Design and performances evaluation of an eco-friendly board applying an alkali activated binder

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Authors Caprai, V., Doudart de la Grée, G.C.H., Q.L. Yu, H.J.H Brouwers

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1   Introduction

There is an increased interest in bio-based composites around the world, because of the renewable nature of e.g. wood that perfectly fits in a sustainable society. One of these composites is wood-wool cement boards (WWCB). Generally, WWCB are consisting of 30-70 % by weight of wood-wool strands mineralized with 30-70 % of inorganic binder and water as a reactant [1]. Currently, a large portion of the WWCB market is using Ordinary Portland Cement (OPC) as binder, generating medium density boards (400-450 kg/m^3). WWCB can have various properties but are mostly applied as a sound absorbing material with 60-120 minutes of fire resistance, depending on the density of the board. Although the application of OPC as binder has been successful for more than 100 years, its manufacturing process contributes to an increasing greenhouse effect. Its production develops around 1 ton of CO_2 for each ton of cement (4 % of the global annual CO_2 emissions) and therefore is less suitable in a sustainable environment. In order to reduce the environmental impact, some alternative industrial minerals are already successfully applied in preliminary studies as a partial replacement of by-products e.g. Fly ash (FA), Ground Granulated Blast Furnace Slag (GGBFS) and Limestone powder (LP). However, the replacement is ≤ 40 %, as above this quantity their requirements contribution is insufficient concerning reactivity/strength, hence, cannot meet the 24 h and 10 days strength performance. Therefore, at higher replacement levels the material need to be activated by using e.g. additional industrially manufactured alkalis. However, there are certain aspects that need to be considered when applying such alkalis like: (1) the production of alkalis e.g. Na_2SiO_3 also influences the environmental impact (400 kg/CO_2 per ton); (2) they are costly; (3) the use of an alkali-activated binder in combination with organic compounds is not straightforward as the aggressive condition (high pH) can decompose the organic compounds in WWCB [2].

In this study, an alkali activated binder (AAB) is used, composed of industrial minerals which are selected based on their mineral composition. By this a high content of alkali is present in the raw material and the necessity of additional alkalis is reduced, leading to a significant lower environmental impact compared to OPC. Furthermore, the choice of introducing this alkali activated binder is related to: (1) a low pH characterizing the reaction process; (2) the reutilization of industrial minerals; (3) the reduced CO_2 footprint of the binder due to its composition (10 times lower than OPC); (4) the possible increase of thermal and sound absorbing properties, due to its refined microstructure and lower amount of chemically bound water.

Firstly, characterization of the main binders are performed. Secondly, interaction with a retarder (glucose) and influence of the water/binder ratio (w/b) are studied. Thirdly, the main issues concerning the board manufacture process are summarized. Finally, a hybrid system is introduced with a lower environmental impact compared to 100 % OPC, consisting of 30 % OPC with 70 % AAB system to verify the board performances e.g. mechanical strength while fulfilling the EN 13168.

2   Experimental program

2.1   Materials characterization

The binders used in this study are a commercial available OPC type: CEM I 52.5 R White (WOPC) (provided by ENCI, the Netherlands) and two types of alkali activated binders that due to their low environmental impact are termed Eco N and Eco Q. The chemical composition of the materials are presented in Table 2.1. Further features concerning the main materials, as PSD, density, water demand are summarized in Table 2.2, measured with Malvern Mastersize 2000, Micrometrics Accupyc II 1340 and Puntke test, respectively. The choice of using WOPC is due to the high quantity of C_3A and C_3S that increases the early strength development of the binder. Eco N differentiates itself from Eco Q by a slower reaction and due to its mineral composition. Eco Q is later added to this study and is therefore only used in the material characterization, board production and hybrid system section. The choice of including Eco Q is related to the adjustment of the binder requirements along this study in terms of a higher dissolution and reaction as will be explained later. The main difference between the alkali systems examined is the calcium content. It can be observed from

Table 2.1 and 2.2 that Eco N has a CaO/SiO ratio close to 1, with a high amount of aluminates, alkalis (K_2O and Na_2O) and sulfates. On the other hand, Eco Q has CaO/SiO ratio of 2.2, a lower rate of alkalis (K_2O) and a high amount of SO_3, due to the high reactivity provided by the Ca.

