# THE EFFECT OF MINING WASTE ON THE DURABILITY INDICATORS OF CEMENT-BASED COMPOSITES

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**Abstract**

The need for infrastructure development is of major importance and the projected global infrastructure investment between 2013-2030 is estimated in the excess of £30 trillion to support the rapid growth of societies and economies worldwide (1). This trend puts civil infrastructure industry under immense stress to plan properly, construct fast and deliver resilient structures. Concrete is the dominant construction material and the key element in most infrastructure assets.

However, concrete's manufacture is extremely energy and resource intensive: >4 Billion tonnes of cement are produced annually, accounting to ~8% of global anthropogenic $CO_2$ and resulting to an annual production of ~2 tonnes of concrete for every person on the planet. The production of concrete is a process associated with very high energy consumption. In Europe, the construction sector alone is responsible for the 36% of $CO_2$ emissions and the 40% of all energy consumption.

The utilisation of mining waste in cement-based composites is an area of growing interest worldwide, with mining and excavation waste increasing considerably the last decade. Our work focuses on the replacement of cement with mineral wastes and the initial findings suggest that even at 20% replacement, the mechanical properties are marginally affected. This contribution will discuss some preliminary data on the effect of mining waste on the durability indicators of cementitious composites (oxygen permeability, capillary sorption and ion diffusion).

Keywords: Mining waste, Silicates, capillary water absorption,

## 1. INTRODUCTION

The fabrication of resilient and durable infrastructure is essential to support the rapid growth of societies and economies worldwide. Specifically, the projected global infrastructure investment is estimated to exceed £30 trillion pounds for the period 2013-2030 (1). This trend puts civil infrastructure industry under immense stress to plan properly, construct fast and deliver resilient structures.

Concrete is the key material for infrastructure construction as its properties satisfy the requirements for fast construction and delivery of resilient elements, while it is abundant and low-cost. The high demand of concrete makes it the second mostly used material in the world (2) but is also associated with high environmental impact. The manufacturing of cement, main component of concrete, is resulting to high energy consumption and natural resource depletion. Furthermore, the $CO_2$ produced due to fuel consumption for the calcination of limestone and decomposition of calcium carbonate reach up to ~8% of global anthropogenic $CO_2$, resulting to an annual production of ~2 tonnes of concrete for every person on the planet [2].

The need to address sustainability issues relating to infrastructure development and concrete production has been highly prioritised by the construction industry and the relevant research community over the past decades. Nonetheless, the demand for high performing materials is more pressing to satisfy the growing demands of the society. Replacing part of cement with pozzolanic or mineral materials, has been proven a viable solution to decrease the environmental impact of concrete, while enhancing some of its properties. For example, the addition of silica fume, a by-product of the silicon metal or ferrosilicon alloys production, leads to higher strength and increases resistance to chloride penetration.

The utilisation of mining waste in cement-based composites is an area of growing interest worldwide, with mining and excavation waste increasing considerably the last decade. Mine tailings are a residual product after the separation process of the valuable fraction from the ore (3). Their particle sizes are very small due to the comminution process and they can be used for cement replacement, either directly or with little processing. Therefore, use of such mining wastes does not only add value to a waste product but could also lead to reduction of energy requirement for concrete production.

At the present study, the use of a siliceous by-product from the graphite mining industry in cementitious composites is investigated. The silicate is added in the mortars to replace cement at percentages up to 20% and its effect on the strength the durability indicators of cementitious composites is evaluated.

## 2. MATERIALS AND METHODS

### Materials and mix design

The replacement percentages of cement by silicate mining waste were 0%, 10% and 20% for corresponding mortar mixes REF, Sil10 and Sil20. The cement used was CEM II 32,5R and the mortars were fabricated with binder to sand ratio 1:1.5 and water to cement ratio 0.5.

**Table 1: Materials for mortar mix design**

| Mix label | CEMI (g) | Silicate (g) | Sand (g) | Water (g) |
|---|---|---|---|---|
| REF | 3.00 | 0 | 4.5 | 1.5 |
| Sil10 | 2.7 | 0.3 | 4.5 | 1.5 |
| Sil20 | 2.4 | 0.6 | 4.5 | 1.5 |

**Testing methods**

The siliceous by-product was characterised by X-ray diffraction (XRD) to obtain chemical composition and estimate the percentage of amorphous material present. The characterisation was performed with a Bruker diffractometer and the parameters used were 2-theta range of 5-80 with step size of 0.02.

