

**WOOD-WOOL CEMENT BOARD: UTILIZATION OF A POROUS BINDER**

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**ABSTRACT**

Ordinary Portland cement is currently widely applied in wood-wool cement boards as binder. In this study, the effect of replacing cement with an industrial by-product as alternative binder (termed PS) is investigated. The benefit of utilizing PS is its high reactivity by the presence of metakaolin that provides earlier strength, its low density compared to cement and a lower CO<sub>2</sub> footprint. It is found that a treatment is required to address the high free lime content that increases the water demand and the porosity of the binder. After the treatment, the potential of the PS as reactive binder was significantly increased and could be used as cement replacement in WWCB. Furthermore, the influence of utilizing PS on the mechanical, thermal, acoustical, and fire resistant properties of the newly developed WWCB is studied.

**KEYWORDS**

WWCB; cement; fly ash; inorganic; coating

**INTRODUCTION**

Mineral-bound wood-wool composites were already produced in the early 1900's, created from spruce or poplar wood-wool, using magnesite as a binder (Aro, 2004; Wolfe, 1999). Around 1920, after the introduction of Ordinary Portland Cement (OPC), cement replaced the magnesite as a binder leading to the creation of wood-wool cement board (WWCB) (van Elten, 2006). The boards became increasingly popular because of their high thermal insulating and sound absorbing properties gained by the high porosity and low density (400-600 kg/m<sup>3</sup>). Consequently, because of the mineralization of the fibres, the boards possess high resistance to bio-degradation (Pereira et al., 2006) and fire (Aro, 2004). Hence, the boards are applied in both buildings and constructions as roof and ceiling material or as an exterior wall where a high durability and low maintenance is requested. (However, there are certain disadvantages of these products that limit their suitability in the market.) The main drawback is that wood and cement do not work well together due to the wood extractives (mainly sugars) that inhibit the cement setting and result in boards with poor mechanical properties. Furthermore, at present, WWCB is still produced, but the question remains if this product still remains competitive in the current global market where both sustainability and durability gains popularity. Therefore, it would be more favourable to lower the environmental footprint of a WWCB. The environmental footprint is derived from: firstly, cement production, which has a high CO<sub>2</sub> footprint (global warming equivalence is 971 kg CO<sub>2</sub>/ ton cement, data originates from life cycle analyses) and therefore contrasts to the increased concern about global warming, air, water, and soil pollution. Secondly, large amounts of wastes generated from agricultural sources, forestry industry or construction, and demolition waste wood, that are either used as a fuel for newly developed bio-power-plant

installations (replacing coal-combustion power plants) or go to landfill sites. At landfill sites, the decomposition of wood results in the release of methane which is a greenhouse gas and is 72 times worse than CO<sub>2</sub> in a timeframe of 20 years (Leliveld & Crutzen, 1992). In general, the above methods of waste disposal are less favoured regarding the waste hierarchy and can be considered as the waste of a primary resource (Madurwar et al., 2013; Karade, 2010).

The need for and the benefits from an optimized mineral bound- wood cementitious composites have already been studied over the past 60 years, by e.g. investigating the interactions between wood and cement, and introducing different kinds of wood/plant fibres as substitutes to the usually applied spruce wood (Aggarwal et al., 2008; Aro, 2004; Ashori et al., 2012; Hachmi et al., 1990; Leliveld & Crutzen, 1992; Pereira et al., 2006; Simatupang et al., 1987; Wei et al., 2003). Nevertheless, there is a lack of implementation of scientific approaches in the industry because of practical problems or lack of acceptance, which will be discussed later. Additionally, in most studies, wood is mainly used for fibreboards fabrication or as a reinforcement agent in concrete, where different parameters apply compared to WWCB.

This paper focuses on lowering the amount of OPC in WWCB. This is done by applying an industrial by-product as partial replacement of cement and investigation of its influence on the mechanical, thermal, sound absorbing and fire resistant properties. The boards produced in this study are prepared according to the EN 13186 and fulfil the CL3 (chloride content  $\leq 0.06\%$  by mass of composite). In other words, no chlorides are used to accelerate the cement hydration. This is due to the application of the WWCB in ceilings e.g. close to reinforced concrete, where possible ion-migration could lead to corrosion of the reinforcement.

