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Advancements in the synthesis and processing of alkali-activated materials: Activator and Precursors

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Abstract

Alkali-activated materials (AAMs) have gained attention in recent years as a sustainable alternative to traditional portland cement-based materials. This is due to their lower carbon footprint and ability to utilize waste materials as raw materials. However, the high embodied carbon and limited local supply of the conventional activators and precursors have limited large-scale deployment and acceptance as a sustainable alternative to portland cement-based materials. Nonetheless, the synthesis and processing of AAMs have been actively researched in recent years, leading to various advancements in the field. This review paper summarizes recent advancements in the synthesis of AAMs in terms of alternative activators and precursors. The paper also discusses briefly the impact of some of these advancements on the properties of AAMs, such as strength, durability, and sustainability. It aims to provide an overview of the current state-of-the-art on materials utilized in the synthesis of AAMs and identify the areas for future research. Current challenges associated with the synthesis of AAMs are also discussed.

1. Introduction

The construction industry plays a significant role in our society's economic and infrastructure development. The industry's most used construction material is concrete being the primary cementitious material used for various construction applications. Cementitious materials are also seen in various forms ranging from concrete, mortar, grout, engineered/modified cementitious materials, etc. The primary binder in these materials is portland cement (PC). Hence, such materials made with PC as binder can be referred to as portland cementitious materials (PMs). On the other hand, alternative binders can also be used to produce these materials that can behave similarly to that of PMs. Of such materials of high interest and promising applications are alkali-activated materials (AAMs) which do not contain PC as a binder. However, in contrast to PMs, the large-scale application of AAMs is limited and its wide acceptance is still impeded by various challenges.

Significant research and development have occurred in the area of AAMs as an alternative for portland cement materials (PMs) in the last 20 years. The high interest in AAMs is due to the viability of using these materials as sustainable alternatives to PMs due to the high carbon dioxide emissions associated with the production of PC which is the primary binder in PMs. The production of PC has been linked to about 5% of the world's anthropogenic emissions [1,2] and a corresponding emission of about 0.8 tons of carbon dioxide for every ton of PC produced [3]. The high emission from PC is due to carbon dioxide resulting as a product of the production process and the high energy involved in the production [4,5]. On the other hand, AAMs are made with solid precursors that are primarily by-products from various processes or natural minerals. The most common types of precursors used are fly ash (FA) and ground granulated blast furnace slag (SL) which are by-products from the production of power from coal plants and steel manufacturing, respectively. AAMs are primarily composed of a binder that is made up of a precursor and activator. The resulting binder which can be referred to as alkali-activated binder (AAB) can be mixed with other conventional components of cementitious material to produce mortar, concrete, grout, etc. In contrast to PMs, AAMs undergo a different reaction mechanism which is primarily dependent on the type of precursor and activator [6,7]. The properties of precursor, dosage and type of activator in addition to other components and synthesis conditions have a significant influence on the resulting performance.

The sustainability benefits of AAMs compared to that of PMs are primarily dependent on the components and conditions used for the synthesis of AAMs. The use of AAMs in place of PMs has been found to reduce between 5% to 65% of the carbon footprint when the same strength group is considered [3,8]. Hence, it is critical that AAMs are produced with low-carbon materials at ambient conditions. In addition to the sustainability benefits of AAMs, AAMs

can exhibit performance similar to or higher when compared to that of PMs [9,10]. Compared to PMs, the large-scale implementation of AAMs is still limited particularly due to various reasons such as consistency in source materials and lack of appropriate guidelines for construction applications. Nevertheless, significant advancement has been made in the last 20 years which has propelled more research and developments of AAMs. This paper explores briefly and discusses some of the recent advancements in terms of alternative activators and precursors in order to create more awareness about the viability of using AAMs as alternatives to PMs. It is also anticipated that this paper would gear more research and development advancement in the sourcing of alternative locally available materials that can be utilized in the synthesis of AAMs.

2. Alkali-activated materials

AAM is a big term which has been used broadly to describe various forms of materials other than PMs. However, it is worth noting that AAM is a big umbrella that encompasses various types of materials with different reaction mechanisms primarily based on the chemical composition of the precursors. **Figure 1** shows a good overview of AAMS. The mechanism involved in the synthesis of various forms of AAMs is based on the precursor and activator properties resulting in different reaction products which however could behave similarly macro-structurally. For example, the synthesis of fly ash-based AAMs would primarily result in the generation of N-A-S-H gel [11]. However, such synthesis requires a higher temperature than ambient in order to accelerate the dissolution of the monomers. On the other hand, the presence of calcium in precursors such as slag enables the ability to synthesize such AAMs at ambient conditions due to the formation of C-A-S-H gel [12]. Hence, the chemical composition of the precursors determines the categories of AAMs as this significantly influences the reaction mechanism and products generated.

