

Article



# Performance Enhancement of Alkali-Activated Electric arc Furnace Slag Mortars Through an Accelerated CO<sub>2</sub> Curing Process

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**Abstract:** The use of electric arc furnace slag (EAFS) as sole precursor to produce alkali-activated mortars has been experimentally investigated. EAFS, a by-product of the steel recycling industry, is a coarse material with unevenly distributed and size-extensive particles. Milling of EAFS was required to achieve a cement-like sized powder before it could be used as precursor. Different combinations of sodium hydroxide (NaOH) and sodium silicate (Na2SiO<sub>3</sub>) were used, by varying the Na2O/binder concentration (4%, 6%, 8%, 10%, 12%) and SiO<sub>2</sub>/Na<sub>2</sub>O ratio (0, 0.5, 1.0, 1.5, 2.0, 2.5) to maximize the mechanical performance. The alkaline solutions were prepared 24 h prior to mixing to unify temperatures for all mixes. The results showed that the SiO<sub>2</sub>/Na<sub>2</sub>O ratio and strength development are directly proportional. The maximum 28-day compressive strength obtained, after being subjected to an initial 24 h thermal curing at 80 °C, was 9.1 MPa in mixes with 4% Na<sub>2</sub>O/binder and 2.5 SiO<sub>2</sub>/Na<sub>2</sub>O. However, after an additional 28 days of accelerated carbonation, the maximum compressive strength (i.e., 31 MPa compared to 3.9 MPa in uncarbonated mixes, corresponding to an 800% increase) was obtained in mixes with 12% and 1.0 for Na<sub>2</sub>O/binder and SiO<sub>2</sub>/Na<sub>2</sub>O, respectively, thus showing an alteration in the optimal alkaline activator contents.

**Keywords:** alkali-activated materials; electric arc furnace slag; mechanical performance; carbonation; mortars

# 1. Introduction

The world's annual population is continuously increasing by 1.05% on average, with an increase of 81 million people in 2020 [1]. This growth seems to be proportionally linked to the rising demand for housing and other infrastructure that will simultaneously increase the need for construction materials, such as Portland cement. In 2016, the amount of cement produced worldwide reached 4.2 billion tonnes, 57% of which was produced by China alone [2,3]. It is a well-known fact that for each one tonne of cement produced, around 0.5–0.6 tonnes of CO<sub>2</sub> (0.59 according to IEA, 2020 [4]) are being released into the atmosphere, meaning that in 2016, the cement industry itself released nearly 2.5 billion tonnes of CO<sub>2</sub>, which accounted for 8% of the global CO<sub>2</sub> emissions of the same year [5,6]. Solving this issue cannot be done by cutting the production of cement alone; other cementreplacing materials need to be found to meet the growing demand for this vital building material. Alkali-activated materials (AAM) are a relatively recent technology as integral cement replacement in the production of concrete and with reduced environmental impact and cost [7]. However, some of the precursors generally used for their production, such as ground granulated blast furnace slag (GGBFS) and fly ash (FA), are facing shortage in availability [8]. To maintain a constant flow in the supply chain, other aluminosilicate waste precursors, prone to little variation in yearly production over time, are needed. One contender is the electric arc furnace slag (EAFS), as the steel industry is shifting from new steelmaking to a recycling system, consequently reducing the amount of GGBFS. It

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**Copyright:** © 2021 by the authors. Submitted for possible open access publication under the terms and conditions of the Creative Commons Attribution (CC BY) license (https://creativecommons.org/licenses/by/4.0/). has an aluminosilicate-rich chemistry giving it the possibility to be the next cement-replacing AAM used in concrete. In terms of availability, about 190–290 million tonnes of steel slag are generated each year, 15–20% of which are EAFS [9]. EAFS generation is based on the recycling of scrap metal and pig iron that can be limited at some locations [10]. It is being used mostly as an aggregate substitute for road base course layers and asphalt pavements [11,12], which is a downcycling process from what they can actually offer.

According to several studies [12–21], EAFS has led to notable improvements in mechanical strength when used as coarse and/or fine aggregates replacement. However, its richness in calcium oxide (free-CaO) and Ferric oxide (FeO) has pushed researchers to further explore the possibility of using EAFS as a cement substitute [22,23]. Xu and Deventer [24] found that any silica–alumina source material that has a pozzolanic component is suitable for dissolution in alkaline solutions. Since EAFS is rich in aluminium and silica, it is desirable to examine whether its activation in alkaline solutions can occur, considering this knowledge [25,26].

The use of EAFS as partial or full cement replacement has led to improvements in mechanical and microstructural performances [25,27]. Muhmood et al. [28] conducted an experimental study to understand the mechanical performance of partially replaced clinker with EAFS. The study concluded that a compressive strength of 50 MPa can be achieved with 30% clinker-EAFS replacement, compared to 58 MPa with 20% replacement and 58.6 MPa for the reference specimen with 100% cement. It has also been found that the substitution of untreated EAFS with treated EAFS can lead to an increase in strength [28]. Another study has recorded a 28-day compressive strength of 44.38 MPa using 25% EAFS as cement replacement compared to 59.34 MPa for the reference specimen with 100% cement [29]. Moreover, Zhao et al. [30] investigated the influence of cement's partial substitution with EAFS, focusing on the EAFS' particle distribution. The results revealed that concrete specimens containing properly ground EAFS showed better microstructure, higher compressive strength, lesser porosity and better durability performance compared to the reference blast furnace slag blended cement specimens. Other studies have reported similar results [29,31–33]. In addition, raising the fineness of EAFS particles, as well as integrating early thermal curing in the curing stages, led to better hydration performance and a faster early-age hydration [22,31,34,35]. Similar studies found that the compressive strength of alkali-activated concrete cured at 70 °C was higher than that of concrete cured at 30 °C [22,32,36]. Furthermore, Roslan et al. [36] reported later-age improvements in compressive, tensile and flexural strengths with a 20% cement-EAFS replacement.

