



The mechanical properties deterioration of rubberwood-latex sludge flour reinforced polypropylene composites after immersing in different water conditions

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Abstract

The hydrophilic nature of wood-plastic composites (WPCs) is a disadvantage that impacts its performance in applications for construction and building products. Therefore, developing the WPCs into composites that reduce water absorption needs further evaluation. The current work investigates the effects of different water types (distilled water, and water from the Gulf of Thailand and the Andaman Sea), latex sludge types, and immersion time on the physical and mechanical properties of rubberwood-latex sludge flour-reinforced polypropylene composites. The composite samples were produced by a twin-screw extruder (mixing) and a compression molding machine (forming). The results revealed that the composites exposed to different water conditions for a long period resulted in significant ($\alpha = 0.05$) reduction in the modulus of rupture, modulus of elasticity, screw withdraw strength, and hardness with maximum percentage reduction values of 79.9, 117.5, 69.9, and 5.84, respectively. However, adding the latex sludge at 25 wt% resisted deterioration. The composites immersed in Andaman Sea water exhibited the least water absorption and deterioration for all the mechanical properties with a minimum hardness loss (3.74%). The composites immersed in distilled water showed the maximum deterioration for all mechanical properties, with 117.5% loss in modulus of elasticity. Overall, adding the latex sludge flour to the composites made them resistant to mechanical properties deterioration, which is favorable for WPCs used in environments involving contact with the seawater, especially the Andaman Sea.

1 Introduction

In recent years, natural fillers have been widely used to reinforce plastic to produce composite material and significantly increase the application of new composites (Phiri et al. 2020;

Srivabut et al. 2022). It is well known that reinforcements or fillers, such as wood sawdust, clay, calcium carbonate, and wastes, can reduce costs while improving the mechanical properties of composites (Akil et al. 2014; Chow et al. 2007; Saeb and Moghri 2014). Natural fillers have several advantages over synthetic fillers (carbon and glass fibers): biodegradability, availability, recyclability, abundance, low density, impact resistance, and performance competitiveness (Guo et al. 2019; Percin and Uzun 2022). In addition, using natural fillers as reinforcement in plastic composites is one of today's fastest-growing industries. The rubber industries in southern Thailand are growing due to improving business conditions from 2021 to 2023 (Industry Outlook 2021–2023). The stronger demand for both types of rubber, namely rubber concentrated latex and cup lump rubber, has been increasing in many industries, such as health products, tire manufacturing, household items, and medical supplies (especially latex gloves) (Phakee and Boochathum 2015). Additionally, research supported by public and private sector funds will boost demand for rubber products for use by elderly persons.

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Presently, Thai rubber products are produced from rubber concentrated latex and cup lump rubber and sold to overseas markets for processing into downstream products. 86.7% of Thai rubber products are sold abroad. The most important export markets (by value) are China (36.6%), Malaysia (22.6%), the United States (6.5%), Japan (5.7%), and South Korea (3.4%). The remaining 13.3% of midstream production is consumed domestically (Industry Outlook 2021–2023; Samyn et al. 2020). Both types of Thai rubber are used extensively to produce the rubber latex glove for medical use, a processed agriculture product made from natural rubber. Latex gloves are elastic and flexible, available in powdered or powder-free form and for general-purpose examinations (Tian and Xu 2022; Samyn et al. 2020). The powder from rubber concentrated latex is also known as “sludge” and is a by-product and waste produced from the manufacturing process of concentrated latex and fresh latex pond (Homkhiew et al. 2018a).

The sludge waste is a semi-solid slurry of opaque white color. The chemical compositions of sludge waste are phosphorus pentoxide (P_2O_5), magnesium oxide (MgO), potassium oxide (K_2O), zinc oxide (ZnO), and a few other chemical compositions (Vichaphund et al. 2012; Homkhiew et al. 2018a). Generally, in the natural rubber latex manufacturing process and rubber-making industries, a large amount of sludge waste is generated at different stages. Most sludge is discharged without further treatment by dumping them at landfills or discharging in the waste treatment pond (Haftkhani et al. 2011). However, previous industrial studies have found that the sludge wastes have high phosphorus and magnesium oxide contents that has beneficial uses, such as organic fertilizer and reinforcement in plastic composites (Homkhiew et al. 2018a; Jose-Trujillo et al. 2018; Nosbi et al. 2010). There is interest in using sludge wastes as reinforcing filler in plastic composites to produce new composite materials because of their potential to improve mechanical, physical, and thermal properties while reducing costs compared to other types of reinforcing filler materials (Zamri et al. 2011; Zhou et al. 2019). Moreover, composites reinforced with latex sludge waste result in lower density, tougher, and stronger composites that are non-abrasive to the cutting equipment or machines compared to commonly used mineral fillers (Phakee and Boochathum 2015; Guo et al. 2019).

The use of composites reinforced with natural fillers has grown rapidly in recent years. The demand for this material is due to its high stiffness-to-weight ratio and superior properties over wood and plastics alone. More importantly, it costs less than conventional reinforcements, easily adapts to existing plastic processing techniques, is eco-friendly, and is easier to recycle (Phiri et al. 2020; Saetun et al. 2015). Generally, the composites are materials made from plastic as a matrix and fibers or natural fillers as reinforcement (Wu et al. 2022).

Among many types of plastic used as matrix, polypropylene (PP) is the most commonly used as plastic base for composites compared to polystyrene (PS), polyethylene (PE), and polyvinyl chloride (PVC) (Akil et al. 2014; Homkhiew et al. 2022; Khamtree et al. 2020; Ramli et al. 2018). PP is a good matrix material because it has a unique blend of qualities not found in other materials. The advantages of PP are that it is a relatively inexpensive material, has high flexural strength, a low coefficient of friction, is moisture resistant, has good chemical resistance, fatigue resistance, good impact strength, resistance to electricity and a good electrical insulator. Additionally, composite performance depends on other compositions, such as additive or coupling agent that improve the composite materials' mechanical, physical, and thermal properties (Xu and Li 2012; Saeb and Moghri 2014).

The dimensional stability of these composites tends to be greater than traditional wood and plastic products because it involves many components, such as plastic and reinforcing fillers or additives (Phakee and Boochathum 2015; Daly et al. 2007). This stability renders them suitable for application in end uses, where dimensional stability is a prerequisite for composite materials. Composite production and research focus on a few selected reinforcing fillers (Saha et al. 2021). Although the performance of these materials in service is widely known, there is little information about how the composites' mechanical and physical properties affect the environment (Guo et al. 2019; Akil et al. 2014). There are restrictions on the use of composites. The effects of water absorption on composite materials' properties need further study (Sahu and Gupta 2020; Zamri et al. 2011). The hygroscopic nature of wood or fillers adversely affects the composite's performance when exposed to high moisture content, which affects their mechanical and physical properties (Prabhu et al. 2022).

After immersing composites under different water conditions, we aimed to evaluate and compare the deterioration of the mechanical properties of rubberwood-latex sludge flour-reinforced polypropylene composites. The effects of sludge wastes on the composites' properties were also investigated. The morphological and visual surface were observed and analyzed using Field Emission Scanning Electron Microscopy (FE-SEM) and optical microscopy. New information benefits the construction and building of WPC products, which are often applied by immersing them in water or high humidity. Moreover, using latex sludge wastes as fillers in WPCs is a promising way to decrease the amount of waste in landfills.

