Journal of Material Cycles and Waste Management (2018) 20:1792–1803
https://doi.org/10.1007/s10163-018-0749-y

ORIGINAL ARTICLE

Potential utilization of rubberwood flour and sludge waste from natural rubber manufacturing process as reinforcement in plastic composites

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Received: 8 June 2017 / Accepted: 15 May 2018 / Published online: 22 May 2018

Abstract

The present study aims to evaluate and compare the potential of rubberwood flour (RWF) and sludge waste from manufacturing process of the concentrated natural rubber latex as reinforcement in recycled high-density polyethylene (rHDPE) composites. Morphological, thermal, mechanical and physical properties were investigated. The results indicated that the tensile modulus (TM), modulus of elasticity (MOE) and hardness of rHDPE composites containing sludge waste flour (SWF) were improved with the increasing amount of SWF; however, the tensile strength (TS), modulus of rupture (MOR) and water absorption (WA) exhibited a negative correlation with the SWF concentration. The composites based on SWF exhibited better TS, MOR, maximum flexural strain and WA than that of based on RWF. The thermal experiments showed that the decomposition temperatures of the rHDPE composites with SWF were higher than that of with RWF. Further, the addition of maleic anhydride-grafted polyethylene improved the mechanical and physical properties of the composites reinforcing the SWF or RWF. The above results showed that the utilization of latex sludge waste could become a promising way for solving the environmental problem as well as improving many properties of plastic composites.

Keywords Wood–plastic composites · Latex sludge waste · Rubberwood flour · Statistical methods

Introduction

In concentrated natural rubber latex manufacturing process, a large amount of sludge waste is generated at different stages. Some sludge waste could be utilized as phosphatic fertilizers [1]. However, most of them is generally discharged without further treatment by sending them to landfill or discharging them in the rubber plantation [2, 3]; see Fig. 1. Because the composition of this material includes phosphorus 14.7% by dry weight (wt%), rubber hydrocarbon 12.5 wt%, magnesium 12.2 wt% and nitrogen 3.3 wt% [4, 5], it is naturally difficult to decompose rubber hydrocarbon in soils [4]. Disposing the sludge waste with these methods will cause both economic and environmental problems since it still contains high volume of natural rubber (which is valuable) and results in lower gas permeability of the soil in landfill sites [2]. Likewise, accumulation of organic substance in the sludge waste also causes air pollution, releasing stench into the air. Currently, recycling the sludge waste to produce useful products is not being used commercially due to its high cost; this had been shown by Taweepreda [4] who recovered rubber from latex sludge waste using sulfuric acid. So, the possibility of utilizing the sludge waste as filler or reinforcement to improve the mechanical and physical properties of plastic composites is an interesting approach because it still has high rubber hydrocarbon content. Furthermore, the addition of latex sludge waste into plastic matrix is significantly different from rubber–plastic blends because apart from natural rubber, the sludge waste not only consists of natural rubber, but also a number of inorganic substances that infiltrates the rubber matrix. In the past, some researchers had experimented on inserting the sludge wastes into plastic matrix to reduce cost and to improve the properties of plastic composites; however, no

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Fig. 1 Sludge waste from manufacturing process of the concentrated natural rubber latex in landfill

research has been done to add the sludge waste from natural rubber manufacturing process into the plastic composites. Soucy et al. [6] applied paper mill sludge as a raw material in wood–plastic composites (WPCs). They revealed that the increasing proportion of paper mill sludge had improved the composite strength. Likewise, Hamzeh et al. [7] found that the addition of paper sludge waste had positively affected the flexural properties and tensile modulus of the WPCs because not only it acts as a filler, but also as a reinforcing element. Ayrilmis and Buyuksari [8] revealed that the increasing addition of olive mill sludge improved the water resistance of the WPCs.

Plastic composites, especially in WPCs, are most commonly found in outdoor building products such as railing, fencing and decking because they have enough strength, high specific strength and stiffness, and positive impact on environmental issues [9, 10]. While most of WPCs produced from virgin plastics have been being used for exterior building components and commercial products, the utilization of recycled plastics for WPCs is limited. Adhikary et al. [11] reported that the composites produced from recycled high-density polyethylene (rHDPE) were similar or, in some cases, had better mechanical properties than that of composites from virgin high-density polyethylene (vHDPE). Yao et al. [12] showed that the modulus and strength properties of rHDPE resin and its composites were better than vHDPE systems because of additives used during initial processing. Other research showed that a huge amount of plastic waste was incinerated or landfilled while only about 10% was recycled [13, 14]. Therefore, the use of recycled plastics or plastic waste to make WPCs would not only reduce the consumption of natural resources, but also provide a safe and effective solution to plastic waste disposal [15, 16].

In wood processing, a large amount of wood waste in forms of chip, sawdust and flour is generated at different stages, and such waste is mainly disposed in landfills and burning, resulting in pollution problems. Utilization of wood waste or natural fibers as reinforcement in plastic composites is a good approach in disposal of such waste, which reduces environmental impacts and improves the properties of the composites [17]. Väisänen et al. [18] reported that the stiffness of the end-composites increased with addition of natural fibers as a reinforcement. The flexural strength and modulus of WPCs increase with increasing wood content since the wood has higher modulus than the plastic [19]. However, the natural fibers have certain disadvantages such as low compression strength, moisture absorption and poor thermal resistance [20, 21]. In spite of extensive research in the area of plastics reinforced with natural fibers, there are few researchers who have used rubberwood flour as reinforcement in plastics, and there is no report of comparative study between latex sludge waste composites and natural fiber composites.

The objective of this work was to evaluate and compare the potential of rubberwood flour and sludge waste from manufacturing process of the concentrated natural rubber latex as reinforcement in rHDPE composites. The sludge waste generated from the manufacturing process of concentrated natural rubber latex as filler was utilized in the plastic composites. The use of sludge waste in the plastic composites would become a promising way for both solving the environmental pollution and increasing the value of this waste as well as replacing the WPCs in some situation.

