PRE-PACKED ALKALI ACTIVATED CEMENT-FREE MORTARS FOR REPAIR OF EXISTING MASONRY BUILDINGS AND CONCRETE STRUCTURES

Coppola[1] L, Coffetti[2] D, Crotti[3] E

[1,2,3] Department of Engineering and Applied Sciences, University of Bergamo, Italy

[1,2,3] Consorzio INSTM, UdR “Materials and Corrosion”, Florence, Italy

[1] *luigi.coppola@unibg.it*

[2] *denny.coffetti@unibg.it*

[3] *elena.crotti@unibg.it*

ABSTRACT

This paper is aimed to study a ground granulated blast furnace slag activated with alkali powder to manufacture Portland-free mortars for conservation, restoration and retrofitting of existing masonry buildings and concrete structures. Activator/precursor represents the key parameter – not only for elasto-mechanical performances – influencing the rheological properties and the shrinkage: the higher the activator dosage, the higher the consistency class and shrinkage. Moreover, elastic modulus of slag-based mortars is lower than that of OPC-mortars at the same strength class. AAMs seem to be more promising for a sustainable future in construction since the GER and GWP are reduced by about 80 % compared with traditional Portland cement mortars with the same compressive strength.

KEYWORDS

Alkali-activated materials; Ground-Granulated Blast-Furnace Slag; cement-free mortars; Sustainability; Gross Energy Requirement; Global Warming Potential

HIGHLIGHTS

- Sustainable alkali-activated slag-based mortars were studied
- The influence of powder activator-dosage was investigated
- Rheological, physical and mechanical properties are affected by activator dosage
- Compressive strength is “tailored” by changing the activator/precursor ratio

INTRODUCTION

One of most important challenge of twentieth century is to achieve a sustainable development model, especially in the building industry. The main challenge is to support the growth of the population, and the subsequent industrialization and urbanization, protecting the environment and reducing the energy consumption and natural resources [1]. Today, Portland cement is the most widely used construction material in the world. The consumption of this material is increasing dramatically in developing countries and expected that, by 2020, the global demand would increase by approximately 400 % by 2050 [2]. Production of Portland cement is energy-intensive (requiring kiln temperatures of 1450-1550°C), consumes 1,5 tons of raw materials each ton of Portland clinker [3,4] and is responsible for global warming, accounting for 7% of worldwide CO_2 emissions.

Sustainability in construction industry can be achieved through several options: a) use alternative fuels and/or alternative raw materials to reduce CO_2 emissions [5,6], b) replace Portland clinker to the greatest extent with low-carbon supplementary cementitious materials (SCM) in concrete production [7,8], c) develop alternative Portland-free low-carbon binders [9], d) reduce natural resources consumption increasing waste utilization in concrete manufacturing [10–13].

Reduction of energy consumption and pollutants emissions is achievable with the use of belite Portland cements, blustfurnace or pozzolanic cements [14], calcium-sulphoaluminate cements, CSA-based ternary binders [15], alkali-activated binders [16] and geopolymers [17]. Finally, depletion of natural resources can be avoided using in concrete production recycled aggregates from demolition of existing structures or industrial processes, replacing natural aggregates [18–20].

Among those above mentioned, alkali activated binders replacing Portland cement could represent in the next future a very promising solution to make the construction world more environmentally friendly. The first use of the alkali activation of aluminosilicate precursors in order to obtain an ordinary Portland cement alternative material is a patent of Kuhl in 1908. But it is only thanks to the studies of Glukhovsky in the 1950s that scientist and researchers began to talk about alkaline cements. Relevant changes took place in the 1970s with the findings of Davidovits [21] who coined the term “geopolymer” in 1979 having patented several aluminosilicate formulations. In 2009 Provis and van Deventer [22] summarized the state of art describing the process of transition from natural or synthetic powders to geopolymer-alumina-silicates. Alkaline cements are cementitious materials formed as the result of an alkaline attack on the amorphous or vitreous aluminosilicates. When mixed with alkaline activators, these materials set and harden, yielding a material with good binding properties. Different alumino-silicate materials can be activated by alkali, but ground granulated blustfurnace slag presents a lot of positive issues. In fact, the worldwide production of iron slag has been estimated around 300 – 360 million tons in 2015 [2]. Moreover, in recent years, due to the growth in the world iron production, the amount of slag has increased considerably. However, the vast majority of this slag is still disposed in landfills. For this reason, a slag-based activated binder represents an attractive alternative to disposal. However, a preliminary grinding of slag is needed to use the waste as precursor in Alkali Activated Mortars (AAMs) and concrete. The energy required to grind granulated blast furnace slag is only approximately 10 % of the total energy required for Portland cement production [14]. Hence, alkali-activated binder based on ground granulated blast furnace slag (GGBFS) represents an eco-friendly alternative to Portland cement due to its lower raw materials consumption and CO_2 emissions [23].

The purpose of the present study is the production of mortars based on ground granulated blast furnace slag activated by a mixture of sodium metasilicate pentahydrate, potassium hydroxide and sodium carbonate in powder form for multipurpose application, from plasters and renders to repair mortars for retrofitting and seismic upgrade of reinforced concrete elements.

MATERIALS AND METHODS

Materials

Ground granulated blast furnace slag with 28-day pozzolanic activity index equal to 0.76 (according to UNI EN 15167-1 and EN 196-5) as precursor and sodium metasilicate pentahydrate : potassium hydroxide : sodium carbonate = 7 : 3 : 1 in powder form as activator were used to produce different mortars with the dosage of activator between 2% and 32 % vs binder mass. The maximum activator/precursor ratio was limited to ensure both environmental and economic sustainability. The physical properties, laser granulometry and XRD analysis of GGBFS are reported in Table 1 and Figure 1 and 2.

