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Effect of inorganic pigments on the hygrothermal properties of wood-plastic composites

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The addition of iron oxide pigments to wood-plastic composites results in an increase in the flexural strength and moduli they exhibit after hygrothermal aging tests.

Due to the introduction of wood fibers, wood-plastic composites (WPCs) are hygroscopic in nature (i.e., they attract and absorb water). This absorption of moisture, which occurs in wet environments and/or under high humidity, leads to dimensional instability, microcracks, degradation, mass loss, susceptibility to bio-deterioration, and significant reduction in the mechanical stiffness and strength of WPCs.[1]

To enhance the market appeal of plastic products, the addition of pigments is a common practice.[2] In general, inorganic iron oxide pigments are characterized by their excellent durability, high inherent opacity, good UV-screening properties, low toxicity, and low cost. However, few studies have been conducted on the incorporation of inorganic iron oxide pigments into WPCs.[3–5]

The hygrothermal aging test is the most robust and reliable method for quickly evaluating the long-term service performance of moisture-affected fiber-reinforced polymer composites. The information obtained from this test can be used to predict the outdoor performance of WPCs that are exposed to rain and environmental moisture.

To test the effect of pigments on the hygrothermal aging properties of WPCs, we fabricated a control sample and two colored samples made of recycled high-density polyethylene (HDPE) highly filled with wood waste, calcium carbonate, and inorganic fillers at a ratio of 27/73.[6] Characteristics of these fillers and the WPC formulations that we employed can be seen in Tables 1 and 2. The WPC lumbers were fabricated in a commercial production line via compounding, followed by an extrusion process. These specimens were then immersed in distilled water at 45°C for 70 days. We subsequently investigated the hygrothermal aging properties of the samples.

We found the pigment-colored WPCs to better withstand the hygrothermal aging test. Compared to the control sample, the properties of these WPCs include lower water absorption, less total color change, and a higher mechanical-property-retention rate. Among our samples, the combination of iron oxide red, yellow, and black pigment exhibited the best aging performance in terms of dimensional stability, color stability, and the retention rate of mechanical properties.

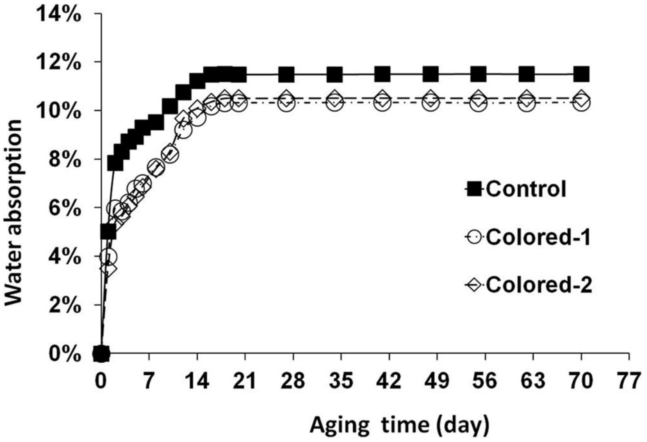

Figure 1. *Water absorption of wood-plastic composites (WPCs) as a function of aging time. Control: No pigment. Colored-1: Red iron oxide pigment. Colored-2: Red, yellow, and black iron oxide pigments (brown).*

The water-absorption curves of different samples are illustrated in Figure 1. Generally, the water absorption of the specimen increases with immersion time and reaches a maximum (equilibrium) value at a saturation point of approximately 20 days, beyond which no more water is absorbed and the water content of the composites remains constant. For the sample without pigments (original color, the control), the equilibrium for water absorption is 11.49%. For the pigment colored-1 (red) and colored-2 (brown) samples, the equilibria are 10.34 and 10.52%, respectively: see Table 3. In addition, the diffusion coefficients of pigment-colored WPCs are slightly lower compared to the control specimens.

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Table 1. *Characteristics of the inorganic fillers.*

Inorganic materials	Molecular formula	Molar mass g/mol	Density g/cm^3	Particle size nm	Particle shape	Suppliers
Iron Red 190	Fe_2O_3	159.69	5.24	700	Spheroidal	YiPin Pigments
Iron Black 722	Fe_3O_4	232.6	5.18	350	Acicular	YiPin Pigments
Calcium carbonate	$CaCO_3$	100.09	2.83	3000	Irregular	Hainan Kunlun

Table 2. *Formulation of the WPCs (wt%). HDPE: High-density polyethylene. MAPE CMG9804: Maleic-anhydride-grafted polyethylene CMG9804. PE-Wax BN-208: Polyethylene wax BN-208.*

WPCs	Wood	HDPE	$CaCO_3$	MAPE CMG9804	PE-Wax BN-208	Iron Red 190	Iron Yellow 313	Iron Black 722
Control	54	27	16.5	1.5	1	0	0	0
Colored-1	54	27	13	1.5	1	3.5	0	0
Colored-2	54	27	13	1.5	1	1.5	0.5	1.5

The hygrothermal aging treatment changed the color of all samples, but the patterns of lightness change (ΔL*) were very different between the control and the colored samples. Figure 2 shows ΔL* to increase for colored WPCs, indicating fading or lightening with aging time. The control, however, darkened with aging time. The lightening rate of the colored WPCs is slightly increased with time, whereas the darkening rate of the control sample was relatively fast over the first 42 days and became slower thereafter. Figure 3 shows the change in total color (ΔE*) of both control and colored WPCs to increase with aging time. The brown sample exhibited the lowest rate of increase for ΔE*, demonstrating that darker WPCs have better resistance to color change during hygrothermal aging.

