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Effects of natural weathering on the properties of recycled polypropylene composites reinforced with rubberwood flour

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ABSTRACT

The effects of natural weathering on the physical and mechanical properties of polypropylene (PP)/rubberwood flour (RWF) composites were investigated for various compositions, with different grades of plastic (virgin and recycled) and varied contents of wood flour and ultraviolet (UV) stabilizer. Composite panels were manufactured using a twin-screw extruder. Weathering sharply changed lightness (L^*) and discoloration, and slightly reduced flexural strength (MOR) and modulus (MOE) of the PP/RWF composites up to 120 days and then clearly decreased after 180 days. Virgin PP had smaller relative changes of lightness and smaller relative loss of hardness, MOR, and MOE than recycled PP (rPP), both in composites and as unfilled plastic. Increasing RWF content from 25 to 45 wt% in composites increased the change of L^* and loss of hardness, MOR, and maximum strain. Addition of 1 wt% UV stabilizer reduced change of L^* and loss of hardness, MOR, MOE, and maximum strain, compared to composites without UV stabilizer.

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1. Introduction

Wood-plastic composites (WPCs) have been gaining popularity in several applications, including door inner panels, seat backs, and headliners in automotive industry; window frames, decking, cladding, and fencing in construction; and in infrastructure as marina and boardwalk (Bajwa et al., 2011). Among these, structural applications are the largest and fastest-growing market for WPCs (Mapleston, 2001; Pilarski and Matuana, 2005) that offer low density, low cost, low maintenance, recyclability, and eco-friendliness with good mechanical properties. Moreover, softwood lumber is increasingly replaced by WPCs and plastic lumber in applications of deck-building because this improves durability (Carroll et al., 2011; Ganguly and Eastin, 2009), and the demand for WPCs is expected to have increased nearly 12% annually between 2000 and 2010 in the United States (Ganguly and Eastin, 2009).

The increasing use of WPCs in construction has resulted in concerns about long-term weatherability and durability (Fabiyi et al., 2008). Generally, the WPC products that are used in ground contact or in aboveground exterior are subject to accelerated material deterioration (Pilarski and Matuana, 2005). In ground contact, biological agents such as fungi and subterranean termites affect

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http://dx.doi.org/10.1016/j.indcrop.2014.02.034 0926-6690/© 2014 Elsevier B.V. All rights reserved. degradation (Mankowski and Morrell, 2000; Pilarski and Matuana, 2005), while in aboveground exterior there is exposure to ultraviolet (UV) rays and moisture (Matuana and Kamdem, 2002; Pilarski and Matuana, 2005). Likewise, WPCs have been reported to discolor when exposed to weathering, affecting esthetics, and to lose mechanical properties critical to performance (Matuana et al., 2001; Selden et al., 2004; Stark, 2006; Stark and Matuana, 2003, 2006). The loss of mechanical properties can stem from matrix crystallinity changes, composite surface oxidation, and interfacial degradation caused by moisture absorption (Stark et al., 2004). Therefore, when a new WPC material is developed, it is important to evaluate changes in its properties under a variety of environmental conditions such as UV light and moisture (Anwar et al., 2011), to assess effects influencing useful service life (Chaochanchaikul et al., 2013; Pilarski and Matuana, 2005).

A number of studies on the properties of WPCs under service conditions have focused on weathering (e.g. aging effects of temperature, moisture, UV light, and biological decay), and its relation to wood content, processing method, and type and content of UV stabilizer and pigments (Chaochanchaikul et al., 2013; Matuana et al., 2001; Muasher and Sain, 2006; Stark, 2006). The addition of wood flour (WF) into plastics accelerates the photodegradation of WPCs (Chaochanchaikul et al., 2013). This is attributed to the deterioration of wood's components (namely lignin, cellulose, hemicellulose, and extractives) (Dence, 1992; Muasher and Sain, 2006). Likewise, the UV component of sunlight affects







primarily the photodegradation of wood. The degradation rates of wood components greatly depend on their abilities to absorb UV light (Matuana et al., 2011). Because most wood chromophores are in lignin, it accounts for 80–95% of light absorption by wood, making its photodegradation a significant contributor to wood discoloration (Matuana et al., 2011; Müller et al., 2003). In addition, when WPCs are exposed long-term in outdoor applications, moisture or water negatively affects their properties. Moisture can accelerate photo-oxidation and mechanical property loss in WPCs by causing the wood fibers to swell, facilitating deeper light penetration into the wood, and inducing cracks in the plastic matrix (Chaochanchaikul et al., 2013; Stark, 2006). As a result the flexural strength and modulus decrease with the deterioration of interfacial bonding between natural fibers and the matrix (Chaochanchaikul et al., 2013; Stark and Matuana, 2004a). Moisture absorption can be influenced by the wood flour loading, wood particle size, and wood species (Stark and Matuana, 2004a). Although the photodegradation and mechanical property loss of WF-reinforced plastics have been extensively examined, little information is available on WFreinforced polypropylene, and there is no prior report on the photodegradation and mechanical property loss of rubberwood flour (RWF) reinforced postconsumer polypropylene that was the focus of this research.