Materials and methods

Materials

There were two types of reinforcement used in this investigation, i.e., sludge waste and rubberwood sawdust waste. Sludge waste from the manufacturing process of the concentrated natural rubber latex was provided by a rubber glove plant in South of Thailand. It composed of moisture 30 wt%, rubber hydrocarbon 20 wt% and inorganic and carbon substances 50 wt%. Rubberwood sawdust, used as a lignocellulosic filler, was supplied from a local rubberwood furniture factory in Songkhla, Thailand. Its main chemical composition was hemicelluloses and cell wall 29%, lignin 28%, cellulose 39% and ash 4% [10, 22]. The rubberwood usually has tensile stress and Young's modulus of approximate 57.12 and 2252 MPa, respectively [23]. The rHDPE pellets (the polymer matrix), under the trade name WT114 with a melt flow index of 14 g/10 min at 190 °C, were purchased from Withaya Intertrade Co., Ltd. (Samutprakarn, Thailand). To improve the interfacial adhesion between plastic matrix and reinforcement, maleic anhydride-grafted polyethylene (MAPE) with 0.5 wt% of maleic anhydride and 500 cP (140 °C) (lit.) of viscosity was chosen as a coupling agent; it was supplied by Sigma-Aldrich (Missouri, USA). Because the sludge waste had high moisture content and large sizes, it was dried in an oven at 120 °C for 48 h and then hammer-milled into particles as shown in Fig. 2. The particles of both sludge waste and rubberwood sawdust were then sieved through a standard sieve of mesh size 70 (passing particles smaller than 212 µm) before compounding.

Composites processing

Production of the composite materials consists of two processes. First, the sludge waste flour (SWF) or the rubberwood flour (RWF) were compounded with rHDPE and MAPE pellets (formulations in Table 1) in a co-rotating twin-screw extruder (Model CTE-D25L40 from Chareon Tut Co., Ltd, Samutprakarn, Thailand). The 7-barrel temperature of the extruder was controlled in the range of 135–170 °C from feeding to die zone. The screw rotation speed was varied between 50 and 60 rpm, depending on the reinforcement content being blended. The extruded strand passed through an air blower and was subsequently pelletized. Second, to produce composite panels, the compounded pellets were dried in an oven at 110 °C for 5 h to minimize their moisture contents. The composite pellets were then compressed in a metal frame size of 200 mm × 250 mm × 6 mm using a hydraulic hot compression machine. The temperature of the press plates was set to 190 °C before placing the mold containing pellets on the lower plate. The pellets were then compressed under a pressure of 250 psi for 5 min; the pressure was then increased to 1000 psi for 10 min. Finally, the mold and the composite panel were removed to a hydraulic

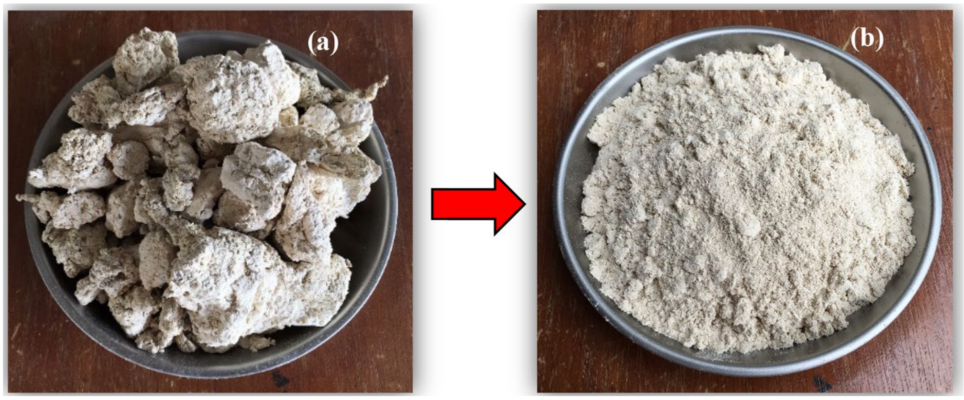

Fig. 2 Sludge waste **a** before and **b** after hammer-milled

Table 1 Formulation of composite materials in experiment

Composite sample code	rHDPE (wt%)	RWF (wt%)	SWF (wt%)	MAPE (wt%)
rHDPE	100	–	–	–
HRWF20	80	20	–	–
HRWF30	70	30	–	–
HRWF40	60	40	–	–
HRWF50	50	50	–	–
HRWF60	40	60	–	–
HSWF20	80	–	20	–
HSWF30	70	–	30	–
HSWF40	60	–	40	–
HSWF50	50	–	50	–
HSWF60	40	–	60	–
HRWF60MA2	38	60	–	2
HRWF60MA4	36	60	–	4
HSWF60MA2	38	–	60	2
HSWF60MA4	36	–	60	4

rHDPE recycled high-density polyethylene, *RWF* rubberwood flour, *SWF* sludge waste flour, *MAPE* maleic anhydride-grafted-polyethylene, *wt%* percent by weight cold press and held under a pressure of 1000 psi for 10 min. Subsequently, the composite panels were machined as specimens according to American Society for Testing and Materials (ASTM) standard for mechanical and physical tests.

Characterizations

Mechanical test

Tensile and flexural tests were carried out on a Mechanical Universal Testing Machine (Model NRI-TS500-50 from Narin Instruments Co., Ltd, Samutprakarn, Thailand) at room temperature (25 °C). Tensile properties, such as tensile strength (TS) and tensile modulus (TM), were conducted using the type-IV tensile specimens with a cross-head speed of 5 mm/min, according to ASTM standard D638-99. Flexural properties, such as modulus of rupture (MOR), modulus of elasticity (MOE) and maximum flexural strain, were also measured in a three-point bending test with nominal dimensions of 4.8 mm × 13 mm × 100 mm, a span of 80 mm and a cross-head speed of 2 mm/min, according to ASTM standard D790-92. All mechanical tests were conducted with five replications for each composite formulation.

Hardness test

The hardness of the plastic composites was measured according to ASTM standard D2240-91. The specimens with nominal dimensions of 30 mm × 30 mm × 6 mm were used for testing. The measurements for five replications were conducted by using Durometer Shore D scales (Model GS-702G from Teclock Corporation, Nagano, Japan), at 25 °C.

