Development of Sustainable Alkali-Activated Mortars Using Fe-Rich Fayalitic Slag as the Sole Solid Precursor

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Vast amounts of water-cooled non-ferrous metallurgy slags are generated yearly, and significant amounts are unutilized or dumped in landfills. To address this issue, in this study, MgO-FeOx-SiO$_2$ fayalitic slag (FS) was used as the sole solid precursor (as an aggregate and binder) in alkali-activated mortars. The performance of the mortar samples was analyzed in terms of workability, density, compressive strength, and ultrasonic pulse velocity. The microstructural properties and binder composition of the samples were studied using a scanning electron microscope (SEM) coupled with energy dispersive X-ray spectroscopy (EDS). Experimental results revealed that mortar samples made with FS aggregates performed better, achieving a 28-day compressive strength of 21 MPa compared to mortars produced with standard sand aggregates, which gained compressive strengths of 9 MPa. Further optimization of the particle size distribution of FS aggregate-based mortar samples using particle packing technology improved the workability, densified the mortar and yielded a mechanical performance of up to 40 MPa. FS aggregates have better interfacial bonding with the binder gel compared to standard sand, and the FS aggregates participate in the hardening reactions, consequently affecting the final binder phase composition, which consists of a Na$_2$O-Fe$_2$O$_3$-SiO$_2$ gel with lower quantities of CaO, MgO, and Al$_2$O$_3$. Therefore, the alkali-activated mortars produced based on the optimization of fully recycled industrial residues can provide a pathway for the sole utilization of metallurgical by-products, which can have a wide range of structural applications.

Keywords: Fe-rich fayalite slag, aggregate, binder, alkali activation, mortar, interface region, ultrasonic pulse velocity, alkali-activated materials

HIGHLIGHTS

- Fayalite slag upcycling as sole solid precursor in alkali-activated mortars was demonstrated.
- Fayalite slag aggregate-based mortars have superior mechanical and microstructural properties than standard sand aggregate-based mortars.
- The properties of fayalite slag aggregate-based mortars were enhanced by particle packing.
- The chemical composition of interface region and binder gel was influenced by the aggregate type.
- The study provides a new sustainable pathway for full recycling of fayalite slag for use in construction industry.
INTRODUCTION

The increasing usage of Portland cement (PC) concrete in the construction industry and its consequences have been widely studied (Ali et al., 2011; Shi et al., 2011). Some of the critical issues identified include CO$_2$ emissions during cement production, depletion of natural resources and the disruption of the natural ecosystem such as air, water, and soil contamination due to mining and the quarrying of aggregates (Mo et al., 2016).

To reduce the environmental impact from the cement and concrete industry, several alternatives have been proposed. Researchers have explored the possibility of replacing PC partially or wholly with various cementitious industrial residues (Alex et al., 2016; Provis, 2018). One notable breakthrough is the development of alternative alkali-activated binders from industrial residues, such as ground granulated blast furnace slag (GGBFS), coal fly ash, and more recently, mineral wools (Mo et al., 2015; Zhao et al., 2015; Provis et al., 2018; Carvelli et al., 2020; Lemougna et al., 2020). While GGBFS and coal fly ash have been widely researched and successfully applied in alkali-activated cements and concrete, they are already utilized almost fully as supplementary cementitious material (SCM) (Scrivener et al., 2016). Thus, other potential industrial residues should be investigated as precursors for alkali-activated materials (AAMs). One recently studied residue is fayalite slag (FS) (Mo et al., 2015, 2018).

Fayalite slag is an Fe-rich industrial residue and a non-ferrous metallurgy slag generated from nickel and copper production, and significant amounts of this slag remain unutilized (Marangoni et al., 2016). This slag is named FS due to its main mineralogical crystalline component. The chemical properties of the slag are influenced by its cooling process and cooling rate. Slowly cooled FS results in a hard, crystalline product, while the water cooling of FS enhances quick solidification, resulting in an amorphous/glassy slag (Fan et al., 2014; Siakati et al., 2020). Water-cooled FS comes in a granular form with particle sizes ranging from 0 to 4 mm, and its production has continued to rise yearly with the increasing demand for nickel and copper (Michailova and Mehandjiev, 2010). For example, in Finland, approximately 200,000 tons of water-cooled FS are produced every year, most of which is currently landfilled but can potentially be utilized as AAM precursors (Saari et al., 2019).

