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Comparative effects of different coastal weathering on the thermal, physical, and mechanical properties of rubberwood–latex sludge flour reinforced with polypropylene hybrid composites

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A R T I C L E I N F O

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Natural weathering
Statistics

A B S T R A C T

The influence of coastal weathering in tropical countries is a concern in terms of applications of wood-plastic composites (WPCs). Therefore, developing the WPCs into hybrid composites for increasing the resistance to the coastal climate needs further investigation. The current work studies the effects of different coastal climates (Gulf of Thailand and Andaman Sea), exposure times, and latex sludge contents on the properties of rubberwood-latex sludge flour reinforced with polypropylene hybrid composites. The hybrid composites were manufactured with a twin-screw extruder for mixing and a compression molding machine for forming. The results revealed that the hybrid composites weathered for 12 months significantly ($\alpha = 0.05$) decreased the modulus of rupture, modulus of elasticity, screw withdrawal strength, and hardness with a maximum reduction of 218.6%, 207.4%, 84.2%, and 11.4%, respectively. However, adding the latex sludge flour of about 25 wt% increased crystallinity degree and thermal stability as compared with the WPCs filling 50 wt% rubberwood flour. The hybrid composites weathered under the Andaman Sea exhibited less loss of all the mechanical properties than that weathered under the Gulf of Thailand. It is therefore suggested that the hybrid composites added to the latex sludge waste have the potential to be used to produce the engineering products that were applied under the coastal climates.

1. Introduction

Wood-plastic composites (WPCs) are composite materials consisting of plastic as a matrix, wood as filler, and other additives [1,2]. WPCs can be used for decorative purposes and as building materials in outdoor paving and wall panels [3]. Moreover, composites could also be used in doors, windows, furniture, deck floors, fences, and automotive industries [4,5]. The manufacturing sector of WPCs has grown rapidly in recent years. North America and China are the two largest producers, Europe being the third [6]. Especially, the global market of bio-composites is expected to reach an estimated $9.5 billion by 2027 [7]. Consequently, natural fiber-reinforced polymer composites have demonstrated great potential for many different applications in various industries [8]. Previously, researchers attempted to find eco-friendly alternative materials and develop composites reinforcing with more agents than one type, resulting in a positive hybrid effect. Hybrid polymer composites are materials made by combining two or more different types of reinforcements in a common matrix [9], which have been gaining popularity with each passing year resulting from environmental, economic, and societal [2]. Moreover, hybrid composites are widely used in engineering, medical, defense, and other industries [10]. However, there are studies on the fillers of hybrid composites to improve their properties. For example, Narupai et al. [11] studied the effect of reinforcing with silica and carbon black hybrid filler of natural rubber (NR) composites. The results found that the physical and mechanical properties of NR composites can be improved by using silica and carbon black hybrid filler. Hayajneh et al. [12] studied the natural waste fillers including lemon leaves and eggshells of the composites with

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polypropylene matrix. The results concluded that the eggshells composites had the best values for both tensile and flexural tests while lemon leaves composites had the lowest values. Shahabaz et al. [13] studied the influence of nano-fillers (Al_2O_3 and SiC) in enhancing hybrid composites. The results confirmed that adding nano-fillers produced superior tensile and hardness properties for carbon fiber-reinforced polymer composites. Phunpeng et al. [14] studied hybrid composite materials which combined carbon fiber/epoxy with waste graphite fillers. The results found that the hybrid composite materials provided higher flexural strength and strength-to-weight ratio than the pure aluminum alloy 6061. Moreover, there has been a growing interest in the use of material waste which is considered reinforcing in the WPCs. Alternate materials or partial replacements should be developed by recycling waste materials. For example, Kaho et al. [15] valorized expanded polystyrene (EPS) waste by developing composite materials from EPS waste and wood waste. Mrowka et al. [16] investigated the impact of wood waste (oak, hornbeam, beech, and spruce trees) on the mechanical and biological properties of silicone-based composites. Basalp et al. [17] studied the use of plastic and wood obtained from municipal bulky waste for industrial manufacturing of WPC products. Nukala et al. [18] developed the WPCs using recycled plastic waste generated from university laboratories and recycled wood waste from construction. Homkhiew et al. [19] evaluated the potential of ground rubber tires and rubberwood sawdust waste for developing wood-polymer composites. The results showed that the addition of wood waste enhanced the modulus of elasticity, compressive modulus, and hardness of the composites. In addition, the rubber industries generate a lot of sludge waste, it is a by-product of the manufacturing process of concentrated natural rubber latex [20]. The use of waste materials also reduces the problem of land-filling, environmental, and health concerns [21]. Sludge waste is incorporated to obtain a new composite material with reasonable mechanical and physical properties as well as low cost. There are a few studies related to sludge waste used as the reinforcement of WPCs and other materials. Homkhiew et al. [22] evaluated and compared the potential of rubberwood flour and sludge waste as reinforcement in recycled HDPE composites. It can be seen that the previous studies have focused on the effect of the reinforcing agents that enhanced the compatibility of the wood and polymer matrix resulting in improved properties of hybrid composites. Moreover, sludge waste has the potential to be a good filler. Therefore, the choice of wood waste and latex sludge waste reinforcements in WPCs is proving to be a good alternative in this work.

WPCs are mainly used in outdoor applications for building and construction purposes such as decking, siding, fencing, and cladding [23]. The outdoor performance of WPCs is particularly concerned with the resistance of coastal weathering in tropical countries. The rising sea levels, rising acidity of the sea, changing the intensity and frequency of storms, and warmer ocean temperatures of coastal weathering affected the properties of the WPCs [24]. In addition, natural weathering is generally done to determine the durability of the material in natural conditions and common aspects of natural weathering including heat, cold, moisture, sunlight, rain, and dew [5]. Fewer studies investigated the effects of natural weathering on hybrid composites or other materials. For example, Wei et al. [25] examined changes in wood/polymer composition, surface chemistry, and thermal properties of WPC decking with and without a co-extruded cap layer after 8–14 years of above-ground outdoor exposure at a sub-tropical site near Hilo, Hawaii. Mohammed et al. [26] concluded that the tensile strength of the composites starts to decrease after the first weathering month through to the weathering periods with a constant reduction of tensile. At the end of the weathering period, almost 85% of the composite's mechanical behavior is lost. Torun et al. [5] reported that the weathering exposure caused a slight decrease in the tensile and flexural strength of composites. Moreover, the polymer band intensities decreased due to the degradation, and the composites are in accordance with the surface roughness of composites after weathering. Ratanawilai et al. [27] reported that polystyrene and polypropylene (PP) are the best choices for applying wood-plastic composites which require resistance to high stresses and natural weathering. In addition, the hybrid composites made from thermoplastic matrix have shown improved mechanical properties [10]. Producing the material with a focus on improving aspects is very important in various applications [26]. This work has been carried out on material waste reinforced plastic composites, wherein PP was chosen as matrix material.

Therefore, the novelty of this work aimed to enhance the performance of the hybrid PP composites reinforced by the addition of rubberwood flour and latex sludge flour from concentrated latex processing and fresh latex ponds. Moreover, the comparison in degradation of the hybrid composites under weathering conditions of different coastal climates between the Gulf of Thailand and the Andaman Sea in the South of Thailand was evaluated. Changes in surface color, thermal properties, and mechanical properties were used as measures of the degradation in the hybrid composites. The results obtained from this work are to increase the efficiency of waste recycling and create more materials to be reused. Especially, hybrid material composites have the potential to be used to produce engineered products that have been tested for use in natural weathering conditions.