The water was adjusted in order to attain the same workability at the end of the mixing procedure, equal to 160 mm ± 10 mm by means of a flow table. Furthermore, sand/binder ratio was fixed equal to 3 (maximum diameter of natural siliceous aggregates equal to 2.5 mm).

Table 1 – Physical properties of GGBFS

	D_{50} [µm]	**Specific surface [cm^2/g]**	**Specific mass [g/cm^3]**
GGBFS	12,42	3440	3,13

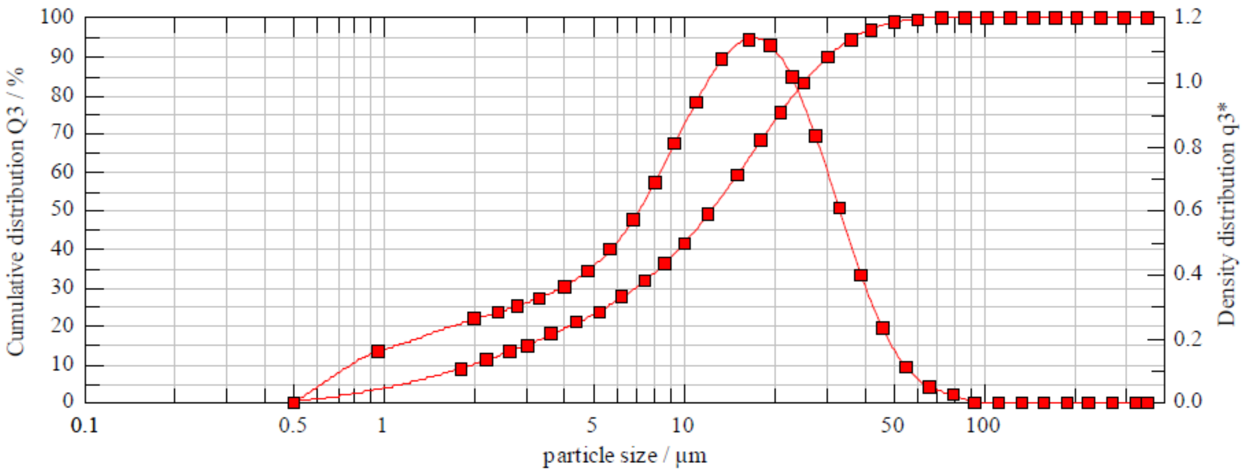


Figure 1 – Laser granulometry of Ground Granulated Blast Furnace Slag

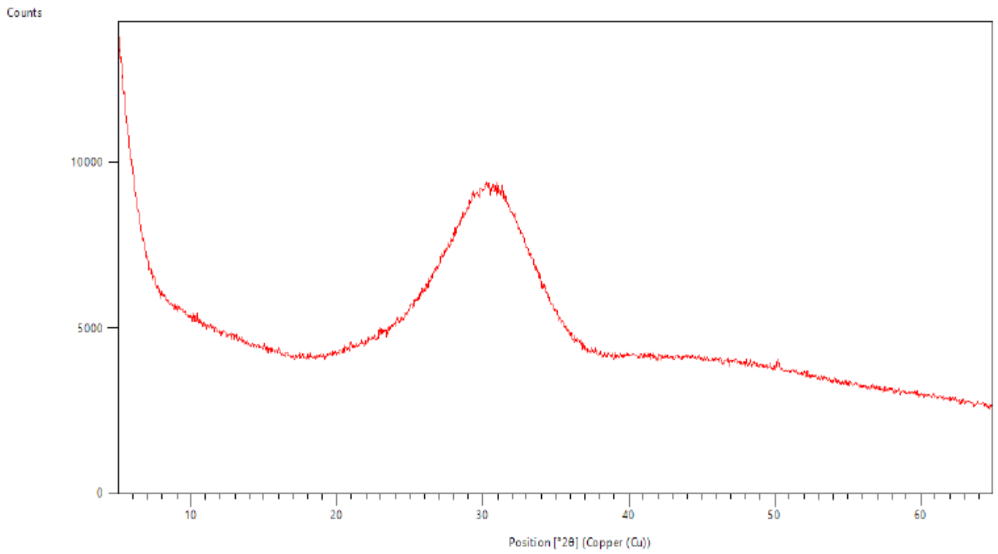


Figure 2 - XRD analysis of Ground Granulated Blast Furnace Slag

Tests on mortars

Workability was measured by means of flow table according to UNI EN 1015-3. The pot-life of the mixture, corresponding the time during which workability by flow table is higher than 140 mm, was also detected. In addition, specific mass on fresh mortars according to EN 1015-6 standard was evaluated. Moreover, pH of the solution obtained mixing the activator with the same amount of water required to produce the mortar was measured. Specimens 40x40x160 mm^3 were produced, cured for 24 hours in mold and stored in a climatic chamber at 20°C and R.H. 60%. Specific mass, compressive and flexural strength at 1, 7 and 28 days of mortars were also determined (EN 1015-11). Drying shrinkage was measured over time on prismatic specimens stored 24 hours after the mixing in a climatic chamber at a controlled temperature and humidity (T = 20°C, R.H. = 60%) according to EN 12617-4. In addition, optical microscopy observations were performed on AA slag- and OPC-based specimens in order to evaluate the micro-cracking formation in binder paste. Finally, elastic modulus (in accordance with method B, EN 12390-13) on 28-day cured cylindrical specimens was measured.

