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Long-term moisture absorption and thickness swelling behaviour of recycled thermoplastics reinforced with *Pinus radiata* sawdust

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Abstract

Composite materials were made from *Pinus radiata* wood flour, and recycled and virgin polyolefin, mainly high density polyethylene (HDPE) and polypropylene (PP) polymer, by using hot-pressing moulding. Long-term water absorption and thickness swelling (TS) kinetics of the composites was investigated with water immersion. It was found that the water absorption and TS increase with wood content and water immersion time before an equilibrium condition was reached. Composites made from the recycled plastics show comparable results as those made of the virgin plastics. It is interesting to find that the water absorption and TS can be reduced significantly with incorporation of a coupling agent (maleated polypropylene) in the composite formulation. Microstructures of the composites were examined to understand the mechanisms for the wood–plastic interaction which affects the water absorption and thickness swelling. Further studies were conducted to model the water diffusion and thickness swelling of the composites. Diffusion coefficients and swelling rate parameters in the models were obtained by fitting the model predictions with the experimental data.

Keywords: Wood–plastic composite; Wood flour; Recycled thermoplastics; Water absorption kinetics; Thickness swelling

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1. Introduction

The worldwide production and consumption of plastics result in a significant contribution to municipal solid waste (MSW). For example, waste plastic accounts for 11.8% of the annual 246 million tonnes of the MSW generated in US during 2005 [1]. The increasing quantities of plastic wastes and the effective and safe disposal has become a serious public concern. Attempts have been made to recycle the consumer plastics in order to reduce the environmental impact and the consumption of the virgin plastics. In Western Europe, plastics recovery was about 8.25 million tonnes in 2004 (39% of total amount of plastics consumed) [2]. The proportion of the recycled plastics in US was 5.7% in 2005 [1] and that in New Zealand was 13.48% [3]. Previous studies have shown that the properties of the recycled high density polyethylene (HDPE) obtained from the post-consumer milk bottles were not largely different from those of virgin resin and thus could be used for different applications [4].

Similarly, a large amount of wood waste was generated at different stages in the wood processing and wood applications such as furniture making and building construction. Such wood wastes have mainly been destined for landfill. In US alone, about 63 million tonnes of wood wastes were generated in 2002 and 26.8 million tonnes of them ended in the MSW [5]. However, studies have also shown that the waste wood in the form of wood flour, fibres or pulp is suitable as a filler for polyolefin's matrix composites [6,7]. Therefore, the increased usage of recycled plastics and waste wood offers the prospect of lessening solid waste disposals and reducing the costs for making the wood–plastic composites.

Wood–plastic composites (WPCs) are becoming a preferred building material due to their better stability and required mechanical properties as compared to wood. Traditionally, WPCs are made from virgin plastics and wood flour or wood fibres [7–12]. However, the studies of WPCs based on recycled thermoplastics are very limited [13–17]. Most of the studied were focused on the use of either a single type of plastic from the waste stream (for example, the milk bottle), or combination of recycled and virgin plastics to produce the composites. In practice, the post-consumer plastic waste stream may contain many

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different grades, colors, and contaminants, leading to varying performance when these plastics are combined with the wood fillers. The impact of post-consumer thermoplastics in WPCs is still not fully understood, leaving open research opportunities for the optimisation of product properties.

Although the use of wood filler in plastic composites has several advantages over inorganic fillers, hydrophilic nature of the wood has a negative effect on the overall performance of the WPCs [18]. Increased moisture content of the composites reduces their mechanical properties and dimensional stability [19]. However, it was found that the incorporation of coupling agents such as functionalised polyolefin can improve the overall properties through enhanced compatibility between the hydrophilic wood filler and the hydrophobic polymer matrix [20–22]. The wood flour is preferred over wood fibre in the WPCs in some applications such as the decking and flooring. The WPCs products typically contain approximately 50 wt.% wood, although some composites contain much less wood and others contain as much as 70 wt.% [23]. The wood flour content in the WPCs mainly depends on the product applications, and the processing methods. In building applications, dimensional stability of the WPCs is critical, especially for outdoor use. To improve the stability, understanding of the water absorption process and impact of the absorbed water on the dimensional change is important [19,24–27]. Previous studies have applied diffusion theory to quantify the water absorption process in the WPCs [28–31]. Most of these studies used mixture of natural fibres and virgin thermoplastics for the composite formulations. This work aims to investigate the characteristics of water sorption kinetics and thickness swelling behaviour in the WPCs made from post-consumer recycled thermoplastics (HDPE and PP) and waste sawdust.

2. Theoretical approach

2.1. Mechanisms of water transport

The transport of water through composite materials may theoretically follow different mechanisms depending on factors such as the chemical nature of the polymer, dimensions and morphology of the wood filler and polymer-filler interfacial adhesion [32]. In general, the water transport behaviour in the polymer matrix composites can be Fickian diffusion, relaxation controlled, and non-Fickian or anomalous. These three cases of water transport can be distinguished theoretically by the shape of the sorption curve represented by the following equation [33,34]:

$$\log\left(\frac{M_t}{M_\mathrm{m}}\right) = \log(k) + n\log(t) \tag{1}$$

where M_t is the moisture content at specific time (t), M_m the equilibrium moisture content (EMC), and k and n are constants. The value of coefficient n shows different behaviour between the three cases of water transport: Fickian diffusion ($n = 0.5$), relaxation ($n \geq 1$) and anomalous transport ($0.5 < n < 1.0$). The coefficients (n and k) can be determined from the slope and the intercept of M_t/M_m versus t in the log plot which can be drawn from experimental data.