2. Materials and methods

2.1. Materials

The PP granules with the trade name 1100NK were obtained from IRPC Public Company Limited (Rayong, Thailand), and they had a melt flow index (MFI) of 11 g/10 min at 230 °C. Rubberwood flour (RWF) with 80-mesh size was acquired from Plan Creations Company Limited (Trang, Thailand). Sludge waste is a semi-solid slurry that is produced from processes of the rubber industries, including from the manufacturing process of fresh latex ponds and concentrated latex processing. There were two latex sludge wastes from the different manufacturing processes of the fresh latex pond (FLSF) and concentrated latex processing (CLSF), which FLSF was from the sedimentation process at the bottom of the fresh latex pond and CLSF was from sludge passing through a centrifuge. They were collected from the rubber latex industry in Southern Thailand (Songkhla, Thailand). The FLSF mainly contains P_2O_5 (36.07 wt%) and MgO (20.43 wt%), while the CLSF is composed of 33.44 wt% of P_2O_5 and 18.21 wt% of MgO, and other components are K_2O, ZnO, CaO, SO_3, SiO_2, Al_2O_3, Fe_2O_3, and Rb [28]. Before the hybrid composites mixing, these materials were oven dried at 110 °C for 24 to reduce the moisture content except sludge waste was dried in an oven at 120 °C for 48 h. Further, the maleic anhydride grafted polypropylene (MAPP) with 8–10% of maleic anhydride (M_w = 9100 and M_n = 3900) was used as a coupling agent. It was obtained from Sigma-Aldrich (Missouri, USA). The MAPP was used to enhance the interfacial bonding between the matrix and reinforcements in the composites.

2.2. Composites processing

The formulation of the hybrid composites in the experiment is shown in Table 1. Firstly, the PP granules, RWF, MAPP, and FLSF or CLSF were mixed using a co-rotating twin-screw extruder (Model CTE-D25L40 from Chareon Tut Co., Ltd, Samutprakarn, Thailand). The barrel temperatures ranged from 170 to 190 °C were controlled from the feeding to die zone. The screw speed is fixed at 50 rpm. The extruded compound was cooled in air and then pelletized. After that, the hybrid composite pellets were placed in the compression molding. The plate mold was 200 mm × 250 mm and 4.8 mm in thickness. The temperature was fixed at 190 °C under a pressure of 6.89 MPa (1000 psi) for 15 min, followed by a water-cooling system under pressure for 5 min. Three replicate plates were produced for each hybrid composite formulation. Finally, the hybrid composite plates were cut according to the American Society for Testing

Table 1
Formulation of the hybrid 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

Notes; PP: Polypropylene; RWF: Rubberwood flour; 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.

and Materials (ASTM) standard for physical and mechanical tests.

2.3. Natural weathering testing

Natural weathering tests under the coastal climate in the South of Thailand were done according to the ASTM D1435 standard. The hybrid composite samples were exposed to coastal weathering including the Gulf of Thailand and the Andaman Sea for a period of 12 months from July 2021-June 2022. The exposure consisted of the climatic conditions from the two annual seasons including the summer and rainy seasons in the South of Thailand. The composite samples were attached to the aluminum racks at a 45° angle and placed on the roof of the Engineering Building at Rajamangala University of Technology Srivijava (Songkhla Province) with latitude 7° 12′ 3.1″ N and longitude 100° 36′ 4.7′'E) and on the roof of the Engineering and Technology Building at Rajamangala University of Technology Srivijava (Trang province) with latitude 7° 31′ 27″ N and longitude 99° 20′ 07′'E), as shown in Fig. 1. Southern Thailand consists of two coastlines including the Gulf of Thailand on the east coast and the Andaman Sea on the west coast. The Southwest monsoon is occurred from May to October to cause lower pressure in China, while the Northeast monsoon is caused by the high pressure from China between October-February. These two monsoons bring humid air from the seas and generate rainfall in Southern Thailand [29]. The average temperature in Songkhla and Trang Provinces ranged from 26.5 - 29.4 °C and 26.2–28.6 °C, respectively, as well as the total rainfall value of 1047 mm and 425 mm, respectively, as shown in Fig. 2. The weather data for each month from July 2021 to June 2022 were collected from the Southern-East Coast Meteorological Center, Ministry of Digital Economy and Society, Thailand.

2.4. Analytical techniques

2.4.1. Thermal analysis

The thermogravimetric analysis of the hybrid composite samples was employed by a Perkin Elmer (TGA-7, USA). The test was conducted in the temperature range of 45 to 700 °C at a heating rate of 10 °C/min in a nitrogen atmosphere. The continuous weight loss and temperature were recorded and analyzed. Additionally, the hybrid composites were performed using differential scanning calorimetry (DSC) analysis with Perkin Elmer (DSC-7, USA). The samples were heated from 20 to 500 °C at the rate of 10 °C/min. The percentage of crystallinity (χ_c) from compounding between plastic matrix and filler wastes was calculated from the DSC thermogram according to Eq. (1):

Fig. 1. Locations of the test specimens being placed in different coastal weathering.

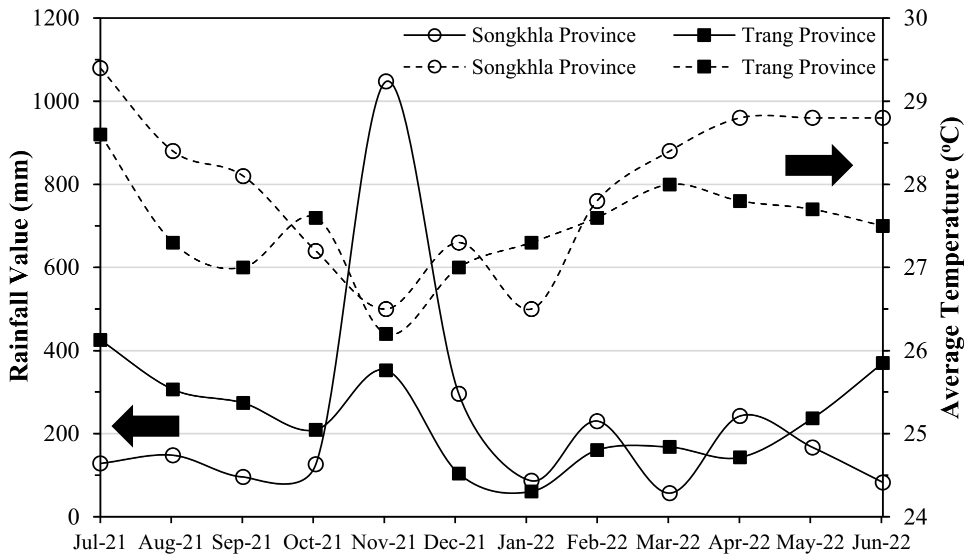


Fig. 2. Climate conditions during different coastal weathering testing showing rainfall values (solid lines) and average temperatures (dash lines).

$$\chi_c = \frac{\Delta H_m}{\Delta H_m^0} \text{ x } \frac{100}{W} \quad (1)$$

where ΔH_m is the enthalpy of fusion determined from DSC, ΔH_m^0 is the theoretical enthalpy of fusion of 100% crystalline plastic melting 148 J/g for PP [30,31], and W is the weight fraction of plastic in hybrid composites.

2.4.2. Visual surface analysis

Changes on the sample surfaces of the composites were analyzed before and after exposure under coastal climates of the Gulf of Thailand and the Andaman Sea for 2, 6, and 12 months. The samples were observed using an optical microscope (Zeiss Axioskop, Oberkochen, Germany).

2.4.3. Colorimetric analysis

The color analysis of the hybrid composite samples was carried out with a CHECK 3 portable spectrophotometer (Datacolor, USA). The surfaces of samples were measured both before and after exposing the coastal climates of the Gulf of Thailand and the Andaman Sea. The color was determined according to the CIE L^* a^* and b^*color system. The L^* displays the lightness and varies from 100 (white) to 0 (gray); the a^*coordinate displays color components from red ($+a^*$) to green ($-a^*$); and the b^* coordinate displays components from yellow ($+b^*$) to blue ($-b^*$). There were three replications of each formulation and condition for 0, 2, 6, and 12 months after weathering, and each sample was measured at three locations. The color difference or discoloration (ΔE) was calculated according to Eq. (2):

$$\Delta E = \sqrt{\Delta L^2 + \Delta a^2 + \Delta b^2} \quad (2)$$

where ΔL, Δa, and Δb represent the component differences between before and after weathering, in respective coordinates of L^*, a^*, and b^*.

2.4.4. Statistical analysis

A student's t-test was applied to detect significant differences between un-weathered and weathered (for 12 months) samples for modulus of rupture (MOR), modulus of elasticity (MOE), screw withdrawal strength (SWS), maximum flexural strain, and hardness of the composites. In addition, comparisons were also conducted within each formulation exposed under the coastal climate between the Gulf of Thailand and the Andaman Sea. Superscript letters were used to denote significance in tabulation; if the letters are the same, the means that are not significant differences; difference superscripts indicate significant differences. All of the statistical analyses were performed at a 5% significance level ($\alpha = 0.05$).

