Alkali-Activated Mortars for Sustainable Building Solutions: Effect of Binder Composition on Technical Performance

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Abstract: There is a growing interest in the construction sector in the use of sustainable binders as an alternative to ordinary Portland cement, the production of which is highly impacting on the environment, due to high carbon dioxide emissions and energy consumption. Alkali-activated binders, especially those resulting from low-cost industrial by-products, such as coal fly ash or metallurgical slag, represent a sustainable option for cement replacement, though their use is more challenging, due to some technological issues related to workability or curing conditions. This paper presents sustainable alkali-activated mortars cured in room conditions and based on metakaolin, fly ash, and furnace slag (both by-products resulting from local sources) and relevant blends, aiming at their real scale application in the building sector. The effect of binder composition—gradually adjusted taking into consideration technical and environmental aspects (use of industrial by-products in place of natural materials in the view of resources saving)—on the performance (workability, compressive strength) of different mortar formulations, is discussed in detail. Some guidelines for the design of cement-free binders are given, taking into consideration the effect of each investigated alumino-silicate component. The technical feasibility to produce the mortars with standard procedures and equipment, the curing in room conditions, the promising results achieved in terms of workability and mechanical performance (from 20.0 MPa up to 52.0 MPa), confirm the potential of such materials for practical applications (masonry mortars of class M20 and M4). The cement-free binders resulting from this study can be used as reference for the development of mortars and concrete formulations for sustainable building materials production.

Keywords: alkali-activated binders; cement-free mortars; environment; resource efficiency; secondary raw materials

1. Introduction

The development of innovative, sustainable, and low-carbon dioxide (CO₂) building materials is essential for the global construction industry, which is currently required to reduce its environmental impact, mainly related to the production of ordinary Portland cement (OPC). The OPC production process is responsible for the destruction of quarries (raw materials extraction), is very energy-intensive and involves high temperatures (1400–1500 °C), with considerable associated costs, and it also releases significant emissions of greenhouse gases, such as CO₂, to the atmosphere [1,2]. The cement production industry is deemed responsible for about 8% of global CO₂ emissions [3,4], mainly due to the thermal decomposition of calcium carbonate (CaCO₃) to generate the reactive calcium silicate and aluminate...
phases, the basis of OPC. The total emissions footprint, approximately 0.8 tonnes of CO₂ equivalent per tonne of OPC produced, include emissions from the combustion of fossil fuels and those released through the conversion of CaCO₃ [5]. Approximately 40% of CO₂ emitted during cement production is related to fuel and electricity, while the remaining 60% comes from the decomposition of the main raw material (limestone, CaCO₃). Great improvements have been made in lowering the “energy-related” emissions; any attempt to reduce the remaining “chemical-related” emissions (decomposition of limestone) will have the inevitable consequence of changing the chemistry of cement. Considering materials’ availability and relevant costs, there are different oxides available as possible candidates for making cement, such as SiO₂, Al₂O₃ or CaO [6].

