

# Recycling Engine Oil Containers to Prepare Wood–Plastic Composites

Yong Lei,<sup>1,2</sup> Qinglin Wu<sup>1</sup>

<sup>1</sup>*School of Renewable Natural Resources, Louisiana State University Agricultural Center, Baton Rouge, Louisiana 70803*

<sup>2</sup>*College of Engineering, Temple University, Philadelphia, Pennsylvania 19122*

Received 24 July 2010; accepted 22 January 2011

DOI 10.1002/app.34212

Published online 19 May 2011 in Wiley Online Library (wileyonlinelibrary.com).

**ABSTRACT:** The collected oil containers were drained, crushed, and used directly to make wood–plastic composites through reactive extrusion, using maleic anhydride (MA) and maleated polyethylene (PE-g-MA) as coupling agents. Oil residue in the container would be controlled at lower than 6 wt % through simple draining, and it played a role as a plasticizer in the plastics and composites. As a coupling agent, MA performed better than PE-g-MA. The comprehensive mechanical properties of oil container plastics (OCP)/wood/MA (60/40/2 w/w) composite were at the same level as those of neat plastic/wood/PE-g-MA

(60/40/3.2 w/w) system. Both PE-g-MA and MA effectively improved water resistance of the OCP/wood flour composites, especially MA. The residual oil in the composites was stable under heat to about 200°C or in the water, and it did not influence the thermal degradation behavior of the composites. The OCP was successfully used to make wood–plastic composite panels. © 2011 Wiley Periodicals, Inc. *J Appl Polym Sci* 122: 964–972, 2011

**Key words:** wood–plastic composites; polyethylene; recycling; engine oil container

## INTRODUCTION

In the United States, consumers buy and use over three billion quart bottles of motor oil every year, and around 150,000 tons of the containers, with as much as 20 million gallons of residual oil, are disposed of each year. Residual oil coating the interior surface of the “empty” motor oil containers constitutes a contaminant that prevents use of the containers. Most plastic recycling programs cannot accept the empty motor oil bottles, which can take 1000 years to decompose in a landfill.<sup>1</sup> Unfortunately, eliminating the dumping of plastics contaminated with motor oil into their local landfills has been the goal of nearly every city and county government, leaking oils into the soil and groundwater, and occupying significant landfill volume. Just one gallon of used oil has the potential to contaminate up to one million gallons of drinking water. Recycling used oil containers not only removes them from the waste stream but also has the added benefit of preventing

leaking oil containers from contaminating other recyclables.

Traditionally, plastic contaminated with oil such as used motor oil containers have been difficult to be recycled. Current available options to land filling the waste plastics include (a) grinding the containers and using them in other plastic recycling processes on a very limited (dilute) basis, (b) using an aqueous process to displace the oil from the plastic, (c) using a solvent to dissolve/dilute the oil from the plastic, or (d) blowing out the residual oil using hot air or carbon dioxide.<sup>2</sup> With these options, removal of the residual oil from the container has been expensive and prone to create additional waste byproducts.

Fillers typically absorb oil in much higher quantities compared to water. Calcium carbonate absorbs 13–21% of oil, aluminum trihydrate absorbs 12–41% of oil, kaolin absorbs 27–48% of oil, and wood flour absorbs 55–60% of oil.<sup>3</sup> Thus, compounding oil containers containing limited residual oil with oil-absorbable wood flour to make wood–plastic composites (WPCs) may be an effective and practical way to solve the problem of recycling used oil containers. However, no effort in this field has been made so far. Much work has been done in studying and developing WPCs, which have successfully proven their application in various fields such as lumber, decking and railing, window profiles, wall studs, door frames, furniture, pallets, fencing, docks, siding, architectural profiles, boat hulls, and automotive

Correspondence to: Y. Lei (yonglei168@hotmail.com) Q. Wu (wuqing@lsu.edu).

Contract grant sponsor: United States Department of Agriculture CSREES; contract grant number: 68-3A75-6-508.

components.<sup>4-9</sup> The global WPC market in North America and Europe has been experiencing double digit growth.<sup>7</sup> Maleated polyethylene (PE-g-MA) has been widely used to improve the compatibility between the hydrophobic polyethylene (PE) and hydrophilic wood flour through the esterification between maleic anhydride (MA) and cellulosic materials.<sup>10</sup> MA was also used to improve the compatibility of blends based on high-density polyethylene (HDPE) and hydrophilic poly(ethylene terephthalate) by *in situ* formed HDPE-g-MA during compounding.<sup>11</sup>

In this work, engine oil containers collected by a oil change station were simply drained, crushed, and blended with wood flour to make composites through reactive extrusion, using MA and PE-g-MA, respectively, as coupling agents. The objectives of the study are (a) to study the influence of residual oil content on the mechanical properties of the container plastics (CPs) and composites and (b) to investigate the effect of coupling agent on the morphology, mechanical properties, and water resistance of the oil container plastics (OCP)/wood composites.

## EXPERIMENTAL

### Raw materials

Castrol<sup>®</sup> engine oil containers made of HDPE collected from a local oil change station (Baton Rouge, LA). They were separated by colors into white, black, and silver containers. Natural color-recycled HDPE (R-HDPE) pellets with a melt index of 0.7 g/10 min (190°C and 2.16 kg) were supplied by Envision Plastics Company (Reidsville, NC). A maleated polyethylene (PE-g-MA) compatibilizer (G-2608) with a melt index of 8 g/10 min (190°C and 2.16 kg) and an acid number of 8 mg KOH/g was obtained from Eastman Chemical Company (Kingsport, TN). MA with purity higher than 99% was from Spectrum Quality Products (Gardena, CA). The initiator of dicumyl peroxide (DCP) and sulfuric acid with a purity of 99.999% were from Aldrich Chemical Company (Saint Louis, MO). Xylene was from Mallinckrodt Baker (Phillipsberg, NJ). Organic lubricant WP 2200 was purchased from Lonza (WilliamSPORT, PA). Pine (*Pinus* sp.) flour with a 40-mesh particle size was from American Wood Fiber Co. (Madison, MI). Pine flour was dried in an oven for 48 h at 90°C before use.

