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# Alkali-Activated Cements— Report

Reported by ACI Committee 242

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## **Alkali-Activated Cements—Report**

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# Alkali-Activated Cements—Report

Reported by ACI Committee 242

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*This report provides a technical overview of alkali-activated cement (AAC) history; chemistry; and mechanical, physical, and durability properties. It also highlights successful case study applications and current technical challenges.*

**Keywords:** alkali-activated cements (AACs); alternative cements; geopolymers.

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### CHAPTER 1—INTRODUCTION AND SCOPE

#### 1.1—Introduction

Concrete has traditionally been made with portland cement (PC) binders and PC binders with supplementary cementitious materials, but several emerging alternative cements offer distinctive properties that may be beneficial for some applications (ACI ITG-10.1R). Alkali-activated cements (AACs) are a distinct class of cements that react using alkali activation to form a cement paste solid. This activation consists of using an alkali- or alkali-salt-based activator to promote the dissolution of an aluminosilicate precursor and subsequent precipitation of cementitious reaction products. Depending on the aluminosilicate precursor (for example, fly ash, ground-granulated blast-furnace slag, calcined clays), alkali activation can initiate a series of either polycondensation reactions, in which water is produced as a result of reaction product formation, or hydration reactions similar to PC, in which water is consumed (Pacheco-Torgal et al. 2014).

AACs are sustainable, low-to-zero-clinker cement alternatives to PC due to their achievable mechanical and durability properties. They also have lower manufacturing carbon dioxide (CO<sub>2</sub>) emissions, which are largely due to the use of industrial by-product materials as precursors (McLellan et al. 2011; Habert and Ouellet-Plamondon 2016). In general, successful precursors used for AACs should contain significant amounts of reactive silica (SiO<sub>2</sub>) and alumina (Al<sub>2</sub>O<sub>3</sub>), whereas the presence of additional oxides, such as calcium oxide (CaO), can result in changes to the resultant phase assemblages (Provis et al. 2014). The most common precursors include fly ash, ground-granulated blast-furnace slag, and natural or calcined clays (for example, metakaolin). The most widely used alkaline solutions include alkali silicates (for example, sodium silicate) and alkali hydroxides (for example, sodium hydroxide). Other activators include alkali carbonate and sulfate salts, which have been demonstrated to have better compatibility with conventional chemical admixtures and are less caustic (that is, lower pH) when implemented in cast-in-place field applications (Bernal 2016). However, the strength development of AACs activated with alkali carbonate and sulfate salts often depends more on the physical (for example, fineness) or chemical (for example, reactive glass) characteristics of the chosen precursor (Wang et al. 1994). Alkali aluminates—especially the by-products of alumina refining—are also capable of activating AAC precursors (van Riessen et al. 2013) and may offer a potentially cost-

effective alternative to other activators produced from virgin materials.

This report provides a technical overview of AAC history, chemistry, mechanical and physical properties, and durability. It also highlights successful case study applications and current technical challenges. The results from extensive research in the field have revealed that the engineering properties of specific formulations of AACs can be comparable to PC or blended hydraulic cements. In some applications, AACs can provide better durability and mechanical performance. Nonetheless, AACs constitute a class of materials that is suitable to use as part of a toolkit of various concrete technologies. Thus, while AACs may not be considered a direct replacement to PC or blended hydraulic cementitious materials for every application, AACs may meet or exceed economic and technical performance requirements in some applications.

#### 1.2—Scope

This report defines relevant AAC terminology and a brief history of AACs. The fundamental materials science and engineering of AAC technology is discussed, along with the definitions of important parameters required for proper mixture proportioning. This report summarizes the fresh- and hardened-state properties of AAC concrete, with a specific focus on reaction kinetics, mechanical properties, and durability. Finally, this report summarizes challenges and opportunities for the AAC field and presents an overview of full-scale industrial applications and safety considerations of AAC concrete.

### CHAPTER 2—DEFINITIONS

Please refer to the latest version of ACI Concrete Terminology for a comprehensive list of definitions. Definitions provided herein complement that resource.

**alkali**—a chemical compound containing one or more elements belonging to Group 1 or Group 2 of the periodic table of elements.

**alkali activating powder**—an alkali powder that can be mixed with a precursor to form one-part alkali-activated cements.

**alkali activating solution**—an alkali-based solution that causes the dissolution of the precursor(s).

**alkali activation**—the process of using an alkali-based solution or powder to promote the dissolution of an aluminosilicate precursor and the precipitation reactions leading to the formation of reaction products.

