



Influence of the spruce strands hygroscopic behaviour on the performances of wood-cement composites

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HIGHLIGHTS

- An absorption model for wood strands is proposed and validated by NMR.
- Different water to binder ratios are calculated for wood wool composites.
- The pre-soaking of strands is quantified for according to the water to binder ratios.
- Flexural strength achieves 3.8 MPa by applying a water to binder ratio 0.5.

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ABSTRACT

In wood wool cement composites, the influence of natural fibres is the main factor leading to the instability of the final product. During the manufacture, small variations in wood properties result in inhomogeneous density and strength of the boards. Among the accountable factors, the hygroscopic behaviour of the wood can deeply affect the cement hydration. However, the competitive water absorption mechanism between wood strands and cement is not fully understood. This paper will address the absorption behaviour of wood wool strands in combination with cement. A model for calculating the water to binder ratio of the paste in presence of wood is proposed and validated by NMR and isothermal calorimetry. Finally, the mechanical performances of the composites are tested for different water amounts, verifying the proposed model and defining the conditions for the optimal bending strength.

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1. Introduction

Due to the growing environmental concern, bio-based materials have become a common substitute for the conventional reinforcement in composites [1]. Natural fibres are often used in cement composites to increase both toughness and mechanical performances [2,3]. Moreover, they are employed due to their ease of production, the low density and the enhanced biodegradability, leading to applications for sustainable purposes in civil engineering [4–6]. On the other hand, the presence of wood introduces some critical factors, which affect the properties of the composites, compromising their final stability. Among those factors, the time of harvest, the wood species, the storage conditions, the composite manufacturing method and the moisture content [7] are playing a major role [8,9]. Therefore, even if the production process does not change, the variation in these other wood parameters creates heterogeneity in the final product properties [10]. Since 1940, the

wood wool (called Excelsior) for wood wool composite boards (WWCB) manufacture is commonly used in Europe and Asia [11]. WWCB are used as fire resistant, sound absorbing walls and ceilings, but also as thermal insulation panels [12]. Although produced on a large scale, the composition and properties of the WWCB are variable, having an apparent density in the range of 300–500 kg/cm³, a wood/cement ratio between 0.4 and 0.6, and a final bending strength lower than 10 MPa [13].

1.1. Factors influencing the stability of wood wool composites

In general, acceptable properties of an inorganic bonded board are dependent on both the amount and nature of the binder and on the wood properties [14]. Among the influencing factors, it is well known that the presence of wood extractives might inhibit the hydration of cement (PC) [15,16]. Granting the impossibility to fully avoid the presence of those extractives, some wood species, such as Spruce, normally do not cause the inhibition of the cement hydration, because of their lower content of those water-soluble compounds [17].

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Numerous factors can affect the bonding mechanism between cement and wood. Among them, the hygroscopic behaviour of the wood plays a major role in the cement hydration, by determining the amount of water available for the binder [18,19]. In presence of water, the ions exchange between cement and wood could lead to a strong bond between the two materials, due to the improved anchoring of the binder [10]. However, for excessively soaked strands, the migration of water soluble extractives is favoured, leading to the partial setting of the cement paste.

Therefore, a low initial moisture content of the wood means that part of the water used for cement hydration will be absorbed into the wood structure, instead [20,21]. Contrariwise, an excessive moisture content of the strands will provide an abundant amount of water for the cement hydration, increasing the final water to binder ratio (w/b) of the paste, but also causing the dissolution of extractives, resulting in a low strength development [22]. This is the reason why the manufacture of a composite must take into account both the water for the distribution and the reaction of the binder [10]. Until nowadays, the liquid absorption mechanism of Norwegian spruce in form of wood wool strands is not often investigated, leading to uncertainties about the quality of the bonding between wood strands and mineral binder. Moreover, in the face of the necessity to limit this ion exchange, the influence of the absorption behaviour of wood strands in presence of PC as well as their “competitive absorption” has not been investigated. The determination of the effective amount of water available for binder hydration is necessary to reduce the inhomogeneity during manufacture. Furthermore, a better understanding of the effective w/b ratio applied will ensure the maximization of the mechanical performances of the composite.

In this study, a characterization of Portland cement and Norwegian Spruce excelsior wood wool is provided. Thereafter, the liquid uptake of spruce strands is measured empirically and by solid state hydrogen nuclear magnetic resonance (NMR) results, which helps to differentiate the water located in the spruce structure and on the strands surface. Based on the results, the quantification of the pre-soaking water necessary for a precise w/b ratio in the binder is computed, considering the wood initial moisture content (MC_i), the binder water demand and the wood/binder ratio of the composite. The validation of the model is performed using isothermal calorimetry, by comparing the cumulative heat of the binder in presence and absence of wood, at 48 h. According to the same w/b ratio used in the validation, WWCBs are manufactured and tested for bending strength, by three-point flexural testing.

1.2. Boundary conditions for the liquid sorption model

The behaviour of wood strands in presence of water is directly related to the wood species, the structure of the wood cell, as well

as their chemical composition. The main regions used for characterizing the wood moisture adsorption capacity in this study are displayed in Fig. 1, together with the water speciation in the wood cell, depending on the different moisture contents (MC). In oven dry conditions (0% MC) the wood structure is composed of cells, characterized by cell walls and empty lumens; no water is detected in the structure [23]. A second region is defined as the “hygroscopic region” [23], where the wood behaviour is mainly influenced by the presence of amorphous cellulose and hemicellulose [1,24]. Compared to crystalline cellulose, amorphous cellulose attracts more liquid and chemically bind the water molecules, due to the numerous available sorption sites ($-OH$ groups) [23]. In the hygroscopic region, the cell walls are filling up until saturation, while no liquid is in the lumens [25]. In this study, the fibre saturation point (FSP) is considered equal to 30% MC (based on the oven dry mass of the wood) [25], which corresponds to the conditions where the cell walls are completely filled and the lumens are completely empty. The region over 30% moisture content is defined as “over-hygroscopic region”, when the water is in the lumens, as the cell walls are already completely filled. This physical water absorption takes place only by capillary suction [23]. As measured in a preliminary study, fully saturated conditions (220% MC) are achieved when all the lumens and smaller pores are filled.

