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Study of synergy between a natural volcanic pozzolan and a granulated blast furnace slag in the production of geopolymeric pastes and mortars

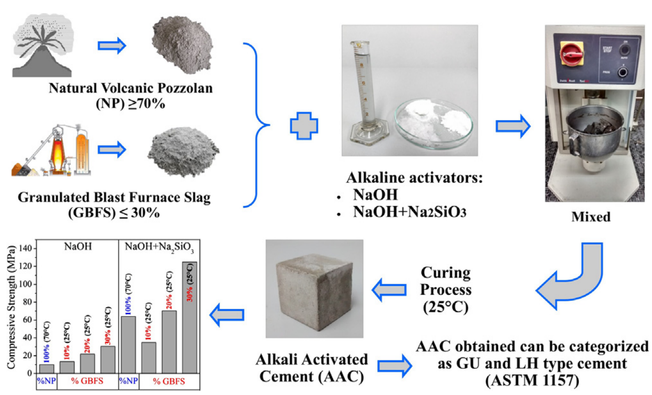
Rafael Andres Robayo-Salazar [a], Mejía de Gutiérrez [a,*], Francisca Puertas [b]

[a] Composites Materials Group (CENM), School of Materials Engineering, Universidad del Valle, Santiago de Cali, Colombia
[b] Eduardo Torroja Institute for Construction Science (IETcc-CSIC), Madrid, Spain

HIGHLIGHTS

- Alkali-activated cements based on high content (≥70%) natural pozzolan are obtained.
- The AACs based on 100% NP obtained a maximum of 64 MPa (70 °C curing).
- Addition of GBFS (30%) contributed to improve the mechanical behaviour (125 MPa) at 25 °C.
- AAC obtained can be categorized as GU- and LH-type cement according to ASTM C1157.

GRAPHICAL ABSTRACT



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ABSTRACT

The production and characterization of a cement and mortar based on the alkaline activation of a high content (≥70%) of natural volcanic pozzolan (NP) and the synergic effect of the addition of granulated blast furnace slag (GBFS) up to 30% were performed. NaOH and a mixture of NaOH + Na_2SiO_3 were used as activators. The results of this study highlight the possibility of obtaining a binder with a compressive strength of up to 125 MPa at 28 days of curing (25 °C). The optimal alkaline cement that was obtained can be categorized as GU- and LH-type cement according to the ASTM C1157 standard.

1. Introduction

“*Alkali-Activated Cements*” (AACs) are binders that result from the chemical interaction between strongly alkaline solutions (activator) and silicoaluminate (precursor), with low and high calcium contents, which may be of natural origin, such as natural clays or pozzolans, or of artificial origin, such as industrial waste and by-products. At relatively low processing temperatures (25–100 °C), this chemical interaction between the alkaline activator and the precursor promotes the formation, subsequent coagulation and precipitation of reaction products, which ultimately define the excellent mechanical and durable properties of this type

* Corresponding author at: Composites Materials Group (CENM), Universidad del Valle, Calle 13 # 100-00, Cali, Colombia.
E-mail address: ruby.mejia@correounivalle.edu.co (M. de Gutiérrez).

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of material. Recently, AACs have undergone extensive development and have gradually attracted the attention of the scientific and industrial world. They are considered materials of high potential, partly due to their technical and environmental advantages, their versatility and the ease of implementing new technologies into their production, which enables their application in various engineering fields. The precursors commonly used for obtaining AACs are metakaolin (MK), fly ash (FA) and Granulated Blast Furnace Slag (GBFS), with extensive and satisfactory studies reported since the past century by various authors worldwide [1–6].

Despite the excellent properties reported for FA-based or GBFS-based AACs, the very nature of these precursors (by-products) makes their chemical and mineralogical composition variable, making it difficult to standardize an alkaline activation process for the achievement of specific mechanical properties and durability. In addition, the global generation of these by-products is considered limited; 500–700 Mt/year for FA [7] and 170–250 Mt/year for GBFS [8], when compared with global production of ordinary Portland cement (OPC) (3–4 trillion tons/year) [9]. This would limit the replacement level of the OPC currently demanded by the construction industry for materials based exclusively (100%) on the alkaline activation of these by-products. The global demand for OPC is projected to increase by 216% by 2030; during this same period, the generation of FA and GBFS is projected to increase by only 15% [10]. As a solution to this limitation, "*Alkali-Activated Binary Cements*" have been developed, which are mixtures of two precursors as a base material and "*Hybrid Cements,*" also called "*Alkali-Activated Portland Blended Cement,*" which combine the two technologies (i.e. the positive effects of the OPC with the AACs). Thus, it would be feasible to produce cement based on a high content of aluminosilicates (≥70%) and a low level of OPC addition (≤30%) [11–13]. It should be noted that standards such as ASTM C1157-11 "*Standard Performance Specification for Hydraulic Cement*" in the USA and, recently, NTC 121 (third update) "*Performance Specification for Hydraulic Cement*" in Colombia no longer classified cements based on their performance but rather on their composition, enabling the introduction of AACs into the market as a real alternative to the OPC.

