Is Wollastonite Capable of Improving the Properties of Wood Fiber-cement Composite?

Ali Hassanpoor Tichi,[a] Behzad Bazyar,[a,*] Habibollah Khademieslam,[a] Hossein Rangavar,[b] and Mohammad Talaeipour [a]

Effects of wollastonite substitution were investigated relative to the mechanical, physical, and microstructural properties of a wood fiber-cement composite. Wollastonite content of 0%, 3%, 6%, and 9% and lignocellulosic material (kraft fibers) content of 10%, 20%, and 30% were used based on the dry weight of cement. Then the lignocellulosic material and the resulting board samples were compared to a control (without wollastonite). Modulus of rupture (MOR), modulus of elasticity (MOE), water absorption, and fire resistance tests were conducted to examine the characteristics of the board composite. The results showed that the mechanical properties of wood fiber-cement composite were improved by the 9% wollastonite substitution. The fire-resistance of the composite board was improved when the wollastonite content was increased. Furthermore, cement boards with 9% wollastonite exhibited lower water absorption in comparison to the other specimens. Scanning electron microscopy (SEM) results showed that the calcium hydroxide formed hydrated calcium silicate gel (C-S-H gel) after the addition of wollastonite. The SEM images showed that the micro-structure of the boards were improved by increasing the nano-wollastonite content.

Keywords: Kraft fibers; Cement boards; Fire resistance; Wollastonite

*Contact information: a: Department of Wood and Paper Science, Faculty of Natural Resources and Environment, Science and Research Branch, Islamic Azad University, Tehran, Iran; b: Department of Wood Industries, Faculty of Civil Engineering, Shahid Rajaee Teacher Training University (SRTTU), Tehran, Iran; *Corresponding authors: bazyar@srbiau.ac.ir*

INTRODUCTION

Wood fiber-cement composite are produced and have been used since 1895 (Alpar 2009). The most known product types include wood wool (excelsior) boards, cement bonded particle boards, fiber-cement products, and building blocks (Alpar 2000). Wood-cement products are already used worldwide for roofs, floors, and walls. They have numerous advantages when compared to panels produced with organic resins, such as high durability, good dimensional stability, good acoustic and thermal insulation properties, and low production cost (Na *et al*. 2014). The main problem with producing WCC is the incompatibility of cement and wood. Hydration and MOR and MOE of WCC are sensitive on wood extractives. Water dissolves water-soluble chemicals of wood and some of these are inhibitors, such as sugars (*e.g.*, hexoses: mannose or glucose), tannin, and hemicellulose (*e.g.*, glucomannan, xylan, arabinogalactan, or galactan), and these hinder or stop this hydration of cement. Hardwoods due to higher amount of wooden extractives (soluble xylans) are generally less compatible than softwoods (Naji Givi *et al*. 2010).

The use of admixtures in wood-cement to accelerate the curing process can be further subdivided into the use of mineral and chemical admixtures. Chemical admixtures include magnesium chloride ($MgCl_2$), calcium chloride ($CaCl_2$), water glass (Na_2SiO_2), and aluminum silicate ($Al_2(SO_4)_3$) (Alpar 2009). The most common mineral admixture used as an accelerator is microsilica, or silica fume, ash, and wollastonite. This material, as cement curing accelerators, helps to improve wood-cement compatibility. Alternatively, the use of pretreatments, such as aqueous extraction, to remove inhibitory substances from wood is another method to improve matrix compatibility (Na *et al*. 2014). Commercial grades of wollastonite are typically high in purity because most ores must be beneficiated by wet processing, high-intensity magnetic separation, and/or heavy media separation to remove accessory minerals. The minerals most commonly found associated with wollastonite are calcite (calcium carbonate), diopside (calcium magnesium silicate), and garnet (calcium aluminum silicate). Wollastonite is hard, white, and alkaline (pH 9.8). It is exploited for its chemistry as a source of CaO and SiO_2, and its low ignition loss, low oil absorption, low moisture absorption, and fire-retarding properties (Ciullo 1997). The effect of wollastonite has been reported to improve the dimensional stability of solid woods (Poshtiri *et al*. 2013), and to increase the thermal conductivity coefficient of medium-density fiberboards (MDF) (Taghiyari *et al*. 2013). Fire-retarding properties of wollastonite have been noted in the literature when it is used in solid wood and wood composite materials (Poshtiri *et al*. 2013; Taghiyari *et al*. 2013). Khosrviyan (2009) found that the physical and mechanical properties of wood-plastic composite were improved with the addition of wollastonite. Moreover, wollastonite has antifungal properties (Taghiyari *et al*. 2014a, 2014b). A wide variety of fibers, from different forms and origins, have been used to reinforce cementitious matrices, while kraft pulp is the most common fiber form used. This is because lignocellulosic fibers are a cheap raw material that are used by the paper industry; they can be easily dispersed in water and can represent a basic component for the production of cementitious materials. ACI 544 recommends that kraft pulps are favored for their use in cement-based materials due to their low lignin and hemicelluloses levels, which are less alkali resistant than cellulose that has been obtained from the kraft pulping process (ACI 544, 2002).