Water absorption test

Water absorption (WA) tests of the composites were conducted according to ASTM standard D570-88. The specimens with nominal dimensions of 15 mm × 30 mm × 6 mm were cut from the compressed composite panels. Before testing, five specimens of each formulation were carefully dried in an oven at 50 °C for 24 h, and then weighed with a precision of 0.001 g and subsequently submerged in water at 25 °C. After 24 h, the specimens were removed, dried with tissue papers and immediately weighed to calculate the percentage of WA.

Analytical methods

Morphological analysis

Scanning electron microscope (SEM) was used to observe and analyze the interface adhesion, voids and dispersion of the fillers in the plastic matrix. Prior to SEM evaluation, all sample surfaces were prepared by sputter coating with gold in order to eliminate electron charging during the imaging. A FEI Quanta 400 microscope (FEI Company, Oregon, USA) imaged the composite surfaces at an accelerating voltage of 20 kV with magnifications of 150× and 1500×, respectively.

Thermogravimetric analysis

Thermogravimetric analysis (TGA) to investigate the thermal stability of the rHDPE, SWF and rHDPE composites containing different fillers and contents was performed with a Perkin Elmer (TGA-7, Massachusetts, USA). Samples of approximate 5–8 mg were scanned at a constant heating rate of 10°C/min from 50 to 700 °C under nitrogen atmosphere. The weight loss was recorded as a function of temperature.

Statistical analysis

Experimental results from five specimens of each formulation and characterization were statistically analyzed. The effects of filler and MAPE contents on the rHDPE composites' properties were evaluated by using analysis of variance (ANOVA) and Tukey's multiple comparison test. ANOVA revealed the significant differences of filler and MAPE contents while Tukey's comparison test indicated a comparison between means. The significant differences between filler types were also conducted with a two-sample *t* test. A 5% significance level ($\alpha = 0.05$) was employed for all statistical analyses.

Results and discussion

Micro-morphology analysis

In general, SEM images of fractured surface of the composites can be used to evaluate pores in composite structures, dispersion of fillers in plastic matrix and interfacial adhesion between polymers and fillers. Figure 3a, b illustrates the SEM micrograph of the fractured surface of rHDPE composites containing 20 wt% RWF (HRWF20) and 60 wt% RWF (HRWF60), respectively. It can be observed that the fractured surface of HRWF20 was smoother than HRWF60. The composites with higher RWF contents exhibited more pores in composite structures as well as larger gaps between plastic matrix and filler, which would result in poorer mechanical properties and accelerate the water absorption [24]. As can be seen in Fig. 3, the globular particles of sludge waste dispersed well in the rHDPE matrix. The rHDPE composites filled 20 wt% SWF (Fig. 3c) showed less pores in composite structures and smoother surface than filled 60 wt% SWF (Fig. 3d). For composites with 60 wt% SWF, there was clearly pull-outs of sludge waste flour in the fracture

