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EFFECT OF NATURAL AND ACCELERATED AGING ON THE PROPERTIES OF WOOD-PLASTIC COMPOSITES

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ABSTRACT

The correspondence of natural and laboratory-accelerated aging of WPC has long been a highly important problem discussed by many scholars. In this work, the changes in moisture content (MC), modulus of rupture (MOR), modulus of elasticity (MOE), screw holding force and creep recovery rates of two groups of wood-plastic composites (WPC) after natural and accelerated aging (high-low temperature cycles and freeze-thaw cycles) were studied to provide guidance for the use of WPC in outdoor applications. The results showed that, after the natural aging and freeze-thaw cycles treatments, MC increased significantly with both 167% of the untreated value of wood-HDPE composites with 30% wood fiber content and a thickness of 25 mm (W_{25}), while 67% and 133% of the wood-HDPE composites with 30% wood fiber content and a thickness of 20 mm (W_{20}), but is almost unchanged after the treatment with high-low temperature cycles. The mechanical strength, including MOR, MOE, screw holding force and creep recovery rate, decreased after natural and accelerated aging. The greatest decreases of MOR, MOE, screw holding force and creep recovery rate were 14%, 13%, 21%, and 7% for W_{25}, while 5%, 8%, 8%, and 14% for W_{20} respectively. Environmental aging can reduce the strength of WPC, but the bending strength retention rate is more than 85%, showing that performance of WPC is relatively stable compared to wood materials, which is one of the reasons for the widely use of WPC in outdoor applications.

KEYWORDS: Natural aging, high-low temperature cycles, freeze-thaw cycles, creep performance.

INTRODUCTION

Wood-plastic composites (WPC) are composites that contain wood fiber and thermosets or thermoplastics. WPCs are environmentally-friendly materials because they reduces the deforestation and the negative impact of large-scale use of plastics on the environment. Wood-thermoset composites were first developed in the early 1900s. An early commercial composite composed of phenol-formaldehyde and wood fiber was reported in 1916 and was used as a gearshift knob for the Rolls Royce automobile (Clemons 2002). Because thermosets are plastics that cannot be melted by reheating once cured, wood-plastic composites are generally materials consisting of wood fiber mixed with thermoplastic and additives in a certain proportion. The WPC industry has experienced tremendous growth in recent years because WPC combine the desirable durability of plastics with the cost-effectiveness of wood fibers as a filler or reinforcing agent (Mengeloglu et al. 2000, Clemons 2002, Pilarski and Matuana 2005, Zini et al. 2011). The addition of wood to unfilled plastic can greatly stiffen the plastic but often makes it more brittle (Clemons 2002). WPC can be used for floors, decking, fencing, landscape timbers, plank roads, leisure chairs, and decorative boards (Pilarski and Matuana 2005, Badji et al. 2017).

The influence of wood species, wood flour content and other additives such as tinder fungus on the physical and mechanical properties of WPC has long been studied in the previous research (Stark et al. 2004, Kaymakci et al. 2016, Chen et al. 2017, Xu et al. 2017). Although the durability of WPC is better than that of the solid wood (Albrektas et al. 2020), researches on the durability are very important because of the wildly use of WPC in exterior applications. Pilarski and Matuana (2005, 2006) examined the effects of accelerated freeze-thaw actions on the durability of wood fiber-plastic composites and showed that the stiffness of the composites decreased significantly after only two freeze-thaw cycles, regardless of both the wood species and content.

Temperature and exposure time have important effects on the mechanical properties of WPC (La Mantia and Morreale 2008). Wang and Morrel (2005) studied the effects of moisture and temperature cycling on the properties of WPC and showed that moisture sorption tends to increase with the number of wet/dry cycles and is associated with a significant reduction in MOR and MOE.

