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THE FREEZE-THAW RESISTANCE AND LEACHING OF IMPREGNATED WOOD WASTE-CEMENT COMPOSITES

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Abstract:

The recycling of wood waste still offers a wide variety of opportunities. The possibilities for wood recycling depend mainly on the substances contained in wood waste; in particular, the recycling of impregnated wood waste, which contains environmentally harmful substances, is strictly regulated. Cement matrix composites could offer new potentials for the utilization of wood-waste fractions. The product safety of wood–cement composites, in terms of leaching and freeze-thaw resistance using de-icing salts, was investigated in this study. In a leaching test, low amounts of copper, chromium, and arsenic leached from a wood–cement composite were observed. The measured concentrations of the studied substances decreased over time. Freeze-thaw resistance of wood–cement composites was studied with four different additives typically used in concrete products. The highest resistance was obtained for the composite without any additives, but, in general, all the studied specimen, with one exception, performed relatively well.

Key words: *cement; composite; freeze-thaw resistance; impregnated wood; leaching.*

INTRODUCTION

Due to tightened environmental regulations and laws concerning waste materials, countries and companies have been forced to improve and encourage material efficiency and the recycling of wastes. For example, in the EU, 70% by weight of non-hazardous construction and demolition waste (CDW) must be prepared for re-use, recycling, or recovery as of 2020 (European Commission 2010). CDW also contains hazardous waste, such as impregnated wood, which must be handled in accordance with provided safety instructions. To meet regulatory guidelines, impregnated wood, consisting of waterborne preservatives, must be separated and removed from waste streams. Due to the significant environmental impacts of landfilling, impregnated wood materials are mainly recycled as a raw material for energy production in which case the flue gases from combustion are also cleaned and the ash is treated appropriately for the removal of metals (Solo-Gabriele et al. 2002; Kakitani et al. 2004; Helsen and Van Den Bulck 2005).

Industrial impregnation of wood is carried out as pressure impregnation using overpressure and vacuum to ensure that waterborne preservatives, i.e., impregnation agents, are absorbed deep into the wood material and thus more effectively protect the wood compared to normal surface treatment impregnation. Previously, impregnating agents containing chromium (Cr), copper (Cu), and arsenic (As) (CCA) were most widely used as preservatives to improve the biological resistance of wood materials as typical applications for impregnated wood products include outdoor and environmental constructions, where the wood is exposed to moisture and/or ground contact, and where higher safety level requirements are set for wooden structures. Since 2004, the use of CCA-treated wood in residential and public use has been forbidden in the EU, USA, and Canada, as well as in a number of Asian countries; however, industrial use is still allowed (Hingston et al. 2001; European Commission 2003; Clausen 2004; Janin et al. 2011; Zheng et al. 2015). Since then, new and more environmentally friendly waterborne impregnation agents, e.g., based on copper compounds, have been introduced. Generally, the leaching of preservative components from wood used in aquatic situations may be harmful to the environment. The leachability of wood may be affected by the characteristics of the impregnation process, as well as the environment to which the CCA-treated wood is subsequently exposed to (Hingston et al. 2001).

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Mainly due to a lack of sustainable reuse or recycling applications, the recycling potential of wood waste is still low (Berger et al. 2020) but offers a wide variety of opportunities. For example, the utilization of wood waste or residues as filler material in cement and concrete could help reduce the carbon footprint and emissions of conventional industrial processes (Amiandamhen et al. 2021). Cement is one of the most widely used materials in the field of composites and cement composites offer the possibility of recycling several types of wastes, e.g., wood and plastic residues (Andrade and Caldeira 2010), to reduce the manufacturing costs, energy, and depletion of virgin materials. Wood–cement composites (WCCs) have been widely used in the manufacture of building materials, for both interior and exterior applications, due to their excellent strength properties, as well as their good acoustic properties (Moslemi 1999). Compared to conventional wood products, WCCs also are significantly more fire-, termite- and water-resistant (Frybort et al. 2008) due to the encapsulation of wood particles.

Different types of additives are used in WCC manufacturing, due to either economic reasons, to reduce raw material costs, or for technical reasons, to improve properties, e.g., improved frost-proofing or a higher density. Freezing and thawing in the presence of salts of is one of the most destructive deterioration processes, and causes a decrease in the service life and performance of a concrete structure (Šelih 2010). The durability of concrete is closely related to its internal pore structure, i.e. the volume, radius, and size distribution of pores (Cai and Liu 1998). A lower porosity results in an improved freeze–thaw resistance for a concrete structure (Li et al. 2020). The density of concrete has a strong effect on its strength properties. A higher density of concrete generally provides a higher strength as well as a lower porosity. The properties of concrete can be improved using proper additives in the concrete paste mixture; WCCs have been extensively studied to improve their performance, as well as raise their general acceptance in different building applications.

