Improvement of Biodegradation of Wood Plastic Composites using Rice-Bran Mixture

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Wood-plastic composites (WPCs) are currently discarded using incineration treatment, which is very expensive. Hence, this study was performed to improve the biodegradation of WPCs, such that they could potentially be buried after use, and to estimate their bending strength. A biodegradation test (determining the ultimate aerobic biodegradability of plastic materials under controlled composting conditions) was performed according to ISO 14855-1. Two groups of specimens were prepared using rice-bran mixture as the bioresource. One group contained rice-bran mixtures of 5, 7.5, and 10 wt.% instead of wood flour contents, and another group contained rice-bran mixtures of 8, 16, and 24 wt.% instead of the talc component. During the 20 days of the biodegradation experiment, the WPC (control) showed 18% biodegradation, and 7.5%-rice-bran-mixture-added specimen showed the highest biodegradation of 32%. Furthermore, the bending strength (MOR) was increased by up to 140% by adding rice-bran mixture as a biodegradable component. Therefore, the rice-bran mixture improved the biodegradation and mechanical properties of WPCs.

Keywords: Wood-plastic Composites; Ultimate biodegradation; Ultimate aerobic biodegradability; Rice-bran mixture; Composting

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INTRODUCTION

Wood-plastic composites (WPCs), which are composed of a mixed material of lignum reinforcement of wood flour and wood fiber and thermoplastic of polypropylene and polyethylene, find their maximum use among current wood-based materials. WPCs have dimensional stability, thermal and moisture stability, and high impact resistance. WPCs are commonly used for window framing, exterior materials, decking construction, automobiles, and flight materials for lightening the weight. However, WPCs are difficult to discard after use because these materials contain synthetic materials such as polypropylene (PP), polyethylene (PE), and polyvinyl chloride (PVC). Thus, WPCs are currently being discarded in incineration facilities.

For the natural burial of WPCs and other plastic materials, biodegradable plastics of polycaprolactone (PCL) and polylactic acid (PLA) were developed to replace existing thermoplastics (PP, PE, *etc.*). However, biodegradable plastics are too expensive (the exact cost of biodegradable plastics could not be obtained) to use as a component for WPCs. Thus, WPCs are difficult to bury (Endres and Siebert-Raths 2009).

The manufacture of ultimate biodegradable WPCs is possible, and biodegradable and recycled polymers are more eco-friendly materials than virgin polymers (Bergman *et al.* 2013).

Many recent studies have reported the improved biodegradation of WPCs without the use of high-cost biodegradable plastics. Significant amounts of wood flour, polymer, and additives (*e.g.*, antioxidants) have been applied in order to enhance the biological durability of WPCs (Chow *et al*. 2002; Verhey *et al.* 2002; Silva Guzman 2003; Klyosov 2007; McDonald *et al*. 2009; Morrell *et al*. 2010). The ratio of wood flour to plastic matrix ratio determines the biological durability of WPCs (Silva Guzman 2003). In addition, adding fish and by-product flour to WPCs can improve biodegradation. This research investigated WPCs with such additives buried under soil for 120 days, and measured the mass loss after 20 days. The test specimens were observed to have lost mass, which verified that fish and by-product flour could improve the biodegradation of WPCs (Ali *et al.* 2014).

This study used rice-bran, which is one of the by-products of the polishing process, and iron sulfate mixture instead of WPC pigment and part of wood flour to improve biodegradation. WPC specimens were evaluated over a range of rise-bran contents. Biodegradation was tested according to ISO 14855-1. Bending strength was also tested as a function of WPC composition.

EXPERIMENTAL

Materials

For this study, WPC was manufactured in two steps. The first step involved manufacturing WPSs in a compound pellet form. The second step consisted of making WPCs as complete products.

