

Characterization of Rice Husk-Incorporated Recycled Thermoplastic Blend Composites

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High-fiber-content composites made from rice husk (RH) (from 50 up to 80 wt%) as well as a recycled thermoplastic blend (rTPB) were fabricated using a two-step extrusion and hot/cold press molding technique. The temperature dependency of the thermal degradation and dynamic-mechanical behavior was investigated using thermogravimetric analysis (TGA) and dynamic mechanical analysis (DMA). The long-term water absorption and orthotropic swelling were analyzed following immersion in distilled, tap, and sea water for 15 weeks. Improvements in the thermal stability, storage, and loss modulus, as well as reductions in dimensional stability, were observed as the alkali content of the RH in the rTPB composites was increased. The composites immersed in sea water showed the lowest water absorption, followed by those in distilled and then tap water. The thickness dimension of the composite specimens exhibited the highest swelling values, followed by width and then length dimensions. The tensile strength and elastic modulus showed the maximum values at 70 wt% RH (21.2 MPa and 1.6 GPa, respectively). The surface morphology, interfacial adherence, and bonding between the matrix-fiber phases in the composites were characterized using a scanning electron microscope (SEM).

Keywords: Biocomposites; Agriculture waste; High fiber loading; Recycled plastic; Extrusion; Melt blending

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INTRODUCTION

The increasing awareness of and demand for greener and more sustainable materials, the soaring prices of petroleum-based plastics (Robertson *et al.* 2013; Arjmandi *et al.* 2015), and environmental friendliness regulations regarding recycling issues have attracted the attention of researchers in the field of composites worldwide. Because rice husk and straw are major waste products worldwide, the utilization of agricultural waste is of prime importance, second only to the utilization of industrial wastes. Globally, the annual total production of paddy rice is approximately 600 million tons, and on average, 20% of this, or approximately 120 million tons, is husk (Louis and Thomas 2013). The incineration of RH is strongly discouraged, as it could generate undesired byproducts such as ash, fumes, and toxic gases, causing serious air pollution. Therefore, a number of researchers have studied and reported on various applications that involve RH (Arjmandi *et al.* 2015).

RH has been introduced into a wide variety of thermoplastic materials to produce high-performance natural fiber-reinforced composites. Kim *et al.* (2006) reported that treatment by a compatibilizing agent (maleic anhydride-grafted polypropylene (MAPP) and maleic anhydride-grafted polyethylene (MAPE)) improved the interfacial bonding and compatibility between the RH (fixed at 30 wt%) and the matrix polymer. In their investigation, polypropylene (PP) and linear density polyethylene (LDPE) were chosen because of their high melting temperature (T_m), superior thermal and mechanical properties, and low density. Zhang *et al.* (2009) investigated the thermal stability and flame retardancy

of RH-filled high-density polyethylene (HDPE) eco-composites with 20, 50, and 70 wt% RH. The main mechanisms of the flame retardant behavior imparted by the RH in the HDPE eco-composite are ascribed to the heat shielding and diffusion barrier effects induced by the silica-based protective layer (due to the high silica content of the RH) that was formed during the combustion process. Rahman *et al.* (2010) reported that 35 wt% RH filler loading-reinforced PE composites exhibited the optimum mechanical properties when compared with those with 25, 30, and 40 wt% RH; additionally, the alkali (pH 10.5) media-treated RH composites yielded the lowest water absorption in comparison with the other composites, RH treated at acidic (pH 6) and neutral media (pH 7).

The different composite preparation methods, namely internal mixer, single-screw extruder, counter or co-rotating twin-screw extruder, compression molding, and injection molding, have been observed to result in composite materials with different physical, mechanical, thermal, and morphological behaviors. Aside from the processability of natural fiber-incorporated thermoplastics, the enhancement of the interfacial interaction between the matrix and the fiber is another important issue that could be an added benefit for the natural fiber composite (NFC) industry (Arjmandi *et al.* 2015). The application of NFC in the automotive, building and construction, marine, and consumer goods industries requires that it withstand exposure to high-moisture environments (Shinoj *et al.* 2010). As established, natural fiber is inherently highly polar and hydrophilic, which causes the fiber to bond more easily with water molecules, leading to an increase in water absorption. To address this, chemical treatment and the addition of a coupling agent are commonly used to reduce the water absorption in a composite (Chen *et al.* 2015b). However, better understanding of the water absorption and swelling behavior of the NFC in different media would provide information relevant to its application in industries where it would be exposed to different environments.

A number of studies have been reported on RH composites based on a single type of polyolefins (Kim *et al.* 2006; Zhao *et al.* 2009; Yao *et al.* 2011); however, there has been limited research investigating RH composites based on two or more thermoplastics matrices, namely, a polymer blend. A recycled thermoplastic blend (rTPB) made from recycled HDPE and recycled polyethylene terephthalate in the ratio of 75/25 (wt/wt) was used as a polymer matrix in this current study. The blend that was chosen for this work was the blend that had demonstrated the optimum mechanical properties when compared against several blends with different composition and compatibilizer contents in the authors' previous work (Chen *et al.* 2014b). In the present work, high fiber-content eco-friendly composites made from plastic wastes (recycled thermoplastic blends) and agricultural wastes (rice husk) were prepared. The main objective was the evaluation of thermal stability, water absorption, orthotropic swelling, and the morphological and tensile performances of the differently composed rice husk composites.

EXPERIMENTAL

Materials

Recycled high-density polyethylene (rHDPE) with a density of 923 kg/m³ and a melt flow index of 0.72 g/10 min (190 °C, 2.16 kg) and recycled polyethylene terephthalate (rPET) with an intrinsic viscosity of 0.68 dL/g were supplied from a local plastic recycling plant, BioComposites Extrusion Sdn. Bhd. (Malaysia). An ethylene-glycidyl methacrylate (E-GMA, a trade name of Lotader AX8840) with a melt index of 5 g/10 min (190 °C, 2.16 kg) and 8% glycidyl methacrylate content was used as a compatibilizer for the rHDPE/rPET blend. The agro waste filler material used was rice husk (RH) with a particle size of 100-mesh. Maleic anhydride polyethylene (MAPE) with a melt index of 5 g/10 min (190 °C,

2.16 kg) and a maleic anhydride content of 1 wt% was used as a coupling agent for the rice husk/polymer blend composite. All raw materials were obtained from BioComposites Extrusion Sdn. Bhd. (Malaysia).