RESULTS AND DISCUSSIONS

Fresh mixture

Compositions and fresh properties of mortars are shown in Table 2. The amount of water to achieve 160 mm spreading depends on the percentage of activator. The water/precursor decreases if the dosage of alkaline activator is at least 4% (Figure 3 and 4). In other words, activator reduces the amount of water to obtain the target initial workability. In particular, the water reduction with respect the slag reference mortars without activator is equal to 15% and 25% when the activator dosage is 16 % and 32 %, respectively. This behavior is in accordance with Kashani et al. [24] that explain the plasticizing and deflocculating effects of sodium silicate on alkali activated slag-based paste with the increasing of the magnitude of repulsive double layer electric forces, causing the reduction in the yield stress at early ages.

Table 2 – Composition and properties of fresh mortars

Composition – Fresh properties	**S0**	**S2**	**S4**	**S8**	**S16**	**S32**
GGBFS [kg/m^3]	475	475	475	485	480	475
Activator [kg/m^3]	0	10	20	40	80	160
% Vs Precursor	0	2	4	8	16	32
Aggregate [kg/m^3]	1420	1420	1420	1460	1445	1430
Water [kg/m^3]	280	275	275	255	240	205
w/b	0.59	0.58	0.58	0.53	0.50	0.43
Flow [mm]	155	160	170	150	165	170
Density at fresh state [kg/m^3]	2170	2185	2185	2220	2240	2280
Pot-life [min]	>360 **	60	60	45	45	30
pH activator solution	7.00	13.26	13.43	13.61	13.70	13.84

Note: ** Pot-life time > 360 minutes

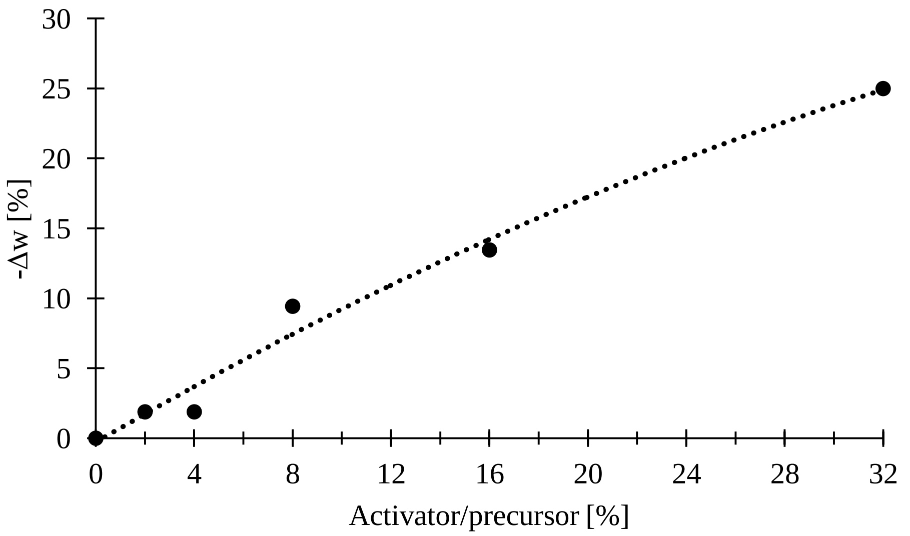


Figure 3 – Water amount reduction with respect to reference mortar as a function of the percentage of the activator, at equal workability

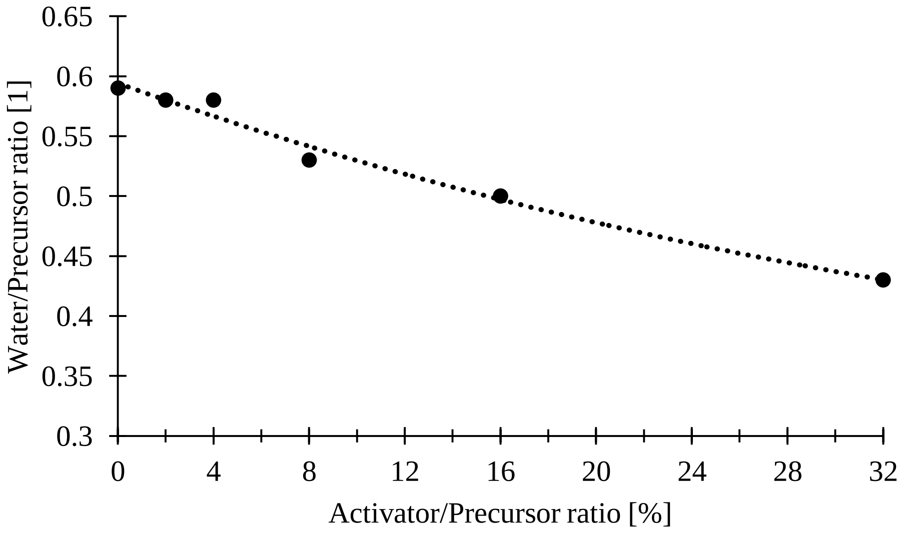


Figure 4 – Water/precursor ratio as a function of percentage of the activator, at equal workability

Pot-life of reference mortar (6 h) is dramatically shortened to 30 and 60 minutes from casting when the activator is added (Figure 5). The reduction of setting times is in agreement with Huanhai et al. [25] and Chang [26] that show as increasing the percentage of activator increases the released heat and shortens the peak time. In fact, a higher concentration of activator helps the resolution of calcium ions from the slag grains and consequently increases the reaction rate.

Moreover, no influence is observed in the fresh state on specific mass values similar to those of traditional Portland cement mortars at the same strength class. The difference between fresh and hardened specific mass decreases as the activator/precursor increases (Figure 6). This behavior could be ascribed to the higher amount of water that can evaporate in reference mixture without activator and in mortars manufactured with 2 and 4 % of the alkaline powder.