WPC raw materials

Generally, wood flour and plastic matrix are the main materials for WPCs, and the coupling agent (GE300; Melt flow index: 2.3 g/min) and additives (*e.g.*, antioxidants (Shimassord 2020)) are minor materials. Softwood flour (less than 60 mesh) was used, and the moisture content was less than 6%. Commercialized polypropylene (Y120; melt flow index: 2.0 g/min; density: 0.96 g/cm^3) and polyethylene (B230A; melt flow index: 0.33g/min; density: 0.96 g/cm^3) as thermoplastic resins. The ratio of PE and PP is 2.2 : 1.

Commercialized rice-bran mixture was used for this study. This material is a rice-bran and iron sulfate mixture (30% of rice-bran and 70% of iron sulfate) which is used as a soil conditioner.

Compound pellet manufacture

Compound pellet for this study was manufactured in Ilsam Co., Ltd. (Eum sung, Korea). First, the wood flour, plastic matrix, and additives were mixed in a Hanschel mixer under 180 °C, and it was cooled down in a cooling mixer until 115 °C. The next stage is compressing (92 mm twin conical extruder, Cincinnati) under a temperature of 150 to 175 °C. Then, the products were cut using a screw cutter for pelletizing. After screw cutting, the compound pellet was cooled by air-cooling. A schematic for compound pellet manufacturing is shown in Fig. 1.

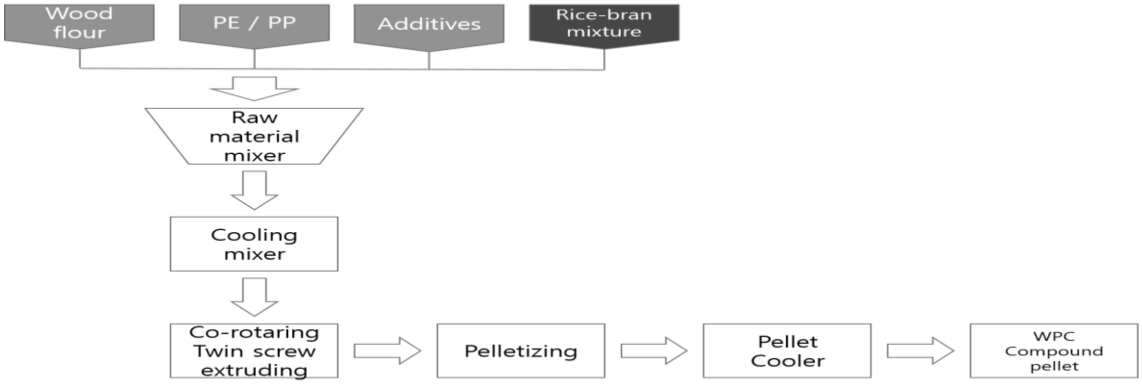

Fig. 1. Process of WPC compound pellet manufacturing (Ilsam Co., Ltd.)

WPC manufacture

WPCs were manufactured with a batch-type extruder using a compound pellet in Jin-seong Co., Ltd. (Ansan, Korea). The compound pellet was fed at 55-rpm feeder speed, and compressed in a mold under a temperature of 250 °C. Molded WPCs were cooled in a water bath for 2 to 3 min. The dimensions of the final products were 150(w) × 25(T) × 1222 (L) mm^3. Figure 2 shows the process of manufacturing of WPC complete product.

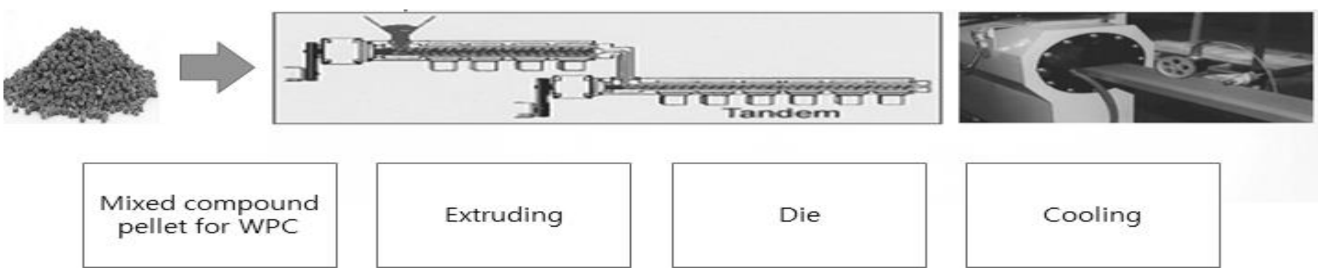

Fig. 2. Process of WPC manufacturing (Jin Seong Co., Ltd.)