In the last few years, several Fe-rich materials have been investigated for their suitability as precursors for AAMs. Such materials include bauxite residue slags, low calcium slags from ferronickel production, lead and zinc slags, laterite soils and Fe-silicate glasses from various mining residues (Komnitsas et al., 2007; Hertel et al., 2016; Machiels et al., 2017; Millán-Corrales et al., 2020; Kaze et al., 2021a; Rodrigue Kaze et al., 2021). Similar to other Fe-rich materials, FS is suitable for use as a precursor for AAMs (Komnitsas et al., 2020). The presence of amorphous material in FS is beneficial for its reactivity in terms of alkali activation (Onisei et al., 2015, 2018). The amorphous phase of FS is the phase with the most substantial reactivity, and it can be used as a binder precursor if milled into fine particle sizes (d$_{50}$ ∼10 µm) and mixed with an alkali activator (mostly alkali hydroxide and/or alkali silicate) (Machiels et al., 2014; Onisei et al., 2015).

The reactivity of FS in alkaline solution is quite low compared to those of aluminosilicate precursors, reasonably due to their distinct chemical and mineralogical composition (Onisei et al., 2015). FS is rich in iron oxide and silica with lower amounts of calcium and alumina, which affects its reactivity and the binder formed. Alkali-activated FS binders have been reported to present a unique reaction pathway compared to other aluminosilicate precursors (Peyes et al., 2019b,c). After the initial dissolution of FS, a complex reaction mechanism results in the formation of two products. The first reaction product is formed as a result of the precipitation of Fe$^{2+}$; Fe$^{3+}$ ions are arranged in an octahedral configuration. The second reaction product is related to the silicate network; it contains an Fe$^{3+}$ charge located in the...
tetrahedral bonding network of the binder gel, with Na playing a charge balancing role (Peys et al., 2019b,c). Thus, iron plays an important role in the binder gel and displays a different nanostructural chemistry from conventional AAMs.

Different studies have reported the use of Fe-rich precursors in alkali-activated mortars. Researchers have shown that alkali-activated mortars can be obtained from a mixture of a milled precursor and standard sand (i.e., a milled Fe-rich precursor is used as the binder while sand is used as an aggregate) (Komititas et al., 2007, 2019; Kalinkin et al., 2012; Machiels et al., 2014; Albitar et al., 2015; Nazer et al., 2016; Iacobescu et al., 2017; Xia et al., 2019). However, the water-cooled FS used in this study can also be utilized without the need for milling as it has a suitable particle size for sand replacement in mortars. Preliminary tests on the use of FS as an aggregate and binder in AAMs showed that the mortar samples with FS aggregates had superior mechanical properties compared to mortar samples with standard sand aggregates (Adediran et al., 2019). However, considering the beneficial use of this industrial residue for dual purposes (i.e., as a natural aggregate and for cement replacement), further investigations are necessary to obtain a comprehensive understanding of the behavior of FS as an aggregate and its influence on the chemical composition of the binder phase formed.

This study provides detailed information on the use of FS as a sole solid precursor (aggregate and binder) in AAMs. The study focuses on the optimization of the mortars based solely on FS to achieve optimal performance and to provide detailed information on the binder gel composition and the microstructure near the aggregates. Three mix compositions with similar binder types and amounts of alkali activators were developed. The reference mix contained standard sand aggregates, and the second mix contained FS aggregates. However, the third mix was formulated using particle packing, and the influence on the properties of alkali-activated slag mortars was investigated. The physical and chemical properties of the prepared mortars were assessed, and the mineralogy was determined using X-ray diffraction (XRD). The workability of the fresh mortar samples was determined using a flow table test. Scanning electron microscope (SEM) and energy dispersive X-ray spectroscopy (EDS) were used to determine the microstructure and elemental composition of the binder phase formed. The ultrasonic pulse velocity (UPV) and compressive strength of the mortar samples were measured at 3, 7, and 28 days of curing. The results of the FS aggregate-based samples obtained were compared with those of the reference mix. This study provides a good understanding of the microstructural evolution that takes place between FS aggregates and FS binders and provides a new pathway for recycling FS as a sole solid precursor in AAMs.