**alternative cement**—an inorganic cement that can be used as a complete replacement for PC or blended hydraulic cements and that is not covered by applicable specifications for PC or blended hydraulic cements.

**geopolymer cements**—a subset of AACs comprising low-calcium (or calcium-free) alkali-activated precursors, such as metakaolin, that produce amorphous inorganic three-dimensional (3-D) aluminosilicate structures that resemble natural mineral systems.

**one-part alkali-activated cement**—an AAC in which the precursor is premixed with an alkali

activating powder that, when mixed with water, causes alkali activation.

**precursor**—a solid powder, usually composed of both alumina and silica, that may dissolve when mixed with an alkali activator and precipitate amorphous or semi-crystalline cementitious binders capable of hardening.

### CHAPTER 3—ALKALI-ACTIVATED CEMENT HISTORY

Although alkali-activated cements (AACs) have been the subject of much research in recent decades, this class of cements represents a century-old technology. The earliest published work related to AACs is a U.S. patent granted to Jasper Whiting for manufacturing a cement using ground-granulated blast-furnace slag, water, and caustic soda (Whiting 1895). In 1908, German scientist Hans Kühl demonstrated the stabilization of powdered ground-granulated blast-furnace slag with caustic soda (Kühl 1908). In 1940, Belgian scientist A. O. Purdon (Purdon 1940) published more extensive research on alkali-activated “clinkerless” cements. Two decades later, in the former Soviet Union, V. D. Glukhovskiy (1965) studied the alkali activation of various soil silicates as structural materials. Glukhovskiy’s alkaline cements were used throughout the former Union of Soviet Socialist Republics (U.S.S.R.) (Provis et al. 2014). In addition to the Trief cements and F-cements developed in Scandinavia in the 1980s (Roy 1999), Davidovits (Davidovits and Sawyer 1985) had made extensive progress in describing the nature of a specific formulation of alkali-activated binders, named geopolymers. These binders were briefly marketed in the United States under the trade name Pyrament (Roy et al. 1989). While the durability and high early strength of this product made it a suitable repair medium, Pyrament production ceased in the mid-1990s. Outside of the United States, AACs have been commercialized and used in select structural settings (Provis et al. 2014), but AACs have not yet experienced the same degree of commercialization and application within the U.S. concrete industry.

### CHAPTER 4—ALKALI-ACTIVATED CEMENT TECHNOLOGY AND MIXTURE PROPORTIONING

#### 4.1—Alkali-activated cement technology

Alkali-activated cements (AACs) are synthesized by dissolving aluminosilicate precursors, such as fly ash, ground-granulated blast-furnace slag, or metakaolin using an alkali activator. The activator tends to be in solution form, although one-part AAC mixtures using a “just add water” approach have been developed (Luukkonen et al. 2018). The most common activators are alkali silicates and alkali hydroxides, where the concentration of alkali and the amount of silicates are tailored according to the precursor(s). In general, sodium or potassium is used as the alkali source. Other activators, such as alkali carbonates and alkali sulfates, have been used, typically when using a high-calcium precursor, such as ground-granulated blast-furnace slag (GGBFS), but strength development can be slow in sulfate-activated systems. AACs

can comprise a single precursor (for example, fly-ash-based AACs) or blended precursor systems (for example, fly-ash- and slag-based AACs). Alkali aluminates are also viable activators, especially for high-silica precursors.

Calcium- and sodium-stabilized aluminosilicate hydrate (for example, C-(N)-A-S-H, C-A-S-H, N-A-S-H) gels are the main reaction products of alkali activation (Provis et al. 2014). High-calcium AACs, such as GGBFS-based AAC, consist of C-(N)-A-S-H gel with silicate chain “dreierketten” structure. Low-calcium AACs that form N-A-S-H exclusively, such as metakaolin-based AAC, are also termed geopolymer cements due to the formation of three-dimensional (3-D) aluminosilicate networks consisting of -Si-O-Si- and sodium-stabilized -Si-O-Al- linkages. N-A-S-H contains minimal bound water within the aluminosilicate network. In addition to the predominately amorphous phases, crystalline phases, including hydrotalcite and zeolites, are common reaction products. Similar to C-S-H in PC, the aluminosilicate hydrate gels are primarily responsible for the strength of hardened AACs.