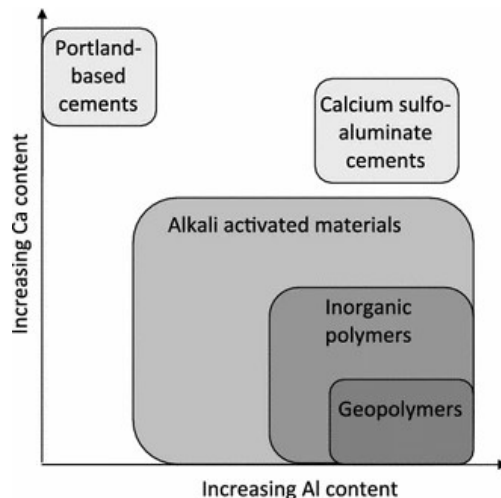


Figure 1 Classification of AAMs and other types of binders [29]

3. AAMs as a sustainable alternative to traditional materials

AAMs can be deemed as a sustainable alternative to PMs depending on their compositions and the synthesis conditions employed. For example, the use of activators and precursors that are processed or produced from an energy-intensive process could eliminate the sustainability benefit of AAMs [13,14]. In addition, the use of elevated heat curing and the use of non-locally sourced materials could also eliminate the sustainability benefits of AAMs due to high-energy usage and the emission associated with the transportation of the materials, respectively. Hence, it is critical to note that in order for AAMs to be used as a sustainable alternative to AAMs; the use of low-carbon precursors and activators in addition to the use of locally sourced material and ambient synthesis conditions is the key.

Nevertheless, as precursors used are primarily waste/by-products of various processes; the use of such materials is sustainable as it offers an effective way to manage wastes provided minimal processing or treatment is required. Similarly, the use of natural minerals with minimal processing and treatment to produce AAMs would be a sustainable

approach for AAMs to be sustainable alternatives to PMs. Generally, AAMs should be treated as a sustainable alternative or supplement to PMs rather than a material that would overhaul or totally replace PMs.

4. AAMs advances

4.1. Precursors

The primary precursors used in the synthesis of AAMs (i.e. fly ash and slag) already have significant applications in the partial replacement of PC in concrete production and clinker in PC production. However, the challenge can come from the availability of these materials that are limited or being limited to specific geographical locations, thus limiting the constant supply. For example, Canada currently has a limited amount of fly ash due to the decommissioning of coal plants. Hence, it is essential that alternative low-carbon materials preferably by-products produced locally in large quantities but not used for other applications in large quantities are used to synthesize AAMs. Some examples of these alternative materials are listed in **Table 1**.

4.1.1. Natural mineral

The natural minerals and by-products utilized as alternative precursors typically usually undergo some form of processing which could be mechanical and/or thermal treatment. The mechanical treatment is generally to reduce the particle size of the materials and corresponding increase their surface area and reactivity. On the other hand, thermal treatment is used to change the mineralogy phases in the materials. A common natural mineral available in large quantities in various parts of the world that can show a promising application in AAMs is clay. However, such clay would need to be processed in terms of mechanical and thermal treatment in order to be suitable as a precursor. Several studies have shown that calcined clay with minimal environmental impact can be produced using various activators and in combination with other precursors [15,16]. However, the variation in the chemical compositions of clay based on geographical locations is still a big challenge.

4.1.2. Processed wastes

In place of the conventional wastes (i.e. slag and fly ash) used as a precursor in AAMs, other processed wastes/by-products as indicated in **Table 1** can be utilized as a precursor in AAMs. Of such promising by-products are mine tailings which are by-products from various mining processes. Studies have shown that the utilization of mine tailings as a precursor in AAMs can be a sustainable approach to managing waste [17,18]. However, these wastes/by-products generally need also need mechanical and/or thermal treatment before they can be utilized as precursors in AAMs. By-products such as red mud are composed of silicate, aluminate and iron compounds which makes them suitable as precursors in AAM synthesis. Some by-products would also need combination with other types of precursors in order to complement the required compounds needed for effective AAM synthesis.