The correspondence of natural and laboratory-accelerated aging of WPC has always been a highly important problem investigated by many scholars in order to enable accurate prediction of the long-term properties of WPCs. However, previous studies did not include investigations of the difference between wet and dry state with various treatments. In this work, the properties such as moisture content (MC), modulus of rupture (MOR), modulus of elasticity (MOE), screw holding force and creep performance of WPC after the natural aging, high-low temperature cycles and freeze-thaw cycles are studied in order to providing guidance for the use of WPC in outdoor applications.

MATERIALS AND METHODS

Materials

Two groups of WPC samples containing poplar flour, plastics and processing aids in the proportions of 30:65:5, were used for the experiments. The samples thicknesses were 25 mm and 20 mm, respectively.

40 mesh flours and high density polyethylene (HDPE) were mixed in a co-rotating twin-screw extruder. The densities of the two specimens were 1.15 and 1.10 $g.cm^{-3}$, respectively. Tab. 1 shows the designations of the specimens.

Treatments for natural aging

The specimens are exposed in an outdoor natural site for 1 year on a galvanized steel rack in Beijing, China. The exposure conditions included four annual seasons (January 10th, 2019 to January 10th, 2020) allowing us to consider the contribution of each season and examine the long-term degradation effects. In 2019, the average temperature in Beijing was 12.5°C, while the annual precipitation was 511.1 mm.

Treatments for high-low temperature cycles

According to the GB/T 24508: 2009, the specimens were placed at room temperature 23 ± 2°C for 1 h, then were placed in a low-temperature test chamber at -20 ± 2°C for 6 h, and then were placed in a dry oven at 60 ± 2°C for 16 h after storing at room temperature 23 ± 2°C for 1 h. This was designated as a single treatment cycle. In this work, the specimens were treated by 3 high-low temperature cycles.

Treatments for freeze-thaw cycles

Freeze-thaw cycles were carried out according to GB/T 24508: 2009. One complete freeze-thaw cycle consisted of three stages: *(1)* water soaking; *(2)* freezing for 24 h; *(3)* thawing for 24 h. The specimens were immersed in the water (20°C) for 24 h, then placed in a low-temperature test chamber at a temperature of -30 ± 2°C for 24 h, and then were removed and placed at room temperature 23 ± 2°C for 24 h. This was designated as single treatment cycle. In this work, the specimens were treated by 3 freeze-thaw cycles. Tab. 1 shows the treatment conditions and testing parameters of the materials.

Tab. 1: Designations, treatment conditions and testing parameters of materials.

Des.	Thickness (mm)	Density ($g·cm^{-3}$)	HDPE (wt%)	Wood flour (wt%)	Treatment conditions	Testing parameters
W_{25}	25	1.15	65	30	Untreated	M_0, MOR_0, MOE_0, S_0, C_0
					natural aging	M_1, MOR_1, MOE_1, S_1, C_1
					high-low temperature cycles (for 3)	M_2, MOR_2, MOE_2, S_2, C_2
					freeze-thaw cycles (for 3)	M_3, MOR_3, MOE_3, S_3, C_3
W_{20}	20	1.10	65	30	Untreated	M_0, MOR_0, MOE_0, S_0, C_0
					natural aging	M_1, MOR_1, MOE_1, S_1, C_1
					high-low temperature cycles (for 3)	M_2, MOR_2, MOE_2, S_2, C_2
					freeze-thaw cycles (for 3)	M_3, MOR_3, MOE_3, S_3, C_3
One complete high-low temperature cycle: 23°C for 1 h, -20°C for 6 h, 23°C for 1 h, 60°C for 16 h; One complete freeze-thaw cycle: 20°C (in water) for 24 h, -3°C for 24 h, 2°C (room temperature) for 16 h.						

Measurement of MC

MCs for two groups of untreated specimens (M_0), specimens after natural aging (M_1), specimens after 3 high-low temperature cycles (M_2), and specimens after 3 freeze-thaw cycles (M_3) were examined according to ISO 16979: 2003. MC values were calculated using Eq. 1:

$$MC = \frac{M_n - m}{m} \times 100\%,$$ (1)

where: M_n - the initial mass of the specimen with different treatments at 23 ± 2°C and a humidity of 50% for 24 h (kg), m - the mass after drying at 103°C for 24 h (kg).