2 Materials and methods

2.1 Materials

IRPC Public Company Limited (Rayong, Thailand) supplied the PP pellets as a matrix in the composites under the trade

name 1100NK with a melt flow index of 11 g/10 min at 230 °C. Plan Creations Company Limited (Trang, Thailand) supplied the rubberwood flour (RWF). Before composite mixing, the RWF were sieved through a mesh of size 40 and dried in an oven at 110 °C for 24 h to reduce the moisture content. The chemical composition of RWF is cellulose 39%, hemicelluloses and cell wall 29%, lignin 28%, and ash 4% (Petchpradab et al. 2009). Two sludge wastes were collected from two manufacturing processes, i.e., concentrated latex processing (CLSF) and fresh latex pond (FLSF), from the rubber latex industry in Southern Thailand and used as reinforcement in hybrid composite materials (Songkhla, Thailand). Before the composites mixing, both latex sludge wastes were dried in an oven at 120 °C for 48 h to minimize the moisture content. The chemical compositions of the latex sludge wastes, namely CLSF and FLSF, are displayed in Table 1. Maleic anhydride grafted polypropylene (MAPP) with 8–10% of maleic anhydride ($M_w = 9100$ and $M_n = 3900$) was purchased from Sigma-Aldrich (Missouri, USA), and used as a coupling agent to improve the interfacial bonding between matrix and reinforcement in composites.

2.2 Composites processing

Composite production involves two stages. Initially, the raw materials (PP, MAPP pellets, RWF as reinforcement, and CLSF or FLSF as filler) were blended using a twin-screw extruder, Model CTE-D25L40 from Chareon Tut Co., Ltd. (Samutprakarn, Thailand). The temperatures of the extruder were within the range of 170, 175, 175, 180, 180, 185, and 190 °C, which was controlled from the feeding to the die zone. The screw rotation speed was set to 50 rpm. The extruded strands were passed through a cutting machine to make composite pellets. Table 2 shows the composite formulations. Afterwards, the mixed composite pellets were dried in an oven at 110 °C for 8 h to reduce moisture content. The composite pellets of each formulation were molded using a compression molding machine into composite panels with dimensions of 200 mm (width) × 250 mm (length) × 4.8 mm (thickness). The temperature profile for press plates was fixed to 190 °C under the maximum pressure of 1000 psi (6.89 MPa) for 15 min, with pre-heating for 5 min and compressing for 10 min. Finally, the composite panels were prepared as specimens into specific dimensions for mechanical and physical properties test per the American Society for Testing and Materials (ASTM) standard.

Table 1 Chemical composition of latex sludge

Type of latex sludge	P ₂ O ₅ *	MgO*	K ₂ O*	ZnO*	CaO*	SO ₃ *	SiO ₂ *	Al ₂ O ₃ *	Fe ₂ O ₃ *	Rb**
FLSF (wt%)	36.07	20.43	1.88	1.16	0.45	0.31	0.21	0.08	0.05	0.10
CLSF (wt%)	33.44	18.21	1.66	1.07	0.40	0.30	0.28	0.08	0.04	0.06

Note: FLSF: Latex sludge flour from fresh latex pond; CLSF: Latex sludge flour from concentrated latex processing; Source of *Homkhiew et al. (2023) and **Siriwong et al. (2009)

Table 2 Formulation of the composites in the experiment

Composite sample code	Composition (wt%)				
	PP	RWF	FLSF	CLSF	MAPP
PP100	100	–	–	–	–
P46R50	46	50	–	–	4
P46R25F25	46	25	25	–	4
P46R25C25	46	25	–	25	4

Note: PP: Polypropylene; FLSF: Latex sludge flour from fresh latex pond; CLSF: Latex sludge flour from concentrated latex processing; MAPP: Maleic anhydride-grafted-polypropylene; wt%: Percent by weight

2.3 Water absorption testing

Long-term water absorption (WA) and thickness swelling (TS) measurements of the composites were performed as per the ASTM standards (ASTM D570). The samples were immersed in different water conditions, namely distilled water (D), Gulf of Thailand (G), and Andaman Sea (A). Water was collected from the Gulf of Thailand from Chalatat beach (Muang District, Songkhla, Thailand) and the Andaman Sea from Radchamongkol beach (Sikao District, Trang, Thailand). Table 3 lists the characteristics of the waters used in the experiment. Five specimens of each formulation and condition were cut from the panels with a dimension of 30 mm (width) × 30 mm (length) × 4.8 mm (thickness) and dried in an oven at a temperature of 50 °C for 24 h to minimize the moisture content. Prior to the WA and TS tests, the weight and thickness of the composite samples were measured to a precision of 0.001 g and 0.001 mm, respectively. Then, the specimens were immersed in water at room temperature for 8 weeks. The composite samples were removed from the waters every week, excess water on the surface was carefully blotted, and measured immediately. WA and TS percentages were calculated using Eqs. (1) and (2):

$$WA_t(\%) = \frac{W_t - W_0}{W_0} \times 100 \quad (1)$$

where WA_t is the water absorption at time t , W_0 is the initial dry weight, and W_t is the soaked weight of the specimen at a given time t .

Table 3 Characteristics of the waters used in experiment

Source of water	Property of water				
	Density (g/cm ³)	Salinity (mg/L)	Sodium (mg/L)	Acid-Alkaline (pH)	Total Suspended Solids (mg/L)
Distilled water (D)	0.995	10	121	7.1	2.4
Gulf of Thailand (G)	1.026	46,640	11,810	7.0	16.8
Andaman sea (A)	1.028	47,550	12,900	6.9	3.1

$$TS_t(\%) = \frac{T_t - T_0}{T_0} \times 100 \quad (2)$$

where TS_t is the thickness swelling at any time t , T_0 is the initial dry thickness, and T_t is the soaked thickness of the specimen at a given time t .

2.4 Mechanical characterizations

2.4.1 Flexural test

The flexural properties, namely modulus of rupture (MOR), modulus of elasticity (MOE), and maximum flexural strain of the composite samples were determined as per ASTM standard (ASTM D790) with a computer-controlled Universal Testing Machine (Model NRI-TS500-50 from Narin Instrument Co., Ltd., Samut Prakan, Thailand). The test on the composite samples was set at a cross-head speed of 2 mm/min and using a span of 80 mm, respectively. The rectangular specimens had a dimension of 13 mm (width) × 100 mm (length) × 4.8 mm (thickness) using five replications of each formulation and condition. The depth and width of the specimen were measured at the center of the support span and the flexural test was evaluated at room temperature (25 °C).

2.4.2 Screw withdrawal test

The composite samples' screw withdrawal strength (SWS) was measured as per ASTM standard (ASTM D1037) using the computer-controlled Universal Testing Machine, the same as the flexural test. The cross-head speed of screw withdrawal was set to 1.5 mm/min. The rectangular dimension of the composite specimens was 50 mm (width) × 50 mm (length) × 4.8 mm (thickness). The wood screws with a diameter of 4.18 mm and threaded length of 50 mm were driven through their faces. The screws were embedded before immersing in the water. All the screw withdrawal strengths were calculated using Eq. (3):

$$\text{SWS (MPa)} = \frac{P_{\max}}{dl_p} \quad (3)$$

where P_{\max} is the maximum load (N) required to withdraw a screw from the specimen, d is the diameter (mm) of a wood

screw, and l_p is the depth (mm) of the penetrated screw in the specimen.

2.4.3 Hardness test

The composites' hardness was measured as per ASTM standard (ASTM D2240) using Shore D Durometer scales (Model GS-702G from Teclock Corporation, Nagano, Japan). The composite specimens were measured with a nominal dimension of 50 mm (width) × 50 mm (length) × 4.8 mm (thickness) at room temperature (25 °C) with five replications for each formulation and condition. The specimens were placed on a horizontal and hard surface with the durometer held vertically.

The flexure, screw withdrawal, and hardness measurements were repeated at 1-week intervals for otherwise continuously immersed samples. The mechanical properties degradation was determined over eight weeks, when the samples were saturated with water and no longer absorbing moisture.

2.5 Analytical methods

2.5.1 Morphological and visual surface analysis

The Field Emission Scanning Electron Microscope with an FEI Apreo microscope (FEI Company, Oregon, USA) was used to observe the voids, interfacial bonding, and dispersion of the RWF and latex sludges (CLSF and FLSF) in the polymer matrix. Prior to the FE-SEM observation, the cross-section surface area was gold-coated using a sputter coater to prevent electron charging during imaging. FE-SEM images were taken at an accelerating voltage of 20 kV with a magnification of 1000×. In addition, optical microscopy (Zeiss Axioskop, Oberkochen, Germany) was used to observe the change on the composite surfaces after immersing them for 4 and 8 weeks.