## **INDUSTRIAL BY-PRODUCT FROM THE PAPER RECYCLING PROCESS**

Paper sludge is a residue from the paper recycling process. In the past it was disposed by landfill or in incineration plants as it was considered to consist of unusable ingredients. Nowadays, through an adjusted burning process with low emissions, an industrial by-product (termed PS) with good pozzolanic properties and without any hazardous contaminants are created. Within the Netherlands, the utilization of industrial by-products is desired because of the increasing landfill costs. The reuse of this material (PS) in concrete is limited because of the increased water demand and content of free lime, which can reduce the properties of concrete. However, the benefit of PS is its high reactivity due to the presence of metakaolin that provides earlier strength, low density compared to cement, and the negative CO<sub>2</sub> footprint, i.e. life-cycle analyses present a global warming equivalent of -120 kg CO<sub>2</sub>/ton cement.

## **INHIBITORY EFFECT OF WOOD**

Before applying a new type of binder, the inhibitory effect of wood on the binding ability needs to be evaluated and taken into account. Currently, cement is the most widely applied binder in wood cement composites. Therefore, most of the studies are related to the retarding effect of wood components on the cement hydration. When wood is in contact with water, it will start to absorb the water, mostly by the hydrophilic hemicellulose. Afterwards, cement is added and different mechanisms initialize:

- Firstly, the hemicellulose and other inhibitory substances such as starches, sugars, phenols and hydroxylated carboxylic acids – that are soluble – start to dissolve and leach out from the wood fiber. This is even accelerated by the rapidly increasing alkalinity (caused by the added cement) of the environment that promotes the formation, dissolution and diffusion of this inhibitory substances;
- Secondly, from the start of migration of inhibitory substances, the alkaline hydrolysis of hemicellulose and other substances result in the formation of calcium salts of lignin, polysaccharides and sugars (Aggarwal & Singh, 1990), which will interfere with the cement hydration forming different hydration products and alter crystalline structures;
- Thirdly, because of the absorption phenomena, potassium ( $K^+$ ) and calcium ( $Ca^{2+}$ ) could be absorbed to some extent by lignocellulosic particles. Thus, calcium, which is an essential part of the hydration products to promote the cement hardening, is removed from the slurry, reducing the availability for cement hardening reactions and causing their impairment to some degree;
- Fourthly, lignocellulosic particles can absorb water that otherwise could be used for binder hydration. Hence, they reduce the amount of water used for binder curing (Pereira et al., 2006; Wei et al., 2004).
- Fifthly, Govin et al. (2006) found that the formation of  $Ca(OH)_2$  is reduced after the first 4 hours and strongly inhibited beyond 48 hours of hydration by natural poplar, while an increasing amount of calcium carbonates was found. This inhibition could be linked to the absorption of water by wood particles which leads to a decrease in the water-cement (w/c) ratio. However, a w/c ratio of 0.33 promoted the  $Ca(OH)_2$  precipitation below 16 hours of hydration. Therefore, the inhibition of  $Ca(OH)_2$  formation is not due to the decrease in w/c ratio, but more likely due to the alkaline degradation of wood that could induce a carbon dioxide release. This carbon dioxide leads to the carbonation of  $Ca(OH)_2$ . Consequently, the amount of  $CaCO_3$  in the paste increases.

According to Hachmi et al. (1990), the inhibitory effect of wood also depends on the  $pK_a$  of the extractives. Extractives with lower  $pK_a$  may inhibit the cement hydration much less than those with high  $pK_a$ . This is probably due to the lack of complex reactions of calcium ions due to salt formation with acidic extractives and, therefore, it has a lower influence on the calcium concentration required for cement hydration.

Studies have shown that both cellulose and lignin have a small influence on the cement hydration (Wei et al., 2000; Pourchez et al., 2006).

As mentioned before, calcium ion migration occurs, leading to the reduction of the available calcium for cement hydration. However, when PS is applied, this influence could be overcome and results in an increased amount of cement hydration. Furthermore, the fast reaction of metakaolin could be used as an accelerator to reduce the inhibitory effect of wood extractives.

## **CHARACTERIZATION**

The chemical properties of PS in comparison with the used CEM I 52.5 R are presented in Table 1 and the physical properties such as the density and BET specific surface area are listed in Table 2.

