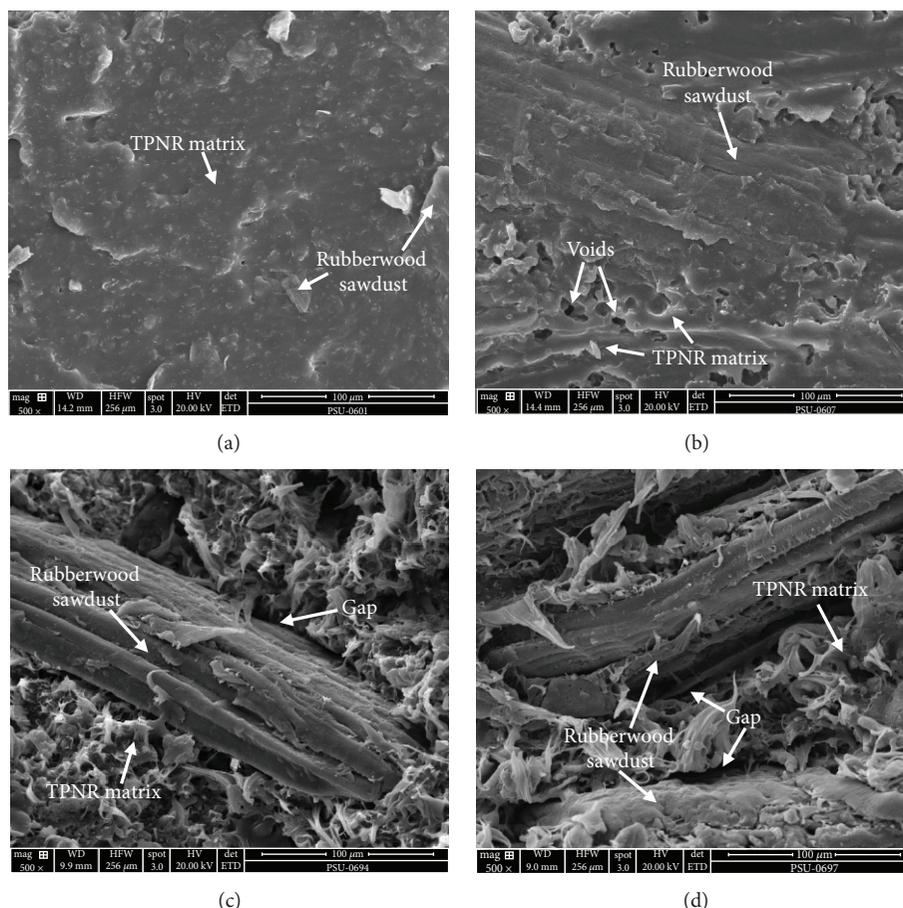


FIGURE 4: SEM images of R60/P40 composites with (a) 30 wt% RWS and (b) 60 wt% RWS and R30/P70 composites with (c) 30 wt% RWS and (d) 60 wt% RWS. All formulations reinforced with RWS 40 mesh.

Particularly for the TPNR/RWS composites with higher plastic content (R30/P70), it was observed that the wood sawdust displayed the shape of irregular short fibers in the composite structure. The composites containing higher wood sawdust content to TPNR blends (Figures 4(d) and 5(d)) seemed to exhibit a higher number of porosities and agglomerations as well as poorer interfacial bonding between the RWS and the polymer matrix.