2.5. Characterizations

2.5.1. Flexural test

The three-point flexural test was carried out using a Mechanical Universal Testing Machine (Model NRI-TS500–50 from Narin Instrument Co., Ltd., Samut Prakan, Thailand) at a crosshead speed of 2 mm/min with a span of 80 mm, according to the ASTM D790. The size of samples was prepared following the standard about 13 × 100 × 4.8 mm^3, which measured both un-weathered and weathered specimens under coastal climates for 0, 2, 6, and 12 months. Data were collected and used to calculate the MOR, MOE, and maximum flexural strain of the composites.

2.5.2. Screw withdrawal test

The composite samples for the screw withdrawal test were prepared according to ASTM D1037 standard using the computer-controlled Universal Testing Machine. The dimensions of the samples were approximately 50 × 50 × 4.8 mm^3 (width × length × thickness). The wood screws with a diameter of 4.18 mm and threaded length of 50 mm were applied, which have been driven through their face. The speed of the loading crosshead was set at 1.5 mm/min for testing the specimens un-weathered and weathered for 2, 6, and 12 months.

2.5.3. Hardness test

The hardness measurement was performed on the un-weathered and weathered composite surfaces for 2, 6, and 12 months using a Shore D Durometer (Model GS-702 G from Teclock Corporation, Nagano, Japan), according to the ASTM D2240 standard. The composite samples were prepared with approximate dimensions of 30 mm (length) × 30 mm (width) × 4.8 mm (thickness). All mechanical characterizations were performed at ambient temperature (25 °C) with five replications for each formulation and condition.

3. Results and discussion

3.1. Differential scanning calorimetry analysis

The DSC values of the hybrid composites reinforcing fillers at ratios of 25 wt% RWF and 25 wt% FLSF or CLSF are presented in Table 2. The

Table 2
Melting temperature, melting enthalpy, and crystallinity degree of the hybrid composites after weathering.

Sample	T_m (°C)		ΔH_m (J/g)		Crystallinity (%)
	Peak I	Peak II	Peak I	Peak II	
P46R50	165.5	465.8	41.86	242.04	61.48
P46R50 (G)	149.3	447.8	88.79	268.70	130.42
P46R50 (A)	149.0	449.2	81.07	282.37	119.08
P46R25F25	165.8	477.7	51.21	229.06	75.22
P46R25F25 (G)	150.5	449.2	95.17	320.31	139.79
P46R25F25 (A)	150.2	449.7	81.55	290.15	119.78
P46R25C25	165.8	477.0	43.02	286.01	63.19
P46R25C25 (G)	149.5	451.0	95.10	327.75	139.68
P46R25C25 (A)	150.0	451.2	91.34	333.91	134.16

Notes; T_m is the melting temperature; ΔH_m is the heat quantity obtained by DSC; G: Gulf of Thailand; A: Andaman Sea.

melting temperature (T_m), melting enthalpy (ΔH_m), and crystallization of the composites were measured before and after weathering under conditions of the Gulf of Thailand and the Andaman Sea for 12 months. Previous studies found that the use of reinforcing fillers into the composites had a positive effect on thermal properties, but may decrease the toughness and ductility of the filled plastic matrix [32,33]. The decomposition profiles of the hybrid composites are characterized by two peaks in both the melting temperature and melting enthalpy. As can be seen from the melting temperature, the hybrid composites reinforced with FLSF had higher melting temperatures than those of the RWF and CLSF as fillers. The composites with 25 wt% FLSF (P46R25F25) indicated values of 165.8 °C for peak I and 477.7 °C for peak II, which gave the highest melting temperature as compared to those of the other samples. Further, the melting temperatures (165.8 °C) in peak I of both P46R25F25 and P46R25C25 were similar to that of P46R50 (165.5 °C). Likewise, the results in peak II showed that the composites with 25 wt% FLSF (P46R25F25) had the highest melting temperature as compared to those of the other samples. This result is in agreement with Homkhiew et al. [28], who concluded that the composites made from FLSF as reinforcing filler had higher melting temperatures than the CLSF. These results could be explained that the FLSF is not combusted in the molding process including extrusion and hot-pressing processes, resulting in high-temperature resistance of such composites in thermal testing.

The melting enthalpy (ΔH_m) was also presented to evaluate the thermal performance of the hybrid composites, as indicated in Table 2. The endothermic peaks of the composites reinforced with rubberwood and latex sludge flour were characterized in the form of two peaks, as same as the melting temperature. The composites made from P46R25F25 under weathering of the Gulf of Thailand and P46R25C25 under the climate of the Andaman Sea had the highest melting enthalpy values of 95.17 °C (peak I) and 333.91 °C (peak II) as compared to those of the other samples. The thermal result of P46R25F25 was good in terms of resisting the high temperature in DSC testing, in which melting enthalpy can be observed from second peaks (peak II). Moreover, the melting enthalpy of both the first and second peaks reduced, but the crystallinity degree rose with an increase of FLSF and CLSF in the hybrid composites. It is a good agreement with Srivabut et al. [34], who concluded that the addition of filler contents into the WPCs resulted in some decreases in the glass transition but increases the crystallization. After exposure to weathering of the Gulf of Thailand and the Andaman Sea, the addition of 25 wt% FLSF (P46R25F25) increased the crystallinity degrees by about 139.73% and 119.78%, respectively, while 25 wt % CLSF (P46R25C25) the crystallization degree increased about 139.68% and 134.16%, respectively. Overall, the result suggests that the hybrid composites reinforced with 25 wt% FLSF effectively improved the melting temperature, melting enthalpy, and crystallization, due to interface occurred and better compatibility between filler and plastic matrix.

3.2. Thermogravimetric analysis

The TGA and DTG curves for rubberwood and latex sludge flour-reinforced polypropylene as the hybrid composites before and after coastal weathering are displayed in Fig. 3(a) and (b), respectively. The study of the thermal stability of the hybrid composites was evaluated to analyze their future applications at high temperatures from the environment including the weathering and melting from the extrusion and compression molding process [35,36]. It is well known that one of the limiting factors of the hybrid composites made from plastic and natural fillers as reinforcement was their low thermal stabilities. Thus, it is necessary to evaluate their thermal stability for identifying filler types by comparing each condition [2,37]. However, the thermal stability behavior of the hybrid composites is studied using TGA and DTG as weight (%) values. According to the TGA curve (Fig. 3(a), the composites reinforced with 50 wt% RWF both un-weathering and weathering had higher weight loss in a temperature range of 100 to 700 °C as compared to those of the other composites. In addition, the hybrid composites produced from P46R25F25 and P46R25C25 had similar TGA curves in a temperature range of 500 to 700 °C, and the hybrid composites un-weathered displayed less thermal stability in the second stage of the decomposition than that weathered under conditions of the Gulf of Thailand and the Andaman Sea, as shown in Fig. 3(a). This result explains that the coastal weathering under the Gulf of Thailand and the Andaman Sea influenced the weight loss percentage of the hybrid composites, resulting in their more thermal stability in the second stage of the decomposition.

The DTG curves of the hybrid composites with rubberwood and latex sludge flour before and after coastal weathering are illustrated in Fig. 3 (b). All of the DTG curves of the hybrid composites are characterized into three peaks in which the first peak is the decomposition of water or oxygen components, while the second peak is the degradation behavior of the reinforcing filler, and the third peak is the decomposition temperature of the plastic matrix [18,38]. As reported by Ratanawilai et al. [39] revealed that the first stage corresponded to the release of moisture content from materials and the second stage is the decomposition of components in the composites. The results indicated that, in the third peak, the hybrid composites with 25 wt% FLSF (P46R25F25) after weathering under the Gulf of Thailand showed the largest derivative weight as compared with the other formulation and condition. Compared to the hybrid composites weathered under different coastal weathering, the composites weathered under the Gulf of Thailand presented a higher derivative weight than that under the Andaman Sea.