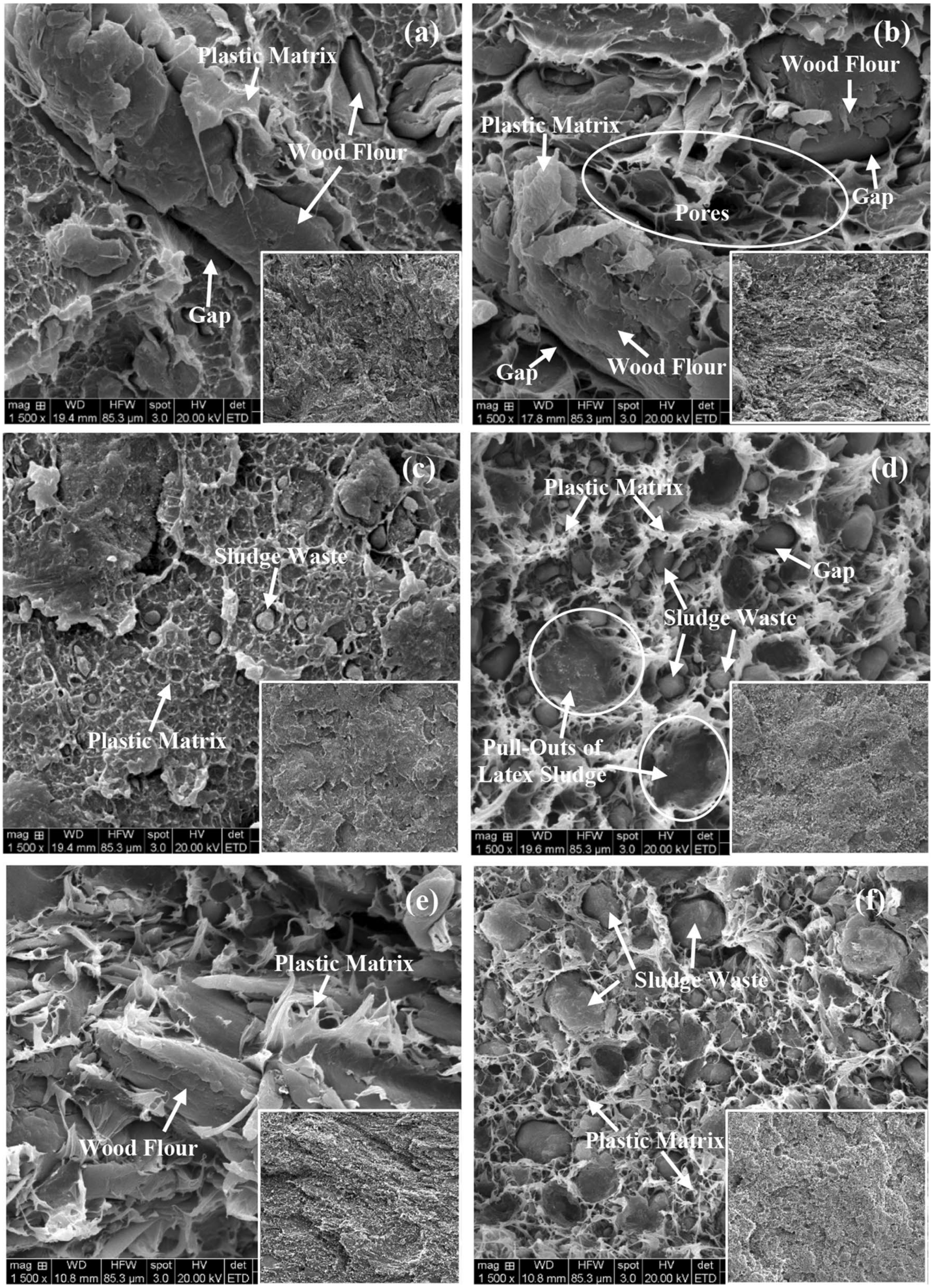

Fig. 3 Scanning electron micrographs of rHDPE composites reinforced with rubberwood flour **a** 20 wt% and **b** 60 wt%, with sludge waste flour **c** 20 wt% and **d** 60 wt%, with rubberwood flour **e** 60 wt% and MAPE 4 wt%, and with sludge waste flour **f** 60 wt% and MAPE 4 wt%

surface. Meanwhile, the interfacial adhesion between sludge waste and polymer matrix was weak due to bad compatibility. Further, a rougher surface of rHDPE/RWF composites was obvious from their SEM micrographs compared with rHDPE/SWF composites. The microstructure of rHDPE composites with 60 wt% SWF seemed to be more plastic matrix than the composites with 60 wt% RWF. In addition, the shape of rubberwood in the rHDPE composites is flakes; it is obviously different from the globular particles of the sludge waste. Thus, the rubberwood flour with a higher aspect ratio would have a higher interfacial area that led to have better stress transfer between the filler and plastic matrix [25].

The SEM micrographs of 4 wt% MAPE addition in the rHDPE composites containing 60 wt% RWF and 60 wt% SWF are shown in Fig. 3e, f, respectively. The addition of MAPE improved the interfacial adhesion and compatibility between RWF or SWF and plastic matrix compared with the composites without MAPE. The composites added MAPE had fewer voids, stronger interfacial adhesion and better dispersion of the filler in the plastic matrix. It would therefore enhance the efficiency in transferring load from matrix to fillers. In addition, it can also be observed that microstructure of the composites with 60 wt% SWF and 4 wt% MAPE had fewer and smaller pores than the composites with 60 wt% RWF and 4 wt% MAPE, which led to higher load-resistant capacity. This happened because the MAPE could also improve the compatibility between natural rubber in SWF and plastic matrix. Ponnamma et al. [26] revealed that the domain size or particle size of nitrile rubber phase dispersed in HDPE matrix decreased with an increase of MAPE concentration. Likewise, Sadasivuni et al. [27] found that maleic anhydride-grafted poly(isobutylene-*co*-isoprene) could improve the dispersion of clays in the poly(isobutylene-*co*-isoprene) composites.

Thermal stability

TGA and derivative thermogravimetric (DTG) provide important information on weight change process or weight loss and they are essential to investigate and prove the thermal stability of new materials or the polymer composites. Figure 4 shows the thermal degradation of latex sludge waste and recycled high-density polyethylene. The first 30% weight loss of latex sludge waste in the range 90–190 °C was due to moisture release while the 20% weight loss between 190 and 480 °C was due to the decomposition of rubber hydrocarbon. The remaining, which is 50% of the weight from 50 to 700 °C, were inorganic and carbon substances which did not decompose. Further, the decomposition of the rHDPE quickly occurred when the temperature reached 400–520 °C due to chain scission of rapid random carbon resulting from the formation of free radicals [28, 29].

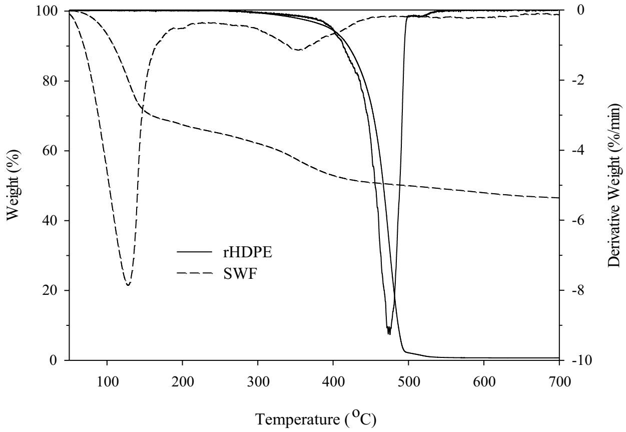

Fig. 4 Curves of TGA and DTG for rHDPE and SWF

The weight loss percentages during the heating of rHDPE/SWF and rHDPE/RWF composites clearly occurred in two states; see Fig. 5. In the rHDPE/SWF composites, the first stage, which corresponded to the decomposition (TGA) of latex sludge waste (rubber hydrocarbon), started at about 198 and 189 °C. It had the maximum derivative weight (DTG) at 349 and 342 °C for the composites with 20 and 60 wt% of SWF, respectively. In the second stage, rHDPE decomposition (TGA) occurred in range of 400–510 °C with the DTG at 473 and 469 °C for composites containing 20 and 60 wt% of SWF, respectively. The decomposition of the rHDPE was caused by the decomposition of main chains of high-density polyethylene matrix. It can also be observed that weight loss clearly increased with an increase of SWF content in the temperature range 190–455 °C because the composites with 60 wt% SWF contained higher volume of rubber hydrocarbon. However, after 455 °C, the composites with higher SWF content had larger thermal stability due to more inorganic substance.