### Treatment of oil containers

OCP-containing residual oil was made by draining for 1 min followed by crushing and granulating to pass a 3-mm screen, using a Granu-grinder S10/9GF (C.W. Brabender Instruments, Hackensack, NJ).

The clean CPs were also made. Each engine oil container was first drained for 2 min and then collected and weighed. The drained container was first washed three times in a hood, using xylene at room temperature, and then was cut into several pieces and dried at 105°C for 24 h. Finally, the dried pieces were weighed and granulated.

### Preparation of blends with different oil-loading levels

For each blend, the CPs and engine oil were melt blended and extruded in a corotating twin-screw extruder CTSE-V/MARKII (C.W. Brabender Instruments, Hackensack, NJ) with a screw length/diameter of 25 : 1. The temperature profile used was 140–160–160–160–150°C, and the screw-rotating speed was fixed at 50 rpm. A strand die with two 3-mm strand openings was used. After cooled in a water bath, the strands were cut into pellets by a BT 25 Strand Pelletizer (Scheer Bay Co., Bay City, MI). Based on the plastic weight, the loading levels of engine oil were 0, 3, 6, and 9%, respectively.

Test samples for mechanical properties were made through injection molding, using a PLUS 35 injection system from Battenfeld of American (South Elgin, IL). The blends were injection molded at 170°C, with a mold temperature of 38°C.

### Preparation of wood-plastic composites

The CP or OCP, wood flour, and additives were melt blended and extruded in the twin screw-extruder CTSE-V/MARKII. The temperature profile used was 140–160–170–170–170°C, and the screw-rotating speed was 50 rpm. After cooled in a water bath, the strands of the extrudate were cut into pellets by the pelletizer. For composites containing PE-g-MA, the weight ratios of plastics to wood flour were 80/20, 70/30, and 60/40, respectively. The loading of PE-g-MA was fixed at 8% based on the wood flour weight. For composites containing MA, the weight ratio of plastics to wood flour was fixed at 60/40. The loadings of MA were 1, 2, and 3%, respectively, based on the total weight of plastic and wood flour. The loading level of DCP was 1% based on the weight of wood flour.

Test samples for mechanical properties were also made through injection molding, using the PLUS 35 injection system. The composite pellets were injection molded at 180°C, with a mold temperature of 58°C.

### Extrusion of wood-plastic composite panel

The OCP, R-HDPE, wood flour, and additives were compounded at targeted proportions through A

**TABLE I**  
**Oil Content in Containers Before and After 2-Min Drainage**

Container color	Clean container weight (g)	Drained oil weight (g)	Residual oil weight (g)	Residual oil content (%)
Black	63.9 (1.3)	24.5 (7.3)	1.8 (0.2)	2.8
White	46.6 (0.5)	26.0 (12.4)	2.7 (0.7)	5.8
Silver	46.5 (0.4)	10.5 (8.2)	1.6 (0.3)	3.4
OCP <sup>a</sup>	–	–	–	6.0

<sup>a</sup> The crushed and granulated silver oil container plastic after 1-min drainage. The values in parentheses are standard deviation. The percentage of residual oil content is based on the container weight.

Micro-27 extruder from American Leistritz Extruder Corp. (Somerville, NJ) with a temperature profile of 130–150–160–170–180–190–190–180–180–180°C and a screw-rotating speed of 100 rpm. A profile die with a size of 75 × 5 mm was used. The weight ratio of OCP/R-HDPE/wood flour was fixed at 25/25/50. MA or PE-g-MA and lubricant-loading levels were 2, and 7%, respectively, based on the total weight of plastic and wood flour. DCP was only used for composites containing MA, and its loading was 1% based on the weight of wood flour.

### Characterization

Flexural and tensile properties were measured according to the ASTM D790-03 and D638-03, respectively, using an INSTRON machine (Model 1125, Boston, MA). A TINIUS 92T impact tester (Testing Machine Company, Horsham, PA) was used for the Izod impact test. All samples were notched at the center point of one longitudinal side according to the ASTM D256. For each treatment level, five replicates were tested.

The melt flow indices (MFI) of the blends were measured (ASTM D1238) using an extrusion plastometer MP600 (Tinius Olsen, Horsham, PA) at 190°C with a load of 2.16 kg. The crystallization behaviors of HDPE in the blends were measured using a differential scanning calorimeter (DSC Q100, TA Instruments, New Castle, DE). Samples of 4–5 mg were placed in aluminum capsules and heated from 40 to 270°C at 10°C/min and melt annealed for 5 min at 270°C to eliminate the heat history before cooling at 10°C/min. The crystallinity levels corresponding to the crystallization of HDPE in blends were normalized to the mass unit of the specimens. Thermogravimetric analysis (TGA) was used to observe the thermal degradation behaviors of the blends and composites on a Thermogravimetric Analyzer Q50 (TA Instruments, New Castle, DE), in a nitrogen environment at a scan rate of 10°C/min from room temperature to 650°C.

The morphology of the wood flour-reinforced composites was studied by a Hitachi VP-SEM S-3600N (Hitachi, Tokyo, Japan) scanning electron microscope. The fracture surfaces of the specimens after impact test were sputter-coated with gold before analysis.

Water absorption and swelling tests were done through two steps. After conditioning the samples at 100°C to a constant weight, they were held under vacuum for 30 min at  $-1.0 \times 10^4$  Pa and then impregnated with water. The impregnated samples were subsequently submerged in water at room temperature. At 10-day intervals, the samples were taken out, weighed, and measured for their dimensions after the surface water was removed. Three and nine replicates were measured to determine weight and thickness, respectively. After test, the samples were conditioning again at 100°C to a constant weight and then weighted.

