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Geopolymers: from origins to advanced applications of sustainable and adaptive materials

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Abstract

With the increasing urgency to reduce greenhouse gas emissions, geopolymers and alkali-activated materials (AAMs) present a low-carbon alternative to traditional cement-based construction materials. The production of standard cement contributes to 5-7% of global CO_2 emissions, so finding sustainable substitutes is crucial. Geopolymers, derived from aluminosilicate sources such as fly ash and metakaolin, are created through alkaline activation, forming a stable three-dimensional structure that offers durability and resilience. Similarly, AAMs are synthesized by activating calcium-rich materials in highly alkaline conditions, forming compounds like C-S-H gels that provide structural integrity. These materials demonstrate potential in a range of construction applications, including building restoration, fire-resistant materials, and environmental remediation. Geopolymers are especially promising for cultural heritage restoration due to their ceramic-like structure, durability, and compatibility with traditional materials. Despite their benefits, challenges remain, such as controlling the setting time, ensuring consistent precursor supply, and managing their behaviour in fresh states. Standardization is progressing, with several countries developing guidelines to support their use in infrastructure projects. Overall, geopolymers and AAMs are seen as viable paths toward sustainable building practices, though further research is needed for broader application.

Keywords: geopolymer, sustainability, aluminosilicate materials, amorphus micro-structure.

1. Introduction

As climate change impacts intensify, manifesting in rising sea levels, shifting precipitation patterns, and increased desertification, there is a mounting urgency to reduce greenhouse gas (GHG) emissions.

The construction industry, a substantial contributor to global emissions, demands new solutions to minimize environmental harm. Specifically, traditional concrete production, with cement as a primary component, is responsible for 5-7% of global CO_2 emissions (Worrell, 2001). The growing demand for concrete, driven by population growth and infrastructure needs, exacerbates the need for sustainable alternatives.

In this context, geopolymers and alkali-activated materials (AAM) emerge as a viable, low-carbon substitute, holding the potential to cut CO_2 emissions and resource usage in concrete manufacturing dramatically (Provis et al., 2010; Duxson et al., 2007).

2. Composition and Properties of geopolymers and AAMs

The geopolymers and AAMs are aluminosilicate ceramic systems prepared through an alkaline activation reaction between aluminosilicate-rich source materials and alkaline activating solutions. Particularly, geopolymers consist of an inorganic aluminosilicate polymer with a network of tetrahedrally coordinated Si^{4+} and Al^{3+} ions, forming polymer chains (Thapa et al., 2018; Aversa et al., 2023; Aversa et al., 2024). These geopolymer precursors link chemically, forming oligomers and eventually creating a three-dimensional structure. In contrast, alkali-activated binders are synthesized by activating calcium-rich source materials under high-alkaline conditions. When precursors dissolve in the alkaline solution, they produce a hydraulic binder paste, leading to the concurrent formation of C-S-H gels and aluminosilicate polymers. Figure 1 illustrates the differences in the reaction mechanisms between alkali-activated binders and geopolymers.

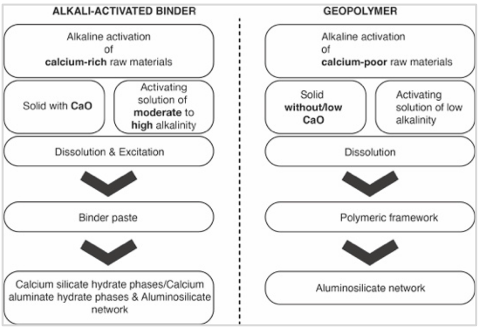

Fig. 1- Reaction mechanisms of geopolymers and AAMs (Thapa et al., 2018).

Studies on alkali-activated materials and geopolymers identify two main types of chemical reactions occurring between alkaline activators and aluminosilicate sources: high-calcium/magnesium and low-calcium/magnesium systems (see Fig. 1). In high-calcium/magnesium systems, calcium/magnesium aluminosilicate hydrates are produced, commonly referred to as C(M)-A-S-H, typically forming a Q2 layered structure. Conversely, in low-calcium/magnesium systems, the silicon and aluminum atoms coordinate in a tetrahedral arrangement, primarily Q3 or Q4. In Q4 structures, positively charged alkaline ions balance the negative charge from tetrahedral aluminum and silicon coordination, resulting in a three-dimensional gel structure. The presence of Q1, Q2, Q3, and Q4 structures is influenced by the Si/Al ratio (Duxson et al., 2005) and the specific reaction environment.

