Upgrading and Evaluation of Waste Paper Sludge Ash in Eco-Lightweight Cement Composites

G. C. H. Doudart de la Grée[1]; Q. L. Yu[2]; and H. J. H. Brouwers[3]

Abstract: The upgrading and evaluation of waste paper sludge fly ash (PsFA) as a cement substitute in wood-wool composite boards (WWCB) is investigated in the present study. This type of board is widely applied in the building environment because of its excellent thermal and acoustical properties; however, cement is still widely used as a binder although environmentally its use leads to a high CO_2 footprint of the composite and is therefore less favorable for sustainable building designs. Paper sludge fly ash could be a feasible replacement of cement; however, its high water demand limits the substitution level to 10% by mass of binder. Therefore, different upgrading methods including water treatment and thermal treatment are performed, and their effects are evaluated using various analytical methods. Afterward, PsFA's suitability as a binder replacer in WWCB is investigated with the hypothesis to refine the microstructure of the boards, resulting in extra lightweight properties. A substitution level of binder with the upgraded PsFA by 20–60% in WWCB can be achieved and is confirmed by the improved mechanical, thermal-insulating, and sound-absorbing properties. **DOI: 10.1061/(ASCE)MT.1943-5533.0002186.** *© 2018 American Society of Civil Engineers.*

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Introduction

Paper sludge is a residue from the paper recycling process consisting of very short and brittle fibers, inorganic materials, and ink that cannot be further used for proper paper production. The paper sludge residue from the paper recycling process consists of 50% water, 25% cellulose fibers to strengthen the paper, and 25% minerals such as calcium oxide and kaolin clay to make the paper smooth. A number of studies have been performed in order to reuse the paper sludge in clay bricks (Cusidó et al. 2015) or other industrial applications (Frías et al. 2015). Still, a large proportion of paper sludge is directly disposed of at landfill sites. A more favorable processing route is the incineration of paper sludge, which reduces the waste volume and in the meantime produces energy in special power plants or bioenergy power plants (Doudart de la Grée et al. 2016). Moreover, the produced paper sludge fly ash (PsFA) can be applied as a substitute for soil stabilization under roads and as a supplementary cementitious material. The suitability depends on the mineralogical composition of the PsFA and the applied combustion temperature. Often on an industrial scale, temperatures between 850 and 1,200°C are used, leading to a material with hydraulic properties (Ferrándiz-Mas et al. 2014). Fava et al. (2011) observed a slight increase in compressive strength in mortar when 5 and 10% by mass of cement was replaced with PsFA. Research has shown that calcining paper sludge at 700–750°C produces a reactive pozzolanic PsFA by forming metakaolin (Bai et al. 2003; Banfill and Frias 2007; Giménez et al. 2015; Pera and Amrouz 1998; Segui et al. 2012). García et al. (2008) reported an increase in the early strength of mortars produced by replacing 10% by mass of cement. Characteristics of the PsFA such as the pozzolanic activity and resulting products are well known (Frías et al. 2008, 2013; Goñi et al. 2012; Vigil de la Villa et al. 2007); however, the origin of the high water demand (Goñi et al. 2012; Wong et al. 2015; Wu et al. 2016) is studied less extensively, and it seems that it is caused by the high microporous structure, which increases the surface area, and speculated high free lime content (Bai et al. 2003; Mozaffari et al. 2009). This reduces the mechanical properties of mortars at higher replacement levels by problematic expansion (Bai et al. 2003; Mozaffari et al. 2009).

Nevertheless, the superior characteristics of PsFA concerning its pozzolanic reactivity, low density compared to cement (e.g., CEM I 52.5 R white), and the negative CO_2 footprint [i.e., lifecycle analyses presenting a global warming equivalent of $-120\ \mathrm{kg\,CO_2/t}$ (Ewijk 2008)] make PsFA a potentially promising material. The PsFA characteristics make it of interest to use as a binder substitution to design a lightweight wood-wool composite board (WWCB) with a refined microporous structure. Meanwhile, wood is available worldwide in large quantities, and its lightweight porous structure makes it of value as a filler, providing thermal and sound-absorbing properties (Ashori et al. 2011; Badejo 1988; Jorge et al. 2004; Lee 1985). The binding agent of cement mineralizes the wood, adding high resistance to biodegradation (Pereira et al. 2006) and fire-resistant properties (Doudart de la Grée et al. 2014; Simatupang and Geimer 1990; Wei et al. 2000). The production process of commercial WWCB involves first the mixing of wood-wool and cement, then a pressing procedure for 24 h, followed by a curing period of 10 days, and finally, the removal of the remaining free water (Doudart de la Grée et al. 2014). Therefore, applying a binder with a fast early strength development property is desirable, i.e., the main strength development needs to be achieved within 24 h of pressing; otherwise, the boards will be

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[1] Ph.D. Candidate, Dept. of the Built Environment, Eindhoven Univ. of Technology, 5623 CB, Eindhoven, Netherlands.

[2] Assistant Professor, Dept. of the Built Environment, Eindhoven Univ. of Technology, 5623 CB, Eindhoven, Netherlands (corresponding author). ORCID: https://orcid.org/0000-0001-8578-3945. E-mail: q.yu@bwk.tue.nl

[3] Professor, Dept. of the Built Environment, Eindhoven Univ. of Technology, 5623 CB, Eindhoven, Netherlands.

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dimensionally unstable and will gain insufficient mechanical properties. Although WWCBs have been produced since 1920 (Elten 2006) and used in the building environment for multiple applications such as roofing of garage boxes, acoustic ceilings in nonresidential buildings, and in residential housing as complete wall elements, the used binder still primarily consists of portland cement leading to a high CO_2 footprint. Because of the previously mentioned requirements, replacement of cement is not straightforward. Moreover, under the alkaline condition of cement slurry, soluble extractives from the wood, which withdraw water from the cement slurry and interact with Ca^+ ions released from cement (Doudart de la Grée et al. 2017), will be leached out, resulting in the retardation of cement hydration.

Hypothetically, PsFA, white in color, as an active filler for substitution of cement in WWCB, is suitable (1) because the low density and porous nature of PsFA can be beneficial to increase the thermal insulation and sound-absorption properties; (2) for aesthetic and environmental reasons, because often white portland cement is applied to obtain a nice appearance but nevertheless has a high carbon footprint; and (3) because fast early strength development is achievable and is attributed to the presence of metakaolin and portlandite (Doudart de la Grée 2012; Joshi and Lohtia 1997; Subas and Emirog 2015).