2.2. Diffusivity determination

The most commonly used method in determination of the Fickian mass diffusivity in polymer composites is developed by Shen and Springer [35]. This method assumes one-dimensional, unsteady diffusion through the thickness of the composite panel where the flat face is much greater than the thickness as expressed by Eq. (2). The analytical solution of this equation for a plane sheet with uniform distribution of initial moisture concentration through the thickness can be obtained as expressed by Eq. (3) which is for the overall moisture change *via* both faces of the composite panel for a short period of time [33,34]:

$$\frac{\partial C}{\partial t} = D_x \frac{\partial^2 C}{\partial z^2} \tag{2}$$

$$M_t = 4M_\mathrm{m}\sqrt{\frac{D_x}{\pi h^2}}\sqrt{t} \tag{3}$$

where M_t is the average moisture content over the panel at any time (t), M_m denotes the corresponding EMC after sufficient time when the mass loss is insignificant, D_x denotes diffusion coefficient, and h is the thickness of the composite panel. Eq. (3) indicates that the mass of the sample increases with time without an upper limit. The surface equilibrium moisture content is in this case the average moisture content when the sample is kept sufficient time in the water immersion. This moisture content may not be the truce equilibrium moisture content but is very close to the surface moisture content when the composite is in contact with the water. Therefore, the diffusion equation here was only used to describe the moisture ingress dynamics at the initial stage of the process provided that the sample geometry mimics an infinitely wide slab. For such cases, one can utilize this equation to determine diffusion coefficient by equating the slope of Eq. (3) with respect to the root square of the time to the slope determined from experimental data. The resulting closed form solution for the diffusion coefficient from Eq. (3) with correction for edge effect [30] is given as follows:

$$D_x = \pi\left[\frac{\theta}{4M_\mathrm{m}}\right]^2\left[1 + \left(\frac{h}{l}\right) + \left(\frac{h}{b}\right)\right]^2 \tag{4}$$

where θ is the slope of linear portion of M_t versus $\sqrt{t}/h$ plot, l the panel length and b is the panel width. Thus, the moisture absorption of the WPCs can be determined by using either Eqs. (1) or (4). However, it should be noted that these two equations are only applicable for a certain period of time which can be estimated once the EMC is known.

Once the diffusion coefficient (D_x) is known, the permeability (P) and thermodynamic solubility (S) parameters of the composites can be determined. The solubility is the amount of water absorbed per unit mass of the composite at equilibrium while the permeability is defined as the product of the diffusion coefficient and solubility [33]:

$$P = D_x \cdot S \tag{5}$$

2.3. Prediction of thickness swelling

The thickness swelling (TS) is an important property that represents the stability performance of the composite. Generally, the swelling rates for polymer matrix composites are low during the initial stages of moisture absorption due to the viscoelasticity of the polymer matrix. In addition, any pores or voids that are present after fabrication will help to accommodate some of the swelling of the added wood. Based on these considerations, the thickness swelling of the composite panel has been determined by the following equation as proposed by Shi and Gardner [36]:

$$\mathrm{TS}(t) = \left(\frac{h_\infty}{h_0 + (h_\infty - h_0)\mathrm{e}^{-K_{\mathrm{sr}}t}} - 1 \right) \qquad (6)$$

where h_0 is the initial composite panel thickness at $t=0$, h_∞ the ultimate thickness of the panel at equilibrium and K_{sr} is the intrinsic relative swelling rate parameter. The value of K_{sr} in Eq. (6) depends on how fast the composites swell and reach the ultimate thickness swelling at equilibrium. The values of K_{sr} for both the wood-recycled plastic composites and the wood–virgin plastic composites will be determined through non-linear regression curve fitting to the experimental data.

3. Experimental methods

3.1. Materials

Radiata pine (*Pinus radiata)* sawdust was collected from a local sawmill. The fresh sawdust was dried at 102 °C for 24 h to moisture content of 2–3% (dry-base) and then ground to finer flour of 35–45 mesh sizes (diameter between 0.18 and 0.5 mm). Recycled HDPE (rHDPE) and recycled polypropylene (rPP) granules were provided by a local plastics recycling company derived mainly from post-consumer beverage packaging, containers and films, milk bottles and janitorial-grade packaging. The plastic granules were thoroughly washed with water and then dried. Virgin plastics of both high density polyethylene (vHDPE) and polypropylene (vPP) were also used for manufacturing of the wood–plastic composites for comparison studies. A maleated polypropylene (MAPP) coupling agent used was Epolene G-3015 polymer.