One type of cement-free binder that is attracting special attention consists of alkali-activated materials resulting from metakaolin (calcined clays), or industrial by-products such as pulverized fuel ash (PFA) and ground-granulated blast-furnace slag (GGBS or GGBFS) [7,8]. Metakaolin is produced from natural clays (kaolin) by calcination at 600–850 °C, while fly ash and furnace slag are materials recycled from industrial wastes produced, respectively, when coal is burnt during power generation, and iron is manufactured in a blast furnace [9]. Recycling industrial by-products to produce new materials is a driving path to promote sustainability. Alkali activation can be a very effective tool to reach this goal, especially when carbon fly ash and blast furnace slag instead of calcined natural clays (e.g., metakaolin) are used as starting materials [10,11]. Both fly ash and furnace slag are highly effective for alkaline reactions, providing readily soluble alumina and silica that undergo a dissolution–reorientation–solidification process to form new products [12–14]. The binding phases are derived by the reaction of coal fly ash, metallurgical slag or calcined clays, with an alkaline solution. The nature of precursors may result in different alkali-activated systems: (a) high-calcium systems, in particular, those based on metallurgical slag; (b) low-calcium systems, predominantly dealing with alumino-silicates and including materials known as geopolymers, or (c) intermediate systems, by blending calcium-based and alumino-silicate precursors [5]. The CO₂ savings, achieved by the use of such alkali-activated binders, are mainly due to the avoidance of CaCO₃ precursors and the high-temperature processing of conventional cements. These materials offer technical properties comparable to those of OPC, but with a much lower CO₂ footprint and with the potential for performance advantages over traditional cements in specific applications [4,15,16]. If formulated optimally, alkali activated cements made from fly ash or metallurgical slags could reduce, by 80%, the CO₂ emissions associated with the manufacturing of OPC [17]. These binders are not expected to offer a like-for-like replacement of OPC across its full range of applications, for reasons related to supply chain limitations, need for careful control of formulation, and curing or practical challenges in application mode. However, when produced using locally-available raw materials, with well-formulated mix designs and production under adequate levels of quality control, alkali-activated binders result in sustainable and cost-effective construction materials. Alkali-activated binders can be generated from a wide range of precursors, with differing availability, reactivity, cost, and value worldwide. This diversity means that these materials are locally adaptable and very versatile [18]. There are major opportunities for alkali-activated binders based upon substantial knowledge of properties and mechanisms, performance evaluations in various applications, and future orientation as environmentally friendly materials, by making use of substantial amounts of by-product and waste materials, thereby consuming less energy, generating less waste, and increasing resource efficiency [19].

The use of alkali-activated binders, to achieve environmental savings in the production of construction materials, is currently an extremely active area of research and development [20–24]. Research works carried out in developing alkali-activated binders showed their enormous potential to become an alternative to OPC. These binders are still at the early stage of development, and hence, there is a need for further investigations to become technically and economically viable construction materials [25,26]. Despite different advantages (e.g., reduced environmental impacts, technical properties, such as good early strength, especially when heat cured, or in terms of
heat and acid resistance) widespread use of alkali-activated materials in the construction industry has encountered technical, economic, and institutional barriers [27,28]. Although alkali-activated materials appear promising building materials for the future, there is still a range of concerns that need to be addressed, and some challenges to be met before they can go into mass production (e.g., curing, rheology) [29]. Research priorities in the area of alkali-activated materials include, for instance, development and optimization of mix designs based on an ever-broader range of raw materials, performance-based specifications applicable to alkali-activated cements, and validation and standardization of testing methods [18]. The main reasons for the on-going research on these materials are partially environmental, in order to reduce CO₂ emissions relative to OPC, and partially economical, in particular, when unexploited alumino-silicate sources can be used. Alkali-activated materials have been used for certain niche applications, however, only few examples were found of large-scale applications as construction materials. Hence, there is a need for further research in order to launch this technology for real [29]. The aim of this paper is to report on the technical feasibility of sustainable and robust mortars cured in normal lab conditions, and based on alkali-activated binders of metakaolin, fly ash, and furnace slag (locally sourced by-products). With the aim to maximize and promote the use of precursors from by-products, the metakaolin was replaced or combined with fly ash and furnace slag. The effect of different binder compositions on the technical performance of mortars (workability, mechanical performance) is analyzed and discussed in detail. The main objective consists in the fine tuning of material combinations that allow the preparation of innovative mortars with optimized technical performance, with high content of precursors from by-products, suitable workability for practical uses, and high mechanical resistance by curing in room conditions. Several promising formulations of alkali-activated mortars are proposed; these were developed in the view of bringing the materials on a large-scale implementation as innovative and sustainable building solutions. The research data presented in this work can be also useful to understand the effect of each typology of alumino-silicate precursor on alkali-activated binders, thus contributing to the large-scale application of sustainable construction products.