**Table 1 – Oxide composition determined by XRF.**

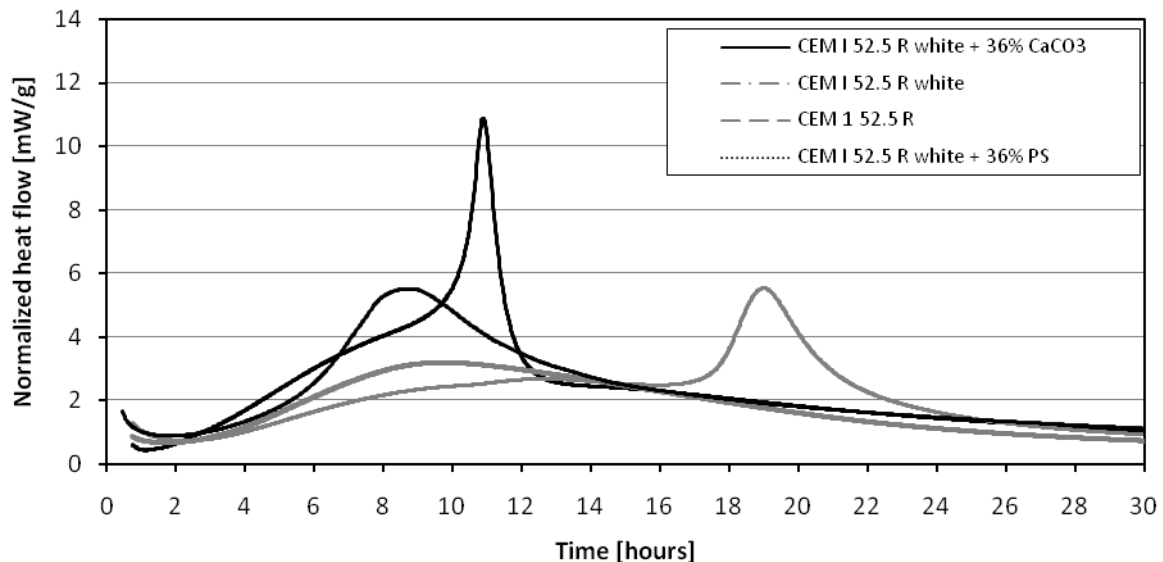
Chemical composition	PS[%]	CEM I 52.5 R [%]	CEM I 52.5 R white [%]
CaO	50	63.71	67.19
SiO <sub>2</sub>	19.7	20.27	20.86
Al <sub>2</sub> O <sub>3</sub>	9.7	4.80	3.91
Fe <sub>2</sub> O <sub>3</sub>	1.98	3.43	0.45
MgO	2.37	1.58	0.4

It is shown in Table 1 that PS has a similar elemental composition as that of CEM I 52.5 R. Only the CaO in PS is composed of 28% CaCO<sub>3</sub>, 2% Ca(OH)<sub>2</sub>, and 20% CaO (free lime) and the latter leads to an increased water demand. Furthermore, the Al<sub>2</sub>O<sub>3</sub> and SiO<sub>2</sub> are mainly present as metakaolin (23%), making the binder a fast reactant, which significantly contributes to the early strength development of the WWCB during the 24 h pressing. The reactivity of PS is confirmed by the isothermal calorimetry test. The mixture consists of 36.63% PS and 63.37% CEM I 52.5 R and is compared to the mixture consisting of 36.63% CaCO<sub>3</sub> and 63.37% CEM I 52.5 R, pure CEM I 52.5 R and CEM I 52.5 R white respectively. From the results presented in Figure 1, it can be seen that the material has a fast reaction with a T<sub>max</sub> at around 9 hours and with the main reaction within 24 hours.

