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Mechanical performance after high-temperature exposure and Life Cycle Assessment (LCA) according to unit of stored energy of alternative mortars to Portland cement

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ABSTRACT

Decoupling energy demand has led to the importance of energy storage for increasing the capacity of renewable energy power plants. In this field, Portland cement (PC) concrete is proving to be a promising way to store energy as it can be used as sensible thermal energy storage (TES) medium in concentrated solar power (CSP) technology. However, the high energy and water consumption involved in the PC manufacturing process makes it necessary to develop new alternatives. Thus, alkali-activated materials (AAM) and hybrid materials (HM) were manufactured using blast furnace slag and glass waste (GW) to replace the PC and the sand in concretes respectively in order to study their feasibility as TES media in parabolic through CSP systems. The viability of these proposed new systems was tested from a mechanical point of view, while taking into account the environmental aspect using Life Cycle Assessment (LCA) methodology to study carbon and water footprints. The new systems were exposed to high temperature (up to 500 °C), showing better performance than the ordinary PC under high temperatures, and their mechanical properties were not affected at all. After thermal treatment the alternatives show improvements of up to 79% compared to the PC reference sample. Furthermore, in terms of LCA analysis, it was concluded that TES systems with partial (HM) or total (AAM) substitution of PC by using by-products improve water use up to 40% when an AAM material includes GW as a recycled aggregate in its composition. Results likewise revealed a more than 100% reduction in the carbon footprint. These results open a new gate for the study of materials as TES since the alternatives to PC are more promising from an operational and environmental point of view.

1. Introduction

In recent years, climate change has become an imminent environmental emergency, due to the highly polluting gases emitted by industrial activities and the energy needs of transport and buildings. In addition, the population growth rate, leading to an important increase in future energy demand, requires the study and development of new alternatives for energy production that reduce the environmental impact [1].

To deal with this global emergency, numerous Climate Change Conferences have been held during recent decades. The latest one (COP26) took place in Glasgow (2021) where nations adopted the Glasgow Climate Pact, which agreed upon the reduction of CO_2 , the removal of inefficient fossil subsidies, and the promotion of renewable energies [2].

To achieve the aforementioned aims, the Portland cement industry needs to be reexamined because of its negative consequences. Specially, due to its high consumption of fossil fuels, which represent 12–15% of the total energy consumption [1], it emits a large amount of polluting gases, specifically CO_2 , which is the most common greenhouse gas (GHG) contributing to global warming [3]. CO_2 emissions by the PC industry comprise about 7–9% of global CO_2 emissions [4]. These consequences stem from the high energy demand required in heating the raw material (limestone, clays and other additives) to obtain burnt lime and then reacting this compound with silica, alumina and iron oxides, at temperatures between 1400 and 1500 °C, to form the main constituents of clinker [5,6]. On the other hand, the manufacturing process of PC also requires a large amount of water in each step.

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In order to avoid these adverse effects, PC should be replaced. There are two research lines to develop construction materials substituting PC in a total or partial way. First, alkali-activated materials (AAM), socalled geopolymers, are an alternative that proposes the total substitution of PC by using aluminosilicate precursors that are activated by alkaline solutions. These raw materials can be natural minerals, such as kaolinite or clay, or by-products of industrial processes, such as blast furnace slag, fly ash or silica fume. Precursors can be used individually or can be mixed, where the most important factor to consider is the content of silicon and aluminum, as it will determine the strength of the material [1,7,8]. Nevertheless, other parameters such as the alkaline activator, the concentration of the solution or the curing conditions, influence the properties of the material [7]. Second, due to limitations of workability that arise with alkaline solutions [9,10], another alternative emerges: hybrid materials (HM). In this case, there is not a total removal of PC, because HM are mixes made of PC (about 20-30%) and mostly composed of aluminosilicates (70-80%) [11,12]. Additionally, these multi-component systems include soft alkaline salts in their mixtures that are usually present in a solid state (to avoid the use of highly corrosive liquid solutions). A solid activator allows the material to be hydrated using only water (avoiding alkaline solutions), providing good mechanical properties at an early age [12].

Both solutions offer significant cost reduction and environmental improvement because of a decrease in the energy needed in the manufacturing process. This is due to the use of by-products (wastes) that otherwise would end up in dumps, following the principles of the circular economy [1,7,13-16].

Industrial wastes and by-products can be incorporated into construction materials, not only as part of the binder phase, but also as part of the aggregates replacing the sand [17]. This kind of substitution also contributes to environmental improvement since the Life Cycle Assessment (LCA) approach of cement industries reveals that natural aggregates (NA), such as sand, contribute significantly to abiotic depletion, eutrophication, acidification [18] and to the increase of CO₂ [19], among other environmental problems. Consequently, some research is focused on replacing fine NA by wastes from different sources [20] because of the negative environmental impacts caused by the extraction of the NA, and their limited supply in nature [19]. Thus, wastes become important because avoiding the landfill and the energy required in the recycling step improves the overall environmental profile. All of this contributes to reducing the use of new raw materials, helping to maintain limited resources and avoid exploitation and its negative environmental consequences [19,21].