Prior to compounding, RH flour was dried in an oven for 24 h at 100 °C. The raw RH was immersed in a 4% sodium hydroxide solution (w/v) at room temperature for 24 h. The RH was then washed with water several times to eliminate excessive sodium hydroxide and dissolved impurities. After rinsing, the RH flour was oven-dried at 90 °C for 24 h to eliminate the trapped moisture.

Methods

Preparation of RH composites

The melt-blending of the RH-filled recycled thermoplastic blend was performed using a laboratory-scale co-rotating twin-screw extruder (Thermo Prism TSE 16 PC, Thermo Electron Corporation, Germany) with a diameter of 16 mm and a length/diameter ratio of 25. During the first extrusion, a recycled thermoplastic blend of rHDPE and rPET with a weight ratio of 75/25 (wt%) and 5% EGMA (based on the total weight of rHDPE/rPET blend) were extruded at a screw rotating speed of 30 rpm. The barrel temperature profiles were set at 190, 240, 270, and 250 °C, from the feeding to die zones, respectively. The extrudates were cooled and then granulated into pellets with a crusher.

During the second extrusion, the pre-extruded recycled thermoplastic blend (rTPB) granulates and the dried RH filler were melt-compounded thoroughly, in the presence of MAPE, using the same twin-screw extruder. The extrusion temperature profiles of 195, 210, 215, and 170 °C and the screw rotating speed of 30 rpm were used. The RH filler contents were varied at 50, 60, 70, and 80 wt%, whereas the content level of MAPE was fixed at 3% based on the total weight of composites in this study. The extrudates were then pelletized after being cooled at room temperature.

A hot- and cold-pressing process (compression molding machine model LP50, Labtech Engineering Company Ltd., Thailand) was used to produce the composite panels for testing. The temperatures of the upper and lower plates were set at 200 °C, and a pressure of 6.9 MPa was applied. The period of preheating, venting, full pressing, and cold pressing were set at 3, 2, 5, and 5 min, respectively. The molded panels were stored and stabilized at ambient conditions for 40 h before testing.

Characterization Techniques

Thermogravimetric analysis (TGA)

The thermal degradation and stability of the specimens was examined using a thermogravimetric analyzer (Mettler Toledo TGA/SDTA851e, United States). The specimens, approximately 10 to 15 mg, were heated from 25 to 600 °C at a heating rate of 10 °C/min under atmospheric air flow conditions.

Dynamic mechanical analysis (DMA)

The storage and loss modulus values of the test samples were evaluated using a dynamic mechanical analyzer (TA Instruments, Inc., New Castle, Delaware) in dual cantilever mode. Measurements were carried out in the linear viscoelasticity region at a frequency of 1 Hz and strain amplitude of 40 μm , with a heating rate of 5 °C/min, from 25 to 150 °C.

Water uptake and orthotropic swelling behaviors

Three specimens were cut to the dimensions of 76.2 x 25.4 x 3.2 (length x width x thickness) mm^3 following the ASTM D570-98 (2010). Before they were submerged into water, the specimens were oven-dried at 90 °C for 24 h to get rid of the trapped moisture.

The measured weight and dimensions of the oven-dried specimens were recorded. All specimens were submerged in three types of media: distilled, tap, and sea water, respectively. At the end of the immersion periods (periodically for 105 days), the specimens were removed from the water, their surface water wiped with tissue paper, and weighed. The water uptake (WA) and orthotropic swelling (OS) of the composites were computed using the following formulas,

$$WA (\%) = (W_t - W_d) / W_d \times 100 \quad (1)$$

$$OS (\%) = (D_t - D_d) / D_d \times 100 \quad (2)$$

where W_d and D_t are the weight and dimension of the oven-dried specimen and W_t and D_t are the weight and dimension, respectively, of the wet specimen after water immersion at time t .

Scanning electron microscopy (SEM)

The surface morphology of each specimen was examined using scanning electron microscopy (VPSEM LEO 1450VP, Carl Zeiss Inc., Germany) with an accelerating voltage at 10 kV. The fracture surfaces of the specimens were sputter-coated with gold using a sputter coater (Q150R, Quorum Technologies Ltd., London) before being subjected to SEM examination at a magnification of 150 times.

Mechanical property measurements

Prior to tensile testing, compression-molded specimens were cut according to ASTM D638-03 (type I) specifications (2003). A universal testing machine (Model Testometric M350-10CT, Testometric materials testing machines, UK) set at a crosshead speed of 5 mm/min and operated at room temperature was used for the tensile measurement. The average result of five replicates was reported for each formulation.

RESULTS AND DISCUSSION

Thermal Degradation and Stability

The TGA and derivative thermogravimetry (DTG) curves of the investigated composites are depicted in Fig. 1(a) and (b), respectively. The neat rTPB without the presence of the RH underwent dramatic weight reduction in a single-stage degradation process in the temperature range of 374 to 508 °C, whereas there were three stages of weight reduction for the RH; the first stage of weight reduction occurred in the range of 37 to 151 °C, which corresponded to the evaporation of approximately 7% of the moisture, and the second and third stages of weight reduction happened at 175 to 370 °C and 375 to 486 °C, respectively, because of the decomposition of hemicellulose, cellulose, and lignin components (Zhao *et al.* 2009). As a combination of thermal degradation behaviors, the rTPB/RH composites experienced a three-step degradation process similar to that of RH, which occurred at 140 to 145 °C, 200 to 350 °C, and 355 to 500 °C, respectively (Fig. 1). Because RH was introduced into composites as a filler, its composition did affect the thermal degradation characteristics of polymer matrix. Therefore, only the weight reduction stage involving the degradation of polymer matrix (third stage for rTPB/ RH composites) is described in Table 1. Three values of temperatures were recorded: (1) T_o , is determined at the temperature where the weight reduction started, (2) T_{max} , is determined at the temperature where the highest weight reduction happened, and (3) T_e , is determined at the temperature where the weight reduction ended. For neat rTPB without the RH filler, the initial decomposition started at $T_o = 374.8$ °C, the maximum decomposition rate (T_{max})

occurred at 467.1 °C, and the final decomposition ended at $T_e = 508.4$ °C. As the RH content increased in the composite materials, T_o and T_m increased up to 46.5 °C and 14.3 °C, respectively, in contrast to the neat rTPB. These observations indicated that the addition of the RH filler had improved the thermostability of the rTPB matrix, as reported in the authors' previous study on an untreated RH/polymer blend (Chen *et al.* 2014a). Above T_e , it was found that the remaining residual content gradually increased with the RH content because of the high silica content in the RH, which formed silica ash upon heating.