A preliminary study revealed that the substitution of cement in WWCB by >10% by weight of PsFA leads to insufficient hardening of the binder caused by the high water demand of both the PsFA and the wood-wool. Therefore, in this study, two treatment pathways are evaluated to increase the usability of industrially calcined PsFA by reducing its water demand. The first treatment consists of prewetting the PsFA with water vapor to turn the free lime into portlandite. Moreover, a further decrease in water absorption is expected because of the modification of the surface area and micropore structure by the treatment. The second treatment method is a thermal treatment, to turn all the calcium into free lime and then prewet it with water vapor. The PsFA, both treated and untreated, is physically and chemically characterized. By monitoring the heat release, the reactivity of the binder is examined. Furthermore, the efficiency of these treatment methods is assessed, and the suitability of PsFA as a cement substitution is then evaluated in wood-wool composites, considering the environmental footprint and main properties such as thermal insulation and sound absorption.

Materials and Methods

Materials

The PsFA used in this study is provided by Van Gansewinkel Minerals (Netherlands), produced by calcining paper sludge at 780°C [International Patent No. WO96/06057 (1996)]. The CEM I 52.5 R white (C52.5w) portland cement used in this study was provided by ENCI (Maastricht, Netherlands). For the production of the wood-wool composite boards, wood-wool (also known as excelsior) made from spruce wood is used with a width of 1.5 mm and varying thicknesses of 0.24–0.35 mm and a length of 250 mm having an initial moisture content of 9%. Because recent developments regarding WWCB have shown positive results when implementing 36% of commercially available limestone powder (LP) as a cement replacement, this combination is used as the reference binder in this study, considering further sustainability.

Upgrading of PsFA

The treatment of PsFA resulted in the following samples: (1) prewetted, termed PsFA-w; (2) thermally treated at 750°C, termed PSFA-h; and (3) the thermally treated sample and then prewetted, termed PsFA h/w (Doudart de la Grée 2012). The amount of water to initiate the chemical upgrading of PsFA is determined based on the conversion of free lime to portlandite.

This water dosage would be a minimum quantity because it only takes into account the transition of free lime to portlandite, although it is reasonable to expect that simultaneously other phases will also react. Furthermore, part of the water will be physically absorbed on the surface of the powder (Yu and Brouwers 2011). The water demand of powders is evaluated by using the Puntke test (Hunger and Brouwers 2009). The test is performed by adding water to 50 g of powder and then stirring. The water is continuously added, and the amount is measured until a shiny surface is obtained that indicates that the system has reached saturation. Using this method, the packing density of the binder can be determined as follows:

$$P = \frac{V_p}{V_p + V_w} = \frac{m_p}{m_w \times \frac{\rho_p}{\rho_w} + m_p} \tag{1}$$

where P = packing of a material in the range of 0–1, where 1 = solid perfectly packed and 0 = no packing; V_p = volume of the powder (cm^3); V_w = volume of the water (cm^3); m_w = mass of the applied water (g); m_p = mass of the measured powder (g); ρ_d = density of the powder (g/cm^3); and ρ_w = density of the water (g/cm^3). The packing of a material provides insight about the binder: a higher packing value leads to a denser matrix.

Characterization of the Powders

Physical Properties

The physical properties, such as the BET specific surface area, the specific densities, and the particle size distribution, are measured by N_2 adsorption using a Tristar II 3020 (Micrometrics), an AccuPyc 1340 II gas pycnometer (Micrometrics), and a Mastersizer 2000 (Malvern), respectively. The bulk density is determined by using the mass of the powder divided by the filled cup with a known volume.

Chemical Properties

The chemical composition of the powdered samples is analyzed with X-ray fluorescence (XRF) using an EDXRF Epsilon 3^X (PANalytical). In the case of PsFA, all calcium-based compounds are considered by XRF, although they differentiate in their chemical nature, such as free lime (CaO), portlandite [$Ca(OH)_2$], calcite ($CaCO_3$), and other amorphous Ca-containing phases. Thermogravimetry (TG) analysis (Netzsch STA 449 F1 Jupiter) is performed to determine the free lime amount. The original and treated samples are heated with 10°C/min up to 1,000°C in order to assess the content of $Ca(OH)_2$ and $CaCO_3$ from the mass loss caused by the removal of H_2O and CO_2, respectively. Three main mass loss intervals are expected: the loss of physical water under 120°C (Arlacon Ruiz et al. 2005; Castellote et al. 2004), the decomposition of $Ca(OH)_2$ between 350 and 600°C (Alonso and Fernandez 2004; Shui et al. 2008), and the decomposition of $CaCO_3$ between 600 and 900°C (Shui et al. 2008). Based on this, the effect of the treatment can be identified by observing the increase in $Ca(OH)_2$. X-ray diffraction analysis is performed using a Rigaku-Geigerflex spectrometer using Cu-Kα radiation and a 2θ angle between 15 and 55° with a step size of 0.02° and a 1-s count time to evaluate the effect of the treatments. The pattern processing was done using *EVA* software (Bruker) and the international center for diffraction data (ICDD) pattern database.

Table 1. Mix Proportions of the Tested Mixtures (W/B Is the Wood-to-Binder Ratio, and W/P Is the Water-to-Powder Ratio)

Sample	Composite recipes	C52.5w (%)	LP (%)	PsFA-w (%)	W/B	W/P
B1	0.64 C52.5w + 0.36 LP	64	36	0	0.75	1.0
B2	0.57 C52.5w + 0.33 LP + 0.1 PsFA-w	57	33	10	0.75	1.0
B3	0.51 C52.5w + 0.29 LP + 0.2 PsFA-w	51	29	20	0.75	1.0
B4	0.64 C52.5w + 0.36 PsFA-w	64	0	36	0.75	1.0
B5	0.4 C52.5w + 0.6 PsFA-w	40	0	60	0.75	1.0

Hydration Kinetics

The hydration kinetics of PsFA-w is examined by using an isothermal calorimeter (TAM Air, TA Instruments). The tests were performed for 70 h with a water-to-powder ratio of 0.5 under a fixed temperature of 20°C. The powder samples were first transferred in an ampoule and then well mixed before water was added. After the addition of water, the mixture was mixed for 2 min, and the ampoule was loaded to the isothermal calorimeter. The mixtures consisting of 36% LP with 64% C52.5w (reference binder) were compared to the mixture consisting of 36% PsFA-w with 64% C52.5w to evaluate the nucleation and seeding effects that are reflected by an accelerated hydration rate.