The production of waste biofiber wood fiber-cement composite reinforced with nano-SiO_2 particles as a substitute for asbestos cement composites showed that the addition of silica nanoparticles to mixtures increases mechanical properties. Additionally, the physical properties was improved by the addition of up to 1% silica nanoparticles (Hosseinpourpia *et al*. 2012). Increasing the wollastonite microfiber content resulted in a compressive strength comparable to or higher than that of the control mixture without microfibers. Wollastonite microfibers reduced shrinkage strains and increased cracking resistance compared to that of the control mixture. However, no noticeable improvement in the flexural behavior was achieved with the addition of wollastonite microfibers due to a sudden rupture of microfibers within the matrix (Soliman 2011). Del Meneéis *et al*. (2007) produced oriented strand board (OSB) from pine (*Pinus tadea*) and Portland cement by mixing them in a 1:1 ratio. Cement was partly substituted by silica fume (SiO_2) in proportions of 0%, 10%, or 20%. The best results were observed for the board made with cement that had 10% SiO_2 substitution.

This research reports the preparation and the properties of wood fiber-cement composite that were reinforced with wollastonite. The morphological, mechanical, physical, and fire retardancy properties of the wood fiber-cement composite with different compositions were also investigated.

EXPERIMENTAL

Materials

The cement used in this study was type II Portland cement; it was purchased from Abyeik Company (Qazvin, Iran). The natural fibers used in this research were kraft pulp fibers obtained from Mazandaran Wood and Paper Industry (Sari, Iran). Commercial wollastonite (WOLL) and calcium chloride were purchased from Vard Manufacturing Company of Mineral and Industrial Products (Tehran, Iran) and Merck Company (Darmstadt, Germany), respectively.

Sample preparation

Specifications of the wollastonite composition are given in Table 1. Kraft pulp fibers were applied at three proportions (10%, 20%, or 30%, with respect to the total content of binder in each mixture), and different wollastonite substitution levels (0%, 3%, 6%, or 9%) for cement.

Table 1. Inorganic Composition of Wollastonite Used in This Study (Taghiyari *et al.* 2013)

Inorganic Component	Percentage (%)
SiO_2	46.96
CaO	39.77
Fe_2O_3	2.79
Al_2O_3	3.95
TiO_2	0.22
K_2O	0.04
MgO	1.39
Na_2O	0.16
SO_3	0.05

Composite production

First, the cement powder and kraft pulp fibers were mixed together by hand. Next, water and wollastonite were mixed using a mortar mixer at a moderate speed (200 rpm) for 2 min. Finally, all the materials were stirred at a high speed (600 rpm) for 4 min. The mixture was then poured into molds with the dimensions of 40 cm × 30 cm ×4 cm. The obtained mat was pressed (30 kg/cm^2) using a Burkle LA-160 flat press (Babol, Iran) for 10 min at room temperature to obtain a target thickness of 12 mm. The density was kept constant for all treatments (1.1 g/cm^3). The boards were conditioned at standard conditions (20 ± 1 °C, 65 ± 2% relative humidity) for an additional 28 days for the cement to cure.

Methods

Mechanical tests

The samples were then evaluated for flexural moduli of elasticity and rupture according to DIN EN 634 parts 1 and 2 (1995) using a universal testing machine (model GT-TCS-2000; Taichung Industry Park, Taichung, Taiwan) at the speed of 10 mm/min. The specimens were trimmed to the dimensions of 350 mm × 50 mm × 12 mm for the mechanical tests. Three samples were evaluated for each treatment. The load and deflection were continuously recorded and the data were used to calculate flexural modulus of elasticity (MOE) and modulus of rupture (MOR) according to Eqs. 1 and 2,

$$MOR = 1.5\frac{FL}{bd^2} \tag{1}$$

$$MOE = \frac{FL^3}{4bd^3D} \tag{2}$$

where F is the maximum force (N), L is the span length (mm), b is the sample width (mm), d is the sample thickness (mm), and D is the deflection.

Physical tests- Water absorption

The effect of composite formulation on the water uptake was determined in samples with the dimensions of 50 mm × 50 mm × 12 mm according to DIN EN 634 parts 1 and 2 (1995). The sample was initially oven-dried for 48 h at 103 °C, and the mass was recorded. The sample was then soaked in distilled water for 2 h and 24 h. Water uptake was then calculated based on the initial oven-dried mass and the wet mass values.

Mass loss due to fire activation

The specimens (150 length × 100 width × 12 thickness, mm^3) were prepared according to the ISO 11925 (2010) specifications for the fire resistance tests. Mass loss of the samples due to fire exposure was vertically mounted on a holder up-straight and exposed to a Bunsen-type burner (with the internal diameter of 11 mm) hold at 45 degrees to the surface of the specimen for 120 seconds in accordance with method described by Ayoub Esmailpour *et al*. (2017).

