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Application of recycled paper sludge and biomass materials in manufacture of green composite pallet

Sumin Kim^a, Hyun-Joong Kim^b, Jin Chul Park^{c,*}

^a Green Building Materials Lab, School of Architecture, Soongsil University, Seoul 156-743, Republic of Korea

^b Laboratory of Adhesion & Bio-Composites, Program in Environmental Materials Science, Seoul National University, Seoul 151-921, Republic of Korea

^c School of Architecture & Building Science, Chung-Ang University, Seoul 156-756, Republic of Korea

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ABSTRACT

Rice straw, rice husk and paper sludge are by-products and industrial waste, and are beneficial resources as raw biomass materials used for manufacturing value-added composite products. We investigated the effect on selected mechanical properties of adding rice straw, rice husk and paper sludge to wood composites to replace wood particles for manufacturing green pallets. Results showed that increasing the contents of rice straw and rice husk dramatically decreased the mechanical strength of the composites. This is because the wax and silicate coating of these materials obstructed the strong bonding with UF resin. When 10 wt.% of wood particle was replaced with 10 wt.% of paper sludge, the wood-paper sludge composites showed similar mechanical properties to those of wood particle. Wood particle can be replaced by 10 wt.% of dried paper sludge in accordance with the minimum requirement recommended by standards in green pallet manufacturing.

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1. Introduction

Pallets are rigid horizontal platforms that are easily transported using special equipment. They serve for storing, stacking, handling and transporting goods as a unit load. Different materials are used for the production of pallets, such as solid wood, woodbased composites, paper, plastic and metal. Wood from many hardwood species is used for the production of pallets and containers. Wood is the predominant pallet material in the U.S.; wood pallets command a market share of approximately 95%, and 97% of U.S. pallet producers manufacture only wood pallets. In the U.S., an estimated 450 million new pallets are produced annually and 1.9 billion pallets are in use at any moment in the U.S., most of them wooden pallets. While material preferences vary based on performance requirements and costs, solid wood pallets account for an estimated 90-95% of all pallets in use in the U.S. (Patricio and Maravall, 2007; Buehlmann et al., 2009; Bush et al., 1996; Kabir et al., 2003).

To produce 450 million new pallets annually, the pallet industry consumes vast quantities of resources. In 2005, it was estimated that 33% (3.8 billion board feet) of the total hardwood lumber produced in the United States was used for pallet manufacturing,

making it the single largest use of hardwood lumber (Hardwood Market Report, 2006). Ground pallet material is being used to furnish a variety of composite products. Wood composites include particleboard, hardboard, fiberboard, and insulation board. However, pallet material has little, if any, inherent advantage over other sources used to furnish these products (e.g., sawmill wastes and small diameter roundwood) and so must compete on the basis of relative delivered cost (Bush et al., 1996). Green pallets are manufactured from recycled wood pallets in the form of particleboard. A number of different wood raw materials are used for particleboard production, ranging from logs to sander dust. The use of agricultural and industrial residues to replace wood as raw materials for particleboard has received considerable attention in recent years (Wang and Xiuzhi, 2002; Papadopoulos and Hague, 2003).

Lignocellulosics from field crop residues such as cereal straw, flax straw, corn stalks, cotton and sorghum, bagasse and grass, represent a potentially valuable source of fiber which could be used either as a supplement to, or as a direct substitute for wood in the manufacture of forest products, e.g. particleboard, fiberboard or pulp for paper-based products. Approximately 2.5 billion mt of these agricultural residues are annually produced worldwide (Grigoriou, 2000). Rice straw fiber can be considered to have important potential as an alternative material for green composite because of its lignocellulosic characteristics. Global paddy production reached 628 million tons in 2005 with an additional 1% increase in 2006 (FAO, 2006). The U.S. rice production in 2006/2007 is projected to be 10 million tons (USDA, 2006). With an approximate

^{*} Corresponding author. Tel.: +82 2 820 5261; fax: +82 2 816 5261.

E-mail addresses: skim@ssu.ac.kr (S. Kim), hjokim@snu.ac.kr (H.-J. Kim), jincpark@cau.ac.kr (J.C. Park).

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Table 1	
Characteristics of the used paper sludge (oven-dry weight basis) (Yang et al., 2003)).