MATERIALS AND METHODS

Experimental

Industrial water-cooled FS, which originated from nickel production, was obtained from Bolden Harjavalta Oy (Finland). The chemical composition of FS as determined using X-ray fluorescence (PANalytical Omnium Axiosmax) from a melt-fused tablet and loss on ignition (LOI, measured with PrepAsh, Precisa) is shown in Table 1. The major oxides present in FS are Fe2O3, SiO2, and MgO along with other minor oxides, and it is similar to other slags obtained from copper production (Michailova and Mehandjiev, 2010; Adediran et al., 2021). In FS, iron may exist as Fe2+ and Fe3+, but it is reported here as Fe2O3 due to the X-ray fluorescence sample pre-treatment using thermogravimetric analysis at 950°C during which the iron oxidizes mostly into Fe3+, depicted by the negative LOI in Table 1.

X-ray diffraction patterns of FS were determined using a Rigaku Smartlab diffractometer, with a Cu K-beta radiation and a scanning rate of 0.02°/step from 5–80°, operated at 135 mA and 40 kV. The quantification of the amorphous phases was done with 10 wt.% rutile (TiO2) as the internal standard. The Rietveld refinement method was used for the qualitative and quantitative analysis of the crystalline phase (Rietveld, 1969).

X-ray diffraction results in Table 2 revealed that FS consisted mainly of fayalite [(Fe2SiO4), pdf. 04-007-9022] and magnetite [(Fe3O4), pdf. 04-008-8145] as the crystalline components while the rutile [(TiO2), pdf. 04-008-7847] peak present in Figure 1 is as a result of the internal standard used. The amorphous fraction (55.5 wt.%) in FS is higher than that of the crystalline phase, and the amorphous phase is the only phase with significant reactivity (Iacobescu et al., 2017; Onisei et al., 2018).

Standard sand conforming to European standard EN 196-1 (EN 196-1, 2016) with a particle size distribution (PSD) between 0.08 and 2 mm and a maximum moisture content of 0.2%, was used as the reference aggregate. The FS was moist when received and then dried in the oven at a temperature of 60°C overnight to remove the moisture. The slag was then sieved using a 2 mm mesh to remove particles >2 mm, and the PSD was determined using the sieving method (EN 933-1, 2000). The sieve analysis results revealed that the PSD of FS was different from that of standard sand. Thus, the PSD of FS was modified to achieve a PSD similar to that of standard sand (named "FSi"). To achieve optimized particle packing, the FS was ball-milled for 30 min to achieve a median particle size (d50) of 248 μm; this was optimized with FS by particle packing technology using the Elkem Material Mixture Analyzer (EMMA) based on a modified Andreasen’s particle packing model. A distribution coefficient (q) of 0.29 was used, and the particle size was fixed between 0.1 and 2000 μm. The optimization curve for the aggregate fraction can

| TABLE 1 | Chemical composition of FS as weight-%. |
|---------|----------------------------------------|
| Fe2O3   | 52.5                                   |
| SiO2    | 34.4                                   |
| MgO     | 6.8                                    |
| Al2O3   | 2.4                                    |
| CaO     | 1.9                                    |
| Others  | 1.8                                    |
| LOI at 950°C | −0.2        |

| TABLE 2 | Mineralogical composition of FS as weight-%. |
|---------|---------------------------------------------|
| Fayalite| 42.4                                        |
| Magnetite| 2.1                                      |
| Amorphous| 55.5                                    |
be found in the Supplementary Material. This aggregate fraction is named “FS2”.

Fayalite slag to be used as a binder precursor (named “FS binder”) was ball-milled for three hours in a laboratory tumbling ball mill (Germatec, Germany) to achieve a $d_{50}$ of 10 µm. The PSD was analyzed using a Beckman Coulter LS 13320 laser diffraction particle size analyzer. Water was used as a dispersion medium during measurement. The PSD of the aggregates and the binder are presented in Figure 2.

