**Low-Carbon Composite Mortar with high Cement Replacement Ratios Using Volcanic Pozzolan and Silicon Manganese Slag**

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**Abstract.**

With the rising requirements of lowering global CO2 emissions, cement production, which accounts for nearly 8% of global CO2 emissions, is a key target for reduction. One of the most effective methods of reducing emissions is to reduce its use. This leads to the encouragement of finding alternative, less environmentally damaging cementitious materials to reduce reliance on emission-producing cement. The most effective strategy is to reduce cement consumption by incorporating alternative, low-carbon cementitious materials. While fly ash has been widely used as a supplementary cementitious material (SCM), declining availability necessitates the exploration of viable alternatives. This study investigates the performance of Icelandic Volcanic Pozzolan (VPI) as a partial replacement for Ordinary Portland Cement (OPC), both in binary and ternary blends. Silicon manganese slag (SiMn slag), a byproduct of silico-manganese alloy production, is used as a co-replacement material to further enhance sustainability and resource utilization. Six different mortar combinations were tested, incorporating 50% to 90% total OPC replacement levels. Compressive strength development was studied at 3, 7, 28, and 56 days. The scanning electron microscopy (SEM) and X-ray diffraction (XRD) analysis of the 28-day samples were conducted to study the composition of the microstructure. Isothermal calorimetry was used to compare the heat development changes due to the incorporation of SCMs. The results indicate that VPI and SiMn slag show promising potential as partial replacements for OPC, with 50% VPI replacement yielding a notable increase in compressive strength and favorable microstructural characteristics. The results imply that VPI and SiMn combination could serve as a viable and sustainable alternative to conventional cement in certain applications, potentially reducing clinker use and associated CO₂ emissions.

**Keywords:** low-carbon, Volcanic Pozzolan, SiMn slag

**1. Introduction**

Cement production is responsible for approximately 8% of global CO₂ emissions [1]. Considering the escalating impacts of climate change and Norway’s ambitious target to reduce carbon emissions by 90–95% [2], there is an urgent need for significant emission reductions across all sectors. Moreover, with a variety of contributing CO2 sources in the construction process, from transport, waste, assembly, and material production, both in collecting raw materials and their processing [3], especially with concrete. Portland cement production requires multiple resource-intensive processes [4], starting with the damaging quarrying of limestone, where it is ground and mixed with other materials to the desired composition. To address the environmental challenges associated with cement production, several strategies have been proposed. These include the incorporation of alternative supplementary cementitious materials (SCMs), optimization of mix design for reduced clinker content, implementation of carbon capture technologies, development of novel binders such as geopolymer and alkali-activated cements, application of life cycle assessment to inform sustainable decision-making, and enhancement of energy efficiency across the production and construction processes.

Industrial by-products such as ground granulated blast-furnace slag (GGBFS), silica fume, metakaolin, and fly ash are commonly used as partial replacements for cement, offering the potential to reduce greenhouse gas emissions significantly [5]. The use of SCMs to reduce cement consumption dates back as early as 1911, when approximately 45% of the cement was replaced with locally available silica-rich materials, primarily composed of granite, silt, and sandstone [6]. Fly ash has a long history of use as a supplementary cementitious material, first applied in 1942 during the repair of a spillway at the Hoover Dam in the United States. Its use was further expanded in large-scale infrastructure projects such as the Hungry Horse Dam, initiated in 1948, where fly ash replaced up to 32% of cement in selected structural elements [6]. Ground Granulated Blast Furnace Slag (GGBFS), a widely used supplementary cementitious material, has been incorporated into concrete at replacement levels as high as 95% in specific applications, though typical usage ranges between 30% and 80% [5]. However, the amount of slag availability is limited, with the current supply capable of meeting only 5–10% of global cement production [7]. Consequently, there is a pressing need to find alternative SMCs that offer higher replacement potential derived from more sustainable and abundant sources.