рН	Ash (%)	Cellulose (%)	Extractives (%)	Lignin (%)
7.78 (0.02)	27.76 (0.25)	20.83 (4.07)	6.37 (1.26)	17.41 (1.98)

rice-to-straw ratio of 1.0, an equivalent amount of rice straw (i.e., 10 million tons) is produced (Yao et al., 2008).

Straw could be available in very large amounts to new industrial applications. Amongst these, the production of particleboard panels that are at present almost exclusively produced from timber or timber by-products such as saw dust seems to be feasible. Lowdensity straw panels have already been suggested for applications in thermal and acoustic insulation. Panels that have properties corresponding to furniture industry standards have also been reported (Dalen and Shorma, 1996; Boguillon et al., 2004). The author of this present study has reported that the rice straw-wood particle composite is suitable as a sound absorbing insulation material in wooden construction (Yang et al., 2003). While a longer press time is needed and the method for coating the epidermis of straw stems causes problems in bonding straw to wood particle with conventional UF resin (Grigoriou, 2000), rice straw was used in this study as an alternative material to wood chip for green pallets. The goal of this research was to maintain more than 90% of the mechanical strength of wood chip pallets when using a combination of rice straw and wood chip in composites. Because the existing mechanical strength of pallets that comprise only wood chips was sufficiently high based on the standard, 90% of the mechanical strength will still satisfy the strength required of pallets, with the added economic value gained by replacing wood chips with rice straw as a by-product of agriculture.

Paper sludge was also considered as an alternative material to wood chips in green pallets. Paper sludge is a by-product from the pulp and paper industry. Since this sludge paper is mainly composed of short cellulose fibers and inorganic materials, there will be wide commercial application and development prospects for paper sludge as a new kind of filler in polymers. The application of paper sludge in polymer composites is also very important in terms of recycling (Ismail et al., 2005). Paper sludge is mainly composed of fibrous fines and some inorganic materials, such as kaolin clay and calcium carbonate. Thus, the use of paper sludge to produce green pallets may be an innovative way to recycle paper sludge (Taramian et al., 2007; Baziramakenga and Simard, 2001) because the primary methods of disposal for this type of sludge have been land application and landfilling. Landfilling costs in the EU are rising because of increasingly stringent regulations, taxes, and a declining capacity. With landfill space becoming scarce and expensive, some sludges are being burned or incinerated to reduce their volume and to recover part of the energy they contain (Jesús and Ochoa,



Fig. 1. Wood particles-rice straw and wood-rice husk composites as rice straw and rice husk loading contents. (a) Rice straw 5 cm: 10 wt.%, (b) rice straw 5 cm: 20 wt.%, (c) rice straw 2 cm: 10 wt.%, (d) rice straw 2 cm: 20 wt.%, (e) rice husk: 10 wt.%, (f) rice husk: 20 wt.%.

2008). However, the adverse effect of paper sludge on the mechanical properties of the particleboard produced is a serious concern (Taramian et al., 2007).

This study was conducted to evaluate rice straw and paper sludge as alternative materials for green pallets that consist of wood particles. The objective of the research was to replace wood particles with rice straw and paper sludge without reducing mechanical properties.

2. Experimental

2.1. Materials

Commercial wood particles were used in this study. For rice straw samples, after removing the top 10 cm, the rice straw stalks were cut into three sections (top, center and bottom), and rice straw particles were prepared by cutting each of the sections of the rice straw into 2 and 5 cm lengths. The particle depended on the native straw stem, which was wider at the bottom than at the top. The rice husk used as reinforcing material was supplied by Saron Filler Co., South Korea. It was crushed once after harvest and the mean particle diameter was 4–5 mm. The characteristics of the paper sludge used in this study are given in Table 1. Paper sludge was provided



Fig. 2. Bending strength of wood particles–rice straw and rice husk composites as rice straw length and loading contents.

by a local paper manufacturing company. Because paper sludge is very sensitive to fungal attacks, it was immediately dried in an oven at 100 °C for 24 h to adjust it to a moisture content of approximately 2–3%. It was then ground in a laboratory mill and screened through a mesh screen to obtain a desirable paper sludge size. Paper sludge was classified by size; small (2–3 cm \times 2–3 cm \times 1–2 cm) and large (3–4 cm \times 3–4 cm \times 1.5–2.5 cm). Commercial UF resin adhesive (65 wt.% of solid content) was used as the composite binder added with 10 wt.% NH₄Cl solution as a hardener.