2. Methodology

2.1. Materials

In this study, Norwegian spruce is used as reference wood in the form of Excelsior wood wool, provided by Knauf Insulation (NL). Spruce is analysed in the form of strands 2 mm wide, 0.4 mm thick and 250 mm long. The binder applied in the study is CEM I 52.5 R white (PC) provided by ENCI (NL).

2.2. Methods

2.2.1. Materials characterization

The dry mass of the wood wool has been measured by drying the sample in an oven at 105° C, for 24 h [25]. Visualization of the spruce structure is performed by Scanning Electron Microscopy (SEM, Quanta 650 FEG, FE), coupling large field detector, GSED detection (LFD) and BSE- detector (low vacuum, chamber pressure 0.6 mbar, spot size 4.0, voltage 10 kV). The analysis of the chemical composition of spruce is done accordingly by HPAEC to Tappi T222, Tappi UM250 and Tappi T264 standards. A general chemical composition of the binder is measured by X-ray Fluorescence spectrometer (PANalytical Epsilon 3 range, standard less OMNIAN method), on pressed powder. The specific gravity is determined,

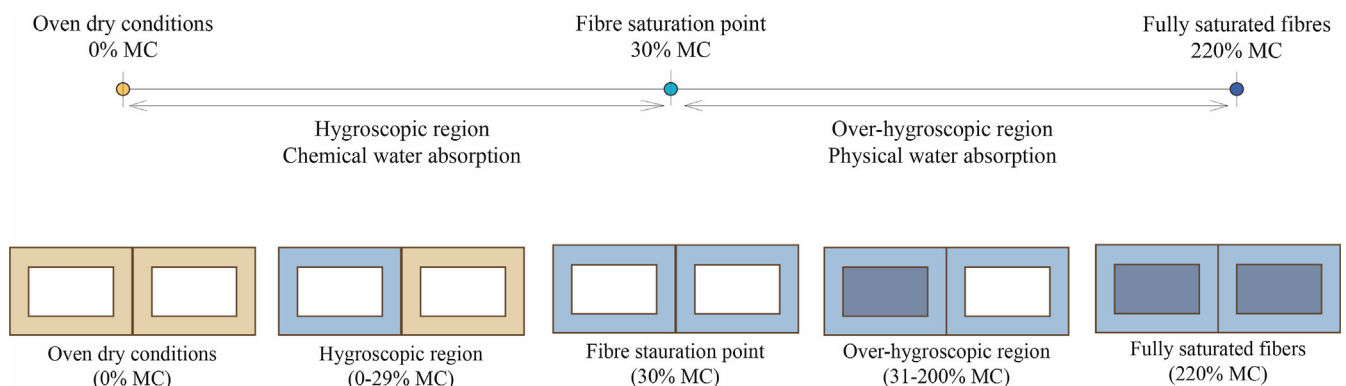


Fig. 1. Representation of the water speciation in an enlargement of wool wood cells, according to the increasing moisture content [19–20]. Wood moisture capacity is characterized by two main ranges: the hygroscopic region (0–29% MC) and the over-hygroscopic region (between 31 and 220% MC).

using a helium pycnometer (Micromeritics Accupyc II 1340) and the bulk density, by a conventional pycnometer. BET surface area is measured by using a nitrogen adsorption measurements (Micromeritics, Tristar II 3020V1.03). The water demand of the binder is measured by the Punkte test [26].

2.2.2. Water absorption measurement for spruce wood wool

Due to the fast water uptake of strands compared to cubic specimens, the water absorption of wood wool cannot be investigated by submersion. Therefore, a different water absorption measurement is carried out, which is displayed in Fig. 2 and repeated three times:

- Knowing its MC_i (14%), 2 g of wood wool are sprayed with ≈ 0.5 g of water (m_w), considering an absorption time of 3 min. This absorption time represents the time between the pre-wetting treatment of the wood wool and the application of the mineral binder, during the WWCB manufacture.
- For the quantification of the water located on the strands surface, the wet strands are dried carefully with absorbent paper, without pressing them, in order to remove only the water located externally, and then weighed.
- The final mass of the wood strands after being dried is also weighed, to evaluate the mass of water located internally.

Four main parameters are tracked during the measurement: the liquid sorption (LS) representing the liquid uptake internal to the wood cells in every step (chemically and physically bonded water), the moisture content of the strands (MC), the surface water (SW) indicating the water located externally on the strand surface and the evaporation water (EW), describing the water mass loss during the measurement. The parameters are quantified per step as a percentage of the mass of the initially applied water (m_w) (Fig. 4) or of the oven dry mass of the wood (m_{od}) (Fig. 7), as a function of the increasing moisture content. For clarification purposes Table 1 resumes the parameters used for the evaluation of the water speciation in this study.