Fayalite slag has a higher specific gravity than that of standard sand (3796 vs. 2674 kg/m³) due to its high iron content. In addition, the bulk density of FS (2192 kg/m³) is higher than that of standard sand (1544 kg/m³), indicating that the use of FS as an aggregate may produce an alkali-activated mortar of higher density. The fineness modulus of FS is similar to that of standard sand, which can be obtained from the PSD (Figure 2). The water absorption of FS is 0.4% compared with 0.2% for standard sand. The shape of the particle as viewed using the optical microscope indicated that FS particles are angular in shape with no roundness, but those of standard sand are mostly rounded in shape and less angular (Figure 3).

Sample Preparation

The alkali-activated mortars were prepared according to the following procedure. The alkaline solution utilized for the synthesis was a mix of a 10 M sodium hydroxide solution and an analytical grade sodium silicate solution (Merck, United States). The sodium silicate solution used was composed of 65.5% H₂O (Na₂O: 7.5–8.5%, SiO₂: 25.5–28.5%, density: 1.296–1.396 g/ml). A sodium hydroxide solution was prepared using analytical
grade sodium hydroxide pellets (>98% purity; VWR Chemicals) dissolved in deionized water. The choice of a 10 M sodium hydroxide solution for the synthesis was based on the results of the preliminary experiment and on previous studies on the alkali activation of FS (Adediran, 2017; Adediran et al., 2019). The mass ratio of sodium silicate to sodium hydroxide was 50:50. The final molar ratios of the activating solution were (SiO$_2$/Na$_2$O = 1.0) and (H$_2$O/Na$_2$O = 15.7). The solution was prepared 24 h before mixing.

Three mix designs were developed based on the knowledge of the properties of all ingredients and presented in Table 3. The water to binder ratio for all the mortars was held constant at 0.30. The reference mix consisted of FS binder and standard sand aggregates. However, systematic modification was performed on the mix design of the samples that had FS aggregates. The mass fraction of aggregates used in AAFS1 and AAFS2 were more than that of standard sand used in the reference mix due to the higher density of FS. Meanwhile, the volume fraction of the aggregates for all the mixes remained the same and was kept constant at 3. The type and amount of the binder component used was also held constant for all the mixes regardless of the aggregate type to ensure a similar amount for sample preparation.

The mixing of all the mortar samples was done mechanically in accordance with the EN 196-1 standard (SFS-EN 196-1, 2011). The workability of fresh mortar mixtures was evaluated using a flow table test in accordance with the EN 1015-3 standard (SFS-EN 1015-3, 1999). The mixture was then cast into a 20 × 20 × 80 mm$^3$ oiled rectangular prism mold for strength measurement and a 40 × 40 × 160 mm$^3$ mold for UPV measurement, and a jolting machine (60 shocks, 1/s) was used for compacting. All the samples were sealed in a plastic bag and cured at 60°C for 24 h. The samples were then kept at room temperature to cool down before being tested. The density of hardened samples after 28 days of curing was determined according to ASTM C642 recommendations (ASTM C642, 2013). All the mortar samples were first dried in the oven at a temperature of 110°C for 24 h. The samples were then kept at room temperature to cool down before weighing.

Microstructural Characterization
The microstructure of the samples was studied using SEM-EDS analysis. The reliability of the results obtained from SEM analysis depends on the quality of the surface of mortar samples.
 RESULTS AND DISCUSSION

Workability and Density of Alkali-Activated Mortars

The workability of all the mortar samples prepared is presented in Table 4. AAFS2 had the maximum workability (169 mm). The workability of AAFS1 was 146 mm and that of the reference mix was 160 mm. The high workability of AAFS2 can be attributed to the packing effect (Supplementary Material) as the higher content of fine particles in the matrix of AAFS2 was able to fill up the spaces between the larger particles, allowing lubrication of the mix. The difference in the workability values of AAFS1 and the reference mix could have been as a result of aggregate properties. The higher workability of the reference mix compared to AAFS1 may be due to the rounded particle shape of standard sand aggregates. Similar observations of high workability have also been reported when round-shaped natural river sand was used as an aggregate unlike the angular-shaped crushed limestone aggregate in fly ash geopolymer mortar (Mermerdaş et al., 2017).