Moreover, geopolymers and AAMs can be prepared starting from natural aluminosilicate sources (metakaolin) and industrial by-products such as rice husk ash, fly ash, slag, waste glass, copper mine tailings, red mud, zeolite, pure Al_2O_3–$2SiO_2$ powder with Na_2SiO_3, magnesium-containing minerals and palm oil fuel ash (POFA) obtained from Malaysia. The commonly used alkaline activators are NaOH, potassium hydroxide (KOH), calcium hydroxide ($Ca(OH)_2$), NaOH and Na_2SiO_3, KOH and K_2SiO_3 (potassium silicate), K_2CO_3 (potassium carbonate) and $Ca(OH)_2$, NaOH and Na_2CO_3 (sodium carbonate) (Wang et al., 2019; Singh et al., 2020).

Geopolymers and AAMs can find applications in conventional sectors like construction, repair, restoration, marine engineering, pavement base layers, 3D printing, fire-resistant and high-temperature materials, as well as thermal and acoustic insulation. Specialized uses include immobilizing heavy metal contaminants, pH regulation, catalysis, conductive materials for moisture sensors, and thermal storage. Functional applications include specific applications such as fire-resistant structures, insulation walls, and nuclear facilities, extend to fire protection, isolation, heat retention, adsorption of hazardous ions, and bioscaffolds (Burduhos Nergis et al., 2018; Ricciotti et al., 2020; Ricciotti et al., 2023).

3. History of geopolymers and AAMs

The origins of geopolymers and AAMs technology trace back to the early 20th century when Kuhl (Kuhl, 1930) developed AAMs by combining alkalis (NaOH and KOH) with ground slag to examine its durability and strength relative to traditional OPC concrete. Kuhl earned the first patent for AAB technology in 1908 by pioneering the alkali activation of aluminosilicate-based materials (Kuhl, 1908). Later, in 1939, Feret explored partially replacing OPC with ground granulated blast furnace slag (GGBFS), known as alkali-activated slag (AAS), though he was unable to demonstrate practical application (Feret, 1939). Significant advancements in AAB research began in 1940 through extensive laboratory studies by a Belgian scientist (Purdon, 1949). Purdon's research highlighted that AAS could achieve rapid hardening in two stages with NaOH as the activator. First, $Ca(OH)_2$ and alkaline activators induce the release of Si and Al atoms from precursor materials through various physicochemical processes. Then, the alkali metals form hydrated aluminosilicate (A-S-H) gel. In 1955, Victor Trief introduced 'Trief Cements', the first AAS brand utilized in large-scale construction (The Trief Cement Process, 1955). Victor Glukhovsky, a Russian scientist (Glukhovsky, 1959), examined ancient Roman and Egyptian construction, noting that the primary binding products were the anaclitic phase and C-S-H (alongside C-A-S-H), suggesting that AAB could be developed using calcium-free aluminosilicate in an alkaline medium. He further supported his theory by synthesizing 'soil cement' from powdered aluminosilicate-based industrial materials and potent alkaline activators from group 1 elements (Na and K). A breakthrough in AAMs occurred in the late 1970s when a French researcher (Davidovits, 1979) created aluminosilicate-based composite binders by combining limestone, dolomite, and kaolinite with alkaline substances. Davidovits, who first coined the term *geopolymers* for these binders, observed that their composition and structure resembled zeolites, with polymeric frameworks. His work on geopolymer binders was first documented in a U.S. patent by Davidovits and Sawyer (Davidovits and Sawyer, 1986).