Table 2.1: Chemical composition of the applied compounds [%].

Binders	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$_5$
Eco N [%]	34.41	32.29	12.6	5.71	1.16	1.9	5.16	4.92	0.92	0.16	0.29
Eco-Q [%]	45.19	19.95	9.14	7.47	0.76	-	8.22	7.37	0.78	-	0.48
WOPC [%]	67.19	20.86	3.91	0.45	0.12	0.11	2.92	0.4	0.33	0.02	0.51

Table 2.2: Eco-binder physical characterization.

Binders	Water demand [g]	Density [g/cm^3]	PSD [um]	Packing [-]
Eco N	15.10	2.75	0.27-355	0.55
Eco Q	16.05	2.92	0.27-355	0.53
WOPC	14	3.11	0.39-80	0.53

For the board manufacture spruce wood-wool strands are applied with dimensions of 1.5 mm in width, 25 mm in length and 0.1-0.3 mm in thickness. The use of spruce wood is attributed to its minimal inhibitory effects on the cement hydration [3]. Figure 2.3 displays differences in the boards' appearance. During the board manufacture small quantities of additional compounds as a glenium 51 superplasticizer and waterglass are applied to ensure the distribution of the paste on the wood-wool strands and to increase the reaction e.g. at the interfacial transition zone between the wood-wool and binder.
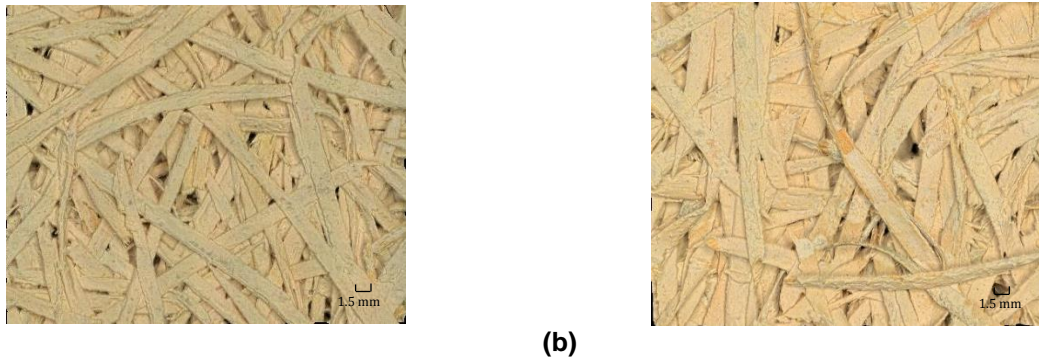

(a)   (b)

Figure 2.3: WWCB characterized by different thicknesses (a) 1.5 mm fibers and (b) 2 mm fibers.

The waterglass enhances the polymerization process of the ionic species present in the system, developing a higher mechanical strength [4]. During the analysis of the hydration process, the presence of D-glucose as a retarder is investigated.

2.2   Measurements

The early stage reaction process of the WOPC and AAB types are analyzed by evaluating the cumulative heat and the heat flow at constant temperature of 25 °C using a thermometric TAM air isothermal calorimeter. The samples are characterized by 6.5 g binder, with w/b ratio 0.5. Furthermore, the addition of 2 % glucose by mass of the binder termed WOPC g and Eco Ng are studied. The measurements are recorded for 72 h.

The pH measurement into the paste are performed by using a pH meter. The mortar including sand (1350 g) and binder (450 g), designed with w/b ratio 0.67 which is also used for board manufacturing. (The pastes termed Eco Q and Eco QW70 are located into a manual mortar mixer for 15 minutes for homogenizing the mixture. The test is performed by diluting 1 g of paste into 100 g of distilled water, repeating it each 15 minutes).