3.3. Tensile Properties of TPNR/RWS Composites. The effect of RWS contents and sizes on tensile strength and modulus of the composites from NR/HDPE blends of R60/P40, R50/P50, R40/P60, and R30/P70 ratios is shown in Figures 6(a) and 6(b), respectively. The tensile strength of the TPNR/RWS composites with a blend of R30/P70 was clearly reduced with the increasing content of RWS, whereas the additions of RWS in the range of 30 to 50 wt% into the blends of R50/P50 and R60/P40 improved the tensile strength of the composites. These phenomena are caused by the domination of plastic in the blending which is strong; therefore, the incorporation of wood sawdust is less effective [4]. In contrast, for blending with rubber dominant which is softer than plastic, the addition of wood sawdust effectively improves the properties of TPNR composites [4]. These reasons can be confirmed by considering the morphology of TPNR/RWS

composites in Figure 4 (Figures 4(a) and 4(b) for TPNR composites blending with R60/P40 and Figures 4(c) and 4(d) for TPNR composites blending with R30/P70). It is evident that the TPNR/RWS composites with R60/P40 clearly displayed smoother surfaces, lower voids, and stronger coupling between the TPNR and the RWS than the composites blending with R30/P70. Thus, the composites blending with R60/P40 possess better stress transfer from the polymer matrix to the wood sawdust. However, at the same RWS contents and sizes, the TPNR/RWS composites with blends of R30/P70 gave clearly better tensile strength and modulus than the composites with blends of R60/P40. It is rationalized because the HDPE plastic has more strength and stiffness than the natural rubber; thus, the composites with larger plastic concentration exhibited higher strength and modulus.

Figure 6(b) exhibits the increment of the tensile modulus with an increase of RWS contents in all systems of the composites. Because these composites have high stiffness wood sawdust and low stiffness polymer matrix, the increasing addition of wood filler volume into the composites increases the stiffness [22]. Jamil et al. [2] revealed that an increment of modulus causes a decrease in the flexibility or elasticity of the soft matrix. In addition, the TPNR composites based on RWS 80 mesh (solid lines) seem to give higher tensile strength than those based on RWF 40 mesh (dashed lines) for the same