## RESULTS AND DISCUSSION

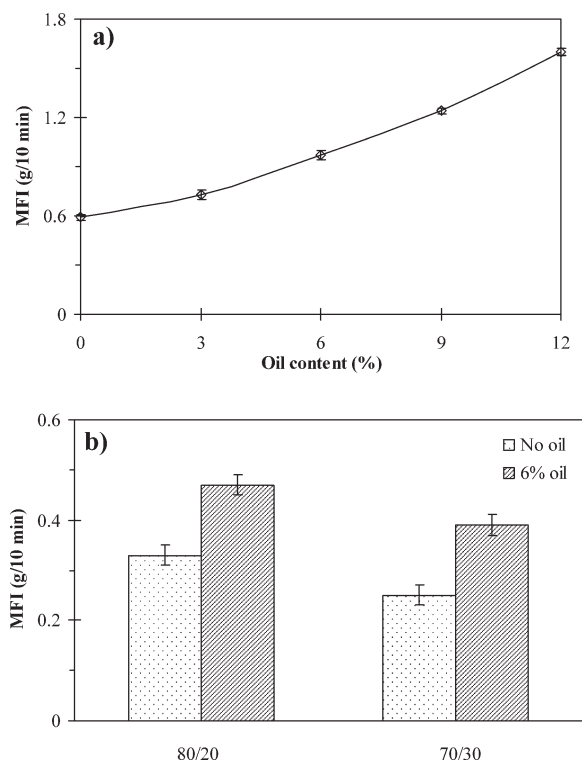
### Oil contents before and after drainage

The weights of residual oil in oil containers before and after drainage are listed in Table I. Before drainage, the average oil content was as high as above 40% for the black container, and the silver container had the lowest oil content, which was 26%, based on the CP weight. Although the oil content of containers after use depends on the person who adds the oil to a vehicle, the container size and shape, the materials and additives from which oil containers are made et al., obviously, the dumped oil containers cannot be used without any treatment because of the very high contents of residual oil.

After 2-min drainage, the average residual oil contents of the black, white, and silver containers were 2.8%, 5.8%, and 3.4%, respectively, based on the container weight (Table I). Thus, the residue oil contents could be controlled at a level of lower than 6 wt % by simple 2-min draining. These 2-min drained containers were washed by the solvent xylene to prepare clean CPs, namely CPs. The residual oil of the OCP, the crushed and granulated silver CPs, was 6%, as listed in Table I.

### Effect of residual oil on melt flowability of the plastics and composites

MFIs at 190°C of the CP of silver containers and its composites are shown in Figure 1. The MFI of CP linearly increased with the increase of oil-loading level [Fig. 1(a)]. Residual oil in the oil containers played a role of a plasticizer, which usually embed themselves between the chains of polymers, and thus increased polymer melt flowability.<sup>12</sup> The MFIs of



**Figure 1** Effect of residual oil-loading level on melt flow index of silver container plastic and its wood flour-reinforced composites. Oil percentage was based on the plastic weight.

the CP/wood flour composites were also increased by the addition of oil. As shown in Figure 1(b), the MFIs of CP/wood flour 80/20 (w/w) and 70/30 (w/w) composites were increased by 42.4 and 56%, respectively, when the 6% oil was added. Obviously, the residual oil in the composites increased the composite melt flowability as an external lubricant or plasticizer.<sup>13</sup>

#### Effect of residual oil on morphologies of the composites

The fractured surfaces of CP/wood and OCP/wood composites are presented in Figure 2. Without coupling agents, there were obvious separation between the plastic matrix and the wood fibers, as shown in Figure 2(a), because of the incompatibility between hydrophobic HDPE and hydrophilic fibers. The 6% residual oil in the matrix made the compatibility worse [Fig. 2(c)]. The addition of 3.2% PE-g-MA improved the compatibility [Fig. 2(b,d)], especially for the CP/wood composite. There still appeared obvious gap between the OCP matrix and the wood fibers [Fig. 2(d)], owing to the 6% hydrophobic oil. However, adding 2% MA significantly improved the compatibility between the OCP matrix and the wood fibers [Fig. 2(e)]. The interfacial bonding between the plastics and wood fibers was even better than that

in the CP/wood/PE-g-MA system in which there was no residual oil [Fig. 2(b,e)].

Engine oil mostly consists of hydrocarbons, organic compounds consisting entirely of hydrogen and carbon. The grafting reaction between the double bond of MA and macroradicals of polyolefins, which are also hydrocarbon compounds, by using peroxide initiators in a molten state has been largely studied and confirmed.<sup>14</sup> Thus, the double bond of MA would react with radicals of hydrocarbons in engine oil initiated by DCP, as shown in Figure 3. The compatibility between the matrix and wood phase was improved after the anhydride groups reacted with the hydroxyl groups in the wood phase to form ester linkages. The combination of radicals of hydrocarbons in engine oil with other radicals to form a new molecule with a larger molecular weight also played an important role in coupling free oil molecules.