The workability of fresh mortars was evaluated immediately after casting, using the flow table method according to BS EN 1015-3 (4). The effect of the silicates on the porosity of the mortars was evaluated indirectly through water absorption by capillary action. The test was performed on half prism specimens (prisms dimension 40x40x160mm) after 28 days of water curing. The specimens were dried to constant mass, then all other sides were sealed with aluminium tape and they were immersed in 3mm water with the 40x40 mm cast side being in contact with water. The mass changes were recorded for a total of 256 minutes (4:16 hours). The compressive and flexural strength of the mortars were tested at 28 days to evaluate the effect of cement replacement by silicate waste, BS EN 1015-11 (5). The compressive strength results are also discussed against those of equivalent mortars with same replacement percentages of silica fume (SF). This is done to highlight the different effect of an amorphous and crystalline fine filler. The nomenclature of the comparative mixes is SF10 and SF20.

## 3. RESULTS AND DISCUSSION

### 3.1 Material characterisation

The silicate waste was characterised by XRD and the identified pattern is presented in Figure 1. It appears the waste is mainly composed of crystalline silicon oxide (S) as indicated by the sharpness of the peaks. The hump observed at low 2-theta angles could correspond to a minimal percentage of amorphous material. Also, the peak corresponding to graphite (C) at 26$^o$ 2-theta angle was identified. This is expected as the silicate used in the present study is a waste of graphite mining. Even though a separation process is used for the two materials, it is assumed that small amounts of graphite could have remained unseparated. Lastly, the peaks at low angles were attributed to a complex, carbon-containing phase (C’). The two most probable phases are $C_{48}$ $H_{62}$ $Cl_0$ Mn $N_{60}$ Na $O_{79}$ and $C_{10.5}$ $H_{7.5}$ $Cl_{0.75}$ $N_{0.75}$ $O_{17.88}$ $V_3$. The particle size distribution of the silicate waste as provided by the supplier is presented in Table 2

**Table 2: Physical properties of silicate waste**

| Properties | Silicate waste |
|---|---|
| Mean (μm) | 8.016 |
| $D_{10}$ (μm) | 0.802 |
| $D_{50}$ (μm) | 3.611 |
| $D_{90}$ (μm) | 18.86 |

It is expected that the silicate waste could contribute to the cement hydration mainly physically. As seen by the X-ray diffraction analysis in Figure 1, the silicon oxide present in the waste is mainly crystalline and the average particle size of the silicate is around 8 μm according to **Table 2**. Even though it is much smaller compared to that of cement, given the level of $SiO_2$ crystallinity the specific surface area might be not sufficient to promote pozzolanic reaction of the material. Therefore, the silicate is considered unlikely to react and contribut chemically in the hydration reaction. Fine materials added in cement, even if they are inert chemically, could contribute to strength development due to the filler effect. This means increase of potential nucleation sites and also more refined porosity (6). Similar inert additions, such as silica powder (3) have been found to favour the strength development of the cementitious matrix in a similar manner; by improving packing, nucleation and refining porosity.