The use of EAFS as a precursor for AAM production usually shows some shortcomings in performance in comparison with FA or GGBS. Ozturk et al. [25] studied the influence of the optimization of SiO<sub>2</sub>/Na<sub>2</sub>O ratio and Na<sub>2</sub>O/binder percentage on the mechanical performance of mortars with 100% EAFS as a precursor. For a silicate modulus of 1, 1.5 and 2, the highest compressive strengths reported were 16.55 MPa, 17.11 MPa and 22.02 MPa, respectively. In addition, an increase in flexural strength was noticed with the increase in silicate modulus resulting in values of 2.85 MPa, 3.09 MPa and 4.18 MPa for SiO<sub>2</sub>/Na<sub>2</sub>O ratios of 1, 1.5 and 2, respectively. Moreover, the highest mechanical strengths were all reached under an 80 °C curing temperature and 12 h curing time. Nikolic et al. [37] investigated the influence of the NaOH and KOH concentration, solid to liquid ratio and temperature on the dissolution kinetics of Si and Al in EAFS. In the study, the increase in alkaline solution concentrations (NaOH or KOH) increased the dissolution of Si and Al of EAFS. Similarly, a decrease in the solid to liquid ratio and an increase in temperature also catalyzed this reaction. Furthermore, Turker et al. [35] studied the influence of thermal curing on the microstructural performance of alkali-activated EAFS mortars activated by a solution with 5% and 1.0 of Na<sub>2</sub>O/binder and SiO<sub>2</sub>/Na<sub>2</sub>O ratios, respectively. A compressive strength of 25.6 MPa was reported for specimens cured at ambient temperature (21 °C), compared to 40.7 MPa for a similar sample cured at 60°C for 6 h (59% increase). A similar study also demonstrated the benefits of early-age thermal curing on the strength development and shrinkage of alkali-activated EAFS mortars, after reporting an increase in compressive strength from 14 MPa to 21 MPa after a thermal curing stage at 70 °C with durations ranging between 3 h and 24 h, respectively [38]. Cesnovar et al. [39] studied the influence of different slag mixtures containing EAFS and ladle furnace basic slag (LS) with different mixing ratios and activated using potassium silicate with a fixed ratio to slag of 1/2. A compressive strength of 56.7 MPa was reported using an EAFS/LS ratio of 1/1. This optimal mixture was used to understand the influence of thermal curing on strength development. The results indicated that the specimens cured at 70 °C for 3 days attained similar compressive strength (~56 MPa) to that of specimens cured at room temperature for 28 days.

Despite the effort made, little is known on the use of EAFS as full binder replacement, and more experimental studies should take place to further explore this interesting research idea. In addition, the optimization of the alkali activators may pave the way toward more encouraging results, but the mechanical performance values recorded so far are not market competitive. This eagerness toward achieving better performance raised the idea of carbonation as a curing method. Exploiting the possibility of curing the EAFS concrete with CO<sub>2</sub> [40–42], by taking advantage of its CaO-rich chemistry, will not only lead to better mechanical performance but also an added value to CO<sub>2</sub>-capturing revolutionary technologies. Monkman and Shao [40] assessed the carbonation behavior of six cementitious materials (cement, FA, GGBFS, EAFS, and hydrated lime) to determine and compare the strength development, %CO2 uptake and carbonation degree for each one of them. The samples were exposed to 100% CO<sub>2</sub> at a pressure of 5 bar for 2 h. Afterward, X-ray diffraction (XRD) and scanning electron microscopy (SEM-EDS) analyses were performed to determine the products of carbonation and to observe the morphology of the carbonation products. The results showed a 12% CO2 uptake for EAFS, FA and cement, unlike lime and BFS that presented 40% and 7% CO2 uptake, respectively. Similarly, cement and EAFS were comparable in terms of carbonation degree with a value of around 25%. The compressive strength (at 2 h) of EAFS reached 16.6 MPa, while that of FA was only 3.5 MPa.

In this paper, the use of alkali-activated electric arc furnace slag (AAEAFS) as a sole precursor in the production of mortars has been investigated. Its main objective is to reach the optimization of the activator's composition based on maximized mechanical performance. Twenty-one different mixes were produced to study the influence of different alkali activator concentrations and ratios, to find the best alkaline solution for the EAFS mortars. Different silicate modulus ratios (0, 0.5, 1.0, 1.5, 2.0, 2.5) and Na<sub>2</sub>O/binder concentrations (4%, 6%, 8%, 10%, 12%) were selected for this study. Their effect on the performance was evaluated through comprehensive testing methodologies, including slump, shrinkage, flexural strength (FS), compressive strength (CS) and carbonation degree. A 28-day curing period, including a 7-day accelerated carbonation stage, followed. Finally, the correlations between the distinctive preparation conditions and the different tested parameters were graphically illustrated, serving the upcoming research studies toward the best future use of EAFS as a more sustainable cement-replacing material.

#### 2. Materials and Methods

#### 2.1. Electric Arc Furnace Slag (EAFS)

The EAFS used in this study is a by-product of steel recycling, collected from Siderurgia Nacional de Portugal and provided by HARSCO (Portugal). It presents an extensive and coarse particle size distribution that requires preparation and grinding before it can be used as a binder.

#### 2.2. Alkaline Activator

The alkaline activator was prepared in the form of a liquid solution. Reactive grade anhydrous sodium hydroxide pellets (NaOH) from Crimolara (Portugal), with 98% purity and a density of 2.13 g/ml, then dissolved in tap water, complying with Directive 98/83/CE

[43]. A commercial solution of sodium metasilicate (Na<sub>2</sub>SiO<sub>3</sub>) from Crimolara (Portugal) was then added, containing  $8 \pm 0.6\%$  of sodium oxide (Na<sub>2</sub>O),  $26.4 \pm 1.5\%$  of silicon oxide (SiO<sub>2</sub>) and  $65.6 \pm 2\%$  of water (H<sub>2</sub>O). The Na<sub>2</sub>SiO<sub>3</sub> has a relative density of 1.355 g/ml.

#### 2.3. Fine Aggregate

The mortars were produced using two types of fine siliceous aggregates (i.e. coarse and fine river sands) to maximize compacity through an extensive particle size distribution. The particle size distribution showed maximum nominal sizes (NS) of 1 mm and 4 mm for fine and coarse aggregates, respectively. The 24 h water absorption (WA<sub>24</sub>), including the values of the apparent ( $\rho_a$ ), rodded-dry ( $\rho_{rd}$ ), saturated surface-dry ( $\rho_{SSD}$ ), and bulk ( $\rho_b$ ) densities are shown in Table 1.