3 Results and discussion

3.1 Reaction mechanism of OPC and AAB

As far as the OPC reaction process is concerning, the hydration is a chemical reaction where the main compounds are created due to the formation of chemical bonds among water molecules and the major cement phases. In the beginning of the reaction the tricalcium silicate quickly reacts with water, increasing the pH till optimal values (around 12) because of the release of alkali hydroxide (OH-) ions, such as NaOH, KOH. After the complete consumption of these hydroxides the pH level drops, due to the saturation of the solution with respect to $Ca(OH)_2$ [5]. The pH of the paste is dependent on the release of OH- groups from the compounds, so that the amount of water applied is not influencing the pH of the paste.

On the other hand, in AAB the initial step of the reaction is the rupture of the covalent bonds of the primary materials due to the increasing pH of the alkaline solution [6]. Contrary to the WOPC mechanism, in the AAB the solution has to provide alkaline hydroxide ions to accelerate the reaction of the solid precursor [7]. The higher the amount of dissolve alkali activator is, the greater will be the amount of reacted particles due to the high pH will be. A low OH- concentration will not allow the pH to reach optimal values, and thus part of the primary material will not be activated. In this case the amount of water is determining the homogeneous distribution of the alkali activator into the paste, as well as it is responsible for the creation of optimal condition for the reaction process.

Although both the systems have an exothermic reaction, the heat developed during the consumption of phases is different. The AAB is characterized by a high heat development during the initial phase (40 mW/g) and a low one during the phase consumption (1-2 mW/g). On the other hand, the hydration of WOPC is not reaching high dissolution peak, but the C_3S and C_3A consumption are clearly visible, as they provide an increase in the heat development of 8 mW/g. Comparing the cumulative heat at 24 h, the Eco N supplies almost half of the heat developed by WOPC in the same time (106.64 J/g against 200 J/g).

3.2 Influence of Retarder

Simatupang & Geimer (1992) states that the glucose could have an inhibitory influence on the cement hydration. This effect of glucose originates a delay, or the complete annulment up to 48 h, depending on the amount of compound inserted into the system [9]. In spite of the difference in the activation, the presence of glucose into the wood can also influence the reaction process of the AAB. Assumptions concerning this phenomenon are: (1) The surfaces of the hydrating cement particles absorb glucose, (2) The surfaces of the hydration product absorb glucose, forming a barrier against further hydration [9].

Figure 3.1 (a) and (b) displays the heat evolution and heat flow rates of Eco N and Eco Q during the first 150 h of reaction, in presence and absence of glucose. A more extended study on the behavior of OPC with glucose can be found elsewhere [10].

Compared to other AABs [11] [12], Eco N illustrates a greater main dissolution peak (63 mW/g). As for WOPC, the addition of glucose influences the AAB reaction, as it causes a slower activation of the paste (Figure 3.1 (a)), as well as a reduced dissolution peak (30 % lower) in the initial phase (Figure 3.1 (b)). Furthermore, it is observed, that the rate of retardation of the alkali activated binder is longer than that of OPC. After a period of 3 days the reaction slowly starts to increase exceeding the Eco N without glucose leading to a higher total heat release and more reaction products. The longer retardation and the slow reaction after can be attributed to a lower dissolution of ions and cover of the particles, reducing the ability to react further. However, slowly more ions are released and the products formed can eventually overcome the effect of glucose, leading to a high ion concentration in the paste and much higher total heat release.
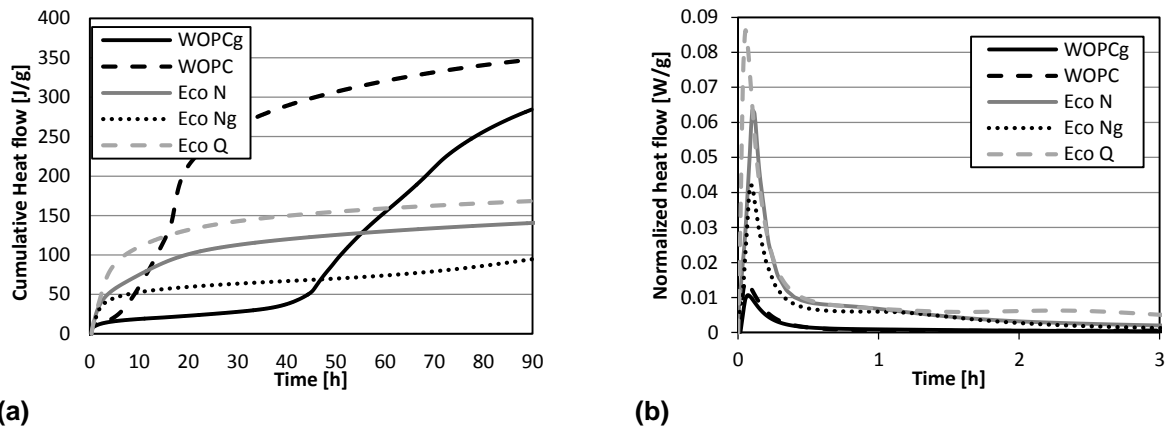
(a)   **(b)**