**Table 2 – Specific surface areas measured by N<sub>2</sub> adsorption (using a Micrometrics Tristar II 3020) and specific density measured by an AccuPyc 1340 II gas Pycnometer.**

Material	T-plot external surface area [m <sup>2</sup> /g]	T-plot micropore area [m <sup>2</sup> /g]	BET surface area [m <sup>2</sup> /g]	Density [kg/m <sup>3</sup> ]
CEM I 52.5 R	1.3167	0.1854	1.5022	3150
CEM I 52.5 R white	1.2577	-	1.29	3150
Limestone powder	0.9621	0.2804	1.2425	2700
Spruce wood (internal and external)	0.0013*	-	0.69	750
Paper sludge fly ash	6.8614	1.3401	8.2015	2850

\* External surface area spruce wood is not measured but calculated



**Figure 1 – Normalized isothermal calorimetric curves of mixtures consist of 36.63% PS and 63.37% CEM I 52.5 R, 36.63% CaCO<sub>3</sub> and 63.37% CEM I 52.5 R white, pure CEM I 52.5 R and CEM I 52.5 R white respectively. Mixtures are produced with a water to powder ratio of 0.5 and measured up to 30 hours.**

The water demand of the powders is tested by using the “Puntke test”, in which water is added to a prescribed amount of powder and then stirred. The amount of water is continuously added and measured until a shiny surface is obtained that indicates that all grains are covered with a layer of water, and more water would only fill capillary pores. The packing of a material using the Puntke test can be calculated as:

$$P = 1000 / (1000 + D * (WD/M)) \quad [1]$$

where  $P$  is the packing of a material in the range of 0-1, with 1 is solid perfectly packed and 0 no packing,  $D$  is the density of the powder ( $\text{kg/m}^3$ ),  $WD$  is the water demand of the powder measured (g),  $M$  is the mass of the measured powder (g).

**Table 3 –Packing of materials using the Puntke test. Water demand is measured for 50 grams of material.**

Material	Water demand[g]	Packing
CEM I 52.5 R	13.2	0.55
CEM I 52.5 R white	14	0.53
63.37% CEM I and 36.63% limestone powder	11.8	0.59
Limestone powder (LP)	10.8	0.63
PS	34.6	0.34

To reduce the water demand of the applied PS powder a treatment is applied, by which the free lime content is reduced and turned into  $\text{Ca}(\text{OH})_2$ .

## BOARD PRODUCTION

In order to investigate the suitability of the PS as cement replacement in wood-wool cement boards, boards are produced applying the mixture of PS plus cement with a water to powder ratio by mass of 1.0 and a wood to powder ratio by mass of 0.75.

Firstly, the wood is pre-wetted, to make the wood-wool more flexible and less vulnerable to breaking. Secondly, the binder is mixed with water to create a homogenous slurry, which is then applied on the wood-wool. Next, the wood-wool cement composite is transferred to a mould to form the board. Subsequently, the mould is removed and the composite is compressed to the desired height. The height is determined by using distant holders of 15 mm. The composites are then compressed for 24 hours, sealed and afterwards stored for 10 days. Finally, after 10 days curing the boards are put into an oven at  $100^\circ\text{C}$  to remove the capillary water.

## RESULTS

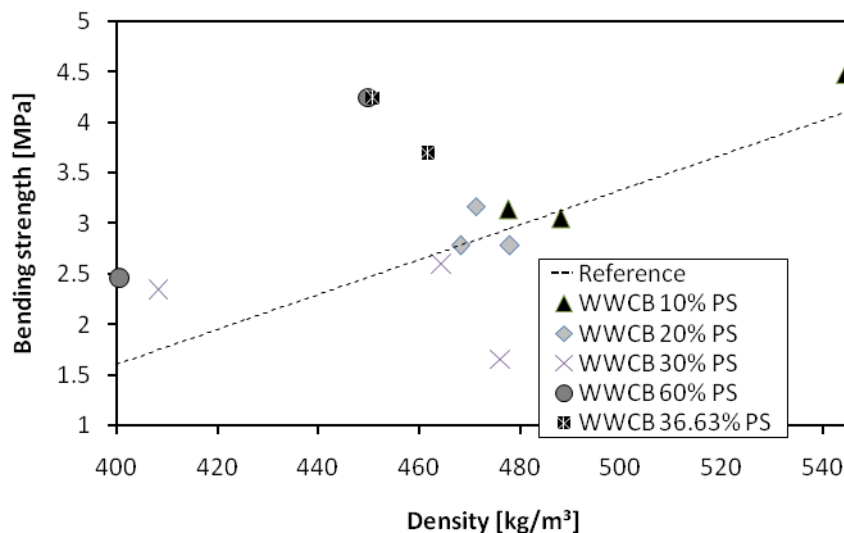
This study aims to create a board with similar mechanical properties of a commercial WWCB but with partial replacement of cement by PS. In an earlier study, the use of limestone powder as 36% cement replacement was successfully tested and the same tests were also successfully performed with the PS as a 36% cement replacement. As presented in Figure 1, a mixture with limestone powder accelerates the reaction of cement probably because it acts as a nucleation site. Therefore, the possibility of further reducing the cement content by first replacing the cement with 36% limestone powder, and then subsequently replacing this new mixture with PS. Levels of 10%, 20% and 30% of this combined composite of limestone powder and cement (from now on termed reference) is replaced and boards