Mechanical Properties

To investigate the suitability of PsFA as a cement replacer in wood-wool composites, boards are produced by applying the mixture PsFA-w and C52.5w according to the method proposed by Doudart de la Grée et al. (2017). The target density of the board is 450 kg/m^3 using a water-to-powder ratio by mass of 1.0 and a wood-to-powder ratio by mass of 0.75. The recipes are presented in Table 2. The substitution level of the reference binder (64% C52.5w + 36% LP termed B1) by 10 and 20% is evaluated, termed B2 and B3, respectively. Moreover, substitution of cement (no LP) by PsFA-w using 36 and 60% as an active filler is investigated, termed B4 and B5, respectively (Table 1).

First, the wood is prewetted with water, corresponding to 30% of the dry mass of wood-wool, to make the wood-wool more flexible and less vulnerable to breakage. Second, the binder is mixed with water for 1 min to create a homogenous slurry, which is then applied on the wood-wool and then mixed for approximately 30 s. Third, the wood-wool cement composite is transferred to a mold with a size of 30×50 cm^2 (W × L). Fourth, the mold is removed, and the composite is compressed to the desired height, which is determined by using distant holders of 15 mm for 24 h, followed by an ambient curing for 10 days. Finally, the boards are dried in an oven at 60°C for 22 h to remove the capillary water, and three samples of 15×20 cm^2 (W × L) are extracted. The apparent density of the boards was determined by the measured mass and bulk volume. The flexural strength was measured using a three-point flexural testing instrument in accordance with EN 12089 (CEN 1997).

Thermal Insulating Properties

Fly ash and biomass ashes have shown a positive contribution to lower thermal conductivity (Jafari and Jung 2016). The effect of the PsFA-w on the thermal properties of the WWCB was evaluated by determining the thermal conductivity and porosity of the produced boards. The thermal conductivity was measured using a commercial heat transfer analyzer (ISOMET 2104, Applied Precision, Bratislava, Slovakia); the porosity of the board was measured using an AccuPyc 1340 II gas pycnometer (Micrometrics); and the morphological properties were studied by scanning electron microscopy (SEM) analysis.

Sound-Absorbing Properties

Because sound absorption is one of the main features of wood-wool composite boards, the influence of PsFA on the sound-absorbing properties of the boards was investigated. The sound-absorption properties of a wood-wool composite using the reference binder with a thickness of 15 mm (B1) were measured and compared with a 15-mm sample by replacing the same recipe by 20% PsFA-w (B3). The sound absorption was calculated using a "slit pore model" by measuring the impedance using an impedance tube. Only 20% PsFA-w is studied because of the required sample size and limitations of the measurement setup. The composite with a very low density turns brittle and can be possibly broken by the cylindrical cutting force for preparing the sample with a size of 40 mm in diameter for the acoustical test. This data cannot be compared to the ISO 354:2003 (ISO 2003) reverberation room test because it does not take into account a diffuse field. Nevertheless, it provides a fair comparison between the sound absorption of the different mixtures, and it can be used as a primary indicator for the sound-absorbing properties of the boards.

Thermal Analysis

The mineralization of wood-wool by the binder leads to an improved fire resistance by reducing the combustibility of the wood-wool; however, the fire behavior of the composites is altered when different mineral compositions are applied. Therefore, TG tests were performed after the boards were cured and dried to estimate the rate of wood-wool combustion. The mass loss was measured using a heat rate of 10°C/min to a temperature of 620°C for wood at which cellulose, hemicellulose, and lignin will decompose (Drysdale 2011) and 1,000°C for the hydrated binders. The total

Table 2. Cumulative Volume; Specific Surface Areas; Specific Density (ρ_s) and Bulk Density (ρ_b); and the Mean Particle Size of the Powders Used in This Study

Material	Cumulative volume of pores (cm^3/g)	T-plot micropore area (m^2/g)	BET surface area (m^2/g)	ρ_s (kg/m^3)	ρ_b (kg/m^3)	D_{50} (μm)
PsFA	0.030	1.34	8.20	2,850	490	30
C52.5w	0.004	—	1.29	3,150	1,090	17
LP	0.005	0.28	1.24	2,700	1,000	10

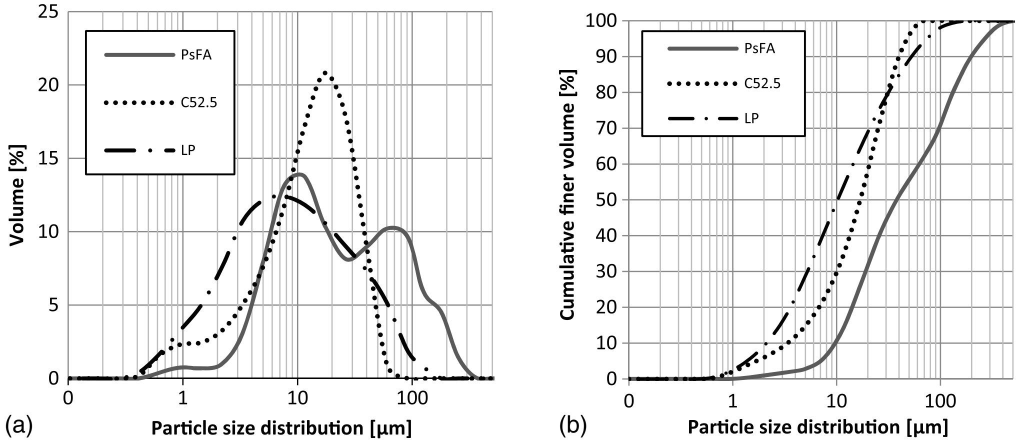

Fig. 1. Particle size distribution: (a) fraction volume; (b) cumulative volume

mass loss of the composites up to 620°C is attributed to the combusted wood-wool and dehydrated binder. Hence, the inherent percentage mass loss of binder was measured and normalized to the amount of binder present in the composite. This quantity was subtracted from the mass loss of WWCB, and the difference accounted for the mass loss of wood.

Results and Discussions

Characteristics of the Binders

Paper sludge fly ash has a BET surface area of 8.20 m^2/g (Table 2). The high surface area and the presence of a high amount of micropore area (1.34 m^2/g) and cumulative volume of pores (0.03 cm^3/g) testify to its porous microstructure. Fig. 1 presents the particle size distribution and cumulative volume of the materials, illustrating that PsFA is coarser compared to the other materials. The specific density (ρ_s) of PsFA is in the range of cement; however, the loosely packed bulk density (ρ_b) is very low—more than a factor of two lower (490 kg/m^3)—compared to that of cement (1,100 kg/m^3), confirming again the high porosity.

From Table 3, it is evident that PsFA has a high calcium content, resulting from the filler used for paper making. The moderate silica and aluminum contents are from the calcining of the kaolin clay used for paper making (Bai et al. 2003; Banfill and Frias 2007; Pera and Amrouz 1998; Segui et al. 2012).