2. Materials and Methods

The binder formulations were developed using the following materials: commercial metakaolin, fly ash (by-product resulting from thermal-power plants) and furnace slag (by-product resulting from metallurgical plants), both from local sources, were used as solid alumino-silicate precursors; a commercial alkaline solution was used as activator (Baucis L160, produced by České lupkové závody, a.s.—Nové Strašecí, Czech Republic, typically used to activate metakaolin based binders), this is classified as sodium activator, and consists of Na₂O and SiO₂ similarly to sodium silicate solutions typically used for alkaline activation of alumino-silicate precursors for green binder preparation. The composition of the above-mentioned materials is reported in Table 1.

<table>
<thead>
<tr>
<th>Chemical Compounds</th>
<th>Unit Measure</th>
<th>Metakaolin (MK)</th>
<th>Fly Ash (FA)</th>
<th>Furnace Slag (FS)</th>
<th>Activator</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al₂O₃</td>
<td>%</td>
<td>42.0</td>
<td>49.0</td>
<td>28.2</td>
<td>0.0</td>
</tr>
<tr>
<td>SiO₂</td>
<td>%</td>
<td>53.0</td>
<td>23.0</td>
<td>13.9</td>
<td>21.7</td>
</tr>
<tr>
<td>CaO</td>
<td>%</td>
<td>0.0</td>
<td>2.0</td>
<td>27.0</td>
<td>0.0</td>
</tr>
<tr>
<td>Na₂O</td>
<td>%</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
<td>12.9</td>
</tr>
</tbody>
</table>

The binders—resulting from alkaline activation of metakaolin (MK), fly ash (FA), furnace slag (FS), or their blends—were combined with CEN standard sand (specifications reported in [30]) used as aggregate for mortars development. There are no specific standards for alkali-activated mortar preparation, therefore the UNI EN 196-1 national standard [31], specific for cement-based mortars was
applied, with some modifications. The reference standard recommends specific dosages of cement (binder), water (‘activator’) and sand (aggregate) for mortar preparation; the proportions, by mass, consist of 1 part cement, 3 parts sand and \( \frac{1}{2} \) part water (water/cement ratio 0.50). In the present study, the cement was totally replaced by the aluminosilicate precursors (metakaolin, fly ash, or furnace slag) and the water by the sodium activator, increasing the liquid content from 250 g of water to 302 g of alkaline activator. Similarly to the reference standard, the protocol for mortar preparation consisted in the binder/activator mixing (1st step) followed by sand addition (2nd step); the speed of mixing and the timing of each step were slightly modified, with respect to the standard, and adapted to the specific nature of the materials investigated. The total mixing time was in the order of 5 min.

The design of the investigated mortars is detailed as follows. A reference mortar, where the binder consisted of a commercial metakaolin precursor, was initially designed. Based on this formulation, the binder was replaced by other precursors from by-products (e.g., fly ash, furnace slag from local sources) or their blends, with the aim to assess the technical feasibility of mortars based on recycled materials in place of the commercial one. For each mortar formulation, the following design parameters were kept constant: total mass of solid subdivided among different precursors (Table 2), dosage of activator, and total amount of sand. The binders, depending on the composition, were grouped as: one-component binders, based on one solid precursor (Mix 1 consists of 100% MK, Mix 2 of 100% FA, and Mix 3 in 100% FS), aiming at the assessment of the performance of each component used alone; two-component binders, based on combinations of two solid precursors (Mix 4 and Mix 5 are FA/FS blends, while Mix 6 and Mix 7 are MK/FA blends) and a three-component binder, based on three solid precursors (Mix 8 consists of MK/FA/FS), aiming at the optimization of the performance of each component used alone, and trying to maximize precursors from by-products in the view of the resource efficiency.

Table 2. Design parameters of alkali-activated binders for mortars development (all the percentages are intended by mass).

