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Effect of nanoclay, talcum, and calcium carbonate as filler on properties of composites manufactured from recycled polypropylene and rubberwood fiber

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HIGHLIGHTS highlights and the second second

- Effect of filler contents on rPP and rubberwood flour composites was evaluated.
- The WPCs were analyzed with one-way ANOVA and Comparing Treatment Means with a Control Method.
- The WPCs with filler contents had statistically enhanced mechanical and physical properties.
- 7 wt% content of CC was an optimum level of filler regarding overall properties of WPCs.

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ABSTRACT

The mechanical and physical properties of wood-plastic composites (WPCs) were determined as function of different contents of nanoclay, talcum, and calcium carbonate as filler. It appears that the specimens made with 7 wt% calcium carbonate content resulted in the highest strength values. The modulus of rupture of the samples linearly increased with increasing amount of filler. The WPCs specimens having 9 wt% nanoclay showed the lowest dimensional stability in the form of water absorption and thickness swelling. An addition of 3 wt% talcum content also presented the highest cooling temperature and crystallization. Content of 7 wt% of calcium carbonate was determined as optimum level of filler regarding overall mechanical and physical properties of WPCs specimens.

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1. Introduction

Forest resources are rapidly depleting in many countries due to growing demands for use of wood for furniture, manufacture, composite panel production, and structural applications. Waste material from wood manufacturing processes could be recycled to produce value-added products. Wood plastic composite (WPCs) is panel or lumber product made from recycled plastic and small wood particles or fibers. Wood plastic composites are relatively new products as compared to the long history of natural lumber or traditional wood composites such as particleboard or fiberboard.

Wood plastic composites are widely used in the United States (U.S.) and other countries. They are manufactured by mixing wood particles as fine as flour and recycled plastics. The economical demand of WPCs in the U.S. between 2000 and 2010 is expected

⇑ Corresponding author. E-mail address: thanate.r@psu.ac.th (T. Ratanawilai). to have increased to nearly 12% each year, with an annual growth of 30%. In 2011, it reached approximately \$6.5 billion. As WPCs capacity increases new products are being developed such as door stiles, rails, and window lineal $[1]$. WPCs could be produced by mixing wood flour with plastics and adhesives, then molded and formed such as by extrusion, injection molding, compression molding, and hot-press [\[2\].](#page--1-0) Additives are mixed to enable the optimized characteristics of WPCs, so that they can be lightweight, biodegradable, and recyclable $[3,4]$. The manufacturing of WPCs from recycled high density polypropylene, polyethylene, and polyvinylchloride is typical manufacturing process. It is very sustainable approach that recycled materials can be used to manufacture the value-added wood plastic composite panels [\[5\].](#page--1-0)

In general, the properties of WPCs depend on various factors, including the inherent properties of the raw materials, processing methods and interactions among these materials. In general WPCs have relatively high stiffness and strength characteristics, low density, low cost, low $CO₂$ emission, biodegradability, and

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renewability. Fiber content in the member plays an important role influencing overall processing parameters and properties of the final product [\[6–8\].](#page--1-0) The composites reinforced with wood flour have shown a great growth due to many advantages presented [\[9\]](#page--1-0). The mechanical and physical properties of plastic composites have been improved with wood wastes from various wood species including rubberwood $[10]$ and other species $[11-14]$. The quality of wood fibers depends upon many factors such as the distribution of the particles, chemical nature of the surface, shape, porosity, and content of impurities $[2,11]$. However, the major problems to manufacture composite samples are the immiscibility with different polymers and the decreasing interfacial adhesion between polymeric matrix and wood flour, resulting in the inferior composites. In order to improve the uniform mixture of the raw materials with the polymers, a third component called coupling agent is applied to the compound [\[10,12\]](#page--1-0). Different types of coupling agent, namely maleic anhydride grafted polypropylene (MAPP), ultraviolet stabilizer (UV), lubricant (Lub), and other chemicals are widely used to improve mechanical and physical properties of the WPCs [\[13–15\].](#page--1-0)

Different types of filler are also used to enhance the physical, mechanical as well as other properties of WPCs. Among many composite precursors, nanoclay, talcum, and calcium carbonate are the most commonly used as filler in plastic-based composites [\[16\].](#page--1-0) The use of nanoclay as filler in WPCs receives a wide interest as innovative materials because of its potential to enhance dimensional stability, mechanical, and physical properties, while reducing costs comparing to other types of reinforcing materials [\[12,17\]](#page--1-0). The surface characteristics of nanoparticles play a key role in their fundamental properties from phase transformation to reactivity. A dramatic increase in the interfacial area between WPCs and nanoclay filler can significantly improve the properties of WPCs [\[18\].](#page--1-0) The use of talcum also creates a positive effect on strength and modulus but may decrease ductility and toughness of filled plastic. In WPCs manufacturing, talcum is chosen to reduce material cost and improve durability and stiffness [\[16,19\]](#page--1-0). The use of calcium carbonate as a filler or an extender is prevalent [\[20\].](#page--1-0) Nano size calcium carbonate is a kind of new high-grade functionality filler with low cost, which is widely used in rubber, plastics, paint, and other industrial applications. It is a fact that the shape, size, and content of calcium carbonate can affect the overall properties of composites. Regarding size, the inorganic fillers have much smaller than the wood fibers, therefore they could be easily filled into polymeric matrix between wood fibers [\[21\].](#page--1-0) In past studies it was determined that nanoclay, talcum, and calcium carbonate provided a positive impact on strength, modulus, processing efficiency, creep, and elastic recovery performance of WPCs [\[18–21\]](#page--1-0). Although polypropylene based WPCs using rubberwood had been manufactured and their properties have been evaluated there is very limited or no information of characteristics of the WPCs having nanoclay, talcum, and calcium carbonate as filler. Therefore the main objective of this work was to manufacture experimental WPCs samples using rubberwood fiber as function of contents of above materials as fillers.