As can be seen in Table 4, PsFA shows a higher water demand of 69% of the dry mass compared to 26.4% for C52.5w and 21.6% for LP. The packing fraction of PsFA because of the high water demand was significantly low, indicating that especially at higher replacement levels, the mechanical properties could be strongly affected because of the reduced densification of the matrix. Moreover, the temperature of PsFA started to increase after the addition of a certain amount of water, which is attributed to the fast hydration of free lime that would lead to thermal cracks and expansion (Bai et al. 2003; Mozaffari et al. 2009).

The mass losses to determine the stoichiometrically calculated amounts of $Ca(OH)_2$ and $CaCO_3$ are presented in Table 5. For PsFA, 2.47% of $Ca(OH)_2$, 37.18% of $CaCO_3$, and 26.30% of Ca-containing phases were found. For PsFA-h, a low amount of water (2.30%) was still present after the thermal treatment, representing the rehydration of the free lime, whereas almost all $CaCO_3$ was turned into CaO, and the remaining 0.39% was possibly resulted from the carbonation of $Ca(OH)_2$, both of which occurred during storage. Based on the remaining CaO amount of 26.30% for PsFA and 47.04% for PsFA-h, the minimum required water amount was estimated for the transition to portlandite, indicating that 8.4% and 15.1%, respectively, of water was needed, which would be valid if all CaO is free lime.

Applying 8.4% of water did not lead to any noticeable formation of portlandite, which can be explained by the high water demand of

Table 3. Oxide Composition Determined by XRF after Subtraction of the LOI

Chemical composition	PsFA (%)	C52.5w (%)	LP (%)
CaO	49.0	67.2	89.6
SiO_2	19.7	20.9	4.4
Al_2O_3	9.7	3.9	1.0
Fe_2O_3	2.0	0.5	1.6
MgO	2.4	0.4	1.0
Other oxides[a]	17.2	7.1	2.4

[a] Oxides lower than <0.5%.

Table 4. Packing of Materials Using the Puntke Test [Eq. (1)]

Material	Water demand (g)	Packing
PsFA	34.6	0.34
C52.5w	13.2	0.55
LP	10.8	0.63

Note: Water demand is measured for 50 g of material.

Table 5. Mass Loss of Original and Treated PsFA and Predicted Percentage of Ca-Containing Compounds

Temperature range and calcium compounds	PsFA (%)	PsFA-h (%)	PsFA-w (%)	PsFA h/w (%)
20–150°C	0.14	0.05	1.72	1.53
350–600°C	0.6	0.56	2.71	9.08
600–900°C	16.35	0.17	14.07	0.93
$Ca(OH)_2$	2.47	2.30	11.15	37.34
$CaCO_3$	37.18	0.39	32.00	2.12
CaO[a]	22.70	1.96	26.36	29.45
CaO[b]	26.30	47.04	22.64	19.55

[a] Percentage of CaO content from $Ca(OH)_2$ and $CaCO_3$.

[b] Percentage of CaO after subtraction of the CaO present in $Ca(OH)_2$ and $CaCO_3$ from the CaO obtained by XRF.

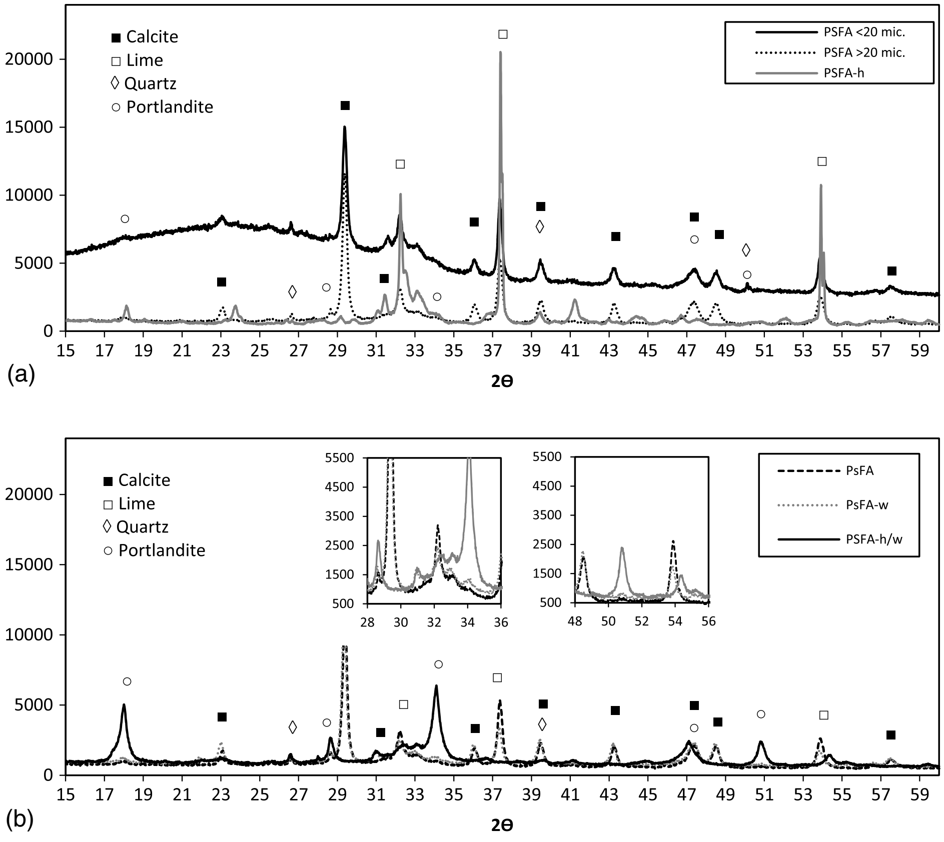

Fig. 2. Highlighting the crystal Ca-containing phases: calcite, lime, quartz, and portlandite for (a) XRD-pattern of original (<20 μm and >20 μm) and thermal PsFA; (b) original, water, and thermal water treated PsFA

the powder, as shown by the Puntke test. Hence, a water treatment was chosen based on the 15.1% of possible free lime when heating the sample and an addition 10% accounting for the part of the water that will be surface related. Therefore, a prewetting of 25% of the dry mass of powder for both PsFA and PsFA-h was further evaluated for portlandite formation using TG. In the case of the PsFA-w sample (Table 5), the treatment led to a $\mathrm{Ca(OH)_2}$ amount of 11.15%, which is 8.68% more compared to the untreated PsFA. In the case of PsFA h/w, the conversion of free lime with the same amount of water led to a $\mathrm{Ca(OH)_2}$ content of 37.34%, an increase of 34.88% compared to PsFA. Moreover, both results indicate that not all the CaO in PsFA-w and PsFA h/w is turned to portlandite, indicating the existence of free lime or other calcium-containing phases.