2.5.2 Statistical analysis

A statistical analysis of the experiment results was performed. Significant changes in mechanical properties were analyzed for an immersion period between 1 and 8 weeks

using a two-sample *t*-test. The effect of different water conditions was evaluated using analysis of variance (ANOVA) and Tukey's test, and a 5% significance level ($\alpha=0.05$) was used for the statistical analysis.

3 Results and discussion

3.1 Mechanical properties of the composites

Figure 1 shows the MOR, screw withdrawal strength, and hardness of rubberwood-latex sludge flour-reinforced polypropylene composites. The results show that the flexural strength of P46R50 (41.6 MPa) was similar to that of P46R25F25 (41.5 MPa). It was also greater than the value of P46R25C25 (38.1 MPa). Adding 25 wt% of FLSF gives it a flexural strength similar to 50 wt% of RWF. Further, the composites reinforced with latex sludge waste from fresh latex pond had higher experimental flexural strength value than composites enforced using concentrated latex processing. This is because latex sludge flour from fresh latex pond absorbed large amounts of energy and good stress transfer during the deformation process (Phiri et al. 2020). In addition, it was found that the result of screw withdrawal strength property on P46R50 had the highest value (40.9 MPa), followed by P46R25F25 (33.2 MPa) and P46R25C25 (29.9 MPa), respectively. These results showed that the reinforcement with RWF had better screw withdrawal strength than the composites with latex sludge flour, both of fresh latex pond and concentrated latex processing. The flexural strength and screw withdrawal strength of the composites with 50 wt% RWF were higher than that of latex sludge flour. This result of the experiment agrees with Phakee and Boochathum (2015), who reported that

the addition of rubber markedly decreases flexural strength. Moreover, wood flour in composites has greater mechanical strength due to its rigid structure compared to flexible rubber molecules.

Generally, rubber comprises long flexible chain-link molecules with high elasticity properties (Zhao et al. 2010). For the hardness test, it was found that P46R25F25 (80.1 shore D) and P46R25C25 (80.0 shore D) gave similar results, which were greater than P46R50 (78.7 shore D). The latex sludge waste reinforcement improved the hardness property of polypropylene composites compared with RWF. A possible reason for this observation is that latex sludge waste consists of main substances, such as magnesium oxide, phosphorus pentoxide, potassium oxide, and zinc oxide, which are more rigid than polypropylene matrix, making the composites more rigid as well.

Figure 2 shows the MOE and maximum flexural strain of rubberwood-latex sludge flour-reinforced polypropylene composites. The results showed that all proportions of the composites gave MOE that was drastically higher than PP because natural fiber and filler are naturally more rigid than polymer matrix. Therefore, the composites' modulus increased with the filler content in plastic composites (Khamtree et al. 2020). It agrees with Phakee and Boochathum (2015), who reported that Para rubber wood sawdust slightly increased the flexural modulus of thermoplastic elastomer composites. P46R50 gave the maximum value (3.40 GPa), followed by P46R25F25 (3.20 GPa) and P46R25C25 (3.06 GPa). Adding latex sludge reinforcement reduced the composites' MOE, which also agrees with Tian and Xu (2022), who concluded that rubber filler composites had lower modulus. Additionally, the maximum flexural strain values of P46R50, P46R25F25, and P46R25C25 were 1.55%, 1.57%, and 1.59%, respectively. These values were

Fig. 1 MOR, screw withdrawal strength, and hardness of rubberwood–latex sludge flour reinforced polypropylene composites

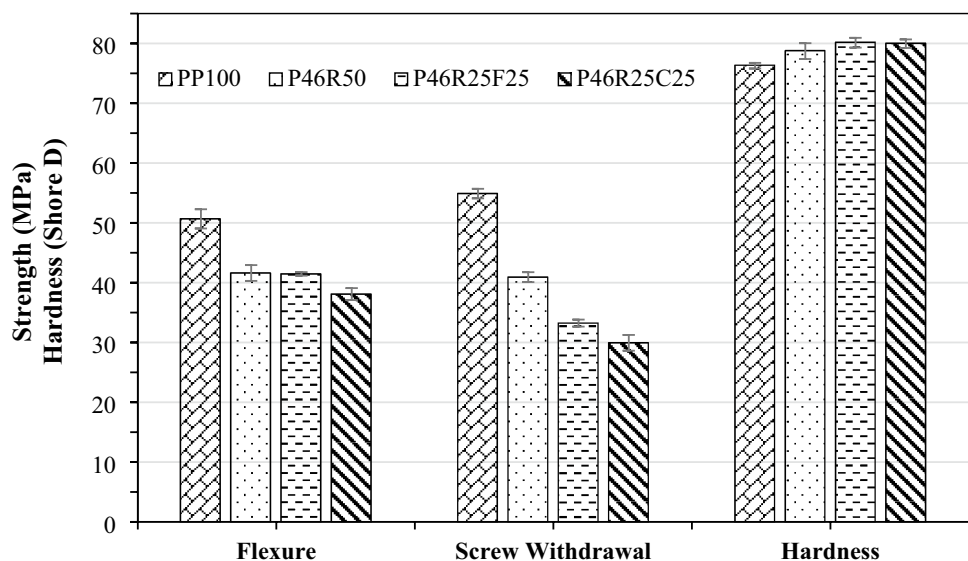
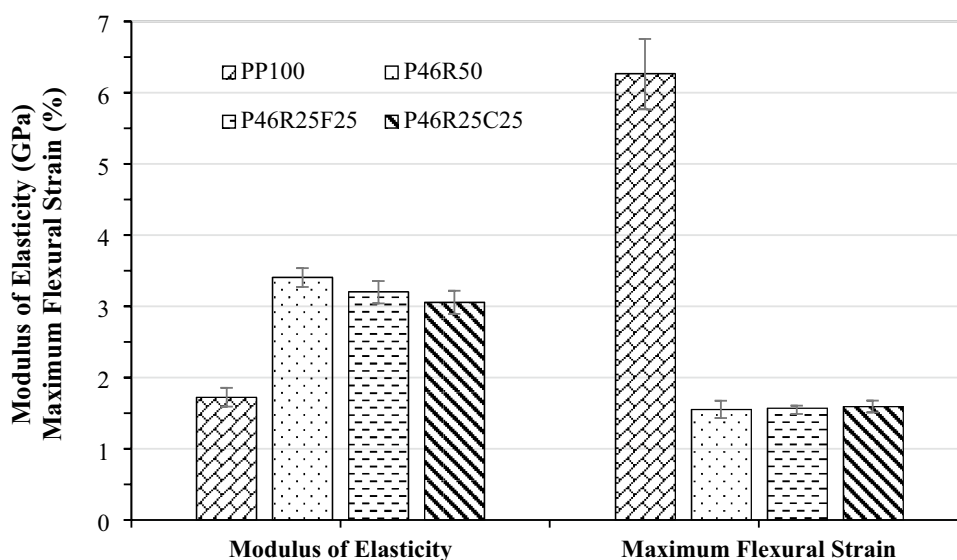


Fig. 2 MOE and maximum flexural strain of rubberwood–latex sludge flour reinforced polypropylene composites



very similar and lower than PP100 (6.26%). RWF and latex sludge waste reinforcement gave lower flexural strain than the polymer matrix.

3.2 Morphology of the composites

The interfacial adhesion in the composites was investigated using FE-SEM. Figure 3(a), (b), and (c) show the PP composites reinforced with 50 wt% RWF, 25 wt% RWF and 25 wt% FLSF, and 25 wt% RWF and 25 wt% CLSF, respectively. Figure 3(a) shows the composites consisting of RWF and polypropylene. It also displayed strong interfacial adhesion between wood flour and polymer matrix. Figure 3(b) and (c) show the latex sludge waste types, LSF and CLSF, used to reinforce the composites. Generally, latex sludge waste is more dispersed and homogeneous (Homkhiew et al. 2018a).