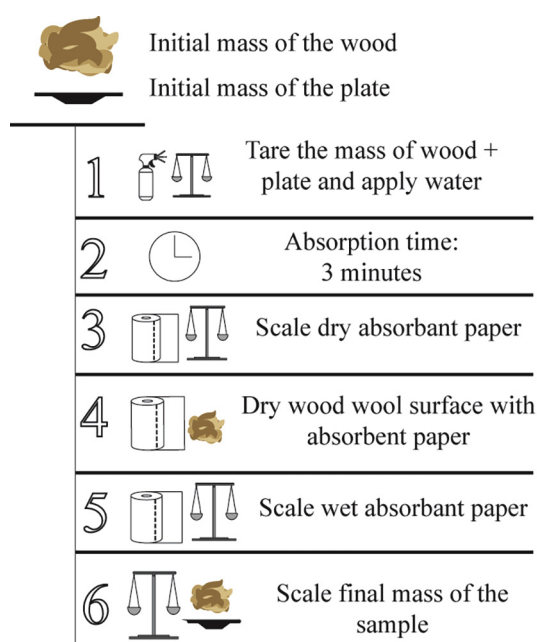


Fig. 2. Schematization of the liquid sorption measurement for wood wool strands, at ambient temperature. The measurement is repeated three times.

Table 1

Parameters used in the study for the calculations, and their description.

Parameter	Description
LS_w	Liquid sorption of wood normalized for the m_{od}
LS_B	Water demand of the binder normalized for the m_{od}
LS	Liquid sorption of wood normalized for the m_w
m_{BB}	Mass of binder used in the board manufacture
m_{WP}	Mass of water used in the Punkte test
m_{od}	Oven dry mass of the wood
m_i	Mass of the wood at initial moisture content
m_w	Mass of water applied during absorption measurement, per step
MC_{equ}	Moisture content conditions when LS_w equals LS_B
MC_{opt}	Optimal moisture content for the manufacture of the composite
MC_i	Initial moisture content of the strands
w/b	Water to binder ratio

2.2.3. Solid state hydrogen nuclear magnetic resonance (NMR)

The determination of the location of the water absorbed into the wood wool strands is performed by T_2 relaxometry NMR for different moisture contents, in absence of binder. Spin-echo decay curves are measured by the CPMG method using 30 MHz Maran Ultra (Resonance Instrument LTD, Witney, UK) TD NMR. A standard Carr-Purcell-Meiboom-Gill (CPMG) pulse sequence is used. Per decay curve, a total of 1024 echoes are acquired with an inter-echo time of 300 μ s, 16 averages and repetition time (TR) of 4 s. The data are analysed by numerical inverse Laplace transformation, as implemented in CONTIN [27], to get T_2 -spectra that show the relative intensity as a function of the T_2 relaxation time. According to different studies [28–31], soaked wood is characterized by different relaxation times, depending on its moisture content. In this study the strands are tested at different moisture contents, by pre-soaking them with water (11% MC, 50% MC, 150% MC, 150% MC_{dry} and 220% MC). The sample 150% MC_{dry} is obtained by drying the pre-soaked strand with absorbent paper as shown in Fig. 2. Small differences among the relaxation times observed in literature and the one reported in this study are attributed to the different wood geometry (strands instead of blocks) and by the different Spruce species taken into account. Evaporation is not taken into account, due to the short time of the measurements (30 s) and to the sealed conditions of the strands, during the experiment.

2.2.4. Isothermal calorimetry

TAM AIR isothermal calorimeter is used as an indicator for the hydration degree of the binder, both in presence and absence of wood [32,33]. The cumulative heat (Q) provides information concerning the reaction degree of the PC: by increasing the w/b ratio of the paste, a higher cumulative heat is recorded, as the greater availability of water eases the formation of reaction products [34]. The reaction degree of the pure binder with different w/b ratio is measured for 48 h, as a reference, in order to ensure the complete reaction of the main PC phases. The w/b ratios considered are 0.5, 0.8, 1, named PC0.5, PC0.8, PC1 respectively. The composite mixtures used are based on a wood/binder ratio of 0.75. These samples are named PCW0.5, PCW0.8, PCW1, according to the w/b ratio applied in the references. All the samples are measured at ambient temperature (20° C) and normalized for the mass of cement. In order to avoid the influence of mixing, the evaluation of cumulative heat is performed by removing the first 20 min of the measurement. The amount of water necessary for the achievement of the desired w/b ratio is applied by pre-soaking the wood wool strands, accordingly to the w/b ratio chosen. The calculations for the determination of the amount of water are clarified in Section 3.4. Achieving the same cumulative heat in the sample in presence and absence of wood will indicate the same water availability for the binder hydration, and thus same w/b ratio and

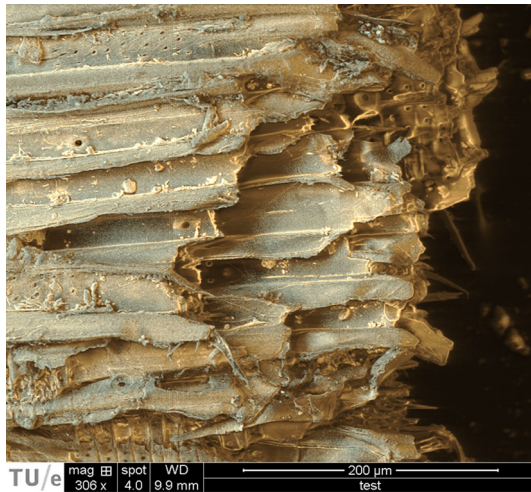


Fig. 3. Wood cell walls structure for spruce wood wool strands, in absence of binder.

hydration degree. In the comparison of the heat released between the two systems (in presence or absence of wood), the reaction involving the water and wood is not taken into account, due to its low intensity measured by calorimetry (13.7 J/g in 48 h).

2.2.5. Manufacture of the composites

Based on the understanding of the liquid sorption dynamics, the optimal w/b ratio of the system can be applied in the composite design. In order to evaluate the mechanical performances as a function of the w/b ratio, WWCB are manufactured with 3 different chosen ratios (0.5, 0.8, 1) and named PCW0.5, PCW0.8, PCW1, respectively. In this study, the manufacture of the composites is based on the dry method, usually applied in WWCB production plant [13,35]. The final product is placed into a mould 15×30 cm and pressed for 24 h, using a mechanical press. Successively, the sample is cured in a plastic sheet for 7 days and then left drying at ambient conditions for another 3 days. In order to achieve the same moisture content before testing, the boards are dried in the oven for 2 h at 50 °C.