The density of hardened AAFS2 mortars was higher compared to that of AAFS1 mortar samples due to the better packing of particles. Furthermore, the density of the mortar samples prepared with FS aggregates (AAFS2 and AAFS1) were higher than the reference mortars with standard sand aggregates. This can be attributed to the high specific gravity of FS.

| TABLE 3 | Mix proportion of mortar samples. |
| Sample name | Aggregates (g) | FS binder (g) | 10 M NaOH + Na-Sil (g) | w/b** |
|-------------|----------------|--------------|----------------------|-------|
|             | Sand | FS1 | FS2* | |
| Reference   | 804  | –   | 396  | 200  | 0.3 |
| AAFS1       | –    | 1139.9 | –   | 396  | 200  | 0.3 |
| AAFS2       | –    | –   | 1139.9 | 396  | 200  | 0.3 |

*FS2 consists of 560.5 g of as-received FS and 579.4 g of milled FS with d50 of 248 μm.
**w/b is water to binder ratio.

| TABLE 4 | Workability and density values of the alkali activated mortar samples. |
| Sample name | Mean workability (mm) | Density (kg/m$^3$) |
|-------------|----------------------|--------------------|
| Reference   | 160 ± 1.3            | 2228               |
| AAFS1       | 146 ± 1.2            | 2688               |
| AAFS2       | 169 ± 1.2            | 2931               |

Compressive Strength

The compressive strength of AAFS2 was higher than those of AAFS1 and the reference mix as presented in Figure 4. The compressive strength of both AAFS2 and AAFS1 after 28 days of curing was relatively good (i.e., 40 and 21 MPa) and can be suitable for structural clay load-bearing wall tiles, pedestrian and light traffic paving bricks (ASTM C34-03, 2003), for example. The compressive strengths also satisfied the ASTM C62.99 recommendation of 20.7 MPa for building bricks (ASTM C62, 1999).

The dense packing of aggregates and binders had a remarkable positive effect on the compressive strength of AAFS2 mortar samples. AAFS2 had the highest early age strength at 3 days followed by AAFS1, and a much lower early age strength was observed in the reference mix. The introduction of small-sized aggregates in the matrix formulation of AAFS2 via particle packing might have contributed to its mechanical behavior at early and late age when compared to AAFS1. The optimum packing of aggregates and the binder is believed to have reduced porosity and void in the mortar and increased the affinity between the mortar components, which is very important for the mechanical properties of the mortar. The strength achieved in AAFS2 is comparable to those obtained for alkali-activated composites made from calcined iron-rich laterite soil cured at elevated temperature (Kaze et al., 2021b).

The difference in compressive strength between the reference and AAFS1 samples could be ascribed to the angular shape of the FS aggregates and chemical bonding between FS aggregates and the FS binder (later shown in section “SEM-EDS”). Angular particles have been reported to have higher bonding characteristics, which create stronger interlinking of the particles as a result of the higher surface to volume ratio (Mermerdaş et al., 2017; Lyu et al., 2019). The observation in this study is consistent with the findings of other researchers on the use of other iron-rich slags, such as copper slag and ferronickel slags, as aggregate replacements in cement-based mortars and concrete paving blocks (Ayano and Sakata, 2000; Al-Jabri et al., 2011; Dimitrioglou et al., 2015; Mithun and Narasimhan, 2015). When copper slags were used as fine and coarse aggregates, the mechanical properties were similar or superior to those of the reference mixtures containing sand. Additionally, copper slags were found to have higher compressive strength as coarse aggregates in high strength concrete than limestone aggregates as a result of the stronger bonding between copper slag aggregates and the matrix (Khanzadi and Behnood, 2009). Thus, the lower strength exhibited by the reference mix may likely be due to the low chemical interaction between aggregates and the binder (see section “SEM-EDS”) and the round shape and smooth surface of standard sand, which led to a lower bonding strength with the FS binder.