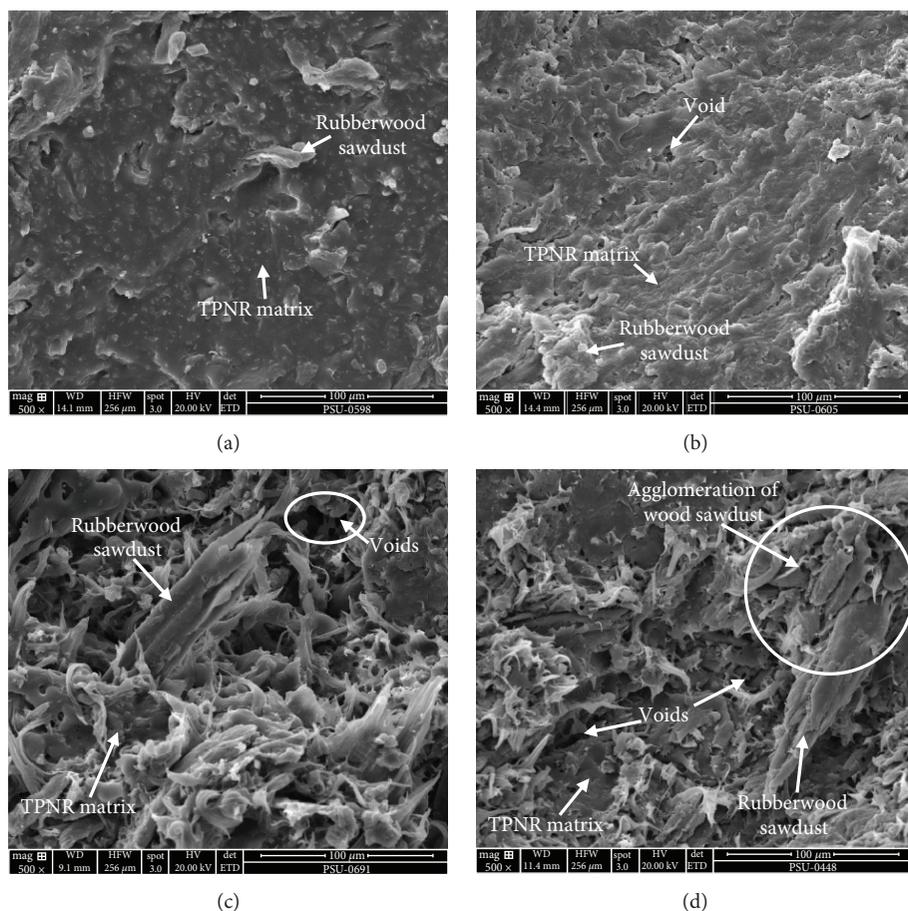


FIGURE 5: SEM images of R60/P40 composites with (a) 30 wt% RWS and (b) 60 wt% RWS and R30/P70 composites with (c) 30 wt% RWS and (d) 60 wt% RWS. All formulations reinforced with RWS 80 mesh.

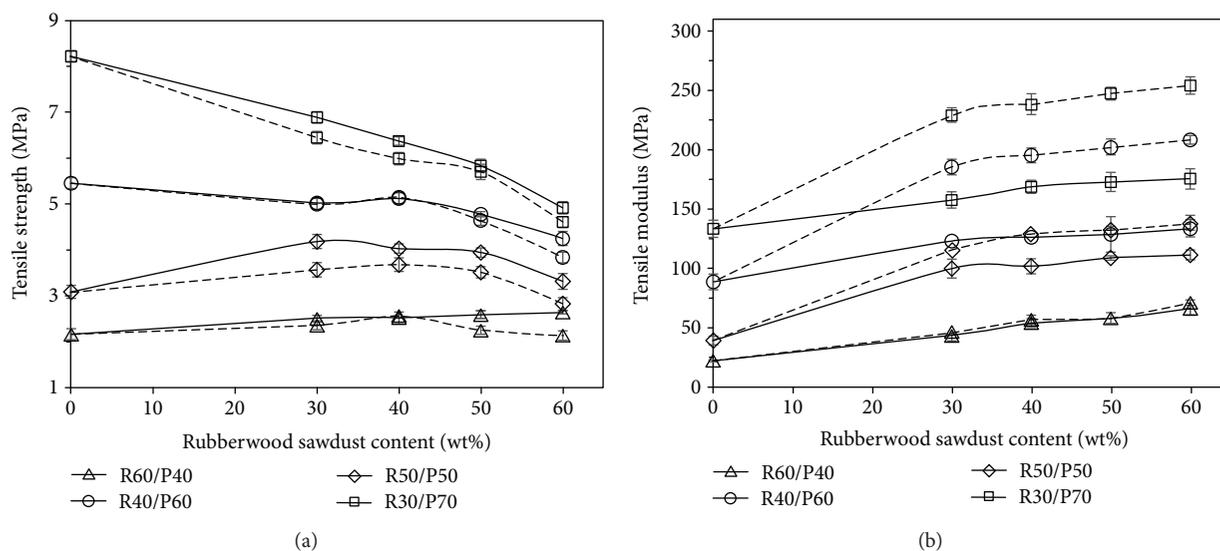


FIGURE 6: Effect of wood sawdust contents and sizes on (a) tensile strength and (b) tensile modulus for TPNR/RWS composites. Dashed lines show RWS 40 mesh, and solid lines represent RWS 80 mesh.

blend ratio of NR/HDPE. Probably, small sawdust size distributes more homogeneous than that of large sawdust size in the polymer matrix, resulting in a uniform stress transfer

from the TPNR matrix to the dispersed wood sawdust [13, 27]. Thus, it could increase the load resistance performing from the tension. However, for the tensile modulus, the