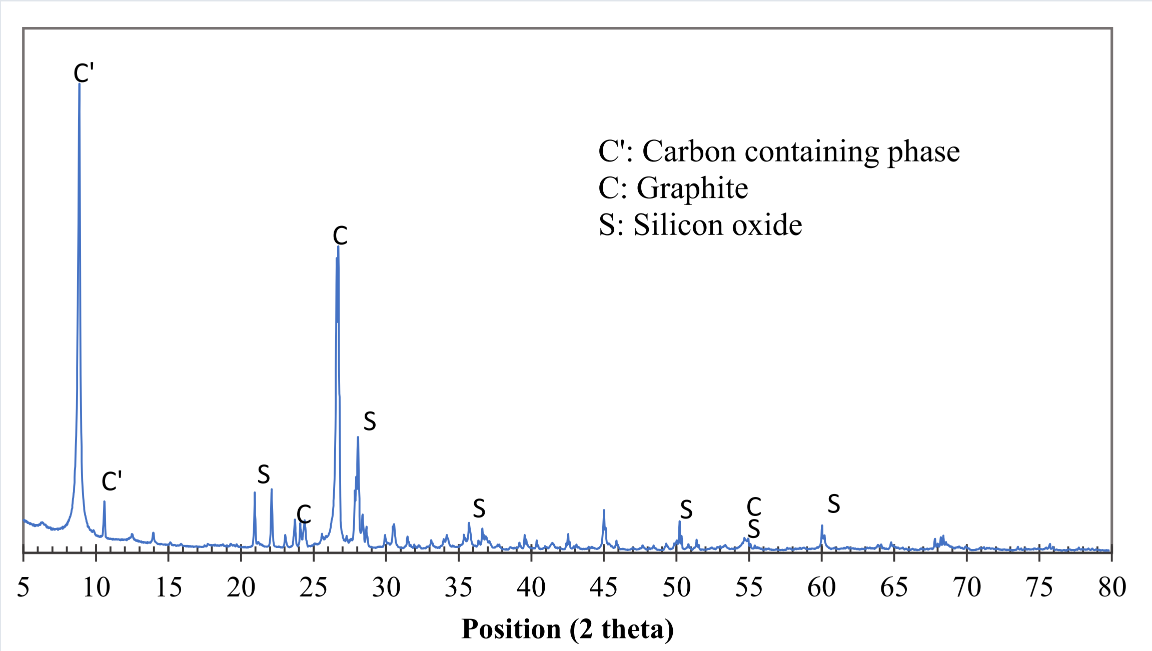


**Figure 1 X-ray diffraction pattern of silicate waste**

## 3.2 Workability

The addition of increasing silicate percentages in the mortars resulted to stiffer mixes as seen in **Table 3**. This was expected due to the particle size of the silicate waste which is significantly smaller than that of cement. Also, the shape of the silicate could have contributed to the flow reduction as plate-like or sharply-shaped powders hinder flowability of paste (7). The reduction in flow is not proportional to the replacement percentage, as for 10% silicate waste the reduction is 11% while for double replacement the percentage is 15%.

**Table 3: Flow of mortars with silicate replacement**

| Mix label | Flow (mm) | Reduction percentage |
|---|---|---|
| REF | 244.5 | 0% |
| Sil10 | 216.5 | 11% |
| Sil20 | 207.0 | 15% |

This reduction would be expected due to the particle size of the silicate waste, as discussed before. Specifically, the addition of a finer particle in the mix is expected to increase water demand due to the increased specific surface area (8). Nonetheless, the reduction exhibited by the mixes did not hinder proper compaction and it is considered that in case of specific requirement for workability level, flow can be maintained by the use of a plasticizer and superplasticizer admixture.

### 3.3 Capillary water absorption

Water absorption through capillary action was used as an indicator to evaluate effect of silicate waste on mortar durability. The test was performed on mortars cured for 28 days and the results are presented in **Table 4**. It is observed that increasing silicate percentage resulted to reduced water absorption. This could be expected as the increased percentage of finer material leads to stiffer mixes and less porous matrices, due to the filler effect (6). Specifically, addition of fine inert material such as silica powder plays a significant role in refining cementitious matrix porosity and can be used for the production of high-performance mixes (9). The capillary water absorption results can be associated with the observed reduction of flow and attributed to the particle size of the mineral addition. Specifically, increasing silicate percentages lead to lower flow values. Nonetheless, given that the mix design was performed to ensure a fairly workable mortar, this slight reduction in workability could have led to better compaction and reduced amount of entrapped air in the mix. This alongside with the filler action of the silicate waste are the main reasons the mixes are exhibiting reduction of water absorption by capillary action.

**Table 4: Sorptivity coefficient of mortars with silicate replacement**

| Mix label | Sorptivity Coefficient ($mm/min^{1/2}$) | Reduction percentage |
|---|---|---|
| REF | 0.8754 | 0% |
| Sil10 | 0.8700 | 1% |
| Sil20 | 0.8506 | 3% |

### 3.3 Strength of mortars

In order to ensure resilient infrastructure, both durability and strength requirements need to be satisfied. To perform a full preliminary evaluation of silicate mining waste as a it is essential to examine the effect on strength development. The results of 28-day compressive and flexural strength tests are presented in Figure 2 and Figure 3 respectively. In Figure 2 the strength of equivalent mortars with silica fume is plotted to perform the comparison between the different effects of the two fillers.