3.3. Color changes and cracked surface analysis

The lightness (L^*) and total color changes or discolorations (ΔE) of the hybrid composites are displayed in Fig. 4(a) and (b), respectively. The L^* and ΔE values of the control sample (PP100) and those of the composites reinforced with RWF and latex sludge flour (both FLSF and CLSF) un-weathered and weathered in different coastal weathering (Gulf of Thailand and Andaman Sea) for 2, 6, and 12 months are investigated. Theoretically, the surfaces of the composites exposed to the natural weathering observed changes in the form of color fading, in which the color changes in the L^* values increased with the increment of weathering exposure time in a similar way to the ΔE values [40]. The L^* and ΔE values are generally raised with an increase in the exposure time, but the characteristics of surface color also depend on the reinforcing filler contents [5,23,41]. This study revealed that the composite samples rapidly increased the L^* values in 2 months of exposure, except the control sample slightly increased. After that, all of the composite samples insignificantly increased the L^* values until 12 months of the coastal exposure. This result is in agreement with Mohammed et al. [26], who concluded that natural weathering reduced some physical properties such as the color performance of composites, and more biodegradable composites had less natural weathering resistance. Additionally, the

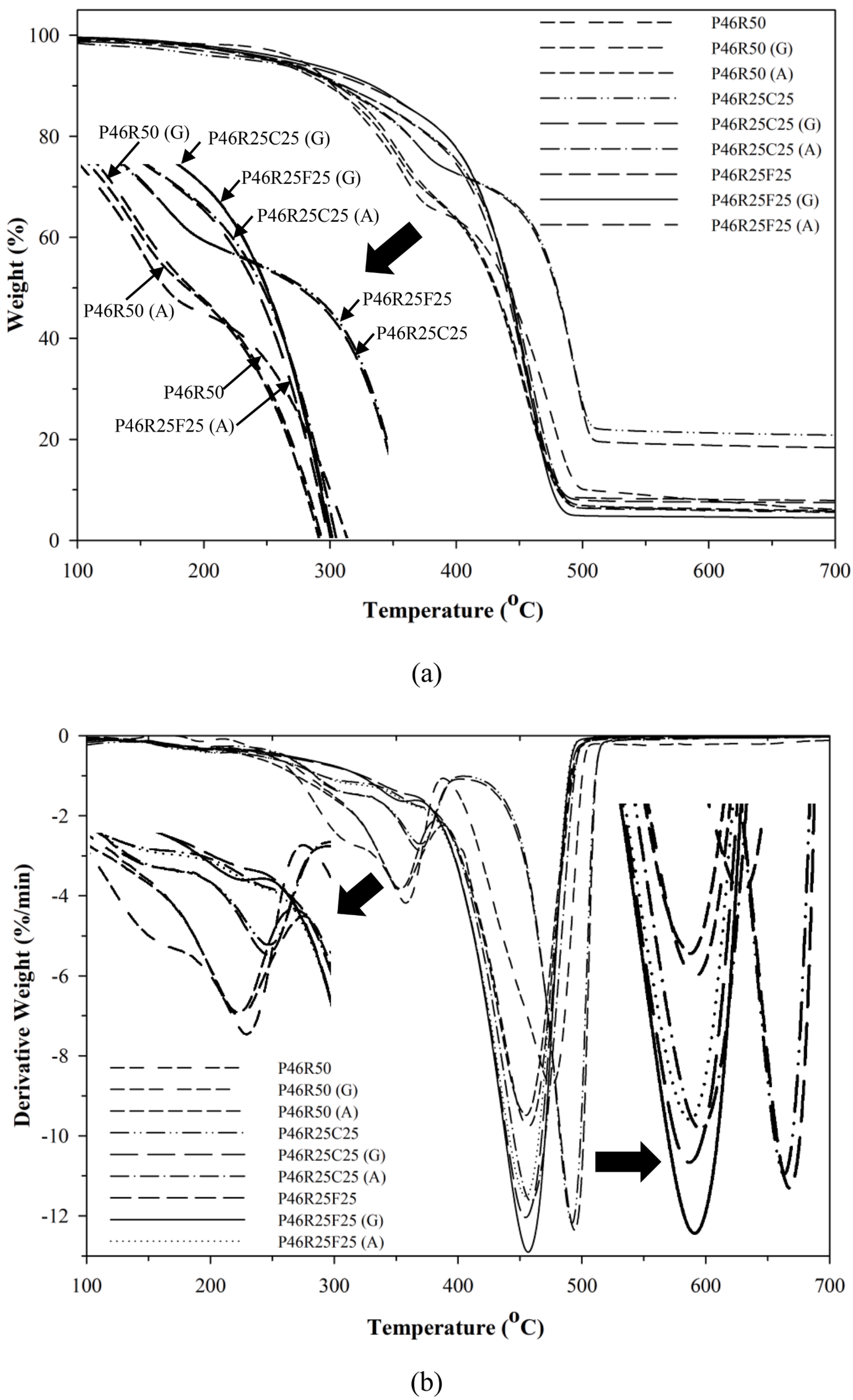
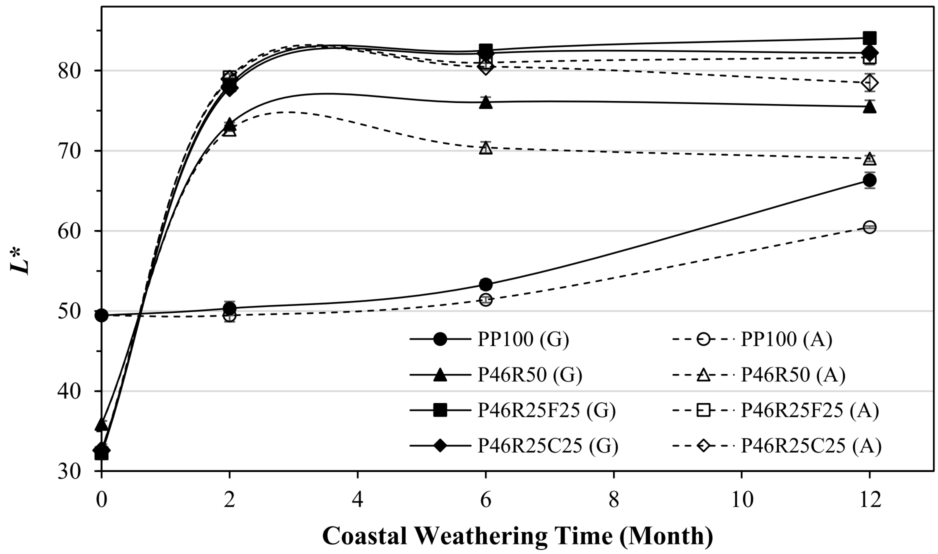
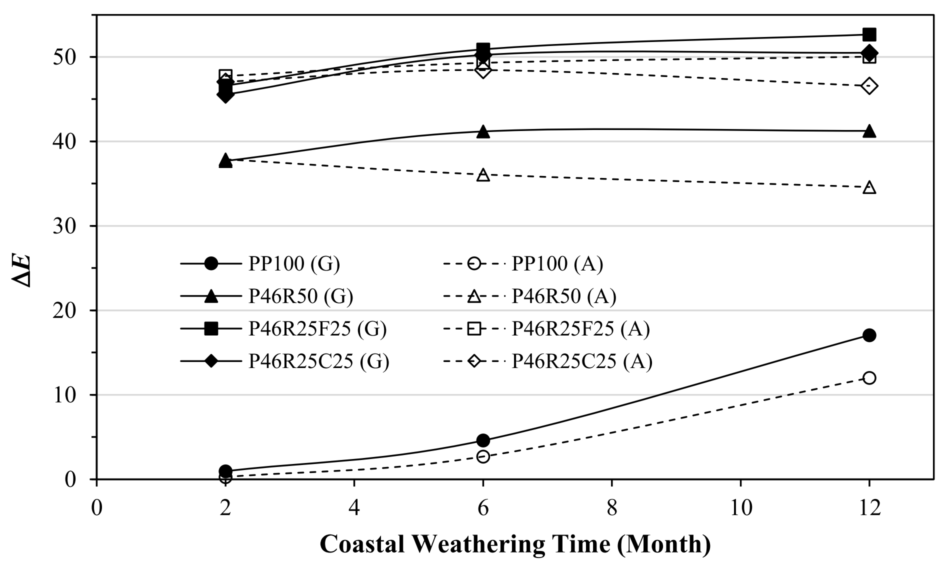


Fig. 3. Curves of (a) TGA and (b) DTG for rubberwood-latex sludge flour reinforced with polypropylene hybrid composites before and after coastal weathering.