The activation energies of the composites were evaluated for the major decomposition only of the polymer matrix component using the Coates and Redfern method (Alwani *et al.* 2013; Mahmoud and Makled 2012),

$$\ln \left[\frac{-\ln(1-\alpha)}{T^2} \right] = -\frac{E_a}{R} \frac{1}{T} + \ln \frac{AR}{\beta E_a} \quad (3)$$

$$\alpha = \frac{m_i - m_a}{m_i - m_f} \quad (4)$$

where α is the decomposition fraction, E_a is the activation energy (kJ mol^{-1}), R is the gas constant ($8.314 \text{ JK}^{-1}\text{mol}^{-1}$), T is the absolute temperature in Kelvin (K), A is the pre-exponential factor (min^{-1}), and β is the heating rate (10 °C/min in this study). The E_a values were calculated from the slope ($-E_a/R$) of the linear plot of $\ln[-\ln(1-\alpha)/T^2]$ against $1/T$. As can be seen in Table 1, all the composites containing RH had much lower E_a than that of neat rTPB. The lower values of E_a indicate the lesser energy required for the degradation process. This could be related to the composition of the composites, the thermal behavior of individual components, and the chemical bonding of the composites. As is well known, RH is a type of natural fiber that possesses a low degradation temperature (John and Thomas 2008), and the incorporation of RH as a filler at high contents (more than 50 wt%) into an rTPB matrix affected the degradation process of resultant composites. Therefore, low values of E_a were obtained for the RH composites. This result is in agreement with Albano *et al.* (1999) who reported a sudden reduction in E_a was observed when the sisal fiber was mixed with PP and PP/HDPE blend.

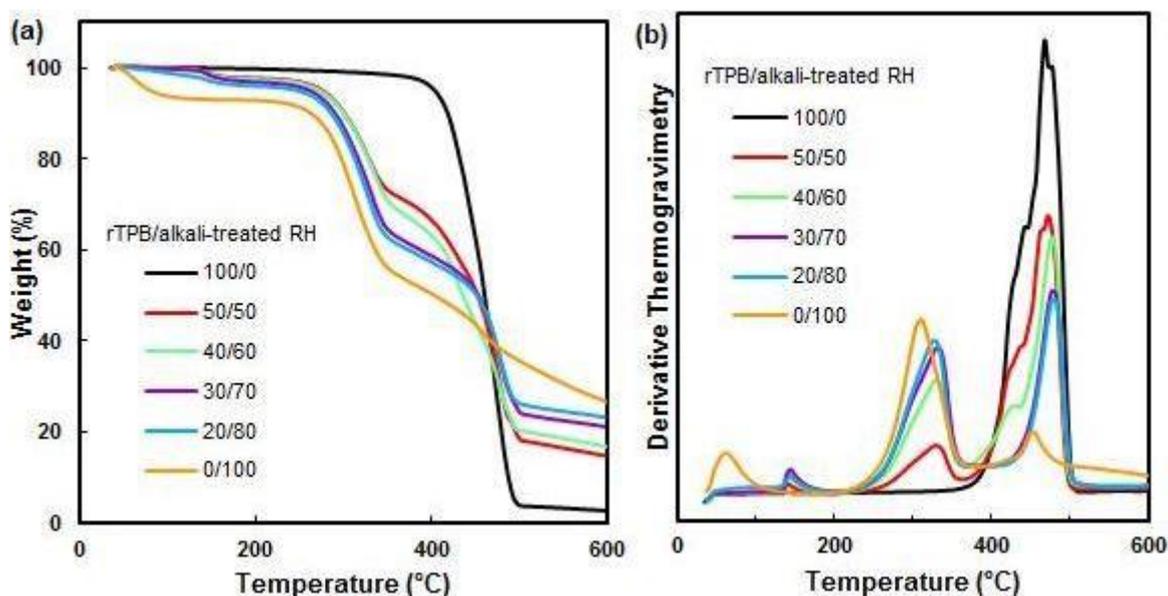


Fig. 1. (a) TGA and (b) derivative thermogravimetry (DTG) curves of rTPB/RH composites with different compositions

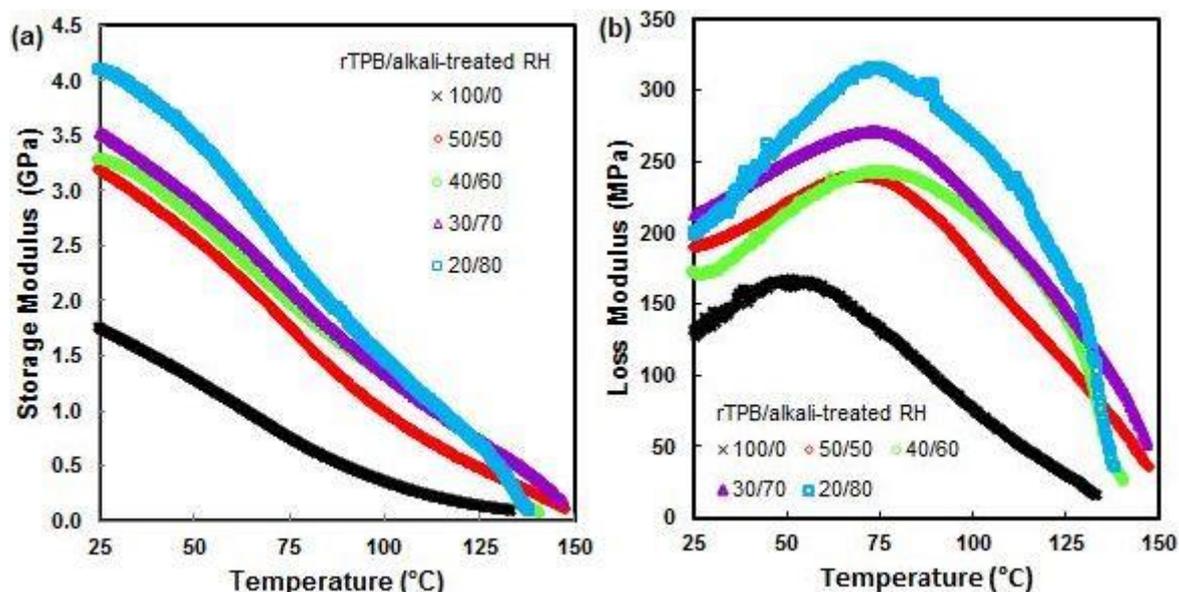
Table 1. Effect of RH Incorporation and Its Composition on Thermal Degradation Characteristics of rTPB Matrix