The local availability of Natural Pozzolans (NP) in significant quantities is promoted as a viable alternative from the commercial point of view (industrial scale) for the production of AACs since better quality control of the raw materials is possible [14,15]. While natural resources would be used, as in the production of OPC, AACs are obtained at relatively low temperatures (25–100 °C), and unlike the OPC, AACs do not require a clinkerization process (1350–1450 °C), which leads to a significant reduction in CO_2 emissions linked to this process. It is estimated that this reduction would reach values close to 70%, which is interesting considering that the cement industry is now responsible for 5–7% of the CO_2 emitted into the atmosphere, and it is estimated to exceed 10–15% in 2020 [16–21].

NP deposits account for approximately 0.84% of the soils worldwide (124 million ha), which are distributed predominantly (60%) in tropical regions [22]. These deposits have an important commercial value for the cement industry because they are suitable for the production of "*Blended Cements,*" especially in countries with high demographic and economic growth, such as Iran, Cameroon, Japan, China, Saudi Arabia, Turkey, Jordan, USA, Mexico, Chile, Ecuador and Colombia [23]. For example, in Colombia, NPs constitute 11% of their territory [24] and are concentrated in the central and southwestern areas; in Chile, they represent 50% of the country's total arable land, concentrating significantly among metropolitan regions [25], and in Japan, the NP covers 18% of its territory [26].

Several authors, including; Allahverdi et al. [27], Lemougna et al. [28], Bondar et al. [29] and Tchakoute et al. [30], report the potential of NPs as raw material for obtaining AACs. In general, NPs have a degree of amorphousness (≤40%) that makes them reactive in the presence of strongly alkaline solutions [31–34]. However, due to its predominant semi-crystalline nature and its deficiencies in CaO and reactive Al_2O_3 contents, applying calcination treatments that are thermal cured (40–90 °C) or above 700 °C is common in order to promote the mechanical performance of these AACs at early curing ages [35–38]. These practices limit the technological application of these materials on an industrial scale. An alternative to substitute the thermal curing of these binders is the incorporation of modifiers or secondary sources of CaO and reactive Al_2O_3 from mineral additions such as GBFS and MK [39–44]. The addition of GBFS, in low proportions (≤30%), allows the setting and hardening of "*Alkali-Activated Binary Cements*" at room temperature, promoting the formation of calcium silicate hydrated (C-S-H) and calcium aluminosilicate hydrated (C-A-S-H) [45,46].

By incorporating 5% (by weight) of GBFS, Allahverdi et al. [47] achieved compressive strengths of up to 36 MPa at 28 days curing (25 °C) for an Iranian NP (Taftan pumice) activated with NaOH + Na_2SiO_3 (MS = 0.6). In a later study, the authors emphasize that incorporating GBFS significantly reduces the presence of efflorescence [40]. From this same NP (Taftan Pumice), Nadoushan and Ramezanianpour [48] obtained compressive strengths of up to 42 and 55 MPa after 28 days of curing (25 °C) using alkaline solutions based on NaOH (8 M) and KOH (8 M), respectively, and a GBFS addition level of 25% (by weight).

In an earlier study, Robayo-Salazar et al. [15] demonstrated the feasibility of obtaining an "*Alkali-Activated Binary Cement*" based on a high content of a Colombian natural pozzolan rich in montmorillonite, reaching values of compression resistance (at the level of paste) up to 48 MPa at 28 days of curing at room temperature with the addition of 30% GBFS (by weight relative to NP) and the use of NaOH + Na_2SiO_3 as activator. However, prior to its alkaline activation, this NP required a calcination process at 700 °C to reduce or transform the montmorillonite phase.

This study presents a further advance in this line of research, such as the use of another Colombian natural pozzolan free of montmorillonite and rich in sodium-calcium feldspar as a more viable alternative for the preparation of the "*Alkali-Activated Binary Cements*" and its implementation at industrial scale. For its alkali activation, natural pozzolan does not need to undergo the calcination process that was required in the previous study, which is a limiting stage from a technological point of view. Additions (GBFS) are also used in contents of less than 30% by weight to replace the heat curing of the blends. This study aims to determine the effect of the addition of GBFS on the synergy and kinetic behaviour, mechanical performance, porosity and final microstructure of the mixtures. Finally, the potential application of this alternative binder in the construction industry through the preparation of a mortar, its classification based on NTC 121 (equivalent to ASTM C1157-11) and a comparison of its mechanical performance with a mortar based exclusively on OPC are shown.

2. Experimental methodology and materials

2.1. Materials

As precursors for obtaining the AAC, a NP of Colombian provenance and blast furnace steel slag (GBFS) were used. The chemical composition of these materials, which is presented in Table 1, was determined via X-ray fluorescence (XRF). The high SiO_2/Al_2O_3 molar ratio (6.79) and the low alkali content (CaO, Na_2O and K_2O) are notable for NP, whereas GBFS has a SiO_2/Al_2O_3 molar ratio of 4.93 and a high CaO content (40.3%).

Fig. 1 shows the pattern of X-ray diffraction of the precursors. The amorphous phase content of the NP was determined by