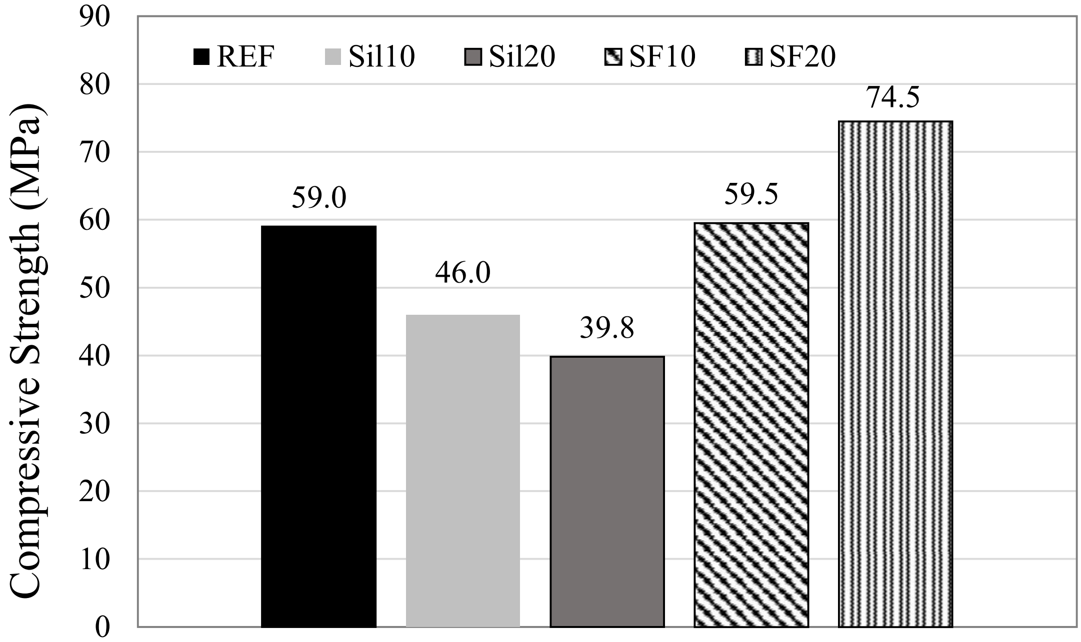


**Figure 2: Compressive strength of mortar with silicate waste at 28 days compared to mortars with silica fume**

Increasing percentage of cement replacement by silicate is leading to a decreasing trend of compressive strength. Specifically, the Sil10 mortar with 10% silicate waste presents a reduction of about 22% on 28-day strength. The corresponding strength reduction percentage for 20% silicate in the mortar is 32%. In the study of Pyo *et al*. (3), silica powder is replaced by siliceous crystalline mining waste at 50% and 100%, and it was similarly found to reduce workability and strength while no contribution in the hydration products was identified. The reduction of workability was attributed to the particles shape, which was plate-like and the reduction in strength was associated with inert nature of the mining waste. It appears that the silicate waste used here has a similar action, as no contribution on strength development is identified.