Despite the environmental disadvantages described above, recent research highlights the benefit of using PC concrete as a thermal energy storage (TES) medium to operate in concentrated solar power (CSP) technology [22–25]. PC concrete is proving to be a viable option with a significant cost reduction and high operational efficiency in CSP, which boosts renewable energy, increasing the capacity factor of solar power plants [22–25].

The most relevant properties of TES material are its density, specific heat and thermal conductivity, which defines thermal diffusivity. Also to consider are operational temperatures and pressure, compatibility and stability between the materials used for heat transfer (block and exchanger), heat loss coefficient and cost [22,25].

Following these main characteristics, PC concrete is currently under study due to its low cost, availability, and ease of processing. It has a high specific heat, so it is able to maintain it during a longer time. At the same time, PC presents high values of compressive strength and maintains them after exposure to high temperatures or thermal cycles (normally withstanding temperature loads and discharges between 200 °C and 400 °C [24]). In addition, its thermal expansion coefficient is similar to that of steel, so problems with swelling are minimized at the interface created with heat exchangers [22,26].

The stability of PC systems has been tested up to 500 $^{\circ}$ C, showing that the mass loss and the mechanical properties finally stabilize after a

period and several thermal cycles [27]. It has been proven that PC concrete is more favorable as a TES medium than any other solid medium because of its low cost, good mechanical properties and ease of use. High power levels were able to be achieved during cooling (discharge of the block) and the operation run without problems, even with the high temperature difference between the block and the heat transfer fluid (HTF) at a certain operating conditions [28]. Large-scale simulations conclude that the overall CSP performance could be improved if concrete is used as TES medium instead of the conventional two-tank, indirect, molten salt TES [29]. Molten salts are commonly used in modern CSP technology, specifically the NaNO3 (60 wt%) -KNO3 (40 wt%) mixture [30]. The main drawback of using molten nitrates is that the operating TES are composed of metallic materials. These large TES containers require the application of Ni-based superalloys or austenitic stainless steels, which are very expensive and, in addition, present corrosion problems within approximately 30 years, compromising the mechanical structure of the tanks [31]. For this reason, PC concrete is a promising avenue for developing clean energy due to its simplicity of installation and operation, low investment cost, modularity and scalability of plants from small to large, and the possibility of commercial deployment because of the basic materials needed (steel and concrete) [29]. An improvement in the TES systems based on PC not only reduces the storage volume and the area of the solar power plant, but also increases the capacity factor of the solar power plant [24].

However, because of the serious environmental drawbacks of PC and NA mentioned earlier, it is necessary to develop new alternatives based on the principles of the circular economy using waste and industrial byproducts. Thus, in this study, AAM and HM were studied from a mechanical and environmental point of view, to improve the sensible solid thermal storage systems. This work studies the effect in the systems after their exposure to high temperatures, between 300 °C and 500 °C, due to parabolic trough collectors are currently the most utilized CSP technology (in approximately 95% of CSP installations) [30] with operation temperatures between 290 °C and 390 °C when molten salts are used [32]. Mechanical tests after the exposure of the mortars to temperature provides information about the durability of the alternatives to work as TES media in CSP, as a recent study showed that the heat transfer within the blocks was higher in the alternatives than in the conventional PC systems [4]. There is an existing body of research about the environmental impacts of PC, NA, AAM and HM in terms on mass [3,33]. However, very little is known about the environmental impacts of effectively storage energy in those mass systems. To determine if the alternatives are truly viable from an environmental point of view, the mass required to store a specific unit of energy was calculated to compare the environmental impacts of the energy storage in the new alternatives and to measure it against PC concrete. To this aim, in our work, we selected carbon footprint and water use as the environmental impact categories to analyze by means of LCA methodology.

2. Material and methods

2.1. Materials and sample preparation

Two raw materials were used as binders for the mortars. Portland cement I 42.5R, supplied by the company "Cementos Portland Valderrivas", and an industrial by-product, a blast furnace slag from ENSIDESA (Avilés, Spain). Focusing on the aggregates to substitute NA in all the systems, sand (S) was replaced in percentage by weight with glass waste (GW) from "Ecovidrio" (Ajalvir, Spain). Furthermore, to resemble the material that was replaced, GW was ground using a cross beater mill. Thus, the particle size used, as in the sand, was under 2 mm.

Table 1 shows the main chemical composition of the raw materials (PC and SLAG), where the content of CaO, SiO_2 and Al_2O_3 is a majority in both of them. The GW chemical composition is also shown. It can be observed that GW is mostly composed of SiO_2 .

Table 1

Chemical composition of the raw materials (%wt) by XRF.

%wt	CaO	SiO ₂	Al_2O_3	MgO	Fe ₂ O ₃	SO_3	Na ₂ O	K ₂ O	*LoI
Portland cement Blast furnace slag	61.94 35.73	21.28 36.15	6.45 11.75	<0.003 12.75	2.53 0.38	5.87 1.75	<0.012 <0.010	1.01 0.27	2.35 2.10
GW	11.75	70.71	2.05	1.17	0.52	-	11.71	1.08	0.83

*LoI: loss on ignition.