Figure 3.1: (a) Cumulative heat production and (b) Normalized heat flow for Eco and Eco G.

3.3 Influence of water/binder ratio

The final products and Eco reaction degree are evaluated by using Scanning Electron Microscopy (SEM) on sample, after 10 days.

As for WOPC, also in Eco composites, the w/b ratio influences the mechanical performances of the reacted paste. Figure 3.2 (a) and (b) displays the SEM picture of specimens with respectively high and low w/b ratio, by varying the binder amount.
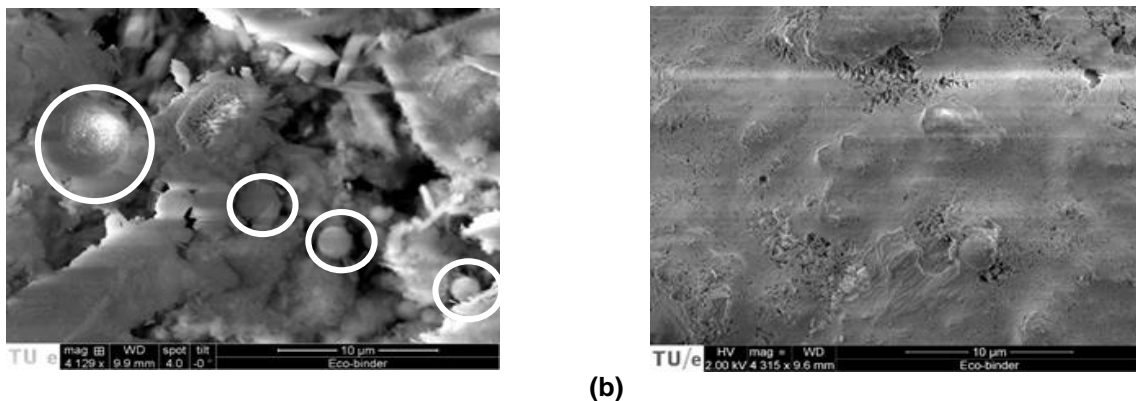

(a)   **(b)**

Figure 3.2: SEM picture of (a) reacted Eco with w/b ratio 1 and (b) w/b ratio 0.9.

FA particles are visible in correspondence of white circle, testifying the impossibility to complete the reaction process within 10 days and instead it acts as a filler reacting in a later stage. Application of an increasing amount of binder results in a denser and thicker layer on each wood fiber, increasing final mechanical strength [13]. However, the increasing amount of binder is only feasible until a certain limit, otherwise the increasing boards' density will not comply the stated requirements (450 kg/m^3).