The melt flow index (MFI) of the recycled and the virgin plastics were measured by GunnLab, Christchurch, New Zealand. HDPE was tested using conditions of 190 °C and 2.16 kg load while the PP was tested at conditions of 230 °C and the same load (2.16 kg). The MFI value for the rHDPE was 0.072 g/10 min and that for the rPP was 21 g/10 min. The corresponding MFI values for vHDPE and vPP were 0.1 g/10 min and 25 g/10 min, respectively.

3.2. Composite preparation

The wood flour was compounded with either the recycled or the virgin polyolefin granulates in a co-rotating twin screw extruder. The extruded strand was passed through a water bath, palletised, and dried at 75 °C for 4 h to remove any moisture. The composite granules were consolidated by moulding in an electrically heated platen press using an aluminium mould to produce panels with final dimensions of 165 mm × 152 mm × 6.4 mm (thickness). The composite panels was hot-pressed first for 4 min under a pressure of 1.0 MPa and then for 5 min at a pressure of 5.0 MPa. This was followed by cold pressing under the pressure of 5.0 MPa. After the cold press, test specimens were conditioned at a temperature of 23 ± 2 °C and relative humidity (RH) of 50 ± 5% for at least 40 h in accordance with ASTM D618-99 [37]. The experimental variables studied were polymer type (HDPE, PP), polymer form (virgin, recycled), wood flour content and coupling agent content. Based on the polymer form, the composites were divided into two major series (HDPE and PP) to give a range of different formulations as described in Table 1.

Table 1
WPCs formulation (percent by weight)

Composite sample code	Polymer type	Polymer content (%)	Wood flour (%)	Coupling agent (%)
Wood flour–HDPE composites				
rHDPE100	Recycled	100	0	0
vHDPE60W40	Virgin	60	40	0
rHDPE70W30	Recycled	70	30	0
rHDPE60W40	Recycled	60	40	0
vHDPE50W50	Virgin	50	50	0
rHDE50W50	Recycled	50	50	0
rHDPE47W50CA3	Recycled	47	50	3
rHDPE45W50CA5	Recycled	45	50	5
rHDPE57W40CA3	Recycled	57	40	3
Wood flour–PP composites				
rPP100	Recycled	100	0	0
vPP50W50	Virgin	50	50	0
rPP50W50	Recycled	50	50	0
rPP47W50CA3	Recycled	47	50	3
rPP60W40	Recycled	60	40	0
rPP45W50CA5	Recycled	45	50	5

3.3. Measurement for water absorption and thickness swelling

Water absorption and thickness swelling tests were conducted in accordance with ASTM D570-98 [38]. Long-term water immersion tests were conducted at 23 ± 1 °C for 63 days. In total, five replicates from two different manufactured panels were tested for each composite formulation. The weight and thickness of the specimens were measured periodically until the samples attained equilibrium. For each measurement at preset time, the specimens were first taken out of the water and the liquid water attached on the surface was then removed using blotting paper. The moisture absorption was determined as the weight gain over the oven-dry weight of the samples, and calculated using the following expression:

$$M(\%) = \frac{(m_t - m_0)}{m_0} \times 100 \tag{7}$$

where, m_0 and m_t denote the oven-dry weight and weight after time t, respectively. The EMC was considered to be attained when the increase in weight over a week was less than 1% in three consecutive weeks in accordance with ASTM D570-98 [38]. Thickness swelling (TS) was assessed by measuring the average thickness of each specimen during the immersion tests (h_t) and after oven drying (h_0), and then calculated using the following equation:

$$\mathrm{TS}(\%) = \frac{(h_t - h_0)}{h_0} \times 100 \tag{8}$$

3.4. Micro-structural examination of fracture surfaces

The fracture surfaces of the test specimens were examined with high resolution field emission scanning electron microscopy (FESEM) (JSM-7000F). The FESEM was operated at an accelerating voltage of 1–5 kV and emission current of 47 μA. Fracture surfaces were sputter-coated with a gold coating of approximately 50 nm in thickness. Specimens were analysed at magnifications of 100× and 200×.

4. Results and discussion

4.1. Long-term water absorption behaviour

The long-term water absorption of the composites panels were monitored by full water immersion over a period of 63 days (1512 h) as shown in Figs. 1 and 2. Generally, the water absorption increased with the wood filler content and immersion time until equilibrium conditions were reached. During this period, the polymers exhibited negligible water absorption while the wood flour induced significant water absorption. Water absorption was the maximum for composites made of wood flour–rHDPE and wood flour–vHDPE with 50 wt.% wood content, having water contents of 19.5% and 23.5%, respectively after 1512 h (Fig. 1). However, at the same wood contents, addition of 3–5 wt.% coupling agent (MAPP) to the composites significantly reduced the water absorption. This was true for the composites using both the virgin plastics and the recycled plastics. For the composites of 50 wt.% of wood flour, addition of 3 wt.% coupling agent (rHDPE47W50CA3) reduced the EMC to 7.8% as compared to 19.5% EMC of the corresponding composite without coupling agent (rHDPE50W50). Similar trends were observed for the wood flour–PP series (Fig. 2). The PP based composites with 50 wt.% wood flour exhibited the highest water absorption values both for the virgin and for the recycled matrices, and the addition of MAPP again reduced the water uptake significantly.