#### Effect of residual oil on HDPE crystalline behaviors

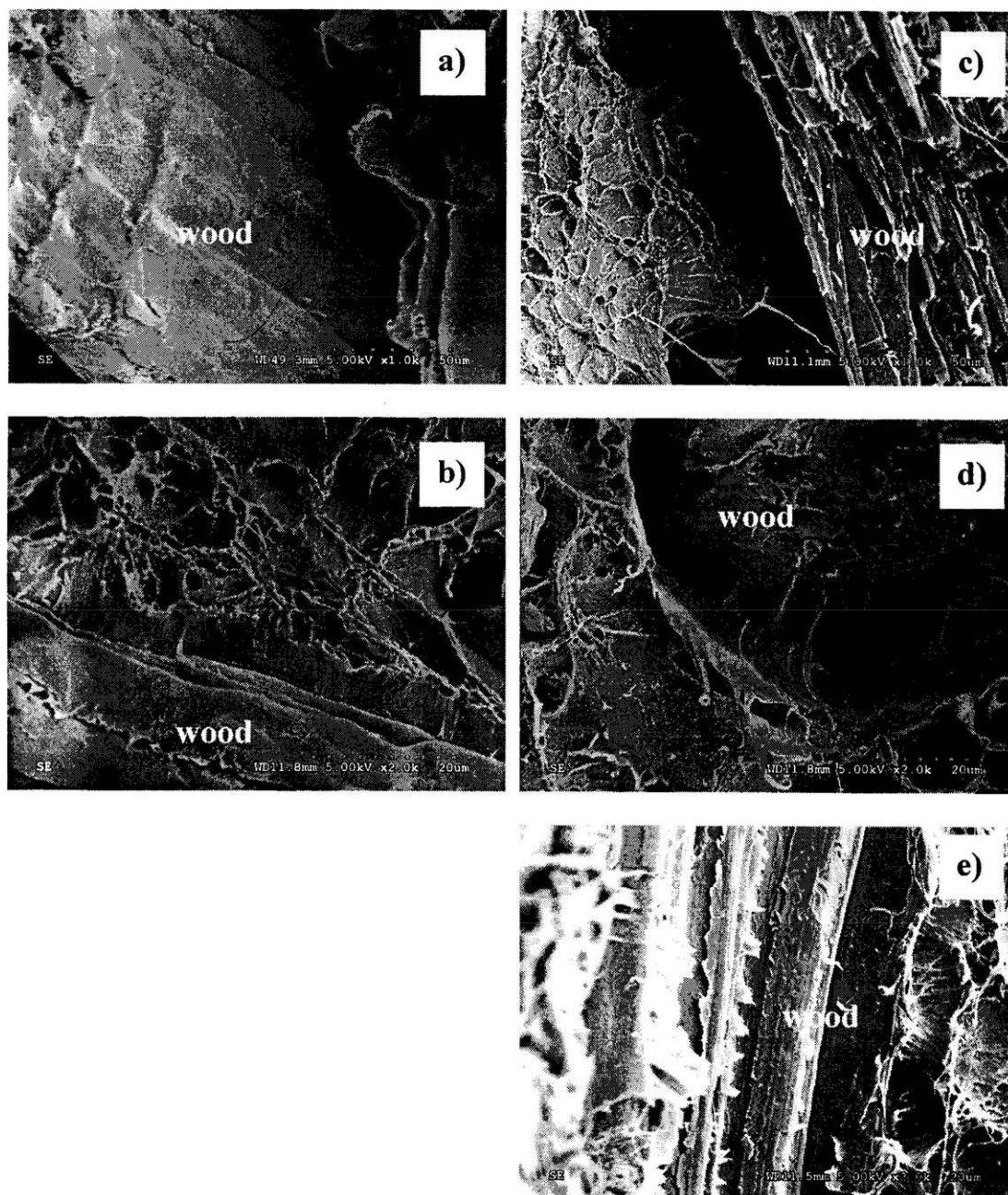
Figure 4(a) shows the DSC heating traces for the CP of silver containers, OCP, and their composites. The melting peak of OCP was wider and stronger, and the melting peak temperature ( $T_m$ ) was lower, compared to those of CP. The  $T_m$  of OCP/wood/PE-g-MA composite was also lower than that of CP/wood/PE-g-MA system. Thus, the residual oil lowered the melting temperature of the materials. Compared to OCP/wood flour/PE-g-MA composite, OCP/wood flour/MA composite had a wider melting peak and the same  $T_m$ , as shown in Figure 4(a).

After melt annealing, the DSC cooling traces at 10°C/min for the CP, OCP, and their composites were also collected and shown as Figure 4(b). The crystallization peak temperature ( $T_c$ ) found was 118.0°C for the CP and 116.1°C for the OCP, respectively. OCP had a wider and stronger crystalline peak and a lower  $T_c$  than the CP. For the CP/wood composites, the addition of 6% residual oil also lowered the  $T_c$ . The residual oil performed similarly to a plasticizer, which lower the  $T_m$  and  $T_c$  of a polymer.<sup>15</sup>

The crystallinity level ( $\chi_c$ ) of high-density polyethylene (HDPE) matrix was evaluated from the following relationship:

$$\chi_c = \frac{\Delta H_{\text{exp}}}{\Delta H} \times \frac{1}{W_f} \times 100\% \quad (1)$$

where  $\Delta H_{\text{exp}}$  is the experimental heat of crystallization,  $\Delta H$  is the assumed heat of crystallization of fully crystalline HDPE, and  $W_f$  is the weight fraction of HDPE in the samples. For fully crystalline HDPE, the heat of crystallization is 276 J/g.<sup>16</sup> The



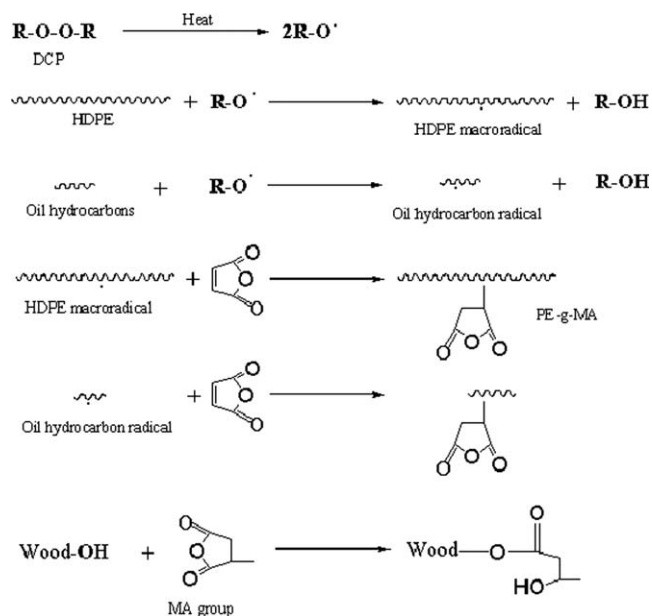
**Figure 2** The fractured surface of (a) CP/wood, (b) CP/wood/3.2% PE-g-MA, (c) OCP/wood, (d) OCP/wood/3.2% PE-g-MA, and (e) OCP/wood/2% MA composites.