rTPB/RH	Polymer Decomposition Temperature (°C)			Residues after T_e (%)	Activation energy, E_a (kJ/mol)
	T_o	T_{max}	T_e		
100/0	374.8	467.1	508.4	3.74	165.99
50/50	375.9	476.7	498.6	19.49	61.20
40/60	376.6	478.4	497.3	20.49	83.21
30/70	421.2	478.8	496.7	25.66	84.95
20/80	421.3	481.4	496.8	26.72	82.39

T_o denotes the onset temperature; T_{max} denotes the maximum decomposition rate of the polymer; T_e denotes the endset temperature.

Storage and Loss Modulus

Figure 2 shows the temperature dependency of the storage and loss modulus values of the rTPB/RH with different compositions. Figure 2 (a) presents a general trend of the rise of storage modulus (E') with the increase in RH fiber content in the composites at any particular temperature. This result is expected, as E' is closely related to the elastic and stiffness characteristic of a material, such that the reinforcement imparted by the incorporated RH allowed greater stress distribution at the interface between the rTPB and RH filler (Kim *et al.* 2005). With the increase in temperature, the E' values of the rTPB and the composites decreased remarkably, and subsequently converged to a narrow range at high temperatures. This reduction was because of the increased movement of the polymer chain in the matrix at higher temperatures, which softened the matrix and initiated the relaxation process (Kim *et al.* 2006; Singh and Mohanty 2007).

**Fig. 2.** Temperature dependency of (a) storage and (b) loss modulus with varying loading levels of RH in rTPB matrix

The loss modulus (E'') corresponds to the viscous modulus of viscoelastic material and provides relaxation transition information about the composite materials (Yao *et al.* 2011). In Fig. 2 (b), the loss modulus (E'') of the composites is seen to increase with the increase in fiber loading and to generate a relaxation transition peak at approximately 49.5 °C and 64.6 to 71.2 °C for neat rTPB and rTPB/RH composites, respectively. This peak could be attributed to the alpha (α)-relaxation transition of the resultant thermoplastic blends, which Yao *et al.* (2011) reported for HDPE to be at 47 °C, while Konieczna *et al.*

(2010) reported for PET to be at 105 °C. It is established that α -relaxation is related to the complex multi-relaxation process, which is primarily related to the molecular mobility of the crystalline region of the polymer. The higher α -relaxation temperatures for the rTPB incorporated with RH filler were attributed to the restriction effect of the segmental motion of the polymer molecules at the earlier relaxation temperature (Yao *et al.* 2011).

Water Uptake

The experimental results for water uptake percentage as a function of immersion time for rTPB/RH composites with (a) 50/50 and (b) 40/60 (wt%) compositions in three different mediums are depicted in Fig. 3. The relationship of water content with time appeared to conform to Fickian diffusion behavior, in that the water uptake increased rapidly during the initial stage and then gradually reached an equilibrium state (Chen *et al.* 2015a).

Table 2 lists the water uptake values of the rTPB/RH composites at 2 h, 24 h, and at maximum water uptake (M_m). Generally, the percentages of water uptake seemed to increase with the increase in RH content. This result was as expected given the presence of the free hydroxyl (OH) groups in the RH fibers, which contributed a strong hydrophilic character to the fibers, allowing their combination with water molecules via hydrogen bonding and thus resulting in increased water uptake for the composites (Shinoj *et al.* 2010). In contrast, the rTPB matrix was confirmed for retaining hydrophobic nature as a result of its negligible water uptake values.

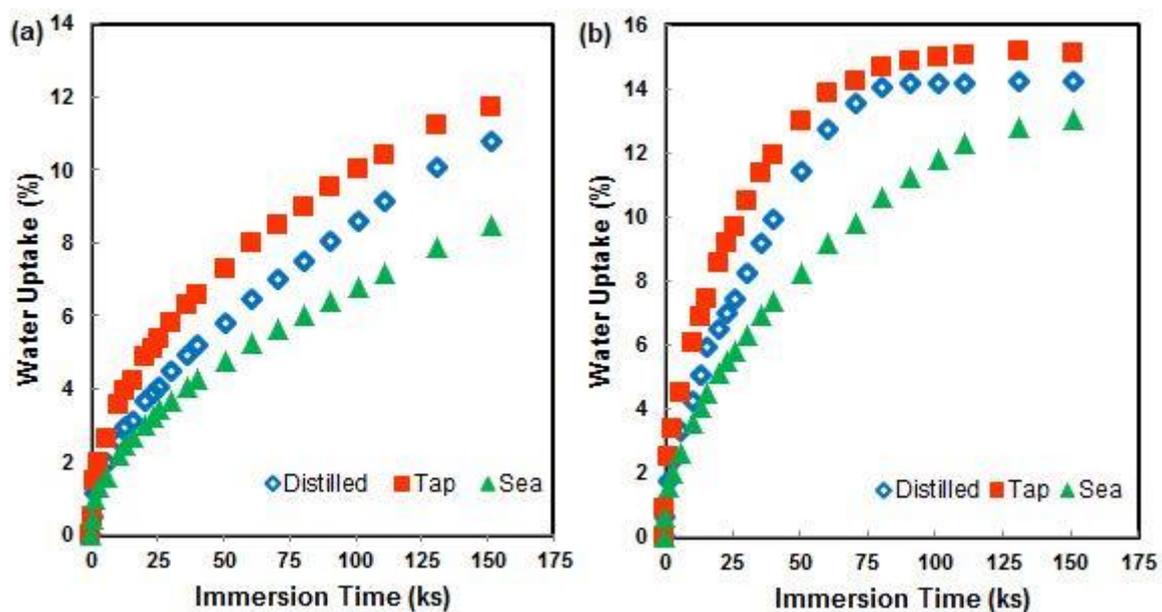


Fig. 3. The percentages of water uptake for rTPB/RH composites with (a) 50/50 and (b) 40/60 (wt%) compositions