Moreover, the rubber surface seemed clean, intact and free from adhering wood flour and matrix (Zhou et al. 2019). FE-SEM was also employed to investigate the composite bonding and it was observed that FLSF sludge made the composites more compatible, which was explained by the smaller pores or gaps, whereas there was filler pulled out at CLSF composites. It could be also noticed that the particles of FLSF are smaller than CLSF, which could increase the interfacial bonding between matrix and filler. These results are in good agreement with Homkhiew et al. (2023), who revealed that 90% of the FLSF and CLSF particles is below 227.60 μm and 364.20 μm , respectively. This indicates a more compatible interface interaction between wood filler and polymer matrix, influencing the composites' mechanical properties (Elamin et al. 2020; Akil et al. 2009). Therefore, the interfacial bonding between filler and matrix affected their mechanical properties due to loading transfer to the

polymer matrix (Mungamurugu et al. 2017; Saha et al. 2021).

3.3 Long-term water absorption and thickness swelling behavior

The water absorption (WA) and thickness swelling (TS) behaviors of the composites related to three different aqueous environments were investigated in this study. Table 3 shows the characteristics of the water sources: density, salinity, sodium, acid-alkaline, and total suspended solids. Generally, the composites' water absorption behavior is one of the main concerns in structural applications of composites, particularly for natural fiber-reinforced composites (Nosbi et al. 2010; Guo et al. 2019; Candelier et al. 2019). It might cause polymer matrix hydrolysis because the fiber and matrix interface is degraded (Jose-Trujillo et al. 2018; Davies et al. 2022; Mourad et al. 2019). Figures 4 and 5 show the WA and TS behaviors of rubberwood-latex sludge flour-reinforced polypropylene composites, immersed in the three types of water. The results showed that the composites' WA and TS increased rapidly in the initial period and then slowly until they reached saturation point. All composite proportions (P46R50, P46R25F25, and P46R25C25) had the lowest WA and TS for all water sources. As a result, sample P46R25C25 had the maximum WA values at 6.38%, 7.10%, and 8.65% for the Andaman Sea, Gulf of Thailand, and distilled water, respectively. It was observed that the composite components were affected by different water absorption. In addition, the composites with the best water absorption resistance were immersed in water from the Andaman Sea due to the presence of large salt molecules in the seawater (notably sodium chloride), which slowed down the diffusion process in the composite structure, resulting in lower

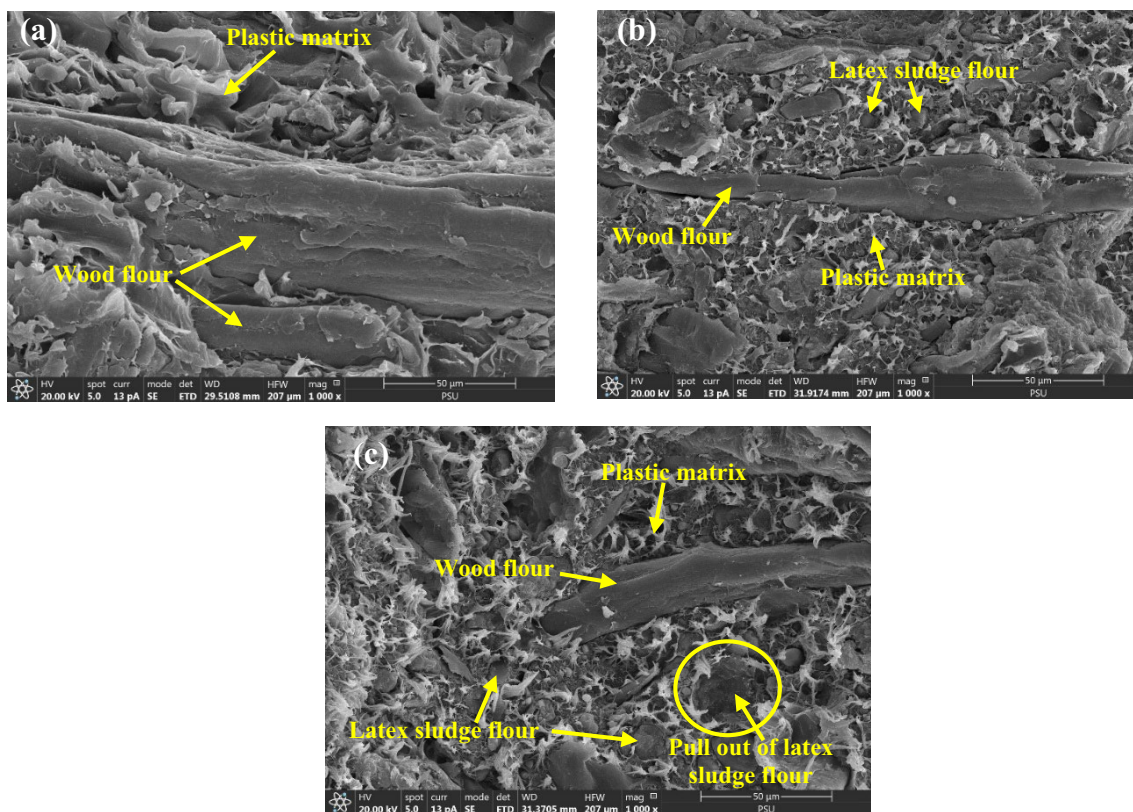
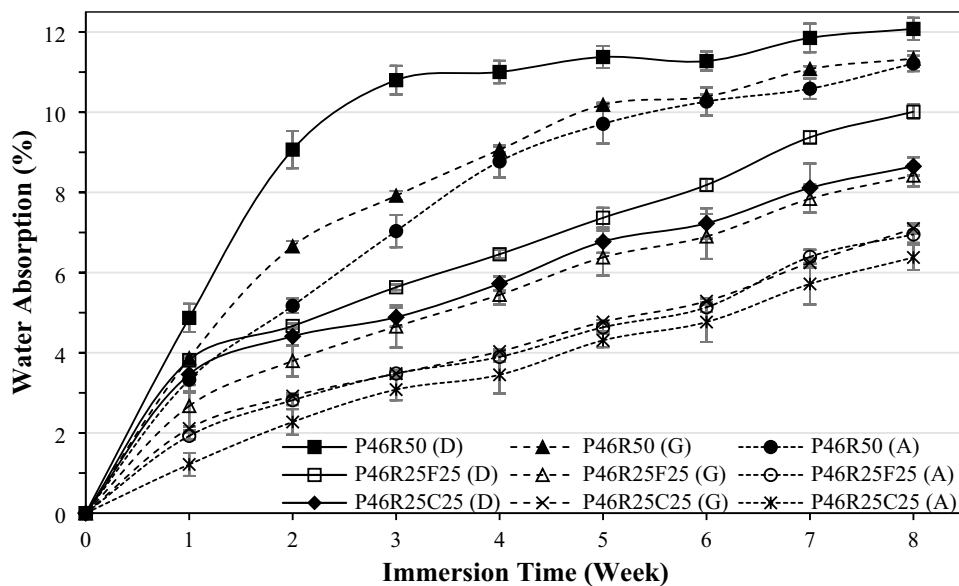


Fig. 3 FE-SEM images (magnification 1000×) of (a) PP composites reinforced with RWF 50 wt%, (b) PP composites reinforced with RWF 25 wt% and FLSF 25 wt%, and (c) PP composites reinforced with RWF 25 wt% and CLSF 25 wt%

