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# Alkali activated composite binders of waste silica soda lime glass and blast furnace slag: Strength as a function of the composition

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## HIGHLIGHTS

- Composite binders of urban waste glass and blast furnace slag were studied.
- The Taguchi method was useful to optimize the formulation of the composites.
- The addition of $Na_2CO_3$ promoted high dissolution of glass particles.
- The reaction products were C–S–H and silica gel.
- Glass is a promising addition to alkaline slag cements.

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## ABSTRACT

Composite binders of urban waste glass/blastfurnace slag activated by NaOH and NaOH/$Na_2CO_3$ were studied using statistical methods (Taguchi); the factors were: %glass, curing temperature, %$Na_2O$ and ratio of alkalis; the compressive strength was the response variable. The optimal levels at 28 days were %0glass-60 °C-6%$Na_2O$ using 2$Na_2CO_3$/NaOH; after 90 days these changed to 100%glass-60 °C-10%$Na_2O$ using $Na_2CO_3$. The microstructure and composition of the hydration products varied depending on the glass content. The reaction mechanism was mainly of dissolution-precipitation of reaction products, in some cases rims of reaction products formed around the slag particles. The confirmation experiments matched the statistical predictions.



## 1. Introduction

The sustainability in construction materials requires the research and development of alternative binders; one way to achieve this, is using postconsumer wastes, as well as industrial and urban wastes. Some of such raw materials are promising in terms of allowing the production of green materials with good mechanical performance. The incorporation of byproducts such as ashes (rice-husk, wood, sugar cane, coal, etc.), various slags, silica fume, and other pozzolanic materials as a partial or total replacement of Portland cement, can help to produce alternative cementitious matrices towards the reduction of the need for Portland cement, in addition to creating more durable concretes and reducing greenhouse gas emissions [1]. Alkali-activated cements represent a sustainable alternative to exploit some wastes and byproducts, which would otherwise become environmental passives. Several studies [2–8] report on the use of different materials (naturals or byproducts), to replace PC partial or totally. Blast furnace slag (BFS) and fly ash (FA) are the most studied materials, due to their abundance [9,10], although their availability is in many areas regionally constrained. The use of alkali activated BFS and FA has been widely reported [11–14]. Leong et. al [13]. reported the effect of different $Na_2O$ and $K_2O$ ratios of alkali solutions on FA based-geopolymers; the compressive strengths achieved were up to 35 MPa at 7 days. Nazari and Sanjayan [12]; they studied the possibility of synthesis of geopolymeric pastes through the alkaline activation of aluminum and grey cast iron slags, four different ratios of grey cast iron slag to aluminum slag were considered including 80/20, 70/30, 60/40 and 50/50; the best 28 day strength of 45 MPa was for pastes with a $SiO_2/Al_2O_3$ ratio of 3.0 activated with NaOH = 16 M. These last reports describe some examples on the use of different slags and FA as a cementitious materials to completely replace Portland cement. Nonetheless, other wastes are also attractive especially if their landfilling involves environmental issues; such is the case of urban waste

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glass (UWG). The glass wastes from human activity, for which more than 90% is silica soda lime glass, represent an environmental problem in urban areas. The estimations of glass currently landfilled are of more than 46 million tons yearly, and could reach 77 million tons by the year 2025 [15]. On the other hand, the recycling of UWG in the glass industry is limited due to difficulties related to the classifying of type and color of the glass.

Glass has been used as aggregate and as a partial substitute on PC in mortars and concretes [16–19]; however, in some cases, the results were not favorable due to the deleterious reactions between the binder and the aggregate, i.e. the alkali silica reaction (ASR) [16–18]. Other studies reported the use of waste glass as a source to obtain sodium silicate (waterglass) on binders made of BFS and binders of fly ash; the dissolution of the glass and its subsequent conversion to waterglass by the alkaline attack using sodium hydroxide (NaOH) resulted in a better activation of the BFS and the FA [20,21]. Avila-López et al. [2]. worked on alkali activated binders systems made of limestone and waste glass, where limestone were used to compensate the deficiency of CaO in the glass; they reported that the best mechanical strength at 28 days (38.8 MPa) were obtained employing a $NaOH/Na_2CO_3$ mixture solution with$\%Na_2O = 9$, the found phases were C-S-H and silica gel with crystalline phases such as pirssonite and gaylussite. On the other hand Torres et.al [22]. reported the use of glass wastes and BFS as cementitious raw materials activated with $5\%Na_2O$ (relative to the slag content) using $NaOH/Na_2CO_3$ mixtures, they found that the highest 28 days mechanical strength (27.7 MPa) was obtained by the composite 70/30 (BFS/waste glass respectively); they concluded that the low compressive strength developed on systems with high glass content were due to the low activation of the glass by the alkaline solution used, compared to a control mix of 100%BFS.

The number of reports related to the use of glass as a binder in alkaline cements is reduced, and considering the opportunity that represent the amount of glass landfilled, this research brings new data about the feasibility of incorporating urban waste glass as a potential cementitious material in combination with BFS to develop Portland cement free composites binders. A systematic statistical study was carried out to identify the effect of the % UWG, activator type, $\%Na_2O$ and curing temperature in the strength and microstructures of pastes of alkaline cements.