For the rHDPE/RWF composites, the first weight loss around 257–382 °C was attributed to the decomposition of the rubberwood components (e.g., hemicelluloses, cellulose, lignin). In general, the thermal decomposition of natural wood depends on its main components; it occurs at 250 °C for hemicelluloses, at 358 °C for cellulose and at 476 °C for lignin and their ash [30]. The second weight loss occurred in the range of 383–495 °C was due to the decomposition of rHDPE. The weight loss of rHDPE grew rapidly under this temperature range. In this range, lignin in the structure of wood flour also decomposed [31, 32]. Furthermore, the thermal stability of rHDPE composites markedly decreased with increasing RWF content throughout the whole thermal degradation. This was mainly due to more decomposition of RWF than the occurrence of rHDPE [33] and due to poorer interfacial adhesion properties of the 60 wt% RWF composites. The space between the wood flour and plastic matrix

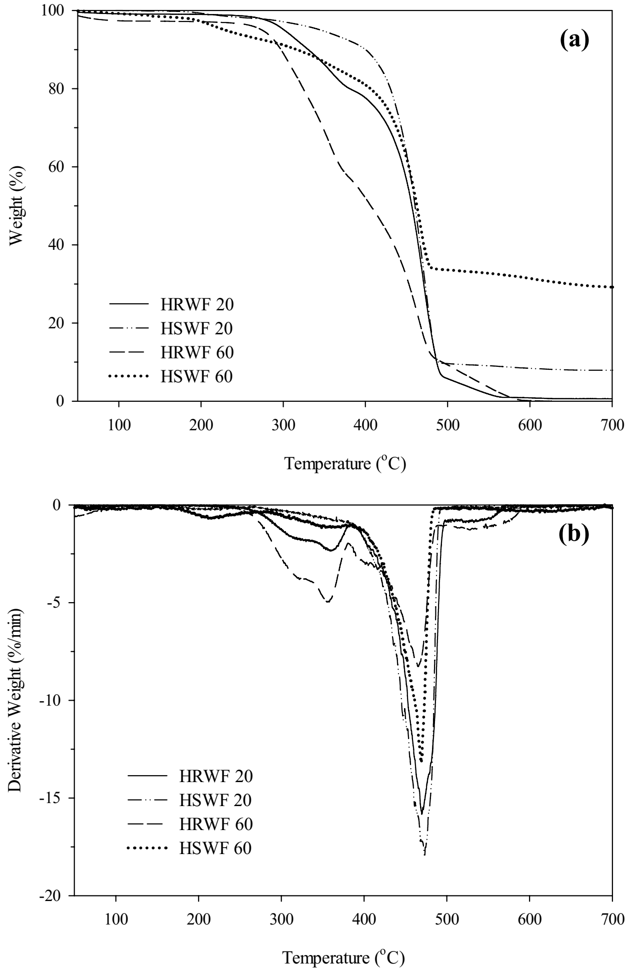

Fig. 5 Curves of **a** TGA and **b** DTG for rHDPE composites containing different fillers

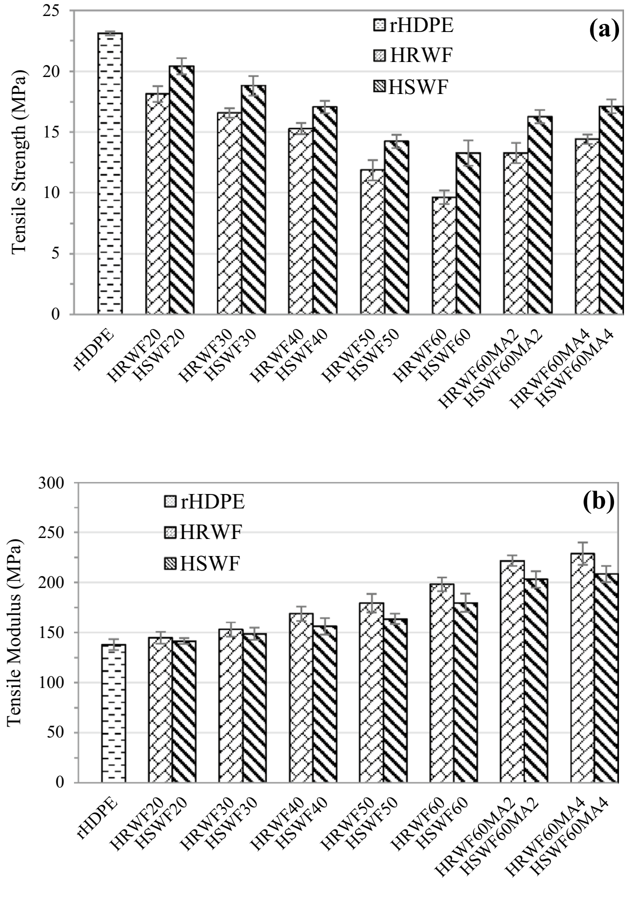

Fig. 6 Tensile property of rHDPE composites with different RWF, SWF and MAPE contents: **a** tensile strength and **b** tensile modulus

could accelerate the decomposition of volatile composites [24, 34].

Obviously, the rHDPE/SWF composites had a higher thermal stability than that of rHDPE/RWF composites for the same filler content as shown in Fig. 5a. Because latex sludge waste components consist of large inorganic substance content and have higher decomposition temperature than that of the RWF and rHDPE, the addition of SWF can improve the thermal stability of the polymer composites.

Mechanical and physical properties of the composites

Tensile properties

Variations of the tensile strength and modulus with different filler contents for rHDPE composites reinforcing with both RWF and SWF are shown in Fig. 6a, b. The TS of the rHDPE composites made from both RWF and SWF clearly reduced with an increase of filler content. It is well known that any filler or natural fiber filled into plastic matrix causes poor dispersion and weak adhesion of the filler in the matrix, which in turn reduces mechanical strengths of the composite materials. In contrast, an increment of filler contents (both RWF and SWF) in the composites slightly increases the TM because fillers have stiffer phase than the plastic matrix. Väisänen et al. [18] reported that the natural fibers had high modulus, many times than that of the plastics, so their addition in the plastic matrix increased the modulus of the composite materials. Furthermore, the composites based on SWF exhibited higher TS and lower TM than those based on RWF, for the same plastic to filler ratio because the natural rubber in the SWF improved bonding in the composite structures, resulting in an increment of load resistance. However, due to more rigid phase of the RWF, the composites containing RWF showed higher tensile modulus.