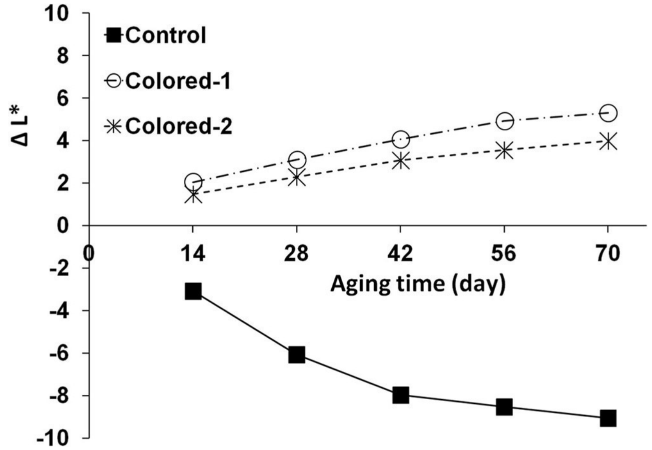

Figure 2. *Brightness change (ΔL*) of the WPCs as a function of aging time.*

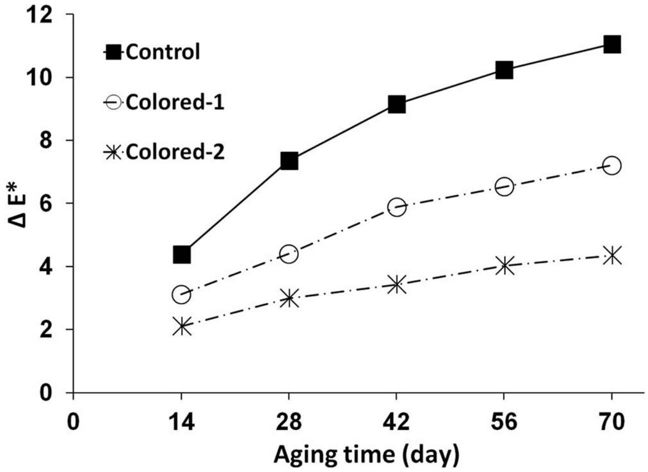

Figure 3. *The total color change (ΔE*) of WPCs as a function of aging time.*

We found the addition of pigments to result in comparable initial flexural properties compared to the control specimen: see Tables 4 and 5. All WPCs exhibit gradually decreasing flexural properties during the hygrothermal aging test, indicating that the degradation effect of interfacial bonding due to water and heat plays an important role in this

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Table 3. *Water absorption (M_m) and diffusion coefficient (D) of WPCs.*

WPCs	Control	Colored-1	Colored-2
D ($\times 10^{-14}$m^2/s)	3.83 (0.18)	3.76 (0.32)	3.46 (0.21)
M_m (%)	11.49 (0.46)	10.34 (0.31)	10.52 (0.15)

Table 4. *Effect of aging time on the flexural modulus of rupture of the WPCs.*

Aging time day	Control MPa		Colored-1 MPa		Colored-2 MPa	
	Ave	Stdev	Ave	Stdev	Ave	Stdev
0	29.7	2.73	28	1.96	28.9	1.61
14	27.8	1.83	26.8	2.41	27.9	2.05
28	25.9	2.11	24.7	1.66	26.1	1.35
42	23.2	0.96	21.9	1.63	23.5	0.54
56	22.1	0.58	21	1.42	22.6	2.91
70	20.2	1.19	20.5	1.37	22.1	2.22
Retention rate (%)	68.01		73.21		76.47	

Table 5. *Effect of aging time on the flexural modulus of elasticity of the WPCs.*

Aging time day	Control MPa		Colored-1 MPa		Colored-2 MPa	
	Ave	Stdev	Ave	Stdev	Ave	Stdev
0	5124	365	4685	89	4845	290
14	4365	200	3958	139	4195	205
28	3893	140	3620	93	3836	138
42	3560	74	3372	297	3576	319
56	3161	147	2990	267	2995	173
70	2605	289	2573	98	2713	255
Retention rate (%)	50.84		54.92		56.00	

phenomenon. It is also interesting to note that the deterioration of the flexural modulus (modulus of elasticity, MOE) caused by hygrothermal aging is more severe than that of flexural strength (modulus of rupture, MOR). The retention rate of MOR was in the range of 68–76.5%, whereas the MOE was only 50–56% for all WPC specimens after 70 days of aging.

In summary, we found WPCs colored with iron-oxide pigments to exhibit better performance after hygrothermal aging than an uncolored sample. In addition, the water-absorption equilibrium and total color change of the colored WPCs were lower. In the next stage of our research, we intend to fabricate WPCs from wood-flour residues and recycled HDPE with nanocellulose derived from banana- or coconut-palm-petiole fibers as reinforcement. We will then investigate the dose and surface morphology of the nanocellulose; the content and species of inorganic pigments; and the effect of the resin/filler ratio on the mechanical and hygrothermal aging properties of the obtained WPCs.

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