(a)



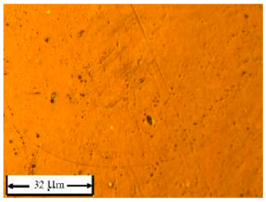
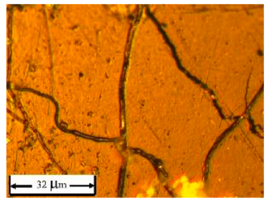
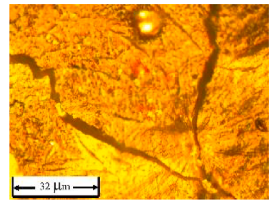
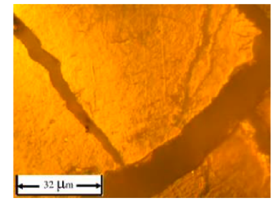
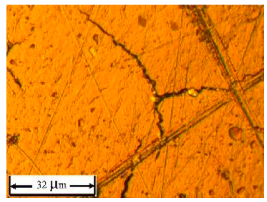
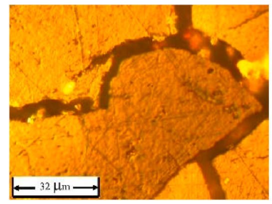
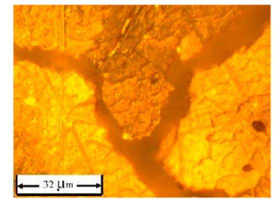
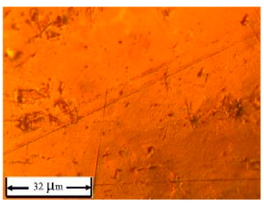
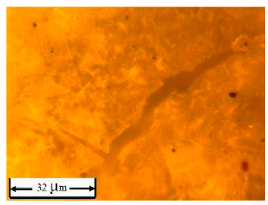
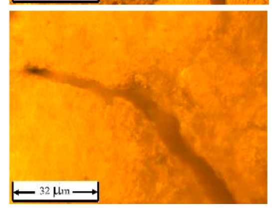
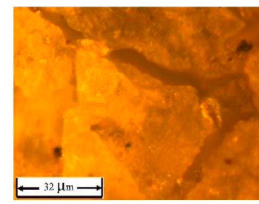
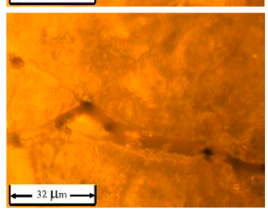
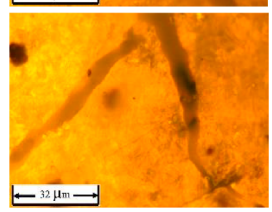
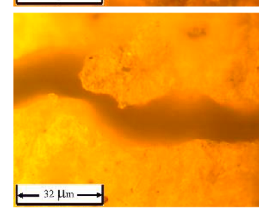
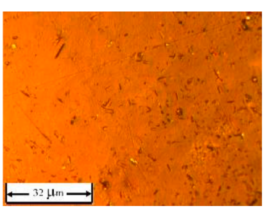
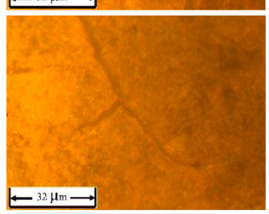
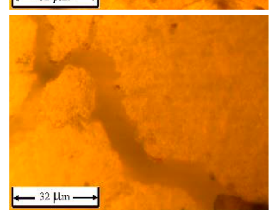
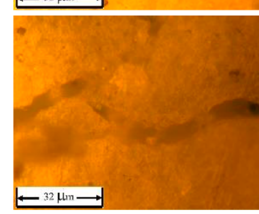
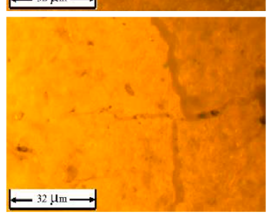
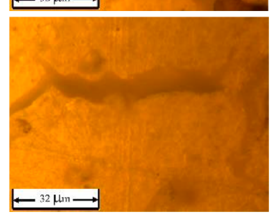
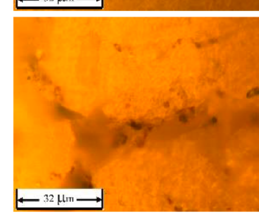
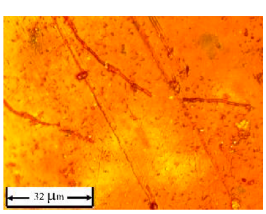
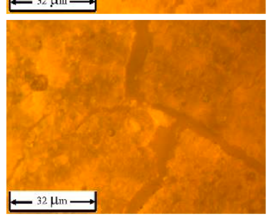
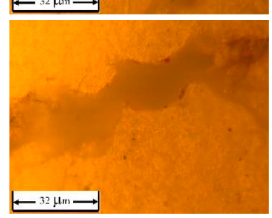
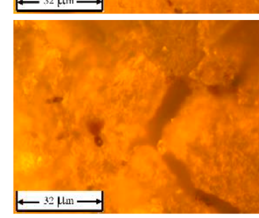
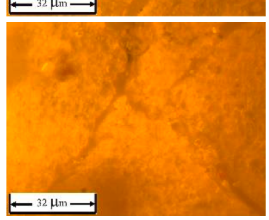
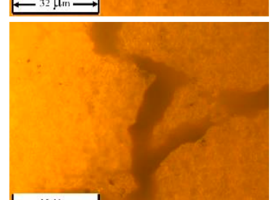
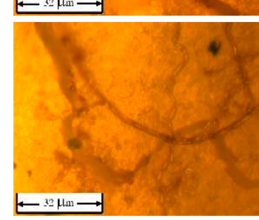
(b)

Fig. 4. Effects of different coastal weathering on (a) lightness and (b) total color change of rubberwood-latex sludge flour reinforced with polypropylene hybrid composites.

results showed that the L^* and ΔE values of the composites samples weathered for 2 months under the Gulf of Thailand were similar to those of the Andaman Sea; however, after 6 months the hybrid composites weathered under the Gulf of Thailand gave larger L^* and ΔE than that weathered under the Andaman Sea. The composition of P46R50 gave the lowest surface lightness under coastal weathering of the Andaman Sea, as displayed in Fig. 4(a). Further, the surface color change of P46R50 was more pronounced at the initial stage, whereas that other displayed a slight color change at the later stage of weathering exposure, which depends on the composition of the hybrid composites, as illustrated in Fig. 4(b). It means that the composition types and content, exposure times, and different coastal weathering conditions strongly affected the lightness and discolorations of the hybrid composites.

The microscopy images of the composite surfaces un-weathered and weathered in different coastal weathering conditions for 2, 6, and 12 months are shown in Table 3. In general, the poor surface performance of the composites is displayed by the presence of roughness and cracks on the surface [24]. As can be seen from the micrograph, the crack and fracture were observed on the surfaces of the control sample and the hybrid composites reinforced with RWF and latex sludge flour after exposure to the coastal climate. This study revealed that the fracture and crack on the surface increased with an increase in the exposure time, due to the increasing number of chain scissions. The polymer chain scission results in surface cracks and embrittlement of the plastic composites [41]. The control sample had a higher fracture and crack on the surface than those of the hybrid composites weathered under the climate of the Gulf of Thailand and the Andaman Sea. This is most likely due to the brittleness of plastics when exposed to light during time of initial stage.

Table 3
Optical microscopy images of the hybrid composite surfaces at different weathering stage.

Code	Un-weathered	Weathered for 2 months	Weathered for 6 months	Weathered for 12 months
PP100 (G)				
PP100 (A)				
P46R50 (G)				
P46R50 (A)				
P46R25F25 (G)				
P46R25F25 (A)				
P46R25C25 (G)				
P46R25C25 (A)				

This result is in agreement with Wei et al. [25], who reported that long-term outdoor weathering resulted in the poor surface in virgin plastic during time ranging from 1 to 2 months. Furthermore, the microscopy images showed that the hybrid composites, namely P46R50, P46R25F50, and P46R25C50, gave similar fracture and crack on the surfaces, whereas the surface of the hybrid composites displayed lower cracks than the control sample.