The compositions of composite specimens, which were used to determine the optimal composition for biodegradable WPCs, are shown in Table 1. The manufacturing process was described in the Experimental section.

As shown in Table 1, two different groups of specimens were manufactured.

Table 1. Components of Composite Specimens

No.	Polymer	Wood Flour	Rice-bran Mixture	Talc	Additives	Density (g/cm^3)	Moisture Content (%)
WPC	24	55	-	8	13	1.20	0
RM5*	24	50	5	8	13	1.23	0
RM7.5*	24	47.5	7.5	8	13	1.20	1
RM10*	24	45	10	8	13	1.20	1
NTRM8**	24	55	8	-	13	1.22	0
NTRM16**	24	55	16	-	13	1.20	0
NTRM24**	24	55	24	-	13	1.21	0

* RM: Instead of wood flour content, rice-bran mixture is added
**NTRM: Instead of talc content, rice-bran mixture is added (wood flour content is maintained)

One group (RM) contained rice-bran mixture instead of the wood flour component, and another group (NTRM) contained rice-bran mixture instead of the talc, while maintaining the wood flour content. Table 1 shows that the density and moisture contents after manufacturing were almost the same for the manufactured WPC specimens.

Methods

Bending strength test

A bending strength test was performed according to ASTM D 790:10 with three-points loading using a universal testing machine (H50KS UTM; Hounsfield Co., Ltd.) at room temperature. A crosshead speed of 10 mm/min, a span length of 500 mm, and a load of 50 kN were applied for the test.

Biodegradation test

A biodegradation test was performed according to ISO 14855-1. All specimens were subjected to a degradation experiment in soil and water, as shown in Fig. 3. The incubation conditions were as follows: 58±2 °C, dark or diffused light, and free from vapors inhibitory to microorganisms. The duration of the experiment was 20 days, with an interval for titration every 24 h. For this experiment, thin-layer chromatography (TLC)-grade cellulose with a particle size of less than 20 µm was used as the control reference material.

Potassium hydroxide (0.4 N) and barium chloride (2.0 N) solutions were mixed to dissolve carbon dioxide after biodegradation. After the addition of phenolphthalein, until the pink is changed colorless titrated with 0.2N hydrochloric solution while stirring.

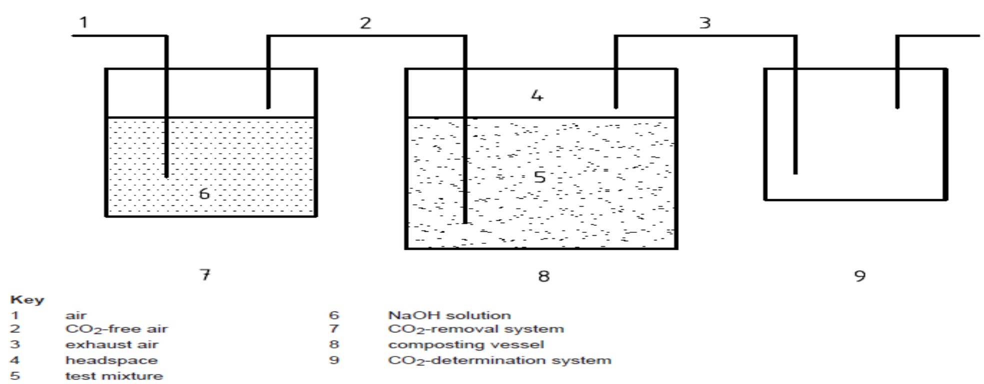

Fig. 3. Layout of the biodegradation testing system (ISO 14855-1)

Scanning electron microscopy (SEM)

The surface biodegradation of the manufactured and biodegradation-tested composites were examined using SEM (EM-30 COXEM) under vacuum-pressure conditions at a temperature of 23 °C. The specimens were coated with a thin layer of Au about 20 nm thick before conducting the SEM. The results of SEM of different types of specimens are presented in the Results and Discussion section.