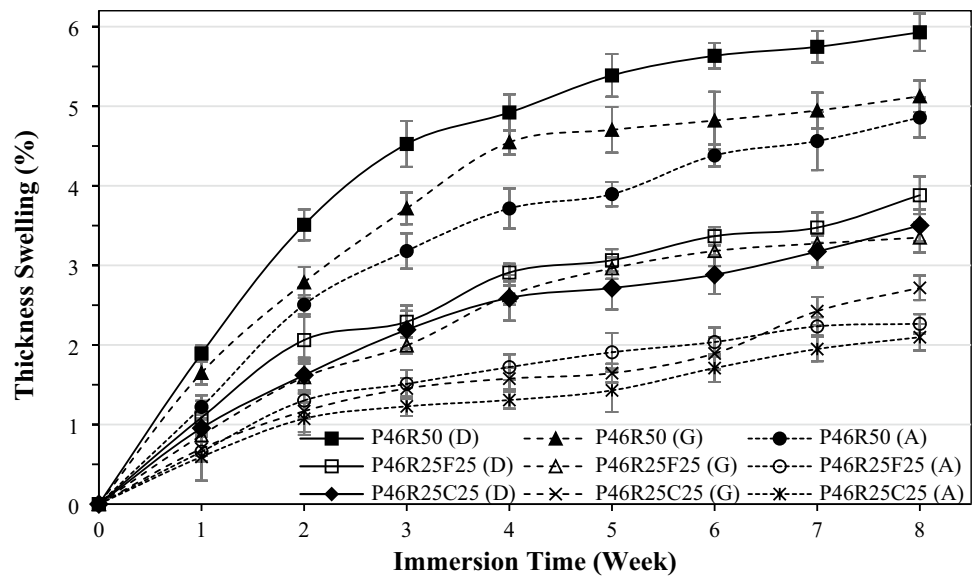
Fig. 4 Long-term water absorption behavior of rubberwood–latex sludge flour reinforced polypropylene composites



absorption (Zamri et al. 2011; Homkhiew et al. 2016). These results were as expected because the Andaman Sea has a higher sodium level (12,900 mg/L) than the Gulf of Thailand and distilled water (Table 3). Samples P46R50, P46R25F25, and P46R25C25, which were immersed in water from the

Andaman Sea, had the maximum WA values of 11.22%, 6.95%, and 6.38%, respectively. The maximum TS values were 4.86%, 2.27% and 2.10%, respectively. The WA and TS values for composites immersed in distilled water and water from the Gulf of Thailand were similar to those immersed in

Fig. 5 Thickness swelling as a function of water immersion time for rubberwood–latex sludge flour reinforced polypropylene composites



water from the Andaman Sea. Sample P46R50 comprising 50% RWF contained hydrophilic components, like cellulose and hemicellulose, with many hydroxyl groups (Phiri et al. 2020), which reduced interfacial adhesion of matrix and fiber, leading to fiber swelling. Water penetrated easily into the composites at the poor interfaces between the polymer matrix and fibers (Homkhiew et al. 2016), and the water molecules decreased the interfacial bonding strength (Law and Ishak 2011).

Additionally, composites P46R25C25 and P46R25F25 reinforced with latex sludge waste as a constituent resulted in lower WA and TS. A possible explanation is the rubber powder improved the composites' water resistance, leading to its hydrophobic property. Moreover, the rubber powder might limit water molecules' access to celluloses, and the number of hydroxyl groups was reduced by increasing the amount of rubber powder, thus decreasing TS (Xu and Li 2012; Saetun et al. 2015).

FE-SEM images and optical microscopy images supported the experimental results. The composites containing latex sludge waste in Fig. 3(b) and (c) are more compatible than the WPCs in Fig. 3(a). Therefore, the PP composites with 50% RWF allow easier water penetration into the cellulose. In fact, more porosities and poorer interfacial bonding provide more water residence sites (Adhikary et al. 2008). Table 4 shows the optical microscopy images of the composite surfaces at different immersion stages. When comparing the immersed composites (P46R50) in different water conditions, the surface of P46R50 (A) immersed in the Andaman Sea water had very few traces of water infiltration. When the composites were immersed for 8 weeks, the traces of water infiltration were clearer than at 4 weeks.

Further, when samples P46R50, P46R25C25, and P46R25F25 were immersed in distilled water (D), it showed

clear water infiltration into the composites' material texture and swelling caused by the fibers. This corresponds to the results of the P46R50 experiment, where WA had the highest value of 12.08% and TS was 5.93%. Therefore, we expect the rubberwood-latex sludge flour-reinforced polypropylene composites to be suitable for construction or engineering applications that require water resistance, especially in the Andaman Sea.

3.4 Deterioration behavior in the mechanical properties under water immersion

3.4.1 Flexural properties

Generally, WPCs show high moisture absorption when exposed to a humid environment or immersed in water due to hydroxyl groups in the cellulosic fibers (Sahu and Gupta 2020). Figures 6, 7 and 8 show MOR, MOE, and maximum flexural strain deterioration of the rubberwood-latex sludge flour-reinforced polypropylene composites, respectively. The composite samples were immersed in the three water sources for 8 weeks in this experiment.

Figure 6 shows that MOR deterioration continued to decrease until week 8. Alomayri et al. (2014) reported a similar observation, where the composites' flexural strength decreased markedly because of water absorption. Water absorption degraded the fiber and matrix interface region, transferring stress from the matrix to the fiber, resulting in low flexural strength (Alamri and Low 2013). Comparing P46R50, P46R25F25, and P46R25C25 immersed in the three water sources, P46R25C25 showed the highest durability in MOR, followed by P46R25F25 and P46R50. Because latex sludge flour had smaller particles that could insert well into the matrix, causing high compatibility. Therefore, the

Table 4 Optical microscopy images of the composite surfaces at different immersion stages

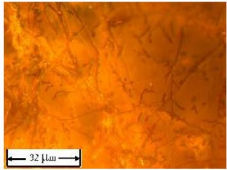
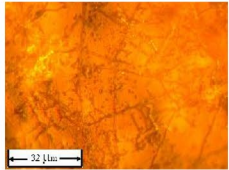
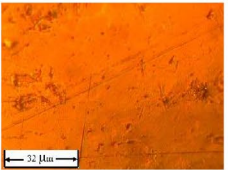
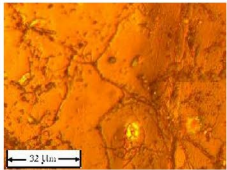
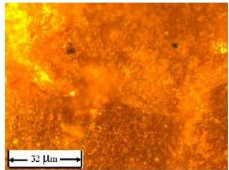
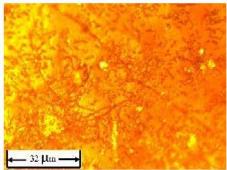
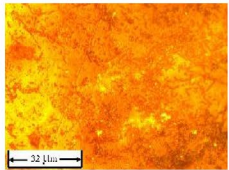
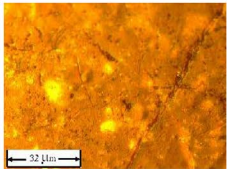
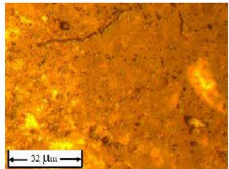
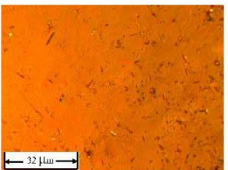
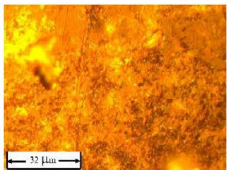
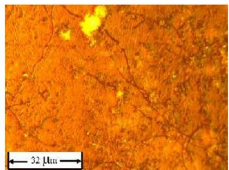
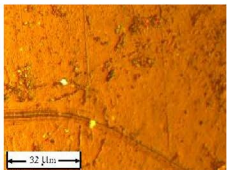
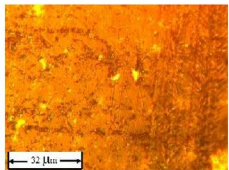
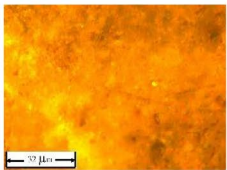
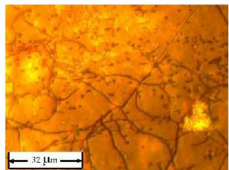
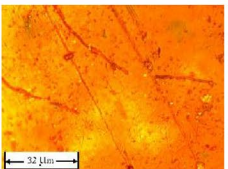
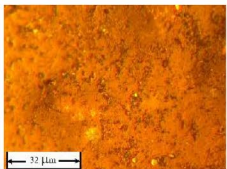
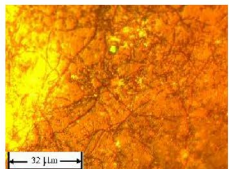
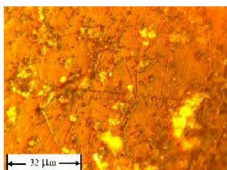
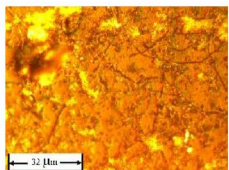
Condition	Un-immersed	Immersed for 4 weeks	Immersed for 8 weeks
P46R50 (D)			
P46R50 (G)			
P46R50 (A)			
P46R25F25 (D)			
P46R25F25 (G)			
P46R25F25 (A)			
P46R25C25 (D)			
P46R25C25 (G)			
P46R25C25 (A)			