Volcanic pozzolan, a natural pozzolanic material, and SiMn slag, an industrial by-product of ferromanganese production, have emerged as promising SMCs. Volcanic pozzolans are abundant in several regions and possess high amorphous silica content, which contributes to pozzolanic activity. SiMn slag, although less explored in cementitious applications, has potential as a reactive or filler component due to its complex mineralogy and availability in large volumes as industrial waste [8]. This study focuses on the investigation of VPI as a SMC, which requires minimal preparation, reducing its environmental impact. Moreover, SiMn slag was used as a SMC to manufacture tertiary mortar by combining with VPI. During the study, compressive strength tests were conducted on seven experimental mixtures and one control mix at 3, 7, 28, and 56 days to evaluate strength development over time. To assess the viability and underlying mechanisms of each binder system, a comprehensive suite of supplementary tests was performed. Characterization of raw materials included Particle Size Distribution (PSD), Brunauer Emmett Teller (BET) surface area analysis, and sieve analysis. PSD and BET analyses were conducted on the binder materials to determine particle size and specific surface area, while sieve analysis was applied to aggregates to ensure consistency and repeatability across mixtures. Further analysis involved X-ray diffraction (XRD) to identify crystalline phases and microstructural development, and scanning electron microscopy (SEM) to visually examine the morphology of the hydration products. Isothermal calorimetry (IC) was employed to monitor the heat evolution of the binder systems, allowing comparison of reaction kinetics and setting characteristics across mixtures.

**2.0 Materials**

The following information pertains to the materials utilized in the experiment. In addition to the conventional ingredients of mortar, cement, sand, and water, SiMn slag and GGBFS were added. The particle size distribution of each material used was observed using the Mastersizer -v3.88 analyzer and wet analysis was performed on all solid materials other than sand.

Conventional ingredients, including cement, sand, and water were utilized and followed the NS-EN 196-1 standard for the preparation of mortar. Additionally, volcanic pozzolanic material from Iceland (VPI) and locally available silicon manganese (SiMn) slag were used as SCMs to replace cement at different percentages to produce binary and ternary mortar. Both INDUSTRISEMENT, CEM I 52.5 R cement conforming to NS-EN 197-1:2011 standard, which served as the primary cement, and excavated, ground VPI material from Iceland, sourced from Heidelberg Materials- Norway. Granulated air-cooled and ground SiMn slag, which is a by-product of ferromanganese steel production of ERAMET Norway, was used to produce ternary mortar. Locally available manufactured sand that conforms to the requirements of NS-EN 2620:2002+A1:2008 was purchased from Heidelberg Materials Aggregate Årdal, Stavanger, and sieved to adjust the particle size distribution within the range of 0-4 mm. The consistency of the mortar was maintained by adding an admixture, named Dynamon SX-N by MAPEI, based on modified acrylic polymers. Locally treated potable tap water was used in the mixing process.

Accordingly, among the binder materials, VPI showed the lowest average particle size, while SiMn slag has the largest particle size. Fig.01 explains the particle size distribution of these three raw materials, which might give a significant contribution to strength development when mixed together with a good packing density. Further, this observation can be verified by the BET fineness values, where the fineness is increasing from VPI, cement and SiMn slag, respectively. It can be clearly understood that the reactivity should be higher in VPI compared to SiMn slag due to its high specific area.

**Table 01**. Physical properties of raw materials.

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| Raw material | Cement | VPI | SiMn slag | Sand |
| Dv,90 (µm) | 38.70 | 20.20 | 51.70 | 1690 |
| Dv,50 (µm) | 15.20 | 7.25 | 17.70 | 658 |
| Dv,10 (µm) | 4.10 | 1.60 | 3.90 | 152 |
| BET, Specific surface area, (m2/g) | 1.76 | 2.84 | 0.96 | - |

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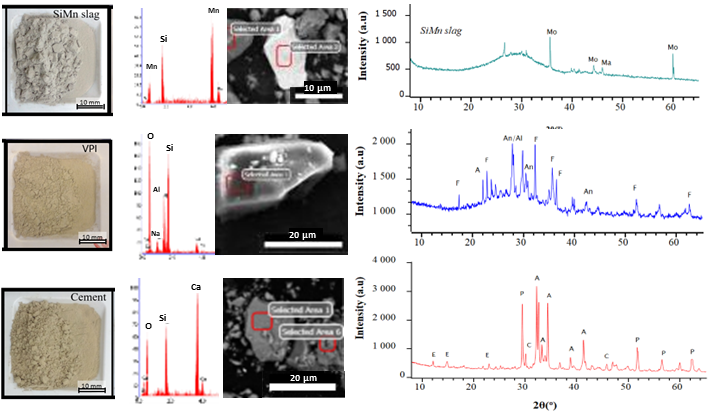
**Fig.1.**particle size distribution curves of materials.