Scanning electron microscopy (SEM)

To confirm the distribution of wollastonite, SEM imaging was carried out at the thin-film laboratory, FE-SEM lab (Field Emission), School of Electrical and Computer Engineering, University of Amir Kabir, Tehran, Iran. One specimen with dimensions of 10 mm × 10 mm was prepared for SEM imaging for each treatment.

Statistical analyses

Statistical analyses were performed using Statistica software (Dell Inc., v.13, Landolock, TX, USA) at a significance level of $\alpha = 0.05$. The obtained results were analyzed statistically, and an analysis of variance (ANOVA) was performed to determine the significance of the tested parameter. A Duncan's multiple range test (DMRT) was performed to compare treatment means.

RESULTS AND DISCUSSION

Mechanical Properties

The results from the mechanical testing are shown in Figs. 1 and 2. As shown, the MOR and MOE values increased in the cement boards that contained wollastonite compared to the cement boards without wollastonite.

For the sample containing 9% wollastonite and 10% kraft pulp fibers, the MOR value increased up to 7.84 MPa, which was 36% higher than the control sample (0% wollastonite). The MOE values were higher with the wollastonite treatments. The highest MOE value (12,400 MPa) was observed with 9% wollastonite substitution and 10% fibers, which was 29% higher than the control. Because water and cellulosic materials were added to the cement, the pH of the wood–cement mixture increased to approximately 12.5, which facilitated the dissolution of wood constituents, particularly low molecular-weight carbohydrates, and extractives materials (phenolic and sugars). These compounds could have inhibited cement hydration (Sandermann and Kohler 1964), which could have reduced the wood fiber-cement composite strength. The high silica (SiO_2) level of wollastonite cause the formation of calcium silicate hydrates, which increases the heat of hydration of cement and improves the connection between fibers and cement (Naji Givi *et al*. 2010).

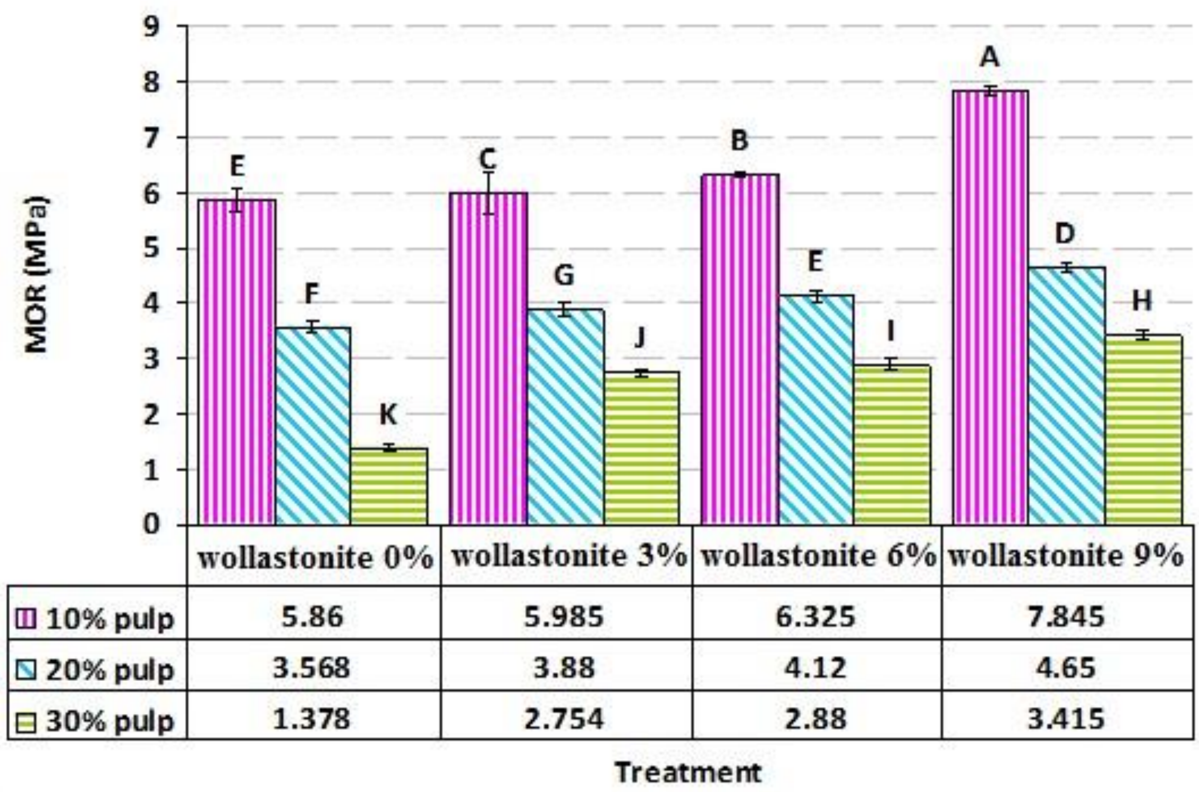
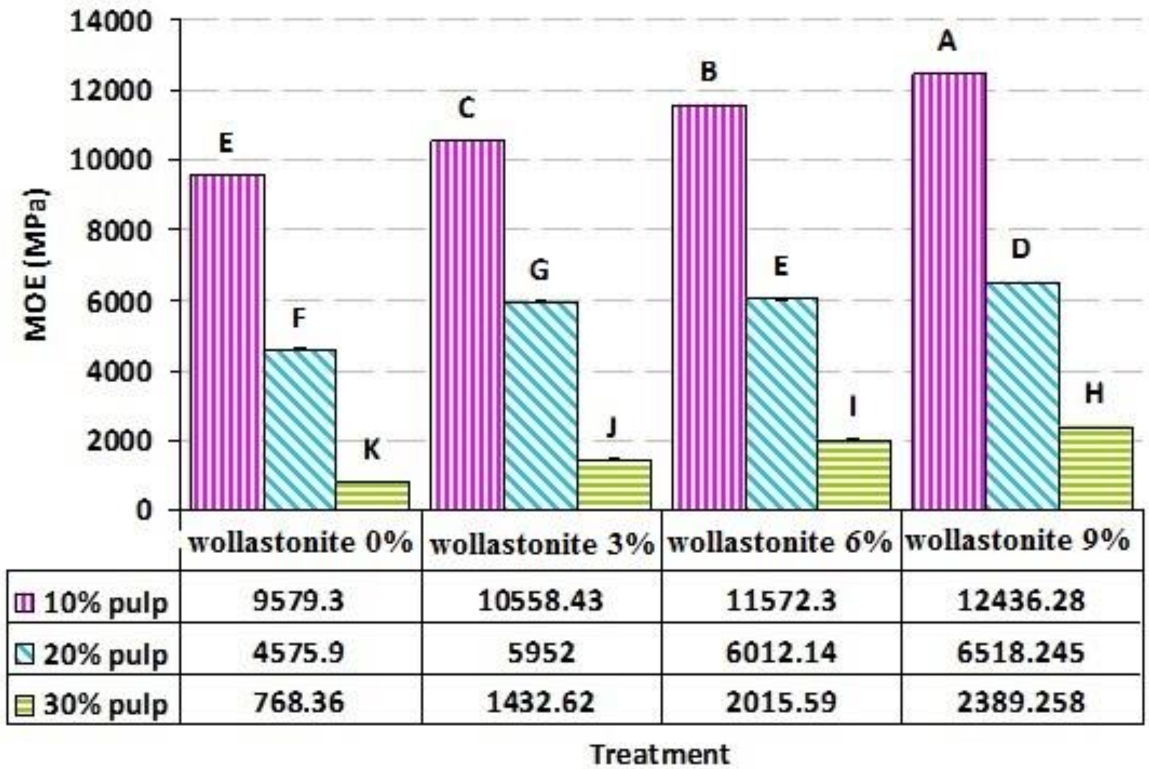