Statistical analysis

Analysis of variance (ANOVA) was conducted ($p < 0.05$) to evaluate the effect of rice-bran mixture on the bending strength of the WPCs. Significant differences among the average bending strength the WPC groups were observed using Duncan's multiple range test. The SPSS 11.5 package program was used for the statistical analyses.

RESULTS AND DISCUSSION

Biodegradation of WPC Components

Before the biodegradation experiment of WPCs, each components of WPCs' (*e.g.* wood flour, PP, PE, rice-bran mixture) biodegradation was tested. As shown in Fig. 4, for the same duration, biodegradation of the reference material of TLC-grade cellulose was approximately 40%, while that of the rice-bran mixture, wood flour, and plastic matrix were 170%, 21%, and 13%, respectively.

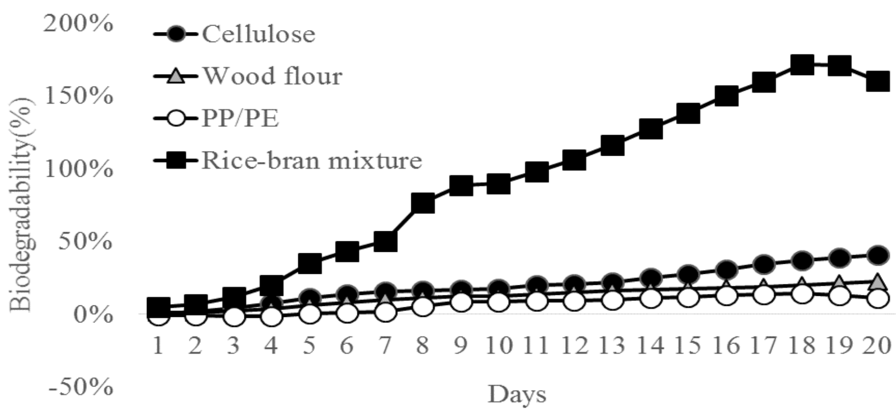

Fig. 4. Biodegradation results of components

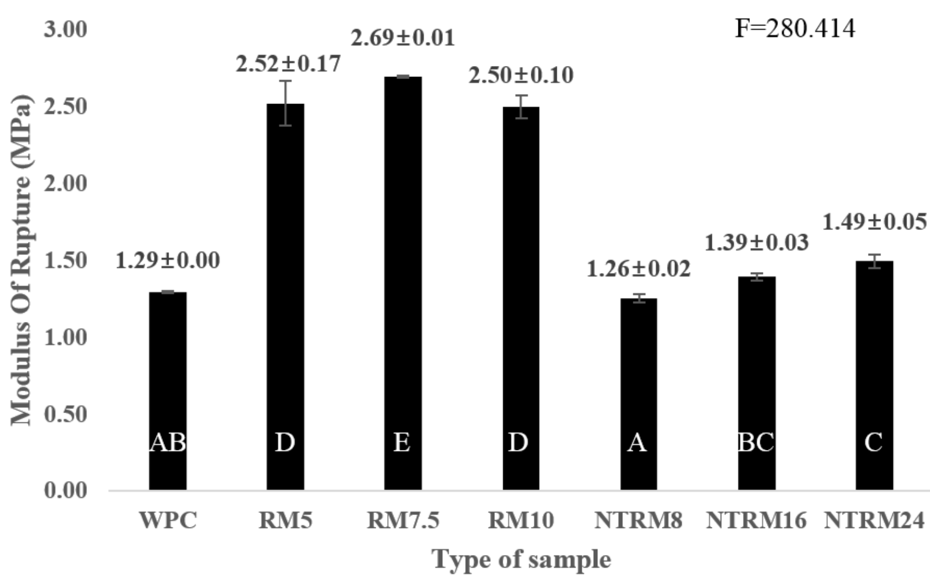

Fig. 5. Modulus of rupture values of different specimens

Bending Strength

The results of bending-strength measurement showed that the WPCs that had been prepared with the addition of rice-bran-mixture had an approximately 130% improvement in strength over the WPC control. In WPC, wood flour is a hydrophilic radical material, but plastic is a hydrophobic material.