Measurement of MOR and MOE

Mechanical characterizations were carried out at 20 ± 2°C and 65 ± 5% RH. The MOR and MOE values for the untreated specimens (MOR_0 and MOE_0), specimens after natural aging (MOR_1 and MOE_1), specimens after 3 high-low temperature cycles (MOR_2 and MOE_2), and specimens after 3 freeze-thaw cycles (MOR_3 and MOE_3) were obtained according to ISO 16978: 2003.

A Universal Mechanical Testing Machine (SHIMADZU AG-IS 50KN, Japan) with the maximum load of 50 kN was used for the measurements. The bending load was applied at a rate of 10 $mm \cdot min^{-1}$ until the failure of the specimen. MOR and MOE were calculated using Eqs. 2 and 3:

$$MOR = \frac{3F_{max}L}{2bh^2}$$ (2)

$$MOE = \frac{FL^3}{4bh^3 s}$$ (3)

where: F_{max} - the maximum load (N), L - the length of span (mm), b - the width of the specimen (mm), h - the thickness of the specimen (mm), F - the load increment on the straight-line portion of the load-deformation curve (N), s - the deformation corresponding to F (mm).

Measurement of screw holding force

The screw holding force for the untreated specimens (S_0), specimens after natural aging (S_1), specimens after 3 high-low temperature cycles (S_2), and specimens after 3 freeze-thaw cycles (S_3) were examined according to ISO 27528: 2009.

Measurement of creep performance

The creep recovery rate for the untreated specimens (C_0), specimens after natural aging (C_1), specimens after 3 high-low temperature cycles (C_2), and specimens after 3 freeze-thaw cycles (C_3) were examined according to GB/T 29418: 2012. These values were obtained using Eq. 4:

$$C = \frac{d_1 - d_2}{d_1 - d_0} \times 100\%$$ (4)

where: d_0 - deformation before loading (mm), d_1 - deformation after loading for 24 h (mm), d_2 - deformation after taking off loading for 24 h (mm).

Data analysis

The SPSS statistical software, (SPSS Inc, Chicago, IL, USA), was used for data analysis. Duncan multiple comparison tests were carried out to analyze whether the effect of natural aging or artificial aging was significant.

RESULTS AND DISCUSSION

Effects of aging on MC

Fig. 1 shows the MC results for the two groups of WPC specimens including untreated, treated by natural aging, high-low temperature cycles and freeze-thaw cycles. MC values increased significantly after the natural aging and freeze-thaw cycles. Compared to that of the untreated sample, the MC of W_{25} and W_{20} increased by 167% and 67% after the natural aging, and by 167% and 133% after the freeze-thaw cycles, respectively. However, it is almost unchanged after the treatment with the high-low temperature cycles.