Portland cement and blast furnace slag were used separately in order to fabricate the reference sample (PC) and the AAM alternative (SLAG), and together to manufacture the HM system. All mortars have been prepared under the UNE EN 196–1 standard:

- PC mortar (reference sample referred to as PC): composed of Portland cement CEM I 42.5R and aggregate (ratio 1:3 respectively). Water was used for its hydration.
- AAM mortar (referred to as SLAG): blast furnace slag and aggregate (1:3 ratio) were used. It was activated with a commercial solution of sodium silicate (waterglass) that was diluted with water and NaOH to achieve a SiO₂/Na₂O ratio equal to 0.8.
- Hybrid slag mortars (referred to as HSLAG 5%): 80% weight of slag and 20% weight of CEM I 42.5R. In addition, the HM systems contain a soft alkaline activator, specifically, 5% of Na₂SO₄ was added to accelerate the hydration process [11]. These systems were also prepared with a precursor/aggregate ratio of 1:3.

The liquid/solid ratios (L/S) were decided following the UNE EN 1015–6 standard [34] which establishes that mortars must have the same workability [35]. Table 2 shows in detail the components of each manufactured mortar and their required quantity. The sand was gradually replaced, by 25 wt%, with GW.

2.2. Equipment

After curing the mortars for 28 days, their properties were measured:

- Compression tests were carried out using the Microtest universal testing machine with the 200 kN load cell to register the necessary force to cause compression failure of the specimen.
- Specific heat (Cp) of mortars was measured using differential scanning calorimeter (DSC) equipment, model 822 (Mettler Toledo GmbH, Greifensee, Switzerland). Each sample weighed approximately 15 mg and went into aluminum crucibles of 40 μ l. The samples were heated in the temperature range of 0–400 °C and nitrogen (35 ml/min flow) was used as purge gas. Two ramps were performed from 0 to 400 °C to calculate Cp after the complete evaporation of water. The heating rate was established at 10 °C/min and liquid nitrogen was used to cool the samples. Three different measurements were carried out in order to obtain an average.
- To calculate Cp after a first heating and in the range of 290–390 °C, it was necessary to study previously the mass losses when mortar is exposed to high temperature. Differential thermal analysis (DTA, SETSYS Evolution, SETARM Instrumentation, France) was carried out to study mass losses because of the decomposition of phases. DTA/TG were tested from 50 °C to 500 °C with a heating rate of 2 °C/min.

2.3. Samples treatment before compression test

• To measure the mechanical properties, the mortars were poured into rectangular molds (4x4x16 cm). Samples were tested without any treatment and after their exposure to high temperatures (300 °C, 400 °C and 500 °C) (Fig. 1). Exposure to 100 °C and 200 °C before the highest temperatures was done to avoid the spalling risk. This was higher in the first heating due to the thermal gradients between the

Table 2	2
Mortar	preparation

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Nomenclature	Raw material (g)	Addition	L/S	Water (g)	Activator solution (g)
PC 100 S	450-PC	-	0.50	225- H ₂ O	_
PC 75 S – 25 GW	450-PC	-	0.57	256.5- Н ₂ О	-
PC 50 S – 50 GW	450-PC	-	0.60	270- Н ₂ О	-
PC 25 S – 75 GW	450-PC	-	0.62	279- Н ₂ О	-
PC 100 GW	450-PC	-	0.64	288- H ₂ O	-
SLAG 100 S	450-SLAG	_	0.55	-	247.5- Na ₂ SiO ₃ Solution
SLAG 75 S – 25 GW	450-SLAG	_	0.54	-	243- Na ₂ SiO ₃ Solution
SLAG 50 S – 50 GW	450-SLAG	_	0.65	-	292.5- Na ₂ SiO ₃ Solution
SLAG 25 S – 75 GW	450-SLAG	-	0.73	-	328.5- Na $_2$ SiO $_3$ Solution
SLAG 100 GW	450-SLAG	-	0.76	-	342- Na ₂ SiO ₃ Solution
HSLAG 5% 100 S	348.75-SLAG; 78.75-PC; 22.5-Na ₂ SO ₄	5% Na ₂ SO ₄	0.46	207- Н ₂ О	-
HSLAG 5% 75 S – 25 GW	348.75-SLAG; 78.75-PC; 22.5-Na ₂ SO ₄	5% Na ₂ SO ₄	0.48	216- H ₂ O	-
HSLAG 5% 50 S – 50 GW	348.75-SLAG; 78.75-PC; 22.5-Na ₂ SO ₄	5% Na ₂ SO ₄	0.51	229.5- Н ₂ О	-
HSLAG 5% 25 S – 75 GW	348.75-SLAG; 78.75-PC; 22.5-Na ₂ SO ₄	5% Na ₂ SO ₄	0.55	247.5- Н ₂ О	-
HSLAG 5% 100 GW	348.75-SLAG; 78.75-PC; 22.5-Na₂SO₄	5% Na ₂ SO ₄	0.58	261- Н ₂ О	-

surface and the core and because of the water vapor release which causes pressure in the material [36]. Five specimens were used for each compression test.



Fig. 1. Exposure time to different temperatures.