## 2. Experimental procedure

### 2.1. Raw materials and characterization

The materials used were granulated blast furnace slag (BFS) and urban waste glass (UWG); these were ball milled to a specific surface area of 4000 $cm^2/g$. Mixes of sodium hydroxide flakes and sodium carbonate powder where used to prepare the alkaline activators. The UWG was compounded by a mixture of green, blue, amber and clear glasses. The chemical composition of the BFS and UWG are shown in Table 1 and the X-ray diffraction patterns presented in the section of results (Fig. 4) indicated that both are predominantly amorphous; the UWG displayed an amorphous halo at 15–40°θ without any crystalline phases, while the BFS showed a broad hump at approximately 20–35 °2θ with weak reflections corresponding to calcite and akermanite.

### 2.2. Sample preparation

Composites of BFS/UWG were prepared with various UWG contents as described below. The dry powders were mixed thoroughly in blender with a planetary movement. The pastes were prepared using a water/solids (w/s) of 0.3; the solution of activator was added into the bowl and the pastes were mixed for 3 min. The pastes were poured into a set of cubic molds of 25 mm per side, the set was afterwards covered with a plastic film to avoid the water evaporation and then allowed to cure in isothermal chambers at the temperatures described below. After 24 h the samples were extracted from the molds and tested to compressive strength at 1, 3, 7, 14 and 28 days.

**Table 2**
Factors and levels for the experimental design.

| Factor | Level 1 | Level 2 | Level 3 | Level 4 | Level 5 |
|---|---|---|---|---|---|
| %UWG | 0 | 25 | 50 | 75 | 100 |
| Alkaline compound ratio[a] | 1:0 | 0:1 | 1:1 | 2:1 | 1:2 |
| % $Na_2O$ | 4 | 6 | 8 | 10 | 12 |
| Curing temperature | 20 | 40 | 60 | 70(12)-20[b] | 70(24)-20[c] |

[a] Weight ratio of $NaOH:Na_2CO_3$.
[b] The initial 12 h of curing at 70 °C then at 20 °C.
[c] The initial 24 h of curing at 70 °C then at 20 °C.

### 2.3. Design of experiments by Taguchi method

The Taguchi method is a technique used to find the optimal parameters among a set of conditions in an experiment using orthogonal arrays (OA) which randomly combine the factors and its levels. This method proposes an analysis of variance in order to identify the optimal combination of levels for the parameters considered in the experimental array [23–25]. The Taguchi method was implemented using the factors and levels indicated in Table 2. The set of experiments, was an $L_{25}$ ($5^4$) array, so a total of 25 trials were prepared; the configuration of the OA is shown in Table 3. The %UWG represents the weight percentage of glass in the binders. The second factor is the ratio of alkaline compounds which includes the NaOH (NH) and $Na_2CO_3$ (NC) in various ratios. The $\%Na_2O$ levels were 4, 6, 8, 10 and 12% relative to the weight of UWG + BFS. The curing temperatures included isothermal curing at 20, 40 and 60 °C and two temperature treatments, namely 12 h at 70 °C and subsequently 20 °C and the other for 24 h at 70 °C and subsequently 20 °C.

The Taguchi method randomly combine the factors and its levels in order to reduce the number of experiments and to minimize the influence of external factors that cannot be controlled; the experimental results are interpreted by calculating a factor called signal-to-noise ratio (S/N), where the signal corresponds to the compressive strength, and the noise that correspond to the set of parameters for a given experiment. The S/N ratio definitions depend on the optimization target with three possible criteria: larger is better, smaller is better or nominal is better; the criterion selected in this case was *larger is better,* which is defined by Eq. (1).

$$\frac{S}{N} = -10\log_{10}\left[\frac{1}{n}\sum_{j=1}^{n}\frac{1}{Y_{ij}^2}\right] \quad (1)$$

#### 2.3.1. Characterization

The compressive strength was used as the response variable for the analysis of variance (ANOVA) and was measured by testing 4 samples of each formulations at ages from 1 day and up to 180 days. The test were performed using an hydraulic press and a cell of 10 ton, fractured pieces were collected afterwards and placed in a plastic container and immersed for 72 h in methanol together with a small portion of an untested specimen for characterization by X-ray diffraction (XRD), scanning electron microscopy (SEM) and energy dispersive spectroscopy (EDS). The XRD analysis was carried out on a Philips D-Expert 3040 diffractometer using CuKα radiation and a scanning rate of 2°/s from 7° to 80°2θ. A Philips XL30 scanning electron microscope, operated at 20 keV was used to characterize the microstructure on polished samples using backscattered electron imaging, while EDS was used to determine the chemical composition of the products conforming the microstructure.

## 3. Results and discussion

### 3.1. Compressive strength

Fig. 1 shows the strength evolution from 1 to 28 days for the pastes grouped by the %UWG. The compressive strength in general

**Table 1**
Chemical composition (%wt.) of blast furnace slag and urban waste glass by X-ray fluorescence.

| Raw material | $\%SiO_2$ | %CaO | $\%Al_2O_3$ | $\%Na_2O$ | %MgO | $\%TiO_2$ | $\%K_2O$ | %MnO | $Fe_2O_3$ | $SO_3$ |
|---|---|---|---|---|---|---|---|---|---|---|
| BFS | 32.3 | 39.4 | 10.5 | – | 8.7 | – | 0.8 | 0.5 | 0.5 | 3.2 |
| UWG | 69.7 | 13.8 | 1.4 | 12.9 | 0.2 | 0.1 | 0.5 | – | 0.2 | – |