**Table 5: Strength Activity Index (SAI) for Silicate waste and Silica fume mortars at 28 days**

| Mix Label | Sil20 | SF20 |
|---|---|---|
| SAI | 0.67 | 1.26 |

This is further corroborated by examining the quite distinct effect of silica fume addition in the mortars, as increasing percentages lead to increasing strength values. Specifically, the SF10 with 10% silica fume demonstrates a slightly higher value than the REF. The mortar with 20% replacement presents 26% percent increase. Apart from filler effect, silica fume contributes chemical to the mix resulting to a significant increase of strength due to its strong pozzolanic action. To access pozzolanic reactivity of the silicate waste, the Strength Activity Index of the mortars was calculated and compared to the values for silica fume replacement, as shown in ***Table 5***. The SAI is calculated by diving the unconstrained compressive strength of a pozzolan-containing mortar by that of the control. According to ASTM C618, a SAI greater than 0.75 after 7 and 28 days for FA and natural pozzolans at a cement replacement of 20% is required to indicate positive pozzolanic activity (10). The values for silica fume demonstrate a clear addition to the strength of the mortars. On the contrary the value for 20% replacement of silicate waste is below the threshold. The SAI indicator further supports the hypothesis of the chemical contribution from silicate waste in the mix is not sufficient to enhance strength. Similar observations were made by Donatello *et al*. when the SAI was calculated for mortars with 20% silica sand replacement (10). Nonetheless, the SAI of the silicate waste mortars is not dramatically lower than the threshold values. This could indicate that pozzolanic reactivity of the material could be enhanced by processing such as ball milling for reducing the particle size.

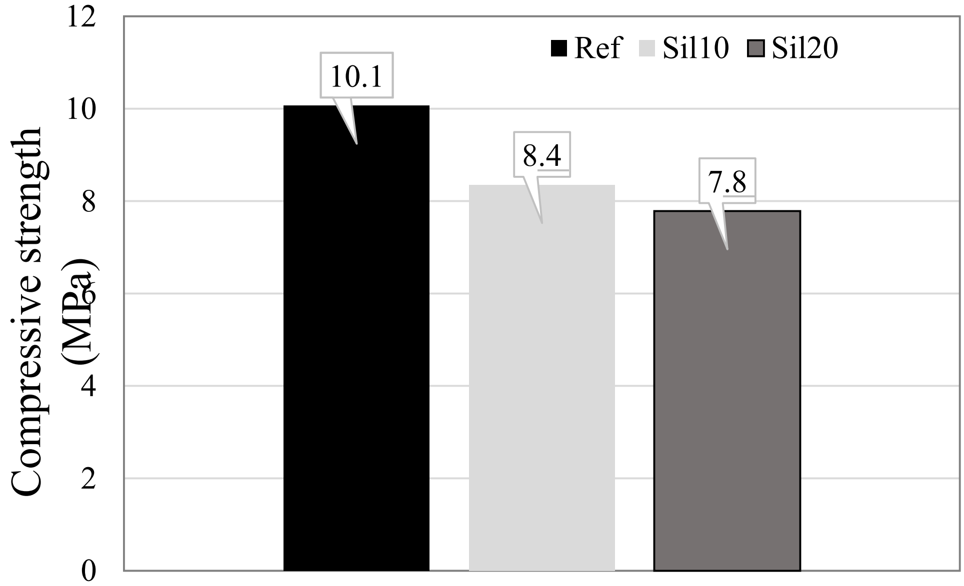


**Figure 3: Flexural strength of mortar with silicate waste at 28 days**

Regarding the flexural strength, a similar but milder reduction trend is observed. The addition of 10% and 20 % of silicate waste lead to 17% and 23% lower strength values for the 28-day mortars. The reduction in flexural strength could be associated with the increase in the mortars' stiffness and also the overall weaker matrix due to low reactivity of the silicate.

## 4. CONCLUSIONS

Examination of the silicate mining waste showed that it consists of mainly crystalline silica, with graphite residue and a carbon containing crystalline phase. The high crystallinity of the silicate waste and particle size were associated with low pozzolanic reactivity when added as a cement replacement in mortars. This was confirmed by reduction in compressive and flexural 28-day strength with increasing silicate waste percentages in the mortars. This was further highlighted by comparing the silicate waste effect to that of silica fume, a fine siliceous but mainly amorphous mineral addition.

As expected, the addition of a finer powder reduced the workability of the mortars.

Nonetheless, addition of silicate waste resulted to 1% and 3% reduction of the mortars capillary water absorption. The reduction as associated with the smaller particle size of the silicate waste which resulted to finer pores of the matrix potentially being filled up and the overall filler effect observed.

The results overall demonstrate that addition of the silicate mining waste presents a potential to improve durability of cement mortars through porosity refinement. The impact of the replacement on strength can be addressed by mild processing such as milling to a greater fineness. This would not only enhance the pozzolanic activity of the waste but could lead to further porosity refinement. Further work will be performed on low energy processing of the silicate waste to improve reactivity and optimising the mix design for achieving sufficient strength values while maintaining adequate workability.

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