The effects of filler contents are also assessed by the ANOVA analysis. According to one-way ANOVA for the rHDPE composites reinforcing RWF or SWF in Table 2, the filler contents significantly ($p < 0.05$) affected the TS and TM of the composite materials. Tukey's test in Table 2

Table 2 Results of statistical analysis for the mechanical and physical properties of rHDPE composites with different RWF and SWF contents

Property	Filler type	Filler content (wt%)					*p* value
		20	30	40	50	60	
Tensile strength (MPa)	HRWF	18.1aF	16.6bF	15.3cF	11.9dF	9.6eF	0.002*
	HSWF	20.4aI	18.8aI	17.1bI	14.2cI	13.3cI	0.000*
Tensile modulus (MPa)	HRWF	144.9aF	153.0abF	168.9bcF	179.4cF	198.3dF	0.015*
	HSWF	141.6aF	148.8abF	156.2bcF	163.7cF	179.8dF	0.023*
Modulus of rupture (MPa)	HRWF	23.9aF	22.8bF	21.8cF	20.8dF	16.5eF	0.014*
	HSWF	23.3aF	22.7abF	22.0bcF	21.1cdF	20.8dI	0.021*
Modulus of elasticity (MPa)	HRWF	1026.1aF	1160.1bF	1273.7bcF	1326.8cF	1374.2cF	0.000*
	HSWF	919.9aI	976.9abI	1037.3bcI	1140.3cI	1321.4dF	0.000*
Maximum flexural strain (%)	HRWF	2.82aF	2.65aF	2.01bF	1.23cF	0.87dF	0.000*
	HSWF	3.60aI	3.56aI	3.45abI	2.99bI	1.39cI	0.026*
Hardness (Shore D)	HRWF	66.1aF	67.2bF	68.4cF	69.6dF	70.1dF	0.000*
	HSWF	65.4aI	65.9aI	66.7bI	67.3bI	67.8cI	0.013*
Water absorption (%)	HRWF	0.96aF	1.06abF	1.15bF	2.48cF	4.68dF	0.000*
	HSWF	0.26aI	0.39abI	0.50bI	0.73cI	0.92cI	0.018*

*The effect of filler contents is significant at $p < 0.05$. Likewise, means within each property with the same letter (suffixes a–e for effect of filler contents and suffixes F–I for effect of filler types) are not significantly different ($\alpha = 0.05$)

also verified that the rHDPE composites based on SWF with filler content of 20 wt% (suffix a) have insignificantly higher TS than the composites with filler content 30 wt% (suffix a), which is in turn have significantly higher TS than the composites with filler content 40 wt% (suffix b), which is also have significantly larger TS than the composites with filler content 50 wt% (suffix c). However, TS of the composites with filler content of 50 wt% (suffix c) is insignificantly different with filler content of 60 wt% (suffix c). Further, two-sample *t* test in Table 2 also reveals that the composites made from SWF (suffix I) show significantly higher TS than those made from RWF (suffix F) but while TM of the composites based on SWF (suffix F) is insignificantly lower those based on RWF (suffix F), for the same filler contents.

In addition, Fig. 6a, b also shows the effects of MAPE contents on the TS and TM, respectively, of the composites reinforcing with RWF or SWF. The additions of 2 and 4 wt% MAPE in the rHDPE composites containing 60 wt% RWF or SWF showed that the TS and TM increase with an increase of MAPE contents. Since chemical bond between the fillers and rHDPE polymer chains was improved, the modified filler surface increased the compatibility of hydrophilic filler and hydrophobic polymer [35, 36]. These results could be proved with morphological analysis as explained earlier that the composites with MAPE showed fewer voids and stronger interfacial bonding compared to the composites without MAPE. Likewise, in the ANOVA analysis, Table 3 indicates that the MAPE contents significantly ($p < 0.05$) affected the TS and TM of the composites with 60 wt% RWF or SWF. The addition of 2 wt% MAPE (suffix b) gives significantly higher TS and TM than the composites without MAPE addition (suffix a), while the composites with 2 wt% MAPE addition (suffix b) show no significantly lower TS and TM than with 4 wt% MAPE (suffix b).

Flexural properties

The ANOVA results in Table 2 indicate that the effects of the filler (both RWF and SWF) concentration on the flexural properties are statistically significant for rHDPE composites. The MOR, MOE and maximum flexural strain of rHDPE composites with various contents of RWF and SWF are shown in Fig. 7a, b, c, respectively. The MOR was slightly reduced with the increased proportions of RWF or SWF in the plastic matrix. These results could be explained with SEM micrographs (as shown in Fig. 3), that the composites reinforcing with higher RWF or SWF contents had more voids in composite structures and poorer interfacial bonding between filler and plastic matrix, resulting in a decrease of mechanical properties. It was also found that the composites reinforcing with SWF over 40 wt% gave larger MOR than reinforcing with RWF. However, the composites based on RWF (suffix F in Table 2) showed significantly higher MOE than those based on SWF (suffix I in Table 2) for the same filler contents since the rubberwood has higher stiffness or modulus than the sludge waste. Likewise, the increasing additions of both fillers in the rHDPE composites significantly ($p < 0.05$ in Table 2) increased the MOE. In contrast, the maximum flexural strain clearly reduced with an increase of filler contents in the rHDPE composites. Adhikary et al. [11] concluded that the composite material became stiffer with the increasing addition of filler, which resulted in a

Table 3 Results of statistical analysis for the mechanical and physical properties of RWF or SWF-rHDPE composites with different MAPE contents