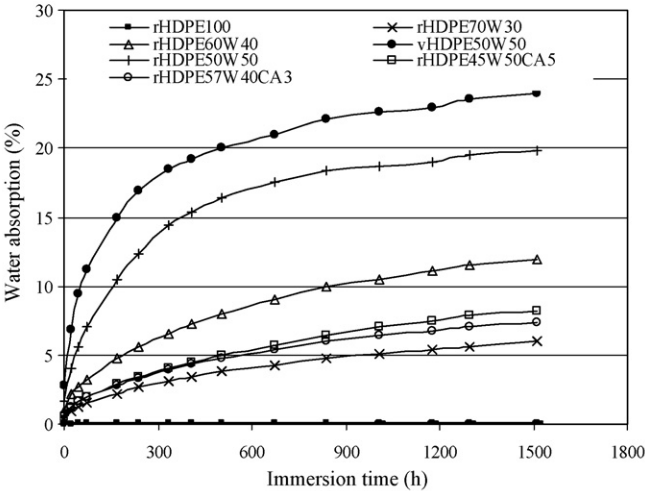

Fig. 1. Water absorption behaviour for wood flour–HDPE composites.

The water absorption increase with the wood content can be explained by the water–wood interaction. When the content of wood flour is increased in the composite, the number of free OH groups of wood cellulose increases and, hence, the water absorption increases. These free OH groups come in contact with water and form hydrogen bonding, which results in weight gain in the composites. Also the composite with lower wood content reaches the equilibrium moisture content more quickly. The addition of the coupling agent can enhance the bonding in WPCs, due to the improved compatibility between the polymer and the wood particles thus more wood surface

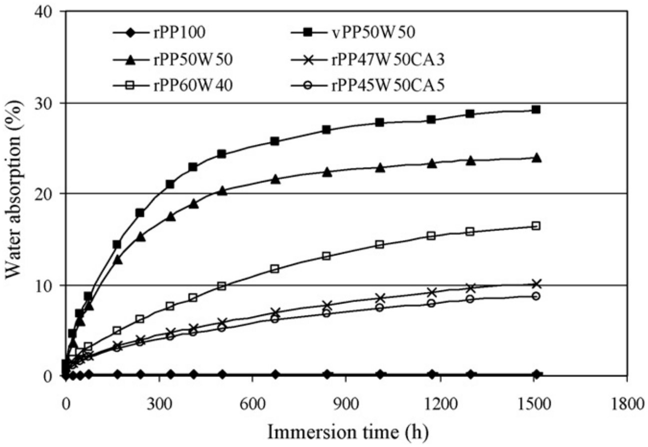

Fig. 2. Water absorption behaviour for wood flour–PP composites.

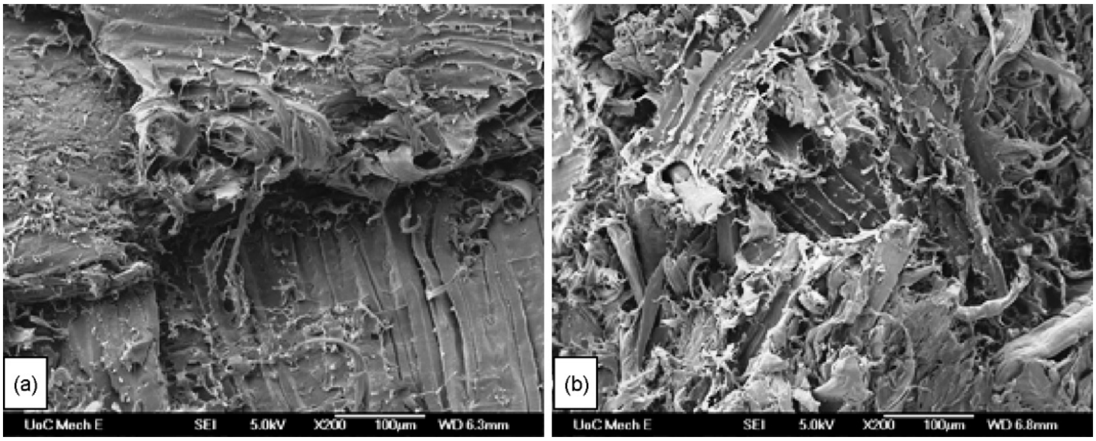

Fig. 3. SEM micrographs of fracture surface of composites at 200× (a) vHDPE50W50 and (b) rHDPE50W50.

areas being covered by the polymer during compounding. The addition of the coupling agent increases the ester linkages between the hydroxyl groups of wood flour and the anhydride part of MAPP [39]. Therefore, the amount of free OH^- in the wood cellulose is reduced because some of them are interacting with succinic anhydride. Due to these changes, the water absorption increment is rather less, compared to the composite formulation without MAPP. The other possible reason for less water absorption could be the change in crystallinity of WPCs coupled by the coupling agent (MAPP). It was reported that such crystallinity of the WPCs is much greater than that of the corresponding WPCs without the MAPP modification [10]. As the crystalline regions are impermeable to the penetrant, the water absorption is less in the coupled composites.

