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One-part alkali-activated materials: A review

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ABSTRACT

Alkali-activated materials (AAM) are recognized as potential alternatives to ordinary Portland cement (OPC) in order to limit CO₂ emissions as well as beneficiate several wastes into useful products. However, the alkali activation process involves concentrated aqueous alkali solutions, which are corrosive, viscous, and, as such, difficult to handle and not user friendly. Consequently, the development of so-called one-part or “just add water” AAM may have greater potential than the conventional two-part AAM, especially in cast-in-situ applications. One-part AAM involves a dry mix that consists of a solid aluminosilicate precursor, a solid alkali source, and possible admixtures to which water is added, similar to the preparation of OPC. The dry mix can be prepared at elevated temperatures to facilitate the reactivity of certain raw materials. This review discusses current studies of one-part AAMs in terms of raw materials, activators, additives, mechanical and physical properties, curing mechanisms, hydration products, and environmental impacts.

1. Introduction

Ordinary Portland cement (OPC) contributes significantly to the global CO₂ emissions: in 2016, the estimated value was 1.45 ± 0.20 Gt CO₂, that is, approximately 8% of the total anthropogenic CO₂ release [1]. Although the cement industry has been able to improve energy efficiency significantly, increasing cement demand (estimated as 3.36–3.48 Gt in 2015 and 3.68–4.38 Gt per year by 2050) has outpaced much of the positive impact [2]. Furthermore, approximately 50–60% of OPC-production-related CO₂ emissions are released from the calcination (decarbonation) of limestone at 1400–1450 °C (Reaction 1), which cannot be influenced by improving energy efficiency [3,4].



Consequently, the development of alternative low-carbon binders is recognized as one option to reduce CO₂ emissions [5,6]. Geopolymers, which are sometimes considered a sub-group of alkali-activated materials (as originally suggested by Irene Beleña) are promising materials in this regard [7]. While they are unlikely to completely replace OPC, they may serve as an alternative and supplementary binder depending on the local availability of raw materials [8]. Alkali-activated materials could be designed to have superior properties compared to binders prepared from OPC, namely better resistance to acids and sulfate [9–11], better heat resistance [12–14], lower drying shrinkage and creep [15], and higher strength [16]. However, efflorescence formation can be a potential problem if not properly controlled by, for example,

mix design [17]. In terms of unwanted alkali-aggregate reactions, alkali-activated materials have frequently showed better performance than OPC, as documented, for instance, by García-Lodeiro et al. [18] but attention should be paid for the proper selection of type and dose of activator, type of binder, and type of aggregates in this regard [19]. Alkali activation technology also allows a waste beneficiation route for utilizing several industrial by-products [20]. In addition, many of the current standards for cementitious materials do not recognize the use of alkali-activated materials as they have been written from the viewpoint of OPC use (i.e., they include prescriptive compositional limits instead of performance-based specifications) [21]. However, in some countries, such as Ukraine [22], the use of alkali-activated cements is already included in the legislation. Also, supply chains for raw materials, suitable admixtures for alkali-activated materials, and testing protocols are still inadequate [23].

Binders prepared by reacting an alkali source and solid aluminosilicate were patented as early as 1908 [24]. Later, important pioneering work was done by Glukhovskiy [25], Krivenko [26], Davidovits [27], and Palomo et al. [28] among many others. A complete historical overview of alkali-activated materials is available in several reviews [29–32]. However, there is still some ongoing debate about the terminology related to alkali-activated materials and geopolymers: the former is generally characterized by a lower coordination of Si, that is, Q² and Q²(1Al) and higher calcium content, whereas the latter contains more Q⁴(2Al) and Q⁴(3Al) centers, has low calcium content, and is essentially three-dimensional [7,33–35]. However, the terms

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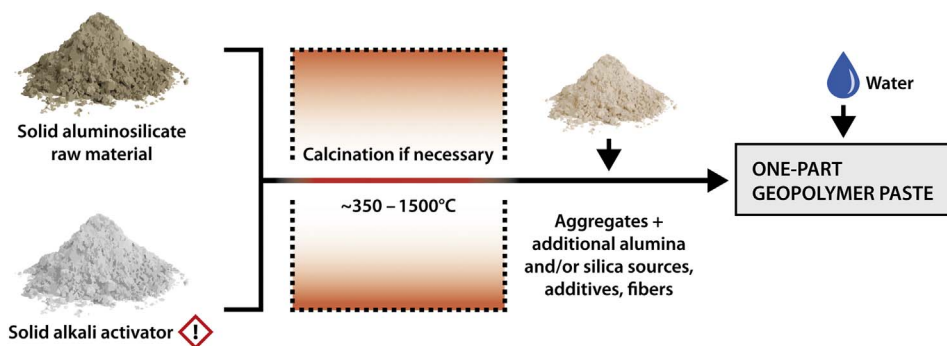


Fig. 1. The general procedure of one-part geopolymer preparation.

geopolymer and alkali-activated material are used in the literature somewhat interchangeably and occasionally even as synonyms. In this review, materials are referred to as using the same terminology as used in the cited references.

