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# Setting and curing of mortars obtained by alkali activation and inorganic polymerization from sodium silicate and silica aggregate

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## HIGHLIGHTS

- Water insoluble silicate mortars were produced.
- Optimum properties were obtained by controlling processing temperature and composition.
- Higher level of mechanical properties were achieved in silicates aggregates.
- Compared to cement, lighter and better sustainable silicate mortars were produced.
- Thermal conductivity of silicate mortars was less compared to Portland cement mortar.

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## ABSTRACT

This research is focused on sodium silicate bonded silica aggregates for making sustainable construction materials such as bricks and precast products. Different compositions are investigated to produce castable mortars. The mortars are cured at temperatures ranging from 150 to 300 °C and characterized, in particular microstructural and mechanical properties are investigated. Very high compressive strength of 100 MPa and elastic modulus of 5 GPa are obtained for samples with optimized compositions and heat treatments. Solubility and degradation study of the samples in water demonstrate that alkali silicates are prone to be soluble if not treated at 200 °C or above. Transformation of Si–OH to Si–O–Si not only increases the strength but also makes it insoluble in water. It is concluded that sodium silicate bonded bricks and blocks are very promising and affordable materials for construction. They represent an alternative to Portland cement concrete bricks and to sintered clay bricks, providing higher strength and representing an eco-friendly material.



## 1. Introduction

Currently infrastructure and housing sector make use of huge amounts of cement concrete and clay bricks. Both the production of cement and ceramic construction materials require extensive energy and result in $CO_2$ emissions. Therefore environmental activists and government regulations are continuously forcing scientists and engineers in development of new alternatives to cement and sintered ceramics which could be able to fulfil the desired requirements [1,2]. The earth crust is mostly composed of aluminosilicates, and most of the researches are trying to mimic the natural stone formation processes in the labs which can be viable for commercial production. Among the various processes available, the ones using alkali silicate binders are particularly interesting. When an aluminosilicate material and an activating agent like sodium hydroxide are combined, a partially crystalline solid is obtained. The class of these materials is designated as geopolymers. The aluminosilicate sources can be naturally occurring or industrial wastes like fly ash, clays tailing, kaolin and pozzolans [3,4]. Geopolymerization reaction also occurs without aluminum, forming Si–O–Si type network. This Si–O–Si bonding gives much higher strength but the drawback of these kinds of materials is that they are water soluble [1]. These types of bonds (Si–O–Si) can be stabilized by carbonation from atmospheric $CO_2$ which is a well-known process to harden the inorganic silicate paints [5]. If a similar process is achieved in-situ by the use of sodium carbonates and mild thermal treatments (above 200 °C), the mortar may be

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hardened and stabilized to water. This in-situ carbonation has already been successively used for the hardening process of geopolymers [6]. It is also known that the higher the amorphous silica content, the higher is the geopolymerization reaction due to higher dissolution of the silica and other contents [7,8]. To initiate the chemical reaction between silica aggregates and alkali, three components are necessary namingly, a critical amount of reactive mineral phase in the aggregates, pH of the alkali solution and moisture [9]. For the chemical reaction to take place, the alkali concentration should be high; pH should be higher than 11. For that purpose usually a 14 M solution of sodium silicate is used for the dissolution of the aggregates [10,11]. Higher pH of activating solution results in higher geopolymerization and strength [7].

In this research, we have studied the binding effect of sodium silicate on naturally occurring silica aggregates rather than aluminosilicate minerals as it is done for geopolymers. The investigated binding process is similar to geopolymerization but with lower amount of aluminum hence without true geopolymerization reaction. We have used compositions as reported by Temuujin et al. and Sarkar for the fly ash geopolymers to make the silica inorganic geopolymer bricks by using silica sand aggregates instead of fly ash or any other aluminosilicate [10]. With respect to geopolymerization reaction more emphasis is given to Si–O–Si polymerization rather than Si–O–Al (geopolymerization) hence named differently as an inorganic polymerization.

We also study the effects on the silica inorganic polymerization by increasing amounts of sand aggregates and in-situ carbonation using sodium bicarbonate. The physical and mechanical properties of as prepared silica inorganic polymer samples are carefully reported.

## 2. Experimental

### 2.1. Materials and preparation

Silica sand aggregates by Norman sand (Beckum/Germany CEN Standard Sand EN 196-1, ISO-679) were used as silica source for which the chemical composition (measured by X-ray fluorescence spectrometer (Bruker M4 Tornado)), is given in the Table 1. Sodium bicarbonate (CRUCIANI DAB E500/Italy) was used as in-situ $CO_2$ source. Water glass with 9% $Na_2O$, 30% $SiO_2$ and 61% $H_2O$ (Prochin, Italy) and specific gravity of 1.35 g/cm$^3$, was used in the formulation. 14 M sodium hydroxide solution was used as an activator solution.