Accesso	NS	ρα	ρrd	pssd	ρь	$WA_{24}$
Aggregates	mm	kg/m³	kg/m³	kg/m³	kg/m <sup>3</sup>	%
Fine sand	0/1	2652	2624	2637	1544	0.4
Coarse sand	0/4	2636	2601	2617	1556	0.5

## 2.4. Water-Reducing Admixture

The water-reducing admixture (WRA) used in this research was SikaPlast-717, consisting of a synthetic organic water-based naphthalene-based dispersant, with a density of  $1.21 \pm 0.03$  kg/dm<sup>3</sup> and a pH of  $10 \pm 1$ .

## 2.5. Mortar Mix Design

The experimental research focused on optimizing the alkaline activator based on the mortar's mechanical performance. To achieve the optimum activator for EAFS, different mixing combinations were used. The Na<sub>2</sub>O/precursor concentrations used were 4%, 6%, 8%, 10% and 12%, while the adopted SiO<sub>2</sub>/Na<sub>2</sub>O mass ratios were 0, 1.0, 1.5, 2.0 and 2.5 (Table 2). The amount of each constituent was calculated based on the volumetric and mass ratios of the components, in accordance with their densities. The binder/aggregate volumetric ratio (V<sub>B</sub>/V<sub>A</sub>) was 0.33, while the mass ratio of water/binder (M<sub>W</sub>/M<sub>B</sub>) was fixed at 0.3. The mass WRA/precursor ratio varied from 0% to 1% to achieve constant workability.

$N_{22}O(9/)$	SiO <sub>2</sub> /Na <sub>2</sub> O									
1Nd2O (70)	0	0.5	1	1.5	2	2.5				
4	N4S0	N4S0.5	N4S1	N4S1.5	N4S2	N4S2.5				
6	N6S0	N6S0.5	N6S1	N6S1.5	N6S2	-				
8	N8S0	N8S0.5	N8S1	N8S1.5	-	-				
10	N10S0	N10S0.5	N10S1	-	-	-				
12	N12S0	N12S0.5	N12S1	-	-	-				

Table 2. Mix code of EAFS mixes.

## 2.6. Production Method

The production of mortars was based on EN 196-1 [44]. The alkaline solution was prepared by gradually dissolving the NaOH pellets in water and then leaving them to cool down for 24 hours. The  $40 \times 40 \times 160$  mm<sup>3</sup> three-gang steel moulds were wrapped entirely with thin plastic film to demould the samples without the need for a release agent. The alkaline solution was poured first into the mixer followed by WRA and the precursor, before mixing for 3 minutes. After that, the mixer was paused to add the fine aggregates, and then mixing was resumed for another 2 minutes, followed by one additional minute of high-speed mixing. Next, the slump was tested using the slump table according to EN 1015-3 [45]. Afterward, the mix was moulded and covered with plastic film and immediately placed in the thermal curing chamber. Finally, the specimens were demoulded, and each specimen was sealed and left to cure in a dry chamber at a temperature of  $23 \pm 2$  °C and relative humidity (RH) of 65%. The samples were left in the dry chamber until the testing day.

## 2.7. Curing Conditions and Testing Methods

As stated previously (section 2.6), the moulded mix was thermally cured at 80 °C for 24 hours. After that, the mortar specimens were demoulded and placed in their designated curing condition depending on the desired test method, as shown in Table 3. The loading rate for the flexural and compressive strength tests had a constant value of 30 N/s and 300 N/s, respectively, complying with its corresponding standard (Table 3). The compressive test was applied to both halves resulting from the flexural test. The shrinkage values of the specimens were measured using a mortar shrinkage apparatus following a certain measuring regime in accordance with EN 1015-13 [46].

Table 3. Curing conditions and testing methods assigned for the specimens.

Test	Standard	No. of specimens	Curing conditions
Flexural strength	ENI 101E 11 [47]	(	Sealed specimens in a dry chamber until
Compressive strength	EN 1015-11 [47]	6	testing age.
			14 days sealed + 14 days unsealed in the
Accelerated carbonation	EN 13295 [48]	4	dry chamber; then placed in the carbona-
			tion chamber until testing age.
Charinghan	ENI 101E 10 [4/]	2	Sealed specimens in a dry chamber after
Эпгіпкаде	EIN 1015-13 [46]	2	demoulding until the end of the test.

### 3. Results

## 3.1. Characterization of EAFS

This material presents an apparent density of 3770 kg/m<sup>3</sup>. The oxide chemical composition of the raw material, obtained from X-ray fluorescence (XRF), is shown in Table 4. The studied EAFS contains 28.5% of Fe<sub>2</sub>O<sub>3</sub>, 28.2% of CaO, 17.7% of SiO<sub>2</sub>, and 10.1% of Al<sub>2</sub>O<sub>3</sub>. The high amount of iron in EAFS could induce magnetic properties on AAEAFS concrete [49] and it was confirmed to be a strongly magnetic precursor with the use of neodymium magnets.

Table 4. Chemical composition of EAFS obtained from XRF (%)
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e <sub>2</sub> O <sub>3</sub>	CaO	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	MgO	MnO <sub>2</sub>	Cr <sub>2</sub> O <sub>3</sub>	TiO <sub>2</sub>	P2O5	SO <sub>3</sub>	Na <sub>2</sub> O	BaO	K <sub>2</sub> O	$V_2O_5$	CuO	ZnO
28.48	28.18	17.66	10.13	5.66	5.44	2.38	0.65	0.42	0.33	0.19	0.17	0.03	0.11	0.02	0.02

Figure 1 presents the results of the particle size distribution of the EAFS after the milling process. Ordinary Portland and fly ash (FA) from a coal power plant were tested as reference materials. EAFS showed a bimodal particle distribution curve similar to FA and cement. The highest peak of both the EAFS and cement curves indicated a similar particle diameter of 25  $\mu$ m. However, the peak corresponding to cement's curve was sharper than the one of EAFS, showing a higher distribution percentage. EAFS and cement showed another peak at 0.35  $\mu$ m, indicating the presence of very fine particles. In contrast, FA showed a wider peak at 20  $\mu$ m and a lower distribution percentage at 0.35  $\mu$ m compared to EAFS and cement.