It is interesting to note that composites based on the recycled matrix absorbed less water compared with the virgin polymer matrix. The possible reason is the enhanced dispersion and interfacial adhesion due to the presence of chemical impurities, different molecular and compositional differences (MFI and crystallinity) between the virgin and the recycled plastics. This can be confirmed by comparing the SEM micrographs of fractured surfaces of composites made of virgin plastics and recycled plastics (Figs. 3 and 4). The wood flour dispersion and adhesion was slightly better in the WPCs with rHDPE as shown in Fig. 3b as compared the WPCs of vHDPE (Fig. 3a). Similarly, wood flour dispersion and adhesion in composites with rPP (Fig. 4b) was better than those with vPP (Fig. 4a). The composites with the virgin plastics shows more isolated fibres, and some gaps and flaws which provide more water residence sites. However, it was observed that in the composites with recycled plastics, polymer penetrated into the fibre lumens, pit holes, and other void spaces in the matrix. This was not obvious in the composites made of virgin polymers.

The water sorption kinetics in the WPCs specimens can be evaluated through the diffusion constants (k and n) obtained from the experimental data. It was observed that except for one formulation (vHDPE60W40), the value of n is close to 0.5 for all of the other composites (Table 2). This confirms that the Fickian diffusion can be used to adequately describe water transport in the composites, which is consistent with previous studies [28,31]. A higher value of n and k indicates that the composite needs shorter time to attain equilibrium water absorption. The value of k was found to increase with increasing wood content for both the virgin and the recycled polymer matrices. The value of k for rPP was higher that of wood flour–PP composites, which may be due to the presence of some impurities in the recycled PP resulting higher moisture absorption initially. The k tends to be less for the MAPP coupled wood flour–PP composites, confirming the

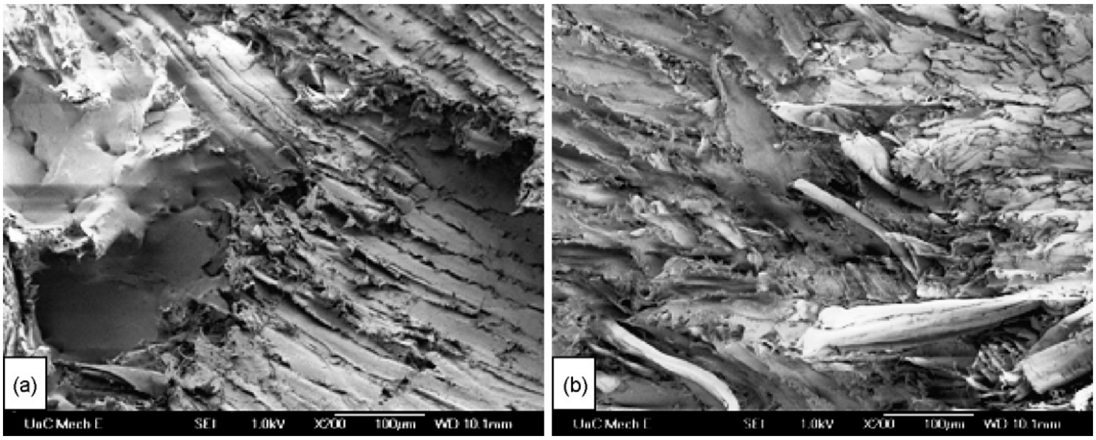

Fig. 4. SEM micrographs of fracture surface of composites at 200× (a) vPP50W50 and (b) rPP50W50.

Table 2
Diffusion case selection parameters

Composite specimen code	n	k (h^{-n})
vHDPE60W40	0.3530	0.0839
rHDPE70W30	0.4165	0.0495
rHDPE60W40	0.4037	0.0599
vHDPE50W50	0.3865	0.0887
rHDPE50W50	0.4210	0.0605
rHDPE47W50CA3	0.4072	0.0525
rHDPE45W50CA5	0.4253	0.0435
rHDPE57W40CA3	0.4049	0.0562
rPP100	0.5300	0.0332
vPP50W50	0.5535	0.0295
rPP50W50	0.5687	0.0285
rPP47W50CA3	0.4260	0.0384
rPP60W40	0.4906	0.0246
rPP45W50CA5	0.4379	0.0381

reduction in water absorption. However, MAPP coupled wood flour–HDPE composites had higher k value than non-coupled composites, and the reason for this is not clear.

From the measured water absorption and the sample mass and the determined diffusion coefficient (D_x), the composite thermodynamic solubility (S) and the permeability (P) have been calculated. It is seen that D_x, S and P all increase with the wood flour content for the composites examined (Table 3). These results are consistent with previous findings on wood and natural fibres composites [29,31].

The addition of MAPP decreases the D_x compared with the non-coupled composites. The virgin HDPE composites have higher diffusion coefficients compared to the corresponding recycled HDPE composites. In contrast, the recycled PP composites exhibit higher diffusion coefficients compared with composites based on the virgin PP. This is possible due to contaminants present in the rPP; however, the observed difference is within the experimental error and thus may not be significant.