Fig. 6 Deterioration in MOR as a function of water immersion time for rubberwood–latex sludge flour reinforced polypropylene composites

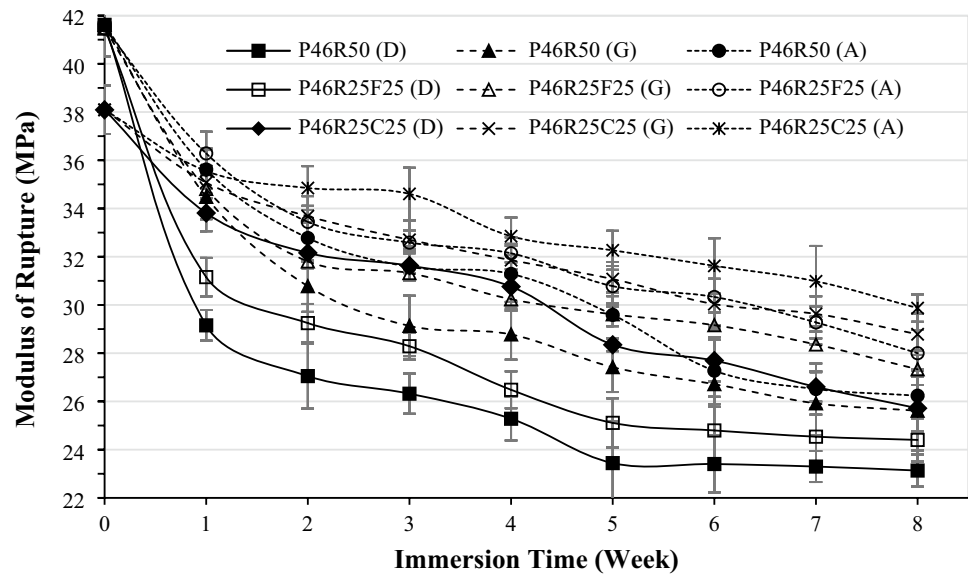
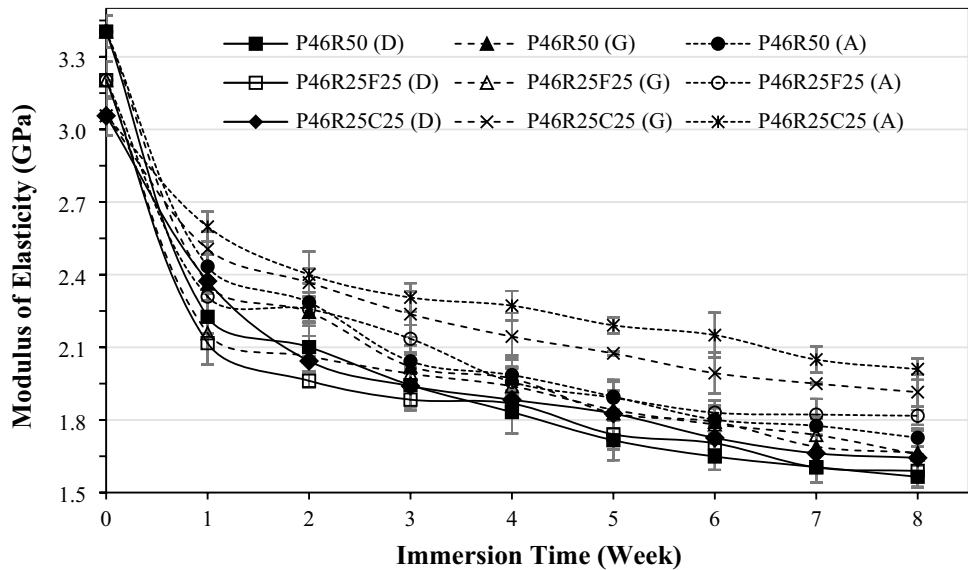


Fig. 7 Deterioration in MOE as a function of water immersion time for rubberwood–latex sludge flour reinforced polypropylene composites



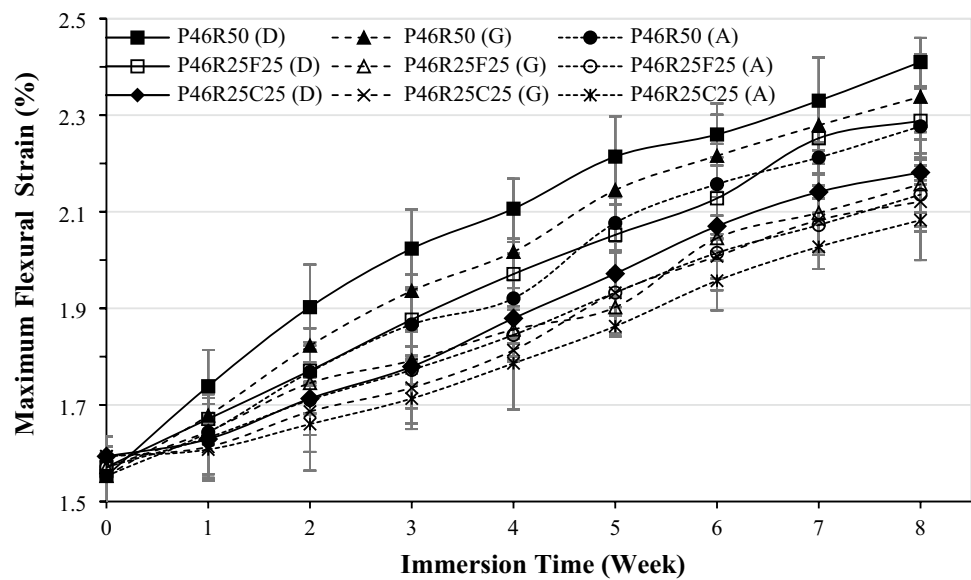
composites made from latex sludge flour enhanced water resistance compared to unfilled composites. Further, comparing composites P46R25C25 immersed in different water sources revealed the least MOR deterioration in the Andaman Sea, followed by the Gulf of Thailand and distilled water. This is because sea water slowed down water diffusion into the matrix of composite materials (Daly et al. 2007; Homkhiew et al. 2022). Therefore, composites tested in the Andaman Sea and Gulf of Thailand water exhibited lower strength degradation than distilled water. The same was observed for P46R50 and P46R25F25 composites.