#### 4.2—Mixture proportioning

The following are important considerations in the design and proportioning of AACs.

**4.2.1 Physical and chemical properties of precursors—**Increased precursor fineness and lower degrees of crystallinity (higher amorphous content) accelerate precursor dissolution during alkali activation. The chemical composition can vary greatly between different precursors (refer to Fig. 4.2.1), which in turn also affects initial dissolution kinetics and reaction products. One important consideration is the presence of calcium in the precursors. The more calcium present in the reactive amorphous phase of the precursor, the lower the overall alkali concentration required to achieve sufficient alkali activation and resulting fresh- and

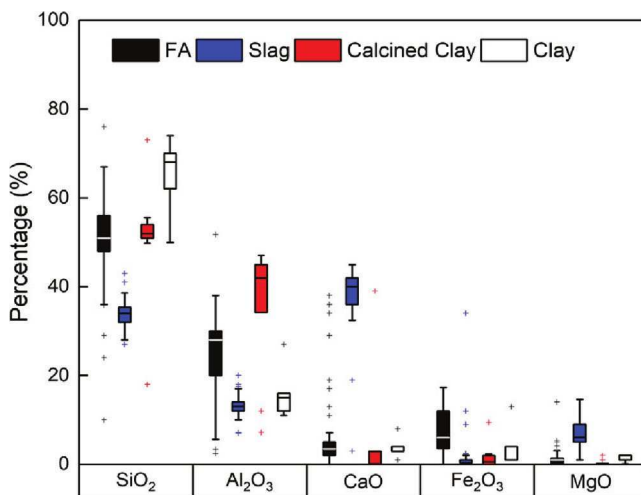


Fig. 4.2.1—Average chemical composition (wt. %) of common aluminosilicate AAC precursors. Data compiled from more than 65 studies of AACs. Median data are represented by centerlines in the box plots (Oslo-Norgaard et al. 2018).

hardened-state properties. Higher calcium content can thus accelerate the setting time of the resultant AAC.

**4.2.1.1 Fly ash**—Fly ash is primarily composed of silicon dioxide ( $\text{SiO}_2$ ), aluminum oxide ( $\text{Al}_2\text{O}_3$ ), iron oxide ( $\text{Fe}_2\text{O}_3$ ), and varying amounts of calcium oxide ( $\text{CaO}$ ). Most fly ash is in the form of amorphous aluminosilicate glass. Other oxides are typically contained in four crystalline mineral phases: quartz, mullite, hematite, and magnetite, which together can form 10 to 50% of the bulk ash (Dananjayan et al. 2016). Fly ash is classified according to its chemical composition into two major categories: low calcium and high calcium (Class F and Class C, respectively, as specified by ASTM C618). These classifications depend on the calcium content and do not necessarily address the degree of crystallinity. Upon alkali activation to produce a fly-ash-based AAC, the formation of C-A-S-H, C-(N)-A-S-H, and N-A-S-H phases is common. For unclassified fly ash, it is important to determine the relative amount and chemical composition of the amorphous aluminosilicate glass in the fly ash source prior to its use.

**4.2.1.2 Ground-granulated blast-furnace slag**—Granulated slag is a highly vitreous, highly reactive by-product of the iron-making industry that is produced with a consistent chemical composition. Primarily composed of a glassy phase consisting of the oxides (for example,  $\text{CaO}$ ,  $\text{SiO}_2$ ,  $\text{Al}_2\text{O}_3$ , and  $\text{MgO}$ ), slags usually contain high  $\text{CaO}$  (30 to 50%), but not as high as PC (greater than 60%). Out of all precursors shown in Fig. 4.2.1, slags consistently contain the lowest quantities of both  $\text{SiO}_2$  and  $\text{Al}_2\text{O}_3$ . Upon alkali activation to produce a slag-based AAC, C-(N)-A-S-H forms, which is the predominant cementitious phase (Myers et al. 2014).

**4.2.1.3 Metakaolin and other calcined clays**—Metakaolin is a distinctly pure aluminosilicate precursor high in  $\text{SiO}_2$  and  $\text{Al}_2\text{O}_3$ . Natural clays, such as kaolin (consisting of the crystalline mineral phase kaolinite), contain varying amounts of  $\text{SiO}_2$  and  $\text{Al}_2\text{O}_3$  due to the presence of impurities such as anatase and quartz. Calcination of kaolinite (that is, heated to 932 to 1472°F [500 to 800°C]) leads to the formation of metakaolin with approximately 50%  $\text{SiO}_2$  and 50%  $\text{Al}_2\text{O}_3$  by weight. Calcination induces crystalline-to-amorphous structural changes and, consequently, increases the reactivity of kaolin clays by lowering the degree of crystallinity. Upon alkali activation, the formation of amorphous N-A-S-H is common. Crystalline zeolites can also form, especially if the cements are heat-cured, alkali-activated, or both, using sodium hydroxide (Gevaudan et al. 2017).