3.4. Mechanical properties of the hybrid composites

3.4.1. Flexural properties

The flexural properties (MOR and MOE) of the control sample (PP100) and the hybrid composites reinforced with RWF and CLSF or FLSF un-weathered and weathered under different coastal weathering (Gulf of Thailand and Andaman Sea) for 2, 6, and 12 months are displayed in Table 4 and Fig. 5. The experimental results in different contents, coastal weathering conditions, and exposure times were analyzed to compare the performance of the composites. As can be seen in Table 4, this study revealed that the compositions of the hybrid composites significantly affected the flexural properties. Overall, the control sample un-weathered had higher MOR values than those of the hybrid composites. The addition of RWF and latex sludge flour as reinforcement reduced the MOE values of the hybrid composite, as compared with the composites reinforced only RWF. It is a good agreement with Tian and Xu [42], who concluded that the wood filler in composites brings a pronounced lower modulus. Further, comparing to P46R50, P46R25F25, and P46R25C25 un-exposed to the coastal weathering, the MOR of the P46R50 (41.6 MPa) was the best tolerated, followed by P46R25F25 (41.5 MPa) and P46R25C25 (38.1 MPa), respectively. This result is in agreement with Srivabut et al. [34], who reported that rubberwood flour slightly increased the MOR in wood-plastic composites. For MOE values, the composites made from P46R50 had higher MOE values than those of all the hybrid composites. The samples having the composition of P46R50, P46R25F25, and P46R25C25 gave MOE values of 3.40 GPa, 3.20 GPa, and 3.06 GPa, respectively.

In addition, the flexural properties decreased with coastal weathering exposure time for 2, 6, and 12 months, as displayed in Fig. 5. As expected, the degradation in MOR and MOE values of the hybrid composites continually increased until 12 months, in which the MOR and MOE values slowly decreased in the initial period (2 months), and then they rapidly decreased until the 12 months. As reported by Rashdi et al. [43], it was concluded that the flexural properties slowly decreased in the initial period until 2 months of the natural weathering. Further, the hybrid composites (P46R25C25) gave the maximum MOR value as compared to those of the other composites after weathering for 12 months. The MOR values of 14.3 MPa and 15.9 MPa were determined for the hybrid composites having 25 wt% CLSF after exposure to weathering of the Gulf of Thailand and the Andaman Sea, respectively. Moreover, the hybrid composites made from P46R25C25 after weathering under the Gulf of Thailand and the Andaman Sea for 12 months showed the highest MOE values of 1.30 GPa and 1.42 GPa, respectively. The highest loss in MOR and MOE values about 448% and 207.4% was determined for the control sample and the composites with 50 wt% RWF (P46R50), respectively, under the condition of the Gulf of Thailand.

The maximum flexural strain of the control sample and the hybrid composites is also displayed in Table 4 and Fig. 6. The addition of the reinforcing fillers into the hybrid composites resulted in a lower flexural strain than the control sample. This could be possible due to the elastic deformation in which the atoms that moved and to the effects of stress moved back to their original positions [9,44]. As can be seen in Table 4, the maximum flexural strain of 6.26% was determined in the control sample before coastal weathering, whereas after weathering for 12 months under the climate of the Gulf of Thailand and the Andaman Sea, it gave the flexural strain of 2.20% and 2.46%, respectively. Comparing the flexural strain in P46R50, P46R25F25, and P46R25C25, it is evident that P46R25C25 had higher flexural strain than the P46R50 and P46R25F25 in both of the coastal weathering. For example, the maximum flexural strain of P46R25C25, P46R25F25, and P46R50 weathered under the Gulf of Thailand for 12 months was 1.18%, 1.09%, and 1.05%, respectively. All of the strain values in the hybrid composites that were un-weathered were similar to those that were weathered, and they were lower compared to the control sample, as shown in Fig. 6. The composites reinforcing with fillers either RWF or latex sludge waste gave lower flexural strain as compared to the plastic matrix.

A two-sample *t*-test was applied to analyze the effects of different coastal weathering conditions after weathering for 12 months of the control sample and the hybrid composites, as shown in Table 4. This study revealed that the coastal weathering conditions significantly ($p < 0.05$) affected the flexural properties of the hybrid composites. For example, in the formulation of the P46R25F25, the composites after weathering under the Gulf of Thailand having 13.2 MPa (suffix a) had significantly lower MOR than that under the Andaman Sea with 14.5 MPa (suffix b). Further, the two-sample *t*-test was also applied to analyze the effect of exposure time. The experimental results verified that the flexural properties (both MOR and MOE) of the hybrid composites significantly ($p < 0.05$) reduced after weathering for 12 months. For example, in the formulation of the P46R25C25, the composites un-weathered had a MOE of about 3.06 GPa (suffix A) which is a significantly higher MOE than that weathered under the Andaman Sea having 1.42 GPa (suffix B).

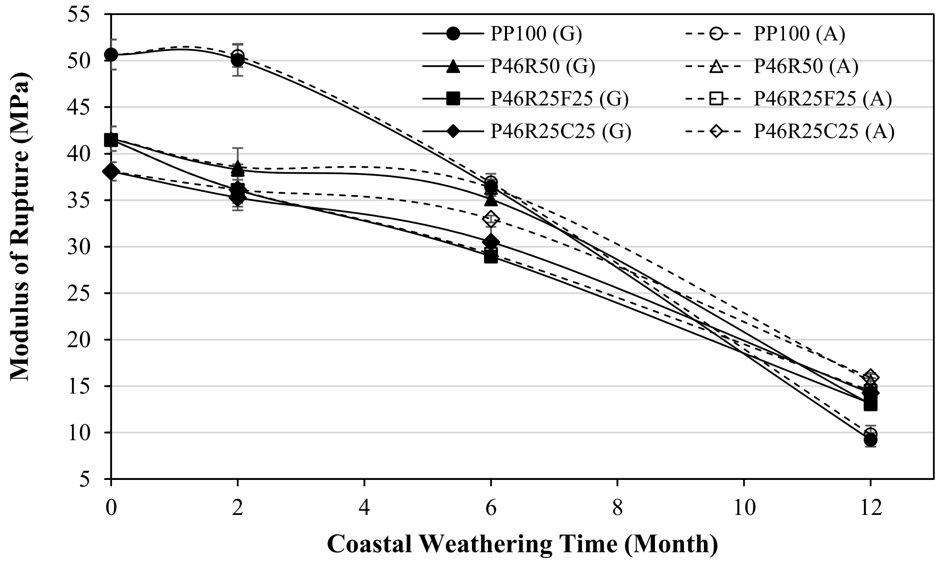
3.4.2. Screw withdrawal property

The screw withdrawal strength of the control sample and the hybrid composites un-weathered and weathered in different coastal weathering conditions (Gulf of Thailand and the Andaman Sea) for 2, 6, and 12 months is exhibited in Table 4 and Fig. 7. Generally, the aim of the screw withdrawal test in the WPCs is to measure the maximum strength

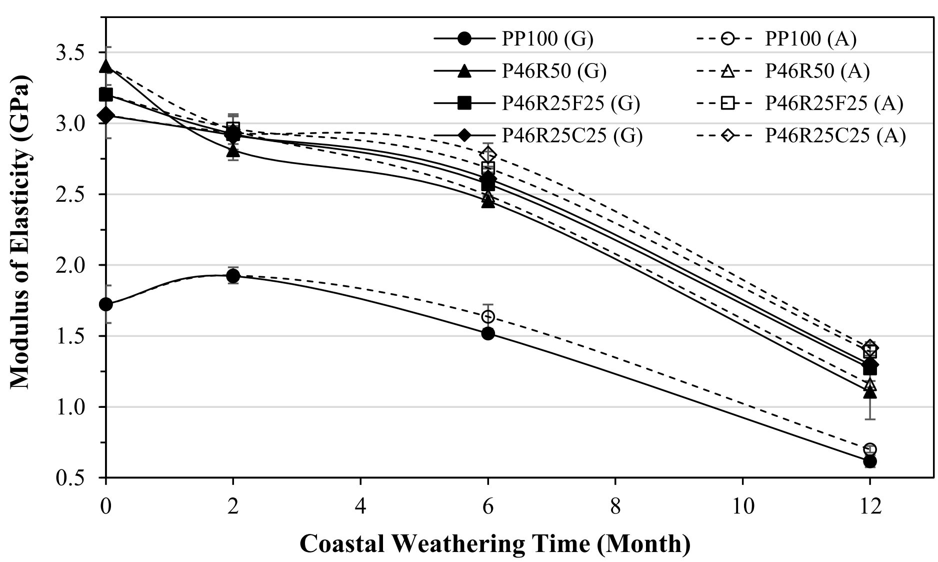
Table 4
Effects of different coastal weathering on mechanical properties of the hybrid composites.