<table>
<thead>
<tr>
<th>Materials</th>
<th>Mix 1</th>
<th>Mix 2</th>
<th>Mix 3</th>
<th>Mix 4</th>
<th>Mix 5</th>
<th>Mix 6</th>
<th>Mix 7</th>
<th>Mix 8</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>One-Component Binders</td>
<td>Two-Component Binders</td>
<td>Three-Component Binder</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Metakaolin MK</td>
<td>100%</td>
<td>0%</td>
<td>0%</td>
<td>0%</td>
<td>0%</td>
<td>20%</td>
<td>50%</td>
<td>25%</td>
</tr>
<tr>
<td>Fly ash FA</td>
<td>0%</td>
<td>100%</td>
<td>0%</td>
<td>50%</td>
<td>80%</td>
<td>80%</td>
<td>50%</td>
<td>25%</td>
</tr>
<tr>
<td>Furnace slag FS</td>
<td>0%</td>
<td>0%</td>
<td>100%</td>
<td>50%</td>
<td>20%</td>
<td>0%</td>
<td>0%</td>
<td>50%</td>
</tr>
</tbody>
</table>

The technical performance of the mortars were assessed in the fresh state, in terms of density and workability (flow spread) by flow table—UNI EN 1015-3 [32]. The workability evaluation involves placing a truncated conical mold in the center of the flow table, and filling it with fresh mortar; the mold is then removed, and the table jolted (15 times in about 15 s), and the diameter of the spread mortar is finally measured in two directions at right angles. Immediately after the preparation of the mortars, prismatic specimens (40 mm × 40 mm × 160 mm) were molded [33] and cured in room conditions (25 °C temperature and 50% relative humidity); these were demolded after 24 h and stored in lab conditions until mechanical testing. In the hardened state, the mortars were assessed in terms of density, and flexural and compressive strength at different curing stages—UNI EN 1015-10 [34] and UNI EN 196-1 [35]. The flexural strength evaluation involves a three-point loading method, with a vertical load applied on the mortar specimens until fracture; prism halves resulting from flexural strength test are therefore used for compressive strength assessment (Figures 1 and 2).
were tested just after the mixing operations (T0) to measure the flow spread; all the mortars were also monitored during time, and the tendency was to maintain a good degree of workability, thus showing their potential for real scale applications. The hardened mortars were tested in flexural and compressive mode at different curing stages (1, 7, 14, 28, and 60 days) and the development of mechanical performance during time was therefore assessed.

3. Results

3.1. Technical Performance of Alkali-Activated Mortars

The performance related to the developed mortars are reported in Table 3. The fresh mortars were tested just after the mixing operations (T0) to measure the flow spread; all the mortars were also monitored during time, and the tendency was to maintain a good degree of workability, thus showing their potential for real scale applications. The hardened mortars were tested in flexural and compressive mode at different curing stages (1, 7, 14, 28, and 60 days) and the development of mechanical performance during time was therefore assessed.

Table 3. Performance of optimized alkali-activated mortars.

<table>
<thead>
<tr>
<th>Performance</th>
<th>Mix 1</th>
<th>Mix 2</th>
<th>Mix 3</th>
<th>Mix 4</th>
<th>Mix 5</th>
<th>Mix 6</th>
<th>Mix 7</th>
<th>Mix 8</th>
</tr>
</thead>
<tbody>
<tr>
<td>Flow spread @ T0 (mm)</td>
<td>235.0</td>
<td>240.0</td>
<td>135.0</td>
<td>225.0</td>
<td>300.0</td>
<td>230.0</td>
<td>195.0</td>
<td>290.0</td>
</tr>
<tr>
<td>Density @ 7 days (kg/m³)</td>
<td>2291.0</td>
<td>2185.0</td>
<td>2130.0</td>
<td>2155.0</td>
<td>2134.0</td>
<td>2017.0</td>
<td>2100.0</td>
<td>2205.0</td>
</tr>
<tr>
<td>Compressive strength @ 7 days (MPa)</td>
<td>45.1</td>
<td>19.2</td>
<td>39.2</td>
<td>28.2</td>
<td>16.6</td>
<td>31.3</td>
<td>43.9</td>
<td>26.1</td>
</tr>
<tr>
<td>Density @ 28 days (kg/m³)</td>
<td>2244.0</td>
<td>2155.0</td>
<td>2190.0</td>
<td>2108.0</td>
<td>2100.0</td>
<td>2079.0</td>
<td>2205.0</td>
<td></td>
</tr>
<tr>
<td>Compressive strength @ 28 days (MPa)</td>
<td>51.7</td>
<td>29.5</td>
<td>48.0</td>
<td>34.3</td>
<td>20.2</td>
<td>39.0</td>
<td>45.8</td>
<td>40.2</td>
</tr>
<tr>
<td>Density @ 60 days (kg/m³)</td>
<td>2200.0</td>
<td>2110.0</td>
<td>-</td>
<td>2198.0</td>
<td>2067.0</td>
<td>2070.0</td>
<td>2048.0</td>
<td>-</td>
</tr>
<tr>
<td>Compressive strength @ 60 days (MPa)</td>
<td>52.0</td>
<td>29.2</td>
<td>41.7</td>
<td>35.5</td>
<td>21.1</td>
<td>45.2</td>
<td>51.5</td>
<td>-</td>
</tr>
</tbody>
</table>