Because of the different properties of the main raw materials of wood flour and plastic, WPC typically shows low strength. However, adding inorganic substances improves the surface activity of wood flour and plastic in WPCs and results in strength improvement (Park and Kim 2011). The RM7.5 specimen showed the highest strength (2.69 MPa). RM5, RM 10, and the WPC control showed strengths of 2.49 MPa, 2.48 MPa, and 1.29 MPa, respectively. The WPC control showed low bending strength because these specimens were manufactured for the pilot production. NTRM specimens also demonstrated improved strength. NTRM8, NTRM16, and NTRM24 showed strengths of 1.26 MPa, 1.39 MPa, and 1.49 MPa, respectively. Between RM and NTRM groups, the RM group showed higher strength than the NTRM group; this is because NTRM specimens had more wood flour content than the RM specimens, which causes the strength to decrease (Lee and Byon 2013).

Biodegradation Test

As shown in Figs. 6 to 8, in the 20-day biodegradation test, the reference material of the TLC-grade cellulose showed 41% biodegradation, and RM7.5 showed the highest biodegradation (31%) among the test specimens. RM5 and RM10 showed 30% and 25% biodegradation, respectively, in the same duration.

In the NTRM group (rice-bran mixture added instead of the talc component), NTRM16 showed the highest biodegradation (28%), and NTRM8 and NTRM24 showed 25% and 15% biodegradation, respectively. The control WPC showed only 18% biodegradation. Hence, adding rice-bran mixture instead of the WPC component improved biodegradation in WPCs.