A preliminary XRD analysis shows that no amorphous compounds are detectable in PsFA, whereas metakaolin may exist in the fine particles in the range of 0–20 μm (Tafraoui et al. 2009). Therefore, for a more representative analysis, PsFA was separated to two fractions of (1) particles smaller than 20 μm, and (2) particles larger than 20 μm. The obtained XRD results are presented in Fig. 2. It can be seen that PsFA originally consisted of a high content of $\mathrm{CaCO_3}$ and some small peaks of CaO and $\mathrm{SiO_2}$. After the water treatment, the CaO was reduced, and the formation of $\mathrm{Ca(OH)_2}$ was detected.

After the thermal treatment, almost no $\mathrm{CaCO_3}$ or $\mathrm{Ca(OH)_2}$ could be detected in PsFA-h, and only CaO was identified. For PsFA h/w, the $\mathrm{CaCO_3}$ was turned into CaO and later into $\mathrm{Ca(OH)_2}$. However, not all lime was transformed into $\mathrm{Ca(OH)_2}$, which confirms the TG analysis. Moreover, the TG results show that compared to PsFA, the amount of $\mathrm{Ca(OH)_2}$ in PsFA-w was increased, and the XRD analysis indicates that an amount of free lime was still present after the treatment. Menéndez et al. (2012) studied the rehydration of CaO when $\mathrm{CaCO_3}$ decomposed at 650 and 1,000°C and concluded that at 650°C, 80% of CaO could rehydrate, whereas at 1,000°C, only 20–40% will rehydrate, which is in agreement with the present study. From the XRD results, no additional crystalline phases were observed after the treatments. Additional analysis of PsFA-w indicated that by the water treatment, the BET surface area was reduced to 6.91 $\mathrm{m^2/g}$ and the micropore area and pore volume to 0.91 $\mathrm{m^2/g}$ and 0.28 $\mathrm{cm^3/g}$, respectively, suggesting that a reaction of portlandite and other amorphous phases occurred that could influence the reactivity of the material.

The applied water vapor is perhaps unable to reach all the free lime in the porous microstructure of PsFA, and the heat release caused by the transition of free lime to portlandite contributes to the evaporation of part of the applied water. Nevertheless, prewetting the PsFA with the required water amount to turn free lime into portlandite, coupled with the reduced surface area, significantly reduces the water demand. Based on the obtained results, it is determined that only PsFA-w will be further evaluated because this binder type can be easily created in practice and is environmentally a better option compared to the thermal treatment.

Hydration Kinetics

The heat release results, shown in Fig. 3, present a fast reaction for C52.5w with a $T_{\max}$ at 20 h because of the second-stage reaction of $\mathrm{C_3A}$ and ettringite, forming monosulfate. Replacing C52.5w with 36% LP (binder used in B1) led to a faster reaction, showing a $T_{\max}$ at 11 h attributed to the nucleation effect of LP

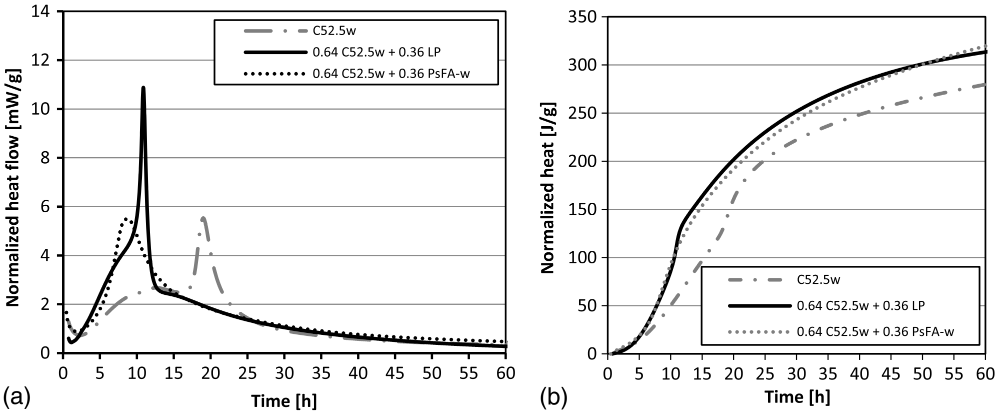

Fig. 3. (a) Normalized heat flow and (b) normalized heat of mixtures consist of C52.5w compared to 0.64 C52.5w + 0.36 PsFA-w and 0.64 C52.5w + 0.36 LP; mixtures are produced with a water-to-powder ratio of 0.5 and measured up to 60 h

(Bonavetti et al. 2001). Replacing C52.5w with PsFA-w (binder used in B4), a further acceleration is visible, with $T_{\max}$ occurring at 9 h, indicating that indeed the material leads to a fast early reaction, providing similar heat compared to the use of LP but having a coarser particle size. This is possibly due to the provided extra C-S-H seeding (Thomas et al. 2009), created after the water treatment as part of the water, which is also used to form gel or reaction of metakaolin.

Mechanical Properties

Fig. 4 displays the bending strength of WWCB versus the density manufactured with the composition as described in Table 1. In most of the replacement levels, the material behaves similarly compared to the reference sample B1, up to the replacement level of 20% (B3). At higher replacement levels, the binder matrix is found too weak because the presence of limestone powder insufficiently contributes to the gel formation, leading to a bending strength that could not fulfill the stated requirement of 1.7 MPa (J. Mencnarowski, personal communication, 2014), and therefore these results are excluded.

The effect of PsFA-w as an "active filler" is further confirmed by the bending strength of PsFA-w when replacing cement by 36% (B4) and 60% (B5), as presented in Fig. 4. Both samples gave higher strength than B1. Thus, the potential of using PsFA-w as a cement replacement is demonstrated for wood-cement composites.

