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ORIGINAL ARTICLE

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Surface deterioration of wood-flour polypropylene composites by weathering trials

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Abstract The market for wood-fiber plastic composites (WPCs) is expanding rapidly in many countries including Japan, where WPCs are mainly used for exterior products. In such applications, WPCs undergo undesirable color change, chalking, and shrinkage and swelling, and accordingly there is a need to better understand the mechanisms responsible for the weathering of WPC and develop methods of improving their weathering resistance. In this study, weatherability of WPC was assessed by natural and accelerated weathering trials. Discoloration (whitening) of WPC during exposure was caused by degradation of both wood and plastic. Darker color pigments as additives improved the color stability of WPC; however, chalking on the surfaces still occurred. The color stability of WPC was improved by application of exterior coatings. Preweathering of WPC (before coatings were applied) increased the absorption of coatings by the WPC and had a positive effect on the color stability and prevented chalking of the composites.

Key words WPC · Weathering · Discoloration · Chalking · Finishing

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Introduction

Wood-fiber polymer composites (WPCs) have been used for manufacture of interior parts such as flooring, handrails, and decorative components in Japan.[1] WPCs offer good water resistance, hardness, smooth surface, easy maintenance, easy processing, and so on. The market for exterior products made from WPCs such as decks and garden furniture is expanding rapidly in Japan.[2] WPCs are considered to be environmentally friendly, because they can be produced from waste wood and waste plastics, and are recyclable. Many house builders, wood companies, and plastics companies are interested in WPCs. However, the weathering resistance of WPCs is generally poor, and discoloration, chalking, and dimensional change caused by outdoor exposure are major problems for their use outdoors.[3] The discoloration problem seems to be caused by photodegradation of wood components in WPCs.[3,4] Lignin in wood strongly absorbs ultraviolet (UV) light and this leads to radical-induced depolymerization of both lignin and cellulose at wood surfaces.[5,6] Photodegraded components are washed away by rain and the wood surfaces become lighter in color. Therefore, to prevent discoloration of WPCs used outdoors, it is important to improve the photostability of wood fiber. To date, the most effective method of preventing the photodegradation of wood involves treatment with aqueous solutions of inorganic salts, particularly hexavalent chromium compounds,[7] but health concerns about the use of hexavalent chromium have discouraged commercial development of this concept. Some alternative methods of stabilizing wood when it is subjected to exterior exposure or aggressive environments are addition of photostabilizers[8] or pigments,[9] finishing,[10] and chemical modification of wood.[11–13]

The aim of this study was to clarify the mechanisms responsible for the weathering of WPCs and to propose methods of preventing discoloration and chalking of WPC during exposure.

Materials and methods

Natural weathering of WPCs

WPCs used in this study were made from wood flour [WF; spruce (*Picea* spp.) passing 50–100 mesh] and polypropylene (PP). The ratio of WF to PP was 55:45 (w/w) and several percent of compatibilizer (maleated PP) and about 10% of pigments were added. These test specimens used in this study were hollow in structure and were prepared by EIN Engineering with an extrusion method. The dimensions of specimens tested were 120 (longitudinal) × 100 (width) × 20 mm (thickness). Five different colored products (brown, yellow, black, natural, and no pigment) were prepared, and then exposed outdoors in a horizontal configuration for 24 months in Gifu Prefecture near Nagoya, Japan, for 24 months from December 2002 to December 2004.

Accelerated weathering of WPCs

The weatherabilities of three kinds of WPCs (brown, yellow, and no pigment) produced by the extrusion process were assessed in an accelerated weathering chamber (Atlas Ci4000). The mixing ratio of WF to PP was 55:45 (w/w) and the pigment content was about 10% of the total weight of WF and PP. The dimensions of the specimens were 60 × 40 × 4 mm. Furthermore, extruded WPCs with different ratios of WF and PP (20:80, 30:70, 40:60 without pigment) were also prepared. Specimens were placed in an accelerated weathering chamber and tested for up to 2000 h. The accelerated weathering regime involved continuous exposure to xenon arc UV light (0.5 W/m^2 at 340 nm = 550 W/m^2 for 290–800 nm), and 18 min of deionized water spray every 2 h with a black panel temperature of 65° ± 2°C and chamber temperature of 40° ± 2°C.