**4.2.1.4 Other**—Other aluminosilicates, such as other ashes, virgin glasses, ground waste glass, red mud, other calcined clays (nonkaolinitic), and uncalcined clays have been used for alkali activation (Valentini et al. 2018; Tian et al. 2021; He et al. 2021). While naturally occurring—crystalline aluminosilicate clays can be used to produce AAC binders in their raw form—it is well understood that calcination (heat-treatment between 752 and 1832°F [400 and 1000°C]) induces dehydroxylation and crystalline-to-amorphous structural changes, which consequently increases reactivity (Valentini et al. 2018). Thus, uncalcined clays are not typically used as precursors for alkali activation.

**4.2.2 Alkali activator**—The alkali activator may be added to a mixture in the form of a liquid solution or in powder form. In general, increasing available silica in the alkali activator has been shown to lead to finer pore sizes and reduced porosity independent of precursor chemistry (Osio-Norgaard et al. 2018). Similarly, increasing alkali content (generally  $\text{Na}/\text{Al} \geq 0.75$  for low-calcium AACs) promotes rapid dissolution of precursors and leads to reductions in bulk porosity. It is important to note, however, that these results are highly dependent on the chemical composition and physical properties of the specific aluminosilicate precursor.

Although alkali-silicate or alkali-hydroxide solutions are the most common activator systems found in the scientific literature, waste streams or secondary sodium aluminate solutions, carbonates, and sulfates can produce viable AACs (for example, sodium aluminate solutions produced by aluminum refineries) (van Riessen et al. 2013). The cost and sustainability advantages of using industrial by-products instead of virgin silicate/hydroxide solutions offer intriguing commercialization prospects for high-volume applications.

**4.2.3 Common binder design parameters**—Important mixture design parameters include:

(a) **Si/Al (or  $\text{SiO}_2/\text{Al}_2\text{O}_3$ ) molar ratio**: Ratio of the total quantities of silica to alumina in an AAC system

(b) **Si/Na ( $\text{SiO}_2/\text{Na}_2\text{O}$ ) or Si/K ( $\text{SiO}_2/\text{K}_2\text{O}$ ) molar ratio**: Ratio of the total quantities of silica to sodium or potassium in an AAC system

(c) **Na/Al ( $\text{Na}_2\text{O}/\text{Al}_2\text{O}_3$ ) or K/Al ( $\text{K}_2\text{O}/\text{Al}_2\text{O}_3$ ) molar ratio**: Ratio of the total quantities of sodium or potassium to alumina in an AAC system

(d) **Silica modulus (Ms)**: The  $\text{SiO}_2/\text{Na}_2\text{O}$  ratio of activator

(e)  **$\text{Na}_2\text{O}$  wt. %**: The mass ratio of  $\text{Na}_2\text{O}$  to precursor

(f) **Water-to-precursor**: The mass ratio of water to solid precursor

(g) **Liquid-to-solid ratio (L/S)**: A less frequently reported parameter that includes the total mass percentage of all liquids (including dissolved alkalis and silicates) to all solids in the mixture

It is critical that AAC formulators indicate whether these mixture design parameters refer to bulk compositions or indicate the chemical composition of the binders. In most cases, these mixture design parameters are reported in oxide molar ratios (for example,  $\text{SiO}_2/\text{Al}_2\text{O}_3$ ) to indicate the bulk chemical composition of the material, whereas the elemental ratio (for example,  $\text{Si}/\text{Al}$ ) is reserved to describe the cementitious binder chemical composition. The difference between bulk and binder-specific ratios arises from the partial dissolution of the precursor due to the alkali-activating process. It is also critical to note that care should be taken when measuring oxide compositions. Different chemical composition analysis methods can yield different results due to the accuracy or the assumptions used within the method. Some methods, such as X-ray fluorescence (XRF), report elemental compositions and those elements are assumed to be present in oxide form. Researchers should note which method was used to characterize the chemical composition of the precursors and activator materials.