In Figure 2a, the EAFS particles are irregular in shape, dispersed, and have angular

morphology. In addition, finer particles were observed on the surface of the larger ones (Figure 2b). On a larger scale view, vitrified surfaces are seen resembling those of materials exposed to elevated temperatures (i.e. greater than 1600 °C) suggesting the possibility of pozzolanicity and reactivity to alkali activation (Figure 2c). Moreover, the EDS test was carried out on the area presented in Figure 2d and the results were graphically represented in Figure 3. The corresponding EAFS particle is composed of around 50.6% oxygen, 18.0% calcium, 7.1% silicon, 6.2% of iron, and other components. This primary composition matches the mineralogical composition of the EAFS XRD results (section 3.1.3) showing high calcium content seen as white particles (Figure 2d), as well as silicon oxide and iron oxide.



Figure 1. Particle size analysis.



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Figure 2. SEM micrographs of EAFS sample (magnification of a - 100×; b - 500×; c - 5000×; d - 1000×).

Figure 4 shows the EAFS' mineralogical composition obtained by XRD. The results showed three fundamental phases: wustite (FeO), gehlenite (Ca<sub>2</sub>Al<sub>2</sub>SiO<sub>7</sub>), and dicalcium silicate (Ca<sub>2</sub>SiO<sub>4</sub>) (Figure 4a). Other minor phases appeared as magnesioferrite. After heating the sample to 1000 °C in thermogravimetric analysis (TGA), the fundamental phases were gehlenite and magnesioferrite, as presented in Figure 4b; wustite was no longer present and hematite was formed as a result of the former's oxidation (mainly between 400 and 620 °C).



Figure 3. EDS coupled to SEM of EAFS sample.



Figure 4. XRD pattern; (a) EAFS; (b) EAFS - TGA.

3.2. Fresh State Mortar Properties

The mortar's workability was evaluated by their slump according to the EN 1015-3 standard [45]. The target slump had been initially set at 140 ± 20 mm. The superplasticizer was used and adjusted accordingly between 0% and 1.0% depending on the Na<sub>2</sub>O/binder and SiO<sub>2</sub>/Na<sub>2</sub>O ratios to achieve constant workability. Increasing the Na<sub>2</sub>O/binder ratio led to a reduction in workability. This was due to the higher viscosity of the alkaline solution after the addition of a solid solute. It was also expected that the mixes would lose workability after a short time for higher SiO<sub>2</sub>/Na<sub>2</sub>O ratios due to flash setting; Ca<sup>2+</sup> ions from the EAFS quickly react with the silicate ions from the solution, leading to the precipitation of an initial C-S-H, which is responsible for the setting [34]. Therefore, it can be stated that there is a linear correlation between the Na<sub>2</sub>O/binder and SiO<sub>2</sub>/Na<sub>2</sub>O ratios and the workability. However, mixes N8S1, N10S1 and N12S1 presented a slightly lower slump between 109 and 117 mm (Figure 5a) due to the increase in viscosity of the alkaline activator. Therefore, the superplasticizer content was increased to 1.0% for mixes with silicate modulus ratios of 1.5 and higher, to adjust the workability and the slump which ranged between 130 and 147 mm (Figure 5b).



Figure 5. Slump values for mortar mixes (WRA content of a - 0.5% and; b - 1.0%).

## 3.3. Hardened State Mortar Properties

## 3.3.1. Compressive Strength

The average compressive strength of the AAEAFS uncarbonated samples is presented in Figure 6, which shows how the change in parameters affects the mechanical performance of the samples. The values presented in the figure have coefficients of variation ranging between 0.5 and 7.5%. Mixes N4S0.5, N6S0.5, N10S0.5, and N12S0.5 were eliminated due to inconsistent values and unexpected outcomes during the mixing process.

The alkaline solutions of mixes N10S0.5 and N12S0.5 showed precipitation, possibly due to the saturation of silica, during its 24 hours of cooling before the mixing day. Thus, the solution must undergo different conditions to avoid any fluctuation in the results. Correspondingly, the alkali activator is the most important factor controlling compressive strength.



**Figure 6.** Average compressive strength: (a) 4% sodium concentration; (b) 6% sodium concentration; (c) 8% sodium concentration; (d) 10% and 12% sodium concentration (Mix code-C represents carbonated specimens).

Different studies on alkali-activated EAFS used at least one of the alkaline activators stated in this study; Turker et al. [35], Ozturk et al. [25], and Peys et al. [50] used both sodium hydroxide and sodium silicate to achieve compressive strength values of 40.7 MPa, 22.0 MPa, and 16.0 MPa, respectively. Abdollahnejad et al. [51] used only sodium hydroxide and recorded 27.0 MPa. This study presented maximum average compressive strength for uncarbonated specimens of 9.61 MPa for mixes with 4% and 2.5 of Na<sub>2</sub>O/binder and SiO<sub>2</sub>/Na<sub>2</sub>O ratios, respectively. Mixes N10S0 and N12S0, which were specimens with insufficient stability, exhibited values close to zero and thus were not re-

ported. This could be due to the excess amount of sodium hydroxide and the lack of sodium silicate. Increasing the amount of Na<sub>2</sub>O to a given percentage increases the strength, after which the performance starts to deteriorate. Nevertheless, excess amount of OH<sup>-</sup>, due to the Ca(OH)<sub>2</sub> across the particles of EAFS, reduces the interaction of Ca<sup>2+</sup> ions from the surface of EAFS [34]. Therefore, it can be stated that the low strength results from the inadequate amount of C-S-H gels produced by the reaction of Ca<sup>2+</sup> with Si<sup>4+</sup>.

Sodium hydroxide and sodium silicate have a significant influence on the mechanical properties of AAEAFS. Si<sup>4+</sup> and Al<sup>3+</sup> ions within the EAFS dissolve much more with a high concentration of OH<sup>-</sup> [52]. Song et al. [53] observed that increasing the concentration of the alkali activator increases the reaction rate as a result of a high alkali medium. Wang et al. [54] stated that sodium hydroxide and sodium silicate are directly proportional to compressive strength only to a given level (from 3-5% by weight of the mix). The authors also stated that, if the amount of sodium concentration increased over a given limit, it would cause efflorescence, as shown in Figure 7. This is due to the migration of Na<sup>+</sup> ions to the surface of the specimens, leading to the precipitation of sodium carbonate.