Property	Filler type	MAPE content (wt%)			*p* value
		0	2	4	
Tensile strength (MPa)	HRWF60	9.6aF	13.3bF	14.4bF	0.025*
	HSWF60	13.3aI	16.3bI	17.1bI	0.028*
Tensile modulus (MPa)	HRWF60	198.3aF	221.7bF	228.8bF	0.032*
	HSWF60	179.8aF	203.2bI	208.7bF	0.029*
Modulus of rupture (MPa)	HRWF60	16.5aF	19.4bF	19.9bF	0.038*
	HSWF60	20.8aI	22.1bI	22.9bI	0.029*
Modulus of elasticity (MPa)	HRWF60	1374.2aF	1456.2abF	1509.4bF	0.044*
	HSWF60	1321.4aF	1384.1abF	1431.2bF	0.047*
Maximum flexural strain (%)	HRWF60	0.87aF	0.92abF	1.16bF	0.041*
	HSWF60	1.39aI	1.64bI	1.88cI	0.036*
Hardness (Shore D)	HRWF60	70.1aF	71.1bF	71.9cF	0.008*
	HSWF60	67.8aI	69.9bI	70.1bI	0.027*
Water absorption (%)	HRWF60	4.68aF	4.21bF	4.06bF	0.015*
	HSWF60	0.92aI	0.83abI	0.71bI	0.034*

*The effect of MAPE contents is significant at $p < 0.05$. Likewise, means within each property with the same letter (suffixes a–c for effect of MAPE contents and suffixes F–I for effect of filler types) are not significantly different ($\alpha = 0.05$)

decrease of strain at failure. This is due to reduction in the ductility of the material. The rHDPE/SWF composites showed significantly higher flexural strain than the rHDPE/RWF composites because the natural rubber in the SWF retains the elasticity of the rHDPE composites, resulting in less decrease of the strain than rHDPE/RWF composites.

Figure 7 also illustrates the effect of the MAPE contents on the flexural properties of the composites. It is clear that all mechanical characteristics (MOR, MOE and maximum flexural strain) of rHDPE composites containing the RWF or SWF are improved with the addition of MAPE. Likewise, the increasing concentrations of MAPE enhance all flexural characteristics of these composites due to the formation of ester bonds between hydroxyl groups of the natural fibers and the anhydride carbonyl groups of MAPE [11]. Furthermore, the ANOVA results in Table 3 show a statistically significant ($p < 0.05$) effect of MAPE content on the flexural properties of the rHDPE composites reinforcing RWF or SWF.

Hardness property

Figure 8 illustrates the hardness values of both RWF and SWF/rHDPE composites with different amounts of filler. It can be seen that the average hardness of both composites linearly increased with the reinforcing filler content because both fillers (RWF and SWF) have higher hardness than that of the weak plastic matrix [37] while the flexibility of the composites decreases with the increasing additions of fillers, resulting in stiffer composites [38]. Moreover, WPCs made with RWF filler exhibit higher hardness value than that with SWF filler because the SWF consists of high rubber hydrocarbon content (approximate 12.5 wt%), and it has lower hardness than that of the RWF.

The increasing additions of MAPE in both RWF and SWF/rHDPE composites increased the hardness values. The addition of 4 wt% MAPE increased the hardness values about 2.6 and 3.4% for the composites reinforcing 60 wt% RWF and SWF, respectively, compared with the composites without MAPE addition. This is because of both stronger interfacial adhesion between the filler and plastic matrix and minimization of voids [38]. Furthermore, the ANOVA results in Tables 2 and 3 show that the effects of filler contents (both RWF and SWF) and MAPE contents significantly ($p < 0.05$) affected the hardness property of WPCs.

Water absorption

Water absorption is another important characteristics of WPCs needed to evaluate, which determines their end-use applications [39]. ANOVA analysis of WA values after 24 h of the composites indicates that the independent variables of filler contents had significant effect at the 5% significance level; see Table 2. Figure 9 illustrates the WA rates of the rHDPE composites reinforced with RWF or SWF. The WA of rHDPE composites slowly increased with increasing RWF content in the range of 20–40 wt% RWF because with the increase of the wood cellulose content, there is an increase of free OH groups interacting with polar water molecules, resulting in the increasing weight gain of the composites [40, 41]. Likewise, in reinforcing 50 and 60 wt% RWF, the WA of the composites

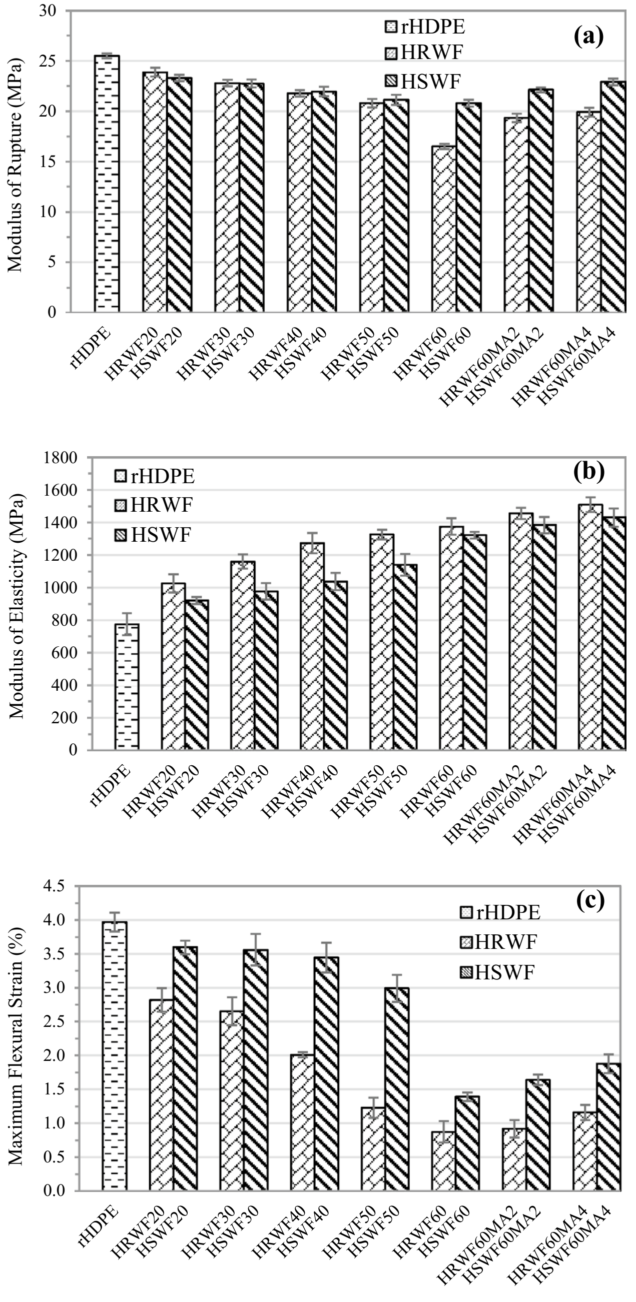

Fig. 7 Flexural property of rHDPE composites with different RWF, SWF and MAPE contents: **a** modulus of rupture, **b** modulus of elasticity and **c** maximum flexural strain