To obtain a better understanding of the behavior of the composite specimens in applications of varying environmental conditions, a comparison of the water media (distilled, tap, and sea water were chosen for investigation) for each submerged specimen was considered. Distilled water was used as the control medium, and tap water was used because it was closer to the natural service condition than was distilled water. The water uptake by the composite was reported to be more significant in tap water than in sea (salt) water (Tamrakar and Lopez-Anido 2011). It was concluded that rTPB/RH composites exhibited water uptake in the following descending order: tap water > distilled water > sea water. The lowest water uptake, exhibited by the specimens submerged in sea water, was attributed to the presence of the large-size ionic salt molecules such as sodium chloride that

blocked the pathway of the diffusion process (Akil *et al.* 2009) by reducing the activity of the water molecules into the composites and thus inhibiting the water uptake. Furthermore, the presence of salt ions would reduce the osmotic pressure effects and thereby resulting in less swelling (Nair *et al.* 2016). The highest water uptake, obtained for the specimens submerged in the tap water, and the reason on this result needs further investigation. By comparing Fig. 3 (a) and (b), it can be observed that the water uptake of the composites converged towards a narrow range at higher filler content (60 wt%). Further additions of RH fibers into the composites led to an indefinite trend.

Table 2. Water Uptake of rTPB/RH Composites

rTPB/ RH	Water Uptake (%)								
	Distilled			Tap			Sea		
	2 h	24 h	M_m	2 h	24 h	M_m	2 h	24 h	M_m
100/0	0	0.02	0.14	0.02	0.05	0.18	0	0.02	0.13
50/50	0.49	1.19	10.77	0.51	1.50	11.74	0.45	1.02	8.48
40/60	0.66	1.79	14.23	0.87	2.50	15.13	0.61	1.57	13.07
30/70	1.08	3.67	18.47	1.33	4.05	19.24	1.30	4.07	19.02
20/80	14.04	37.76	50.45	13.37	36.14	47.73	13.22	32.16	52.88

M_m denotes maximum water uptake

Orthotropic Swelling

Figure 4 presents the orthotropic swelling for rTPB/RH composites with (a) 50/50 and (b) 40/60 (wt%) compositions.

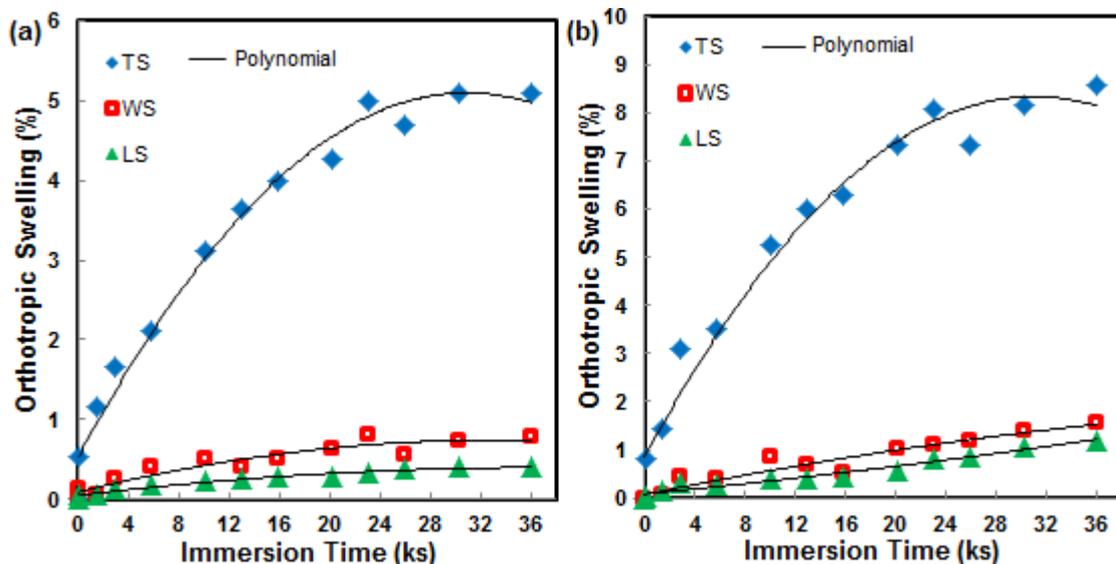


Fig. 4. The percentages of orthotropic swelling for rTPB/RH composites with (a) 50/50 and (b) 40/60 (wt%) compositions are presented. TS denotes thickness swelling; WS denotes width swelling; LS denotes length swelling.