2. Materials and method

2.1 Materials

The recycled polypropylene (rPP) was used as polymeric matrix with a melt flow index of 12 g/10 min, and a density of 0.83 $g/cm³$ supplied from Withaya Intertrade Co., Ltd, Thailand for the samples. Rubberwood flour (RWF), used as the reinforcing material, supplied by a local furniture manufacturer in Trang, Thailand. The size of the rubberwood flour particles was smaller than $180 \mu m$ (<80 mesh) and dried in an oven at a temperature of 110 °C for 8 h, to reduce the moisture content of less than 3% [\[8\]](#page--1-0). Maleic anhydride-grafted polypropylene (MAPP) with a rate of 8–10% Sigma-Aldrich, Missouri, USA was used as the coupling agent. Paraffin wax as lubricant was purchased from Nippon Seiro Co., Ltd. Yamaguchi, Japan. The ultraviolet stabilizer supplied by TH Color Co., Ltd. Samutprakarn, Thailand was also used as light stabilizer additive in the samples. Nanoclay (NC) surface modified, talcum (Talc), and calcium carbonate (CC) were also added in each sample as filler.

2.2. Manufacture of composite samples

The ratios of the materials used in this study were rPP: 50.3%, RWF: 44.5%, MAPP: 3.9%, UV: 0.2% and Lub: 1% by weight [\[22\].](#page--1-0) Nanoclay, talcum, and calcium carbonate were also added into WPCs samples at the ratios of 1%, 3%, 5%, 7%, and 9% by weight to improve their mechanical and physical properties as displayed in Table 1. In the first stage WPCs pellets were manufactured before they were compounded into composite samples employing a twin-screw extruder, Model CTE-D25L40. The temperature from feeding to die zone was controlled in the range of 170 to 200 \degree C, while the screw rotation speed was fixed at 50 rpm. In the next stage the WPCs pellets were carefully dried prior to use in an oven at a temperature of 50 \degree C for 24 h. The samples were compressed in a hot-press having a temperature of 190 C at a pressure 870 psi for 30 min with sequence of pre-heating, compressing, and cooling. Finally, the specimens were machined complying with ASTM standards prior to any tests were carried out.

2.3. Mechanical properties of the samples

Tensile and flexural properties of composites were measured on an Instron Universal Testing Machine, Model 5582. The specimens were dried in an oven at a temperature of 50 °C for 24 h. prior the tests $[10]$. Tensile test for the samples was performed accordance to ASTM D 638-91 using a crosshead speed of 5 mm/ min. For flexural test, three-point bending test was followed based on ASTM D790-92 at a cross-head speed of 2 mm/min, using a span of 80 mm. Specimens for bending test had nominal dimensions of 13 mm \times 100 mm \times 4.8 mm. All tests were performed at a room temperature with five replications of each combination.

2.4. Hardness test of the samples

Hardness measurements of the samples were carried out based on ASTM D2240-91. The experiments were conducted using a mechanical Shore D Durometers, Model GS-702G from Teclock Corporation, Nagano, Japan. Five replications of the specimens with dimensions 30 mm \times 30 mm \times 4.8 mm were tested at a room temperature.

2.5. Water absorption and thickness swelling of the samples

Water absorption (WA) and thickness swelling (TS) of the specimens were carried out according to ASTM D570-88. The specimens with dimensions 10 mm \times 20 $mm \times 4.8$ mm were cut from the panels. Five replications of each combination were dried in an oven at a temperature of 50 \degree C for 24 h. until a constant weight. The weight and thickness of dried specimens were measured to a precision of 0.001 g and 0.01 mm, respectively. The specimens were then placed in water and kept at a room temperature for 11 weeks. For each measurement, the specimens were removed from the water tank, wiped off with tissue paper, and immediately weighed. The percentage of water absorption and thickness swelling were calculated using Eqs. (1) and (2),

$$
WA_t(\%) = \frac{W_t - W_o}{W_o} \times 100
$$
\n⁽¹⁾

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

$$
TS_t(\%) = \frac{T_t - T_o}{T_o} \times 100
$$
\n
$$
\tag{2}
$$

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

Table 1

Formulation of WPCs materials (wt%).

Composites code	rPP	RWF	X,	MAPP	UV	Lub
WPCs	50.3	44.5		3.9	0.2	
R43.5X _i 1	50.3	43.5		3.9	0.2	
$R41.5X_i3$	50.3	41.5	3	3.9	0.2	
R39.5X _i 5	50.3	39.5	5	3.9	0.2	
$R37.5X_i7$	50.3	37.5		3.9	0.2	
$R35.5X_i9$	50.3	35.5	q	3.9	0.2	

Note: X_i : $i=1-3$ where X_1 , X_2 and X_3 are NC, Talc, and CC, respectively. R43.5NC1 is rubberwood flour 43.5 wt% and nanoclay 1 wt%.

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