2.2. Methods

2.2.1. Fabrication of wood-rice straw composite and wood-paper sludge composite

Wood-rice composite of straw boards $270 \text{ mm} \times 270 \text{ mm} \times 10 \text{ mm}$ (length \times width \times thickness) were manufactured at a specific gravity of 0.85 with rice straw contents of 0, 5, 10, 20, 25 and 30 wt.%. Rice straws were cut as above to examine the effect of rice straw particle width (as the straw width), and length. The wood particles and rice straws were placed in a rotary drum mixer and 10 wt.% (based on the weight of the oven dried raw material) of commercial UF resin adhesive used as the composite binder was sprayed onto them while rotating the mixer. The mixture was cold pressed at 10 kg/cm^2 for 2 min to ensure the stability of the mat and to obtain the proper density gradient of the composites prior to hot pressing. The mixture was then hot pressed to form composite boards at a peak pressure of 100 kg/cm² and temperatures of 180 °C. The press time was 3 min, and the pressure was released in two steps of 1 min each. The fabricated composite boards were pre-conditioned at 25 °C and 65% RH for two weeks before testing. Wood-rice husk and Wood-paper sludge composite boards were manufactured using the same method. Rice husk loading contents were 10 and 20 wt.% while paper sludge loading contents were 10, 15 and 20 wt.%.

2.2.2. Mechanical properties

3-point bending strengths were determined using a Universal Testing Machine (Zwick Corp.) using the ASTM D 1037 method (1999). Each value represents the average of ten samples.

2.2.3. Thermogravimetric analysis (TGA) and contact angle study

For the TGA test of paper sludge, 10 mg of dried paper sludge sample was placed on a balance located in the furnace. Heat was then applied in the temperature range of room temperature to 800 °C using a Thermogravimetric Analyzer (Rheometric Scientific TGA 1000). High-purity nitrogen gas consisting of 99.5% N₂ and 0.5% O₂ was used as the inert purge gas to displace air in the pyrolysis zone in order to avoid unwanted oxidation of the sample. The heating rate was 10 °C/min.

Due to the concern for the water absorption of paper sludge, the contact angles of paper sludge and wood particles were measured. An image analysis system calculated the contour of the drop from an image captured by means of a video camera. About 10–12 measurements of the sessile drop contact angle (SEO 300A, Surface & Electro-Optics Corp.) were taken on each of the three drops of distilled water per liquid placed on the sample. The temperature during measurements was 23 ± 1 °C and the relative humidity was $55 \pm 3\%$.



Fig. 3. Wood-paper sludge composites as paper sludge loading contents. (a) Dried paper sludge: 10 wt.%, (b) dried paper sludge: 20 wt.%.

3. Results and discussion

3.1. Wood-rice straw and wood-rice husk composites

Fig. 1 shows wood-rice straw and wood-rice husk composites in terms of rice straw and rice husk loading contents; a) Rice straw 5 cm: 10 wt.%, (b) Rice straw 5 cm: 20 wt.%, (c) Rice straw 2 cm: 10 wt.%, (d) Rice straw 2 cm: 20 wt.%, (e) Rice husk: 10 wt.%, and (f) Rice husk: 20 wt.%. 5 cm of rice straw was classified as shown in Fig. 1a and b, while 2 cm of rice straw was too difficult to classify. Although the wax and silicate coating of straw obstructs the development of bonding with UF resin (Grigoriou, 2000), each composite was well processed because pallets are required to withstand high pressure. However, the 3-point bonding strength data shown in Fig. 2a reveals a decrease in bonding strength. The replacement of wood particle with rice straw and rice husk resulted in a decreasing bending strength as the loading content was increased.

The case where the rice straw was shorter showed much lower strength than the case of the longer rice straw. However, rice husk, which is the smallest of the alternative materials, showed the highest bending strength at a loading content of 20 wt.%. In this study, the replacement of wood particle with rice straw and rice husk was not expected to result in an increase in the mechanical properties of the composites. The goal was to remain within a 10% reduction rate of mechanical properties when wood particles are replaced with alternative materials. As shown in Fig. 2b, the reduction rate of bending strength was more than 50% at a 10 wt.% loading content in both cases of rice straw and rice husk. Furthermore, at a 5 wt.% loading content for the case of shorter straw there was already more than a 50% reduction rate. It is therefore demonstrated that because there was no advantage in replacing wood particle with rice straw and rice husk, rice straw and rice husk are unacceptable as alternative materials for green pallets.