It can be observed that the thickness dimension of the composites exhibited the highest swelling percentage, followed by the dimensions of width and then length. This result indicated the orthotropic nature of the dimensional instability of natural fiber-reinforced composites. During hot press compression, most of the natural fibers aligned in the direction of matrix flow, that is, parallel to the length dimension (Ho *et al.* 2012). The lignocellulosic fibers were normally swelled in the diameter direction, that is, perpendicular to the length direction. Somehow, a minority of the fibers remained randomly-oriented, which contributed to the comparatively lower width and length swelling (Bajwa *et al.*

2011). In general, the experimental results for the orthotropic swelling represented by symbols were found to fit closely with the polynomial curve. The relationships of thickness, width, and length swelling with the respective immersion times were established as follows:

For rTPB/RH at 50/50 (wt%):

$$\text{Thickness swelling (TS)} = -5 \times 10^{-9} t^2 + 0.0003 t + 0.5241, R^2 = 0.9831$$

$$\text{Width swelling (WS)} = -6 \times 10^{-10} t^2 + 4 \times 10^{-5} t + 0.0932, R^2 = 0.8951$$

$$\text{Length swelling (LS)} = -2 \times 10^{-10} t^2 + 2 \times 10^{-5} t + 0.0530, R^2 = 0.9556$$

For rTPB/RH at 40/60 (wt%):

$$\text{Thickness swelling (TS)} = -8 \times 10^{-9} t^2 + 0.0005 t + 0.8592, R^2 = 0.9744$$

$$\text{Width swelling (WS)} = -3 \times 10^{-10} t^2 + 5 \times 10^{-5} t + 0.0711, R^2 = 0.9290$$

$$\text{Length swelling (LS)} = 2 \times 10^{-10} t^2 + 3 \times 10^{-5} t + 0.0830, R^2 = 0.9624$$

Scanning Electron Microscopy (SEM)

Figure 5 illustrates the surface morphologies of the rTPB/RH composites with (a) 50/50, (b) 40/60, (c) 30/70, and (d) 20/80 (wt%) compositions. The red arrows in the figure represent the RH fillers. As the RH content increased from 50 to 80 wt%, it can be observed that the polymer components were getting lesser. The composite with rTPB/RH at 20/80 wt% exhibited comparatively loose morphological structure with the appearance of clear gaps, which these observations were probably related to the insufficient interaction between larger amount of RH fillers and lesser amount (one fifth) of polymer resin.

In the aspect of filler dispersion and distribution within the polymer matrix, the morphology of composites filled with 80 wt.% RH showed no significant differences with the composites containing 50 wt.% RH (Fig. 5). This observation indicated that the natural fibers reinforced composites can be manufactured in high filler loading up to 80 wt.%, which can give an advantage to the composite industry in the economic and environmental friendly aspect.

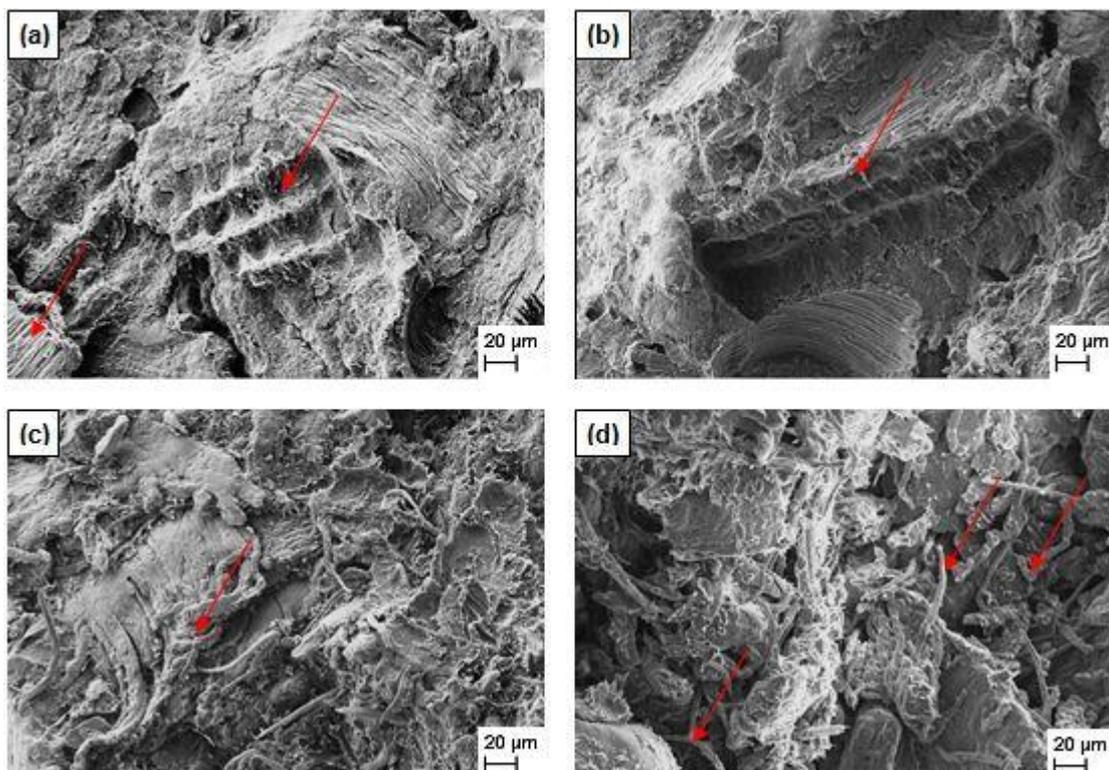


Fig. 5. SEM micrograph of rTPB/RH composites with (a) 50/50, (b) 40/60, (c) 30/70, and (d) 20/80 (wt%) compositions (150x)

Tensile Properties

The tensile strength, elastic modulus, and strain at break of the rTPB/RH composites at various compositions are depicted in Fig. 6. As can be observed in Fig. 6(a), the tensile strength and elastic modulus of the composites gradually increased with the increase in RH content up to the composite composition of 30/70 (wt%), and then they were reduced back for the composites with the composition of 20/80 (wt%). The tensile performance of the composite specimens could be affected by the type of constituent added, whether filler or polymer matrix, by the interfacial interaction, by modification *via* the addition of a compatibilizer, coupling agent, or surface treatment, and by the specimen processing method (Jayaraman and Bhattacharyya 2004; Yao *et al.* 2011). The increase in tensile strength and elastic modulus was attributed to the enhanced interfacial interaction and bonding between the hydrophobic rTPB matrix and the hydrophilic fillers *via* the mechanical interlocking induced by alkali treatment on the surface of RH fibers, and also to the interfacial modification by the addition of a MAPE coupling agent. The improvement of mechanical interlocking by the alkali treatment could be explained with the facts where: (1) the NaOH treatment increased the surface roughness of RH and promoted the fibre fibrillation, as observed in our previous work (Chen *et al.* 2015b); and (2) when the fibre surface is rough, the mechanical interlocking could occur to a greater extent (Pickering *et al.* 2016). On the other hand, the latter reduction in tensile strength and elastic modulus was attributed to the insufficient filler-matrix adhesion and interaction for the higher difference ratio of polymer matrix: filler in the rTPB/RH, which was about one fifth, as indicated by the SEM micrograph (Fig. 5(d)). At high filler contents, there was not enough interaction between filler and polymer matrix as it was dominated by the filler-filler interaction.