Fig. 1. Effect of varying levels of kraft pulp fiber and wollastonite on the MOR; letters on each column indicate Duncan's grouping at the 95% level of confidence



	wollastonite 0%	wollastonite 3%	wollastonite 6%	wollastonite 9%
10% pulp	9579.3	10558.43	11572.3	12436.28
20% pulp	4575.9	5952	6012.14	6518.245
30% pulp	768.36	1432.62	2015.59	2389.258

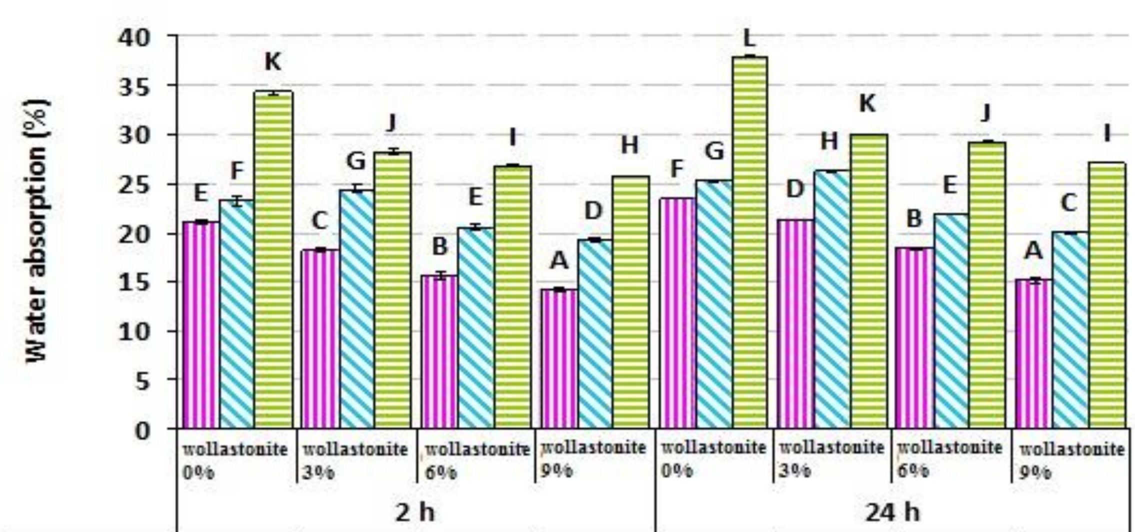
Fig. 2. Effect of varying levels of kraft pulp fiber and wollastonite on the MOE; letters on each column indicate Duncan's grouping at the 95% level of confidence