corresponding results are listed in Table II. The  $\chi_c$  of CP was 44.7% and that of OCP was 52.3%. The residual oil obviously increased the  $\chi_c$ . The increased  $\chi_c$  was resulted from the improved melt flowability of CP (Fig. 1) and the wide crystalline temperature range [shown in Fig. 4(b)]. It was noticed that the HDPE  $\chi_c$  was kept at the same level after 40% wood flour was compounded with the CP. For the CP/wood/PE-g-MA composite, the HDPE  $\chi_c$  was lowered by the 6% residual oil, and the lowered value was made up when PE-g-MA was replaced by MA, as shown in Table II.

#### Effect of residual oil on mechanical properties

Table III lists the measured mechanical properties of CPs and their blends with engine oil. For the three CPs with different colors, tensile and flexural properties are almost at the same level, but the impact strength was different. The silver CP had the highest impact strength, and the black one had the lowest value.

The increase of residual oil content linearly reduced the tensile and flexural properties of CPs. The impact strength of the silver CP decreased sharply with the oil content increase up to 6% and



**Figure 3** Main chemical reactions involved in OCP/wood/MA composites during processing.

then decreased slowly. However, the effect of oil on the impact strength of the other two CPs was small. The residual oil had the same effect on mechanical properties as plasticizers for thermoplastics, which space thermoplastic molecular chain apart (increas-

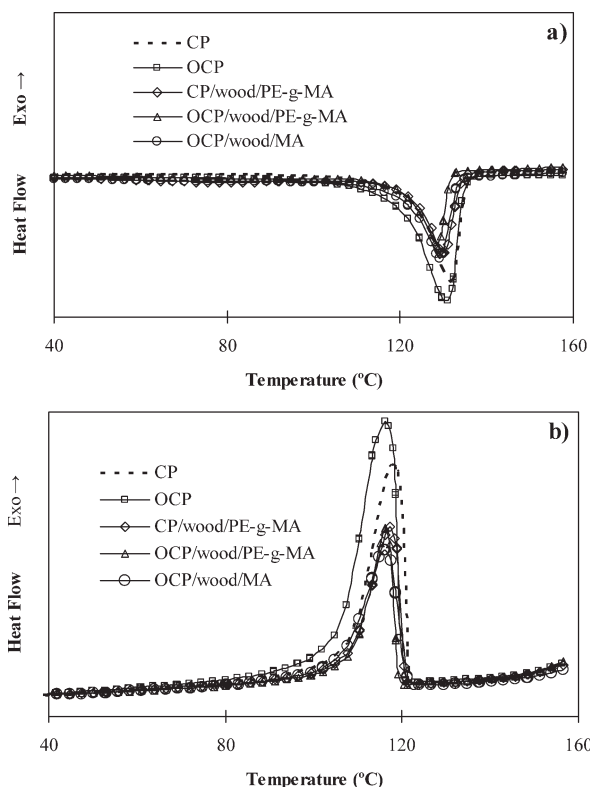
**TABLE II**  
Crystalline Level and Temperature of the Plastic and Composites

System <sup>a</sup>	Peak temp. (°C)	$\chi_c$ (%)
CP	118.0	44.7
OCP	116.1	52.3
CP/wood/PE-g-MA	117.3	44.5
OCP/wood/PE-g-MA	116.2	38.9
OCP/wood/MA	116.3	45.0

<sup>a</sup> CP, clean plastic of silver containers; OCP, oil/plastic (6/100 w/w); plastic/wood, 60/40 (w/w). Loading levels of PE-g-MA and MA were 3.2% and 2%, respectively, based on the total weight of plastic and wood.

ing of the “free volume”) and thus lower their glass transition temperatures as well as their strength and hardness.<sup>17–19</sup>

Effect of residual oil content on mechanical properties of silver CP/wood (70/30 w/w) composites is shown in Figure 5. Both the flexural strength and tensile strength of CP/wood composite were lowered by the 6% residual oil. However, the impact strength of the composite increased when the 6% oil was introduced [Fig. 5(a)]. Both the flexural and tensile moduli of the CP/wood composite were also lowered by the residual oil, especially the tensile modulus [Fig. 5(a)]. The residual oil also had the similar effects on mechanical properties as plasticizers for thermoplastics.



**Figure 4** DSC (a) heating and (b) cooling traces for the plastic, OCP, and their composites.

### MA influence on mechanical properties of the OCP/wood composites

Figure 6 shows the influence of MA content on mechanical properties of OCP/wood (60/40 w/w) composites. Both tensile and flexural strengths increased with the increase of MA-loading level, and their maxima appeared at about 2% MA. With the addition of 2% MA, the tensile and flexural strengths were increased by 49.2% and 35.7%, respectively. The addition of MA hardly changed the impact strength of the composite. The tensile modulus of the composite did not decrease until the MA-loading level increased up to 2%, and the flexural modulus decreased slowly with the increase of MA-loading level, as shown in Figure 6(b).