3.4 Influence of pH

As previously mentioned, the low pH condition of the AAB generates a favorable environment for the maintenance of the wood integrity as well as for the binder reaction. However, the development of reaction products in alkali activated binder as FA, is linked with the pH level, as when 45-50 % of the hydration process is reached, the kinetics of these reactions plunge to very low levels [14]. On the other hand a too aggressive alkaline environment can lead to the decomposition of the wood –wool strands. A comparison is made between Eco Q and a mixture of Eco Q (70 %) and WOPC (30 %) termed Eco QW70. The pH conditions in time is displayed in Figure 3.3. The decreasing trend of the pH in the reaction of Eco reaches the maximum value at 45 min. with a pH of 11.7. The early drop in the pH level creates an unsuitable environment, reducing the particles dissolution reaction [7]. The presence of OPC favors the pH conditions, by keeping them stable in time (pH $\simeq$11.8) for the first 2 h, because of the production of Ca(OH)$_2$. The enhancing rate of reacted particles into the binder results in a denser and thus stronger matrix.
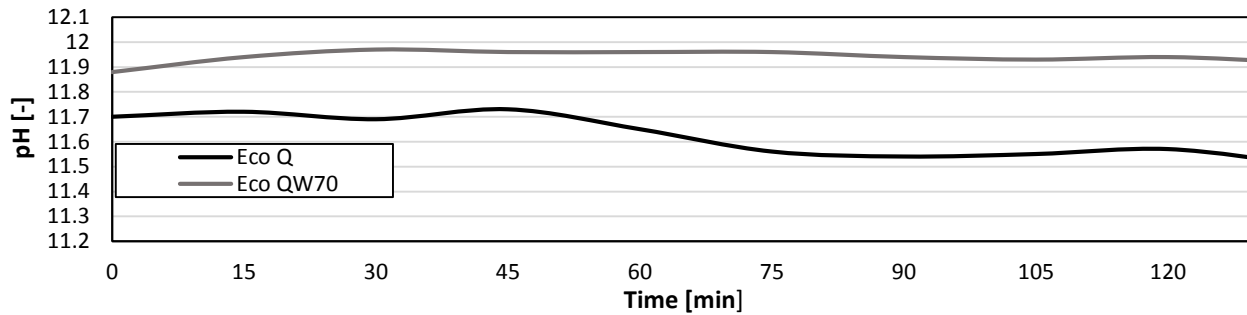
Figure 3.3: pH development of the mixture Eco Q and Eco QW70 in time.

3.5 Boards production

The manufacture of wood-wool binder boards (WWBB) can be performed by using two different methods, e.g. wet and dry method, depending on the type of binder applied. For cementitious binders like OPC, the dry method is successfully used. Firstly, the wood-wool is pretreated using a solution of water and waterglass. Secondly, the binder is distributed on the wood-wool by sprinkling. The wet method (applied in case of AAB) is characterized by a pre-wetting treatment of the wood-wool strands to make them more flexible, in which the waterglass is included. At a later stage, the binder is spread on the wood-wool by using a slurry consisting of binder, water and SP (created separately). The addition of water into the AAB aims to distribute and dissolve homogeneously the alkali activator, increasing the pH and leading to a proper ion and cation distribution for the development of the reaction [7]. The application of the wet method is thus, necessary for starting the reaction process, before the distribution on the wood-wool strands. In comparison with AAB, OPC does not need the preparation of the slurry, because it is able to create optimal conditions for the reaction locally (pH $\simeq 12$), as they depend on the particle dissolution [5]. In this study, the wood-wool is pretreated with ⅓ of the total amount of water and waterglass (2.4 % of mass of the binder), while the slurry is prepared by using a w/b ratio 0.67 with the addition of superplasticizer (2.4 % of mass of the binder). The mixture applied is consisting of 30 % WOPC + 70 % Eco Q by mass of binder. After mixing, the slurry is applied on the wood-wool strands and mixed. A final amount of water (⅛ of the total) is distributed on the covered wood-wool in order to cover them homogeneously before the mineralized wood-wool is placed into molds. Subsequently, pressing is applied for 24 h and boards are afterwards sealed and cured for 10 days.