In Fig. 6(b), the strain at break of the composites showed a negative correlation with the RH content as compared with the neat rTPB. This trend was as estimated because the RH is a type of stiff organic filler that possesses a brittle nature, and hence, the incorporation of RH in a high filler loading (more than 50 wt%) tended to decrease the ductility characteristic of the polymer matrix (Chen *et al.* 2015b). Therefore, the strain at break of the composite decreased with the different rTPB/RH compositions, corresponding to the following order: 100/0 < 50/50 < 40/60 < 30/70 < 20/80 (wt%).

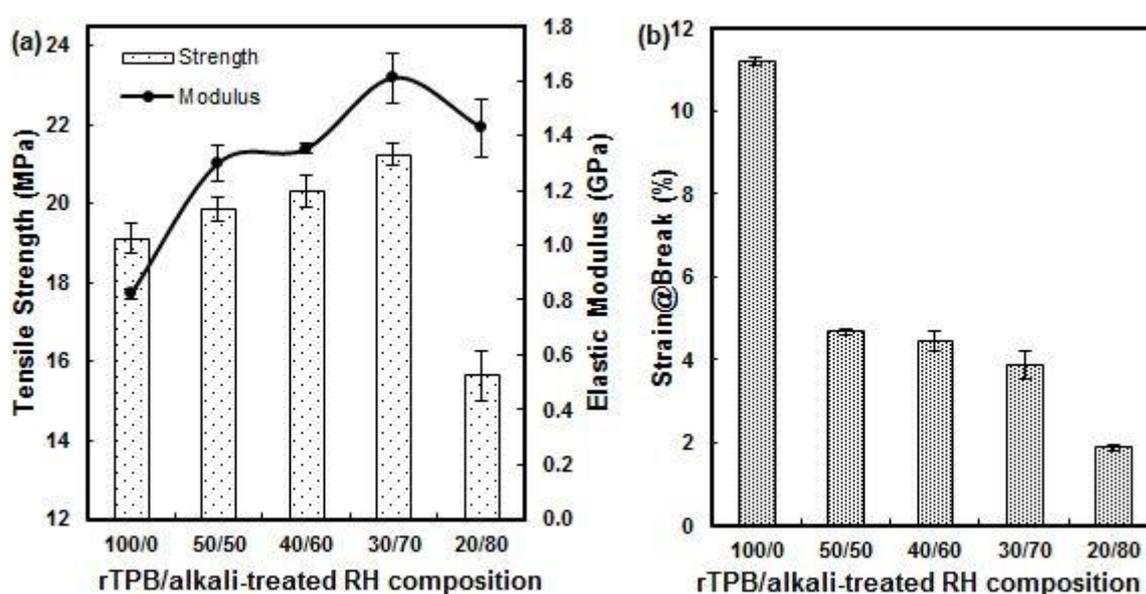


Fig. 6. Tensile properties of rTPB/RH composites with different compositions

CONCLUSIONS

1. The thermal stability of the recycled thermoplastic blend-rice husk (rTPB/RH) composites was significantly improved with the addition of the agricultural-waste fiber (RH).
2. With the increase in the RH content in rTPB, the E' of rTPB was noticeably increased at lower temperatures. Meanwhile, the E''_{max} peak temperature (α -relaxation) of the composites was shifted slightly higher for the rTPB incorporated with RH fibers.
3. The water absorption and orthotropic swelling of the composite were increased with the increase in immersion time and increase in RH fiber content, and reduced accordingly with different immersion media as follows: tap water > distilled water > sea water. The highest swelling percentage was obtained in the thickness dimension, followed by those in the width and then length dimensions.
4. The maximum tensile strength and elastic modulus values were obtained for rTPB/RH composites at the composition of 30/70 wt%, whereas the strain at break was dramatically decreased with the incorporation of high-fiber content RH into the rTPB matrix, and then continued to reduce gradually with the increasing RH contents.

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REFERENCES CITED

- Akil, H. M., Cheng, L. W., Mohd Ishak, Z. A., Abu Bakar, A., and Abd Rahman, M. A. (2009). "Water absorption study on pultruded jute fibre reinforced unsaturated polyester composites," *Compos. Sci. Technol.* 69(11-12), 1942-1948. DOI: 10.1016/j.compscitech.2009.04.014
- Albano, C., González, J., Ichazo, M., and Kaiser, D. (1999). "Thermal stability of blends of polyolefins and sisal fiber," *Polym. Degrad. Stab.* 66, 179-190. DOI: 10.1016/S0141-3910(99)00064-6
- Alwani, M. S., Khalil, H. P. S. A., Sulaiman, O., Islam, M. N., and Dungani, R. (2013). "An approach to using agricultural waste fibres in biocomposites application: Thermogravimetric analysis and activation energy study," *BioResources* 9(1), 218-230. DOI: 10.15376/biores.9.1.218-230
- Arjmandi, R., Hassan, A., Majeed, K., and Zakaria, Z. (2015). "Rice husk filled polymer composites," *Int. J. Polym. Sci.* 1-32. DOI: 10.1155/2015/501471
- ASTM D638-03 (2003). "Standard test method for tensile properties of plastics," American Society for Testing and Materials (ASTM), West Conshohocken, USA.
- ASTM D570-98 (2010). "Standard test method for water absorption of plastics," American Society for Testing and Materials (ASTM), West Conshohocken, USA.
- Bajwa, S. G., Bajwa, D. S., Holt, G., Coffelt, T., and Nakayama, F. (2011). "Properties of thermoplastic composites with cotton and guayule biomass residues as fiber fillers," *Ind. Crops Prod.* 33(3), 747-755. DOI: 10.1016/j.indcrop.2011.01.017
- Chen, R. S., Ab Ghani, M. H., Ahmad, S., Salleh, M. N., and Tarawneh Ma, A. (2014a). "Rice husk flour biocomposites based on recycled high-density