Figure 1. Alkali-activated mortars: casting of prismatic samples for mechanical testing.

Figure 2. Testing on alkali-activated mortars: (a) flow spread measurement; (b) prismatic sample tested in flexural mode and aspect of a sample after a compression test.
3.2. Preliminary Durability Evaluations

Mix 8 was selected as binder formulation to further extend the experimental study. This alkali-activated binder is based on three different aluminosilicate precursors, and therefore, representative of all the materials investigated in this study (metakaolin, fly ash, and furnace slag). By a proper proportioning, this binder was combined with fine and medium aggregates (0–4 mm and 4–8 mm) for the preparation of a self-compacting concrete; this was then compared with another self-compacting concrete, based on a cementitious binder, and designed targeting the same mechanical performance. The durability was assessed by a water penetration test (UNI EN 12390-8); this showed how the alkali-activated material allows a reduction of water absorption, thus meaning a more compact structure less susceptible to eventual attacks from external factors. Technical properties (e.g., water penetration, compression strength (UNI EN 12390-3), and elastic modulus (UNI 6556)) of the alkali-activated material, monitored over time and compared with the traditional one, are included in Table 4.

Table 4. Performance comparison of alkali-activated and traditional materials.

<table>
<thead>
<tr>
<th>Performance</th>
<th>Alkali-Activated</th>
<th>Traditional</th>
</tr>
</thead>
<tbody>
<tr>
<td>Water penetration @ 28 days (mm)</td>
<td>4</td>
<td>14</td>
</tr>
<tr>
<td>Compressive strength @ 7 days (MPa)</td>
<td>37.1</td>
<td>32.0</td>
</tr>
<tr>
<td>Compressive strength @ 28 days (MPa)</td>
<td>54.6</td>
<td>42.5</td>
</tr>
<tr>
<td>Compressive strength @ 60 days (MPa)</td>
<td>58.9</td>
<td>44.0</td>
</tr>
<tr>
<td>Elastic modulus @ 28 days (GPa)</td>
<td>31.97</td>
<td>32.50</td>
</tr>
</tbody>
</table>

4. Discussion

The effect of binder composition on technical performance (e.g., consistence, compressive strength) of the prepared mortars is analyzed in this section. At first, the performance of one-component mortars (Mix 1, Mix 2, and Mix 3), totally based on metakaolin, fly ash, and furnace slag, were compared. Later, the performance of two-component mortars (Mix 4, Mix 5, Mix 6, and Mix 7) and a three-component mortar (Mix 8) of different compositions, with prevalence of by-product precursors, were discussed. The discussion deals with the relative percentage of precursors in the binder that allows the maximization of technical performance (consistence, compressive strength), taking also into consideration environmental aspects. A preliminary analysis of the binder composition is also included. The goal of this discussion is to identify cement-free binders usable for sustainable mortars, but also for green concrete development.