3.6 Bending strength

During preliminary studies with AAB, it was found that the binder could not sufficiently develop strength/where only partly reacted. This was not only the case with the AAB systems presented in this paper but also with other alkali binder systems studied. Although sufficient strength results were obtained in a mortar (binder + sand) the combination with wood was found to be unfavorable. Therefore, the addition of WOPC, and thus the application of a hybrid system (HS) is studied further. The aim is to enhance the activation of Eco, by favoring the process with an internal heat curing made by the hydration process. Figure 3.4 illustrates the comparison between the reference WWCB trend and the 30-70 1.5 mm and 30-70 2 mm. The HS is characterized by Eco Q and WOPC, 70 % and 30 % respectively. The use of hybrid system results in successful board performances compared to 100 % AAB. The presence of WOPC has a beneficial effect on the Eco reaction. The heat released in the early hydration acts in conjunction with the high pH in the aqueous medium, accelerating particles dissolution. This acceleration of hydration favours the precipitations of a larger amount of reaction products, enhancing the mechanical strength of the matrix [15].

Although lower than the reference, the hybrid system Eco QW70 1.5 mm provides enhanced properties respect to the 100 % AAB, guaranteeing the stiffness and thickness requirements for the board. However, the reduced bending strength for such high density does not allow the comparison with WWCB. On the other hand the Eco QW70 2 mm board results in performance overcoming the reference, due to the greater binder/wood ratio.
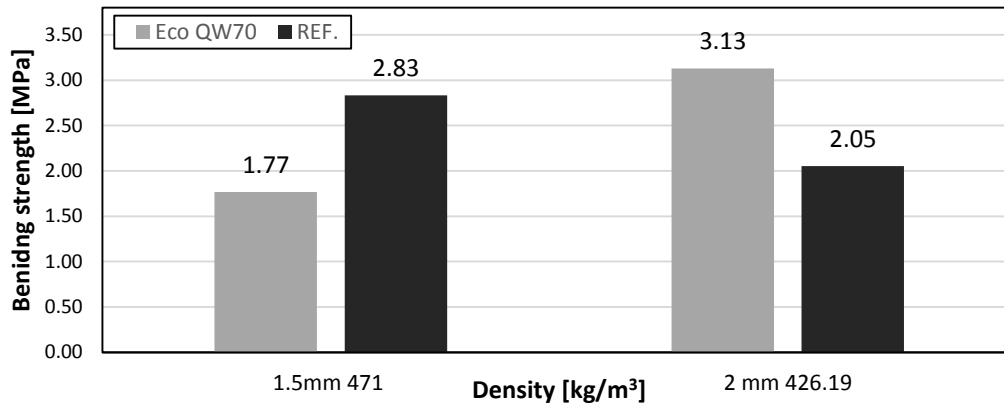
Figure 3.4: Bending test results comparison among Eco QW70 and the REF. WWCB.

4   Conclusions

In this research the application of an alkali activated binder into the wood wool composite is studied. The combination of an alkali system together with an organic material has never been developed, due to the sensitivity of organic materials in combination with the alkaline solution in which the binder should react. The following conclusions can be derived from the study:

- The reaction mechanism of an AAB, due to the dissolution of the activator and its ion distribution, makes the system much more sensitive to the amount of water applied, compared to OPC that only needs to be in a humid environment to be activated.
- As OPC, AABs are sensitive for retarders like glucose, as it prevents the ion dissolution and the further reaction of the binder. However, this phenomenon can increase the binder final mechanical performance in a later stage, rising the heat released compared to the reference mix.
- In the board performances, the decreasing w/b ratio results in a denser and stronger matrix. The fibers results more bonded, due to the thicker, and stronger layer of binder covering them.
- The heat released during the WOPC reaction favors the dissolution of particles in the AAB, increasing the amount of reaction products and thus the mechanical strength.
- The more stable pH conditions created by the presence of WOPC enhance the reaction of the Eco Q, resulting in a stronger matrix.
- The optimal conditions for the manufacture of a board by using a hybrid system involve a high calcium content AAB and *WOPC*. The combination of the AAB system with the cementitious one provides a good mechanical strength and respect the thickness requirements due to their positive interaction and stable pH level compared to a wood-wool binder board made of only AAB.

5   Acknowledgment

6 References

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