This behavior can be explained by the treatment. First, the preadded water turned part of the free lime into $Ca(OH)_2$, which led to a reduction in the surface area and water demand of the PsFA-w. Therefore, when creating a slurry consisting of PsFA-w, cement, and water, more water is available for the cement hydration, whereas without treatment, a higher water amount would be needed, leading to an increased ion migration to the wood because of the hydroscopic nature of the wood-wool (Pereira et al. 2005, 2012; Wei et al. 2004). Second, by the reaction of metakaolin (Pavlík and Užáková 2016) present in PsFA-w (Frías et al. 2015), the binder reacts fast, as indicated by the isothermal calorimeter results. Simultaneously, C_3A in cement reacts with water and $Ca(OH)_2$ (the latter is provided by the reaction of cement or PsFA-w), forming calcium aluminate hydrates in the form of C_2AH_8 and C_4AH_8 that later turn into a more stable hydrate C_3AH_6 (Taylor 1990). Because of this conversion, water is removed, decreasing the porosity of the binder (cement with PsFA-w). Third, the low bulk volume of PsFA-w leads to a better surface coverage of the wood-wool [Fig. 5(a) compared to Fig. 5(b)] when applying the same mass amount as cement or limestone powder (Table 2). This increases the interlocking between the wood-wool strands by

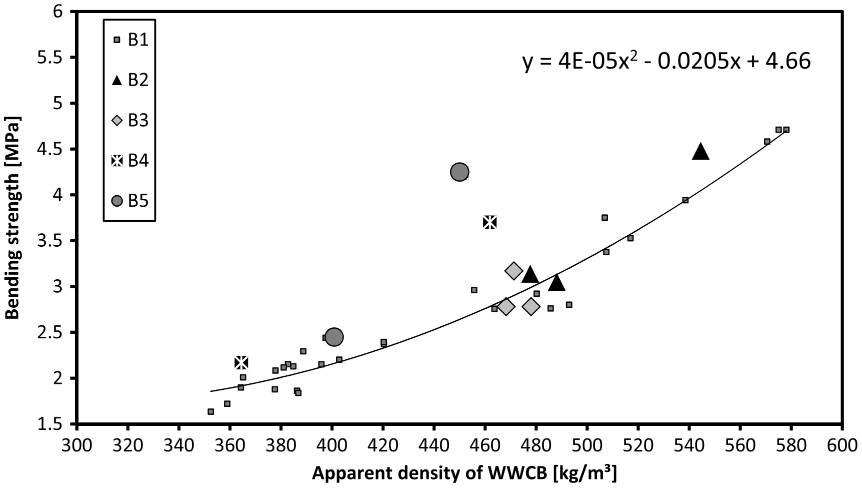

Fig. 4. Bending strength versus apparent density of boards produced with the treated PsFA as 10 and 20% replacement (B2 and B3) of the reference binder (B1); moreover, a mixture of 36% (B4) and 60% (B5) PsFA-w as cement replacement

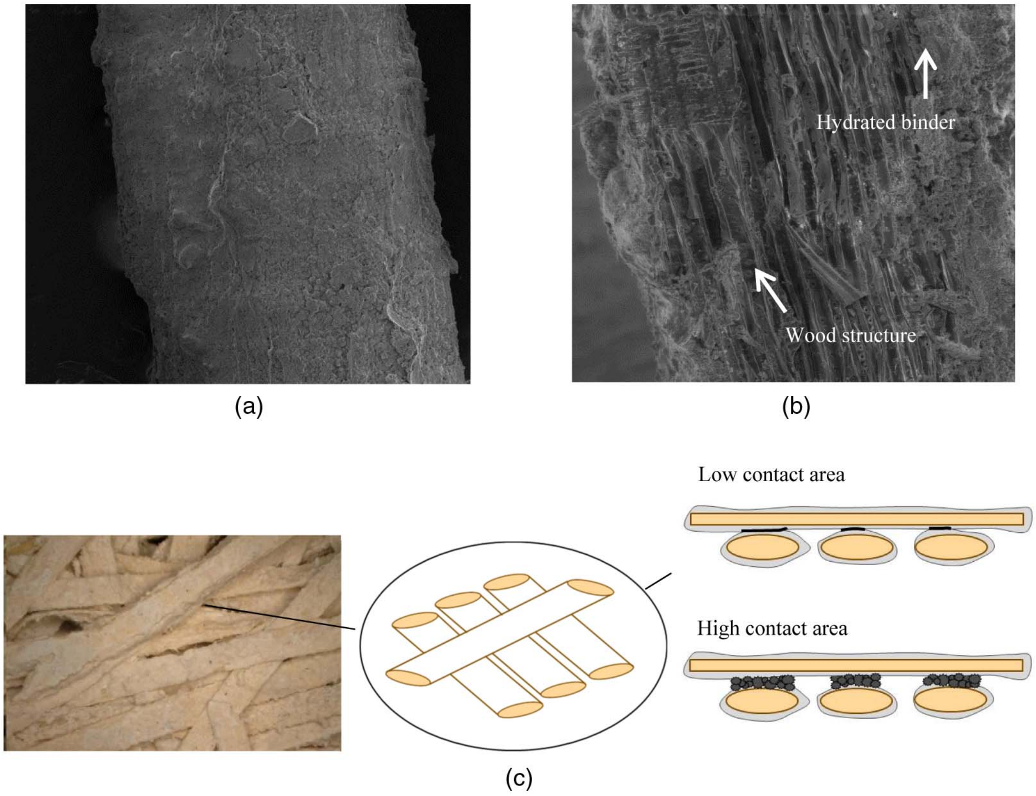

Fig. 5. (a) Well-distributed binder over the wood-wool surface; (b) poorly covered wood-wool surface with binder; (c) improved interlocking of wood-wool because of enhanced contact between binder and wood-wool

means of a higher contact area [Fig. 5(c)]. Furthermore, the high porosity of the boards would allow a small expansion caused by the high amount of PsFA, whereas in mortars, this might lead to reduced mechanical properties.

Finally, the concentration of calcium ions in cement is compensated by adding an additional source of calcium from PsFA-w. Although the interaction between Ca^+ ions and wood forming complexations still occurs, the inhibitory effect on the binder hydration is reduced. When the strength results of the boards in this study are compared with boards using only CEM I 52.5 white as binder (Doudart de la Grée et al. 2017), the boards made with PsFA in a density class of 450 kg/m^3 had a similar or higher bending strength depending on the replacement level.

Thermal Properties

The thermal conductivity results of the tested samples are summarized in Table 6. It can be seen that the thermal conductivity of the WWCB was decreased when increasing the replacement percentage of cement by PsFA-w. The board density was reduced, and the porosity was accordingly increased. The reduction in density can be explained by the microstructure of PsFA-w using SEM, as illustrated in Fig. 6. The matrix of B4 [Figs. 6(a and b)] consists of a large amount of needle-shaped particles, identified as ettringite, together with hexagonal plates of $Ca(OH)_2$ and gel-like structures. During the drying of the material in the oven at 60°C to remove the extra water, part of the chemically bound water in the ettringite was also released, increasing the mass loss and reducing the density of the boards. The microporous structure is clearly visible compared to a denser structure of B1 [Figs. 6(c and d)]. The low thermal conductivity improves the thermal insulation properties, allowing for the implementation of this type of composite in large wall structures, which have been recently developed for low-cost housing.