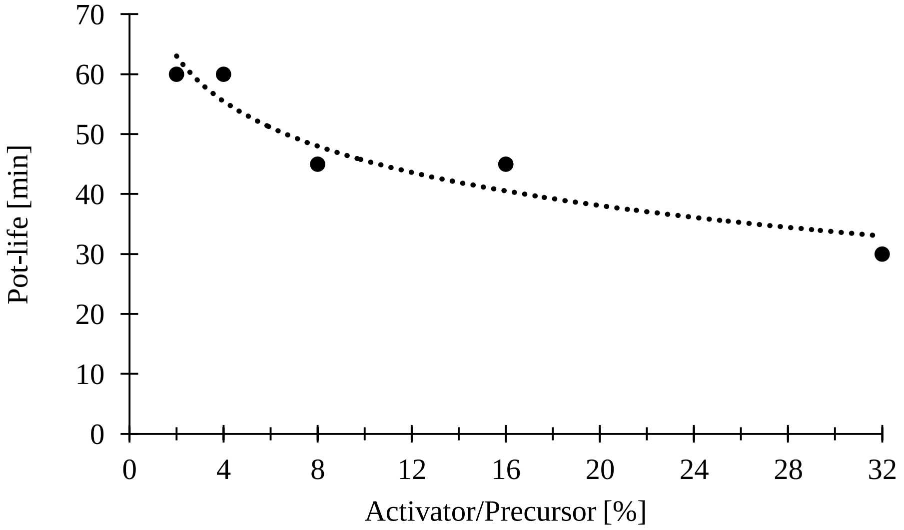


Figure 5 - Pot – life at selected activator/precursor

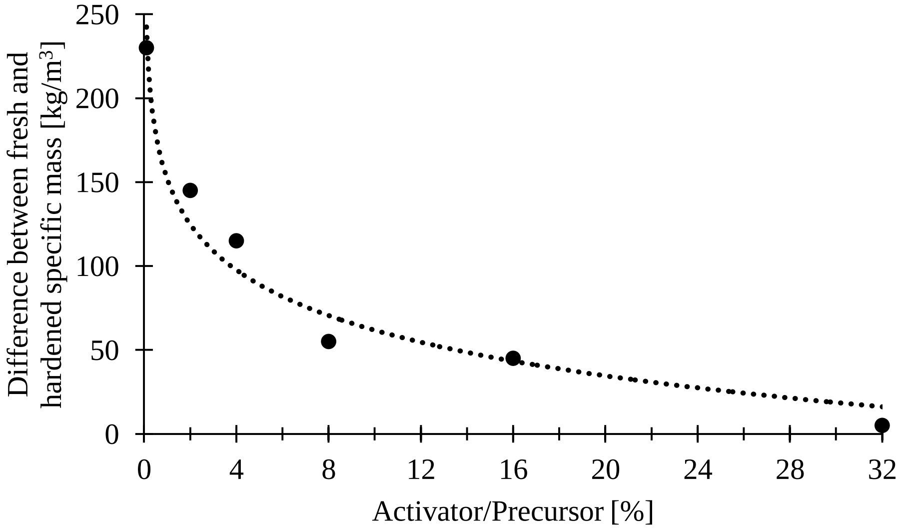


Figure 6 - Difference between fresh and hardened specific mass as a function of activator/precursor ratio