2.4. LCA methodology

LCA is a useful standardized methodology for assessing the potential environmental impacts of a product, practice or a process throughout its whole life cycle [37,38]. Different research studies highlight the usefulness of this tool to evaluate the environmental and energy performance of a material/product during its lifespan [39,40]. LCA provides an overview of the potential environmental damage considering associated upstream (e.g. extraction of raw materials) and downstream (e.g. use of waste) processes.

Following data reported in previous research [3], the carbon and water footprints were analyzed, following the ISO LCA phases, as indicators of the environmental impact of the alternative mortars, and were compared with the conventional systems. Carbon footprint and water use were the two indicators analyzed in this study due to their environmental importance and the motivation to improve and to optimize those alternative mortars, for example, replacing sand after finding that the NA was the component having the highest impact on the water footprint.

ISO 14040/44 standards [41] establish four phases to be accomplished: (i) goal and scope definition; (ii) life cycle inventory (LCI); (iii) life cycle impact assessment phase and the (iv) interpretation phase. The four phases are described below.

(i) Goal and scope definition: the intended application, system boundaries and the functional unit are defined. This study evaluates the environmental impacts of the production of alternative mortars (SLAG and HSLAG 5% systems) to compare with the reference PC system. To enable comparison of the mortars, 1 MJ of stored energy was considered as the functional unit. In previous studies with these types of systems, as is common practice, a unit of mass was used to compare the mortars [3]. However, in this case, the unit of mass would not be the most accurate way because for a unit of mass each mortar would store a different amount of energy. Thus, a unit of stored energy was chosen, which is what really matters in a material when using a system such as TES. The system boundaries were limited to a cradle to gate approach.

(ii) LCI consists of identifying and quantifying the inputs and outputs (i.e. energy, raw materials...) needed to define the functional unit In this case, 1 MJ of stored energy in a mortar. Table 3 shows the LCI of the four systems (Fig. 2) which present the highest and more acceptable mechanical properties after the compression test. Material flows were directly extracted from experimental results. To model these flows, datasets were taken from the Ecoinvent 3.6 database [42]. The use of common waste (such as glass) and industrial by-products (blast furnace slag) were modelled as avoided products, thereby following a consequential approach. This approach was chosen because the alternatives are governed by the principle of circular economy: recovering waste materials to include them in new products.

(iii) Life cycle impact assessment: characterization and assessment of the environmental impacts. For this purpose, environmental threat values were calculated with SimaPro 9.1 software. SimaPro can model a consequential approach by including avoided material in the input/ output section. Modelling in this way allows slag or GW to be introduced as waste instead of commercial material. In our work, the LCA study is focused on two indicators: the Global Warming Potential (GWP 100) and the Available Water Remaining (AWARE).

GWP was developed by the Intergovernmental Panel on Climate Change (IPCC) in 2007 and is updated continuously [43]. GWP is measured in kg CO₂ equivalent (eq.) which is a relative measure of how much heat can be absorbed by a given GHG over a period. For this study, IPPC 2013 was used with the climate change factors of IPCC for a period of 100 years, sufficient time for evaluating the cumulative effects of GHG on the environment. GWP is also known as the carbon footprint (CF) and is one of the most frequently reported environmental indicators.

Regarding the water footprint, an agreement has been reached to recommend an indicator for calculating the environmental impact of water use by the United Nations Environment Programme (UNEP) and the Society of Environmental Toxicology and Chemistry (SETAC) [44]. Specifically, AWARE is the recommended method for water scarcity footprint assessments [45]. It is a water use midpoint indicator, which represents the Available Water Remaining per area after human and ecosystem requirements have been satisfied, thus assessing the potential of water deprivation. The characterization is done using the water consumption needed in the product life cycle (inventory data), as well as the regionalized characterization factors[46], as can be seen in Equation (1):

$$Water use[m^{3}] = \sum_{i} LCI_{i}[m^{3}] \cdot \frac{AMD_{world}[m^{3} \cdot m^{-2} \cdot month^{-1}]}{AMD_{region i}[m^{3} \cdot m^{-2} \cdot month^{-1}]}$$
(1)

where AMD refers to the Availability Minus the Demand. Specifically, the AMD world average and the AMD of each region is used, where the term availability is used in the sense of precipitation minus evaporation [45]. Every input presents its LCI extracted from Ecoinvent database focused on a specific geographic location. Consequently, the region used in the present paper for water is Europe without Switzerland.

iv) Interpretation: in this last step, significant issues, conclusions and recommendations are identified.

3. Results and discussion

3.1. Mechanical properties

Fig. 2 shows the results of compression strength for the three systems evaluated: reference mortar (PC system) and the alternative mortars, SLAG and HSLAG 5% at different percentages of sand substitution by GW.

The three systems present a similar behavior with GW addition. As the content of GW is increased, the compressive strength values decrease, which is consistent with the literature [47–49]. This is produced due to the weak bonding between the binder and GW [48] and also because of the increase of the L/S ratio (Table 2), which generates porosity, when GW is increased [49].

Table 3

LCI for the four systems with the best mechanical performance, for 1 MJ of stored energy.