Rubber tree (Hevea brasiliensis) is widely planted in South and Northeast Thailand. It is major economic importance because the latex extracted from these trees is the primary source of natural rubber. However, the trees become unproductive at about 25 years of age and are cut down (Rimdusit et al., 2011). Rubberwood lumber and root are mainly utilized to manufacture furniture, toys, and packing materials. In these rubberwood industries, a large amount of wood waste in the forms of flour, sawdust, and chips is generated at different stages of processing. Generally, rubberwood waste is dumped in landfills or burned, resulting in pollution issues, but some of the waste is also used to produce medium-density fiberboard and particle board (Homkhiew et al., 2013). The utilization of rubberwood waste as a reinforcement in polymer composites could decrease environmental impacts from the waste, as well as increase value of the waste material. Moreover, rubberwood waste reinforced thermoplastics also offer advantages including biodegradability, renewable character, absence of associated health hazards, low density, low cost, and low equipment wear during their processing (Ashori et al., 2013; Nourbakhsh and Ashori, 2010), when compared to inorganic or synthetic fillers (Yemele et al., 2013).

The quality of filler (wood flour or wood fiber) is an important factor affecting the photodegradation and mechanical properties of WPCs, because different wood species have different contents of cellulose, lignin, hemicelluloses and extractants (Li et al., 2008). Hence, the effects of our filler (rubberwood flour) and the selected grade of plastic (virgin or recycled PP) on the composites need to be characterized. The ultimate goal of current work was to determine the effects of material compositions (including different grades of plastic; and contents of RWF and UV stabilizer) on the physical and mechanical properties of RWF reinforced PP composites exposed to weathering tests. The new information helps target the most suitable end-use applications of such composites.

2. Materials and methods

2.1. Materials

Recycled polypropylene (rPP) pellets, WT170 with a melt flow index of 11 g/10 min at $230 \,^{\circ}\text{C}$ and load of 2160 g, were purchased from Withaya Intertrade Co., Ltd (Samutprakarn, Thailand). Virgin polypropylene (vPP) granules, HIPOL J600 with a melt flow index

Table 1

Wood-plastic composite formulations (percent by weight).

| Composite sample code | rPP | vPP | RWF | MAPP | UV | Lub |
|-----------------------|------|------|------|------|-----|-----|
| rP100 | 100 | | | | | |
| vP100 | | 100 | | | | |
| rP70R25M3U1 | 70 | | 25 | 3 | 1 | 1 |
| vP70R25M3U1 | | 70 | 25 | 3 | 1 | 1 |
| rP60R35M3U0.5 | 60.3 | | 35.3 | 3 | 0.5 | 1 |
| vP60R35M3U0.5 | | 60.3 | 35.3 | 3 | 0.5 | 1 |
| rP50R45M3U1 | 50 | | 45 | 3 | 1 | 1 |
| vP50R45M3U1 | | 50 | 45 | 3 | 1 | 1 |
| rP51R45M3U0 | 51 | | 45 | 3 | 0 | 1 |

Note: The sample codes summarize the formulation, as in rP70R25M3U1 having 70 wt% rPP, 25 wt% RWF, 3 wt% MAPP, and 1 wt% UV.

of 7 g/10 min at 230 °C and load of 2160 g, were supplied by Mitsui Petrochemical Industries Co., Ltd (Tokyo, Japan). Rubberwood flour obtained from the cutting process in local furniture industry (Songkhla, Thailand) was used as reinforcement. Its main chemical constituents were cellulose (39%), hemicellulose (29%), lignin (28%), and ash (4%) (Petchpradab et al., 2009). Before compounding, the RWF was sieved through a standard sieve of mesh size 80 (passing particles smaller than $180 \,\mu m$) and was dried in an oven at 110 °C for 8 h. Maleic anhydride-grafted polypropylene (MAPP) with 8-10% of maleic anhydride was supplied by Sigma-Aldrich (Missouri, USA), and used as a coupling agent to improve the interfacial adhesion between filler and matrix. Hindered amine light stabilizer (HALS) additive, chosen as the UV stabilizer, was supplied by TH Color Co., Ltd (Samutprakarn, Thailand) under the trade name MEUV008. A paraffin wax lubricant (Lub) was purchased from Nippon Seiro Co., Ltd (Yamaguchi, Japan).

2.2. Composite processing

WPCs were produced in a two-stage process. In the first stage to produce WPC pellets, RWF and PP were dry-blended, meltblended, and pelletized into wood-plastic composite pellets using a twin-screw extruder (Model SHJ-36 from En Mach Co., Ltd, Nonthaburi, Thailand). The 10 temperature zones of the extruder were controlled at 130-170 °C from feeding to die zone, to reduce degradation of the compositions, while the screw rotating speed was maintained at 70 rpm. In the second stage to produce WPC panels, the WPC pellets were again dried prior to use, in an oven at 110°C for 8 h. The WPC pellets, MAPP, UV stabilizer, and lubricant (formulations in Table 1) were then dry-mixed and fed into the twin-screw extruder. The processing conditions for extruding were as follows: (1) temperature profiles: 130–190°C; (2) screw rotating speed: 50 rpm; (3) melt pressure: 0.10-0.20 MPa depending on wood flour content; and (4) vacuum venting at 9 temperature zones: 0.022 MPa. The WPC samples were extruded through a rectangular die with the dimensions of $9 \,\text{mm} \times 22 \,\text{mm}$ and cooled in ambient air. After cooling, the specimens were cut according to American Society for Testing and Materials (ASTM) for physical and mechanical testing.

2.3. Natural weathering testing

The unfilled PP and PP/RWF composite specimens were cut from extrudates to dimensions dependent on the type of testing. The composite specimens were placed on the roof of 4th floors building in Hat Yai, Songkhla, Thailand, for 360 days (from July 1st 2012 to June 25th 2013). All the specimens were placed on wood exposure racks according to ASTM D1435-03, and were attached on the racks at a 45° angle, facing in a southerly direction (Chaochanchaikul

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