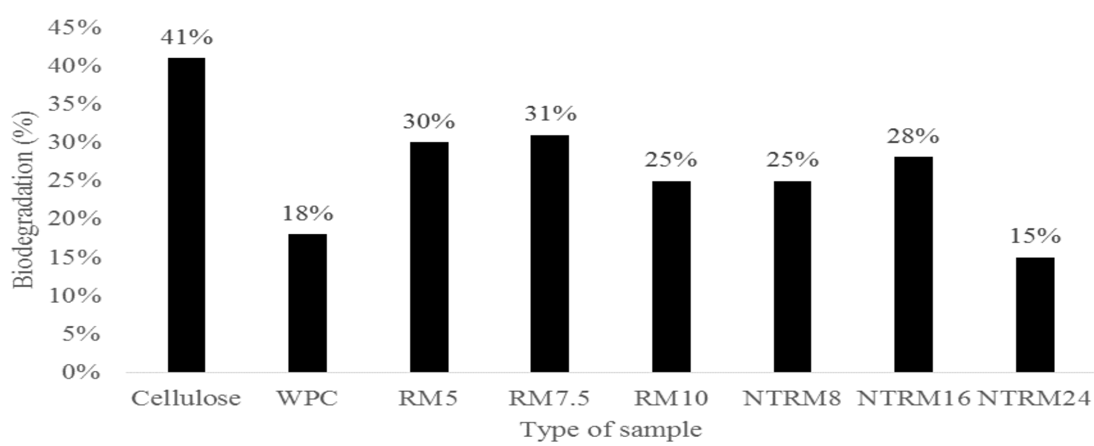

Fig. 6. Biodegradation results

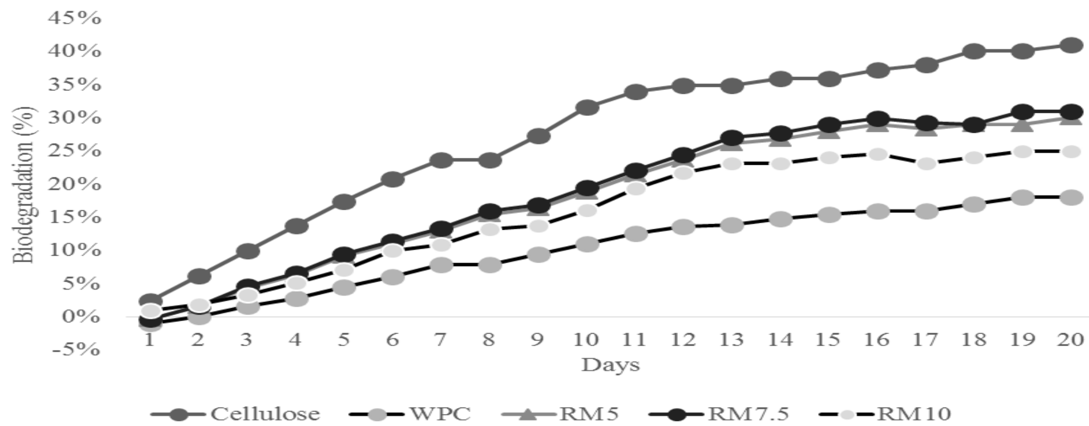

Fig. 7. Biodegradation results of RM group

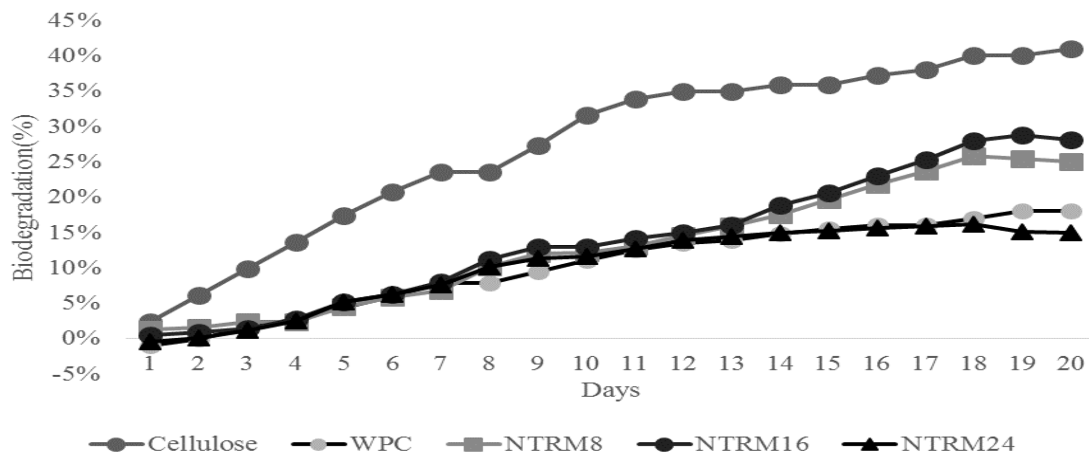

Fig. 8. Biodegradation results of NTRM group

Surface Morphology

Figures 9 and 10 show the surface morphologies of each specimen on a weekly basis. Before the biodegradation experiment, SEM was performed, and the result is shown in Fig. 9 and 10.

In the beginning of the experiment, both of specimens (control WPC and RM7.5) showed relatively smooth surfaces, and there were no holes on the surface. However, both specimens (control WPC and RM7.5) showed the growth of hyphae (in the red circle, it shows hyphae).

With the passage of time, hyphae continued to grow and adding rice-bran mixture accelerated growing hyphae in RM7.5. Especially, there were differences of hyphal growth in the 3rd week between specimens. RM7.5 showed faster growth and decay than the control WPC.

After the first week, both of the specimens showed biodegradation (holes, decay). In particular, RM7.5 showed more decay and holes than the control WPC. Based on a comparison between Figs. 9 and 10, both specimens were decaying and undergoing biodegradation; however, RM7.5 was decaying faster than the control WPC. This is confirms that the rice-bran mixture accelerates WPC biodegradation.