Today, geopolymers have been further refined for practical infrastructure applications and are marketed under names like 'Pyrament' (Roy, 1999). Similar materials, such as 'F-cement' in Finland (Forss, 1983) and 'geocement' in Ukraine (Krivenko, 1994a), were also introduced. Since then, AAMs have seen rapid development, widespread research, and global acceptance, making them some of the most commonly used sustainable construction materials. For example, in Australia, Victoria's state road agency, VicRoads, has constructed several rigid pavements using geopolymer concrete (Deventer, 2012), and around 40,000 m^3 of geopolymer concrete was used for various infrastructure projects at Brisbane West Wellcamp Airport in Queensland (Glasby, 2015). Other Australian projects include the first precast use of geopolymer concrete in structural elements at the Global Change Institute building (Aldred and Day, 2012; Bilgh and Glasby, 2013), retrofitting the Northern Wall of Port Kembla harbor with Hanbar armour units (Mahmood et al., 2020), and Pinkenba Wharf in Brisbane (Manalo et al., 2021). Additionally, AAM technology has been standardized and applied in various projects across the UK, USA, Russia, South Africa, Ukraine, and the Netherlands (Cross et al., 2005; McIntosh et al., 2015; Provis et al., 2009).

The recent rise in commercial applications of geopolymer concrete is supported by decades of research since its groundbreaking developments in the 1980. Later research focused on understanding reaction mechanisms, product formation, and the role of different precursor materials and activators that underlie this technology.

According to van Deventer et al. (2010), 'geopolymer' represents a subset within the broader category of AAMs. Inorganic polymers, such as GGBFS, metakaolin, and their blends, fall under AAMs. In 1999, Palomo et al. proposed two models for categorizing AABs: the Si+Ca system, activated by mild alkali and producing C-S-H as the primary reaction product, and the Si+Al system, activated by strong alkalis like those found in fly ash or metakaolin-based binders. Duxson et al. (2006) classified the former as AAB and the latter as 'geopolymer', whose polymeric structure resembles natural zeolite. Other terms for aluminosilicate materials include 'inorganic polymers', 'geocements', 'alkali-activated cements', 'alkali-bonded ceramics', and 'hydroceramics'. Despite the variety in terminology, these names all refer to materials synthesized through similar chemical processes.

4. Applications in Cultural Heritage

AAMs show structural and mineral properties closely resembling ceramics, with chemical, physical, and mechanical characteristics that lend them exceptional durability. These qualities make them suitable as fillers for voids and cracks in tiles and repair mortars, offering an alternative to traditional materials for cultural heritage restoration. Unique properties, such as durability in harsh environmental conditions, strong adhesion to ceramic surfaces (due to their similar structures), rapid drying, and polymerization, make geopolymers particularly promising for conservation and restoration (Geraldes et al., 2016; Moutinho et al., 2020; Baltazar et al., 2019; Clausi et al., 2016; Ricciotti et al., 2017; Glasby et al., 2015; Aldred and Day, 2012). However, they present some challenges, such as the potential corrosion of the artefact's historical glaze or the formation of soluble salts when exposed to air, which may result from the high concentration of alkaline activators that can lead to carbonation processes. Geopolymers and hybrid materials derived from these sustainable binders remain an active research area in cultural heritage, with studies over recent decades exploring their use in stone conservation, tile repair, and structural consolidation.

For instance, Geraldes et al. (2016) examined metakaolin-based geopolymers for tile repair, noting that water evaporation is crucial to the polymerization reaction, influencing both cracking and adhesion of the geopolymer paste to ceramics. Moutinho et al. (2020) also explored using metakaolin-derived geopolymers to repair cracks in Portuguese tiles, while Clausi et al. (2016) and Baltazar et al. (2019) researched formulations involving decorative stones and fly ash to enhance compatibility and durability for stone structures. Clausi et al. (2016) found that adding stone aggregates, such as Pietra Serena and Pietra di Angera, to metakaolin geopolymers reduced mechanical strength slightly but remained within acceptable mortar standards, making them suitable for stone object restoration. Additionally, Baltazar et al. (2019) explored using fly ash-based geopolymers as consolidation materials for older stone buildings.

Beyond simple geopolymer and AAM applications, new materials can be developed by functionalizing geopolymers with various molecules to enhance mechanical, chemical, physical, and aesthetic qualities. For example, Ricciotti et al. (2017) created hybrid materials by combining epoxy resins with metakaolin geopolymers, while this review proposes a synthesis approach for new, enhanced green binders. Studies emphasize the advantages of geopolymers, such as compatibility with diverse materials, low CO_2 emissions, the use of industrial by-products, and resilience to atmospheric pollutants and harsh agents. With tailored formulations, geopolymers can be fire-resistant, breathable, and resistant to rising salts and acid rain, and can mimic natural and stone materials. Although there are numerous studies on the characteristics of these materials, few address cultural heritage applications directly. Still, current findings suggest significant potential for using geopolymers to consolidate, conserve, and restore historical structures, especially within heritage centres where their low carbon footprint and beneficial properties could greatly support environmental and structural preservation.