The direct comparison of the diffusion coefficient obtained from this work with previous studies is difficult due to differences in materials, manufacturing methods and test conditions. Nevertheless, the magnitude of the diffusion coefficients obtained in this work (2.76×10^{-12} to 9.45×10^{-12} m^2/s) are close to the reported values in literature. Wang et al. [31] published a diffusion coefficient of 4.63×10^{-13} m^2/s for hot pressed 50 wt.% rice hull-HDPE composites coupled with MAPP. Espert et al. [29] reported a diffusion coefficient of 1.09×10^{-12} m^2/s for PP composites containing 30 wt.% coir fibre and a diffusion coefficient of 1.83×10^{-12} m^2/s for composites containing 30 wt.% luffa fibre. Tajvidi et al. [40] prepared injection moulded 50 wt.% wood flour–PP composites with 2 wt.% MAPP with a measured diffusion coefficient of 1.33×10^{-11} m^2/s.

4.2. Long-term thickness swelling behaviour of composites

The thickness swelling (TS) of the wood flour–polyolefin composites was determined in the experiment from Eq. (8). The results show that the TS was the highest for the 50 wt.% wood flour–HDPE (5.8–6.6%) and 50 wt.% wood flour–PP (9.3–10.6%), which correspond to the highest water absorption. In a similar trend to the water absorption, the TS increase with wood flour content for non-coupled composites. For example, 30, 40, and 50 wt.% wood filler in the rHDPE-based composites exhibited equilibrium TS values of 2.2%, 4.4% and 6.6%, respectively (Fig. 5). However, the equilibrium TS of 50 wt.% wood flour–rHDPE composite was reduced from 6.6% to 2.8% with the addition of 3 wt.% MAPP. It is interesting to note that swelling of the 50 wt.% wood flour–vHDPE composite was greater than the 50 wt.% wood-rHDPE composite up to 300 h. After this time, the swelling of the 50 wt.% wood-vHDPE composite was almost stable whereas the swelling of the 50 wt.% wood-rHDPE composite continued. The high TS for the wood-vHDPE in the initial stage of water immersion is possible due to the poor fibre dispersion and adhesion in the composite with the vHDPE matrix (Fig. 3b), which allows easy access of water to cellulose. The TS for wood flour–PP composites follows

Table 3
Diffusion coefficients and permeability of wood flour/polyolefin composites

Composite specimen code	EMC (%)	Diffusion coefficient (m^2/s)	Solubility parameter	Permeability (m^2/s)
rHDPE100	0.12 (0.03)	[a]		
vHDPE60W40	11.6 (1.4)	3.95E-12	1.13	4.45E-12
rHDPE70W30	5.63 (1.2)	3.24E-12	1.06	3.42E-12
rHDPE60W40	11.1 (1.7)	3.76E-12	1.11	4.19E-12
vHDPE50W50	23.54 (1.3)	9.45E-12	1.24	1.17E-11
rHDPE50W50	19.49 (0.8)	5.94E-12	1.19	7.10E-12
rHDPE47W50CA3	7.79 (1.4)	3.80E-12	1.09	4.15E-12
rHDPE45W50CA5	7.85 (1.5)	3.49E-12	1.09	3.79E-12
rHDPE57W40CA3	7.08 (1.6)	3.22E-12	1.08	3.47E-12
rPP100	0.21 (0.04)	[a]		
vPP50W50	27.98 (1.7)	5.26E-12	1.29	6.76E-12
rPP50W50	23.23 (1.5)	6.26E-12	1.24	7.77E-12
rPP47W50CA3	9.62 (0.5)	3.43E-12	1.10	3.76E-12
rPP60W40	15.81 (0.4)	2.76E-12	1.16	3.20E-12
rPP45W50CA5	8.29 (0.7)	3.05E-12	1.08	3.30E-12

Note: Values given in the parentheses are average standard deviation of five replicate samples.

[a] Diffusion coefficient was not calculated because gradient of linear portion could not be determined precisely.

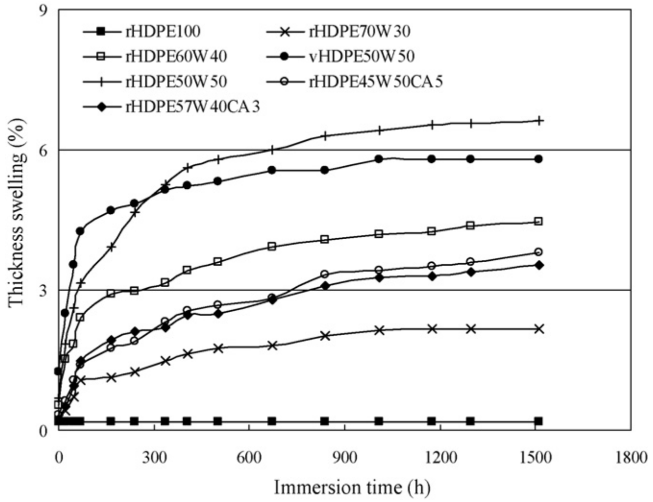

Fig. 5. Thickness swelling vs. water immersion time for wood flour–HDPE composites.