Acoustical Properties

The influence of 20% PsFA-w replacement of the reference binder (B3) on the sound-absorbing properties of the WWCB is presented in Fig. 7 in the frequency range 125–2,000 Hz. Two different kinds of porosities can be defined, namely (1) the measured "open porosity" by using, for instance, a gas pycnometer, and (2) the "acoustically effective porosity" that is defined as the porosity of a material that takes part in the acoustical process. It was found that increasing the bulk density of boards leads to a decrease in the open

Table 6. Measured Physical Properties of Different Composites

Sample	Composite	Thermal conductivity $[W/(m\cdot K)]$	Porosity (%)	Density (kg/m^3)
B1	0.64 C52.5w + 0.36 LP	0.085 ± 0.005	76.7	490
B2	0.57 C52.5w + 0.33 LP + 0.1 PsFA-w	0.083 ± 0.002	79.5	389
B3	0.51 C52.5w + 0.29 LP + 0.2 PsFA-w	0.075 ± 0.001	77.6	440
B4	0.64 C52.5w + 0.36 PsFA-w	0.073 ± 0.002	76.5	448
B5	0.4 C52.5w + 0.6 PsFA-w	0.070 ± 0.002	79.1	400

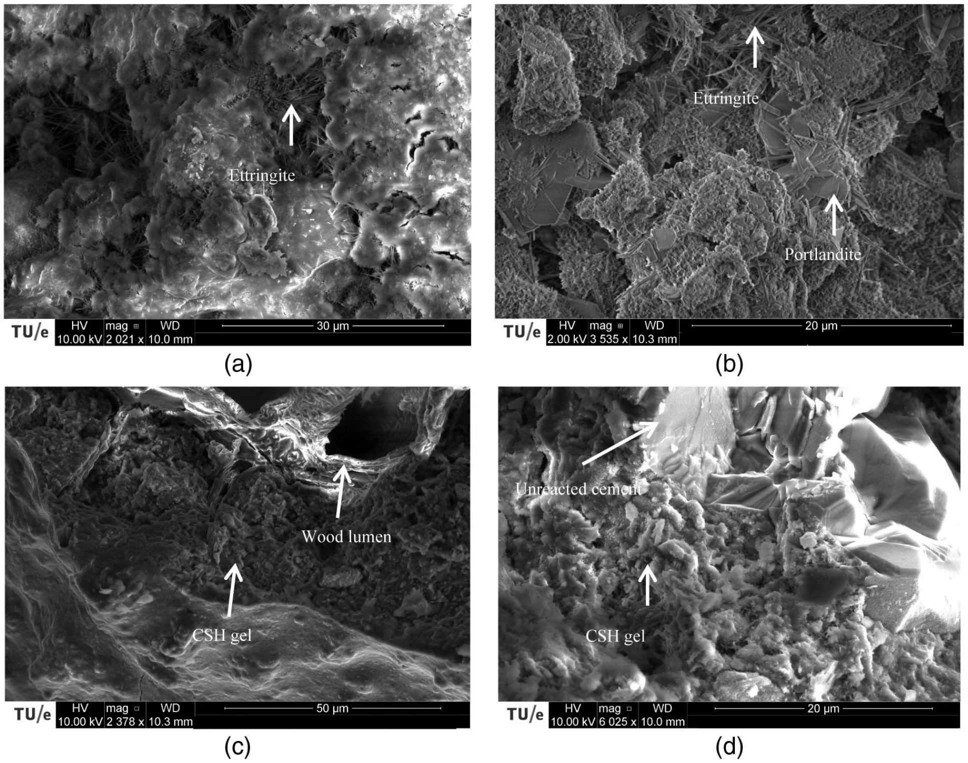

Fig. 6. SEM pictures of the microstructure of (a and b) B4 and (c and d) B1 around wood-wool

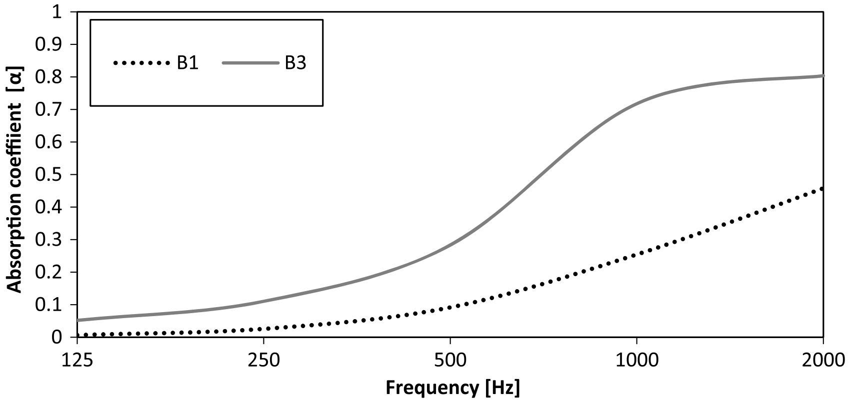

Fig. 7. Sound absorption of 15-mm-thick boards using a slit pore model in which the binder consists of 0.64 C52.5W + 0.36 LP (B1) as reference sample and the binder replaced with 20% PsFA-w (B3)

porosity but an increase in the acoustically effective porosity (Botterman et al. 2018). This can be explained by the following: (1) not all the voids in the wood binder matrix are involved in the acoustical process (Glé et al. 2012; Wassilieff 1996), and (2) not all the voids between the binder-covered wood-wool strands are involved in the acoustical process (Fuchs 2013).

Although the density of the boards using 20% PsFA was reduced, the low bulk density of PsFA led to a similar effect as that of a higher-density board, namely the reduction of the voids between the wood-wool strands by the high volume of the porous binder.

Without PsFA, the voids between the wood-wool strands are not efficiently taking part in the acoustical process. Hence, by using PsFA-w, refinement of the microstructure of the board is achieved because of its low bulk density, consequently resulting in an improvement in the sound absorption of the board—two times better compared to the reference sample.

Performance at High Temperature

Fig. 8 indicates the mass loss of a commercial WWCB (B1), wood-wool, and boards made of 40% PsFA-w as cement replacement (B4), preheated at 100°C and then heated to 1,000°C. The wood-wool was only investigated until 620°C because between 200 and 260°C, hemicellulose is removed, and between 240 and 350°C, cellulose is removed, and lignin starts to decompose between 280 and 500°C (Drysdale 2011). Before 120°C, wood-wool lost approximately 9% of its mass because of the evaporation of water.