In this study, the leaching of cement composite filled with impregnated wood chips is studied. In addition, the effect of four different additives on the durability of wood–cement composites was investigated in terms of the freeze–thaw resistance.

MATERIALS AND METHODS

Materials

The impregnated wood waste used was gathered from a local recycling plant, located in Lappeenranta, Finland. The wood waste was reduced to flakes of approx. 4.0mm using a Shini SG-1635N low-speed granulator. The mixing of cement with impregnated wood flakes and additives was performed using a manual electric mixer (Biltema Mixer PCM 160). Cement, wood flakes and additives were dosed by weight. All specimens contained 5 wt% wood flakes. As the cement, Plus Cement (Finnsementti Ltd, Finland) was used.

Four different commercially available additives were used in this research: alumina cement (A) (Cementos Molins Industrial S.A.), blast furnace slag (B) (Finnsementti Ltd, Finland), Ecofax20 (E) (Fatec Ltd, Finland), and Gepocit™ Surfactant (S) (Apila Group Ltd, Finland). Alumina cement and blast-furnace slag are hydraulic binders, Ecofax20 is a powder-form replacement material for cement, and Surfactant is a surface-active agent that is typically used to improve the workability of concrete and increase the strength and durability properties of concrete structures. The addition amount followed the instructions given by the manufacturer of each additive. A specimen without the additives was used as a reference. The compositions of the wood–cement composites are presented in Table 1.

Table 1

Composition of wood–cement composites, in wt%

	Wood	**Cement**	**Additive**
W-C	5	95	-
W-C-A	5	91	4
W-C-B	5	15	80
W-C-E	5	90	5
W-C-S	5	93	2

The preparation of the specimens was done in accordance with the standard SFS-EN 12390-2. Specimens were made in the form of 150×150×150mm cubes in plastic molds. After 24 hours, the specimens were demolded and placed into a bath filled with tap water and were maintained at a temperature of 20±2°C. The specimens were left in water for 28 days. After this, the specimens were cut with a diamond saw to the specific dimensions required by the tests.

Analysis

The release of dangerous substances from wood–cement composite was determined using a dynamic surface leaching test. The test was performed according to the standard CEN/TS 16637-2:2014. Specimens were cut from prepared cubes and the dimensions of the studied specimens were 40×40×40mm. Measurements were performed after two steps: after 6 hours and after 18 hours. Deionized water was used as a leachant. The volume of leachant (V_l) was defined using Equation (1), and the area release of a substance for each step (r_i) (fraction) was calculated according to Equation (2):

$$V_l = (L/A) \times A \tag{1}$$

where: *L/A* ratio of the monolithic products is 80±10 L/m^2
A is the surface area of the specimen in square meters

$$r_i = (c_i \times V/A) \times 0.001 \tag{2}$$

where: c_i is the concentration of the substance in eluate i, in micrograms per liter
V is the volume of the leachant, in L
A is the area of the test portion, in square meters

The freeze-thaw resistance with de-icing salt of the wood–cement composite specimens was studied in accordance with standard SFS-EN 1338. Specimens were cut from prepared cubes so that the area of the test surface was approx. 8000 mm^2 and the thickness of the specimen was 100mm. Except for the test surface, all the other surfaces of the specimen were covered with a rubber sheet for the entire testing time. The edge of the rubber sheet reached 20±2mm above the test surface. A layer of 3% NaCl solution was added to the test surfaces. The test pieces were frozen and thawed for a total of 28 cycles. After this, the material that had scaled off was collected, rinsed with filter paper to remove salts, and dried in an oven at 105±5°C for 24 hours. The dry mass of the material was weighed to determine the mass loss and the loss of mass (L) was calculated according to Equation (3):

$$L = M/A \tag{3}$$

where: M is the mass of the scaled-off material in kilograms
A is the surface area of the specimen in square meters

RESULTS AND DISCUSSION

The results of the leaching test are presented in Table 2. The cumulative release of the impregnating substances, after the steps of 6h and 18h, from the studied wood–cement composite, were as follows: chromium (Cr) 4.23mg/m^2, copper (Cu) 0.49mg/m^2, and arsenic (As) 0.09mg/m^2. Compared to the reference specimen, which was pure deionized water, the release of chromium in the W-C specimen was 14-fold, and that of copper was 2.5-fold. In the reference specimen, the release of arsenic was not detected at all. The releases after the first step (6 h) of the leaching procedure were higher for all studied wood impregnates compared to the corresponding releases measured after the second step at 18 hours. Based on this, it can be stated that the most significant percentage of dissolution for chromium, copper, and arsenic, as well as other observed dissolutions, occurred during the first step of the leaching procedure.