**Table 2.** Chemical composition of raw materials (weight%) used in the experiment.

|  |  |  |  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| Material | SiO2 | Al2O3 | Fe2O3 | CaO | MgO | Na2O | K2O | MnO | TiO2 | SO3 | P2O5 | SrO2 |
| Cement | 23.59 | 5.05 | 3.84 | 59.14 | - | - | 2.01 | 0.10 | 0.25 | 5.51 | 0.35 | 0.09 |
| SiMn slag | 46.86 | 9.91 | 0.13 | 26.05 | 3.85 | - | 1.03 | 7.15 | 0.05 | 0.55 | - | 0.58 |
| VPI | 53.36 | 7.86 | 16.67 | 16.52 | 3.18 | - | 0.52 | 0.29 | 1.46 | - | 0.13 | - |
| Sand | 79.76 | 8.15 | 3.87 | 2.22 | - | - | 5.31 | 0.07 | 0.32 | - | - | 0.13 |

The results of X-ray Fluorescence (XRF) analysis conducted on these raw materials are presented in Table 2. Accordingly, the primary components of the two types of slags, similar like cement, are SiO2, CaO, and Al2O3, which together make up between 78-83% of the total composition. This constitution contributes to the cementitious properties of the raw materials. However, it's important to note that the CaO content is significantly lower in SiMn slag and VPI compared to cement, while the SiO2 content shows an opposite trend.

The Si/Al ratio is higher in VPI than in SiMn slag, with values of 6.8 and 4.7, respectively. Moreover, VPI contains a notable amount of Fe2O3, which could play a role in forming AFm phases during cement hydration. On the other hand, SiMn slag stands out for its high MnO content and rich with amorphous phases. Comparatively, VPI consists of both crystalline and amorphous phases, but dominantly the crystalline phases likely to that of cement as shown in Fig.2.



**c)**

**b)**

**a)**

**Fig. 2.** Physical shapes and XRD patterns of the raw materials. **(a)** SiMn slag, **(b)** VPI and (**c)** Cement. Where, E-ettringite, F-forsterite, An-anothite, Al-albite, A-alite, P- portlandite, C-calcite, Mo- moissanite, Ma- mavlyanovite.

**3 Experimental program**

**3.1 Mixture proportions and preparation of specimens**

A comprehensive experimental setup was designed to explore the feasibility of developing high-volume cement replacement composite mortar using VPI and SiMn slag as SCMs.

This program consists of a control series using only the cement and three binary mortar mixtures consisting of VPI with cement replaced by 50%, 70%, and 90% by weight. The aim was to assess the maximum achievable cement replacement with VPI without compromising strength properties. Similarly, three ternary mortar mixtures were also prepared with a combination of VPI and SiMn slag where it kept 20% of SiMn slag of the total binder weight fixed, while the percentages of VPI varied accordingly.

A fixed water-to-binder ratio of 0.35 was maintained throughout the study to target a high level of compressive strength after the 28 days. 1% by weight of superplasticizer was added to ensure consistency of workability. This dosage of the superplasticizer was determined based on a series of trials conducted on cementitious pastes prepared using similar mix proportions to those of mortars, aiming to maintain a minimum mini flow table diameter of 200 mm, according to Chinese standard GB/T17671 [9]. The specimens were labeled according to the type of mortar (control, binary, or ternary) and the percentage of replacement as shown in Table 3 and referred to in preceding sections.

**Table 3.** Quantities of material used in each type of mortar mixtures (for 1 kg of batch).

|  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| ID of Mortar type | Cement replaced % | VPI % | SiMn Slag % | Material requirement (g) | | | | | |
| Cement | VPI | SiMn slag | Sand | Water | Admixture |
| Control – (C) | - | - | - | 230.0 | - | - | 695.5 | 73.3 | 2.3 |
| Binary-2 (B2) | 50 | 50 | - | 114.7 | 114.7 | - |
| Binary-3 (B3) | 70 | 70 | - | 68.8 | 160.6 | - |
| Binary-4 (B4) | 90 | 90 | - | 22.9 | 206.4 | - |
| Ternary-2 (T2) | 50 | 30 | 20 | 114.7 | 68.8 | 45.9 |
| Ternary-3 (T3) | 70 | 50 | 20 | 68.8 | 114.7 | 45.9 |
| Ternary-4 (T4) | 90 | 70 | 20 | 22.9 | 160.6 | 45.9 |

**3.2 Casting, Curing and Testing of Specimens**

Casting of specimens, curing, and other relevant testing procedures were carefully carried out to ensure the accurate assessment of the specimens' properties, aiming to minimize the effects of the mortar's inhomogeneity, and are detailed below.