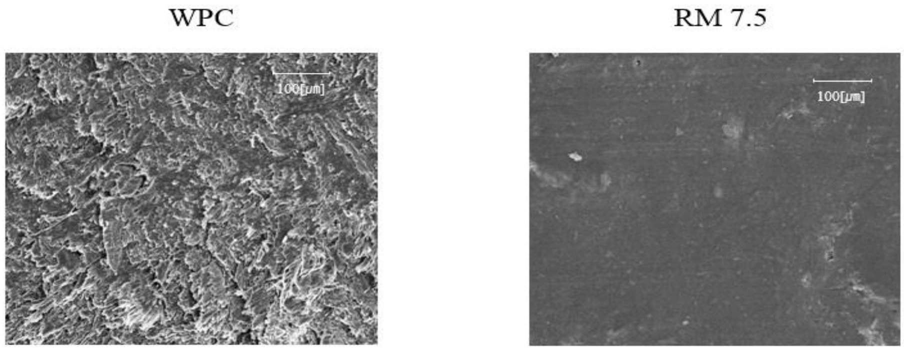

Fig. 9. SEM micrographs of the WPC surface (0 week)

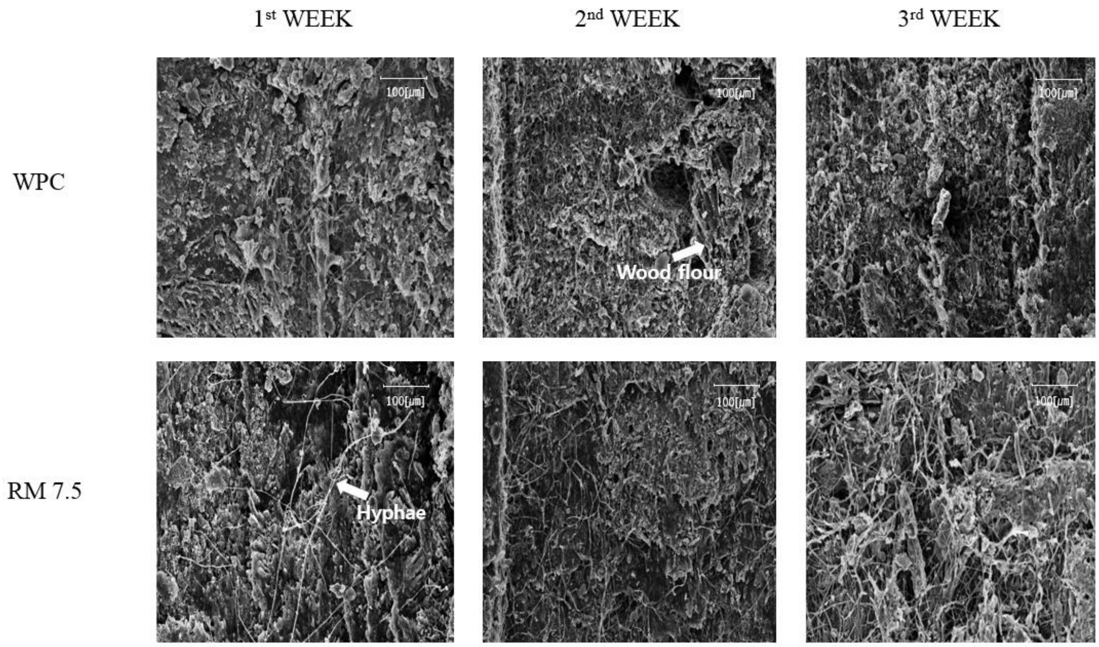

Fig. 10. SEM micrographs of the WPC surface (1-3week)