Finishing of WPCs

Brown color semi-film-forming coating (Sikkens Cetol HLS, Toyo Materia) and penetrating coating (Xyladecor, Japan EnviroChemicals) were applied to weathered WPC specimens. Unpigmented WPCs with the WF to PP ratio of 55:45 were weathered in an accelerated weathering chamber for 0, 500, and 1500 h, and then the specimens were finished with two types of exterior coatings by two coats using a brush. Finished WPCs were put in the accelerated weathering chamber for up to 2000 h and weatherability was assessed again as above.

Surface analysis

A colorimeter (Nihon Denshoku Kogyo, NF333) was used to quantify color changes at the surfaces of WPCs as a result of weathering. Color was expressed using the CIE Lab (L^*, a^*, b^*) system. Color changes due to weathering, ΔE^* and brightness (L^*) were calculated from L^*, a^*, b^* values. Fourier transform infrared (FT-IR) spectroscopy (Jasco, FTIR-470 Plus) by KBr tablet method was used to assess chalking product on the surface of WPCs during weathering.

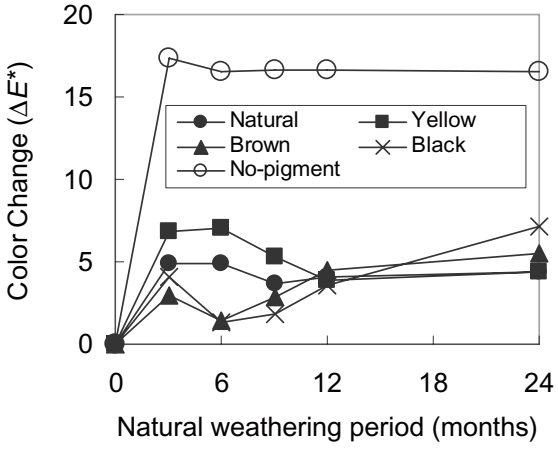

Fig. 1. Color change of pigmented wood-fiber composites (WPCs) exposed to natural weathering for 24 months

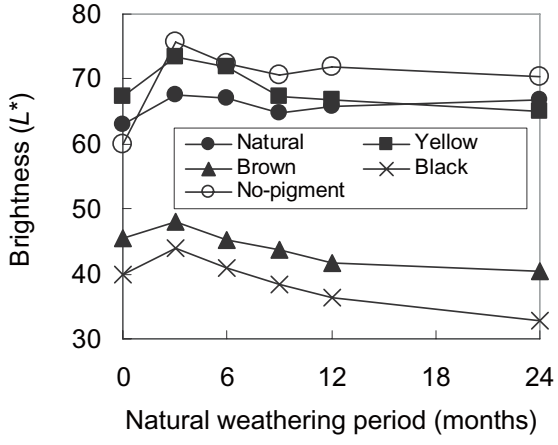

Fig. 2. Change of brightness of pigmented WPCs exposed to natural weathering for 24 months

Results and discussion

Natural weathering of WPCs

Figures 1 and 2 show changes in brightness and color of WPCs during exterior exposure. The more darkly pigmented WPCs showed less color change. The WPC that did not contain pigments showed large color changes and after 2 years of weathering was a greyish color like unfinished solid wood. Brightness of pigmented WPCs was less than that of the unpigmented WPC. Powders caused by chalking on the surfaces were observed in all specimens.