Condition	MOR (MPa)			MOE (GPa)			Maximum strain (%)			SWS (MPa)			Hardness (Shore D)		
	0 M	12 M	% Loss	0 M	12 M	% Loss	0 M	12 M	% Loss	0 M	12 M	% Loss	0 M	12 M	% Loss
PP100 (G)	50.7 ^{A}	9.24 ^{aB}	448.5	1.72 ^{A}	0.62 ^{aB}	179.5	6.26 ^{A}	2.20 ^{aB}	184.0	54.9 ^{A}	17.0 ^{aB}	223.2	76.3 ^{A}	68.9 ^{aB}	10.6
PP100 (A)	50.7 ^{A}	9.82 ^{aB}	415.9	1.72 ^{A}	0.70 ^{bB}	145.9	6.26 ^{A}	2.46 ^{aB}	154.1	54.9 ^{A}	21.2 ^{bB}	159.5	76.3 ^{A}	70.2 ^{bB}	8.66
P46R50 (G)	41.6 ^{A}	13.1 ^{aB}	218.6	3.40 ^{A}	1.11 ^{aB}	207.4	1.55 ^{A}	1.05 ^{aB}	48.0	40.9 ^{A}	24.7 ^{aB}	65.4	78.7 ^{A}	71.4 ^{aB}	10.2
P46R50 (A)	41.6 ^{A}	15.5 ^{bB}	167.8	3.40 ^{A}	1.16 ^{aB}	193.4	1.55 ^{A}	1.16 ^{bB}	33.8	40.9 ^{A}	28.4 ^{bB}	43.9	78.7 ^{A}	73.7 ^{bB}	6.78
P46R25F25 (G)	41.5 ^{A}	13.2 ^{aB}	215.4	3.20 ^{A}	1.27 ^{aB}	151.7	1.57 ^{A}	1.09 ^{aB}	44.1	33.2 ^{A}	18.1 ^{aB}	84.2	80.1 ^{A}	71.9 ^{aB}	11.4
P46R25F25 (A)	41.5 ^{A}	14.5 ^{bB}	186.0	3.20 ^{A}	1.39 ^{bB}	130.4	1.57 ^{A}	1.15 ^{aB}	36.7	33.2 ^{A}	21.4 ^{bB}	55.1	80.1 ^{A}	72.2 ^{aB}	10.9
P46R25C25 (G)	38.1 ^{A}	14.3 ^{aB}	167.2	3.06 ^{A}	1.30 ^{aB}	135.3	1.59 ^{A}	1.18 ^{aB}	35.4	29.9 ^{A}	23.9 ^{aB}	25.1	78.0 ^{A}	72.2 ^{aB}	10.8
P46R25C25 (A)	38.1 ^{A}	15.9 ^{bB}	138.9	3.06 ^{A}	1.42 ^{bB}	115.8	1.59 ^{A}	1.25 ^{aB}	27.5	29.9 ^{A}	25.0 ^{bB}	19.9	78.0 ^{A}	73.0 ^{bB}	9.53

Notes: M: Month; G: Gulf of Thailand; A: Andaman sea; MOR: Modulus of rupture; MOE: Modulus of elasticity; SWS: Screw withdrawal strength; Means within each column of each formulation at 12 M with the same superscripts a-b indicate a insignificant difference ($\alpha = 0.05$) by the *t*-test. Different superscripts A-B of each property and condition indicate significant difference ($\alpha = 0.05$) between mechanical properties of the composites unweathered and weathered for 12 months;% loss was calculated at 12 months of natural weathering.



(a)



(b)

Fig. 5. Effects of different coastal weathering on (a) modulus of rupture and (b) modulus of elasticity of rubberwood-latex sludge flour reinforced with polypropylene hybrid composites.

required to pull a standard-size screw from the specimens, which determines the useful service life in many applications, especially construction and building [45,22]. The experimental results of this research used to evaluate the direct withdrawal of the screw for the hybrid composites were a measure of the resistance of the samples under different coastal weathering conditions and exposure times. According to Table 4, this study revealed that the reinforcing filler types and contents significantly affected the screw withdrawal strength of the composites. It is evident that the addition of reinforcing filler reduced performance in the screw withdrawal strength of the composites. This result is in agreement with Srivabut et al. [45], who concluded that the screw withdrawal resistance was reduced with an increase in wood contents. The control sample un-weathered had the highest screw withdrawal strength as compared to those of the WPCs and the hybrid composites. The average screw withdrawal strength for the control samples was found value of 54.9 MPa. This could be possible due to the thermoplastic matrix encapsulating the thread of the screw, allowing continuous load transfer along the threaded length [46]. In addition, the un-weathered WPCs (P46R50) having a value of 40.9 MPa had a higher screw withdrawal strength than the hybrid composites with 25 wt% of both FLSF (P46R25F25) and CLSF (P46R25C25) having values of 33.2 MPa and 29.9 MPa, respectively.

The screw withdrawal strength of the hybrid composites under different coastal weathering conditions (Gulf of Thailand and Andaman Sea) for 2, 6, and 12 months are also illustrated in Fig. 7. It is observed that the different coastal weathering and exposure times led to more significant changes in screw withdrawal property. The screw withdrawal strength of the control sample and the hybrid composites

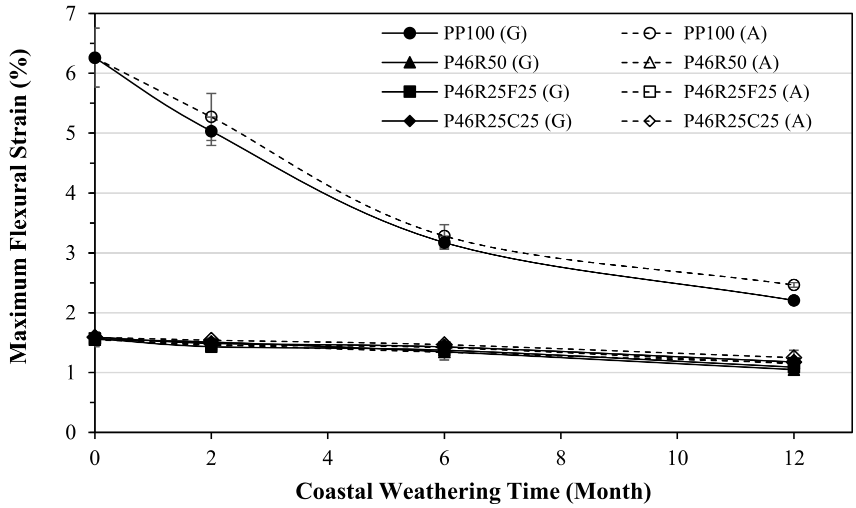


Fig. 6. Effects of different coastal weathering on the maximum flexural strain of rubberwood-latex sludge flour reinforced with polypropylene hybrid composites.

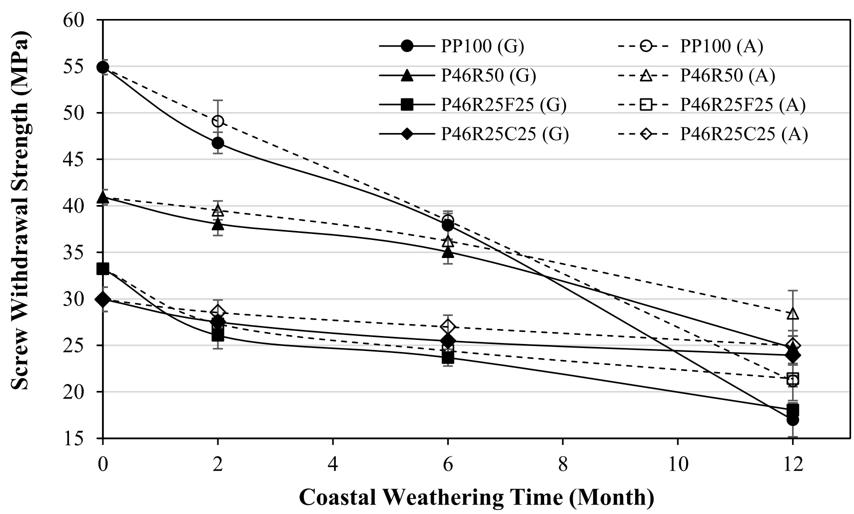


Fig. 7. Effects of different coastal weathering on screw withdrawal strength of rubberwood-latex sludge flour reinforced with polypropylene hybrid composites.

continuously decreased with an increase in exposure time. The highest reduction of screw withdrawal strength values ranging from 54.9 MPa to 17.0 MPa was determined in the control sample after weathering under the Gulf of Thailand for 12 months. The highest loss about 223.3% and 159.5% was found in the control sample after weathering under conditions of the Gulf of Thailand and the Andaman Sea, respectively. The reason for such an extra reduction of the screw withdrawal strength may be found in the fact that the plastic brittleness under natural weathering degraded the surface layer, which contributes less to the strength [23,26,41]. In addition, when comparing the screw withdrawal strength of the hybrid composites with latex sludge flour and RWF as reinforcing fillers under both coastal weathering conditions, the hybrid composites with 25 wt% FLSF (P46R25F25) had a higher loss percentage than those of the WPCs and P46R25C25. The loss in screw withdrawal strength of P46R25F25 after weathering under conditions of the Gulf of Thailand and the Andaman Sea for 12 months was 84.2% and 51.1%, respectively.