4.1. Effect of Binder Composition on Technical Performance of Mortars

The effect of binder composition on fresh and hardened state performance (workability and compressive strength, respectively) of different mortar formulations is reported in Figures 3–5. In order to make a more meaningful comparison among the mortars’ performance, the approach consisted in keeping fixed the total mass of binder, and gradually changing the percentage of each precursor (e.g., metakaolin, fly ash, and furnace slag); also, the activator and the aggregate dosages were kept constant, in order to only evaluate the effect of precursor composition on the final performance of each mortar.

Three mortar formulations based on one-component binders, namely 100% MK (Mix 1), 100% FA (Mix 2), and 100% FS (Mix 3), were compared. In terms of workability (Figure 3a), the use of 100% furnace slag results in the lower workability (135.0 mm), while 100% fly ash allows the maximum of
workability (240.0 mm). In terms of compressive strength (Figure 3b), the use of 100% fly ash results in the lower performance (29.5 MPa), while the use of 100% metakaolin allows the maximization of mechanical performance (51.7 MPa). Even if fly ash and metakaolin have comparable workability, the latter resulted in better mechanical performance; furnace slag has similar mechanical performance of metakaolin, but its workability is lower.

![Figure 3](image_url)

**Figure 3.** Technical performance comparison for mortars based on 100% metakaolin (Mix 1), 100% fly ash (Mix 2), and 100% furnace slag (Mix 3): (a) flow spread and (b) compressive strength at 28 days.

Based on the outcomes of the previous investigations, two-component binders were tested, aiming at maximizing the performance of each of them considered individually, and also adopting sustainability requirements. Taking into consideration the workability of the individual binders, it was decided to combine furnace slag (lower workability) with fly ash being the best performing material with respect to this parameter. Furnace slag was reduced from 100% (Mix 3) up to 50% (Mix 4) or 20% (Mix 5), and replaced by fly ash. Comparisons of workability and strength at 28 days for furnace slag/fly ash blends are reported, respectively, in Figure 4a,b. Overall, with respect to Mix 3 (100% FS), an improvement of workability was obtained for Mix 4 and Mix 5, due to the presence of fly ash; while the mechanical performance was reduced with respect to neat furnace slag with a maximum of 34.3 MPa achieved by Mix 4 (versus 48.0 MPa of Mix 3). Mix 4 (225.0 mm of consistence, 34.3 MPa
of strength) seems the most suitable for practical applications among furnace slag/fly ash blends. This binder combination is also sustainable, being based on by-product precursors. Moreover, taking into consideration the strength of the individual binders, it was decided to combine fly ash (lower mechanical performance) with metakaolin, being the best performing material with respect to this parameter. Fly ash was reduced from 100% (Mix 2) up to 80% (Mix 6) or 50% (Mix 7), and replaced by metakaolin. Comparisons of workability and strength at 28 days for fly ash/metakaolin blends are reported, respectively, in Figure 5a,b. Mix 6 (20% MK) has a workability comparable with Mix 1 (100% MK) and Mix 2 (100% FA), while Mix 7 (50% MK) is slightly below, meaning that fly ash/metakaolin interaction was not beneficial in these dosages, at least in the fresh state. On the other side, the mechanical performance was improved with respect to neat fly ash, due to the presence of metakaolin, with a maximum of 45.8 MPa achieved by Mix 7 (versus 29.5 MPa of Mix 2). Mix 7 (195.0 mm of consistence, 45.8 MPa of strength) seems the most suitable for practical applications among fly ash/metakaolin blends. This binder combination is, in addition, more sustainable than metakaolin used alone, since it also incorporates a by-product precursor.