are produced with the same water addition as in previously conducted studies. It was found that only the board with 10% replacement obtained certain bonding strength but higher levels did not have sufficient strengths (<1 MPa) due to a reduced bond, reduced hydration, and large swelling. The reason for this behaviour is related to the water demand of the PS. Firstly, the particle size of PS in combination with the high specific surface area requires a high amount of water. Secondly, insufficient amounts of water results in insufficient reaction of cement, meaning the produced  $\text{Ca(OH)}_2$  will not be sufficient enough to activate metakaolin. Thirdly, in PS there is a substantial amount of free lime available that requires water to produce  $\text{Ca(OH)}_2$ , which will be beneficial for the activation of the Metakaolin in the PS. Therefore, a pre-treatment was applied to decrease the free lime content and increase the  $\text{Ca(OH)}_2$  content.

**Table 4 – Tested composite mixtures up to 100% binder.**

Composite	Cement [%]	Limestone powder [%]	PS [%]
Reference sample	63.4	36.6	0
10% replacement of reference	57.1	33.0	10
20% replacement of reference	50.7	29.0	20
30% replacement of reference	44.4	25.6	30
36.63% PS	63.4	0	36.6%
60% PS	40%	0	60%

The tested replacement levels were again 10%, 20% and 30% replacement of the reference. Furthermore, an additional test was performed with cement together with 60% treated paper sludge fly ash (no limestone powder involved). These tests were necessary to reveal the difference between PS (as active filler) and limestone powder (as inert filler) using almost the same amount of cement as presented in Table 4.



**Figure 2 – The bending strength of boards produced with the treated PS as 10, 20, 30% reference replacement. Moreover, a mixture of 36.63% and 60% treated PS as CEM I 52.5 R replacement. Results are compared with the reference (36.63% limestone powder and 63.37% CEM I 52.5 R).**

Figure 2 shows that with most of the replacement levels the material behaves similar compared to the reference sample. Only the board with 30% replacement is observed to have lower strength. This can be explained by insufficient pressing which led to a board with a thickness of 17 mm instead of the

target value of 15 mm. This changes the microstructure of the board and therefore reduced the mechanical properties of the board.

The effect of PS as an “active filler” is clearly demonstrated when observing the bending strength results of the pre-treated PS when using both 36.63% and 60% cement replacement. Both the results give higher strength than with limestone powder as replacement. Thus the potential of using PS as cement replacement is clearly foreseen.

The reason for this behaviour can be explained by the treatment which ensures the later added water is applied in a sufficient way, in which high amounts of  $\text{Ca(OH)}_2$  are generated in the PS. When PS is used in a board, different positive effects occur. Firstly, both metakaolin and cement will react faster because of the additional  $\text{Ca(OH)}_2$  that also increases the pH. Secondly, the  $\text{Ca}^+$  ions are significantly compensated that are transported to the wood, and therefore would become unavailable for the cement hydration. This is because more  $\text{Ca}^+$  ions are available, generated by the PS. Thirdly, less water will be needed during the mixing reducing the water uptake by the wood. Fourthly, the increased temperature that normally occurs when water is in contact with free lime did already occur during the pre-treatment and, therefore, less internal heat will be produced when the board is pressed.

