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Carbon dioxide sequestration of fly ash alkaline-based mortars containing recycled aggregates and reinforced by hemp fibres

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HIGHLIGHTS

- Mixture with 8% hemp fibres show a 50% reduction in mechanical properties.
- Accelerated carbonation leads to a carbon sequestration of −102 $kgCO_2eq/m^3$.
- Mixture without hemp fibres show a carbon footprint of 38 $kgCO_2eq/m^3$.
- The use of 8% hemp fibre has a negative global warming potential of −19.7 $kgCO_2eq/m^3$.

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ABSTRACT

Carbon dioxide sequestration is crucial for targets for limiting global warming could be achieved. This paper discloses results of an investigation concerning the performance of fly ash/waste glass alkaline-based mortars with recycled aggregates reinforced by hemp fibres exposed to accelerated carbon dioxide curing. Compressive strength, freeze-thaw resistance, carbon footprint and cost were studied. The results show that hemp fibres lead to a reduction of mechanical properties of alkali-activated materials. A high correlation was found between compressive and flexural strength. The results also show that accelerated curing provides a high carbon sequestration. Furthermore, the use of at least 8% hemp fibres leads to carbon negative emissions −19.7 $kgCO_2eq/m^3$ for fly ash/waste glass alkaline-based mortars with recycled aggregates based composites.

1. Introduction

2016 was the first year with atmospheric CO_2 concentrations above 400 ppm all year round [11]. This means that the 350 ppm boundary set in the [27] global sustainability model was already crossed risking "*abrupt environmental change within continental-to planetary-scale systems*". Therefore, some authors [16] state that carbon dioxide sequestration is crucial so targets for limiting global warming can be achieved. That is why carbon sequestration constitutes one of the Grande Challenges of Engineering [20]. Currently this carbon sequestration is carried out mostly through geologic CO_2 storage in saline aquifers [34]. However, that constitutes a passive strategy has large risks and also has a very high cost. Carbon capture and storage (CCS) from the stream of concentrated CO_2 at fossil fuel burning sites like power plants or steel plants is more efficient and thus less expensive than direct air capture [16]. As a consequence it is important to study how CO_2 generated by power plants and other facilities can be sequestrated in valuable products. Several authors [10,17] have studied the use of CO_2 as accelerated curing of cementitious constructions materials. This technology will in future prevent carbon dioxide to be released into the atmosphere but also to accelerate curing and strength development of those materials. However, so far no studies were performed using alkali activated based materials. These materials are produced though the reaction of an aluminosilicate powder with an alkaline activator, usually composed by hydroxide, silicate, carbonate or sulfate leading to the formation an amorphous aluminosilicate gel and secondary nano crystalline zeolite-like structures [26]. These materials have a particular ability for the reuse of several types of wastes [25,8]. Some wastes like fly ash deserve a especial attention because they are generated in a very high amount and have a very low reuse rate. USA has a reuse rate for fly ash of around 50% meaning that 30 million tons of fly ash are not reused annually [3]. Waste glass is also a waste that is generated in

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relevant quantities and that merits increase recycling efforts. In 2010, approximately 425,000 tons of waste glass was produced in Portugal and only 192,000 tons were recycled. In Hong Kong, approximately 373 tons of waste glass is generated daily in 2010. The high volume of construction and demolition wastes (CDW) also constitutes a serious problem. Eurostat estimates the total for Europe of around 1000 million tons/year, representing an average value of almost 2.0 ton/per capita. The reuse of CDW as recycled aggregates not only constitutes a way to give value to a waste but also prevents the use of river sand being necessary to achieve the 70% target until 2020 in EU [23]. Furthermore, the use of cementitious building materials reinforced with natural fibres could be a way to achieve a more sustainable construction. Natural fibres are a renewable resource and are available almost all over the world. Vegetable fibres cement based composites are as stronger as composites based on synthetic fibres, cost-effective and above all environmental friendly [24]. Moreover, their environmental impact is lower than traditional building materials because relatively large amounts of atmospheric CO_2 can be sequestrated through photosynthesis [30]. Among the new vegetable fibres used, hemp stands out from the rest because of its wide availability, low requirements of fertilizer and irrigation, good humidity control and favourable energy and ecological balances [33]. That is why research on cement composites reinforced by natural fibres constitute an important trend in the sustainability context [21]. Natural fibres can degrade in high alkaline environment of Portland cement composites [15]. However, several authors [2,32], showed that carbonation is associated to a lower alkalinity that can help preserve both the properties and durability of composites reinforced with natural fibres. This means that accelerated carbonation of composites reinforced with natural fibres has not only carbon sequestration advantages but is also especially indicated for such composites. This paper discloses results of an investigation concerning the performance of fly ash/waste glass alkaline-based mortars with recycled aggregates reinforced by hemp fibres exposed to accelerated carbon dioxide curing.