Silica in the wollastonite is converted calcium hydroxide, which is released by the hydration to $CaCO_3$ and accelerates the thermal reaction of cement hydration. Additionally, increasing the mechanical strength *via* increasing the wollastonite addition produces the reaction between calcium chloride, calcium hydroxide, and wollastonite, which provides the required heat for cement hydration and formation of calcium silicate gel. High substitution levels of wollastonite mitigated the negative effect of the extractive materials (Karimi *et al*. 2012; Ma and Wang 2012). Consequently, the mechanical properties were improved.

Physical Tests

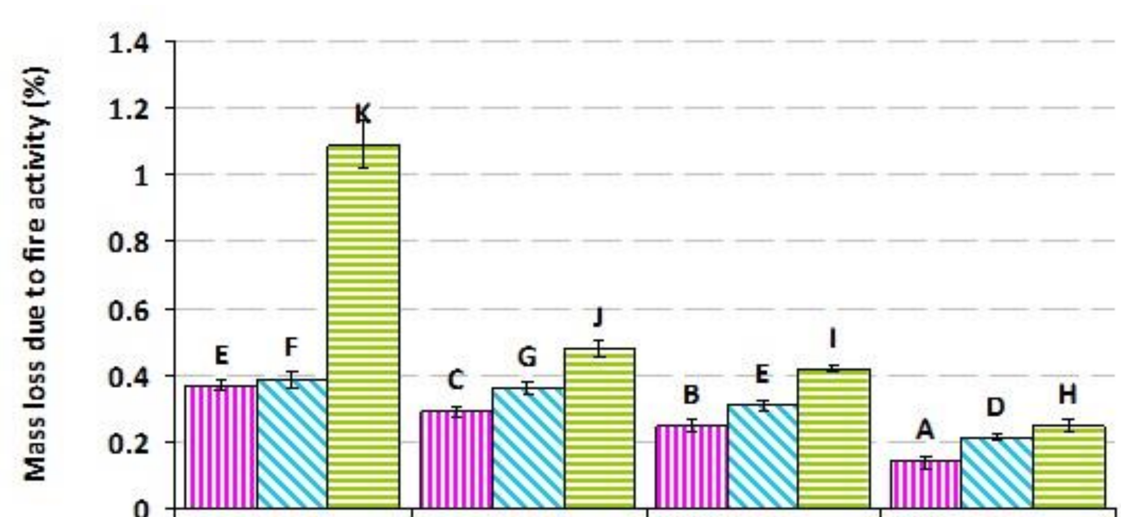
Water absorption

The water absorption of the samples is shown in Fig. 3. The lowest water absorption was observed for 9% wollastonite and 10% kraft fibers, and the highest was observed for the sample without wollastonite and 30% fibers (control). The wollastonite decreased the water absorption of samples at 2 h and at 24 h. A reason for this observation could be related to the hydrophobic character of wollastonite (Hosseinpourpia *et al*. 2012). Khosrviyan (2009) reported similar results for wood-plastic composite and proposed it was associated to the hydrophobic characteristic of wollastonite.



	wollastonite 0% (2 h)	wollastonite 3% (2 h)	wollastonite 6% (2 h)	wollastonite 9% (2 h)	wollastonite 0% (24 h)	wollastonite 3% (24 h)	wollastonite 6% (24 h)	wollastonite 9% (24 h)
10% pulp	21.05	18.23	15.58	14.25	23.45	21.28	18.47	15.12
20% pulp	23.25	24.36	20.59	19.32	25.24	26.25	21.84	20.02
30% pulp	34.23	28.21	26.79	25.71	37.86	30.01	29.14	27.02

Fig. 3. Effect of varying levels of kraft pulp fiber and wollastonite on the water absorption (WA); letters on each column indicate Duncan's grouping at the 95% level of confidence



	wollastonite 0%	wollastonite 3%	wollastonite 6%	wollastonite 9%
10% pulp	0.37	0.29	0.25	0.14
20% pulp	0.39	0.36	0.31	0.215
30% pulp	1.09	0.48	0.42	0.25

Fig. 4. Effect of varying levels of kraft pulp fiber and wollastonite on the mass loss (ML) due to fire exposing; letters on each column indicate Duncan's grouping at the 95% level of confidence