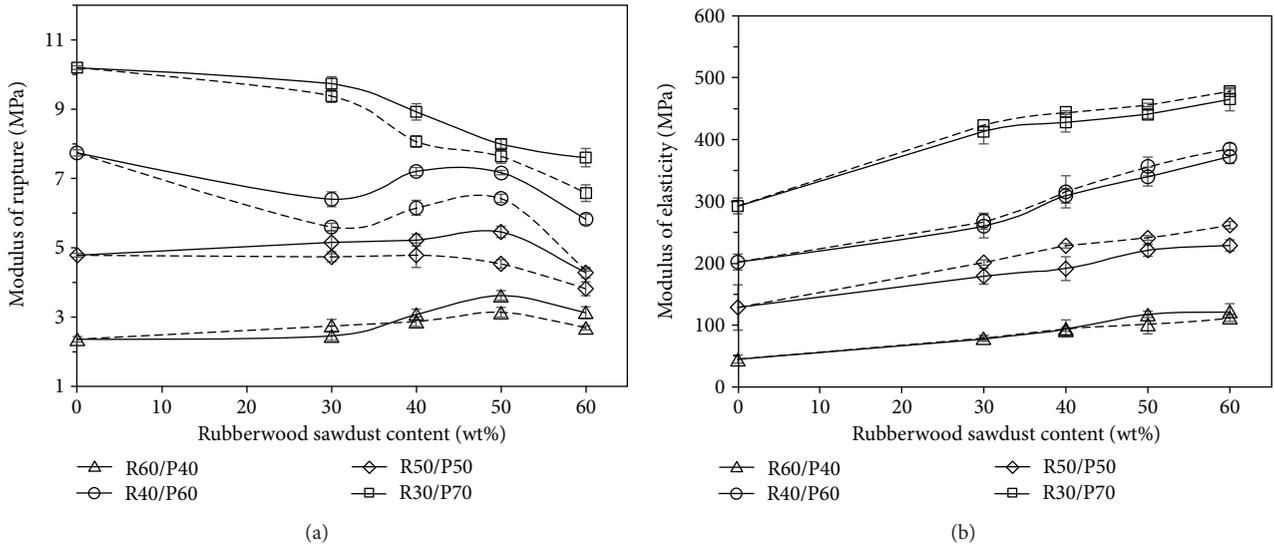


FIGURE 7: Effect of wood sawdust contents and sizes on (a) modulus of rupture and (b) modulus of elasticity for TPNR/RWS composites. Dashed lines show RWS 40 mesh, and solid lines represent RWS 80 mesh.

TPNR composites based on RWS 80 mesh (solid lines) exhibited lower values.

3.4. Flexural Properties of TPNR/RWS Composites. Figures 7(a) and 7(b) show the effect of adding RWS on the modulus of rupture (MOR) and modulus of elasticity (MOE), respectively, of the composites from NR/HDPE blends with R60/P40, R50/P50, R40/P60, and R30/P70 ratios. It can be seen that the MOR of TPNR composites blending with R30/P70 decreased with an increase of RWS content. The optimal RWS loading in the TPNR blending with R60/P40, R50/P50, and R40/P60 is about 50 wt%. The MOR reduced after 50 wt% RWS content indicates that there are small stress transfers from the matrix to the wood due to entanglement and agglomeration of RWS in the polymer matrix. The MOE of TPNR/RWS composites also showed a similar trend to the tensile modulus: the MOE increased greatly with wood sawdust content. Since wood sawdust has higher stiffness or modulus as compared to the polymer matrix, the TPNR composites with higher sawdust content demand higher stress for the same deformation [6, 28].

The MOR of TPNR composites containing RWS 40 mesh (dashed lines) is lower as compared to that of the composites containing RWS 80 mesh (solid lines). This is possible because poorer interfacial adhesion between the wood sawdust and the polymer matrix occurred in the composites with larger wood sawdust [15]. This can be observed in SEM micrographs which show that the composites with RWS 40 mesh (Figures 4(c) and 4(d)) had larger spaces between the wood sawdust and the TPNR matrix than those with RWS 80 mesh (Figures 5(c) and 5(d)). In contrast, the composites with RWS 40 mesh (dashed lines) presented higher MOE than those composites with RWS 80 mesh (solid lines) for the same TPNR to wood sawdust ratio. The composites reinforcing with larger wood sawdust gave a higher fiber aspect ratio. Furthermore, the fiber length-to-diameter ratio determined as the fiber aspect ratio is another factor