2.2.6. Mechanical performances of the composites

The bending strength is measured at 10 days by a three-point flexural test (Zwick 2020) on a sample with dimensions of $15 \times 20 \times 1.5$ cm, using a testing speed of 1.5 mm/min, and a support span of 15 cm (Method A) [36]. Three samples of each w/b ratio are tested. As reference values, the dimensional stability has to be satisfied, by a maximum thickness of 15 mm and a minimum bending strength of 1.7 MPa [35].

Table 3

Chemical and physical characterization of mineral binder used in this study.

A - Chemical characterization of the CEM I 52.5 R white XRF analysis										
CaO	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	K ₂ O	Na ₂ O	SO ₃	MgO	TiO ₂	Mn ₃ O ₄	P ₂ O ₅
% wt.										
67.21	20.93	3.90	0.45	0.12	0.11	2.92	0.43	0.33	0.02	0.51
B - Physical characterization of CEM I 52.5 R white										
Specific gravity [g/cm ³]	Bulk density [g/cm ³]		Water demand g/g		BET surface area m ² /g		Particle size diameter μm			
3.08	1.08		0.22		1.21		0.10–40			

^a Calculated by the ratio between mass of water on mass of binder used during Punkte test.

Table 2

Chemical composition of Spruce strands, performed by the analysis of leachates.

Cellulose [%]	Hemicellulose [%]	Lignin [%]	Extractives [%]
38.60 ± 1.10	34.60 ± 1.01	24.60 ± 0.01	2.20 ± 0.02

3. Results and discussion

3.1. Materials characterization

The micrographs in Fig. 3 display the internal structure of the strands, composed of a tubular cell wall structure and the lumens [23]. Table 2 displays the chemical composition of the Norwegian spruce used in the study. The low amount of leachable extractives present in Spruce (2% wt.) indicates a low inhibition of the cement hydration. The cellulose and hemicellulose content ranging around 35% wt. explains the hygroscopic behaviour of the spruce [37]. Table 3 section A illustrates the chemical composition and main phases of PC, while the physical properties of PC are listed in section B. The reduced BET surface area (1.21 m²/g) leads to a low water demand (0.22 g/g). During the evaluation of the water absorption dynamics (Section 3.4), this value is used as indicator for the binder water absorption, as it takes into account the physical and chemical factors influencing the binder behaviour.

3.2. Study of the spruce strands absorption behaviour

Fig. 4 displays the liquid sorption (LS) of the strands, the surface water (SW) and the steady evaporation water (EW), based on the average mass of water applied in every step as a function of the incremental moisture content in the strands. During measurements, a constant mass loss is registered, due to the evaporation of part of the water applied, calculated as the difference of the total, internal and external applied water. Due to the measuring conditions (25 °C and 50% RH), the evaporation rate is stable at around 20% of the mass of water applied.

Initially, the water applied is mainly absorbed into the strands (LS = 85% m_w for MC = 20%), while only a small amount is located on their surface (SW = 1.7% m_w). The dry cell walls absorb most of the water applied, indicating a maximization of the liquid sorption (within 75% m_w) in the strands for low moisture content conditions (50% MC).

Raising the moisture content in the strands, the liquid sorption decreases and the surface water increases (MC = 100%, SL = 65% m_w and SW = 13% m_w). The reduction of liquid sorption, due to the increasing moisture content, has been already measured on wood blocks [38]. The decreasing liquid sorption rate is explained by a diffusion phenomenon: the liquid diffusion is due to the moisture content gradient between the surface of the strand and its centre. The drier conditions in the centre of the strand favour the migration of water into that area, maximizing the final liquid sorption

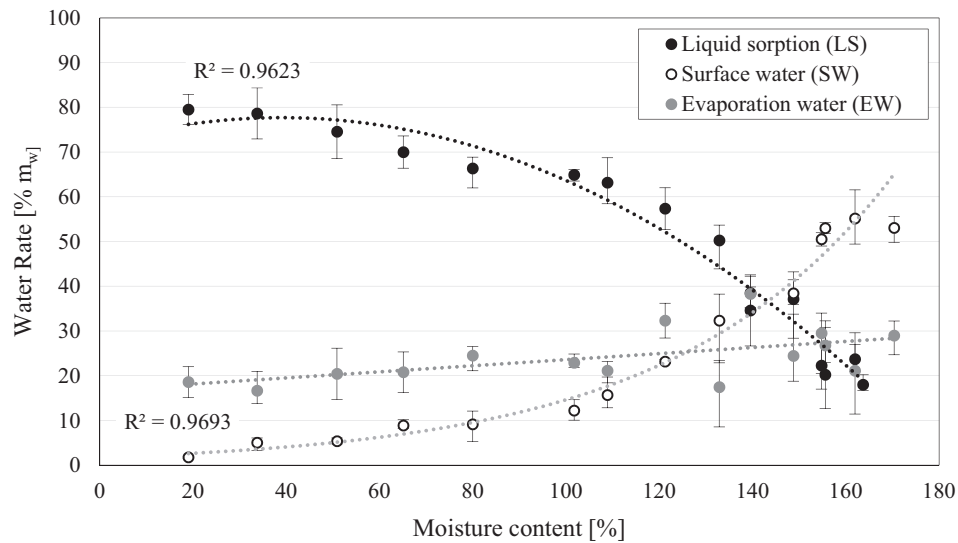


Fig. 4. Normalized liquid sorption (LS), surface water (SW) and evaporated water (EW), normalized for the water applied (m_w). The parameters are represented as a function of the moisture content of the 2 mm strands.