Ecoinvent 3.6 Dataset			Environmental inventory values			
				GWP [kg CO ₂ eq/kg]	Water use [m ³ /kg]	
PC 100 S Cement	1 1.30	MJ kg	Cement, Portland {Europe without Switzerland} market for	0.88	0.0484	
asses	0.65	kg	APOS, U Tap water {Europe without Switzerland}	0.000347	0.0206	
Sand	3.90	kg	market for APOS, U Sand {RoW} market for sand APOS, U	0.0114	0.047	
SLAG 100 S	1	MJ				
Slag	1.37	kg	Cement, Portland {Europe without Switzerland} market for APOS, U (as avoided product)	-0.88	-0.0484	
Tap water	0.48	kg	Tap water {Europe without Switzerland} market for APOS, U	0.000347	0.0206	
NaOH	0.07	kg	Sodium hydroxide, without water, in 50 % solution state {RER} chlor-alkali electrolysis, membrane cell APOS, U	0.824	0.981	
Na2SiO3	0.20	kg	Sodium silicate, solid {RER} market for sodium silicate, solid APOS, U	0.759	0.328	
Sand	4.10	kg	Sand {RoW} market for sand APOS, U	0.0114	0.047	
SLAG 75 S – 25 GW	1	MJ				
Slag	1.18	kg	Cement, Portland {Europe without Switzerland} market for APOS, U (as avoided product)	-0.88	-0.0484	
Tap water	0.41	kg	Tap water {Europe without Switzerland} market for APOS, U	0.000347	0.0206	
NaOH	0.06	kg	Sodium hydroxide, without water, in 50 % solution state {RER} chlor-alkali electrolysis, membrane cell APOS, U	0.824	0.981	
Na2SiO3	0.17	kg	Sodium silicate, solid {RER} market for sodium silicate, solid APOS, U	0.759	0.328	
Sand	2.65	kg	Sand {RoW} market for sand APOS, U	0.0114	0.047	
Glass waste	0.88	kg	Sand {RoW} market for sand APOS, U (as avoided product)	-0.0114	-0.047	
HSLAG 5% 100 S	1	MJ				
Slag	0.93	kg	Cement, Portland	-0.88	-0.0484	

{Europe without

Table 3 (continued)

Ecoinvent 3.6 Dataset				Environmental inventory values			
				GWP [kg CO ₂ eq/kg]	Water use [m ³ /kg]		
			Switzerland} market for APOS, U (as avoided product)				
Cement	0.21	kg	Cement, Portland {Europe without Switzerland} market for APOS, U	0.88	0.0484		
Tap water	0.55	kg	Tap water {Europe without Switzerland} market for APOS, U	0.000347	0.0206		
Na ₂ SO ₄	0.06	kg	Sodium sulfate, anhydrite {RER} market for APOS, U	0.701	0.887		
Sand	3.61	kg	Sand {RoW} market for sand APOS, U	0.0114	0.047		

The comparison between the reference mortar and the alternative ones follows the same tendency observed in previous studies [3,4]. The SLAG mortar activated with sodium silicate shows the best performance at the age of 28 days, especially, without any GW substitution (Fig. 2). Specifically, the SLAG 100 S system reaches resistance values up to 67 MPa which means that this alternative system achieves a 64% higher compressive strength value than the PC reference system (without sand substitution, i.e. PC 100 S). This improvement in the results, compared to the PC mortar, is due to the use of waterglass solution, which contains silicon, as an alkaline activator. The presence of slag as base material causes the formation of the C-A-S-H gel. Moreover, the addition of silicon makes this main hydration product rich in silicon, thus providing high cohesion and polymerisation, as was observed in different studies [9,50]. Although the use of GW reduces the mechanical properties, the substitution of 25% by weight of sand in the SLAG system (SLAG 75 S -25 GW) provides a strength value of 41 MPa, which is the same result obtained in the reference system without GW (PC 100 S). This proves once again the highly cohesive material obtained when slag is activated with sodium silicate.

Regarding HSLAG 5% systems, the simplest one without any GW substitution shows a compressive strength value of 35 MPa, which represents a reduction of only 13% compared to the basic PC mortar. This slight decrease is due to the formation of the main reaction products: the consistency of the mixture of gels in the HSLAG 5% mortar, C—S—H (due to PC) and C—A—S—H (generated by slag), is lower than the consistency of the lesser portion of PC used in the HSLAG 5% system.

Comparing the alternatives, there are clear differences in their mechanical properties. The HSLAG 5% system shows lower compressive strength values. Although both of them have C—A—S—H gel in their structure, their compositions are different, and gel in HSLAG 5% is not as consistent and cohesive as the one that appeared in the SLAG mortar. This is due to the hydration, whereas SLAG used an alkaline solution rich in silicon, HSLAG 5% systems are hydrated just with water thanks to the addition of a soft activator (sodium sulfate).

Although the values of HSLAG 5% 100 S are lower than those of PC 100 S and SLAG 100 S mortars, their resistance values are very close to the ones of the reference sample (PC 100 S). These results should be considered, as HSLAG 5% mortars only use 20% of PC and, in addition, solutions such as waterglass are avoided.