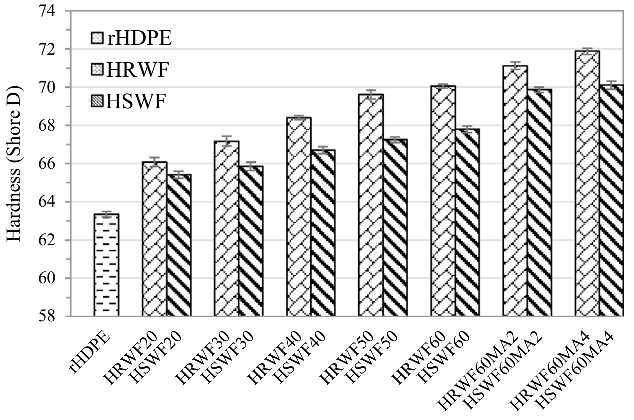

Fig. 8 Hardness of rHDPE composites with different RWF, SWF and MAPE contents

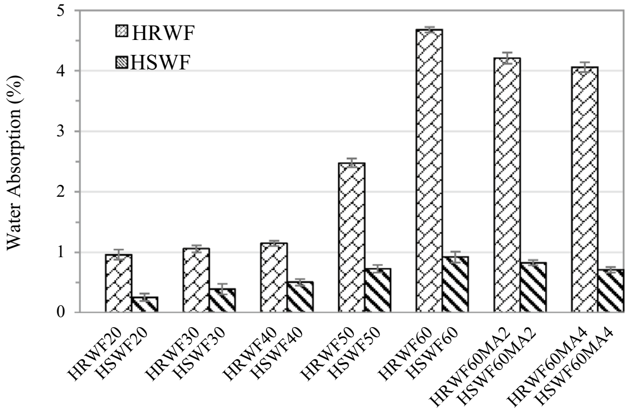

Fig. 9 Water absorption of rHDPE composites with different RWF, SWF and MAPE contents

sharply increased. For example, the composites with 50 and 60 wt% RWF absorbing the water were higher 115 and 306%, respectively, compared with the composites filling 40 wt% RWF. Because the addition of high RWF content in the composites results in large voids, there are many pores and very poor interfacial adhesion between the wood and the plastic matrix as shown in Fig. 3b. Thus, it provides more water residence sites [41]. In the rHDPE/SWF composites, the WA of the composites gradually increased with an increase of SWF contents. This is probably due to an increase of micropores and microcrack formation at the interface of SWF and plastic matrix. Generally, the composites take up a high amount of water because of capillary transport of water in the pores, which flows at the interfaces between filler and plastic matrix, and diffusion of water molecules in the microgaps between polymer chains [40, 42, 43]. Likewise, Sathishkumar et al. [44] concluded that the acceleration of WA of plastic composites consisted of four factors such as microcracks in wood flour, lumen, adhesion between wood flour and polymer matrix and hydrophilicity of wood cellulous.

The effect of filler types (RWF and SWF) on WA of the composites is also shown in Fig. 9. As can be seen, the rHDPE composites containing SWF had significantly lower WA than the composites with RWF. For example, the WA of the composites with 60 wt% RWF was higher 408% than that of the composites with 60 wt% SWF because the RWF mainly contains a high amount of hydrophilic cellulose while SWF consists of rubber and inorganic substance, which makes its WA quite low.

Figure 9 also shows that composites with the MAPE absorbed less water compared to the composites without the MAPE. The addition of 4 wt% MAPE decreased the WA about 13.2 and 22.8% for the composites containing 60 wt% RWF and SWF, respectively. This could be possible due to the improved quality of adhesion between the wood flour and the plastic matrix, resulting in fewer gaps at the interfacial region [45, 46]. Therefore, the velocity of the diffusion processes and water residence sites was reduced in the composite structures [45]. Furthermore, addition of 4 wt% MAPE in the composites reinforced with 60 wt% RWF or SWF gave a lower water absorption than the addition of 2 wt% MAPE.

Conclusions

This research revealed that sludge waste from manufacturing process of the concentrated natural rubber latex could be utilized as a filler or reinforcement in the rHDPE composites. Likewise, the rHDPE composites reinforcing with sludge waste gave better results in some mechanical, physical and thermal properties when compared with the composites with natural fiber (rubberwood flour). The composites based on SWF showed higher TS, MOR, maximum flexural strain and decomposition temperature and lower WA than those based on RWF. The TM, MOE and hardness of rHDPE composites containing SWF were obviously improved with the increasing addition of SWF; however, the TS, MOR and WA showed a negative correlation with the SWF concentration. This is because the composites reinforced with higher SWF contents exhibited more voids in composite structures and poorer interfacial bonding between filler and plastic matrix as shown in SEM micrographs. Furthermore, the mechanical and physical properties of the composites reinforcing the SWF or RWF could be improved with the addition of MAPE, which modifies the compatibility of hydrophilic filler and hydrophobic polymer. Based on the findings of this work, the rHDPE/SWF composites are suitable for applications as fencing, decking and window frames. Because these products require less water absorption for retaining their strengths and extending their lifetimes. It is thus concluded that utilization of latex sludge waste in the plastic composites presents a promising way for both solving the environmental pollution and increasing the value of this waste.

Acknowledgements This work was financially supported from the Government budget Fund (Research Grant Code: 2559A17102098) and the Rajamangala University of Technology Srivijaya (RMUTSV), Thailand. I also want to convey my thanks to Dr. Marwan Affandi from The Faculty of Engineering RMUTSV who has checked my paper.

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