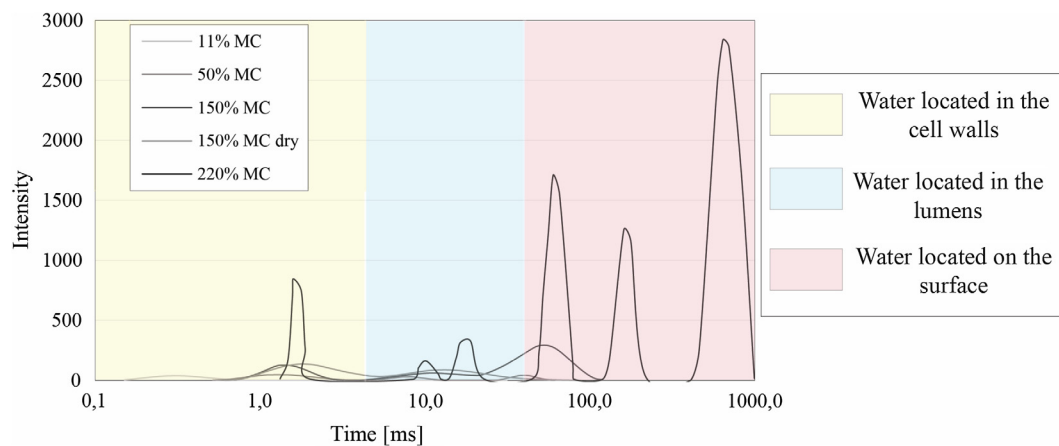


Fig. 5. NMR spectra of spruce strands at different moisture contents [30]. The yellow area corresponds to the wavelength of water located in cell walls, the blue one to the water located in the lumen and the red one to the free water located externally to the strand. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

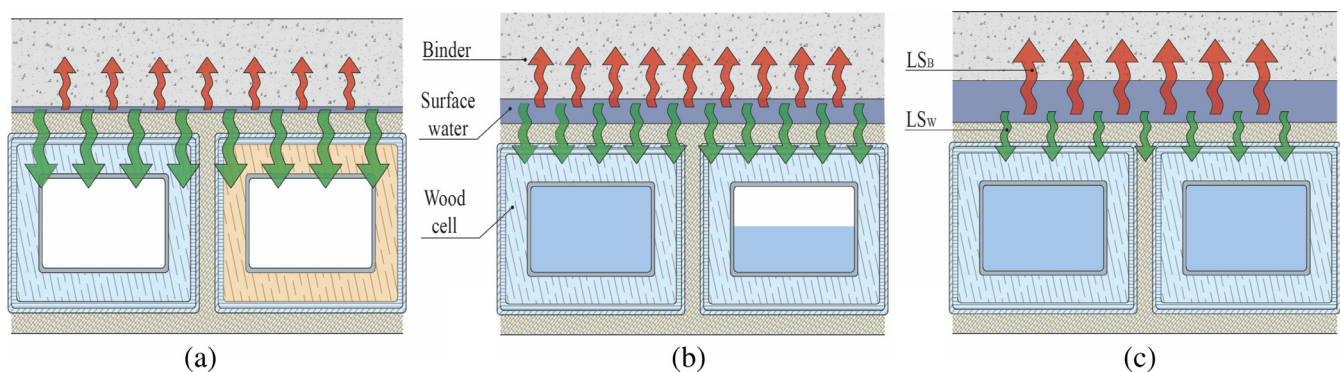


Fig. 6. Enlargement of two wood cells at a) low moisture content, when water absorption is greater than binder ones; b) equilibrium moisture content, which balance the two water absorptions c) high moisture content where wood absorption is lower than the binder water demand. The arrows dimension describes the liquid sorption magnitude of the two materials.

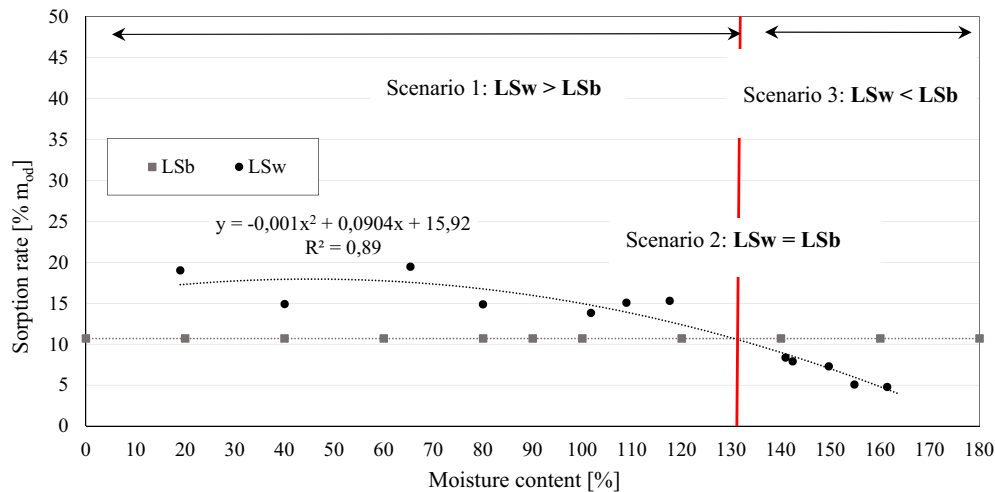


Fig. 7. Water uptake of dry binder and wood wool strands of 2 mm.

of the wood. However, as sorption continues, the moisture gradient between surface and centre decreases, slowing the diffusion process [38]. Unlike the wood blocks where all the water is entirely located internally, in the over-hygroscopic range, the wood strands accumulate part of the water on the external surface, in contact with air. The amount of surface water is related to the liquid sorption of the strands: the greater the liquid sorption, the lower the surface water and vice versa. Therefore, due to this inversely proportional trend, surface water reaches its maximum when the liquid sorption is minimized (MC = 165%, LS = 18% m_w and SW = 50% m_w).