Figure 7 also shows that the composites' MOE deteriorated continuously the longer it was immersed in water. This result agrees with Alomayri et al. (2014), who reported a considerable decrease in the flexural modulus of the wet

composite samples compared to dry samples. The MOE of P46R25C25 composites decreased slightly after water absorption compared to P46R25F25 and P46R50 because water diffusion weakened the interfacial bonding between the fillers and polymer matrix (Guo et al. 2019). As a result, the composites immersed in Andaman Sea water showed a slight decrease in MOE. After composite P46R25C25 was immersed in Andaman Sea water for 8 weeks, its MOE was 2.01 GPa. Its MOE, when immersed in the Gulf of Thailand and distilled water, was 1.91 GPa and 1.64 GPa, respectively.

Figure 8 illustrates that as immersion time increased, the composites' maximum flexural strain increased for all water sources. This finding agrees with the finding of Zamri et al. (2011) that there is a trend of an increased maximum flexural strain of jute/glass fiber-reinforced polyester composites.

Fig. 8 Maximum flexural strain of rubberwood–latex sludge flour reinforced polypropylene composites after immersing under different water conditions



This result is because the plasticization effect in the wood fiber and matrix interface increased ductility from water immersion (Chow et al. 2007). Additionally, the plasticizer used to make composites more flexible will cause the composites' cellulose content to decrease due to water absorption (Akil et al. 2014). The results revealed the lowest degradation for composites in the Andaman Sea water, followed by the Gulf of Thailand and distilled water, respectively. For example, composites P46R25C25 showed the lowest degradation in the maximum flexural strain in Andaman Sea water (30.7%), followed by Gulf of Thailand water (33.1%) and distilled water (36.9%).

Therefore, the results showed that composites P46R25C25 containing 25 wt% RWF and 25 wt% latex sludge flour from concentrated latex processing immersed in the Andaman Sea water gave the lowest MOR, MOE, and maximum flexural strain deterioration. This implies that adding the latex sludge flour from concentrated latex processing into the composites could efficiently prevent composites deterioration.

3.4.2 Screw withdrawal property

Screws are WPCs' most common joint fastener, particularly in decking, outdoor furniture, etc. (Haftkhani et al. 2011). The screw withdrawal strength depends on the wood type, material density, wood moisture content, and screw properties (Percin and Uzun 2022). Figure 9 presents the deterioration results in screw withdrawal strength for composites immersed for 8 weeks in the three water sources. The composites screw withdrawal strength decreased continuously with increasing immersion time for all samples.

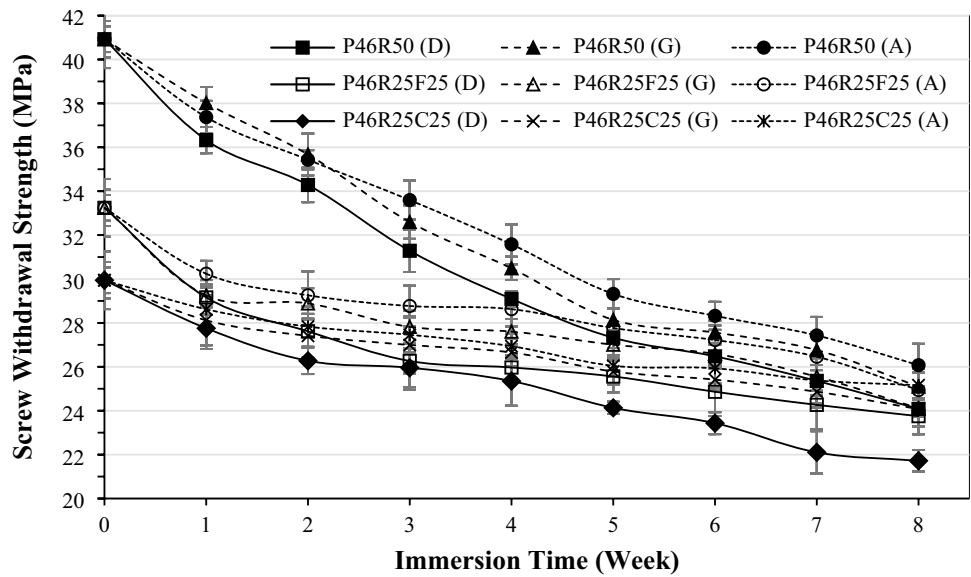
When testing the composites' screw withdrawal strength deterioration, P46R50 had a higher deterioration slope than

P46R25F25 and P46R25C25. After immersing P46R50 composites in the Andaman Sea water for 8 weeks, its ability to hold screws was worse than P46R25F25 and P46R25C25: their loss of screw withdrawal strengths was 26.1%, 24.9%, and 25.2%, respectively. Therefore, the composites with 25 wt% latex sludge and 25 wt% RWF were more resistant to deterioration than those with 50 wt% RWF, which could be due to the thermoplastic matrix's ability to encapsulate the screw thread, allowing continuous load transfer along the threaded length (Haftkhani et al. 2011). On the other hand, this study revealed that filling the latex sludge positively influenced the composites' screw withdrawal strength. This result is because latex sludge absorbs less water than wood, so the composites with latex sludge flour were stronger. Moreover, the composites immersed in the Andaman Sea water were better at resisting deterioration than composites immersed in the Gulf of Thailand and distilled water.

3.4.3 Hardness property

Figure 10 shows the effect of different water types on the hardness deterioration of composites. Composites' hardness decreased as immersion time increased due to the weakening of the interface between the polymer matrix and fiber caused by water absorption (Prabhu et al. 2022). P46R25C25, which contained 25% RWF and 25% latex sludge flour from concentrated latex processing, exhibited the distinguished hardness property. Moreover, it showed a maximum hardness of 80.0 Shore D before immersion in the Andaman Sea water. After 8 weeks of water immersion, the value decreased to 77.1 Shore D, even for composites with improved hardness after adding the latex sludge. This result agrees with Homkhiew et al. (2018b),

Fig. 9 Deterioration in screw withdrawal strength as a function of water immersion time for rubberwood–latex sludge flour reinforced polypropylene composites



who concluded that composites produced with lower rubber content had a much higher hardness value.

Moreover, composites immersed in the Andaman Sea water were more resistant to hardness deterioration than those immersed in the Gulf of Thailand and distilled water. After immersing for 8 weeks, P46R25C25 composites gave the highest hardness value when immersed in Andaman Sea water (77.1 Shore D), followed by Gulf of Thailand water (76.6 Shore D), and lastly distilled water (75.9 Shore D). We conclude that the latex sludge enhanced the composites' hardness compared to wood fiber, which could be because seawater's density and salinity influenced the composites' immersion value, making them less water-absorbent (Prabhu et al. 2022).

One-way ANOVA was performed to evaluate the effect of the water types on the composites' hardness after immersing for 1 week and 8 weeks, as shown in Table 5. The results indicated that after immersing for 1 week and 8 weeks, the water types had a statistically significant effect ($p < 0.05$) on all composites' hardness (P46R50, P46R25F25 and P46R25C25). Turkey's test (shown in Table 5) was also conducted to determine the effect of water types on hardness properties. The results showed that P46R50 composites immersed in distilled water (suffix a) had significantly lower hardness than the Gulf of Thailand water (suffix b) and the Andaman Sea water (suffix b). The hardness difference between P46R50 (suffix b) immersed in the Gulf of Thailand water and Andaman Sea water (suffix b) was insignificant. Then, a two-sample *t*-test was applied to analyze the effect

Fig. 10 Deterioration in hardness as a function of water immersion time for rubberwood–latex sludge flour reinforced polypropylene composites