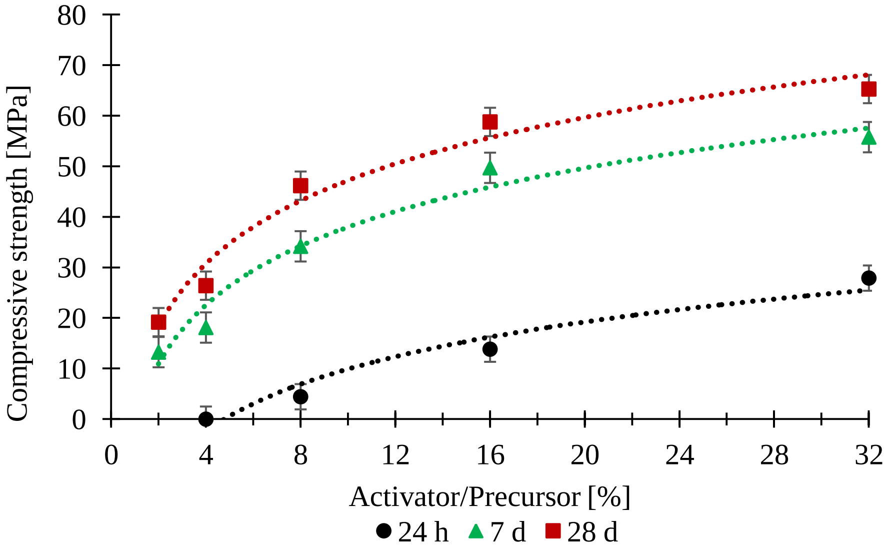
Hardened mixtures

Compressive and flexural strength tests were carried out on prismatic specimens according to EN 1015-11. Table 3 and Figure 7 show compressive and flexural strength at 1, 7, and 28 days. After 24 hours mortars activated with a dosage lower than 8 % were not enough hardened to measure mechanical properties. Flexural (Table 3) and compressive (Table 3 and Figure 7) strength are strictly related with the dosage of the activator. Independently of the age (1, 7, and 28 days), the higher the percentage of the activator the stronger mechanically is the mortar. Compressive strength at 28-days was 6,7 MPa and 63,7 MPa for the reference mortars and the mixture containing 32 % of activator, respectively. This effect is ascribed to the higher amount of silica dissolved, when the activator dosage increase. This assumption is confirmed by the very good correlation between pH of the alkaline activator solution, dissolved silica and 28-day compressive strength of the mortars (Figure 8) according to Pacheco-Torgal [27]. In other words, it is therefore possible to "tailor" the compressive strength of alkali-activated material through the dosage of activator. Specifically, reference mortar (no-activated) can be used for plasters and renders (28-day compressive strength: 6,7 MPa). Weakly alkali-activated (2 - 4 %) GGBFS mortars exhibit compressive strength values specified for seismic retrofitting of masonry buildings (28-day compressive strength equal to 19.2 MPa and 26.4 MPa, respectively). Dosages of the activator higher than 8% (vs precursor mass) allows to manufacture mixtures devoted to structural and/or "cosmetic" repair of existing reinforced concrete elements (Figure 9). Moreover, it is possible to observe how the strength of the activated compounds at 16% and 32% by mass are similar. It can therefore be said that there isn't any advantage in producing mixture with activator/precursor equal to 32%.

Table 3 – Mechanical properties of hardened mortars

Composition – Hardened properties	**S0**	**S2**	**S4**	**S8**	**S16**	**S32**
w/b	0.59	0.58	0.58	0.53	0.50	0.43
Density at hardened state [kg/m^3]	1940	2040	2070	2165	2195	2275
R_f at 24 h [MPa]	**	**	**	1.2	2.9	4.7
R_f at 7 d [MPa]	1.1	3.2	4.0	4.0	6.3	6.6
R_f at 28 d [MPa]	1.2	3.5	4.0	4.4	6.5	6.7
R_c at 24 h [MPa]	**	**	**	4.4	13.8	27.9
R_c at 7 d [MPa]	3.7	13.3	18.1	34.2	49.7	55.8
R_c at 28 d [MPa]	6.7	19.2	26.4	46.2	62.8	63.7

Note: ** The mixture is not hardened enough to be demolded



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167 *Figure 7 - Compressive strength of mortars*

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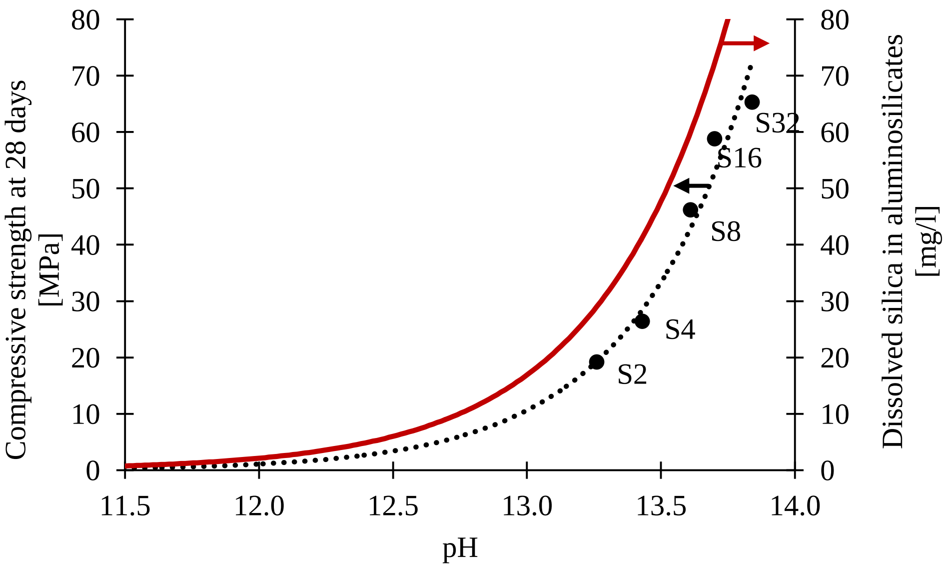

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170 *Figure 8 - Compressive strength of mortars at 28 days vs pH and amount of dissolved silica in aluminosilicate vs pH*

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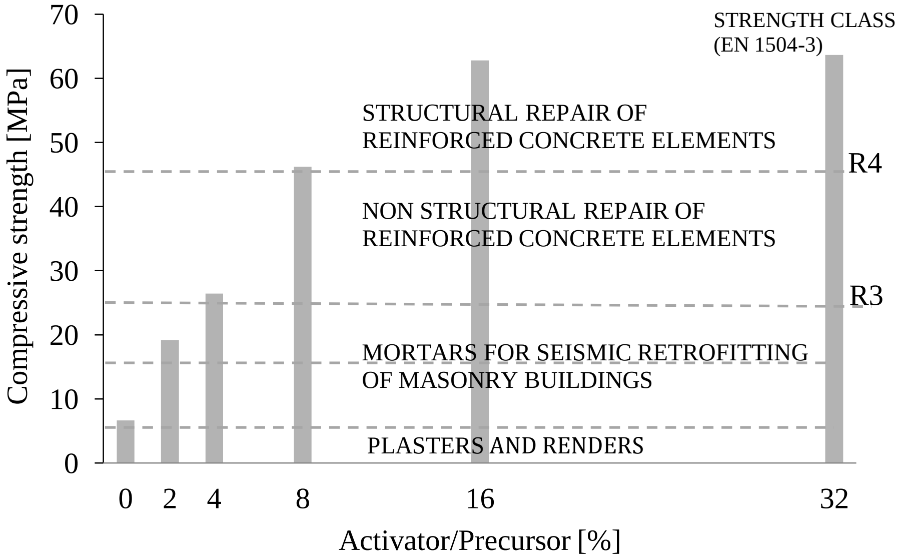