Another parameter affecting the compressive strength for all mixes was the curing time. The specimens in this study experienced 7-day, 28-days, and 91-days of curing in a dry chamber at  $23 \pm 2$  °C and RH of 65%. Since all specimens were sealed, there was no exchange of humidity with the surrounding environment. The maximum compressive strength (9.61 MPa) reported for the uncarbonated specimen N4S2.5, was obtained at 91-days of curing age.



Figure 7. Efflorescence caused by Na<sub>2</sub>CO<sub>3</sub> on the surface of a mortar specimen.

#### 3.3.2. Flexural strength

Figure 8 presents the results reported for the flexural strength of AAEAFS mortars. The values presented in the figure have coefficients of variation ranging between 0 and 11% except in one case that showed 17%. The maximum flexural strength for uncarbonated specimens reached 2.45 MPa for mix N12S1. The surface of the specimens presented some microcracks, possibly caused by the heat curing process and the expedited nature of the reactions at relatively high-temperature levels [55], thus causing a decline in strength. Although a higher SiO<sub>2</sub>/Na<sub>2</sub>O ratio is a good indicator of an improved performance [52,56–58], the low performance in the case of EAFS may be due to the low amount of amorphous phases present in the precursor, which did not react with the alkaline activator. Furthermore, even though one would expect to have improved performance from the interaction of Ca from

EAFS with SiO<sub>2</sub> from the activator to produce C-S-H gels, it is possible that the Ca-bearing mineralogical phases were stable at high pH levels, thereby minimizing the dissolution of Ca<sup>2+</sup> ions to the solution. In this context, flexural strength will only be enhanced when exposed to accelerated carbonation. This improvement in strength could be caused by the reaction of OH<sup>-</sup> from the alkaline activator with the Ca<sup>2+</sup> ions released from the decalcified phases of EAFS to generate Ca(OH)<sub>2</sub> and subsequently CaCO<sub>3</sub>.



**Figure 8.** Average flexural strength: (a) 4% sodium concentration; (b) 6% sodium concentration; (c) 8% sodium concentration; (d) 10% and 12% sodium concentration (Mix code-C represents carbonated specimens).

# 3.3.3. Carbonation

The average compressive and flexural strength for the carbon-cured EAFS mortars are presented in Figure 6 and Figure 8. The specimens in this study, after 28 days of curing in a dry chamber followed by 7 days of exposure to CO<sub>2</sub> (i.e. 35 days), were considered fully carbonated, as there was no indication of a pinkish hue from the phenolphthalein solution pH indicator, thereby making the CO<sub>2</sub> penetration impossible to read. Alkaliactivated materials are known to present a fast decline in pH with ongoing polymerization reactions due to the consumption of the OH<sup>-</sup> ions present in the pore solution. This phenomenon, in combination with the carbonation of Ca-bearing phases, led to an overall decline of the specimens' pH.

The average compressive and flexural strengths of the specimens subjected to accelerated carbonation were tested following EN 1015-11 [47]. A noticeable improvement in the mechanical performance of AAEAFS mortars was observed. The additional 28 days of accelerated carbonation following the 28 days in a dry chamber (i.e. 56 days) led to a near 800% and 500% increases in compressive (i.e. from 3.9 MPa to 31 MPa) and flexural (i.e. from 1.6 MPa to 7.85 MPa) strengths, respectively. The maximum mechanical performance was obtained in mixes with 12% and 1.0 for Na<sub>2</sub>O/binder and SiO<sub>2</sub>/Na<sub>2</sub>O ratios, respectively, thus showing a shift in the optimal alkaline activator contents. It is likely that Ca<sup>2+</sup> ions were released from the EAFS' Ca-bearing phases and reacted with CO<sub>2</sub> forced into the microstructure. This resulted in the precipitation of CaCO<sub>3</sub> polymorphs, which significantly densified the microstructure. The resulting SiO<sub>2</sub> gels from decalcified calcium silicate phases also led to a widespread polymerization thus a more effective binding of the microstructure. These processes resulted in enhanced performance [25,53].

The silicate modulus and sodium concentration also had an obvious effect on the mechanical performance. Contrary to that expected, the flexural strength of mixes with lower performance is often not correlated with the compressive strength, as presented in Figure 9.



**Figure 9.** Flexural vs. compressive strengths of mortars from this study compared with other studies from the literature [59–66].

Carbonated specimens with enhanced performance, in which exponential relationships were observed between the flexural and compressive strength, suggested otherwise. The 28-day relationship of carbonated specimens was close to that obtained by standard 40×40×160 mm cement mortar prims from other studies [59–66]. The improvement in flexural strength between 7 days and 28 days is likely due to the greater polymerization of SiO<sub>2</sub> gels from the greater amount of decalcified calcium silicate phases (e.g., C-S-H). The isotropic growth of these phases, but especially of those growing perpendicularly to the loading vector, led to improved flexural strength.

#### 3.3.4. Shrinkage

In Figure 10, the change in length (i.e. shrinkage) of sealed specimens (with little to no humidity exchange with the surrounding environment) was tested for 91 days. Most specimens presented considerable shrinkage, with one of them close to  $3500 \mu$ m/m, which

is three times what is typically observed in standard cement mortars. All specimens presented at least 45% of their total 91-day shrinkage in the first 28 days, except for mix N8S0.5. Mix N8S0.5 slightly expanded, with minor fluctuations, for the first 28 days and started to shrink later on to settle at 198  $\mu$ m/m after 91 days. This behaviour also occurred in fly ash mortars studied by Atiş et al. [67]. The authors hypothesized that the expansion of mortars containing FA could be from the MgO and the high content of SO<sub>3</sub>, which can result in long-term instability due to the formation of expansive calcium sulphate phases [67]. In Figure 10a, the shrinkage of specimens with a SiO<sub>2</sub>/Na<sub>2</sub>O ratio of 1.0 decreased with increasing Na<sub>2</sub>O/binder ratio and somewhat correlated with their mechanical behaviour. It is likely that the greater dissolution of aluminosilicate phases from the higher pH level of the alkaline solution led to the formation of more strength-enhancing and densifying phases, thus leading to effective restrained shrinkage. However, the opposite was observed for mixes with a SiO<sub>2</sub>/Na<sub>2</sub>O ratio of 1.5. Further research is required to ascertain the trend of specimens with varying silicate modulus and Na<sub>2</sub>O content.