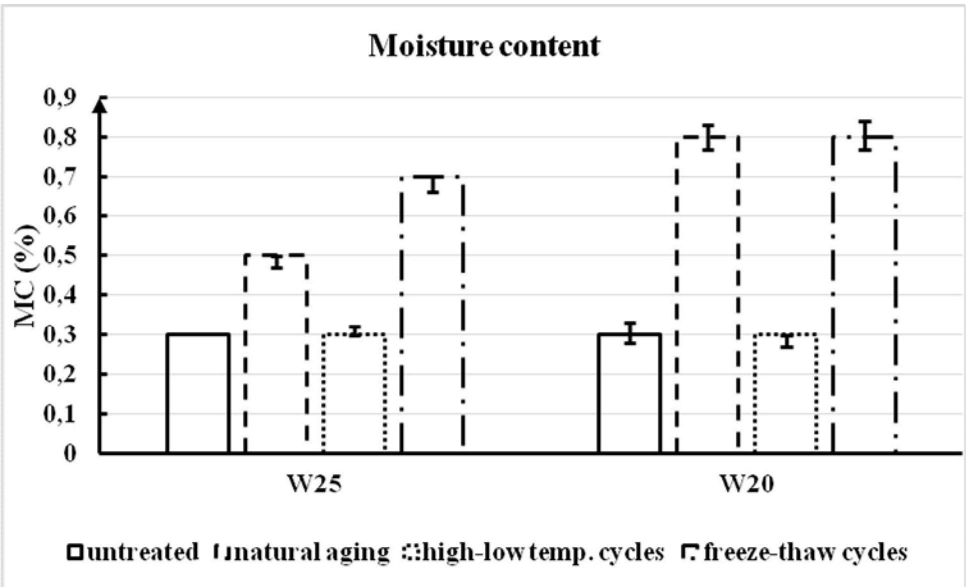

Fig. 1: MC of W_{25} and W_{20} specimens for untreated one, after treatments of natural aging, high-low temperature cycles and freeze-thaw cycles.

The weight of WPC is mainly related to wood fiber or powder and is not affected by PE, PP and PVC (Pilarski and Matuana 2005, Xiao 2010). In the process of natural aging, WPC experienced the effect of rain and snow. Due to the effect of hydroxyl (-OH) in wood fiber, WPC will be accompanied by the breaking and recombination of the hydrogen bond in the process of natural aging (Xiao 2010, Zhao et al. 2015a), resulting in the mass increase after aging that leads to the increase in the MC value. The weight of WPC increases after freeze-thaw cycles due to the existence of water (Pilarski and Matuana 2005). However, when it undergoes high-low temperature cycles, the material does not involve the condensation and melting of water, and therefore the MC value was not changed after the treatment by high-low temperature cycles.

Effects of aging on MOR and MOE

Fig. 2 and 3 shows the MOR and MOE of two groups of WPC specimens for the untreated case, and specimens after treatments of natural aging, high-low temperature cycles and freeze-thaw cycles. The MOR and MOE values of the two groups of WPC decreased after the treatment. This is consistent with the results of previous studies (Badji et al. 2017). Relative to that of the untreated specimen, the MOR values of W_{25} after the treatments of natural aging, high-low temperature cycles and freeze-thaw cycles decreased by 14%, 8%, and 4%, respectively. The corresponding MOR values of W_{20} decreased by 3%, 5%, and 0%, respectively, relative to the MOR of the untreated specimen. The MOE of WPC showed similar changes after the treatment. The MOE of W_{25} decreased by 13%, 8%, and 5%, while it decreased by 8%, 6%, and 3% for W_{20}. These results suggest that these artificial weathering conditions degrade lead to a weaker degradation of the mechanical properties than natural aging, in agreement with the results of Badji et al. (2017).