3.2. Wood particle-paper sludge composites

Paper sludge was considered as another alternative cellulose material. Fig. 3 shows wood-paper sludge composites for the paper sludge loading contents of 10 and 20 wt.%. Magnified figures show the surface of the wood-paper sludge composites. In contrast to the rice straw and rice husk, it was difficult to classify these alternative materials according to the surface of the composites. The mechanical strength of wood-paper sludge composites also decreased as the loading content of paper sludge increased, as shown in Fig. 4a. In particular, in the case of small sized paper sludge, the mechanical strength decreased more than in the case of larger sized paper sludge. The reason for this behavior is attributed to the weak adhesion between the paper sludge and wood particles due to the presence of inorganic materials such as kaolin clay and calcium carbonate, in the paper sludge. Furthermore, paper sludge mainly contains short fibers that cause a decrease in the bending strength of the boards (Taramian et al., 2007). Based on the American National Standard for Particleboard (ANSI A208.1, 1998) and European Standard (EN 312-2, 1996), the minimum requirements for bending strength of particleboard panels for general uses are 11 and 11.5 MPa, respectively. The bending strength of the UF-bonded particleboards containing 10 and 15 wt.% paper sludge satisfied the minimum required by ANSI A208.1 and EN 312-2 standards (Taramian et al., 2007). However, the reduction rate of paper sludge loading was much lower than that of rice straw and rice husk. From the data shown in Fig. 4b, it was found that a reduction rate of less than 10% appeared when there was a 10 wt.% of replacement samples. In particular, in the case of larger sized paper sludge, the bending strength did not decrease. Finally, a 10 wt.% of replacement was successful when large sized dried paper sludge $(3-4 \text{ cm} \times 3-4 \text{ cm} \times 1.5-2.5 \text{ cm})$ was used as an alternative material for green pallets.

In order to utilize paper sludge as an alternative material for wood particle in wood-paper sludge composites, thermogravimet-



(b) Reduction rate of bending strengh

Fig. 4. Bending strength of wood particles–paper sludge composites as paper sludge size and loading contents.



Fig. 5. TGA data of dried paper sludge.

ric analysis needed to be carried out at the probable temperatures for composite manufacture. The hot press temperature was $180 \,^{\circ}$ C. The thermogravimetric behaviors of paper sludge in a nitrogen atmosphere are shown in Fig. 5. The weight change by thermal decomposition of paper sludge at $180 \,^{\circ}$ C was less than 5%. When a 2–3% moisture content for dried paper sludge was considered, paper sludge was sufficient to be used as an alternative material at a high temperature of $180 \,^{\circ}$ C. The first certain thermal composition began at $300 \,^{\circ}$ C. This is the thermal decomposition of cellulose in paper sludge (Kim and Eom, 2001).

From the previous work, the water absorption of particleboard increased as the content of added paper sludge was increased. However, at a lower loading contents level of 15 wt.%, the water absorption decreased because the paper sludge fills up the pits of the boards (Taramian et al., 2007). Therefore, when the replacement of paper sludge was 10%, the water absorption of paper sludge was insignificant in the wood–paper sludge composite. For alternative materials in wood particleboards, the contact angle was measured to check the water repellent property. As shown in Fig. 6, the contact angle on the surface of paper sludge was much higher than that on the surface of wood particle. Although the contact angle decreased with contact time, the contact angle on the wood particle surface.



Fig. 6. Contact angle of wood particle and paper sludge.

4. Conclusion

The results of this study indicate that the mechanical properties of wood-paper sludge composite are positively affected by the use of paper sludge as an alternative material to wood particles in the production of green pallets. In particular, a 10 wt.% replacement with large sized $(3-4 \text{ cm} \times 3-4 \text{ cm} \times 1.5-2.5 \text{ cm})$ paper sludge showed a similar mechanical strength to that of wood particle composite only. However, the replacement with rice straw and rice husk resulted in a decrease in mechanical strength as the loading content was increased. The replacement with paper sludge did not result in a weight loss at the temperature of composite manufacturing and water absorption due to the presence of inorganic materials in paper sludge such as kaolin clay and calcium carbonate. These results suggest that controlled mixing of wood particle and dried paper sludge can generate a suitable wood-paper sludge composite replacement material for limited wood particleboard as green pallet bonded by formaldehyde-based resin. When the mechanical properties for the pallet materials were considered, 10 wt.% of dried paper sludge was the optimum adding content for wood-paper sludge composite green pallets.

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