### Thermal conductivity

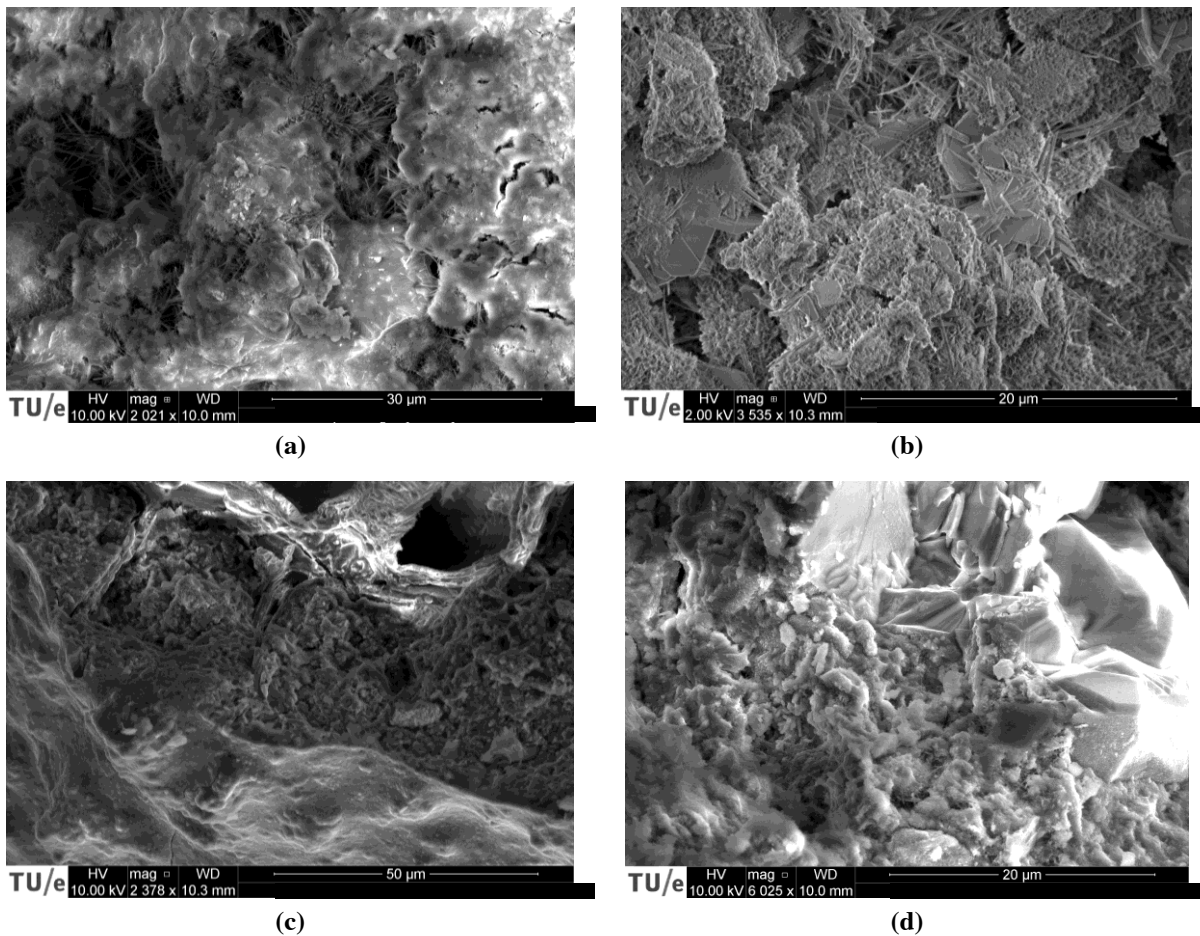
In order to investigate the influence of the new binder on the thermal properties of the new WWCB, the thermal conductivity of the boards was measured and compared with the reference board. The thermal conductivity was measured using a commercial heat transfer analyser (ISOMET 2104, Applied Precision). Furthermore, the porosity of the board was measured using aAccuPyc 1340 II gas Pycnometer and scanning electron microscopy(SEM) analysis was performed to explain the found results. The results of the tests are summarized in Table 5.