In conclusion, the water absorption of wood strands is faster compared to wood cubes and it leads to the formation of a water layer external to the strand. The surface water layer is not formed once that the full saturation of the strand (MC \approx 220%) is reached, but it is created gradually, due to the decreasing of the moisture gradient between the exterior and the interior of the strand (Fig. 4).

3.3. Water location with increasing the moisture content

To locate the water at different moisture contents, strands are analysed qualitatively by NMR, evaluating the relaxation time under different testing conditions. Fig. 5 displays the T_2 distribution for Norwegian Spruce wood wool at different moisture content. Within the hygroscopic region, the interactions with wood extractives affect the relaxation time of the water in the cells. Peaks within a short relaxation time (below 4 ms – yellow area) (11%MC) are attributed to the chemical bond between water molecules and hydroxyl groups (chemically bonded water) [29,39]. As shown in the empirical measurement (Section. 3.2, Fig. 4), within this moisture range the liquid sorption of the strands is maximized due to the dry state of the strands, while any surface water is detected. Generally, entering in the over-hygroscopic range, the free water located in the lumens corresponds to a relaxation time between 10 and 100 ms [29,39]. In this study, it is defined by the blue area (4–50 ms), where most of the water is physically absorbed in the lumens (50% MC). A comparison of the peaks of 150% MC and 150% MC_{dry} indicates that the red area between 50 and 1000 ms characterizes the surface related water, easily removable by drying the strands. Contrary to the lumens and cell walls, which need more absorbent power to be discharged, the surface water is easily removed by the strands surface and available for binder hydration. Therefore, in case of spruce wood wool a relaxation time higher than 50 ms characterizes the water located externally to the strand (red area), but in contact with its surface, named in this study surface water (Fig. 4). As measured in

Section 3.2, the surface water layer on the strand surface is forming gradually before the strand saturation. Finally, for 220% MC the main family peak ranges between 50 and 1000 ms, indicating a maximization of surface water amount on the strands in saturated conditions.

The analysis of NMR results confirms the empirical measurements of wood water absorption in Section 3.2, Fig. 4. The liquid sorption of wood strands is maximized for low moisture (0–50% MC), while the surface water is not able to form in such dry conditions. Increasing the moisture content (50–180% MC) of the strands a surface water layer starts developing, inversely proportional to the liquid sorption of the wood. In fully saturated conditions, the surface water amount is maximized while the liquid sorption reaches its minimum.

3.4. Influence of the binder on the wood water absorption

In order to optimize the manufacture of composites based on Excelsior wood wool, the evaluation of their liquid sorption in presence of binder is necessary. The manufacture of the WWCB involves the application of the dry binder on pre-soaked strands, which will absorb the surface water available, according to its water demand. Due to the hygroscopic behaviour of the materials used, a competitive absorption of the water will start between wood and binder. Consequently, three main scenarios occur, displayed in Fig. 6a), b) and c). There, for visualization purposes, the red arrows represent the binder water demand (LS_B) while the green ones the wood liquid sorption (LS_W).

Scenario 1 ($LS_W > LS_B$): Due to the low moisture content of the fibres, most of the water migrates into the cell walls and lumens, getting chemically and physically absorbed by the structure. This phenomenon causes a limited water availability for the reaction of the binder, leading only to its partial hydration (Fig. 6a)).

Scenario 2 ($LS_W = LS_B$): The moisture content of the strands leads to a liquid sorption that is equal to the water demand of the binder, creating an equilibrium condition (Fig. 6b)).

Scenario 3 ($LS_B > LS_W$): Increasing the moisture content, the liquid sorption of wood is reduced (Fig. 4, Section 3.2), allowing the PC to take the free water available from the wood surface and lumens and causing an excessive water uptake by the binder. The final matrix might result in a high w/b ratio, and thus low strength (Fig. 6c)).

The scenarios presented in Fig. 6a), b), c) are graphically represented in Fig. 7, where the LS_W of the wood is combined with the binder LS_B , using Eqs. (1) and (2). In order to make water demand and liquid sorption comparable, they have been both normalized

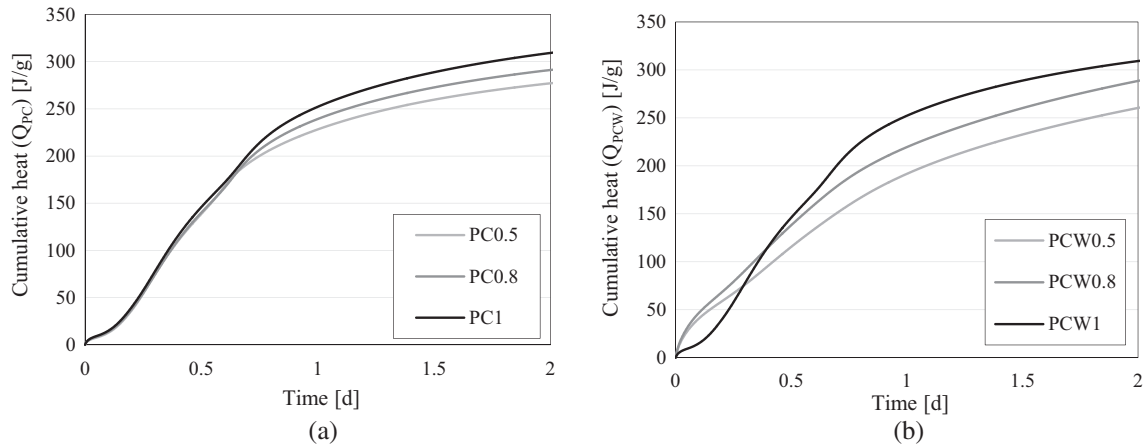


Fig. 8. Isothermal calorimetry analysis used for validation of the proposed model. a) Cumulative heat in absence of wood and b) Cumulative heat in presence of wood for different w/b ratios. Presence of letter W indicates the presence of wood into the sample.