Figure 9 - Compressive strength of GGBFS-based mortars at different activator/precursor

Shrinkage tests were performed up to 150 days on prismatic specimens at 20 °C and R.H. 60%. Figure 10 shows very high free shrinkage of AAMs compared to mixtures manufactured with traditional binders. This phenomenon is due to the different porosity of slag-based pastes compared to traditional cement pastes. Indeed, alkali-activated slag pastes have a much higher proportion of pore size within mesopore region than OPC pastes [28,29]. Further, the radius of pore where the meniscus forms is smaller for AAS than Portland-based pastes, in accordance with the theory that capillary tensile forces set up during drying is a very significant factor for the drying shrinkages of alkali-activated slag materials [30]. In addition, it is possible to note that shrinkage is also influenced by the percentage of alkaline activators, mortar manufactured with 2 % by mass shows free shrinkage equal to 2`000 $\cdot 10^{-6}$ at 150 days from casting, while mixture with 16 % of activators experienced a contraction equal to 4`200 $\cdot 10^{-6}$ at 150 days. Figure 11 shows a linear relationship between the free shrinkage and the dosage of activators. This effect is probable due to the greater amount of silica dissolved in the mortars caused by increasing the activator dosage. An increase of percentage of activator involve in a higher rate of alkali-reaction of the paste and contributed to the formation of more amorphous phases. As a result, the matrix was denser and the shrinkage increased. Moreover, a low water/precursor ratio resulted in a decrease in pore size and subsequently in the water content of the paste, which caused the high capillary stress as described by Kelvin-Laplace equation, strictly related to the autogenous shrinkage [31].

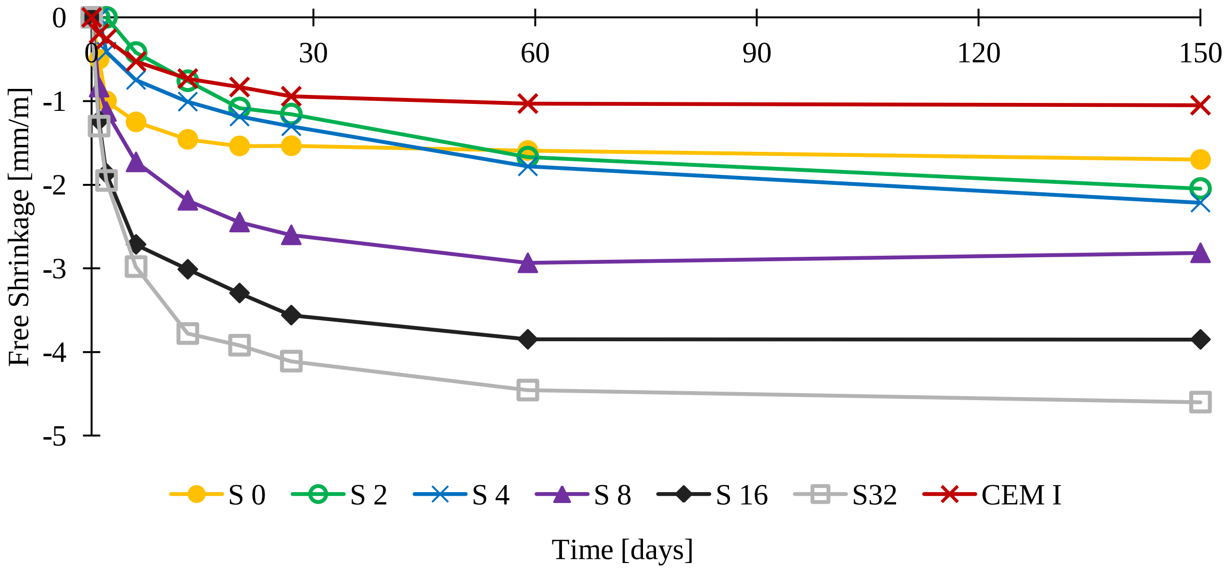

Figure 10 - Free shrinkage of mortars

As far as elastic modulus, results indicated that stiffness of GGBFS-based matrix is significantly lower compared to ordinary Portland cement mortars manufactured with the same aggregates, at equal strength class. These results are in agreement with Thomas et al. [32] which explain the reduction of elastic modulus as a consequence of the high shrinkage of AAM mortars which caused microcrack formation. The presence of micro-cracks is confirmed by observations under an optical microscope on a thin section. In fact, Portland cement-based matrix is dense and compact (Figure 12) while the alkali-activated slag-based compounds show several cracks in binder paste (Figure 13).

Low activator/precursor ratios determine Young's modulus ranging from 10 GPa and 15 GPa, while higher alkaline powders dosages cause an increase in compressive strength and, consequently, elastic modulus grows up to 20 GPa (Figure 14). Since, elastic modulus is significantly lower than that of a Portland-based mortar, tensile stress induced by restrained shrinkage could still be low, preventing the AAM from cracks and detachments. In addition, the use of shrinkage reducing admixtures (SRA) and expansive agents seems to strongly mitigate the problem of excessive shrinkage of alkali activated slag-based mortars [33,34].