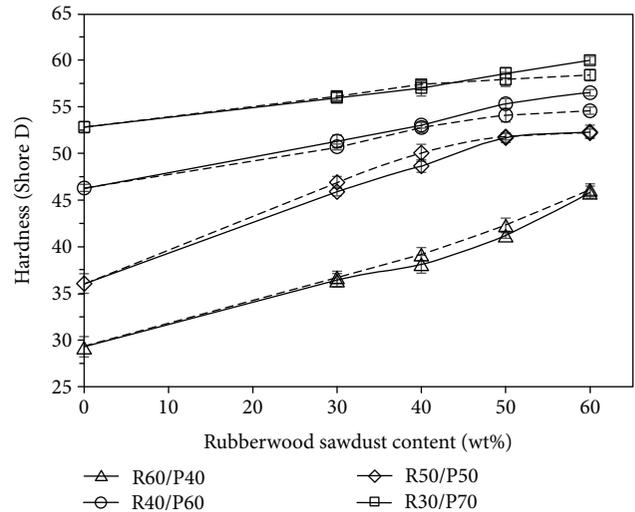


FIGURE 8: Effect of wood sawdust contents and sizes on Shore hardness for TPNR/RWS composites. Dashed lines show RWS 40 mesh, and solid lines represent RWS 80 mesh.

influencing the mechanical properties of the composite material [29]. An increment of the fiber aspect ratio increases the stress transfer from the polymer matrix to the natural fibers, and then the modulus of composites material was improved [29]. In addition, the result obtained indicates that TPNR/RWS composites with a higher HDPE ratio gave higher values of MOR and MOE. It was observed that, for 60 wt% RWS with 40 mesh, the TPNR/RWS composites containing the NR/HDPE blend of the R30/P70 ratio exhibited higher MOR and MOE about 144% and 328%, respectively, as compared to composites containing blends of R60/P40.

3.5. Hardness of TPNR/RWS Composites. The Shore hardness of the NR/HDPE blend and NR/HDPE composites with different RWS contents and sizes is shown in Figure 8. It was

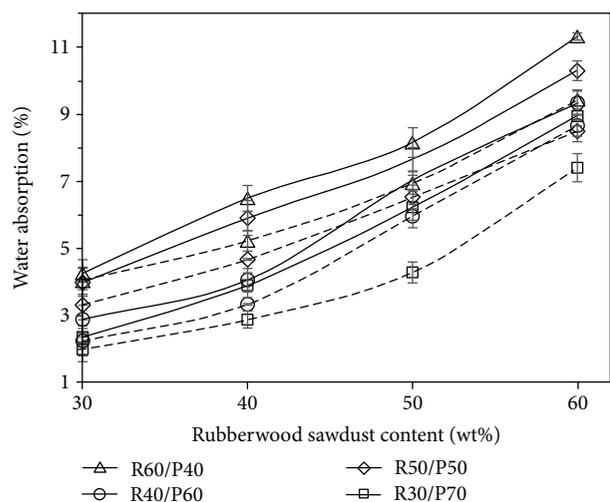


FIGURE 9: Effect of wood sawdust contents and sizes on water absorption for TPNR/RWS composites. Dashed lines show RWS 40 mesh, and solid lines represent RWS 80 mesh.

found that the Shore hardness of NR/HDPE blends increased with an increase of HDPE contents; the NR/HDPE blend of R30/P70 ratio has Shore hardness higher about 80% than the blend ratios of R60/P40. This is because the Shore hardness of natural rubber is low (5 Shore D) as compared to pure HDPE (65 Shore D), leading to higher flexibility TPNR. Moreover, it can be seen that the Shore hardness of the composites progressively increased as wood sawdust content increases in the TPNR/RWS composites. The reason is the wood sawdust has a significantly higher hardness than the thermoplastic elastomer matrix, and the addition of wood particles into TPNR phases decreases the flexibility or elasticity of polymer chains, resulting in more rigid composites [2]. Jamil et al. [2] reported that the presence of more natural rubber in the thermoplastic elastomer results in less Shore hardness.