Accelerated weathering of WPCs

Figures 3 and 4 show changes in color and brightness of pigmented and unpigmented WPCs during accelerated

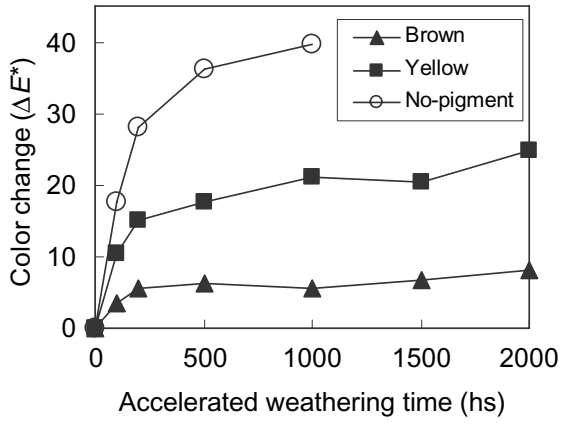

Fig. 3. Color changes of pigmented WPCs exposed to accelerated weathering for 2000 h

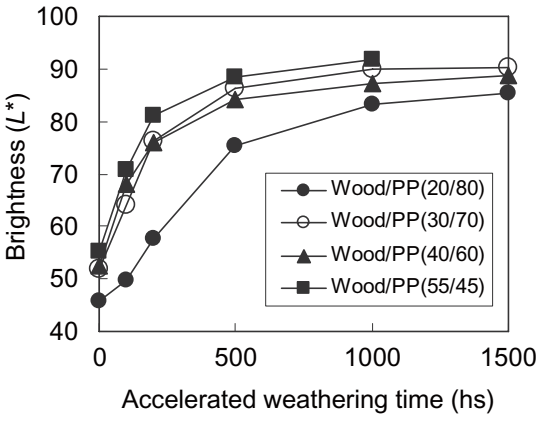

Fig. 5. Brightness of WPCs with different ratios of wood and polypropylene (PP) during 1500 h of accelerated weathering

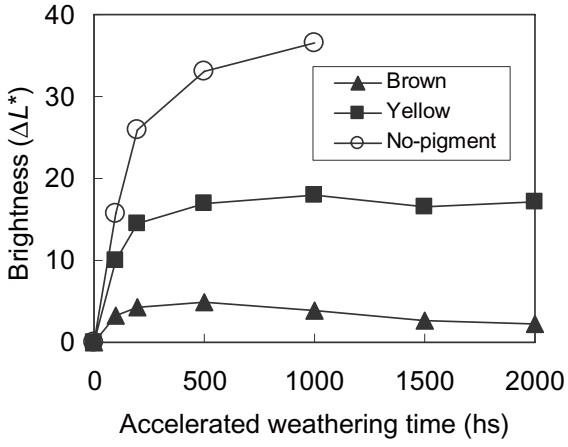

Fig. 4. Brightness change of pigmented WPCs exposed to accelerated weathering for 2000 h

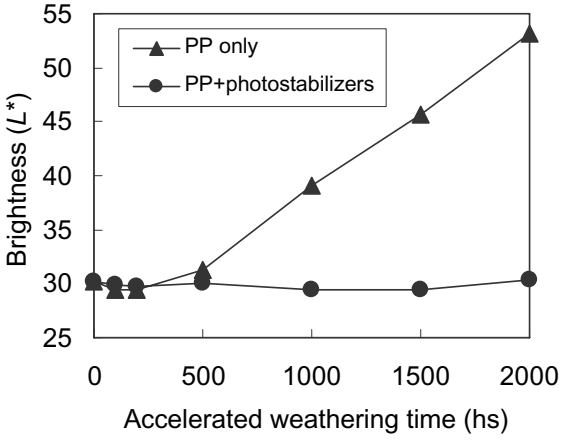

Fig. 6. Brightness of PP with and without photostabilizers during 2000 h of accelerated weathering

weathering. The surface color of the WPC containing no pigment turned white during accelerated weathering. Pigmented WPCs also showed discoloration during exposure mainly resulting from increases in brightness (see Fig. 4). It seems that lignin and hemicelluloses in wood fiber, which were degraded by UV, are leached from exposed surfaces. Subsequently, the main chemical component at the WPC surface is possibly cellulose, which is whitish and comparatively stable against UV. Hence the surface of weathered WPCs was white. Stark et al.[3] reported that lightening of wood–plastic composites originated mainly from photoinduced bleaching of wood fiber.