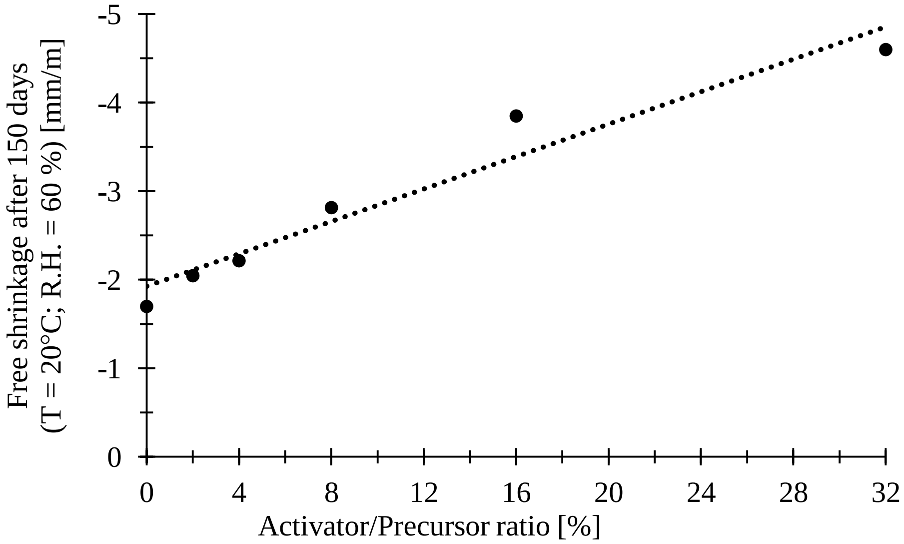

Figure 11 - Free shrinkage of mortars vs activator/precursor

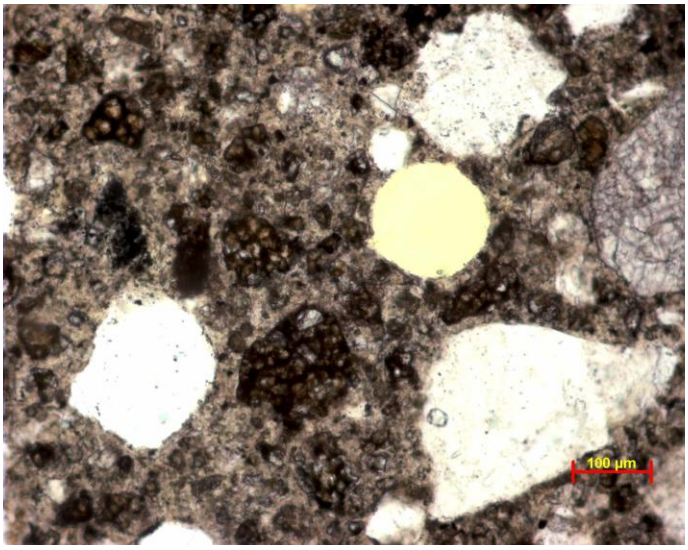

Figure 12 – Optical microscope observation of OPC-based mortar

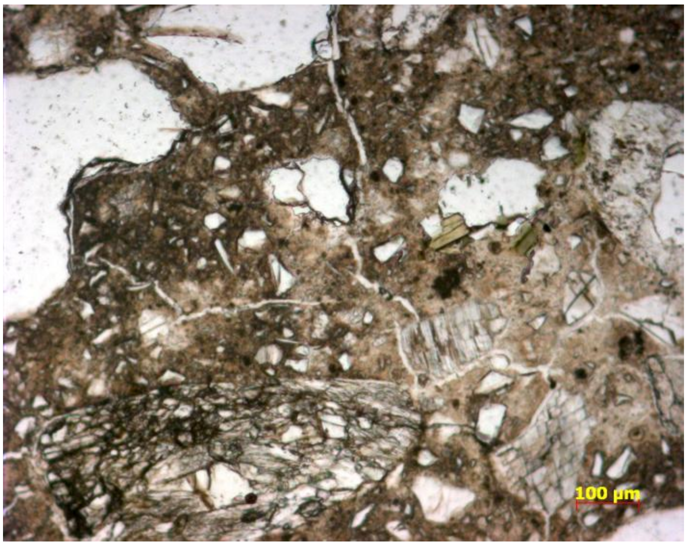

Figure 13 – Optical microscope observation of alkali-activated slag-based mortar (S16 specimen)

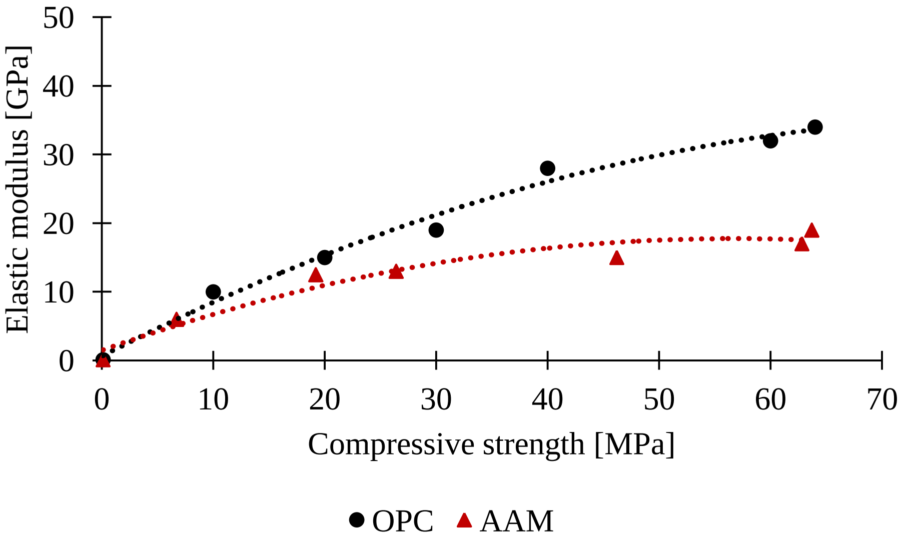

Figure 14 - Elastic modulus as a function of compressive strength

GER AND GWP PARAMETERS

Scientific literature condemns that the use of alkali-activated binders gives enormous benefits from the environmental and ecological point of view [35–37]. For this reason, two fundamental parameters were analyzed: GWP (Global Warming Potential) and GER (Gross Energy Requirement). In particular, the environmental impact of slag-based mortars was calculated on the basis of the data shown in Table 4 and compared to that of OPC mortars at equal 28-day strength class (Table 5). It is possible to observe how, at the same compressive strength, 80 – 90 % and 70 – 80 % reduction in greenhouse gas emission and energy production, respectively, can be achieved compared to mortars produced with Portland cement (Figure 15).