Table 2

Leaching of chrome (Cr), copper (Cu), and arsenic (As) after 6 h and 18 h

	Cr [mg/m^2]	*Cu [mg/m^2]*	*As [mg/m^2]*
W-C 6 h	*2.58*	*0.29*	*0.05*
Ref 6 h	*0.09*	*0.08*	-
W-C 18 h	*1.65*	*0.20*	*0.04*
Ref 18 h	*0.20*	*0.12*	-

Earlier studies related to WCCs (Huang and Cooper 2000; Qi 2001) reported that a cement matrix reduces the leaching of copper and arsenic quite well after CCA-treated wood was mixed with cement, but relatively high concentrations of chromium were still measured. It was also observed that the leaching of chromium in concrete was mainly affected by the water-cement ratio, and that it can be reduced slightly with the addition of silica fume and iron (II) sulfate ($FeSO_4$). The results are consistent with the results of the present study, in which a significantly higher concentration of chromium relative to copper or arsenic was also measured. It has been observed that leaching decreases significantly over time (Hingston et al. 2001). In addition, the leaching of the various components of CCA was not equal over time, or proportional, to their formulation concentrations (Coles et al. 2014). Overmann et al. (Overmann et al. 2021) studied the leaching of fresh and hardened concrete and found that the release of fresh materials was significantly higher than that of hardened materials due to the decreasing diffusion coefficient due to progressive hydration. Kamdem et al. (Kamdem et al. 2004) studied CCA-treated, wood-flour plastic composites, and measured higher leaching concentrations for arsenic and chromium than those observed for copper.

The mass loss of the studied wood–cement composites filled with different additives after 28 freeze-thaw cycles varied heavily, as shown in Table 3. For all the studied specimens, there was a noticeable deterioration of the exposed surfaces. The lowest mass losses were observed for the reference specimen, W-C, and specimens filled with surfactant, W-C-S. The good performance of W-C-S was an expected result because surfactants are typically used to entrain air voids in fresh concrete paste to improve both the workability and freeze–thaw durability of concrete (Qiao et al. 2017). According to the criterion of standard SFS-EN 1338, the mass loss of any specimen must not exceed 1.50kg/m^2. Of the studied specimens, only W-C-B very clearly exceeded this criterion. In addition, the mass loss of W-C-E was barely over the limit.

The results revealed that the loss of mass partly correlated with the density of the specimen; a low density manifested as low loss of mass. Wolfe and Gjinolli (Wolfe and Gjinolli 1999) made similar observations when examining the freeze–thaw resistance of wood-cement composites and found that the lowest mass loss, less than 5%, was reached with a sample of a low density. They explained this with a greater void volume, which provided more space for free water to expand upon freezing, without cracking the cement matrix. Previous research, for example (Ronquim et al. 2014; Castro et al. 2018), has shown that the use of a smaller size of wood particles in wood–cement composites leads to a higher density, i.e., lower void volume, which also results in improved strength properties.

Table 3

The density and loss of mass of the wood-cement composites

	Density [kg/m^3]	*Loss of mass [kg/m^2]*
W-C	*1448*	*0.26*
W-C-A	*1727*	*0.80*
W-C-B	*1870*	*5.28*
W-C-E	*1801*	*1.52*
W-C-S	*1517*	*0.28*

Another valid benchmark for the studied WCCs is concrete, which is the most common and widely used cement-based material. The resistance of concrete to freeze–thaw has been extensively studied using different variables because degradation of concrete exposed to freezing and thawing is the most common cause of destruction of concrete structures (Skripkiunas et al. 2013). Skripkiūnas et al. (Skripkiunas et al. 2013) studied the effect of different cement types on the freeze-thaw resistance of concrete and observed that mass loss varied between 0.1kg/m^2 to 5.5 kg/m^2 after 28 cycles. The lowest mass loss was detected for concrete made from blast-furnace cement. In the study of Šeps et al. (Šeps et al. 2016) a total mass loss of 2.28kg/m^2 after 25 freeze-thaw cycles was measured for fiber-reinforced concrete filled with recycled aggregate. Sáez del Bosque et al. (Sáez del Bosque et al. 2020) found that concrete filled with mixed recycled CDW aggregate was not resistant to freeze-thaw in the presence of de-icing salts, as the mass loss after 56 cycles with de-icing salts was measured at between 3 and 4kg/m^2.

CONCLUSIONS

The leaching and freeze-thaw resistance with de-icing salt of WCCs filled with impregnated wood waste was studied. In terms of leaching, releases were observed for all the studied impregnating preservatives, i.e., chromium, copper, and arsenic. The highest cumulative release leached was measured for chromium. The detected concentrations were quite low for all studied preservatives and they decreased over time. Freeze-thaw resistance with de-icing salt was tested using five different wood–cement composite specimens. The lowest loss of mass was observed for the composite without additives. An equal result was obtained for the W-C-S. Only the result for W-C-B clearly exceeded the standard criterion. Therefore, it can be concluded that the utilization of impregnated wood waste as a filler in cement composites is possible if regulations allow this.

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