Figure 4. Technical performance comparison for mortars based on furnace slag/fly ash blends: (a) flow spread and (b) compressive strength at 28 days.
In addition, the behavior of a three-components mortar (Mix 8) was also investigated. The following composition was selected for the formulation of the relevant binder: 25% of metakaolin, 25% of fly ash, and 50% of furnace slag. From the previous experiments, on one hand, it was evidenced that metakaolin was the best performing in terms of mechanical performance, followed by furnace slag; on the other hand, fly ash improved the workability, but metakaolin also had a satisfactory effect on this property. Taking into consideration the need to reduce the use of natural resources (metakaolin is produced from kaolin clay by calcination, a thermal treatment involving high temperatures) and, in the view of resources efficiency, its amounts were reduced in favor of binders resulting from by-products (fly ash, furnace slag). Based on that, metakaolin was reduced up to 25%, and the remaining 75% consisted in furnace slag and fly ash. The technical performance of Mix 8 resulted in 290.0 mm in terms of flow spread—comparable with Mix 5—and 40.2 MPa of strength at 28 days—comparable with Mix 6. A complete overview of all the mortars developed in this study is reported in Figure 6;
this compares the performance achieved for mortars based on one-component binders (see Figure 3), two-component binders (see Figures 4 and 5), and three-component binder (Mix 8). Overall, all the mortars developed seem suitable for real scale applications; the workability is acceptable in almost all the cases (except for Mix 3), the compressive strength ranges from approximately 20.0 MPa up to 52.0 MPa, after 28 days of curing, that comply with the specifications for masonry mortars [36] of class M_{20} (resistance in compression from 20.0 MPa up to 24.9 MPa) or even M_{d} (resistance in compression higher than 25.0 MPa). Among the mortars tested, a couple of formulations (Mix 3 and 7) with mechanical performance comparable to metakaolin used alone (Mix 1) were identified. These mortars are based, respectively, on furnace slag and fly ash/metakaolin, thus encouraging the use of industrial by-products in place of natural materials for alkali-activated mortar development. It has to be also considered that, based on previous experiments carried out in the lab, mortars based on conventional cementitious binders (OPC) with the same dosages can reach, after 28 days of curing, 49.8 MPa using CEM I 42.5, and 40.7 MPa using CEM IV 32.5. Therefore, the performance of the cement-free mortars developed in this study can be considered promising alternative of conventional mortars.

**Figure 6.** Overview of technical performance of the developed mortars: (a) flow spread and (b) compressive strength at 28 days’ comparison.
4.2. Effect of SiO$_2$, Al$_2$O$_3$, CaO, and Na$_2$O on Technical Performance of Mortars

For the developed mortars, a preliminary analysis, in terms of the main constituents of the alumino-silicate precursors and alkaline activator (e.g., SiO$_2$, Al$_2$O$_3$, CaO and Na$_2$O), was performed and reported in Table 5. For each mortar formulation, the data were calculated based on the composition of constituent precursors and activator (see Table 1). Different parameters were evaluated, considering both binder and activator composition: SiO$_2$ + Al$_2$O$_3$, SiO$_2$/Al$_2$O$_3$, CaO, M+ (alkali dosage) and AM (alkali modulus). M+ was calculated as ratio of Na$_2$O mass in the activating solution to the binder mass; this parameter, being fixed in both the activator and total mass of the binder, is equal to 2.9% for all the binder formulations. AM was calculated as the mass ratio of sodium oxide (Na$_2$O) in the activating solution to total amount of silica (SiO$_2$); this parameter ranges from 0.17 up to 0.26 for the investigated formulations. The trend of workability and compressive strength for the mortars investigated in this study are reported in Figure 7. The minimum workability corresponds to the maximum compressive strength and vice versa; therefore, AM values close to 0.21 allow the maximization of consistence, while AM values close to 0.17 and 0.26 allow the maximization of compressive strength. The present analysis has to be considered just a preliminary evaluation; further investigations are required to assess the actual reactivity of the chemical components provided by the precursor in the alkaline solution, and monitor the reactions among the alumino-silicate precursors and the alkaline activator, as well as the resulting chemical compounds, in the final product (chemical and microstructural characterization).

![Graphs](image-url)

**Figure 7.** Effect of alkali-modulus (AM) on technical performance of the developed mortars: (a) flow spread and (b) compressive strength trend.
Table 5. Analysis of the composition of optimized alkali-activated binders.