Ultrasonic Pulse Velocity

AAFS2 mortar samples had higher UPV values compared to AAFS1 and reference samples as shown in Figure 5. This indicates that there were less voids and defects in AAFS2, and the matrix was denser and more compact compared to AAFS1 and...
reference samples, assuming that all the samples have comparable stiffness. The reason for higher UPV values in AAFS2 can be attributed to densified particle packing of aggregates and the binder, which prevents the formation of internal voids in the mortar thereby increasing the mortar quality. Hence, the internal structure of mortars can be improved by particle packing. Furthermore, the aggregate type used can influence the mortar microstructure. Reference mortars prepared with round-shaped standard sand had lower UPV values than AAFS1 mortars with angular FS aggregates as a result of weaker bonding between the standard sand aggregate and the binder (Figure 7). This is in line with similar observations from other studies, which have reported that aggregate type can influence the prediction of UPV values of composite mixtures (Trtnik et al., 2009). The increase of UPV values for all the mortars correlates with the rate of strength development in Figure 4. A similar correlation between the UPV and compressive strength has been reported in fly ash-based geopolymer concrete and blast furnace slag-based concrete (Omer et al., 2015; Ghosh et al., 2018).

**SEM-EDS**

The SEM micrographs with 500× and 2000× magnifications of representative sections of mortar samples after 28 days of curing are presented in Figures 6, 7. The micrographs of the reference sample (Figure 6A) appear to show a weak interface between the standard sand aggregate and the binder gel formed, which becomes more visible at higher magnifications (Figure 7A). The standard sand aggregate appears intact with bond cracks at the interface, suggesting that the chemical bonding between sand particles and the binder gel is relatively low. The reason for this low interaction might be that standard sand consists of crystalline quartz, which is not dissolved by the activating solution and does not interact with the binder.

In comparison to the reference mix, AAFS1 mortar samples in Figure 6B showed a less distinct interface between the FS aggregates and the surrounding binder gel formed. Only the iron silicate glass phase dissolved to form the binder gel, and the crystalline phases were left over and tightly embedded in the binder matrix (Figure 7). This can be observed particularly in Figure 7C where the region circled with white rings shows excessive dissolution of the glass phase during alkali activation. Thus, FS aggregates seem to chemically bind and adhere more closely to the binder gel (Figure 7B), exhibiting a more compact and stronger interface. The micrograph of AAFS2 mortar samples in Figure 6C appears denser with less crack propagation at the interface when compared to that of AAFS1, indicating higher affinity and bonding between the aggregates and the binder gel formed and is consistent with the compressive strength and UPV results in sections “Compressive Strength” and “Ultrasonic Pulse Velocity.” The reactivity of FS aggregates is further confirmed by the presence of more Fe ions in the interface region compared to reference mortars according to EDS (data not shown). Similar higher concentration of calcium was observed at the interface region when calcium-rich steelwork aggregate was used as aggregate in fly ash based mortar samples (Cristelo et al., 2019).
In all the mortar samples, the partially dissolved and unreacted slag particles acted as small aggregates and fillers in the matrix (Figures 6A–C, 7A–C). These unreacted slag particles, as revealed by the microstructure, are relics of fayalite crystals and magnetite, consistent with the crystalline phase observed in XRD analysis shown in Figure 1. Particularly in AAFS2, a high volume of binder gel formed cover the surface of FS particles at the interface region, ascribed to the beneficial effect of the addition of small-sized aggregates to the matrix. This implies that as AAFS2 contained more small-sized particles compared to AAFS1, the particles did not only act as a filler to improve particle packing, but also partly dissolved, thereby contributing additional Fe ions to the interface region.

Furthermore, there was a difference in the size of particles located in the interface region compared to that of bulk binder region in AAFS2 (Figure 7C). The size of the particles near the interface region appeared smaller, with the core of unreacted particles located in the bulk binder (Figure 7C). One possible explanation could be that due to better particle packing, the smaller particles were able to fill the voids between larger particles and were closely packed through the surface of the aggregates. Smaller-sized particles close to the interface had higher surface areas in comparison to larger particles further out, leading to a higher release of ions such as Fe into the interface region. As a result, a denser microstructure was formed in the interface region, and this is important for the long-term performance of the mortar.

Crack formation, known as “matrix crack,” was noticeable in the binder gel of all the samples, and this became more pronounced at higher magnifications (Figures 7A–C). The cracks formed penetrated through the binder gel and sometimes terminated at the aggregate boundary. The crack formed could likely be associated with specimen preparation, exposure to vacuum conditions during SEM analysis or curing of samples as the samples were cured at an elevated temperature of 60°C. A similar network of cracks has also been reported when copper production-based FS was used as precursor for AAM and cured at 60°C (Iacobescu et al., 2017).