Furthermore, it was found that different coastal weathering conditions (the Gulf of Thailand and the Andaman Sea) significantly influenced the screw withdrawal strength of the hybrid composites. It is observed from the P46R25C25 weathered for 12 months that the hybrid composites weathered under the Gulf of Thailand could hold worse screws than the Andaman Sea, in which loss of their screw withdrawal strengths was 84.2% and 55.1%, respectively. These results could be concluded that the control sample, the WPCs, and the hybrid composites weathered under the Gulf of Thailand degraded more screw withdrawal strength than that weathered under the Andaman Sea.

3.4.3. Hardness property

The evaluation of strength on the surface of the hybrid composites un-weathered and weathered for 2, 6, and 12 months under different coastal weathering conditions (the Gulf of Thailand and the Andaman Sea) is presented by hardness properties. The experimental results of each condition are shown in Table 4 and Fig. 8. It is a known fact that the hardness of the composites made with the reinforcing filler was significantly higher than those of the control sample. As reported by Nukala et at. [18], it was concluded that the hardness values of the composites increased with the increasing wood waste content and higher hardness values than those of recycled plastic waste. As expected, the hybrid composites reinforced with both RWF and CLSF or FLSF as fillers had higher hardness values than the pure PP. As can be seen in Table 4, the reinforcement with FLSF had a better hardness value than the composites with CLSF and RWF. This is due to the latex sludge flour from FLSF absorbing large amounts of energy and good stress transfer during the deformation process [32]. It is also found that the hybrid composites with 25 wt% FLSF (P46R25F25) un-weathered had the highest hardness values of 81.1 shore D, while the average hardness value for the control

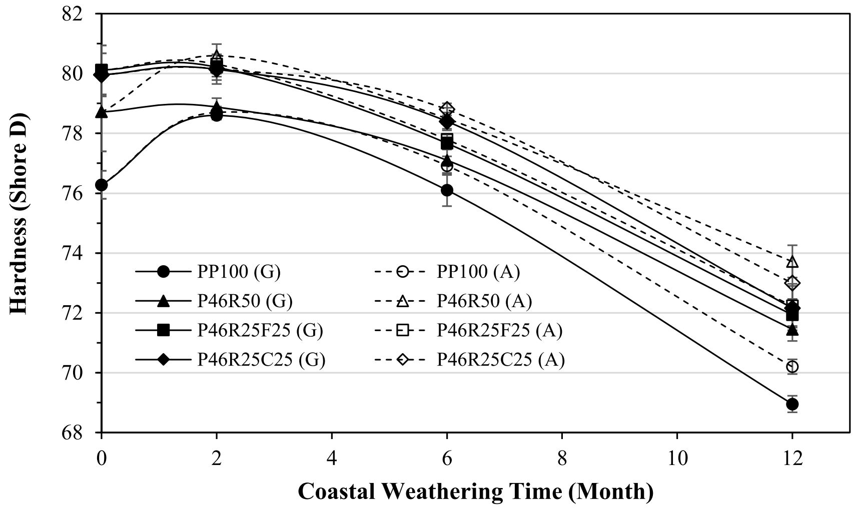


Fig. 8. Effects of different coastal weathering on the hardness of rubberwood-latex sludge flour reinforced polypropylene hybrid composites.

samples was found about 76.3 shore D. These results could be explained that the reinforcing fillers has a considerably higher hardness values than the plastic matrix [18,28,39].

Additionally, the hardness values of the hybrid composites with different coastal weathering (Gulf of Thailand and Andaman Sea) for 2, 6, and 12 months are also displayed in Fig. 8. Theoretically, the hardness values greatly decreased with an increase in the exposure times [41,45]. This study also revealed that the degradation in hardness values of the hybrid composites and the control samples increased after the coastal weathering exposure for 2 months and then decreased rapidly after 6 months until 12 months. Compared to the hybrid composites in the formulation of P46R50, P46R25F25, and P46R25C25 weathered under the Gulf of Thailand and the Andaman Sea for 2, 6, and 12 months, the hardness values of the P46R25C25 were the best tolerated, followed by P46R25F25 and P46R50, respectively. This can be explained that the latex sludge flour as reinforcement had lower moisture absorption than RWF, which could be well inserted into the matrix, resulting in low degradation from coastal weathering.

Moreover, it is also found that the different coastal weathering (the Gulf of Thailand and the Andaman Sea) affected the hardness values of the hybrid composites because climate conditions, as shown in Fig. 2, decrease different toughness and ductility of the hybrid composites [45]. The hybrid composites weathered under the Gulf of Thailand had higher hardness degradation than that Andaman Sea. The hybrid composites weathered under the Andaman Sea in the formulation of P100, P46R50, P46R25F25, and P46R25C25 had a loss of hardness values of about 8.66%, 6.78%, 10.09%, and 9.53%, respectively, while those weathered under the Gulf of Thailand had a loss of the hardness of about 10.6%, 10.2%, and 11.4%, and 10.8%, respectively, as shown in Table 4. Also, the composites with P46R25F25 weathered under the Gulf of Thailand had the highest loss of hardness values of about 11.4%, as compared to those of the other samples. Hence, all of the composites weathered under the Gulf of Thailand for 12 months gave lower hardness values than those weathered under the Andaman Sea.

4. Conclusions

In this study, the hybrid composites' thermal, physical and mechanical properties were characterized before and after weathering under different coastal climates, i.e., the Gulf of Thailand and the Andaman Sea, for 2, 6, and 12 months. We draw the following conclusions based on the results obtained:

- The hybrid composites added to the latex sludge flour had higher crystallinity degree and thermal stability than the WPCs with 50 wt% RWF, while the hybrid composites weathered under the coastal climates gave larger crystallinity degree and thermal stability than those that were un-weathered.
- The hybrid composites rapidly increased the lightness and total color change after weathering under the coastal climates for 2 months, and then insignificantly increased until 12 months of coastal exposure due to bleaching in photo-oxidation of the wood component.
- The degradation in mechanical properties, such as MOR, MOE, SWS, and hardness, of the hybrid composites after weathering under the coastal climates continually increased until 12 months. This is because of continuous chain scissions of the plastic matrix with increasing exposure times.
- The hybrid composites weathered under the Andaman Sea showed higher MOR, MOE, SWS, and hardness than those weathered under the Gulf of Thailand, which resulted in less loss of mechanical properties after weathering under the coastal climate. This is because the weather of the Gulf of Thailand had more average temperature and total rainfall than that of the Andaman Sea.
- The WPCs and the hybrid composites indicated a lower loss percentage of the mechanical properties than the control sample (PP100), after weathering under the coastal climates for 12 months.

The knowledge gained from this study would facilitate informed decisions that were appropriate for applications under the coastal climates of the hybrid composites, especially, the building and construction materials, such as railing, fencing, flooring, and decking, normally applied under the coastal weathering. Additionally, this finding supports the effective application of naturally-sourced fillers in plastic composites. The use of latex sludge waste from the rubber latex industries in reinforcing fillers will also allow the improvement and development of green material products that can be decomposed naturally.

Declaration of Competing Interest

The authors declared that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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