Figure 5 shows lightening of WPCs with different ratios of wood and PP caused by accelerated weathering. Brightness increased during weathering and was positively correlated with wood ratio; however, the difference in brightness of WPCs with wood ratios of 30%, 40%, and 55% was small. Brightness of PP with and without photostabilizers (UV absorbers and hindered amine light stabilizers) during accelerated weathering is shown in Fig. 6. The brightness of PP that did not contain photostabilizers increased greatly after 500 h of weathering. On the other hand, PP that contained photostabilizers did not show any change in brightness during weathering up to 2000 h. In general, adding photostabilizers to PP can improve the photostability dramatically.[14] However, the PP that did not contain photostabilizers showed no color change up to 500 h of weathering and this tendency differed from that observed for WPCs.

Surfaces of weathered WPCs (WF:PP 55:45) were observed by video microscope (Keyence, VH-7000C) (see Fig. 7). At first, wood flour showed lightening during weathering and then the color of PP also turned white after only 200 h of weathering. It seemed that the PP and wood flour together form a heterogeneous mixture and the crystals are unstable, unlike homogeneous PP. It is possible that lignin in the composites generates radicals during photo irradiation that attack PP.[15] Therefore, the heterogeneous PP formed in WPCs showed bleaching even during short periods of exposure. The bleaching of WPCs is not only caused by photodegradation of wood flour but also by degradation of PP.

Figure 8 shows FT-IR spectra of powder on the surface of WPC after 1500 h of accelerated weathering, wood flour

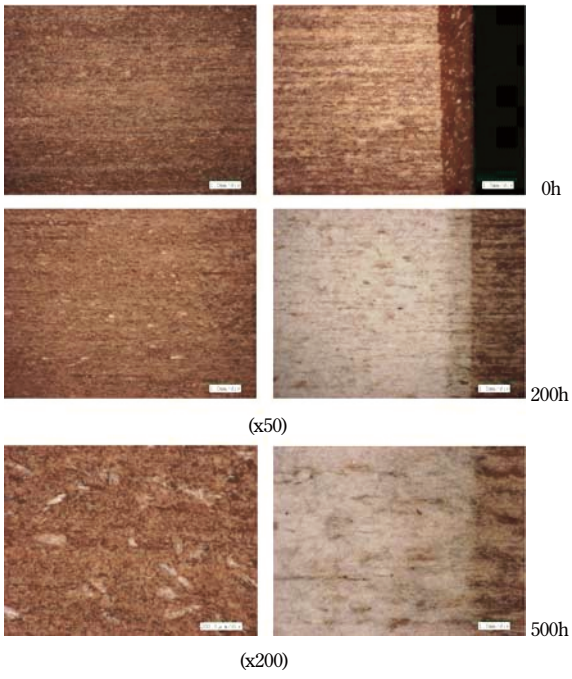

Fig. 7. Surface appearance of WPCs with pigment (*left*) and without pigment (*right*) after accelerated weathering for 0, 200, and 500 h

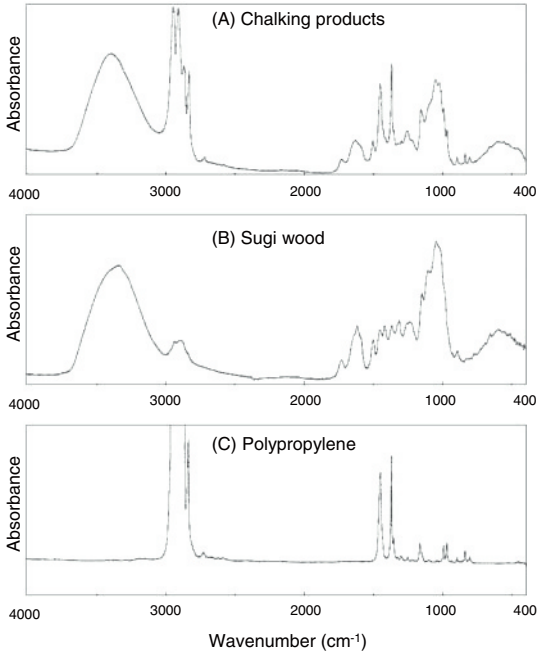

Fig. 8A–C. Fourier transform infrared (FT-IR) spectra of **A** chalking product generated on surface of WPCs by accelerated weathering for 1500 h, **B** wood-flour (unweathered sugi wood), and **C** polypropylene

(sugi, *Cryptomeria japonica* D. Don), and PP. The spectrum of the powder generated by weathering showed characteristics of wood flour and PP. It seemed that chalking during weathering was caused by decomposition of PP following photodegradation and removal of wood flour from exposed surfaces.