**Material mixing procedure**

All mixtures were stirred using a Hobart -A200 desktop mixture of 20-litre, Firstly, the dry materials in correct quantities as in Table.3 were added and stirred for 2 minutes at speed 2 (187 RPM) to obtain a homogenous dry mixture. Measured amounts of water and superplasticizer were then added and continue the mixing for another 4 minutes. Finally, the mixture was immediately transferred to relevant moulds. This procedure was adopted in order to obtain a homogenous mortar mixture fallowing certain guidelines in the ASTM C305-6 standard.

**Curing and testing of specimens**

*Compressive strength test*

Upon completion of mixing, binders were poured into oiled three-gang cube moulds, each measuring 50 x 50 x 50 mm, in two lifts, following the procedure described in ASTM C 109/C 109M – 08, section 10. The top surface of the mortar cubes was levelled with a spatula. The specimens were kept covered with polythene film to prevent moisture loss and left under ambient conditions until demoulding.

After 24 hours of initial curing at room temperature, the moulds were carefully removed. The specimens were then fully submerged in water to maintain 100% relative humidity, inside a curing chamber set at a temperature of 23±2°C until testing [10]. Before testing for compressive strength, the specimens were removed from the water and wiped with a dry paper, measured the weight and dimensions and subjected to compressive strength test with an automatic compressive strength machine (UCS test machine, Toni Technik, 300 kN) with a loading rate of 0.5 MPa/s at intervals of 3, 7, 28 and 56 days. The average compressive strength was calculated from the results of three samples each at least. The test standard followed herein is NS-EN 12390-3:2019.

*Isothermal Calorimeter*

Iso thermal calorimeter studies were conducted on each of the proposed binder proportions by preparing respective pastes without sand. The calorimeter was allowed at least 30 minutes to establish the initial baseline before introducing the sample. For each sample, a representative a batch of approximately 30 grams was prepared.

From this mixture, 6.75 grams was carefully extracted and introduced to the plastic ampule, ensuring minimal mass adhered to the side walls. The ampule was then lowered into the calorimetry machine as quickly as possible in order to minimize the disturbance and start the measurements quickly.

*Microstructural and Mineralogical Analyses*

Microstructural analysis was performed on representative samples collected from each hardened mortar specimen. A SEM (Zeiss Supra 35VP field emission gun SEM) coupled with an EDS detector (EDAX Octane Elite 25) at the University of Stavanger was utilized in this study. Small samples were taken from the inner core of the compressive strength test cubes after the 28-day compressive strength tests for SEM analysis. Additionally, separately cast representative paste specimens were also utilized in this test to accommodate any interference that can be caused from fine aggregates. To eliminate charging effects, the selected specimens were coated with silver using a sputtering process for a 10 or 50 nm thickness in all specimens. Subsequently, the remaining samples were crushed and ground into powder for XRD analysis using a Bruker-AXS diffractometer D8 ADVANCE equipped with a CuKα radiation source (wavelength of 1.5418 Å) and settings of 40.0 kV, 25.0 mA at the same time. The measurement range used was 2θ = 3 degrees to 70 degrees with a step size of 0.0132 [11].

**4 Results and Discussion**

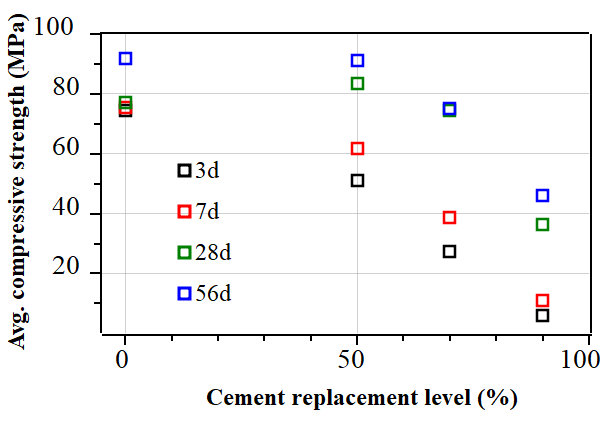
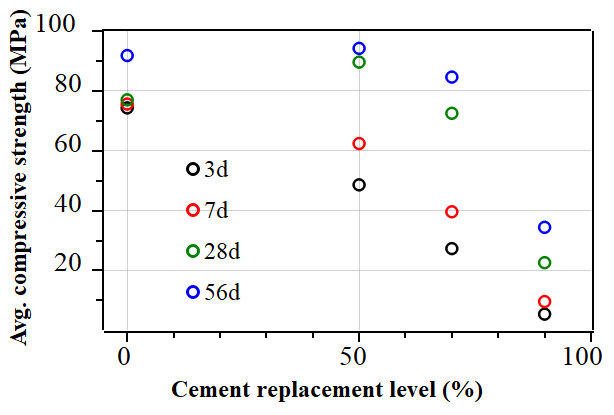
**4.1 Effect of SCMs on Compressive Strength**