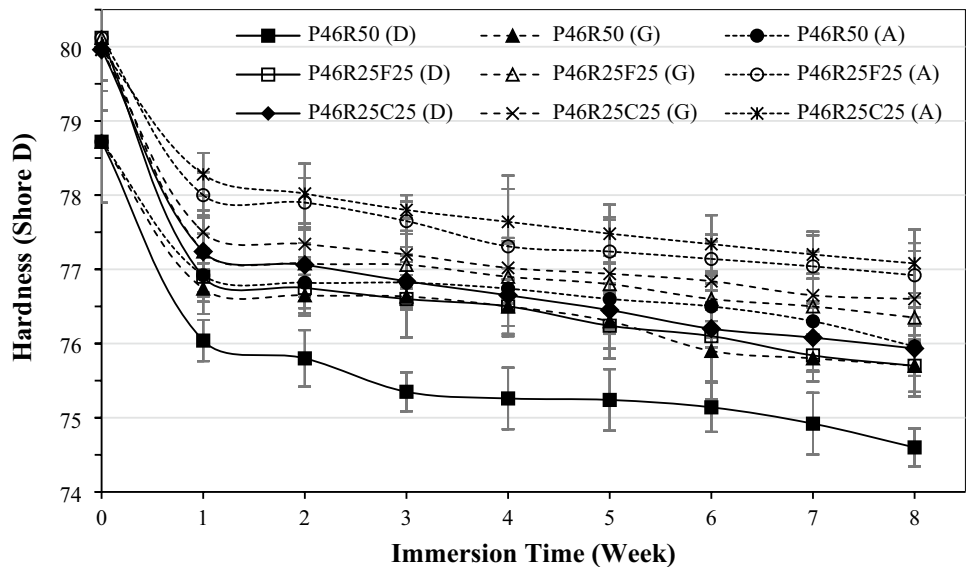


Table 5 Effects of different water conditions on mechanical properties of the composites

Condition	MOR (MPa)			MOE (GPa)			Maximum strain (%)			SWS (MPa)			Hardness (Shore D)		
	1 W	8 W	% loss	1 W	8 W	% loss	1 W	8 W	% loss	1 W	8 W	% loss	1 W	8 W	% loss
P46R50 (D)	29.2 ^{Aa}	23.1 ^{Ba}	79.9	2.23 ^{Aa}	1.57 ^{Ba}	117.5	1.74 ^A	2.41 ^{Ba}	55.2	36.3 ^{Aa}	24.1 ^{Ba}	69.9	76.0 ^{Aa}	74.6 ^{Ba}	5.52
P46R50 (G)	34.5 ^{Ab}	25.6 ^{Bb}	62.5	2.36 ^{Aab}	1.66 ^{Bab}	104.5	1.68 ^A	2.34 ^{Bab}	50.6	38.0 ^{Ab}	25.1 ^{Bab}	63.1	76.7 ^{Ab}	75.7 ^{Bb}	3.99
P46R50 (A)	35.6 ^{Ab}	26.2 ^{Bb}	58.6	2.43 ^{Ab}	1.73 ^{Bb}	97.2	1.64 ^A	2.28 ^{Bb}	46.6	37.4 ^{Aab}	26.1 ^{Bb}	56.9	76.9 ^{Ab}	76.0 ^{Bb}	3.63
<i>p-value</i>	0.000*	0.012*		0.032*	0.029*		0.210	0.041*		0.039*	0.021*		0.039*	0.000*	
P46R25F25 (D)	31.2 ^{Aa}	24.4 ^{Ba}	69.9	2.12 ^{Aa}	1.59 ^{Ba}	101.5	1.67 ^A	2.29 ^{Ba}	45.6	29.2 ^A	23.8 ^B	39.9	76.9 ^{Aa}	75.7 ^{Ba}	5.84
P46R25F25 (G)	34.8 ^{Ab}	27.3 ^{Bb}	51.7	2.16 ^{Aa}	1.66 ^{Ba}	93.3	1.65 ^A	2.16 ^{Bb}	37.3	29.3 ^A	24.1 ^B	37.9	77.2 ^{Aab}	76.4 ^{Bab}	4.94
P46R25F25 (A)	36.3 ^{Ab}	28.0 ^{Bb}	48.1	2.31 ^{Ab}	1.82 ^{Bb}	76.3	1.63 ^A	2.14 ^{Bb}	35.9	30.2 ^A	24.9 ^B	33.3	78.0 ^{Ab}	76.9 ^{Bb}	4.16
<i>p-value</i>	0.018*	0.001*		0.027*	0.014*		0.306	0.025*		0.284	0.103		0.003*	0.027*	
P46R25C25 (D)	33.8 ^A	25.7 ^{Ba}	48.1	2.37 ^{Aa}	1.64 ^{Ba}	85.9	1.63 ^A	2.18 ^B	36.9	27.8 ^A	21.7 ^{Ba}	37.9	77.2 ^{Aa}	75.9 ^{Ba}	5.30
P46R25C25 (G)	35.1 ^A	28.8 ^{Bb}	32.3	2.51 ^{Ab}	1.91 ^{Bb}	59.6	1.61 ^A	2.12 ^B	33.1	28.1 ^A	24.1 ^{Bb}	24.4	77.5 ^{Aa}	76.6 ^{Bab}	4.39
P46R25C25 (A)	35.6 ^A	29.9 ^{Bb}	27.5	2.60 ^{Ab}	2.01 ^{Bb}	52.1	1.61 ^A	2.08 ^B	30.7	28.6 ^A	25.2 ^{Bb}	19.0	78.3 ^{Ab}	77.1 ^{Bb}	3.74
<i>p-value</i>	0.145	0.026*		0.008*	0.012*		0.413	0.264		0.318	0.001*		0.025*	0.014*	

Note: W: Week; MOR: Modulus of rupture; MOE: Modulus of elasticity; SWS: Screw withdrawal strength; *Water types affect significant at *p-value* < 0.05. Means within each column with the same superscripts a–b indicate a insignificant difference ($\alpha=0.05$) by Turkey's test. Different superscripts A–B of each property and formulation indicate significant difference ($\alpha=0.05$) between mechanical properties of the composites immersed for 1 week and 8 weeks; % loss was calculated at 8 weeks of immersion

of 1 week and 8 weeks water immersion. The results show that all mechanical properties changed significantly after a longer immersion. For example, P46R25F25 was significantly harder after 1 week (suffix A) of immersion than after 8 weeks of immersion (suffix B).

4 Conclusion

In this study, composites' physical and mechanical properties were tested after immersion in water from different sources, i.e., distilled water, the Gulf of Thailand, and the Andaman Sea. We draw the following conclusions based on the results obtained:

- FE-SEM micrographs revealed that the latex sludge flour was more homogenous and enhanced the interfacial adhesion between fiber and matrix of the composites.
- Adding the latex sludge flour at 25 wt% increased hardness but decreased MOR, MOE, and screw withdraw strength compared to RWF filler. On the other hand, it did not affect the composites' maximum flexural strain.
- Exposure to different water types for a long period reduced the composites' MOR, MOE, screw withdrawal strength, and hardness. When immersed in distilled water, the maximum percentage reduction was 79.9, 117.5, 69.9, and 5.84. However, latex sludge resisted deterioration due to lower water absorption and smaller particles, leading to composites homogenization, especially the latex sludge flour from concentrated latex processing.
- The composites immersed in the Andaman Sea water exhibited the lowest deterioration of all properties with a minimum percentage loss of 3.74 in hardness due to larger salt molecules in the seawater, which slowed down the diffusion process into the matrix of composite materials.

This study found that adding latex sludge flour to the composites made WPCs resistant to mechanical properties deterioration, making them suitable for applications in an environment that involves contact with seawater, especially the Andaman Sea.

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Author contributions S. Khamtree and C. Srivabut wrote the main manuscript, which S. Khamtree wrote in part of Results and Discussion, and C. Srivabut wrote in part of methodology. C. Homkhiew prepared raw materials, designed of experiment, analyzed the results of

experiment, wrote in part of Abstract and Conclusions. T. Ratanawilai prepared all of the Figures (Figures 1–10) and final proofreading for manuscript. S. Rawangwong prepared all of the Tables (Tables 1–5) and tested the mechanical and physical properties. All of the authors reviewed the manuscript.

Data availability The datasets generated during and/or analyzed during the current study are available from the corresponding author on reasonable request.

Declarations

Conflict of interest The authors declare that they have no competing interests.

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