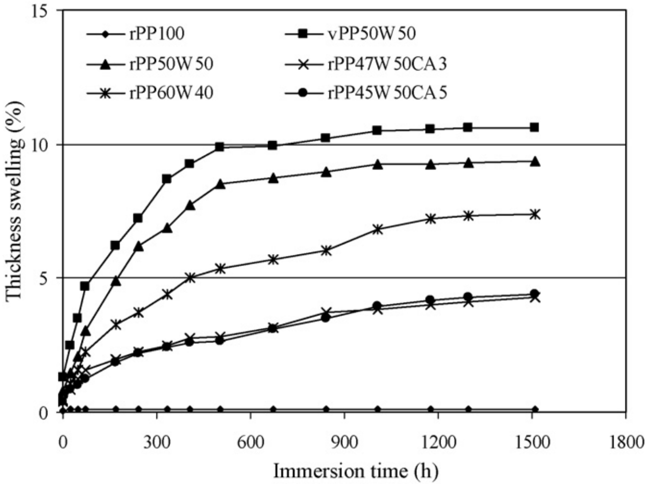

Fig. 6. Thickness swelling vs. water immersion time for wood flour–PP composites.

a similar trend to the water absorption behaviour, increasing with immersion time until an equilibrium condition is attained (Fig. 6). For example, the equilibrium TS of the 40 and 50 wt.% wood filler in the recycled PP matrix were 7.3% and 9.3%, respectively. Similarly to the HDPE-based composites, the TS of the 50 wt.% wood-rPP composite was reduced from 9.3% to 4.1% with the addition of 3 wt.% MAPP.

The experimental data was used to obtain the swelling rate parameter (K_{sr}) in Eq. (6) by using non-linear regression curve fitting. The K_{sr} values, standard error, and coefficient of variation (CV) are given in Table 4. The swelling parameter, K_{sr}, quantifies the rate of the composites approaching the equilibrium value for thickness swelling after sufficient time of water immersion. The higher value of K_{sr} indicates the higher rate of the swelling and thus the composite reaches the equilibrium TS in a shorter period of time. It is found that K_{sr} values for the composites with the virgin polymer is higher than those of the composites made from the recycled polymers (HDPE and PP) in all levels of the wood flour content. For example, the composite of 50 wt.% of wood content and 50 wt.% of virgin PP approached the equilibrium TS about 36% faster than the recycled PP-based composite. The swelling rate of the composites increases with the wood flour content but is reduced significantly with addition of the MAPP, which is due to the improved compatibility between polymer and wood flour through the esterification [39].

Fig. 7 shows the comparison of the predicted TS from the swelling model (Eq. (7)) and the experimental data for the PP series. It is found that the swelling model can fit the experimental data closely for both the wood flour–PP composites (Fig. 7) and the wood flour–HDPE composites although the later graph is not shown in this paper. The model slightly overestimates the thickness swelling for the HDPE composites without the MAPP treatment during the immersion period after the initial

Table 4
Measured TS and predicted swelling rate parameter for wood flour/polyolefin composites

Composite specimen code	Initial thickness (h_0) (mm)	Final thickness (h_∞) (mm)	Equilibrium TS (%)	Swelling rate parameter (K_{sr})$\times 10^{-3}$ h^{-1}	Standard error ($\times 10^{-4}$ h^{-1})	CV (%)	R^2
Wood flour–HDPE composites							
vHDPE60W40	6.61	6.95	4.86 (0.2)	12.76	17.57	18.35	0.91
rHDPE70W30	6.68	6.82	2.17 (0.4)	4.61	5.52	6.76	0.91
rHDPE60W40	6.67	6.94	4.38 (0.6)	9.31	15.95	13.42	0.85
vHDPE50W50	7.17	7.57	5.8 (0.3)	21.62	28.89	28.56	0.89
rHDPE50W50	6.88	7.34	6.56 (0.45)	5.98	6.94	8.15	0.91
rHDPE47W50CA3	6.80	7.13	2.8 (0.2)	1.05	1.74	1.47	0.83
rHDPE45W50CA5	6.84	7.12	3.6 (0.9)	2.37	2.33	3.34	0.88
rHDPE57W40CA3	6.60	6.81	3.39 (1.0)	4.76	5.42	6.98	0.93
Wood flour–PP composites							
vPP50W50	6.84	7.55	10.61(0.2)	6.49	5.48	8.60	0.96
rPP50W50	6.96	7.61	9.31 (1.0)	4.77	2.04	6.26	0.99
rPP47W50CA3	7.19	7.50	4.1 (0.9)	2.76	2.87	3.67	0.89
rPP60W40	6.29	6.75	7.32 (0.96)	3.02	2.06	4.47	0.96
rPP45W50CA5	6.55	6.84	4.30 (0.85)	2.39	2.07	3.49	0.92

Note: Values given in the parentheses are average standard deviation of five replicate samples.