For NR/HDPE blends of R60/P40 and R50/P50 ratios, the addition of RWS 40 mesh (dashed lines) into the TPNR composites seems to result in higher Shore hardness compared to the composites with RWS 80 mesh (solid lines); however, the TPNR composites reinforcing with RWS 40 mesh gave lower Shore hardness than those composites with RWS 80 mesh for the blend ratio of R40/P60 and R30/P70. This could be attributed to more homogeneous dispersion of the small particle size in the polymer matrix, which results in minimum voids [13]. Likewise, a stronger interfacial bonding between the wood filler and the polymer matrix can improve the Shore hardness of the composites.

3.6. Water Absorption of TPNR/RWS Composites. The water absorption of the TPNR/RWS composites was monitored by full water immersion for 24 hours as shown in Figure 9. Normally, the water absorption of the wood-polymer composites increased with wood sawdust content. It was found in the current work that as the amount of rubberwood sawdust increased, the water absorption of TPNR/RWS composites steadily increased due to the hydrophilic nature of wood

sawdust, while the nature of the polymer is hydrophobic [30]. An increment of wood sawdust content increases the free OH groups in the composites, in which the formation of the free OH groups interacts with the H groups from water [30], resulting in the weight gain of the wood-polymer composites [31]. Additionally, an increase of wood sawdust content in the composites resulted more voids and poorer interfacial adhesion between the wood and the polymer matrix and consequently higher water absorption. Wan Busu et al. [30] claimed that the good interfacial bonding between wood particles and polymer can prevent the water molecules infiltrating into the composite system.

The water absorption of TPNR/RWS composites with particle sizes of 80 mesh (solid lines) is higher than that of 40 mesh (dashed lines), which is likely due to harder encapsulation of RWS 80 mesh into the TPNR matrix, which leads to pores in the composite structure. Likewise, the finer wood flour has a much larger surface area per weight [32], and therefore, it has more areas to contact with water molecules. Furthermore, at the same RWS contents and sizes, the TPNR composites with blend ratios of higher plastic content yielded lower water absorption. For example, with a RWS size of 40 mesh, the NR/HDPE blend of the R60/P40 ratio reinforced with 40 wt% RWS showed higher water absorption about 81% than that of the R30/P70 ratio with 40 wt% RWS. This higher water absorption is attributed to an increment of NR content in the plastic which results in larger holes and increases voids in the structure of the TPNR [33]. Therefore, the TPNR with higher rubber content allows easier penetration of water into the composite structure as well as an increase of water residence sites, resulting in larger water absorption.

4. Conclusions

The study has revealed that the RWS size and content as well as the ratio of NR/HDPE blends significantly affect the thermal, physical, and mechanical properties of the composites. The addition of RWS about 30–50 wt% improved the tensile strength and MOR of the composites from the NR/HDPE blend of R60/P40 and R50/P50 ratios, while the modulus and Shore hardness increased with an addition of RWS in the TPNR composites due to a stiffer wood sawdust than the polymer matrix. However, the incorporation of RWS into the TPNR composites negatively affected the thermal and physical properties. An increase of RWS content in TPNR composites decreased the thermal stability and increased the water absorption. In addition, TPNR composites with RWS 80 mesh gave better tensile strength and MOR than the composites with RWS 40 mesh due to less gaps between the wood sawdust and the TPNR matrix. The TPNR/RWS composites with larger plastic content yielded higher thermal stability and tensile, flexural, and Shore hardness properties but lower water absorption. SEM micrographs of the sample also revealed that the microgap formation was reduced by the addition of RWS 80 mesh in the TPNR composites. The TPNR/RWS composites from the NR/HDPE blend of R30/P70 ratio were therefore suggested for applications requiring high strength, modulus, hardness, and low water absorption.

The TPNR composites with a large RWS size is appropriate for applying in conditions of modulus and water resistances. The overall result of the composition will greatly facilitate the development of building materials from thermoplastic elastomer composites.

Data Availability

The data used to support the findings of this study are available from the corresponding author upon request.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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