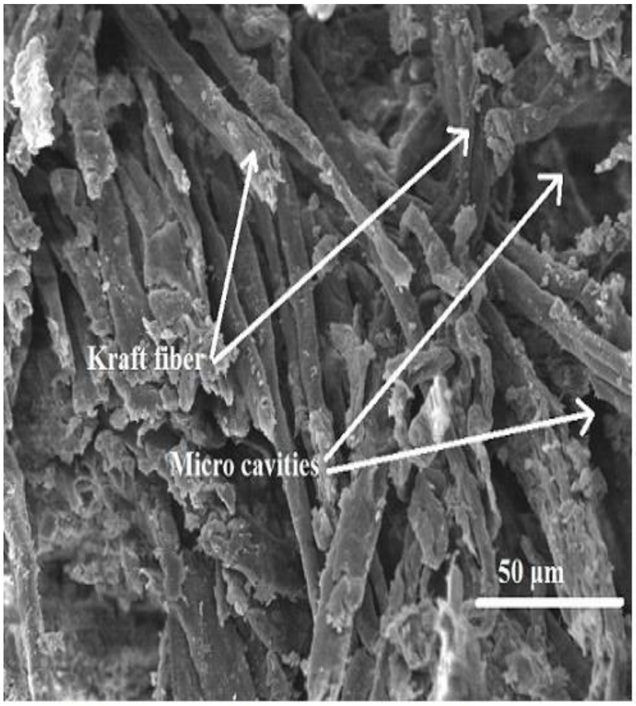


Fig. 5a. Weak structure of cement composites without wollastonite

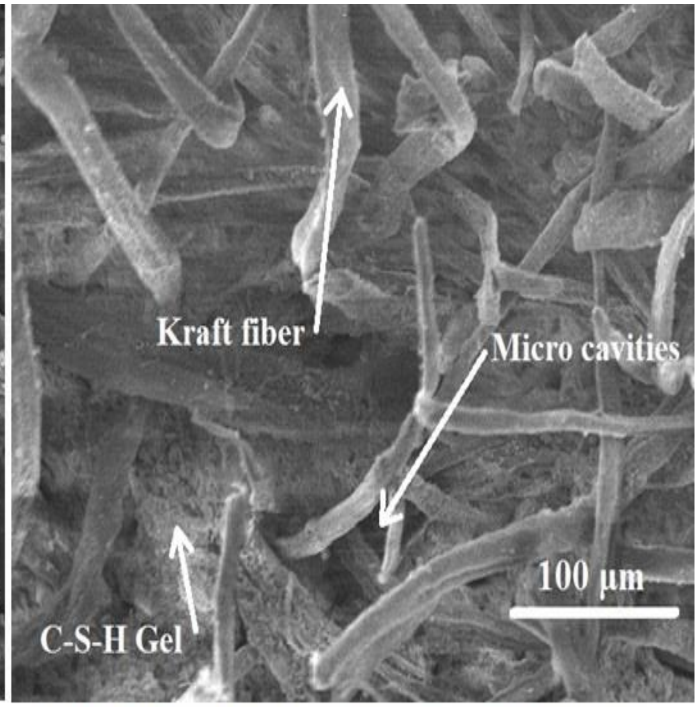


Fig. 5b. SEM image of fiber-cement composites with 3% wollastonite

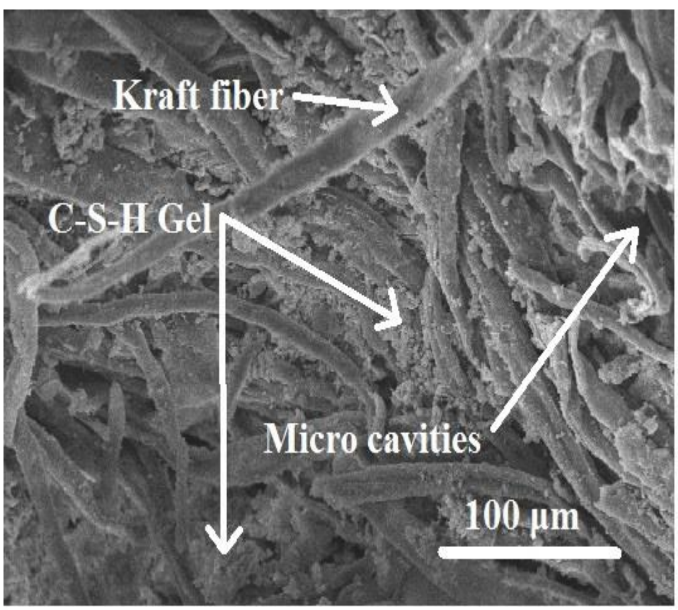


Fig. 5c. SEM image of fiber-cement composites with 6% wollastonite

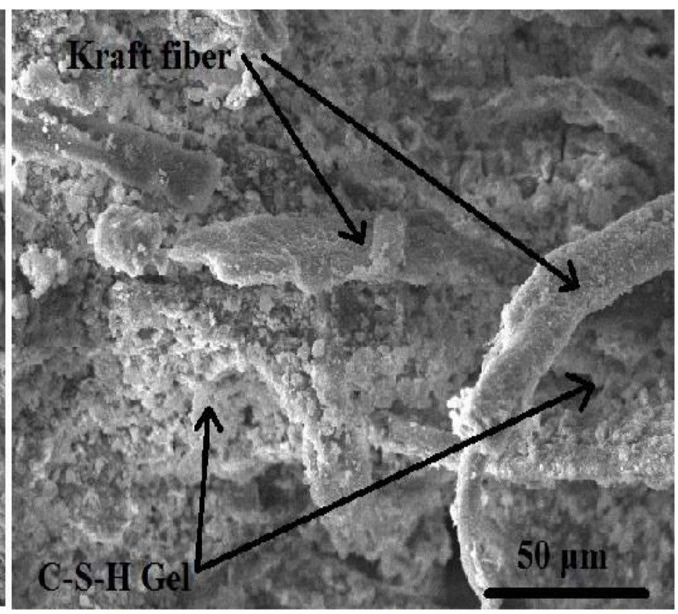


Fig. 5d. SEM image of fiber-cement composites with 9% wollastonite

Mass Loss due to Fire Exposure

Fire retardancy is an important characteristic when using lignocellulosic composite. The results indicated that the fire-retarding properties of the samples were improved by the addition of wollastonite (Fig. 4). The lowest mass loss was noted with the sample containing 9% wollastonite and 10% kraft fibers. An important characteristic of wollastonite is its fire resistance; thus, boards containing wollastonite exhibited higher fire resistance and a lower percentage of mass loss when compared to the control samples without wollastonite (Hosseinpourpia *et al*. 2012). Wollastonite has a high thermal conductivity, which facilitates heat transfer (Taghiyari *et al.* 2014c). Therefore, wollastonite increased fire-resistance in *Populus* wood. The heat-conductivity of wollastonite limited the accumulation of heat at one spot. Furthermore, it acted as a physical barrier to deter heat and mass transfer between the gas and the condensed phase. Consequently, fire-retarding properties were improved, and could decrease the weight loss percentage in the boards with wollastonite (Haghighi Poshtiri *et al*. 2013; Esmailpour *et al*. 2018). Research by Haghighi *et al.* (2013) reported that fir wood saturated by wollastonite had a higher fire resistance than samples without wollastonite, which mirrored the results observed in this research.

Morphology of Composite

The morphology of the wood fiber-cement composites was examined using SEM analysis. In Figures 5a, 5b, 5c, and 5d, can see different amounts of nano wollastonite percentages in the boards. Also, by increasing the percentage of nano wollastonite from zero to 90 percent, the formation of the C-S-H gel increased and in the opposite direction, the micro cavities decreased. The SEM images illustrate a uniform distribution in the wood fiber-cement composite structure with incorporation of 9% wollastonite (Fig. 5d). This can be attributed to the role of wollastonite in bridging the microcracks leading to a delay in micro crack coalescence. Figure 