**Figure 10.** Shrinkage for AAEAFS mortars: (a) values for mixes with silicate modulus of 1.0; (b) values for mixes with silicate modulus of 0.5 and 1.5.

# 4. Conclusions

In this study, electric arc furnace slag (EAFS) was studied as a potential full replacement for cement in the production of alkali-activated mortars. The results obtained in this study allowed a conclusion that EAFS as the sole precursor will result in mixes with relatively low performance. According to the XRD results, this is most likely due to the lower number of amorphous phases compared to other common aluminosilicate pozzolans.

The SiO<sub>2</sub>/Na<sub>2</sub>O ratio and compressive strength are generally directly proportional. The compressive strength was also affected by curing time. The continued reaction was observed after the 24 h thermal curing stage. The specimens in this study experienced 7 days, 28 days and 91 days of curing in a dry chamber at  $23 \pm 2$  °C and RH of 65%. The maximum recorded compressive strength was obtained from specimens tested on day 91 corresponding to 4% and 2.5 of Na<sub>2</sub>O/binder and SiO<sub>2</sub>/Na<sub>2</sub>O ratios, respectively.

Despite the shortcomings of EAFS as a sole precursor, the mechanical performance increased significantly after subjecting the specimens to an accelerated carbonation stage. After subjecting the specimens to a CO<sub>2</sub>-enriched environment for 28 days, AAEAFS showed an average compressive strength increase of ~500%, with one case reaching 800% (i.e., from an initial 3.9 MPa in uncarbonated mixes to 31 MPa for carbonated ones). The maximum performance was observed in mixes with 12% and 1.0 for Na<sub>2</sub>O/binder and SiO<sub>2</sub>/Na<sub>2</sub>O ratios, respectively, thus showing a shift in the optimal alkaline activator contents.

The sealed shrinkage test showed considerable dimensional variability over time. Great autogenous shrinkage may have occurred due to the continuous alkali activation reaction. Nevertheless, this phenomenon is still widely unknown and must be further researched.

The complete replacement of cement with alkali-activated aluminosilicate waste may translate into significant reductions in cost and minimal environmental impacts, especially with incorporating a forced carbonation curing stage using industrial CO<sub>2</sub>-rich flue gases. Therefore, greater focus should be given to this curing technique in future research.

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#### References

- 1. Worldometer, Current World Population, World Population Sections. (2020). Available online: https://www.worldometers.info/world-population/ (accessed on 29 November 2021).
- DBS Group Research, Production of cement in China from 1970 to 2020, DBS Group Research; CEIC; National Bureau of Statistics of China, Statista Research Department. (2018). Available online: https://www.statista.com/statistics/307647/china-production-volume-of-cement/ (accessed on 29 November 2021).
- US Geological Survey. Cement Production Worldwide From 1995 to 2020, USGS Mineral Commodity Summaries; US Geological Survey: Reston, Virginia, 2021, p. 43.
- 4. IEA. Cement; IEA, Paris, France, 2021. Available online: https://www.iea.org/reports/cement (accessed 10 January 2022).
- 5. Andrew, R.M. Global CO 2 emissions from cement production, 1928–2018. *Earth Syst. Sci. Data* 2019, 11, 1675–1710.