CONCLUSIONS

1. Specimens with 7.5% rice-bran-mixture (RM7.5) showed the highest biodegradation (32%), while in same duration, the default WPC showed 18% biodegradation. In another group (rice-bran mixture added instead of the talc component), 16% rice-bran-mixture-added specimen (NTRM16) showed 29% biodegradation. It follows that as rice-bran-mixture is added, biodegradation is improved. Rice-bran-mixture has a high biodegradation property (shown on Fig. 4).
2. As a result of adding rice-bran mixture, the bending strength of WPCs was improved up to 140%. The strength of the existing WPC was 1.29 MPa, while the 7.5% rice-bran-mixture-added specimen (RM7.5) showed the highest strength of 2.69 MPa. There was a significant increase in bending strength when rice-bran mixture was added instead of wood flour. The RM group showed higher strength than NTRM group. It is notable that the RM group had a lower wood flour content.

REFERENCES CITED

Ali Kazemi Tabrizi, Seyed Mohammad Javad Sepidedam, and Amir Nourbakhsh. (2014). "Investigation of waste fish (WF) effects on properties of biodegradable wood-plastic (HDPE) composites." *World of Sciences Journal* 2(1), pp. 44-53.

ASTM International. (2010). "Standard test methods for flexural properties of unreinforced and reinforced plastics and electrical insulating materials," ASTM D790:10. ASTM International, West Conshohocken, Pennsylvania, US.

Bergman, R. D., Han, H. S., Oneil, E., and Eastin, I. L. (2013). "Life-cycle assessment of redwood decking in the United States with a comparison to three other decking materials," *CORRIM Final Report*, University of Washington, Seattle, WA. DOI:10.1061/9780784412626.042

Chow, P., Nakayama, F. S., Youngquist, J. A., Muehl, J. H., and Krzysik, A. N. (2002). "Durability of wood/plastic composites made from *Parthenium* species," *IRG/WP/02-40225*, The International Research Group on Wood Preservation, Stockholm, Sweden.

Endres, H. J., and Siebert-Raths, A. (2009). "Rahmenbedingungenm Marktsituation, Herstellung, Aufbau und Eigenschaften," in: *Techische Biopolymere*, Carl Hanser Verlag, Munich, Germany. DOI:10.3139/9783446421042

International Organization for Standardization. (2012). "Determination of the ultimate aerobic biodegradability of plastic materials under controlled composting conditions," *ISO 14855-1:2012*. Intl. Organization for Standardization, Geneva, Switzerland.

Klyosov, A. A. (2007). *Wood-Plastic Composites*, John Wiley & Sons, 2007.

Lee, K. H., and Byon, S. K. (2013). "A study on the phase separation and mechanical properties of wood flour-polypropylene composites," in: *Elastomers and Composites* 48(3), Rubber Society, Seoul, Korea, pp. 216-220. DOI:10.7473/ec.2013.48.3.216

McDonald, A. G., Fabiyi, J. S., Morrell, J., and Freitag, C. (2009). "Effect of wood species on the weathering and soil performance of wood plastic composites," *Proceedings of the 10th International Conference on Wood and Biofiber-Plastic Composites*, Madison, WI, pp. 84-89. DOI:10.1016/j.compositesa.2010.06.004

Morrell, J. J., Stark, N. M., Pendleton, D. E., and McDonald, A. G. (2010). "Durability of wood-plastic composites," *Tenth Intl. Conf. Wood & Biofiber Plastic Composites and Cellulose Nanocomposites Symposium*, Forest Products Society, Madison, WI.

Park, B. S., and Kim, D. S. (2011). "Effects of coupling agents and clay on the physical properties of wood flour/polyethylene composites," in: *Polymer* 35(2), The Polymer Society, Seoul, Korea, pp. 124-129.

Silva Guzman, J. A. (2003). *Development of an Accelerated Method for Assessing Decay of Wood Plastic Composites (WPCs)*, Ph.D. diss., Oregon State Univ., Corvallis, OR.

Verhey, S. A., Laks, P. E., and Richter, D. L. (2002). "The effect of composition on the decay resistance of model woodfiber-thermoplastic composites," *The Sixth Intl. Conf. on Wood fiber-plastic Composites*, Forest Products Society, Madison, WI, pp. 79-86.

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