5d is a sample containing 9% wollastonite and 10% kraft fibers; this composition was also effective in the microstructural improvement of a cement matrix. As shown, the lower amounts of harmful crystals, such as $Ca(OH)_2$ and ettringite, were observed because of the wollastonite behavior in the cement matrix, which reinforces the matrix and turns the harmful crystals of $Ca(OH)_2$ to a C–S–H gel. The presence of $Ca(OH)_2$ crystals and needle-like ettringite, as well as fibrillated C–S–H gel, can be observed at low levels in the composite without wollastonite in Fig. 5a. In this image, some of the kraft fibers did not perform well in terms of bending strength due to the lack of adhesion with cement hydration products. The micro-cavities in the control specimens (without wollastonite) increased the sample's water absorption. This observation was similar to the reports of Hosseinpourpia *et al*. (2012).

CONCLUSIONS

1. The mechanical and physical properties of wood-cement panels mixed with wollastonite were examined. The results indicated that the wollastonite addition to the board composite improved the MOE and MOR, decreased the water absorption, and decreased the mass loss of the composite when exposed to fire.
2. Wollastonite improved the compatibility of kraft pulp fibers with the cement matrix. The decrease in water absorption of the boards was attributed to the hydrophobic characteristic of wollastonite.

3. The high thermal conductivity of wollastonite increased heat transfer, which improved the fire resistance of the composite boards.
4. It is recommended that wood fiber-cement composite boards contain 10% kraft fibers and 9% wollastonite to obtain the highest values of mechanical properties and the lowest mass loss when exposed to fire.

ACKNOWLEDGEMENTS

The authors express their gratitude to Mr. Mohammad Hossein Vardi, the Managing Director of VARD Mineral and Industrial Products Company, who provided the wollastonite. The authors gratefully acknowledge Sheikh Kazemi and Hadi Ghasemi for their considerable assistance in conducting the tests to prepare the kraft pulp fibers.

REFERENCES CITED

ACI Committee 544 (2002), *State of the Art Report of Fiber Reinforced Concrete* (ACI-544.1R -96) (Reapproved 2002), American Concrete Institute, Farmington Hills, MI, EUA.

Alpar, T. (2000). *Methods on Accelerating Curing of Cement when Producing Cement Bonded Particle Boards*, Ph.D. Dissertation, University of Sopron, Sopron, Hungary (in Hungarian).