- polyethylene/polyethylene terephthalate blend: Effect of high filler loading on physical, mechanical and thermal properties,” *J. Compos. Mater.* 49, 1241-1253. DOI: 10.1177/0021998314533361
- Chen, R. S., Ab Ghani, M. H., Salleh, M. N., Ahmad, S., and Gan, S. (2014b). “Influence of blend composition and compatibilizer on mechanical and morphological properties of recycled HDPE/PET blends,” *Mater. Sci. Appl.* 5, 943-952. DOI: 10.4236/msa.2014.513096
- Chen, R. S., Ab Ghani, M. H., Salleh, M. N., Ahmad, S., and Tarawneh Ma, A. (2015a). “Mechanical, water absorption, and morphology of recycled polymer blend rice husk flour biocomposites,” *J. Appl. Polym. Sci.* 132, 2461-2472. DOI: 10.1002/app.41494
- Chen, R. S., Salleh, M. N., Ab Ghani, M. H., Ahmad, S., and Gan, S. (2015b). “Biocomposites based on rice husk flour and recycled polymer blend: Effects of interfacial modification and high fibre loading,” *BioResources* 10(4), 6872-6885. DOI: 10.15376/biores.10.4.6872-6885
- Ho, M.-P., Wang, H., Lee, J.-H., Ho, C.-K., Lau, K.-T., Leng, J., and Hui, D. (2012). “Critical factors on manufacturing processes of natural fibre composites,” *Compos. Part B Eng.* 43, 3549-3562. DOI: 10.1016/j.compositesb.2011.10.001
- Jayaraman, K., and Bhattacharyya, D. (2004). “Mechanical performance of woodfibre-waste plastic composite materials,” *Resour. Conserv. Recy.* 41(4), 307-319. DOI: 10.1016/j.resconrec.2003.12.001
- John, M. J., and Thomas, S. (2008). “Biofibres and biocomposites,” *Carbohydr. Polym.* 71(3), 343-364. DOI: 10.1016/j.carbpoly.2007.05.040
- Kim, H.-S., Yang, H.-S., Kim, H.-J., Lee, B.-H., and Hwang, T.-S. (2005). “Thermal properties of agro-flour-filled biodegradable polymer bio-composites,” *J. Therm. Anal. Calorim.* 81(2), 299-306. DOI: 10.1007/s10973-005-0782-7
- Kim, H.-S., Kim, S., Kim, H.-J., and Yang, H.-S. (2006). “Thermal properties of bio-flour-filled polyolefin composites with different compatibilizing agent type and content,” *Thermochim. Acta* 451(1-2), 181-188. DOI: 10.1016/j.tca.2006.09.013
- Konieczna, M., Markiewicz, E., and Jurga, J. (2010). “Dielectric properties of polyethylene terephthalate/polyphenylene sulfide/barium titanate nanocomposite for application in electronic industry,” *Polym. Eng. Sci.* 50(8), 1613-1619. DOI: 10.1002/pen.21687
- Louis, N. S. M., and Thomas, S. (2013). “Effect of rice husk ash on mechanical properties of low density polyethylene,” *J. Sci. Ind. Res.* 72(7), 441-445.
- Mahmoud, K. H., and Makled, M. H. (2012). “Infrared spectroscopy and thermal stability studies of natural rubber-barium ferrite composites,” *Adv. Chem. Eng. Sci.* 2, 350-358. DOI: 10.4236/aces.2012.23041
- Nair, M. M., Kamath, M. S., and Shetty, D. N. (2016). “Study on distilled/sea water absorption behaviour influenced by non-uniform long piled up coir fiber composites,” *Int. J. Innov. Res. Sci. Eng. Technol.* 5(9), 15-21. DOI: 10.15680/IJIRSET.2016.0505503
- Pickering, K. L., Aruan Efendy, M. G., and Le, T. M. (2016). “A review of recent developments in natural fibre composites and their mechanical performance,” *Compos. Part A.* 83, 98-112. DOI: 10.1016/j.compositesa.2015.08.038
- Rahman, M. R., Islam, M. N., Huque, M. M., Hamdan, S., and Ahmed, A. S. (2010). “Effect of chemical treatment on rice husk (RH) reinforced polyethylene (PE) composites,” *BioResources* 5(2), 854-869. DOI: 10.15376/biores.5.2.854-869
- Robertson, N.-L. M., Nychka, J. A., Alemaskin, K., and Wolodko, J. D. (2013). “Mechanical performance and moisture absorption of various natural fiber reinforced thermoplastic composites,” *J. Appl. Polym. Sci.* 130(2), 969-980. DOI: 10.1002/app.39237

- Shinoj, S., Panigrahi, S., and Visvanathan, R. (2010). "Water absorption pattern and dimensional stability of oil palm fiber-linear low density polyethylene composites," *J. Appl. Polym. Sci.* 117(2), 1064-1075. DOI: 10.1002/app.31765
- Singh, S., and Mohanty, A. K. (2007). "Wood fiber reinforced bacterial bioplastic composites: Fabrication and performance evaluation," *Compos. Sci. Technol.* 67(9), 1753-1763. DOI: 10.1016/j.compscitech.2006.11.009
- Tamrakar, S., and Lopez-Anido, R. A. (2011). "Water absorption of wood polypropylene composite sheet piles and its influence on mechanical properties," *Constr. Build. Mater.* 25(10), 3977-3988. DOI: 10.1016/j.conbuildmat.2011.04.031
- Yao, F., Wu, Q., Liu, H., Lei, Y., and Zhou, D. (2011). "Rice straw fiber reinforced high density polyethylene composite: Effect of coupled compatibilizing and toughening treatment," *J. Appl. Polym. Sci.* 119(4), 2214-2222. DOI: 10.1002/app.32946
- Zhao, Q., Zhang, B., Quan, H., Yam, R. C. M., Yuen, R. K. K., and Li, R. K. Y. (2009). "Flame retardancy of rice husk-filled high-density polyethylene ecocomposites," *Compos. Sci. Technol.* 69(15-16), 2675-2681. DOI: 10.1016/j.compscitech.2009.08.009

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