Wood plastic composites (WPCs) from multilayer packaging wastes and rHDPE as pallets for green industry

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Abstract. People are becoming more aware of plastic pollution. Consumers, researchers, and manufacturers are seeking ways to reduce their contribution to the problem. Management of plastic wastes has still been severely environmental problems, especially those with multilayer or multicomponent structures due to the difficulty in recycling process. According to the concept of circular economy, this research aimed to overcome these problems by recycling the multilayer plastic film wastes after use from consumers (classified as post-consumer recycled resin, PCR) and transforming those into high-value products. In this work, Wood Plastic Composites (WPCs) made from mixtures of multilayer packaging wastes (PCRs), recycled high density polyethylene (rHDPE) and wood powders were studied in order to be used as pallets in green industry. Studied factors affecting WPC properties included PCR types (linear low density polyethylene (LLDPE), multilayered oriented polyamide (OPA)/PE and polyethylene terephthalate (PET)/PE films) and rHDPE:PCR weight ratios (40:5, 35:10, 30:15 and 25:20). WPC samples were compounded by a two-roll mill and then shaped into samples by a compression molding machine. Morphological, mechanical and thermal properties of WPCs were examined. From SEM study, it was revealed that wood powders and fibers were well distributed and randomly oriented in WPCs. Both PCRs (OPA/PE and PET/PE) were partially melted because the processing temperatures were below the crystalline melting temperatures of OPA and PET. PCR orientation in WPCs was aligned with the compression direction. Mechanical properties of WPCs from OPA/PE were similar to those of WPCs from PET/PE. WPCs from rHDPE:PCR weight ratios of 40:5 and 35:10 had good mechanical properties which passed the industrial target values. Optimum formulas were then used to form WPC samples by extrusion technique and fabricated into WPC pallets. It was found that the WPC pallets from wastes showed promising properties with high potential to replace conventional pallets in warehouses and industrial transport for green industry.

1. Introduction

Mismanagement and lost opportunities in the recycling of plastic wastes have resulted in global concerns, mostly related to widely spread plastic pollution of terrestrial and marine ecosystems [1]. The current management of plastic wastes largely relies on incineration and landfilling, which are typical technologies applied in a linear polymer economy. Recycling plastic wastes could promote economic and environmental benefits and drive the transition to a circular polymer economy [2]. Especially, polymer-based multilayer packaging materials are commonly used in order to combine the respective
performance of different polymers. However, because of multilayer plastic packaging wastes after use from consumers (PCRs) with their poor recyclability, most multilayers are usually incinerated or landfilled, counteracting the efforts towards a circular economy and crude oil independency [3].

Wood Plastic Composites (WPCs) have developed rapidly in recent years and are experiencing fast market growth [4]. Such products can include recycled polymers and fibers as material ingredients. WPCs also contribute to the economy by the use of recycled thermoplastics as HDPE (named as rHDPE). Using rHDPE granulated from post-consumer packaging is economically possible for WPCs. Such steps promote an increase in the value of mixed stream recycled materials and broaden the sustainability message by waste stream diversion towards a circular economy and industrial transport for green industry [5-7]. This research aimed to use mixtures of PCRs, rHDPE and wood powders for WPCs pallets. Pallets were made of wood powder/polymer with the weight ratio of 55:45. Effects of the polymer mixture of rHDPE/PCR weight ratio and PCR type were examined.

2. Experimental
2.1 Materials
Wood powders, which were screened with a 60 mesh sieve, were provided by BBJ Group Co., Ltd., Thailand. rHDPE was supplied by Iamplastic Co., Ltd., Thailand. PCRs (LLDPE, OPA/PE and PET/PE) were crushed to be about 1-5 mm length from Thai Mangkorn Plastic Industry Co., Ltd., Thailand. Calcium carbonate (CaCO₃: C-75(S) grade) from Chinchana Co., Ltd. Thailand was used as a chemical additive. Compatibilizers were given by Dow Chemical Company. Polyethylene wax (PE wax), used as a lubricant, was purchased from Multiple Plus Co., Ltd. Thailand. The anti-oxidants were from BASF Schweiz AG Plastic Additives 4057 Basel, Switzerland.

2.2 Compounding
Mixtures of polymers (rHDPE and PCR), wood powders and other additives (which were CaCO₃, PE wax, compatibilizers, anti-oxidants, proprietary information) were compounded using a two-roll mill at 190°C for 15 min with 12 rpm rotating speed. The test specimens were shaped using a compression molding machine at 190°C. The wood powder/polymer weight ratio was constantly fixed at 55:45. The WPCs formulas with various rHDPE/PCR weight ratios are presented in Table 1.

| Sample name | HD45 | HD40LLDPE05 | HD35LLDPE10 | HD30LLDPE15 | HD25LLDPE20 |
|-------------|------|-------------|-------------|-------------|-------------|
| rHDPE       | 45   | 40          | 35          | 30          | 25          |
| *PCR (LLDPE)| 0    | 5           | 10          | 15          | 20          |

*When PCR is OPA/PE or PET/PE, the sample name is HDxxPAPExx or HDxxPETPExx, respectively.