Alpar, T. (2009). *Wood-cement Compatibility*, Ph.D. Dissertation, Georg August University, Göttingen, Germany.

Ciullo, P. A. (1997). "The Industrial Minerals," in: *Industrial Minerals and Their Uses*, William Andrew – Elsevier, USA, pp. 75-77.

Del Meneeis, C. H. S., Castro, V. G., and Souza, M. R. (2007). "Production and properties of a medium density wood-cement boards produced with oriented strands and silica fume," *Maderas-Cienc. Tecnol.* 9(2), 105-115.

DIN EN 634-1. (1995). "Cement-bonded particleboards - Specifications - Part 1: General requirements," German.

DIN EN 634-1. (1995). "Cement-bonded particleboards - Specifications - Part 2: Requirements for OPC bonded particleboards for use in dry, humid and external conditions," German.

Esmailpour, A., Taghiyari H. R., Nouri P., and Jahangiri A. (2017). "Fire-retarding properties of nanowollastonite in particle-board," *Fire and Materials*. DOI: 10.1002/fam.2493

Haghighi Poshtiri, A., Taghiyari, H. R., and Karimi, A. N. (2013). "The optimum level of nano-wollastonite consumption as fire -retardant in poplar wood (*Populus nigra*)," *Int. J. Nano Dimension* 4(2), 141-151.

Hosseinpourpia, R., Varshoee, A., Soltani, M., Hosseini, P., and Ziaei Tabari, H. (2012). "Production of waste bio-fiber cement-based composites reinforced with nano-SiO_2 particles as a substitute for asbestos cement composites," *Constr. Build. Mater.* 31, 105-111. DOI: 10.1016/j.conbuildmat.2011.12.102

ISO 11925 (2010). "Reaction to fire tests - Ignitability of building products subjected to direct impingement of flame - Part 2," International Organization for Standardization, Geneva, Switzerland.

Karimi, A., Poshtiri, A. H., Taghiyari, H. R., Hamzeh, Y., and Enayati, A. A. (2012). "Effects of nano-wollastonite impregnation on fire resistance and dimensional stability of poplar wood," in: *The International Research Group on Wood Protection*, Kuala Lumpur, Malaysia.

Khosrviyan, B. (2009). *The Study of Mechanical, Physical, Thermal and Morphological Properties of Hybrid Multi-Structures and Nano Hybrid Polypropylene Wood Flour/Wollastonite Multi- Structures*, M.S. Thesis, University of Tehran, Tehran, Iran.

Ma, X. X., and Wang, C. G. (2012). "Hydration characteristics of mixture of grapevine and cement," *Journal of Nanjing Forestry University* 36(3), 157-159.

Naji Givi, A., Abdul Rashid, S., Aziz, F. N. A., and Salleh, M. A. M. (2010). "Experimental investigation of the size effects of SiO_2 nano-particles on the mechanical properties of binary blended concrete," *Compos. Part B- Eng*. 41(8), 673-677.

Sandermann, W., and Kohler, R. (1964). "Studies on inorganic-bonded wood materials. Part 4: A short test of the aptitudes of woods for cement-bonded materials," *Holzforschung* 18(1-2), 53-59.

Soliman, A. M. (2011). *Effect of Wollastonite for Compression Strength of Concrete*, Ph.D. Dissertation, University of Western Ontario, London, Canada.

Taghiyari, H. R, Mobini, K., Sarvari Samadi, Y., Doosti, Z., Karimi, F., Asghari, M., Jahangiri, A., and Nouri, P. (2013). "Effects of nano-wollastonite on thermal conductivity coefficient of medium-density fiberboard," *J. Nanomater. Mol. Nanotechnol.* 2(1), 1-5. DOI: 10.4172/2324-8777.1000106

Taghiyari, H. R., Bari, E., Schmidt, O., Tajick Ghanbary, M. A., Karimi, A., and Tahir, P. M. D. (2014a). "Effects of nano-wollastonite on biological resistance of particleboard made from wood chips and chicken-feather against *Antrodia vaillantii*," *Int. Biodeter. Biodegr*. 90(7), 93-98. DOI: 10.1016/j.ibiod.2014.02.012

Taghiyari, H. R., Bari, E., and Schmidt, O. (2014b). "Effects of nanowollastonite on biological resistance of medium-density fiberboard against *Antrodia vaillantii*," *Eur. J. Wood. Prod*. 72(3), 399-406. DOI: 10.1007/s00107-014-0794-8

Taghiyari, H. R., Ghorbanali, M., and Tahir, P. M. D. (2014c). "Effects of the improvement in thermal conductivity coefficient by nano-wollastonite on physical and mechanical properties in medium-density fiberboard (MDF)," *BioResources* 9(3), 4138-4149.

Article submitted: June 23, 2018; Peer review completed: January 12, 2019; Revised version received: May 5, 2019; Accepted: May 6, 2019; Published: June 17, 2019.
DOI: 10.15376/biores.14.3.6168-6178