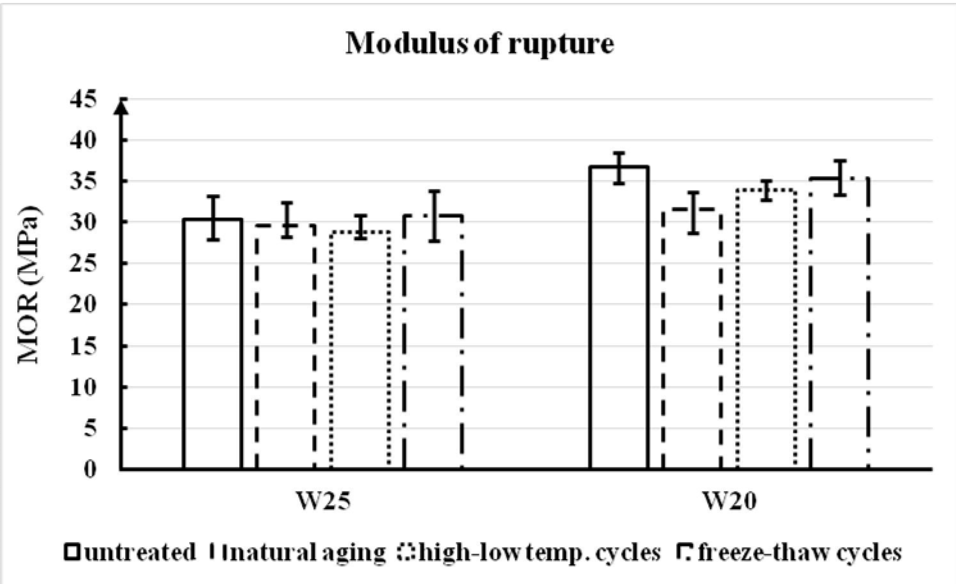

Fig. 2: MOR for untreated specimens, and specimens after treatments of natural aging, high-low temperature cycles and freeze-thaw cycles.

When the material undergoes high-low temperature cycles, the plastic produces larger thermal deformation due to the different expansion of wood fiber and plastic (Chatterji 1999a, Clemons 2002), resulting in the decrease of the adhesion strength between the fiber and plastic. This leads to the lower MOR and MOE values of WPC after the high-low temperature cycles.

Both MOR and MOE also decreased when the material underwent the freeze-thaw cycles. Pilarski and Matuana (2005) showed that the observed property loss was primarily due to the effect of water during the cycling that appears to have led to the decreased interfacial adhesion between the wood flour and the rigid PVC matrix. Freeze-thaw actions had no apparent effect on the density of the composites after exposure. However, these changes led to a moisture uptake that decreased the interfacial adhesion and increased the amount and size of the pores in the composites, resulting in a significant degradation of the flexural properties (Pilarski and

Matuana 2005). However, this mechanism is different from that of the degradation due to the high-low temperature cycles. In the process of freeze-thaw cycles, wood fiber soaks up moisture to expand its volume. At the same time, water molecules destroyed the interface between the wood fiber and the matrix, weakening the original effect of the wood fiber and resulting in the decrease of MOR and MOE (Xiao 2010). The effects on the mechanical strength of WPC after two treatments (high-low temperature cycles in dry state and freeze-thaw cycles in wet state) are different because of their different mechanisms. The water in the cell cavity and micropores of the cell wall freezes at low temperature. The condensation of water at low temperature is a process of breaking the original ice layer followed by the formation of a new ice layer until all of the water freezes (Chatterji 1999a,b). As a kind of porous material, the high bonding strength between ice and wood surface makes the mechanical properties of WPC tend to increase because the hydroxyl group on the molecular chain of wood is a hydrophilic group (Ayrilmis 2007, Zhao et al. 2015a,b). Therefore, the strength of WPC after freeze-thaw cycles is higher than that of WPC after the high-low temperature cycles.

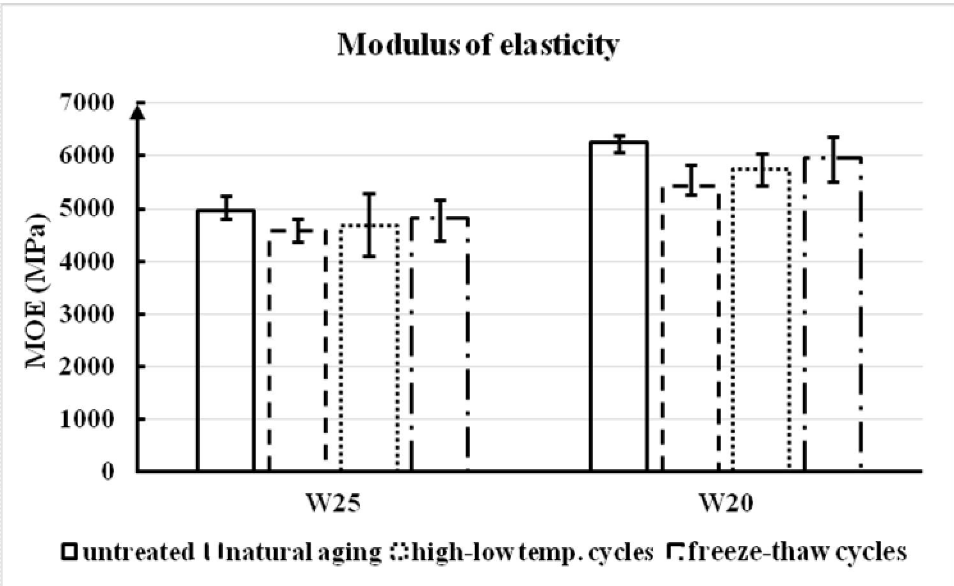

Fig. 3: MOE for untreated specimens, and specimens after treatments of natural aging, high-low temperature cycles and freeze-thaw cycles.