Evolution of compressive strength over curing duration is recorded in Fig. 4 up to 56 days. The early strength at 3 days of control is significantly higher due to its clinker content. It is a known fact that this strength development rate becomes mild with time. In general, all the mortar types investigated showed delayed early strength gain and gradual development up to 56 days of the investigation period. The higher the replacement level of cement lowers the strength development is, as expected due to less contribution coming from the cement hydration process with the replacements.

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**Fig. 4.** Average compressive strength of binary and ternary binders including control.



**a)**

**b)**

**Fig. 5.** Effect of cement replacement to the compressive strength development at different ages of **(a)** binaries and **(b)** ternaries.

The inclusion of high levels of pozzolanic materials led to delayed strength gain, attributed to the slower reaction kinetics compared to the control. However, this has been outweighed by pozzolanic reaction when comparing the strength developments after 28 days, especially with 50% of cement replacement mortars B2 and T2, which overcome the strength of control. Those mortars have 65-70% strength development at 3 days compared with the control, reflecting the effect of reduced cement content and insignificant or minor contribution to early strength development comes from SCMs. The time involve effect on strength development due to cement replacement is shown from another perspective in Fig. 5.for better interpretation the results.

The combined effect of particle packing arrangement, including sand is also a factor to be considered. More interestingly, 70 % cement replaced T3 achieved 92% strength and B3 achieved 80% strength compared to the control after 56 days. Further, mortars where 90% cement replaced, especially T4 containing only 10% of cement gained a strength of 45 MPa at 56 days which is about 50% the strength of control. This is a considerable achievement and highlights the significant capabilities of VPI alone and combinations of VPI with SiMn slag to contribute to the progression of the strength development.

**4.2 Effect of SCMs on hydration kinetics**

Fig.6 and 7 demonstrate the hydration kinetics of proposed binary and ternary binders based on the results obtained from the IC test series conducted on respective paste systems. The initial dissolution phase was not considered, as the mixture was prepared outside of the IC instrument, and this research focused on the following hydration peaks, which govern the setting and onset of strength development.

The normalized heat flow with respect to binder weight and cumulative heat graphs for all binders are shown in Fig.6 and 7, respectively. The highest peak and the highest cumulative heat were scored by the control due to a higher contribution from clinker to the hydration kinetics. One significant peak was observed in all systems is due to the reactions of C3S phase primarily and followed by C2S to form C-S-H and CH [12].

Accordingly, with increased cement replacement reduces the first peak intensity proportionally and the cumulative heat generated. Nevertheless, of the type of SCM, the total cumulative heat follows the same trend for both binary and ternary binders. However, when considers Fig.6., the peak position also shifted towards the time axis with increased levels of cement replacement except for B4 and T4. This observation correlates with the lower early strength development trends observed in those systems. The deviation of 90% of cement replaced binders might be due to the early cessation of the majority of the hydration process due to a very small amount of cement, which facilitates early formation of nucleation sites as the clinker portion is well exposed among the less reactive SCM surrounded environment.

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**Fig. 6.** Normalized heat flow of different binders including control.

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**Fig. 7.** Normalized cumulative heat flow of binders investigated.

According to Fig.7., the majority of heat was generated in the binders with 50-70 % cement replacement during the first 24-30 hours. It is visible during that period that the heat released is reducing with increased cement replacement level. This heat of hydration significantly affects early thermal cracking in the binder matrix and is the dominant factor that needs to be considered for durability aspects. Especially in large volume concrete works, to reduce thermal cracking. Therefore, it is justifiable that the studied composite binders have the potential to reduce the early thermal cracking due to their lower cement content. But further investigations are required to find the optimal level of cement replacement.

**4.3 Effect of SCMs on microstructure by SEM -EDS analysis**

**XRD analysis**

Most peaks with high intensities in XRD diffractograms are assigned to quartz and other co-existing components like feldspar (albite, microcline, etc.) minerals, which are present in sand used as fine aggregate. Mostly, the hydration-associated peaks are masked due to the high abundance of those minerals within all the cementitious mixtures. However, most dominant peaks attributed to cement hydration products, such as ettringite at 2θ angle 9.1°, and portlandite at 2θ angle 18° and 34.1° could be identified and showed a reduction in their peak intensity with increased cement replacement levels as observed in Fig.8 which is a plot contains all the XRD diffractograms with same scales in both axis for comparison purpose.