- 6. Habert, G.; Roussel, N. Study of two concrete mix-design strategies to reach carbon mitigation objectives. *Cem. Concr. Compos.* **2009**, *31*, 397–402.
- 7. Xiao, R.; Zhang, Y.; Jiang, X.; Polaczyk, P.; Ma, Y.; Huang, B. Alkali-activated slag supplemented with waste glass powder: Laboratory characterization, thermodynamic modelling and sustainability analysis. *J. Clean. Prod.* **2021**, *286*, 125554.
- Alberici, S.; de Beer, J.G.; van der Hoorn, I.; Staats, M. Fly Ash and Blast Furnace Slag for Cement Manufacturing. *BEIS Res. Pap.* 2017, 19, 1–34.
- Teo, P.T.; Zakaria, S.K.; Salleh, S.Z.; Taib, M.A.A.; Mohd Sharif, N.; Abu Seman, A.; Mohamed, J.J.; Yusoff, M.; Yusoff, A.H.; Mohamad, M. Assessment of electric arc furnace (EAF) steel slag waste's recycling options into value added green products: A review. *Metals* 2020, 10, 1347.
- 10. Fisher, L.V.; Barron, A.R. The recycling and reuse of steelmaking slags A review. Resour. Conserv. Recy. 2019, 146, 244–255.
- 11. Manso, J.M.; Gonzalez, J.J.; Polanco, J.A. Electric arc furnace slag in concrete. J. Mater. Civ. Eng. 2004, 16, 639–645.
- 12. Autelitano, F.; Giuliani, F. Electric arc furnace slags in cement-treated materials for road construction: Mechanical and durability properties. *Constr. Build. Mater.* **2016**, *113*, 280–289.
- 13. Abu-Eishah, S.I.; El-Dieb, A.S.; Bedir, M.S. Performance of concrete mixtures made with electric arc furnace (EAF) steel slag aggregate produced in the Arabian Gulf region. *Constr. Build. Mater.* **2012**, *34*, 249–256.
- 14. Adegoloye, G.; Beaucour, A.-L.; Ortola, S.; Noumowé, A. Concretes made of EAF slag and AOD slag aggregates from stainless steel process: Mechanical properties and durability. *Constr. Build. Mater.* **2015**, *76*, 313–321.
- 15. Alharbi, Y.R.; Abadel, A.A.; Elsayed, N.; Mayhoub, O.; Kohail, M. Mechanical properties of EAFS concrete after subjected to elevated temperature. *Ain Shams Eng. J.* **2021**, *12*, 1305–1311.
- 16. Coppola, L.; Buoso, A.; Coffetti, D.; Kara, P.; Lorenzi, S. Electric arc furnace granulated slag for sustainable concrete. *Constr. Build. Mater.* **2016**, *123*, 115–119.
- 17. Lam, M.N.-T.; Le, D.-H.; Jaritngam, S. Compressive strength and durability properties of roller-compacted concrete pavement containing electric arc furnace slag aggregate and fly ash. *Constr. Build. Mater.* **2018**, *191*, 912–922.
- 18. Monosi, S.; Ruello, M.L.; Sani, D. Electric arc furnace slag as natural aggregate replacement in concrete production. *Cem. Concr. Compos.* **2016**, *66*, 66–72.
- 19. Rooholamini, H.; Sedghi, R.; Ghobadipour, B.; Adresi, M. Effect of electric arc furnace steel slag on the mechanical and fracture properties of roller-compacted concrete. *Constr. Build. Mater.* **2019**, *211*, 88–98.
- San-José, J.T.; Vegas, I.; Arribas, I.; Marcos, I. The performance of steel-making slag concretes in the hardened state. *Mater. Des.* 2014, 60, 612–619.
- 21. Santamaría, A.; Roji, E.; Skaf, M.; Marcos, I.; Gonzalez, J.J. The use of steelmaking slags and fly ash in structural mortars. *Constr. Build. Mater.* **2016**, *106*, 364–373.
- 22. Lee, J.-Y.; Choi, J.-S.; Yuan, T.-F.; Yoon, Y.-S.; Mitchell, D. Comparing properties of concrete containing electric arc furnace slag and granulated blast furnace slag. *Materials* **2019**, *12*, 1371.
- 23. Jiang, Y.; Ling, T.-C.; Shi, C.; Pan, S.-Y. Characteristics of steel slags and their use in cement and concrete—A review. *Resour. Conserv. Recy.* 2018, 136, 187–197.
- 24. Xu, H.; Van Deventer, J. The geopolymerisation of alumino-silicate minerals. Int. J. Miner. Process. 2000, 59, 247–266.
- Ozturk, M.; Bankir, M.B.; Bolukbasi, O.S.; Sevim, U.K. Alkali activation of electric arc furnace slag: Mechanical properties and micro analyzes. J. Build. Eng. 2019, 21, 97–105, https://doi.org/10.1016/j.jobe.2018.10.005.
- 26. Hafez, H.; Kassim, D.; Kurda, R.; Silva, R. V.; de Brito J. Assessing the sustainability potential of alkali-activated concrete from 2021, electric furnace ECO2 framework. arc slag using the Constr. Build. Mater. 281. https://doi.org/10.1016/j.conbuildmat.2021.122559.
- Bernardo, G.; Marroccoli, M.; Nobili, M.; Telesca, A.; Valenti, G. The use of oil well-derived drilling waste and electric arc furnace slag as alternative raw materials in clinker production. *Resour. Conserv. Recy.* 2007, 52, 95–102.
- 28. Muhmood, L.; Vitta, S.; Venkateswaran, D. Cementitious and pozzolanic behavior of electric arc furnace steel slags. *Cem. Concr. Res.* **2009**, *39*, 102–109.
- 29. Parron-Rubio, M.E.; Perez-García, F.; Gonzalez-Herrera, A.; Rubio-Cintas, M.D. Concrete properties comparison when substituting a 25% cement with slag from different provenances. *Materials* **2018**, *11*, 1029.
- 30. Zhao, J.; Wang, D.; Yan, P. Design and experimental study of a ternary blended cement containing high volume steel slag and blast-furnace slag based on Fuller distribution model. *Constr. Build. Mater.* **2017**, *140*, 248–256.
- 31. Amin, M.N.; Khan, K.; Saleem, M.U.; Khurram, N.; Niazi, M.U.K. Influence of mechanically activated electric arc furnace slag on compressive strength of mortars incorporating curing moisture and temperature effects. *Sustainability* **2017**, *9*, 1178.
- Roslan, N.H.; Ismail, M.; Khalid, N.H.A.; Muhammad, B. Properties of concrete containing electric arc furnace steel slag and steel sludge. J. Build. Eng. 2020, 28, 101060.
- Traven, K.; Češnovar, M.; Ducman, V. Particle size manipulation as an influential parameter in the development of mechanical properties in electric arc furnace slag-based AAM. Ceram. Int. 2019, 45, 22632–22641.
- Bernal, S.A.; Provis, J.L.; Rose, V.; Mejía de Gutierrez, R. Evolution of binder structure in sodium silicate-activated slag-metakaolin blends. *Cem. Concr. Compos.* 2011, 33, 46–54, http://doi.org/10.1016/j.cemconcomp.2010.09.004.
- Türker, H.T.; Balçikanli, M.; Durmuş, İ.H.; Özbay, E.; Erdemir, M. Microstructural alteration of alkali activated slag mortars depend on exposed high temperature level. *Constr. Build. Mater.* 2016, 104, 169–180.