Conventional (two-part) geopolymers are formed by a reaction between a concentrated aqueous solution of alkali hydroxide, silicate, carbonate, or sulfate, for instance, and solid aluminosilicate precursor, that is, two parts in addition to water [36–40]. However, the impracticalities related to handling large amounts of viscous, corrosive, and hazardous alkali activator solutions has put pressure on the development of one-part or “just add water” geopolymers that could be used similarly to OPC [41]. In one-part mixtures, only a dry mixture is needed in addition to water. The dry mixture is prepared by mixing a solid alkali-activator with a solid aluminosilicate precursor with or without a calcination step (Fig. 1).

In 1940, Purdon [42] proposed dry mixing of slag and solid sodium hydroxide and subsequently adding water in order to prepare a mortar mixture. In the 1980s, Heitzmann et al. [43] patented a dry mixture of metakaolin, blast furnace slag, amorphous silica, potassium hydroxide and silicate, and one of the following components: fly ash, calcined shale, or calcined clay that could be blended with OPC before the addition of water. Schwarz and Andre [44] patented a geopolymeric dry mixture in which amorphous silica was prepared by dealuminating fly ash or metakaolin with hydrochloric or sulfuric acid. Davidovits [45] also patented one-part geopolymer consisting of aluminosilicate oxide with Al in IV-V coordination (i.e., metakaolin), sodium or potassium disilicate, and slag. Later, Davidovits [46] pointed out that solid sodium or potassium hydroxide and silicate should be partially replaced with “synthetic lavas” (reactive sodium-potassium aluminosilicate glass) due to large-scale availability and production issues. Synthetic lava could be prepared by mixing volcanic tuff and calcium carbonate, melting at 1200–1350 °C, quenching in water, drying, and grinding [46]. Duxson and Provis [41] outlined some general needs and approaches for one-part geopolymer mixes. They proposed that solid precursor would be prepared either by adding, for instance, calcium-containing feldspar to coal before combustion; melting the previous materials separately and grinding; or preparation of a two-part geopolymer as a precursor to a one-part geopolymer. Currently, one-part geopolymers are considered especially promising for in situ applications where handling alkali solutions can be difficult whereas two-part mixtures appear suitable for precast work [8,23,47]. However, the early examples of one-part geopolymers described above have attained little commercial use. In contrast, two-part geopolymers have already been used in several full-scale implementations, such as the Brisbane West Wellcamp Airport in Australia [48].

The purpose of this review is to present the current state-of-art in the development of one-part geopolymer mixtures. A substantial number of review articles exist discussing various aspects and applications of two-part geopolymers [49–61], but no similar reviews about one-part mixtures currently exist. This paper discusses the employed aluminosilicate precursors, solid activators, admixtures, mix designs,

resulting binding phases, hardening mechanisms, and effects of various parameters on the mechanical properties of one-part geopolymers. Moreover, a brief overview of one-part geopolymers containing OPC as one component (i.e., hybrid cements or blended alkaline cement) is presented. Finally, the environmental impact and cost analysis of one-part mixes is provided.

2. Raw materials and preparation of one-part geopolymers

2.1. Aluminosilicate precursors

The most common solid aluminosilicate precursor in one-part geopolymer mixes is fly ash from coal combustion either alone or in combination with blast furnace slag (Table 1). The majority [62–69] of the fly ashes used as precursors fall within class F (low calcium content) as defined by ASTM standard C618 [70]. Class C (high calcium content) fly ash is less frequently used in geopolymer binders because of too rapid setting [71,72] and less abundant availability [73]. Ye et al. [74] used one-part geopolymer approach on the successful solidification/stabilization of municipal solid waste incineration fly ash (containing metals such as Pb, Cu, Zn, and Cr) by mixing it with red mud, NaOH, and then adding water. While fly ashes have usually been employed without pretreatment in one-part mixes, Matalkah et al. [75] used a mechanochemical activation method in which fly ash was ball-milled with dry-blended activators (CaO, MgO, and NaOH). They observed that Na, Ca, and Mg were incorporated in the fly ash structure due to disruption of the aluminosilicate bonds, which resulted in improved properties (higher strength, greater moisture resistance, and finer microstructure) compared to raw materials that were separately milled and then blended [75]. Similar mechanochemical pretreatments have been used with two-part geopolymers as well with a significant increase in compressive strength [76] and rate of geopolymerization [77].

Ground-granulated blast-furnace slag is commonly used as a calcium-rich aluminosilicate precursor in alkali-activated materials. Blast furnace slag, which is a by-product of pig iron manufacturing at blast furnaces, consists of silicate and aluminate impurities present in iron ore and coke. Blast furnace slag is frequently mixed with class F fly ash in one-part geopolymer mixtures, but it can also be used alone (Table 1). Using blast furnace slag improves the reactivity of low-calcium-content fly ash [41]. Usually calcium-rich aluminosilicate or calcium hydroxide would induce rapid setting and high early strength [64,78], but replacement of a portion of the slag with calcium hydroxide resulted in a decrease in both compressive strength and workability in a one-part geopolymer [65]. Decreasing the particle size of blast furnace slag resulted in an increase of compressive strength [63]. Wang et al. [66] found that a ratio of blast furnace slag to solid water glass of 5:1 (by weight) was optimum in terms of compressive strength, whereas ratios of 4:1 and 6:1 led to decreased strength. Nematollahi et al. [67] used “typical” and “gypsum-free” slags in the design of one-part strain-hardening geopolymer composite. The use of “typical slag” resulted in a lower relative slump value, significantly higher thixotropic

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