After verifying the feasibility of NA substitution, the following mortars with good mechanical properties were selected: PC 100 S, SLAG 100 S, HSLAG 5% 100 S and SLAG 75 S – 25 GW. To analyze the thermal behavior of these four systems, all of them were exposed for 24 h, to



Fig. 2. Variation of compressive strength with the replacement of sand by GW.

ensure the temperature homogeneity throughout the material, at three different temperatures: at 300 $^{\circ}$ C, 400 $^{\circ}$ C and 500 $^{\circ}$ C (typical operating temperature in a CSP power plant), following the cycles in Fig. 1. Fig. 3 shows the compressive strength values of the four mortars after 24 h exposure to high temperatures.

First, as it can be observed in Fig. 3, after their exposure to 300 °C, the compressive strength values increase with respect to the untreated mortars in the PC 100 S (9.3%), SLAG 75 S – 25 GW (19%) and HSLAG 5% 100 S (17.2%) systems, while in the SLAG 100 S system (a drop of 3%), the temperature effect at 300 °C is not relevant. This phenomenon

can be explained by the polycondensation between tetrahedral aluminosilicate gels [51] and also by new binders that can be created with what is known as a "high-temperature curing", where an internal autoclaving is produced due to the evaporation of water that take place between sheets of the gel. In other words, the gradual transportation of the residual moisture allows an accelerated hydration at the early stage of heating mortars to high temperatures [52–55]. Furthermore, in the case of PC, it was seen that there is an increase of the Van der Waals forces during evaporation because the cement gel layers get closer [52] and strain behavior occurs in the cement as a result of the expansion of



Fig. 3. Compressive strength values of the four selected mortars after exposure to 300 °C, 400 °C and 500 °C.

the non-hydrated phases and a shrinkage of the hydrated phases [54].

The SLAG 100 S system maintains its mechanical properties almost constant regardless of the thermal treatment, always providing the highest results. Specifically, its resistance value goes from 67 MPa to 64 MPa (approximately 5% reduction). Up to 500 °C, there is an improvement in the mechanical properties with respect to PC 100 S of almost 79% (Table 4). This good performance derives from the highly cohesive gel (C—A—S—H gel).

The system that includes GW in its composition (SLAG 75 S – 25 GW) shows behavior similar to the PC 100 S system, an increase in their mechanical properties up to 300 °C and then, a decrease of the resistances inversely proportional to the temperature increase. It is worth mentioning that the SLAG 75 S – 25 GW system reaches higher compressive strength values than the PC 100 S system for each thermal treatment (Table 4); the improvement of SLAG 75 S – 25 GW compared to the reference sample is 10% at 300 °C, 15% at 400 °C and 6% at 500 °C.

As regards the HSLAG 5% 100 S system, although the compressive strength values at 300 and 400 °C are still slightly lower than those of the PC system as in the case of the untreated samples, at 500 °C both systems present a resistance value of 35 MPa. The HSLAG 5% 100 S seems to stabilize in the range of 400 to 500 °C. In fact, it is the mortar showing the least reduction after the thermal treatments, not even reaching 1%.

The decrease in the PC and HSLAG 5% values may be related to the dehydration and destruction of CSH gel [51] and to the Ca(OH)₂ decomposition that occurs at about 400 °C [56]. In the case of the SLAG 75 S – 25 GW system, the decrease is due to the formation of microcracks because of the thermal incompatibility between the binder and the aggregates (GW and sand in this case) at the interface zones [51].

3.2. LCA study

Once the operational feasibility of systems such as TES has been demonstrated, the environmental impacts of the most suitable systems are studied to also check their environmental efficiency.

As stated above, our work is focused on the comparison of the mass needed for each system, to store 1 MJ of energy. For this purpose, the DSC method was carried out to calculate the values of the specific heat capacity (Cp) of each system using Equation (2):

$$Q = \int_{T_{min}}^{T_{max}} mCpdT \tag{2}$$

where Q is the heat energy, m is the mass of the sample that was introduced in the DSC equipment, Cp is the specific heat of the mortar, T_{max} is the maximum operating temperature and T_{min} the minimum operating temperature.

Thus, Cp was calculated using DSC (three different measurements of

Table 4

Comparison of the alternative mortars to the reference sample (PC 100 S) after their exposure at different temperatures.

each mortar were carried out to provide an average value) in the range of 290 °C (T_{min} - minimum operating temperature in a CSP technology which works with molten salts) and 390 °C (T_{max} - maximum operating temperature in a CSP technology with molten salts).

Prior to the DSC measurement, TGA was used to calculate the mass loss produced by the decomposition of the phases that could be produced from 0 to 400 °C. Mass loss percentages after the exposure of the mortars to 400 °C appear in Table 5. Most of the mass loss occurred above 100 °C, due to the evaporation of the water contained in the mortars.

Then, in the DSC equipment, a first heating ramp was carried out from 0 to 400 °C to eliminate the possible decomposition that could occur in the mortars as mentioned above. Cp values were calculated using the parameters of Equation (2) extracted from the second heating ramp, when the mass was stable, in the range of 290 °C to 390 °C. Cp results are shown in Table 6. With those Cp values, the mass necessary to store 1 MJ in the operational temperatures range (290 °C – 390 °C) is shown in Table 7.