2.3 Characterization and testing
The samples were observed under scanning electron microscopy (SEM) to obtain high magnification images to show morphology of wood particles and their interaction with the thermoplastic matrix. PCRs were characterized using a thermogravimetric analyzer (TGA). Flexural properties of samples were performed according to the ASTM D790 using a universal testing machine with a test speed of 10 mm/min. Impact property of WPCs with V-notched specimens was evaluated using a Izod impact tester according to the ASTM D256.

3. Results and discussion
3.1 TGA of PCRs components
Thermogravimetric analysis of PCRs was performed under oxygen and nitrogen atmospheres. TGA results revealed that LLDPE PCR consisted of 100% PE, OPA/PE PCR was about 22% PA and 78% PE and PET/PE PCR had about 26% PET and 74% PE.

3.2 Morphology of WPCs

SEM micrographs at the 150x magnification of the cryogenically fractured surface of the WPCs from rHDPE/PCRs weight ratio of 35/10 are demonstrated in Figure 1.

![Figure 1](image1.png)

**Figure 1.** SEM micrographs (150x) of the fractured surface of the WPCs.

Figure 1(a) shows completely molten LLDPE PCR in the WPC matrix. However, OPA/PE and PET/PE PCRs were partially melted as seen film left in the WPC matrix in Figure 1(b-c) because the processing temperatures were below the crystalline melting temperatures of OPA and PET.

![Figure 2](image2.png)

**Figure 2.** SEM micrographs (1000x) of the fractured surface of the HD25PAPE20 WPCs.

Figure 2 illustrates SEM micrographs at the 1000x magnification of the cryogenically fractured surface of the HD25PAPE20 WPCs. Figure 2(a) revealed that wood in forms of powders and fibers were well distributed and randomly oriented in WPCs. In Figure 2(b), it is important to consider that fault zones can influence the properties of the WPCs. These voids or fissure could be generated between the matrix and PCRs at the interface, due to weak interfacial adhesion when compared to strength of the matrix and PCRs. Therefore, voids or holes are from adhesive failure.

3.3 Mechanical properties

3.3.1 Flexural properties

Figure 3(a) shows the flexural strength (solid line) and flexural modulus (dash line) of WPCs versus PCR loading (calculated in rHDPE/PCR part). It shows that mostly the flexural strength and modulus of WPCs made from PCR loading up to 10 wt.% are similar to those of WPCs without PCR. Moreover, the WPCs incorporated with 10 wt.% PET/PE PCR has flexural modulus 23% higher than WPCs without PCR. This was caused by the unmelted PET in PCR acted as filler that could reinforce the WPCs. As the industrial target values of flexural strength and modulus of WPCs for pallets are 45 MPa and 1800 MPa, respectively, these properties of studied WPCs made from PCR loading up to 10 wt.% meet the
industrial satisfaction. Nevertheless, the decreasing trend of both flexural strength and modulus of WPCs are presented when adding 15-20 wt.% PCR, regardless of PCR type. As seen in Figure 2(b), defects in the WPCs are observed due to weak interfacial adhesion between PCR (OPA/PE or PET/PE) and polymer matrix. Therefore, more defects could be taken place at higher PCR contents. In the case of LLDPE (as PCR), it was because of stronger rHDPE as compared with LLDPE.

![Graph](image1)

**Figure 3.** Flexural properties and impact strength of WPCs.

### 3.3.2 Impact strength

Figure 3(b) demonstrates the impact strength of WPCs versus PCR loading (calculated in rHDPE/PCR part). An increase in PCR loading (OPA/PE and PET/PE) increases impact strength of WPCs. This could be contributed to the specimen preparation technique (compression process), resulting in the alignment of unmelted OPA and PET (as PCRs) with the compression direction. Therefore, as the amount of PCRs increased, higher impact force transfer and greater absorbed energy were occurred.

### 4. Conclusion

With the WPCs having the wood powder:polymer weight ratio of 55:45, the use of polymer having rHDPE:PCR weight ratios 40:5 and 35:10 gave good mechanical properties, passing the industrial target values of WPC pallets with regardless of the PCR type studied. This research revealed that PCR, rHDPE and wood wastes could be used as alternative raw materials in the manufacture of WPCs. The designed formulas resulted in sustainable production of WPCs pallets at a lower cost with high potential to replace conventional pallets in warehouses and industrial transport for green industry.

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