Table 1 Alternative precursor for AAMs

Natural mineral	Clay, Volcanic ash, Pumice
By-products	Mine tailings, Palm oil fuel ash, Glass waste, Ceramic wastes, Sludge ash, Brick wastes

4.2. Activators

Sodium hydroxide (SH) and sodium silicate (SS) are the most used activators in the synthesis of AAMs due to their resulting high performance. However, the corrosiveness, high cost and high embodied carbon [19,20] of these conventional materials (i.e. SH and SS) have limited their large-scale application. Studies have shown that the activators used in the synthesis of AAMs could contribute up to about 60% of the overall embodied carbon of AAMs due to the high-energy intensive process involved in their production [3]. Hence, the activator type and dosage have been found to be the most critical factor that influences the environmental impact of AAMs [21]. The activator could also be the most expensive component of AAMs. However, studies have shown that the use of alternative activators such as silicate sourced from rice husk ash (RHA) could result in about a 45% reduction in the global warming potential (GWP) while a reduction of about 20% in GWP was achieved with the use conventional sodium silicate

when compared to that of the GWP of PMs [22]. Nonetheless, significant progress has been made in recent years to utilize alternative low-carbon activators in the synthesis of AAMs. These alternative activators range from natural minerals to processed wastes and this paper would classify these alternative activators would be classified into two groups as summarized below and **Table 2** presents some examples.

Table 2 Alternative activators for AAMs

Natural mineral	Processed wastes
Trona - Sodium carbonate	Bottom ash
Sodium sulfate	Rice husk ash
	Glass waste
	Silica fume
	Desulphurization dust
	Industrial alkali wastes
	Calcium carbide residue
	Bayer liquor

The majority of the advancement related to the source of alternative activators from processed wastes is associated with the production of a silica source to eliminate the need for sodium silicate. Hence, various wastes with high silica are processed to obtain silica that can be utilized as an activator in the synthesis of AAMs. Silica is very critical to the synthesis of AAMs as it is essential for the polycondensation reaction and the corresponding products would improve the overall performance of AAMs [23]. Studies have shown that the use of these alternative sources of silica can result in up to a 50% reduction in the embodied carbon of the activator compared to that of sodium silicate [19,22]. As listed in **Table 1**, some of the common alternative sources of silica used as an activator and processed from wastes are silica fume, rice husk ash, glass waste and bottom ash. These processed wastes typically have a silica content of more than 75% as shown in **Figure 2**. It is worth mentioning that these wastes are also generated in large quantities in various parts of the world making the application on a large scale viable as these alternative activators can be produced locally while managing locally generated wastes. The extraction of silica suitable for the synthesis of AAMs from these wastes can be in various forms of extraction methods. These methods could be in form of the thermochemical process [24], fusion process [25] or hydrothermal process [26]. Nevertheless, more research is still anticipated in this area in order to achieve the production of alternative silica with consistent properties using less energy-intensive processes.

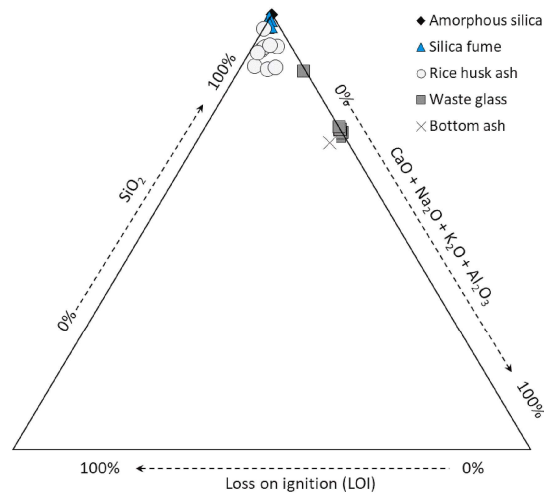


Figure 2 Oxide composition of processed wastes used as silica source (reproduced with permission [30])

Alternative sources of activators have also been explored in the area of using various alkali-based industrial liquid wastes. For example, the study by Fernandez-Jimenez et al. [27] utilized an alkali cleaning solution which is a waste product from aluminum production as an activator alongside fly ash and glass powdered as the precursor. The findings from the study were promising and fly ash activated with the waste cleaning agent exhibited a slightly lower performance compared to when 8M of SH was used as the activator. On the other hand, higher performance was

observed when glass powder was used as the precursor alongside the cleaning agent as the activator[27]. A similar study was carried out by Adesanya et al. [28] where desulphurization dust which is a by-product from the production of steel was used as an alternative to SH as the activator.