On the other hand, the retention values of MOR for W_{25} after the natural aging, high-low temperature cycles and freeze-thaw cycles were 86%, 92%, and 96%, while they were 97%, 95%, and 100% for W_{20}, demonstrating that the performance of WPC was relatively stable despite the decrease of MOR after the treatments. Therefore, WPC is widely used in outdoor applications.

Effects of aging on screw holding force

Fig. 4 shows the screw holding force of the two untreated WPC specimens, and the specimens after treatments by natural aging, high-low temperature cycles and freeze-thaw cycles. The screw holding force decreased after each of the three treatments. Compared to the untreated specimen, the screw holding force of W_{25} after the natural aging, high-low temperature cycles and freeze-thaw cycles decreased by 11%, 21%, and 8%, while the corresponding decreases were 6%, 8%, and 2% for W_{20}, respectively.

Due to the different shrinkage of fiber and plastic at different temperatures, the adhesion force will decrease with temperature changes, leading to the decrease of the screw withdrawal force. The screw holding force after the freeze-thaw cycle is greater than that of the material after the high-low temperature cycle, which is consistent with the MOR and MOE results discussed above.

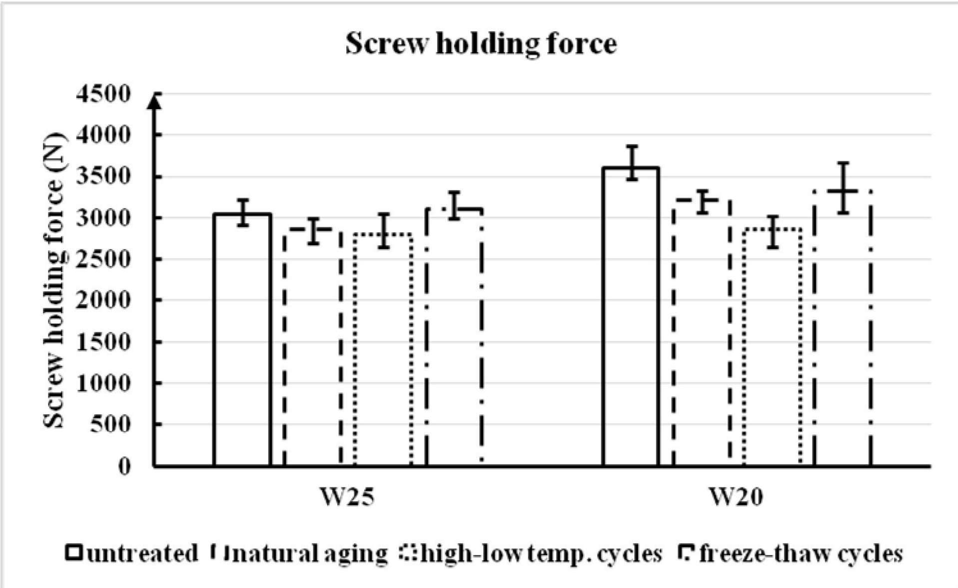

Fig. 4: Screw holding force for untreated specimens, and specimens after treatments of natural aging, high-low temperature cycles and freeze-thaw cycles.

Effects of aging on creep performance

Creep performance is an important factor for the use of WPC materials for long-term loading. The long-term load-bearing capacity of materials can be predicted by the creep performance, and thus, creep performance also describes the material's safety.