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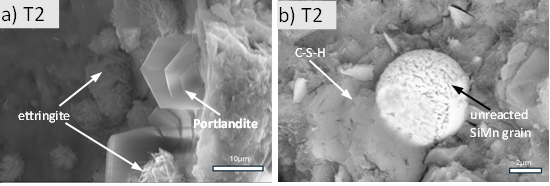
**Fig. 8**. XRD patterns of control, B2, T2, B4 and T4 binders at 28 days

However, in B4 and T4, XRD examination results showed negligible or no portlandite diffraction patterns and very weak intensities of ettringite, which can be attributed to the lowest amount of cement present and the lowest hydration process taking place. Although not qualitative, this observation aligns with the results from the IC tests and indirectly supports the observed trends in compressive strength development.

**SEM-EDS analysis**

Microstructural investigation was conducted after 28 days on both mortar and paste samples using SEM images in order to identify cement hydration products and study the bond formation when it replaces cement with VPI and SiMn slag. Portlandite and ettringite were observed in all binders of B2, T2, B3 and T3 likely to that in controls. Fig.9 (a) shows such observations made on T2 paste. However, in B4 and T4, the formation of such products were observed to be in lower intensities due to lack of cement in those systems. This is in agreement with the XRD peaks identified of such phases in the respective XRD diffractograms.

In addition, Fig.9 (b) explains well integration of unreacted SiMn grains with the C-S-H structure which explains an important factor, where the contribution comes from the microstructure development to the strength gain. Fig.9 provides evidence for the formation of hydration products in SCM incorporated cementitious systems likely to that in controls.



**Fig. 9**. **(a)** hydration products and (**b)** unreacted phases in the microstructure

Moreover, it was evident that higher cement replacement lowers the microstructural densification which leads to deterioration of the strength development characteristics. This also explains the lower strength gain with higher utilization of SCMs. As seen in Fig. 10., in the control, the microstructure becomes more dense, glassy texture due to the formation of C-S-H. This has been slightly outweighed in B2 with increased cement replacement but comparably densified. In contrast, the microstructure of B4 was less developed, and unreacted phases are present. This indicated that less hydration had taken place, and slowly continued the bond formation. However, these results unequivocally reveal the possibility of further growth of compressive strength over time because the continuation of pozzolanic reaction and availability of pore space, which can accommodate developing more C-S-H products unlikely to that in controls.

A close-up of a grey surface

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1a) C

1b) C

A close up of a greyscale image of a greyscale image of a greyscale image of a greyscale image of a greyscale image of a greyscale image

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2b) B2

2a) B2

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3b) B4

3a) B4

**Fig. 10**. Microstructure in **(a)** mortar samples and (**b)** corresponding paste sample at 28 days

**5. Conclusion**

This study evaluated the mechanical and microstructural properties of mortar incorporating variable proportions of VPI and SiMn slag as supplementary cementitious materials. A series of binary and ternary mortar samples were tested at replacement levels ranging from 50% to 90%, alongside a control mix containing 100% OPC. Compressive strength results indicated that replacement levels of 50% outperformed or matched the control mix after 28 days. At 56 days of curing, the 50% binary blend achieved a 102.5% and the ternary blend achieved 99.5% strength over that of the control. Mixtures with 70% replacement exhibited comparable strength of 92-82% to the control, while 90% replacement significantly reduced compressive strength by 50-63%, highlighting performance limitations at high substitution rates.

Isothermal calorimetry revealed delayed early hydration and a reduced secondary heat flow peak in high-replacement mixes, likely associated with lower C3S and slower aluminate reactions due to reduced cement availability and lower calcium hydroxide formation. XRD analysis showed reduced portlandite content in these mixes, indicating both less cement content and ongoing pozzolanic activity. SEM imaging further confirmed microstructural differences, with unreacted phases and less dense microstructures particularly evident at 90% replacement, supporting the observation of slower and less complete hydration.

The findings reveal the use of VPI and SiMn as effective SCMs at moderate replacement levels, supporting both mechanical performance and sustainability goals. While high replacement ratios compromise early mechanical strength, they may be suitable for mass or non-structural applications where long-term durability and sustainability are prioritized.

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