Table 4

Cumulative heat comparison between cumulative heat of PC and PC + wood (Q_{PC} and Q_{PCW} , respectively) at 48 h. ΔQ indicates the difference between the two cumulative heats, expressed both in J/g and % of the Q_{PC} .

w/b	Q_{PC}	Q_{PCW}	ΔQ	ΔQ Deviation rate
-	J/g	J/g	J/g	% Q
0.5	276.7	260.1	17.0	6.0
0.8	290.8	280.6	10.1	3.4
1	309.0	300.1	8.9	2.9

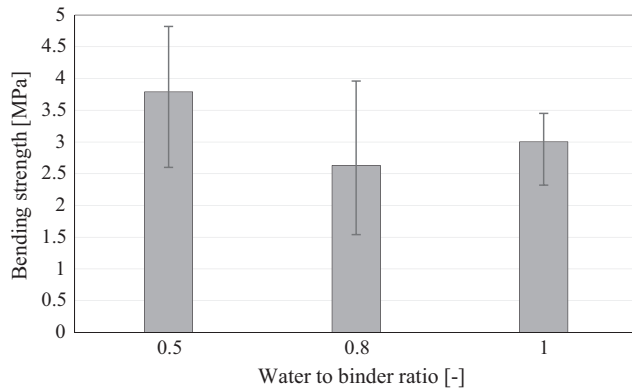


Fig. 9. Bending strength of the WWCB as a function of the w/b ratio applied. The minimum bending strength is set at 3.0 MPa.

for the dry mass of the wood (LS_B and LS_W , respectively), according to the Equations:

$$LS_B = \frac{m_{WP}}{m_{od}} * 100 \quad (1)$$

$$LS_W = LS * \left(\frac{m_i}{m_{od}} \right) \quad (2)$$

where, m_{WP} is the mass of water used for the water demand of the binder [g], m_{od} is the oven dry mass of the wood [g], m_i is the mass of the wood at the initial moisture content [g] and LS is the liquid sorption of wood strands normalized for the mass of the water applied during absorption measurement (Section 3.2, Fig. 4) [g].

Therefore, the moisture content condition for the equilibrium between LS_W and LS_B (MC_{eq}) is computed, by solving the Eq. (3). Those functions are derived from the trend of liquid sorption of

2 mm wood strands, displayed in Fig. 7. Determined the MC_{eq} , the final pre-treatment water necessary for the chosen w/b ratio (MC_{opt}) is calculated by Eq. (4):

$$LS_B = -0.001MC_{eq}^2 + 0.0904MC_{eq} + 15.92 \quad (3)$$

$$MC_{opt} = MC_{eq} + \left(\frac{\left(\frac{w}{b} \right) * m_{bb}}{m_{od}} * 100 \right) - MC_i \quad (4)$$

where,

LS_B is the normalized water demand of the binder [%], MC_{opt} is the optimum moisture content for the reaction of the binder [%], MC_{eq} is the moisture content verifying the system of Eq. (4) [%], m_{bb} is the mass of the binder used for the boards manufacture [g], w/b is the desired water/binder ratio to applied in the board [-], and MC_i is the initial moisture content into the strands [%].

As described in Fig. 4, the liquid sorption of wood (LS_W) is controlling the absorption behaviour of the system, as it is maximized within the 50% MC (sorption rate 18% m_{od}). Within this moisture content the LS_W is higher than LS_B , indicating that all the water available will be driven inside the strands. Overcome the hygroscopic region (between 50% and 130% MC), the liquid sorption of wood decreases, due to the reduction of the moisture gradient between strands surface and centre, until matching the water demand of the binder (130% MC with a sorption rate of 10% m_{od}). Over this MC_{eq} , the binder water demand drives the absorption behaviour, due to the saturation of the strands. In Fig. 7, Scenario 2 is described by the intersection between LS_W and LS_B trends ($MC_{eq} = 130\%$), while Scenarios 1 and 3 are represented by the adjacent areas (higher and lower moisture content than 130%, respectively). If the LS_W has the same intensity of the LS_B , the further amount of water applied will correspond to the liquid available for the binder to react. Once the binder will reduce the moisture content of the strands, by up taking water for hydration, the LS_W in the wood will rise again balancing the LS_B of the binder and avoiding further suction.

The identification of a MC_{eq} to equalize sorption powers of wood and binder (LS_B and LS_W , respectively) is possible by the representation of these parameters, as the rate of the oven dried mass of the wood. For CEM I 52.5 RW and spruce 2 mm strands the MC_{eq} is achieved for 130% MC. Pre-treatment moisture content (MC_{opt}) necessary for the WWCB manufacture can be defined by Eq. (4) according to the chosen w/b ratio.