The comprehensive mechanical properties of OCP/wood flour (60/40 w/w) composites were enhanced by the coupling agents, as listed in Table IV. Compared to the composite containing 3.2% PE-g-MA, the composite with 2% MA had higher strength and moduli, especially the tensile strength, which was about 46% higher. Their impact strengths were at the same level. Therefore, 2% MA performed better than 3.2% PE-g-MA as coupling agents for the OCP/wood flour composite. The higher mechanical properties possibly resulted from the higher

**TABLE III**  
Mechanical Properties of the Plastics

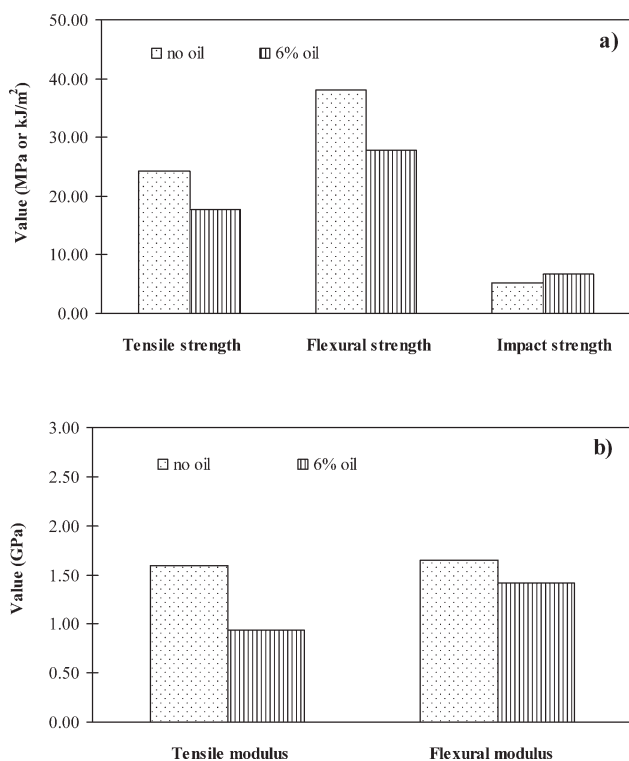
Container color	Oil content (%) <sup>a</sup>	Flexural		Tensile		Impact strength (kJ/m <sup>2</sup> )
		Strength (MPa)	Modulus (GPa)	Strength (MPa)	Modulus (GPa)	
Black	0	24.2(0.7)	0.80 (0.03)	20.6(0.3)	0.38(0.08)	14.7(0.5)
	3	19.7(0.2)	0.61(0.01)	18.9(0.5)	0.31(0.03)	14.6(0.7)
	6	17.0(0.3)	0.51(0.01)	16.7(0.1)	0.27(0.01)	14.0(0.5)
	9	14.7(0.3)	0.42(0.01)	14.3(0.1)	0.22(0.01)	13.0(0.2)
	12	12.8(0.1)	0.35(0.01)	12.9(0.2)	0.20(0.01)	13.9(0.7)
White	0	24.4(0.2)	0.78(0.01)	20.3(0.1)	0.38(0.04)	18.7(0.4)
	3	20.1(0.9)	0.58(0.04)	17.8(0.1)	0.31(0.01)	17.8(0.3)
	6	16.9(0.1)	0.47(0.01)	15.8(0.1)	0.26(0.01)	15.9(0.8)
	9	15.4(0.1)	0.43(0.01)	14.5(0.1)	0.23(0.01)	14.2(0.4)
	12	13.0(0.2)	0.35(0.01)	12.6(0.1)	0.19(0.01)	14.8(0.6)
Silver	0	23.7(0.1)	0.75(0.01)	19.9(0.2)	0.36(0.02)	39.9(2.2)
	3	19.4(0.3)	0.56(0.01)	17.1(0.9)	0.29(0.01)	25.5(0.6)
	6	17.1(0.4)	0.47(0.02)	15.7(0.1)	0.24(0.01)	19.8(0.2)
	9	14.4(0.2)	0.40(0.01)	13.7(0.1)	0.21(0.01)	18.0(0.6)
	12	12.9(0.3)	0.35(0.01)	12.9(0.1)	0.20(0.01)	17.6(0.4)

<sup>a</sup> The percentage of residual oil content is based on the container weight. The values in parentheses are standard deviation.

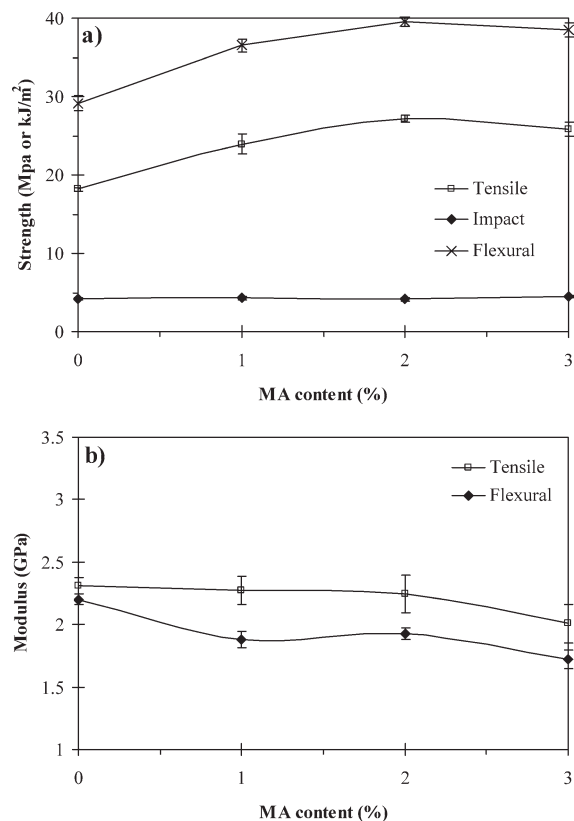
crystallinity level of HDPE [Fig. 4(b)] and better interfacial compatibility [Fig. 2(d,e)]. With the aid of DCP initiator and heat, the residual oil hydrocarbons might take part in chemical reactions during reactive extrusion (Fig. 3), resulting in less and less free oil

molecules, which influenced the mechanical properties as plasticizers.