**Table 5 – The measured physical properties of different composites.**

Composite	Thermal conductivity [W/(m·K)]	Porosity [%]	Density [kg/m <sup>3</sup> ]
Reference sample	0.08-0.09	76.1-76.7	490-515
10% replacement of reference	0.080-0.085	-	-
20% replacement of reference	0.074-0.076	77.6	440
30% replacement of reference	0.072-0.076	-	-
36.63% PS	0.070-0.076	76.5	448
60% PS	0.066-0.073	79.1	400

From Table 5, it can be seen that the thermal conductivity of the WWCB is decreased when increasing the replacement percentage of cement by PS. The board density is also reduced and the porosity is accordingly increased. The reduction in density can be explained by observing the microstructure of PS using SEM as illustrated in Figure 3. The matrix consists of large amount of needle-shaped geometries, which is probably ettringite together with hexagonal plates of  $\text{Ca(OH)}_2$  and gel-like structures (probably C-S-H). Because the hydration products of PS differ in amount compared to that of cement, the mass of the hydration products differ resulting in a lower board density. Furthermore, during the drying of the material in the oven at 100 °C to remove the extra water, the chemically bonded water in ettringite is also released, increasing the mass loss and reducing the

density. From Figure 3, the pores in the microstructure is clearly visible, compared to a more dense structure of the cement matrix.



**Figure 3 – SEM pictures of (a) microstructure of the PS, (b) microstructure of a mixture of PS and CEM I 52.5 R composite (c) dense cement and limestone powder matrix around the wood-wool (d) gel structure of CEM I 52.5 R with limestone powder.**

## Sound absorption

The influence of PS on the sound absorbing properties of the boards and the level of porosity is investigated. It was found that the porosity of the board is not significantly changed as presented in Table 5. Figure 4 illustrates the promising sound absorbing properties, which can be attributed to due to the high specific surface area and quite low porosity of the hydrated binder. The sample 25 mm WWCB represents the measured sound absorption of a commercial WWCB according to ISO 354: 2003. The sound absorption values of 15 mm WWCB, 15 mm 20% PS and the 30 % PS untreated are calculated values using a “slit pore model” by measuring the impedance (-using an impedance tube). Although this data cannot be compared to the ISO 354: 2003 test it provides a good comparison between the sound absorption of the different mixtures and can be used as first indicator for the sound absorbing properties of the boards.



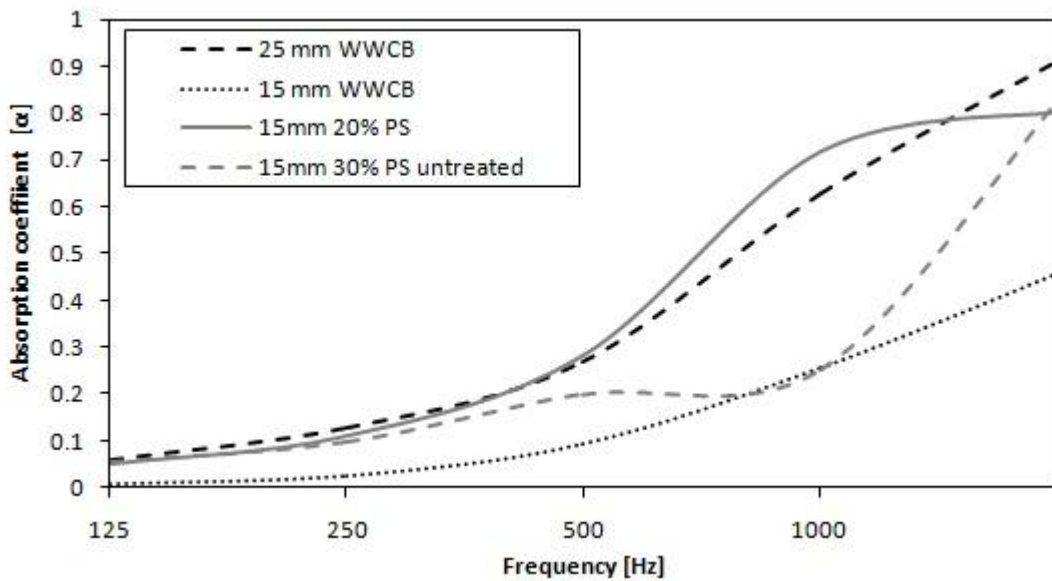


Figure 4 – Sound absorption of 25 mm height reference WWCB and calculated 15 mm height mixtures using a “slit pore model”. The calculated mixtures are the 15 mm height reference sample and mixtures with 20% treated PS as reference replacement and 30% untreated PS as CEM I 52.5 R replacement.