<table>
<thead>
<tr>
<th>Main Constituents/Parameters</th>
<th>Mix 1</th>
<th>Mix 2</th>
<th>Mix 3</th>
<th>Mix 4</th>
<th>Mix 5</th>
<th>Mix 6</th>
<th>Mix 7</th>
<th>Mix 8</th>
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<tbody>
<tr>
<td>Binders composition</td>
<td></td>
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<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>SiO$_2$ + Al$_2$O$_3$ (g)</td>
<td>116.70</td>
<td>93.70</td>
<td>63.80</td>
<td>78.75</td>
<td>87.72</td>
<td>98.30</td>
<td>105.20</td>
<td>84.50</td>
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<tr>
<td>SiO$_2$/Al$_2$O$_3$</td>
<td>1.78</td>
<td>3.07</td>
<td>3.59</td>
<td>3.27</td>
<td>3.14</td>
<td>2.67</td>
<td>2.24</td>
<td>2.64</td>
</tr>
<tr>
<td>CaO (g)</td>
<td>0.00</td>
<td>2.00</td>
<td>27.00</td>
<td>14.50</td>
<td>7.00</td>
<td>1.60</td>
<td>1.00</td>
<td>14.00</td>
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<tr>
<td>Na$_2$O (g)</td>
<td>12.90</td>
<td>12.90</td>
<td>12.90</td>
<td>12.90</td>
<td>12.90</td>
<td>12.90</td>
<td>12.90</td>
<td>12.90</td>
</tr>
<tr>
<td>AM (Alkali modulus)</td>
<td>0.17</td>
<td>0.18</td>
<td>0.26</td>
<td>0.21</td>
<td>0.19</td>
<td>0.18</td>
<td>0.18</td>
<td>0.21</td>
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</tbody>
</table>

<table>
<thead>
<tr>
<th>Mortars performance</th>
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</thead>
<tbody>
<tr>
<td>Flow spread @ T0 (mm)</td>
<td>235.0</td>
<td>240.0</td>
<td>135.0</td>
<td>225.0</td>
<td>300.0</td>
<td>230.0</td>
<td>195.0</td>
<td>290.0</td>
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<tr>
<td>Compressive strength (MPa)</td>
<td>51.7</td>
<td>29.5</td>
<td>48.0</td>
<td>34.3</td>
<td>20.2</td>
<td>39.0</td>
<td>45.8</td>
<td>40.2</td>
</tr>
</tbody>
</table>

5. Conclusions
The effect of binder composition on the technical performance (workability, compressive strength) of sustainable alkali-activated mortars, cured in normal lab conditions and mainly based on alumino-silicate precursors from by-products (fly ash and furnace slag as replacement of metakaolin), was discussed in this study. The main outcomes of this study are listed below:

- general guidelines to monitor the performance (e.g., workability, compressive strength) of alkali-activated mortars by the binder composition are proposed;
- the curing in room conditions, the use of standard procedures and equipment, the suitable workability and the high compressive strength (ranging from 20.0 MPa to 52.0 MPa) confirm the potential of the investigated mortars for masonry applications ($M_{20}$ and $M_{44}$ class);
- overall, the developed formulations seem suitable as alternatives to traditional cementitious mortars, and their performance are competitive with those based on conventional binders;
- the developed alkali-activated binders, when used in concretes, resulted in superior durability performance (reduced water penetration) in comparison with conventional binders;
- the investigated cement-free binders can be used as reference for sustainable building materials (e.g., mortars, concretes) development, thus opening new perspectives for a more sustainable construction sector.

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Author Contributions: Agnese Attanasio analyzed the data and wrote the paper; Livio Pascali and Wanda Arena conceived and designed the experiments; Livio Pascali performed the experiments with the technical contribution of Vito Tarantino; Alessandro Largo supervised the technical activities and the paper writing.

Conflicts of Interest: The authors declare no conflict of interest.

References


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