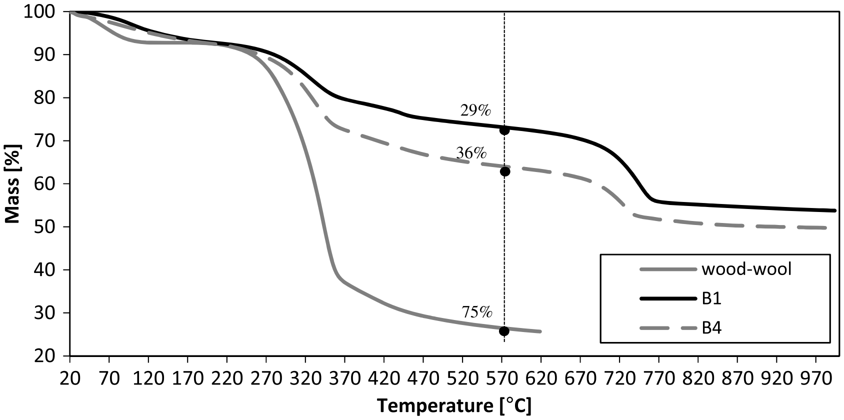

Fig. 8. TG curves of wood-wool, reference board (B1), and 40% C52.5w replaced by PsFA-w (B4)

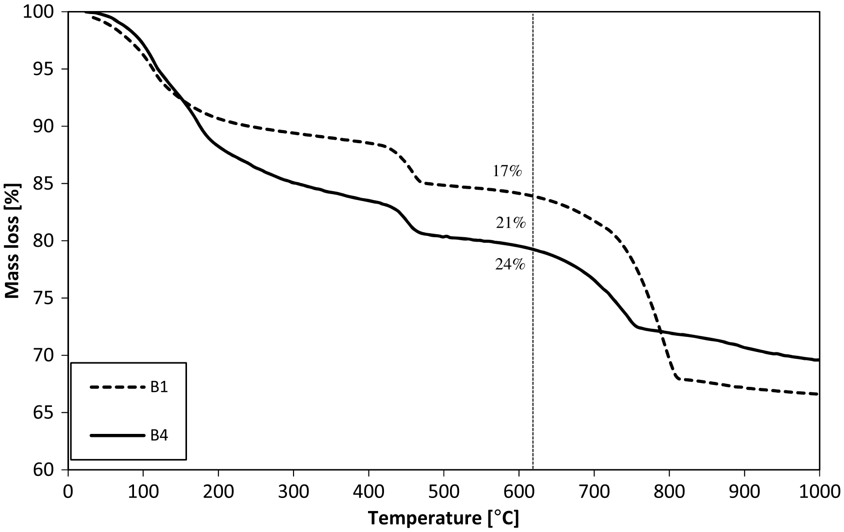

Fig. 9. TG curves of reference (B1) and 60% C52.5 + 40%PsFA-w (B4)

At 620°C, the majority of the wood is combusted, and only 25% of the initial mass remains. The wood-wool composites B1 and B4 lost at 600°C 29 and 36%, respectively, of their mass. Fig. 9 presents the mass loss of the hydrated binders without wood, indicating a loss of 21% when using B4 and 17% when using B1. It is found that 41.2 and 55.3% of the wood-wool was combusted for B1 and B4, respectively, which is lower compared to the 75% mass loss without using any binder. Nevertheless, the fire resistance of a material is not only indicated by its mass loss. As is the case with gypsum boards (Yu and Brouwers 2012a, b), chemically bound water present in PsFA-w is released during heating as moisture, effectively retarding the heat transfer by a so-called gas retention phenomenon (Grossel 2003; Thomas et al. 2009). In densely packed materials (e.g., concrete), the steam causes thermal spalling because the vapors cause expansion of the matrix and increase the pressure in the voids. However, the binder matrix of WWCB has a high pressure capacity because of its high porosity by the incorporation of PsFA-w, and hence, vaporization of the moisture has a lesser effect concerning the thermal spalling. Furthermore, the formed steam migrates toward the colder regions and becomes reabsorbed in the neighboring layer pores. Gradually this leads to a saturated layer hindering the gas flow (moisture clogging) by forming a front between the dry and saturated layers (Chen et al. 2009; Vickers et al. 2015). Moreover, even after complete dehydration, the present calcium carbonate will start to decompose, which requires certain energy, and the binder continues to act as a heat-insulating barrier because of its high thermal mass.

Conclusions

In the present paper, the upgrading and application of waste PsFA as a cement substitute in lightweight wood-wool composite board is investigated. Paper sludge fly ash is physically and chemically characterized, and different treatments, including a water and a combined thermal water treatment, are studied. This resulted in the following samples: prewetted (PsFA-w); thermally treated at 750°C (PSFA-h); and the thermally treated sample and then prewetted (PsFA h/w) to reduce its free lime content and porosity. The influence of PsFA-w on the main board properties, such as mechanical strength, thermal insulation, sound absorption, and resistance to heat, is examined. Based on the obtained results, the following conclusions can be drawn:

- The high surface area (8.2 $\mathrm{m^2/g}$), internal pore structure (1.34 $\mathrm{m^2/g}$), and free lime content lead to a high water demand of PsFA. The most successful and environmentally feasible

treatment route is to treat PsFA by applying a water amount of 25% of the dry mass of PsFA. This resulted in a reduction of the free lime content by an increased $CaOH_2$ content (8.7%), final surface area of 6.9 m^2/g, and internal pore structure of 0.91 m^2/g.
- Replacement of the reference binder used for WWCB by 20% leads to a similar flexural strength of 3 MPa with an apparent density of 470 kg/m^3. When applying 60% PsFA-w as cement replacement, superior flexural strength is obtained (4.3 MPa with an apparent density of 450 kg/m^3).
- Replacement of the reference binder used for WWCB by 20% reduces the board density with 50 kg/m^3 and lowers the thermal conductivity up to 11.8%. When applying 60% PsFA-w as cement replacement, a low density board of 400 kg/m^3 with the reduced thermal conductivity up to 17.6% is achieved.
- Replacement of the reference binder used for WWCB by 20% PsFA-w leads to an increased sound absorption by a factor of two by increasing the tortuosity and flow resistivity of the boards.
- Prewetted PsFA provides a very good thermal profile and potential protection against the ignition of wood-wool by release of chemically bound water.

Future research will be focused on the synergetic effect of the treated PsFA and secondary cementitious materials (e.g., ground granulated blast furnace slag and fly ash) on cement hydration and the performance of lightweight cement composites in order to acquire a further-reduced CO_2 footprint, and in the meantime, improved properties.

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