The necessary mass values to store 1 MJ (shown in Table 7) were considered to identify and to quantify the inputs needed (Table 3) to model the different systems in order to study their carbon and water footprint. Thus, Table 8 shows the numerical results for water use and GWP to store 1 MJ in each type of mortar: PC 100 S, SLAG 100 S, SLAG 75 S – 25 GW and HSLAG 5% 100 S.

To compare the alternatives with the reference sample (PC 100 S), the relative environmental impacts have been plotted in Fig. 4.

Regarding GWP, the results presented in Fig. 4 and Table 8 show that all the alternatives provide very significant improvement over the PC reference system. Specifically, the new developments obtain negative results, which means environmental benefits. Particularly, the AAM systems obtain the highest negative results. Due to the increased use of a by-product (slag), avoiding its accumulation in landfills, the SLAG 100 S system shows the highest improvement, by more than 180%, followed by the SLAG 75 S – 25 GW system with an environmental enhancement of 171%. This decrease in the AAM systems is because the SLAG 75 S – 25 GW mortar does not incorporate the same amount of by-product and furthermore, the substitution of sand by GW does not improve the carbon footprint as much, due to the low contribution of sand to CO_2 emissions. For the hybrid system (HSLAG 5% 100 S), the carbon

Table	5
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Mace	loss porcont	ago aftar ite	ovpocuro	to 400 °C	
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System	Mass loss percentage after its exposure to 400 $^\circ\text{C}$ (%)
PC 100 S	4.5
SLAG 100 S	5.6
SLAG 75 S – 25 GW	5.1
HSLAG 5% 100 S	5.6

	PC 100 S	SLAG 100 S		SLAG 75 S – 25 GW	r	HSLAG 5% 100 S		
	Compressive strength (MPa)	Compressive strength (MPa)	Increase in compressive strength (%)	Compressive strength (MPa)	Increase in compressive strength (%)	Compressive strength (MPa)	Increase in compressive strength (%)	
Without treatment	41.0	67.5	+ 64.6	41.4	+ 1.0	35.4	- 13.7	
Т 300	44.8	65.5	+ 46.2	49.3	+ 10.0	41.5	- 7.4	
т 400	39.1	62.5	+ 59.8	45.3	+ 15.9	35.7	- 8.7	
Т 500	36.0	64.4	+ 78.9	38.2	+ 6.1	35.3	- 1.9	

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Table 6

Cp of the four systems selected.

System	Cp (J/kg °C)
PC 100 S	1710
SLAG 100 S	1602
SLAG 75 S – 25 GW	1867
HSLAG 5% 100 S	1861

Table 7

Necessary mass to store 1 MJ.

-	
System	m (kg)
PC 100 S SLAG 100 S SLAG 75 S – 25 GW	5.85 6.24 5.36
HSLAG 5% 100 S	5.37

footprint improvement is more than 146%. The percentage reduction is because, in contrast to AAM, the HM use PC in their systems.

Focusing on the total water use calculated following the AWARE method, as shown in Table 8, the value for the reference sample (PC 100 S) was 0.25 m^3 , 0.27 m^3 for the SLAG 100 S and 0.15 m^3 and 0.19 m^3 for the SLAG 75 S – 25 GW and HSLAG 5% 100 S, respectively. On the one hand, when SLAG 100 S is used, more water is needed compared to the reference sample (an 8%). This fact is due to the larger amount of mass required (in particular sand, as will be noted later) in order to store 1 MJ. On the other hand, a 40% benefit is obtained in water use if SLAG 75 S – 25 GW is chosen instead of PC 100 S and 24%, if HSLAG 5% 100 S is considered. Both systems store 1 MJ in a lower mass compared to PC 100 S and include by-products in their composition. Moreover, the SLAG 75 S – 25 GW system avoids the use of sand (the component producing the highest impacts on the water footprint), hence its major improvement.

Fig. 5 shows the contribution in percentages for each mortar component to the overall GWP and water use for the four systems. In addition, the numerical results associated to each contribution can be observed in Table 8.

In the PC 100 S system (Fig. 5 (a)), the main contribution to the GWP is derived from Portland cement, specifically, $1.14 \text{ kg CO}_{2 \text{ eq}}$ of the total $1.18 \text{ kg CO}_{2 \text{ eq}}$ are due to this component. This means a contribution of more than 96%. In contrast, the contribution of the sand is more than 3% and therefore the tap water contribution to the GWP is negligible. The water use situation is completely different. As mentioned earlier, the component with the most water consumption is sand rather than the PC. The NA contributes to the water use by more than 70%, followed by the PC with about 24% and then, tap water with just over 5%.

Because of the negative consequences on the environment from the PC 100 S mortar (the conventional construction material), it is important to develop new eco-efficient alternatives which can specifically curb climate change. For this purpose, the focus is on reducing the carbon footprint by totally or partially substituting the PC using the aforementioned alternatives (AAM and HM materials). Removing all PC can be done using AAM systems. Fig. 5 (b) and (c) represents the environmental contribution of the components used in the two selected AAM mortars. The two main differences between SLAG 100 S and SLAG 75 S -25 GW is that more mass is needed in the first case to store the same energy (1 MJ), and the replacement of sand by GW in the second case. This is due especially to the water use because more sand is necessary in the case of the SLAG 100 S, leading to consumption of 0.19 m³ of the total 0.27 m³. Whereas in the SLAG 75 S – 25 GW, in addition to requiring less mass, the NA (component with the highest impact on water consumption) is replaced by GW, consuming 0.12 m³ of the sand out of 0.15 m³. The rest of the components display the same behavior. As for the GWP, the component that affects the carbon footprint the most is sodium silicate, followed by sodium hydroxide (both elements are used to prepare the alkaline solution). This tendency is the same for the water footprint, excluding the sand, which affects it the most. As in the previous case, tap water is negligible for both indicators in the AAM systems.