- 36. Roslan, N.H.; Ismail, M.; Abdul-Majid, Z.; Ghoreishiamiri, S.; Muhammad, B. Performance of steel slag and steel sludge in concrete. *Constr. Build. Mater.* **2016**, *104*, 16–24.
- Nikolić, I.; Drinčić, A.; Djurović, D.; Karanović, L.; Radmilović, V.V.; Radmilović, V.R. Kinetics of electric arc furnace slag leaching in alkaline solutions. *Constr. Build. Mater.* 2016, 108, 1–9.
- 38. Bakharev, T.; Sanjayan, J.G.; Cheng, Y.-B. Effect of elevated temperature curing on properties of alkali-activated slag concrete. *Cem. Concr. Res.* **1999**, *29*, 1619–1625.
- Češnovar, M.; Traven, K.; Horvat, B.; Ducman, V. The potential of ladle slag and electric arc furnace slag use in synthesizing alkali activated materials; the influence of curing on mechanical properties. *Materials* 2019, 12, 1173.
- 40. Monkman, S.; Shao, Y. Assessing the carbonation behavior of cementitious materials. J. Mater. Civ. Eng. 2006, 18, 768–776.
- 41. Pan, S.-Y.; Adhikari, R.; Chen, Y.-H.; Li, P.; Chiang, P.-C. Integrated and innovative steel slag utilization for iron reclamation, green material production and CO2 fixation via accelerated carbonation. *J. Clean. Prod.* **2016**, *137*, 617–631.
- 42. Baciocchi, R.; Costa, G.; Di Bartolomeo, E.; Polettini, A.; Pomi, R. Carbonation of stainless steel slag as a process for CO 2 storage and slag valorization. *Waste Biomass Valori*. 2010, *1*, 467–477.
- 43. Europeia, U. Diretiva 98/83/CE do Conselho, de 3 de novembro de 1998, relativa à qualidade da água destinada ao consumo humano. J. Of. Das Comunidades Europeias. L 1998, 330, 32–54.
- EN-196-1. Methods of Testing Cement Part 1: Determination of Strength; European Committee for Standardization: Bruxelles Belgium, 2005; p. 36.
- 45. EN-1015-3. Methods of Test for Mortar for Masonry—Part 3: Determination of Consistence of Fresh Mortar (by Flow Table); European Committee for Standardization: Bruxelles Belgium, 1999; p. 10.
- 46. EN-1015-13. Methods of Test for Mortar for Masonry—Part 13: Determination of Dimensional Stability of Hardened Mortars; European Committee for Standardization: Bruxelles Belgium, 1993; p. 20.
- 47. EN-1015-11. Methods of Test for Mortar for Masonry—Part 11: Determination of Flexural and Compressive Strength of Hardened Mortar; European Committee for Standardization: Bruxelles Belgium, 1999; p. 12.
- 48. EN-13295. Products and Systems for the Protection and Repair of Concrete Structures. Test Methods. Determination of Resistance to Carbonation; European Committee for Standardization: Bruxelles Belgium, 2004; p. 18.
- Ozturk, M.; Akgol, O.; Sevim, U.K.; Karaaslan, M.; Demirci, M.; Unal, E. Experimental work on mechanical, electromagnetic and microwave shielding effectiveness properties of mortar containing electric arc furnace slag. *Constr. Build. Mater.* 2018, 165, 58–63.
- 50. Peys, A.; Arnout, L.; Blanpain, B.; Rahier, H.; Van Acker, K.; Pontikes, Y. Mix-design parameters and real-life considerations in the pursuit of lower environmental impact inorganic polymers. *Waste Biomass Valori.* **2018**, *9*, 879–889.
- Abdollahnejad, Z.; Jesus, C.M.; Pacheco-Torgal, F.; Aguiar, J. One-part geopolymers versus Ordinary Portland Cement (OPC) mortars: Durability assessment. In Proceedings of the 2nd International Conference on Wastes: "Solutions, Treatments and Opportunities" Braga, Portugal, 11–13 September 2013; pp. 115–120.
- 52. Shearer, C.R.; Provis, J.L.; Bernal, S.A.; Kurtis, K.E. Alkali-activation potential of biomass-coal co-fired fly ash. *Cem. Concr. Compos.* 2016, 73, 62–74.
- 53. Song, S.; Jennings, H.M. Pore solution chemistry of alkali-activated ground granulated blast-furnace slag. *Cem. Concr. Res.* **1999**, 29, 159–170.
- Wang, S.-D.; Scrivener, K.L.; Pratt, P.L. Factors affecting the strength of alkali-activated slag. Cem. Concr. Res. 1994, 24, 1033– 1043, http://doi.org/10.1016/0008-8846(94)90026-4.
- 55. Gu, Y.-m.; Fang, Y.-h.; You, D.; Gong, Y.-f.; Zhu, C.-h. Properties and microstructure of alkali-activated slag cement cured at below-and about-normal temperature. *Constr. Build. Mater.* **2015**, *79*, 1–8.
- 56. Caijun, S.; Yinyu, L. Investigation on some factors affecting the characteristics of alkali-phosphorus slag cement. *Cem. Concr. Res.* **1989**, *19*, 527–533, http://doi.org/10.1016/0008-8846(89)90004-5.
- 57. Criado, M.; Fernández-Jiménez, A.; De La Torre, A.; Aranda, M.; Palomo, A. An XRD study of the effect of the SiO2/Na2O ratio on the alkali activation of fly ash. *Cem. Concr. Res.* 2007, 37, 671–679.
- 58. Qureshi, M.N.; Ghosh, S. Effect of silicate content on the properties of alkali-activated blast furnace slag paste. *Arab. J. Sci. Eng.* **2014**, *39*, 5905–5916.
- 59. Corinaldesi, V. Mechanical behavior of masonry assemblages manufactured with recycled-aggregate mortars. *Cem. Concr. Compos.* **2009**, *31*, 505–510.
- 60. Corinaldesi, V.; Moriconi, G. Behaviour of cementitious mortars containing different kinds of recycled aggregate. *Constr. Build. Mater.* **2009**, *23*, 289–294.
- Vegas, I.; Azkarate, I.; Juarrero, A.; Frías, M. Design and performance of masonry mortars made with recycled concrete aggregates. *Mater. De Construcción* 2009, 59, 5–18.
- 62. Jiménez, J.; Ayuso, J.; López, M.; Fernández, J.; De Brito, J. Use of fine recycled aggregates from ceramic waste in masonry mortar manufacturing. *Constr. Build. Mater.* **2013**, *40*, 679–690.
- 63. Ledesma, E.F.; Jiménez, J.R.; Ayuso, J.; Fernández, J.M.; De Brito, J. Maximum feasible use of recycled sand from construction and demolition waste for eco-mortar production-Part-I: Ceramic masonry waste. J. Clean. Prod. 2015, 87, 692–706.
- 64. Corinaldesi, V. Environmentally-friendly bedding mortars for repair of historical buildings. *Constr. Build. Mater.* **2012**, *35*, 778–784.

- 65. Cuenca-Moyano, G.; Martín-Morales, M.; Valverde-Palacios, I.; Valverde-Espinosa, I.; Zamorano, M. Influence of pre-soaked recycled fine aggregate on the properties of masonry mortar. *Constr. Build. Mater.* **2014**, *70*, 71–79.
- 66. Silva, R.; De Brito, J.; Dhir, R. Performance of cementitious renderings and masonry mortars containing recycled aggregates from construction and demolition wastes. *Constr. Build. Mater.* **2016**, *105*, 400–415.
- 67. Atiş, C.D.; Kilic, A.; Sevim, U.K. Strength and shrinkage properties of mortar containing a nonstandard high-calcium fly ash. *Cem. Concr. Res.* **2004**, *34*, 99–102.