Compared to the CP/wood/PE-g-MA (60/40/3.25) composite without any residual oil, the OCP/wood/MA (60/40/2.0) had higher tensile strength



**Figure 5** Effect of residual oil on (a) tensile, flexural, and impact strength and (b) tensile and flexural modulus of silver container plastic/wood (70/30 w/w) composites containing 2.4% PE-g-MA based on the total weight of plastic and wood.



**Figure 6** Effect of MA content on (a) tensile, flexural, and impact strength and (b) tensile and flexural modulus of silver OCP/wood flour (60/40 w/w) composites.

**TABLE IV**  
**Mechanical Properties of Wood Flour-Reinforced Composites<sup>a</sup>**

System	Flexural		Tensile		Impact strength (kJ/m <sup>2</sup> )
	Strength (MPa)	Modulus (GPa)	Strength (MPa)	Modulus (GPa)	
OCP/wood	29.2 (1.0)	2.20 (0.04)	18.2 (0.2)	2.31 (0.07)	4.2 (0.2)
OCP/wood + 3.2% PE-g-MA	32.1 (0.9)	2.15 (0.07)	18.6 (0.4)	1.88 (0.26)	4.5 (0.2)
OCP/wood + 2% MA	39.6 (0.6)	2.43 (0.03)	27.1 (0.5)	2.25 (0.15)	4.1 (0.2)
CP/wood + 3.2% PE-g-MA	40.7 (0.6)	2.48 (0.04)	23.2 (0.4)	2.33 (0.16)	4.7 (0.2)

<sup>a</sup> Forty percent wood flour was based on the total weight of plastic and wood. The percentages of PE-g-MA and MA were based on the total weight of OCP and wood flour. The values in parentheses are standard deviation.

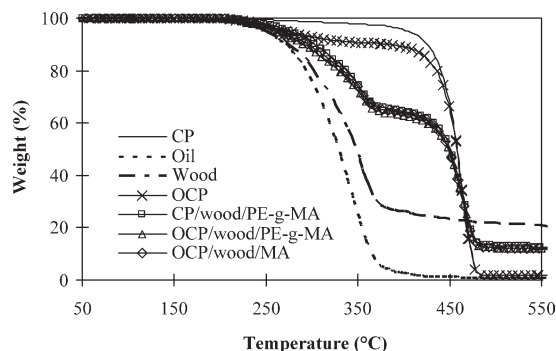
(Table IV). Their other measured mechanical properties were almost at the same level. This result would be attributed to the better interfacial bonding [Fig. 2(b,e)].

### Thermogravimetric behaviors of the plastics and composites

Figure 7 shows the TGA weight loss traces of silver CP, oil, OCP, wood flour, and their composites. The degradation of CP began at about 430°C with a degradation peak temperature ( $T_d$ ) of 466°C. The oil and wood flour had the same thermal degradation temperature range, namely around 220–370°C when heating rate was 10°C/min, and the  $T_{d5}$  were 356°C for the wood flour and 346°C for the oil, respectively. For the OCP, the initial degradation temperature was about 220°C owing to the oil degradation, and the  $T_d$  of plastic component was almost the same as that of CP, as shown in Figure 7. The weight loss versus temperature curves of composites were almost overlaid with each other. The initial weight loss of the OCP/wood composites resulted from the degradation of oil and wood flour. Obviously, the existence of residual oil did not influence the thermal degradation behavior of CP/wood composites.

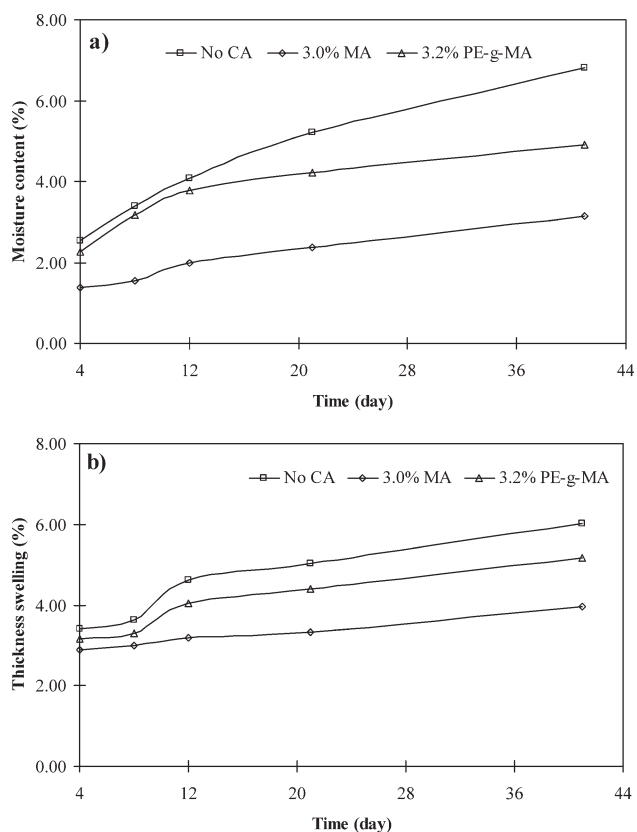
### Moisture stability of the OCP/wood composites

The influence of coupling agents on the moisture stability of OCP/wood flour composites is shown in



**Figure 7** TGA weight loss traces of oil, plastic, OCP, pine flour, and their composites.

Figure 8. Both absorbed moisture content (MC) and thickness swelling (TS) of OCP/wood flour composites was lowered when coupling agents were added, especially the coupling agent MA. After soaking 12 days, the composite without coupling agents absorbed about 4% water, while the 3.0% MA-treated composite absorbed about 2%. Although the MC values of all investigated systems increased with the increase of soaking time, the value difference between the system without coupling agents and the system containing a coupling agent was increased with the increase of soaking time, as shown in Figure 8(a). The TS values of all investigated systems also increased with the increase of



**Figure 8** Influence of coupling agents on (a) moisture content and (b) thickness swelling of OCP/wood flour composites. CA, coupling agent.