With regard to the HM mortar (HSLAG 5% 100 S), Fig. 5 (d) shows that the low percentage of PC mainly affects the GWP. Sand and sodium sulfate are the other components that have a positive environmental impact in GWP, but their contributions are limited to about 5%. As in the previous cases, the water use is mainly driven by the NA, but now, the soft alkaline activator, sodium sulfate, also affects water consumption to a lesser extent.

Although there is a need to introduce new components to develop the alternatives, specifically those that activate the precursors (alkaline solutions and soft alkaline precursors) which have a positive impact on the GWP (Fig. 5 (b), (c) and (d)), the total carbon footprint of the energy



Fig. 4. Relative environmental profile (%) of the case studies.

Table 8

LCA results (water use and carbon foot	print	of the mass needed to store 1 M	MJ o	of energy	y in each of the	4 mortar systems.
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Units	PC 100 S		SLAG 100 S		SLAG 75 S $-$ 25 GW		HSLAG 5% 100 S	
	Water use m ³	IPCC GWP 100a kg CO _{2 eq}	Water use m ³	IPCC GWP 100a kg CO _{2 eq}	Water use m ³	IPCC GWP 100a kg CO _{2 eq}	Water use m ³	IPCC GWP 100a kg CO _{2 eq}
Sand	0.18	0.04	0.19	0.05	0.12	0.03	0.17	0.04
Tap water	0.01	2.10^{-4}	0.01	1.10^{-4}	0.01	1.10^{-4}	0.01	2.10^{-4}
NaOH			0.07	0.06	0.06	0.05		
Na ₂ SiO ₃			0.07	0.15	0.06	0.13		
Slag			-0.07	-1.21	-0.06	-1.04	-0.05	-0.82
Na_2SO_4							0.05	0.04
Glass waste					-0.04	-0.01		
Total	0.25	1.18	0.27	-0.95	0.15	-0.84	0.19	-0.55



Fig. 5. Contributions of the different mortar components to the environmental impacts for a) PC 100 S, b) SLAG 100 S, c) SLAG 75 S – 25 GW and d) HSLAG 5% 100 S.

stored in these proposed mortars is negative which indicates a great benefit. These results are in line with previous research where the functional unit in volume of other eco-efficient systems were compared [3]. In terms of water consumption, it is important to highlight the significant benefits in the water footprint stemming from the NA replacement.

4. Conclusions

The main purpose of this work was the study of eco-efficient mortars from a mechanical and environmental point of view to provide further knowledge on their use as TES media. For that purpose, the following conclusions can be drawn from our work:

- AAM mortars are an alternative to PC systems from a mechanical point of view. The C—A—S—H gel provides significant compressive strength results without any heat treatment and after its exposure to high temperatures due to its cohesion and polymerisation.
- The high resistance of the AAM allows the substitution of NA by GW, resulting in a mortar with better mechanical properties than the PC system after exposure to high temperatures.
- Although the mechanical properties of the HM mortars are slightly lower than those of the PC reference sample at room temperature, after exposure to 500 °C, a stabilization of the compressive strength values of the HM systems occurs, which makes them competitive with the PC system.

- For energy storage, negative carbon footprints are obtained when the process recovers blast furnace slag, being the GWP for the SLAG 100 S mortar of -0.95 kg CO₂/m³, and -0.84 kg CO₂/m³ and -0.55 kg CO₂/m³ for the SLAG 75 S 25 GW and HSLAG 5% 100 S, respectively. In contrast, the GWP for the PC 100 S system is positive (1,18 kg CO₂/m³), which means a carbon footprint reduction of more than 100% if alternatives are used instead of the conventional PC.
- In terms of remaining water available, a 40% reduction in water use is achieved if GW is used instead of sand. Thus, the biggest improvement in water footprint is given by energy storage in the SLAG 75 S – 25 GW system.

Although PC is currently proving to be a promising material to use it as TES, this study has found that its alternative materials, hybrid and alkali-activated materials, can offer better mechanical performance after exposure to high temperatures as well as more environmentally friendly impacts.

Therefore, alternative materials to PC systems can be considered for use as TES media opening new gates towards use of renewable energies.

CRediT authorship contribution statement

I. Ramón-Álvarez: Conceptualization, Investigation, Resources, Formal analysis, Data curation, Writing – original draft. E. Batuecas: Conceptualization, Methodology, Formal analysis, Software, Writing – original draft, Writing – review & editing. S. Sánchez-Delgado: Conceptualization, Writing – review & editing, Validation, Supervision, Funding acquisition. **M. Torres-Carrasco:** Conceptualization, Resources, Writing – original draft, Validation, Supervision, Funding acquisition, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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