The EDS point analysis of binder gel formed for all the mortar samples are summarized in Figure 8. EDS analysis has some drawbacks as it is only semi-quantitative, and it is possible that data points represent a mixture of several binder phases or undissolved particles if they are mixed on a scale smaller than the depth of X-ray generation for the 15 kV accelerating voltage used (i.e., <5 µm³). However, the average and standard deviation of several points located on the studied phase could give an insight into the composition of the phase. The elemental composition of the binder gel formed in all the mortar samples was compared with that of the glass phase of FS. The binder gel formed in all the samples after 28 days of curing consisted mainly of Si, Fe, and Na, with lower quantities of K, Ca, Al, and Mg. This is in line with earlier study of alkali-activated Fe-silicate glasses (Machiels et al., 2014). However, in the mentioned study, the slag composition was slightly different as it had more Ca and less Mg, which may explain why their binder had higher concentrations of Ca in the
binder matrix. As compared to the glass composition of FS, the average concentration of Na and Si are higher in the binder gel formed and these elements are originating from the activating solution and those dissolved from the slag. Similar observation was reported in the binder gel of inorganic polymers made from plasma convertor slag (Kriskova et al., 2015). Additionally, the average concentration of Al and Ca are higher and Mg and Fe lower in the binder gel as compared to the glass composition.

In all the mortar samples, the average Na concentration are similar due to similar sodium oxide composition used for all the mixes. However, the average concentration of other elements such as Fe, Si, Ca, Al, and Mg are higher in AAFS2 and AAFS1 in comparison to the reference mortars. Fe is present in large quantities in the binder gel formed, indicating that Fe-containing phases do dissolve in alkaline environment and is consistent with the observation of other studies on alkali activation of Fe-rich slags and glasses (Machiels et al., 2014; Onisei et al., 2015; Peys et al., 2019a). The higher amount of Fe in the binder gel of AAFS2 could have influenced its properties and this is supported
by some studies where the positive role of Fe in alkali activation synthesis of FS, volcanic ash, Fe-rich glasses, and phyllite have been reported (Lemougna et al., 2013; Onisei et al., 2018; Simon et al., 2018; Adesanya et al., 2020).

**CONCLUSION**

In this study, an Fe-rich water-cooled granular FS from nickel processing was investigated as a sole solid precursor in alkali-activated mortars with and without particle size optimization. The mortar samples were evaluated in terms of workability, compressive strength, microstructural integrity, and the chemical composition of the binder. The properties of the alkali-activated mortars produced were mainly influenced by the aggregate type and particle packing. The use of FS as an aggregate and binder resulted in mortar samples with better mechanical performance compared to mortar samples with standard sand aggregates. The higher compressive strength achieved by FS aggregate mortars was due to the participation of FS aggregates in the hardening reaction, which contributed to the interfacial bonding and binder gel composition of the mortars. In contrast, the lower strength in standard sand aggregate-based mortars was due to the low chemical interaction between standard sand and the binder gel. The binder gel in all samples consisted of Na, Fe, and Si, with lower quantities of Ca, Mg, K, and Al.

Further optimization of the PSD of the FS aggregates significantly improved the workability, microstructural integrity, and fresh and hardened properties of the mortar. The compressive strength increased from 20 to 40 MPa, and the mortar had a denser structure depicted by the higher UPV value and bulk density once the PSD was optimized. In addition, mortar with an optimized PSD had better flowability.

The mechanical performance of FS aggregate-based mortars was 20–40 MPa, depending on the particle packing optimization, demonstrating their suitability as potential construction materials for building bricks, road pavement, and tiles. Overall, the successful utilization of FS as an aggregate and binder in alkali-activated mortar provides a new, sustainable, and cost-effective pathway for recycling industrial residue and can reduce the impact of solid waste on the environment.

**DATA AVAILABILITY STATEMENT**

The raw data supporting the conclusions of this article will be made available by the authors, without undue reservation.

**AUTHOR CONTRIBUTIONS**

AA: conceptualization, methodology, validation, formal analysis, investigation, visualization, writing (original draft), and writing
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SUPPLEMENTARY MATERIAL

The Supplementary Material for this article can be found online at: https://www.frontiersin.org/articles/10.3389/fbuil.2021.653466/full#supplementary-material
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Conflict of Interest: The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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