A dense homogenized slurry was prepared by milling the sand aggregates together with sodium hydroxide solution, water glass and sodium bicarbonate (for in-situ carbonation formulation), for 10 min using a planetary ball mill and alumina balls as grinding media. Table 2 indicates sample codes and compositions utilized for sampling and testing. Water may also be added later to enhance plasticity of the mortar but with much care as it may be detrimental to uniform properties. After mixing, grinding and adjusting water contents, the mortars were cast in to polyethylene plastic molds with a height to diameter ratio more than 2 (0.9 cm diameter and 2.5 cm height). After forming, the samples were demoulded from the cylindrical vessels and cured at different temperatures (150, 200 and 300 °C) for 2 h in muffle furnace and characterized further.

### 2.2. Analysis of samples

Calorimetric properties (Differential thermal analysis) of the prepared mortar samples were measured from 25 to 700 °C by Mettler Toledo instrument. X-ray diffraction patterns were obtained by Rigaku diffractometer with Cu Kα radiation generated at 40 kV and 20 mA. FTIR analysis was performed on ATR (Attenuated Total Reflectance; Perkin Elmer) with diamond crystal as a probe. Thermal conductivities were measured by C-Therm thermal conductivity analyser.

Micrographic analysis was performed using electron microscope (Model: Zeiss, Jena, Germany). Bulk density was calculated by measuring the dimensions and weight of each samples. Mechanical properties of the samples after curing were measured using Lloyd LR5 K instrument in compression mode by applying ASTM C109 standard protocol.

**Table 1**
Composition of the inert sand aggregates by XRF analysis (mass (%)).

| Composition (%) | Inert sand |
|---|---|
| $Al_2O_3$ | 3.76 |
| $SiO_2$ | 94.02 |
| $K_2O$ | 1.07 |
| CaO | 0.16 |
| $TiO_2$ | 0.13 |
| $Fe_2O_3$ | 0.68 |
| MnO | 0.04 |
| $ZrO_2$ | 0.04 |

**Table 2**
Compositions of silicates used.

| Sample ID | Sand (%) | Sodium silicate (%) | Sodium hydroxide 14 M | Sodium bicarbonate (%) | Water |
|---|---|---|---|---|---|
| IG | 85 | 10 | 4 | – | 1 |
| IGBC | 73 | 17 | 7 | 2 | 1 |
| IGC | 73 | 18 | 7 | – | 2 |

## 3. Results and discussion

### 3.1. XRD analysis

The XRD spectra of sand aggregates and prepared samples after curing is shown in Fig. 1. In general, all the spectra indicate the absence of amorphous phases, as there is no hump or broaden peaks visible and the most abundant phase is quartz in aggregates as well as in hardened samples. In pure sand aggregates, the peaks are more intense compared to their silicate mortars, because the percentage of silica was reduced by the addition of sodium hydroxide from alkali activator and water glass. All the compositions made for this work show similar trends. There are no peaks appearing after polymerization as the samples and aggregates were mostly quartz silica. But a very small peak present in aggregates at 27.94 (Anorthite; $CaAl_2Si_2O_8$, JCPDS 89-1462) is diminished after hardening where as another similar small peak reappeared in mortars without $NaHCO_3$ treated, at 27.50 corresponding to CAH10 ($CaO{\cdot}Al_2O_4{\cdot}10H_2O$). Anorthite is a zeolitic compound where Al+ is present in the four fold symmetry and its presence gives the insight of the aggregates where this was formed due to weathering and erosion and remained in contact with water for longer times. [12]. Due to dissolution of anorthite, calcium and aluminum react to form CAH10 which is cementing compound formed by calcium. In IGBC, calcium and aluminum didn't form CAH10 due to higher affinity of calcium oxide toward $CO_2$ to form $CaCO_3$, hence CAH10 peak is absent in the samples treated with sodium bicarbonate. A very small peak for $CaCO_3$ was appeared

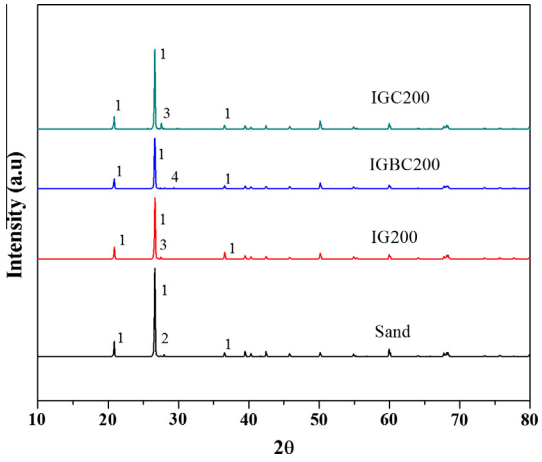


**Fig. 1.** XRD pattern of sand aggregates and different silicate mortar compositions cured at 200 °C. (1 = Quartz, 2 = Anorthite $CaAl_2Si_2O_8$, 3 = $CaO{\cdot}Al_2O_4{\cdot}10H_2O$, 4 = $CaCO_3$).