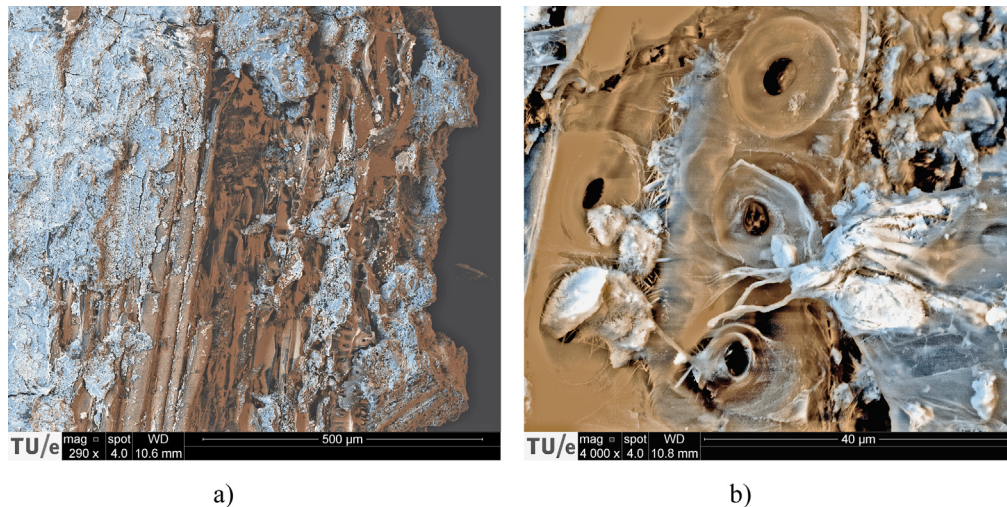


Fig. 10. SEM of a) spruce strands from wood wool cement boards with w/b ratio 0.5 covered by cement and b) bordered pits filled by reaction products.

3.5. Validation of the water absorption behaviour by isothermal calorimetry

Fig. 8 displays the calorimetry results used for the validation of the model proposed, comparing the cumulative heat of PC mixtures in absence (Fig. 8a)) and presence of wood (Fig. 8b)), for the selected w/b ratios. The pre-soaking treatment applied is corresponding to the MC_{opt} determined by Eq. (4), according to the different w/b ratio measured. Generally, all the samples are characterized by the same trend, where a higher availability of water leads to a greater reaction degree in the sample. Raising the moisture content accordingly to the different w/b ratio selected, the same cumulative heat is achieved in absence and presence of wood, with a deviation within the experimental error (Table 4). The increase of the surface water on the strands results in a higher amount of water available for reaction, raising the hydration degree. The development of the same cumulative heat testifies the same availability of water for the cement reaction, leading to the same reaction degree of the paste.

This comparison shows correspondences in the reaction degree of the binder in presence and absence of wood. These data validate the quantification of pre-soaking water calculated by Eq. (4), indicating that the behaviour proposed is resembling the competitive absorption mechanism between wood and cement.

3.6. Validation of the water absorption behaviour by mechanical performances

Manufactured WWCB are tested for bending strength, in order to evaluate the impact of the w/b ratio on the mechanical properties of the composites. Fig. 9 displays the results. The boards show a good dimensional stability independently of the w/b ratio applied.

For w/b 0.5 and 0.8, the boards range around 15 mm thickness, while for w/b ratio 1 a higher thickness is achieved after drying (15.8 mm). The bending strength is the highest for the lowest w/b ratio (PCW0.5), due to the higher density of the matrix created. During a failure, the WWCB distributes the tensile and compressive stresses on the wood and binder, respectively. Therefore, under compressive stresses, a denser matrix favours the development of higher mechanical performances [40]. Increasing the amount of pre-soaking water of the boards (PCW0.8 and PCW1) the final performances are decreasing. The higher availability of water on the strands surface (Section 3.2, Fig. 4) determines the

greater w/b ratio of the binder, leading to the formation of additional porosity [41] and a drop in mechanical performances.

This interaction is evaluated by the presence of reaction products detected into the cell wall structure, caused by the hygroscopic behaviour of the wood. The SEM image in Fig. 10a) displays the strands-cement bonding in the composite PCW0.5, where gel formation is visible on the strands surface. Furthermore, a micrograph at higher magnification of bordered pits reveals the presence of needle shaped reaction products, ascribable to ettringite, inside the lumens of the spruce strands (Fig. 10b). This result confirms the migration of cementitious ions into the structure, and therefore the presence of an exchange phenomena taking place between wood and binder, using water as medium. This leads to an improved bonding between wood and cement [10], justifying the higher performances in bending strength.

The results validates the behaviour predicted by the model presented in Section 3.4. In this system including CEM I 52.5 RW and 2 mm wood wool strands, the fulfilment of the mechanical requirements is overcome (3.8 MPa) for w/b ratio of 0.5, taking into account a MC_{eq} of 130%.

4. Conclusions

A model was developed to calculate the competitive water uptake of natural strands (wood wool) and cement in wood cement composites (WWCB). From the analysis performed the following conclusion can be drawn:

1. During the water absorption of wood wool, the water is distributed both internally to the strands (liquid sorption) and on their surface (surface water). The higher the moisture content, the lower the liquid sorption and the greater the surface water of the strands, used for the cement hydration.
2. A method for the evaluation of wood wool water absorption is studied by T_2 -NMR measurement. For Spruce wood wool, a relaxation time below 4 ms indicates the chemically bonded water, while between 4 and 50 ms the physically bonded one. The relaxation times higher than 50 ms address the presence of water on the externally on the strands surface.
3. The equilibrium moisture content (MC_{eq} = 130%) between the liquid sorption of 2 mm Norway spruce wood wool and the water demand of CEM 52.5 I RW is calculated as well as the optimum pre-soaking water of the strands (MC_{opt}). With these

parameters, maximization of the mechanical performances of the composite was achieved with a bending strength of 3.8 MPa for a w/b ratio of 0.5.

4. The proposed water absorption model is confirmed by isothermal calorimetry. Wood wool cement mixtures were prepared using different w/b ratios, by pre-soaking the wood with the amount of water calculated according to the model. The same heat release and thus reaction degree is achieved in presence and in absence of wood, which indicates the same availability of water for reaction.
5. The proposed model can be adapted for other binders as well as other fibres by measuring the liquid sorption of the strands and the water demand of the binder. MC_{opt} of the new system can be calculated according to the formula proposed.

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