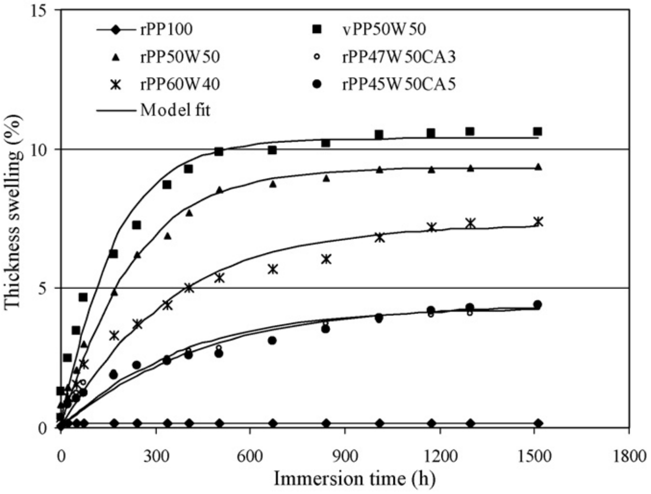

Fig. 7. Thickness swelling model fit for wood flour–PP composites.

fast swelling. The model prediction for the PP composites is satisfactorily accurate ($R^2 = 0.9$). The accuracy of the swelling model prediction appears to be a function of the magnitude of the swelling rate parameter (Table 4). It is also observed that more accurate prediction from the swelling model can be obtained for the composites with lower thickness swelling.

According to Shi and Gardner [36] the swelling rate increased linearly with the decreasing of the composite board density. Our findings support this in principle, but such relationships is not strong as shown in Fig. 8 with the R square value being 0.61. It reflects that the thickness swelling is a complicated behaviour and can be affected by the combination of factors such as polymer type, virgin polymer or recycled, wood content and more significantly the coupling agent treatment. Fig. 9 shows the relationship between the EMC and the equilibrium TS of the composites and very significant relationship is confirmed ($R^2 = 0.91$). The linear correlation can be expressed by an empirical equation as given in the following equation:

$$\mathrm{TS}(\%) = 0.35\mathrm{EMC} + 0.55 \tag{9}$$

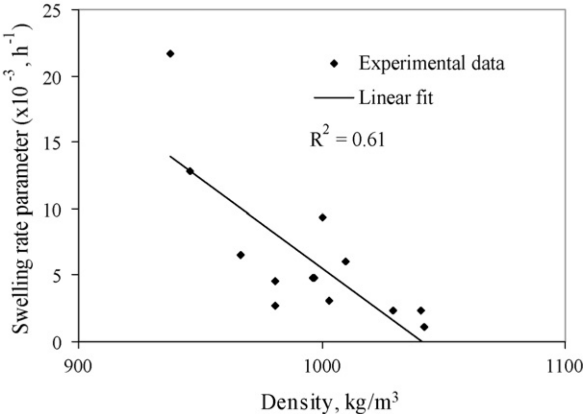

Fig. 8. Swelling rate vs. density of wood flour/polyolefin composites.

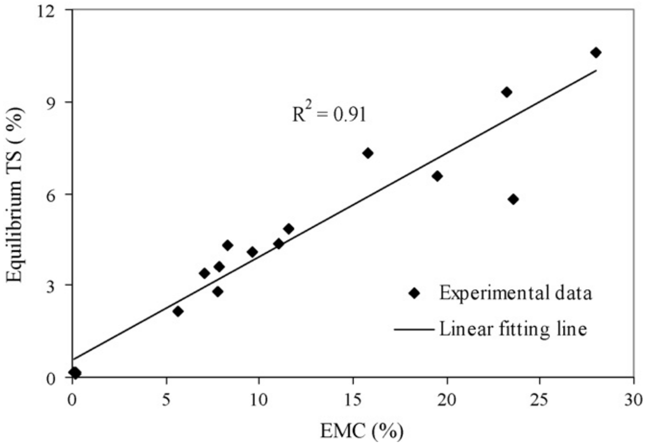

Fig. 9. EMC vs. equilibrium TS for wood flour/polyolefin composites.

5. Conclusion

This paper investigated the long-term water absorption and thickness swelling (TS) properties for the hot-press moulded composites made of wood flour filled-recycled and wood flour–virgin polyolefin (HDPE and PP). With the water immersion tests, the water absorption and TS increase with wood content and immersion time for all types of composites irrespective of polymer type and form. The composites made from the recycled plastics show comparable results to the composites made of the virgin plastics. However, the water uptake and TS can be reduced significantly with the incorporation of coupling agent (MAPP) in the composite formulation. From the experimental results, TS has liner relationship with the water absorption.

Water transport mechanism within the wood flour–polyolefin composites was proved to follow the Fickian diffusion. The kinetics parameters are influenced by the wood flour content, polymer type and form, and coupling agent. The diffusion coefficient, composite thermodynamic solubility and permeability increase with wood flour content but these parameters are all reduced with the addition of MAPP. A thickness swelling model is successfully employed to determine the swelling rate parameter although better predictions are achieved for the composites with lower thickness swelling rate.

The results and findings of this work provide evidence that the recycled plastics (HDPE and PP) can be successfully used to produce stable WPCs suitable for exterior applications in terms of low water absorption and low thickness swelling. The improved performance of the recycled plastics based composites can be achieved by increasing the polymer content and or by addition of coupling agents. Further research work is being undertaken to explore the detail mechanisms of moisture transport, moisture distribution and damage development, and factors influencing kinetics parameters.

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