2. Experimental program

2.1. Materials

The mortars were made of fly ash (FA), calcium hydroxide (CH), waste glass (MG), ordinary Portland cement (OPC), recycled aggregates and a sodium hydroxide solution. The fly ash was obtained from The PEGO Thermal Power Plant in Portugal and categorized as class B and group N regarding the [5]. Table 1 presents the major oxides of fly ash particles. The Portland cement is of type I class 42.5R from SECIL, its composition contains 63.3% CaO, 21.4%SiO_2, 4.0%Fe_2O, 3.3%Al_2O_3, 2.4%MgO and other minor components. The calcium hydroxide was supplied by LUSICAL H100 and contain more than 99% CaO. Waste glass from glass bottles ground for one hour in a ball mill was also used. The final density of the milled waste glass was 1.27 g/cm^3. Solid sodium hydroxide was supplied by ERCROS, S.A., Spain, and was used to prepare the 8 M NaOH solution. Distilled water was used to dissolve the sodium hydroxide flakes to avoid the effect of unknown contaminants in the mixing water. The NaOH mix was made 24 h prior to use in order to have a homogenous solution at room temperature. A recycled sand to binder ratio of 4 was used in all the mixtures. The recycled sand was obtained from the crushing of concrete blocks. The average compressive strength of concrete blocks was around 40 MPa. A preliminary sieving operation was carried out to remove both coarser and dust particles before being used. The dimension of the sieves was 4.75 mm and 0.6 mm. The sand was dried at 105 °C for 24 h until constant mass achieved. After the preliminary sieving a standard sieving was carried out showing that the recycle sand has a fineness modulus of 3.885. The detailed grain size distribution of the recycled sand are presented in Fig. 1. The recycled sand has a water absorption by immersion of 13% having being determined with a 24 h saturation according to EN 1097-6. Before use the recycled sand was carbonated in a carbon chamber from Aralab model Fitoclima S600 (4.2% CO_2, 40% RH, and 20 °C) for 48 h. The recycled sand has a water absorption of 25%. The explanation for the increase of the water absorption relates to the fact that when CSH carbonates its Ca/Si ratio drops and it becomes highly porous. Studies by NMR spectroscopy indicate that decomposition of C–S–H caused by carbonation involves two steps: 1) a gradual decalcification of the C–S–H, where calcium is removed from the interlayer and defect sites in the silicate chains until Ca/Si = 0.67 is reached, ideally corresponding to infinite silicate chains; 2) calcium from the principal layers is consumed, resulting in the final decomposition of the C–S–H and the formation of an amorphous silica phase [28]. The mortars were reinforced by different weight percentage of hemp shiv fibres that were supplied by Canapor. No surface treatment was used for the hemp shiv fibres in order to avoid cost increase and maintain its eco-effectiveness. Table 2 shows the composition of calcined hemp. The characterization of hemp shiv fibres was implemented based on a statistical analysis to evaluate the variability of the fibre length, which was defined by using 200 fibres. Regarding the statistical analysis, most fibre lengths varied in the range of 20–30 mm (Fig. 2).

2.2. Mix design and mortar production

The composition of the mortars is shown in Table 3. In the batching process of the mortars, dry ingredients (fly ash, recycled sand, calcium hydroxide (or cement), metakaolin, and milled glass) were mixed for 2 min. Then, sodium hydroxide was added and again mixed for 3 min. Finally the hemp fibres were added and all the ingredients were mixed for 3 more minutes. Then, the mixed mortars were cast into cubic molds ($50 \times 50 \times 50$ mm^3) to assess the compressive strength and in prismatic beams with dimension ($40 \times 40 \times 160$ mm) to assess the flexural strength. The specimens were cured for 24 h at the lab conditions (averagely 25 °C and 40% RH) and then they were demolded. Then the specimens were cured in the carbonation chamber (4.2% CO_2 concentration and 40% RH) for 7 days and curing in the lab conditions for the remaining days until the age of the test. This is because preliminary experiments showed that all mixtures were fully carbonated during 7 days through a CO_2 preconditioning curing. Three specimens with dimension of $50 \times 50 \times 50$ mm^3 were casted and used to measure the CO_2 sequestration in the mixture without hemp fibres by using a furnace decomposition method [13]. The carbonated specimens were placed initially in the oven at 105 °C during 24 h to evaporate any absorbed water. Then, the weights of the

Table 1
Chemical composition of major oxides in fly ash.

Material	Oxides (wt%)							
	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	Na_2O	K_2O	TiO_2
Fly ash	60.81	22.68	7.64	1.01	2.24	1.45	2.70	1.46