5. Future Challenges

Over the past 30 years, research on geopolymers and AAMs has focused initially on understanding the underlying reaction mechanisms, products, and kinetics, and later on testing mix designs for engineering applications. Despite growing interest in AAMs as a low-carbon alternative to traditional concrete, their adoption has yet to be fully streamlined for widespread development. In Australia, examples like Brisbane West Wellcamp Airport (Glasby et al., 2015), the Global Change Institute building, which pioneered the use of geopolymer concrete in precast structural elements (Aldred and Day, 2012; Bligh and Glasby, 2013), Pinkenba Wharf in Brisbane (Manalo et al., 2021), and various VicRoads projects (Aldred et al., 2015; Andrews-phaedonos, 2015; van Deventer, 2015) highlight cases where geopolymer concrete has been used to replace some conventional concrete.

Mix Design Variables

Designing AAM mixes is more complex than for OPC (Ordinary Portland Cement) concrete, due to the larger number of components involved. Careful proportioning is essential to meet performance specifications, as concrete properties can vary significantly with minor adjustments. For example, while an increase in the water-to-cement (w/c) ratio generally reduces strength in traditional concrete, this rule doesn't apply directly to alkali-activated concrete. In AAC, water primarily dissolves alkali metals like NaOH or KOH to create the alkaline solution, which may include multiple alkali sources. Thus, the water-to-cementitious ratio reflects not only water content but also the concentration of alkali metals, which directly affects concrete properties. Another important parameter is the binder content. The ratios of binary or ternary precursor blends can be adjusted as needed, which affects both fresh and hardened concrete properties, whether they are alkali-activated or OPC-based. In OPC concrete, increasing SCMs (supplementary cementitious materials) often reduces early-age strength but maintains or improves later-age strength and durability. For alkali-activated and geopolymer concrete, increasing slag content over fly ash provides higher strength, especially with calcium-rich precursors. However, this comes with trade-offs: increasing slag can significantly reduce the workable time due to a faster setting time, sometimes leading to a flash set. Highly concentrated alkaline solutions can further reduce workable time. To address this, finding a balance between precursor types and the concentration of the alkaline solution is crucial to maintaining workability and strength development (Glasby et al., 2015; Aldred and Day, 2012).

Fresh and Hardened State Parameters

Another significant challenge identified by the authors, and supported by existing literature, is the extensive range of mix-designs possible with AAMs. From variations in precursor materials (PMs) to the types and dosages of alkaline activators, mixes can range from those that take over 24 hours to set to those that experience flash setting in under 10 minutes. This wide variability complicates defining standardized mixes for commercial applications. The authors also note that identical mix formulations can yield different performance results depending on the source of the precursor materials. For instance, ground granulated blast-furnace slag (GGBFS) from one steel mill may behave differently from GGBFS obtained elsewhere, despite having similar primary oxide compositions. Moreover, research shows that mixing for at least 5-10 minutes ensures that low-calcium precursors in binary blends contribute to the mix, whereas shorter mixing times tend to activate the high-calcium precursors prematurely, leading to flash setting. This restricts the feasibility of using volumetric mixers and mobile batching plants for alkali-activated concrete production (Mahmood et al., 2021). Further studies are essential to understand these behavioural inconsistencies and standardize material supply for infrastructure applications.

Regarding setting time, the tendency of AAMs to flash set, especially in high-calcium, slag-dominant mixes, is well-documented (Zivica, 2007). Research has explored adding gypsum as a setting retardant, a common technique in traditional Portland cement to control rapid setting (Chang et al., 2005). A recent study by Hamdan et al. (2023) confirmed that adding gypsum to alkali-activated slag extends setting time by lowering the system's pH, which slows the rate of slag dissolution. However, additional research is required to identify optimal chemical compositions for precursor materials, specifically regarding their amorphous phases and oxide content, to ensure reliable workability for commercial use.