**TABLE V**  
**Mechanical Properties of Plastic/Wood Flour Composite Panels<sup>a</sup>**

System	Flexural strength (MPa)	Flexural modulus (GPa)	Impact strength (kJ/m <sup>2</sup> )
R-HDPE/OCP/wood/MA (25/25/50/2)	20.8 (1.0)	2.18 (0.13)	3.0 (0.3)
R-HDPE/OCP/Wood/PE-g-MA (25/25/50/2)	16.3 (1.5)	1.82 (0.13)	2.3 (0.2)

<sup>a</sup> OCP, oil/plastic (6/100 w/w). The percentage of coupling agent is based on the total weight of plastics and wood flour. The values in parentheses are standard deviation.

soaking time. The value difference of TS between the composite without coupling agents and composites containing coupling agents became bigger after about 8-day soaking [Fig. 8(b)].

The wetting of hydrophilic wood flour component normally plays a major role in the moisture absorption and TS of WPCs. The moisture stability of the composite largely depends on the interfacial compatibility between the hydrophobic plastic matrix and the wood inclusions.<sup>5</sup> Obviously, 3% MA was better than 3.2% PE-g-MA in improving the compatibility between components in the composite, because the composite containing 3% MA had the lowest values of TS and WC. It was also noticed that the tested composite sample almost had the same weight after conditioned at 100°C to a constant temperature, indicating no oil leakage from the composites when they were soaked in the water.

#### Mechanical properties of plastic/wood (50/50 w/w) composite panels

The main mechanical properties of the extruded composites panels are listed in Table V. Compared to the R-HDPE/OCP/wood/PE-g-MA (25/25/50/2 w/w) system, R-HDPE/OCP/wood/MA (25/25/50/2 w/w) composite panel had higher flexural strength, flexural modulus, and impact strength.

#### CONCLUSIONS

The collected oil containers were simply drained for 1 min and then crushed into small pieces to obtain recycled OCPs. The OCPs contained about 6% residual oil and were used directly to make wood-plastic composites through reactive extrusion. The residual oil played a role as a plasticizer, lowering the melting peak temperatures, lowering the main mechanical properties, and improving the plastic processibility and their wood flour-reinforced composites.

For the OCP/wood composites, coupling agent MA was better than PE-g-MA. The tensile and flexural strengths of the OCP/wood flour composite were increased by 49.2% and 35.7%, respectively, by adding 2% MA. The OCP/wood/MA (60/40/2) composite had better interfacial compatibility than the CP/wood/PE-g-MA (60/40/3.2%) system without any residual oil. The composite also had a

higher tensile strength than the latter, and their other measured mechanical properties were almost at the same level.

The oil and wood flour had the same thermal degradation temperature range, and the existence of residual oil did not influence the decomposition behaviors of CP/wood composites. Addition of both PE-g-MA and MA effectively improved water resistance of the OCP/wood flour composites, especially MA. The residual oil in the composites was stable under heat or in the water. The OCP was successful used to extrude WPC panels, and the R-HDPE/OCP/wood/MA (25/25/50/2 w/w) composite panel had higher mechanical properties than R-HDPE/OCP/wood/PE-g-MA (25/25/50/2 w/w) system.

The simply recycled OCP can be used directly to make wood flour-reinforced polymer composites by reactive extrusion molding using MA as the coupling agent. The low cost and good properties afford this technology and composites with promising potential applications.

#### References

1. Frisman, P. OLR Res Rep 2004, R-0832.
2. Smith, H. M.; Bohnert, G. W.; Olson, R. B.; Hand, T. E. U.S. Pat. 5,711,820 (1998).
3. Klyosov, A. A. Wood-Plastic Composites; John Wiley & Sons, Inc., New Jersey, 2007.
4. Nabi Saheb, N.; Jog, J. P. Adv Polym Technol 1999, 18, 351.
5. George, J.; Sreekala, M. S.; Thomas, S. Polym Eng Sci 2001, 41, 1471.
6. Clemons, C. Forest Prod J 2002, 25, 10.
7. Alireza, A. Bioresour Technol 2009, 99, 4661.
8. Matuana, L. M. J Vinyl Addit Tech 2009, 15, 136.
9. Jin, S.; Matuana, L. M. Polym Int 2010, 59, 648.
10. Lu, J. Z.; Negulescu, I. I.; Wu, Q. Compos Interface 2005, 12, 125.
11. Lusinchi, J. M.; Boutevin, B.; Torres, N.; Robini, J. J. J Appl Polym Sci 2001, 79, 874.
12. George, W. Handbook of Plasticizers; Noyes: Berkshire, 2003.
13. Li, T. Q.; Wolcott, M. P. Polym Eng Sci 2006, 464.
14. Roover, B. D.; Slavovs, M.; Carlier, V.; Devaux, J.; Lecras, R.; Montaz, A. J Polym Sci A 1995, 33, 829.
15. Jang, J.; Lee, D. K. Polymer 2003, 44, 8139.
16. Wunderlich, B.; Dole, M. J Polym Sci 1957, 24, 201.
17. Shen, M. C.; Tobolsky, A. V. Adv Chem Ser 1965, 48, 27.
18. Konstantellos, B.; Sideridis, E. Plast Rubber Compos Process Appl 1994, 22, 261.
19. Pita, V. J. R. R.; Sampaio, E. E. M.; Monteiro, E. E. C. Polym Test 2002, 21, 545.