Table 4 – Parameters GER and GWP of raw materials (source Ecoinvent 3.0 databased)

	GWP [kgCO$_2$/kg]	GER [MJ/kg]
CEM I 52.5 R	$9.8 \cdot 10^{-1}$	5.50
GGBFS	$1.7 \cdot 10^{-2}$	0.31
Aggregates	$2.4 \cdot 10^{-3}$	0.13
Sodium metasilicate pentahydrate	1.24	10.58
Potassium hydroxide	1.94	20.50
Sodium carbonate	2.20	7.23

Table 5 – Parameters GER and GWP of mortars at the same 28-day strength class

28-day compressive strength	Mixture	GER [MJ/m^3]	GER [% vs REF]	GWP [kg CO$_2$/m^3]	GWP [% vs REF]
25 MPa	OPC	2374	--	395	--
	S4	541	23%	38	10%
45 MPa	OPC	3314	--	566	--
	S8	774	23%	66	12%
65 MPa	OPC	3906	--	674	--
	S16	1242	32%	120	18%

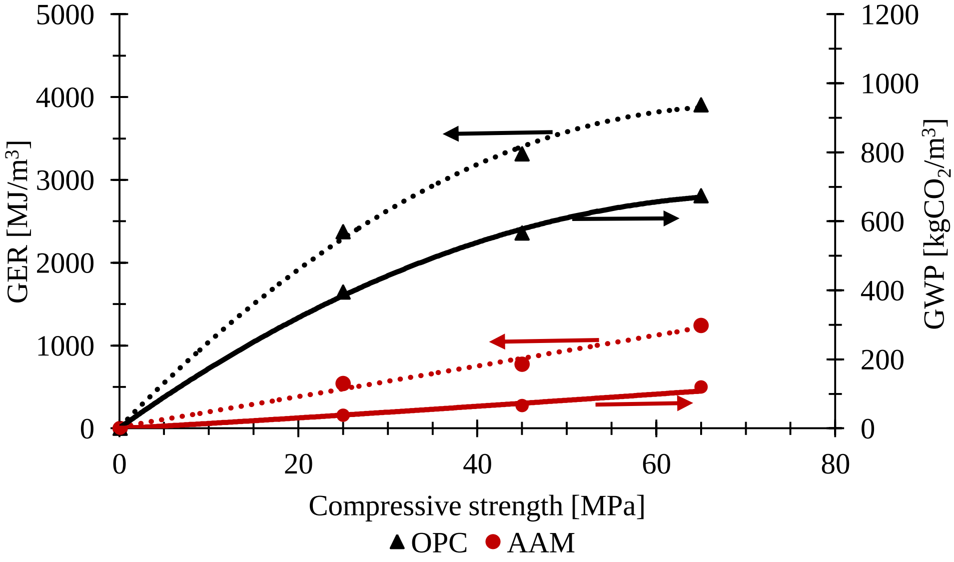

Figure 15 - Variation of GER and GWP of Portland cement mortars and Ground granulated blast furnace slag mortars as a function of compressive strength

CONCLUSIONS

In this paper, performances of alkali-activated binder based on ground granulated blast furnace slag (GGBFS) were evaluated in terms of rheological and physical properties.

Experimental results indicated that:

- The key parameter that regulates most of the properties both in fresh and hardened state of alkali-activated compounds is the activator/precursor ratio.

- The higher the activator dosage, the lower is the water demand at equal workability class due to the effect of sodium silicate-slag particles interaction.
- A higher concentration of activator increase the reaction rate, sharply reducing the setting time of mortars from 6 hours to 30 minutes.
- Slag without activator evidences compressive strength required for plasters and renders. When the dosage of the activator is in the range 2-4% by precursor mass, mortars exhibit compressive strength values specified for seismic retrofitting of masonry buildings. Dosages of the activator higher than 8% (vs precursor mass) allows to manufacture mixtures specifically indicated for structural and/or “cosmetic” repair of existing reinforced concrete elements. No significant benefits are observed in increasing the percentage of activator by more than 16 %.
- Shrinkage values for AAMs are significantly higher (2000 – 4000 μm/m at 150 days from casting) compared to that of a cement-based mortars with the same compressive strength. Moreover, the higher the activator/precursor ratio, the higher the shrinkage due to the different porosity and density of AA slag matrix by varying the activators dosage.
- The modulus of elasticity is about 40% lower than that of a cementitious mortar (at the same strength level) due to the micro-cracking formation caused by shrinkage. This means that tensile stress induced by restrained shrinkage could still be low, preventing the AAM from cracks and detachments.
- At the same 28-day compressive strength, AAMs evidence 80 – 90 % and 70 – 80 % reduction in greenhouse gas emission and energy production, respectively, compared to mortars produced with Portland cement.

In conclusion, from the analysis of the strengths and weaknesses of alkali-activated binders, it turns out that alkali-activated mortars and concretes seem to be a reasonable alternative to natural hydraulic lime-based and/or traditional Portland cement-based mixtures for rehabilitation or restoration of ancient masonry buildings and existing concretes structures.

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