Chemical Formulation

A further hurdle for AAC is the lack of suitable chemical admixtures to manage fresh-state properties, which complicates on-site handling, compaction, and surface finishing. While there are some early efforts in admixture development for alkali-activated concrete (such as MasterCrete® 1000), more advancements are needed, particularly to control the behaviour of AAC in its fresh state.

Alluminosilicate Source Supply

Since AAMs largely rely on industrial by-products and are not manufactured for large-scale supply, careful planning is needed to ensure a sustainable future for this concrete technology, which has been the subject of decades of research. Although there has been substantial research on alkali-activating fly ash, the decreasing availability of fly ash, due to the gradual phasing out of coal-fired power plants in favour of cleaner energy sources, poses a challenge to consistent supply.

Durability

The durability of AAMs has been explored extensively in lab settings. However, most durability tests have been accelerated, raising questions about their accuracy in predicting real-world performance. This lack of long-term, evidence-based data on durability remains a key barrier to the material’s commercial acceptance.

Standardization

AAM standardization efforts are advancing in some countries, particularly those with construction-sector carbon reduction goals. These standards are progressing at an unusually fast pace due to heightened interest in low-carbon materials and significant research efforts. For example, the Concrete Institute of Australia recently held a National Roadshow Series on Low-Carbon Binder Concrete, featuring the new Standards Australia TS 199:2023 "Design of Geopolymer and Alkali-Activated Binder Concrete." This specification has been driven by industry demand rather than regulatory mandates and builds on existing state-level guidelines, such as VicRoads' specifications in Victoria, Australia. These cover alkali-activated concrete across various sections, including 703 (general concrete paving), 701 (drains and pipes), 705 (drainage pits), and 711 (safety barriers), with potential use in structural applications under Section 610 if needed (Andrews-phaedonos, 2014).

In the USA, ASTM C1157 includes performance-based criteria for AAMs, while China's GB/T 29423-2012 standard supports AAMs for applications requiring chemical resistance (British Standards Institute, 2016). In Europe, the UK's BSI PAS 8820:2016 offers guidelines for alkali-activated material and concrete compliance, and Switzerland has its guidelines for alkali-activated slag-based products (SIA, 2014). Meanwhile, the Commonwealth of Independent States (CIS) nations utilize prescriptive standards developed by the Glukhovsky Institute in (Kiev Kavalerova et al., 2014) [56].

6. Conclusions

This paper provides a comprehensive, updated overview of geopolymers and AAMs, covering their historical development, synthesis, reaction mechanisms and key performance factors. It also addresses challenges and future outlooks, leading to the following conclusions:

– Since the 1930s, extensive research has been dedicated to AAM technology, with particular attention in the past five years on synthesis and engineering properties. Despite its promise as a greener alternative to traditional cement, substantial technological advancements remain limited.

– Researchers have investigated the reaction mechanism for geopolymer and AAM formation, generally involving three stages: monomer polymerization, particle growth, and inter-particle bonding. However, the precise chemical process controlling the setting and hardening remains only partly known, likely due to dependencies on the precursor material and alkaline activator.

– A review of precursor materials for geopolymers and AAMs reveals a variety of sources, including industrial, natural, and agricultural, with about 78% originating from industrial by-products. Criteria for selecting precursors should include cost-effectiveness, environmental impact, and construction applicability.

– Influential factors in geopolymers and AAMs synthesis encompass the chemical composition of the precursor (particularly Ca, Si, and Al content in the amorphous phase), physical properties (mainly fineness), type and concentration of alkaline activators (notably molarity and ratio), water-to-binder ratio, and curing conditions (primarily temperature, humidity, and duration).

– Environmental impacts of AAM mixtures are often evaluated based on 1.0 m^3 of concrete, though compiling inventories poses challenges due to the diversity of material sources. The carbon footprint remains a topic of debate, especially concerning the environmental effects of high activator concentrations, despite the use of by-product precursors.

– While geopolymers and AAMs present a more sustainable option compared to OPC-based mixtures, challenges persist regarding their properties and applications. Future challenges include identifying suitable precursors, developing cost-effective and low-carbon alkaline activators, refining mix designs, comprehending core mechanisms, enhancing material and concrete properties, optimizing construction practices, establishing performance standards, and ensuring sustainability in terms of both environmental and economic impact.

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