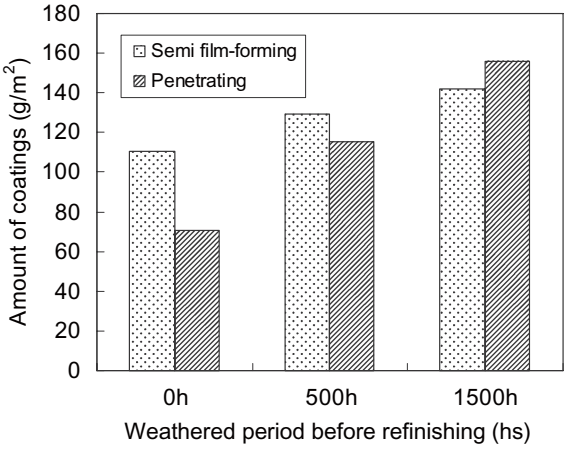

Fig. 9. Amount of penetrating and semi-film-forming coatings on WPCs subjected to accelerated weathering before coating

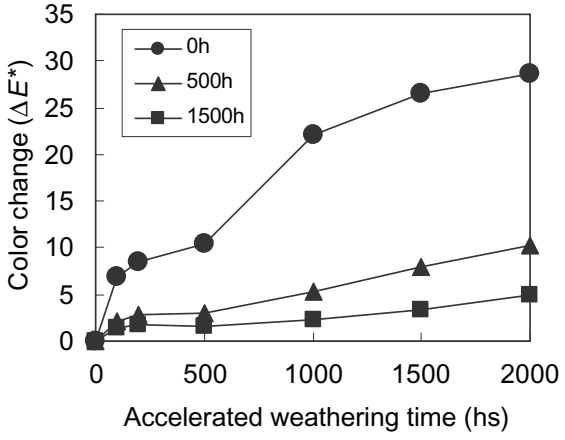

Fig. 10. Color change of preweathered WPCs coated with penetrating finish by accelerated weathering

Finishing of WPCs

In general, finishing of polyolefins such as polypropylene or polyethylene is difficult because of their low wettability. However, finishing with two different commercial pigmented exterior coatings was used to prevent discoloration and chalking of WPCs during weathering. WPCs weathered for different periods of time (preweathering) were finished with semi-film-forming and penetrating coatings. Figure 9 shows the amount of coatings absorbed by WPCs following preweathering. The absorption of both coatings was positively correlated with the time of preweathering. This effect was more pronounced for the penetrating coating, and the amount of coating on the surface preweathered for 1500 h was more than twice that of unweathered surfaces.

Figure 10 shows color changes of WPCs finished with penetrating coatings during accelerated weathering. No preweathered WPCs finished with the penetrating coating showed large color change during accelerated weathering because of the removal of pigments. This was in contrast to the WPCs that were subjected to pre-weathering before

being finished with the penetrating coating; these preweathered specimens showed small color change during accelerated weathering. Because preweathered surfaces absorbed large amounts of the penetrating coatings, the weatherability of WPCs finished with coatings depends on the amount of coatings on the surface. These results show that commercial exterior wood coatings can be used to maintain WPCs weathered for several years.

Conclusions

Discoloration of WPCs caused by weathering was improved by the addition of darker pigments to the composite. Whitening of WPCs (wood and PP components) in the absence of pigments during weathering seemed to be caused by photobleaching of wood fiber and decomposition of PP. Chalking on the surfaces of WPCs during weathering was restricted by finishing the composites with exterior wood coatings. Weatherability of finished WPCs depended on the amount of coatings on the surfaces. Preweathering of the WPC before finishing increased the absorption of coatings and hence improved the resistance of the composites to discoloration during weathering.

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