## Performance at high temperature

The fire behaviour of composites is altered when different mineral compositions are applied. Normally the binder will chemically decompose when heated. Therefore a thermal gravimetric (TG) test is performed to examine the mass loss during the heating after the boards are cured and dried. The mass loss is measured using a heat rate of 10 °C/min to a temperature of 400 °C using a STA 449 F1 Jupiter® thermal analyser. Although, these results cannot be directly compared with the results achieved by a real fire test, it indicates the behaviour and reactions that occur within the sample when exposed to a high temperature. The results are presented in Figure 5.

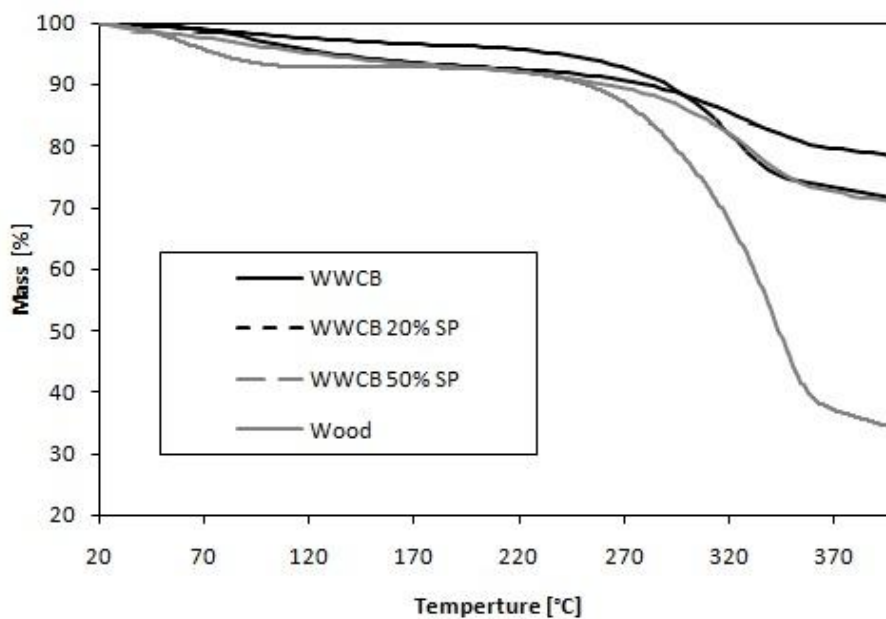


Figure 5 – TG curves of WWCB, wood and 20 and 50% CEM I 52.5 R replaced by PS.

The results show that wood-wool without any binder starts to ignite at around 270 °C and burns. At 400 °C only about 30% of the initial mass remains. With a composite made of cement and limestone powder (reference sample), the coating prevents the incineration of the wood and the mass loss at 400 °C is only 22%. The total mass of the wood is representing 43% of the WWCB. When assuming that the powder has a 5% mass loss due to the evaporation of water and dehydration of Ca(OH)<sub>2</sub>, the totally combusted wood can be calculated as to be only 42%, and the decomposition is significantly improved compared to a 70% combustion of unprotected wood. When using 20% and 50% PS as CEM I 52.5 R replacement, the fire resistance of the wood is also significantly improved, although beyond 320 °C the mass loss is strongly higher increase the reference sample. The reason for this can be related to a different thermal mass of the material but also due to the mass loss through the available water in ettringite.

## CONCLUSION

This paper introduces an industrial by-product (PS) that is tested to replace cement in wood-wool cement boards. The reason for choosing this binder is its low density, comparable chemical composition to cement, available metakaolin and white colour. Before the material could be used successfully, a treatment is required to reduce the free lime content. After the treatment, the material could be used successfully as a cement replacement at different levels. Substitution of different amounts of cement could be achieved up to 36% with improved mechanical properties. Furthermore, the performed experiments show a clear positive effect of PS on the thermal insulating and sound absorbing properties of the WWCB. In addition, the TG analysis of the samples indicates that PS provides a quite good thermal profile and potential protection against the ignition of wood-wool.

## ACKNOWLEDGEMENTS

The authors acknowledge the STW-foundation, Knauf Insulation, Eltomation, ENCI, and Dr. Jan van Dam (Wageningen UR) for their provision of material, knowledge and financial support.

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