Fig. 5 shows the creep recovery rate of two groups of WPC specimens for different treatments. The creep recovery rates for the untreated specimens of W_{25} and W_{20} were 89% and 93%. After natural aging, high-low temperature cycles and freeze-thaw cycles, the rates decreased by 6%, 7%, and 1% for W_{25}, and by 9%, 14%, and 10% for W_{20}, respectively.

The creep recovery rate can reach more than 80% even though it decreases slightly after the treatments, indicating that aging has little effect on the creep performance of WPC. Therefore, WPC is a relatively safe material when it is used for building planks or other structural load-bearing materials.

Correspondence of natural aging and accelerated aging

The type of weathering appears to affect the WPC properties. The correspondence of natural aging and laboratory accelerated aging of WPC has been a problem discussed by many scholars. It was shown that DMA can be used to measure a series of short-term creep or relaxation rates at constant temperature utilizing the time-temperature-transformation software to convert the short-term test results at high temperature into the long-term actual results at room temperature that enable the prediction of the mechanical properties of WPC at room temperature over the time period of several years (Maiti 2016, Lu et al. 2018). This is also an important means for predicting the long-term performance of WPC.

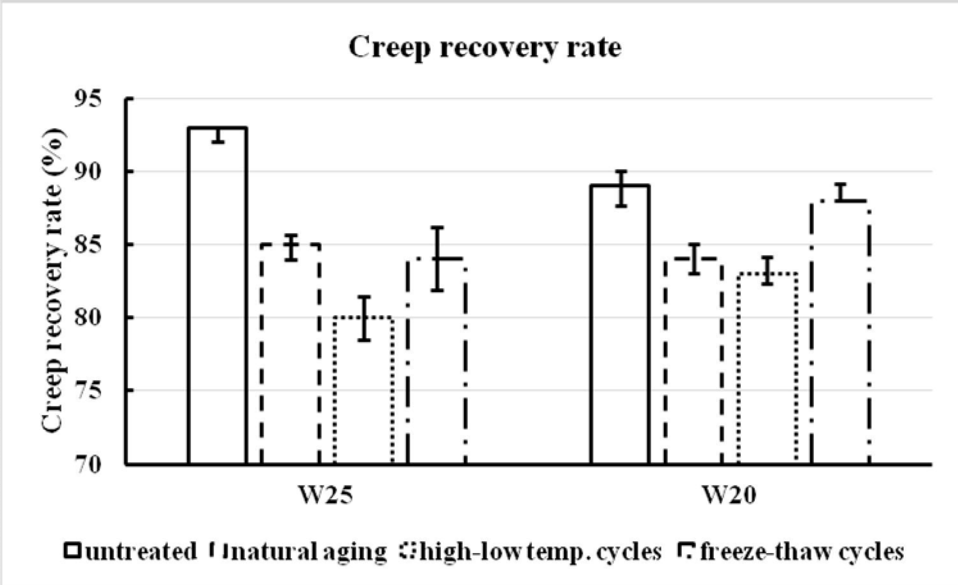

Fig. 5: Creep recovery rates for untreated specimens, and specimens after treatments of natural aging, high-low temperature cycles and freeze-thaw cycles.

CONCLUSIONS

This study evaluated the mechanical properties of WPC after the natural aging and accelerated laboratory aging. The following conclusions were reached: *(1)* After aging treatment, *i.e.*, natural aging and accelerated aging, MC of WPC increased, and mechanical properties such as MOR, MOE, screw holding force and creep recovery rate decreased as well. *(2)* The retention of MOR is greater than 86% after natural aging, high-low temperature cycles and freeze-thaw cycles. *(3)* The creep recovery rate can reach more than 80% after aging, showing that WPC is a relatively safe material for use in building floors or other structural load-bearing materials. *(4)* The mechanical properties of WPC after freeze-thaw cycles are better than those of WPC after high-low temperature cycles. *(5)* The time-temperature- transformation software of the DMA analyzer can be used to convert the short-term test results at high temperature into the long-term actual results at room temperature, so as to predict the long-term performance of WPC.

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