5. Challenges and opportunities in the synthesis and processing of AAMs

5.1. Challenges

Despite the recent advancement in the area of AAMs, several challenges still impede its large-scale and commercial deployment. Some of the critical challenges are summarized below:

- **Standardization:** although AAMs can exhibit similar or better properties compared to PMs, the lack of adequate standards or guidelines to support the mixture design and structure design when AAMs are used is one of the main challenges impeding the progress of AAMs. More large-scale research and deployment would provide support for the generation of these guidelines which will correspondingly promote more large-scale application of AAMs for various construction applications. Another way to overcome this challenge is by enabling more collaboration between various stakeholders in the construction industry and moving design standards/guidelines from a performance-based model to a prescriptive-based model.
- **Inconsistent characteristics of precursors:** the properties of activators can be made similar as much as possible. However, the properties of precursors are influenced by various factors which make the chemical and physical properties vary significantly by location and processes involved in the production of the precursors. The variation in the properties of the precursors would result in a corresponding variation in the performance. Hence, it is essential to develop a framework to classify precursors in terms of physical and chemical properties and linked these properties to the corresponding performance.
- **Durability issues:** despite the ease of strength optimization for various developed AAMs, there still persist various durability issues that need to be resolved in order to propel more acceptance of AAMs as alternatives to PMs. Some of these durability issues are in terms of possible leaching of harmful materials, efflorescence, shrinkage and stability of AAMs in various environments among other durability issues. These durability issues can be resolved with more advanced research techniques and the development of effective test methods to evaluate various durability properties in addition to providing effective mitigation strategies for durability issues.
- **Lack of consistent and uniform carbon intensity quantification methods and guidelines:** significant achievements have been made in effectively quantifying the environmental impacts such as carbon intensity of PMs. However, the majority of the quantifications on AAMs are based on general values and are not representative of the approximate carbon intensity. This challenge has resulted in the being able to effectively deem AAM as a sustainable alternative to PMs.

5.3.5.2. Opportunities for further research and development

With the significant advancement that has been made in recent years in the area of AAMs, several opportunity pathways are open for further research and development in this area. Some of the possible research and development prospects are highlighted below:

- **Optimization of mixtures and performance:** the current availability of high-performance computers and technology such as artificial intelligence offers a platform to use these technologies to develop a design and optimization framework based on the numerous available data. These technologies can be used to optimize both the design of AAMs mixtures and the corresponding performance which can be validated by experimental numerical-physical experimental methods.
- **Sourcing of locally available materials:** the key to achieving AAM with lower environmental impact is the use of low-carbon materials that are available locally in various parts of the world. Hence, sourcing alternative materials preferably natural minerals and waste/by-products which are available in large quantities locally and require minimal processing would enable the production of AAMs on large scale. These alternative materials can be used as precursors and/or activators.
- **Durability and long-term performance:** there is a need to develop accelerated test methods to evaluate the durability and long-term performance of AAMs especially those made with non-conventional activators and precursors. This research and development area should also explore the stability of the phases of AAMs in various

conditions and the long term. Such research and development in this area would provide confidence in the stakeholders using these AAMs and also support the development of appropriate guidelines/standards for the design and performance evaluation of AAMs.

- Understanding synthesis mechanism: there is need for a dedicated research to understand the mechanism involved in the synthesis of various types of AAMs based on the composition and synthesis conditions. Such research and development could be carried out in form of comprehensive experimental and modelling evaluations to connect the physiochemical properties of the activators, precursors and synthesis process with the resulting fresh and hardened properties of the AAMs. This is evidently essential for AAMs not produced with conventional precursors (i.e. fly ash and slag) and conventional activators (i.e. SS and SH).
- Comprehensive life cycle assessment: to promote the application of AAMs as a sustainable alternative to PMs, it is critical that a comprehensive life-cycle assessment preferably cradle-to-cradle is carried out in order to effectively and efficiently quantify the environmental impacts of AAMs. Carrying out a life-cycle assessment would also propel more stakeholders' acceptance.
- Development of AAMs for specialized applications: in addition to the development of various forms of AAMs for various construction applications. Future research and development should also explore the development of AAMs for specialized applications such as protective coatings, repair materials, immobilization materials, functional materials, etc. Such specialized applications could be more acceptable by the construction industry and would propel more research and development for general applications.

6. Conclusions

AAMs offer promising alternatives to PMs due to their ability to achieve similar or higher performance and their ability to source the required raw materials locally. However, there is a need for significant advances to be made in the field of AAMs in order for them to become preferred and more sustainable construction materials for various applications. Nevertheless, recent years have seen significant advancements in the research and development of AAMs. Based on the brief discussion in this paper, it is emphasized that the key to large-scale applications of AAMs and the use of AAMs as a sustainable alternative to PMs is through the use of locally sourced activators and precursors. The use of various by-products or waste as either activators or precursors not only offers a sustainable alternative to conventional materials but offers an economical and effective way to efficiently manage these wastes. In addition, the use of these by-products and wastes embodied them with an economic value which could be an alternative source of income for the waste generator.

In general, significant advancement in the area of AAMs is anticipated in the next decade which would result in more large-scale deployment for various construction applications. A critical key to achieving this large-scale deployment would be to source alternative precursors and activators with associated low cost that would produce AAMs with similar or higher performance compared to that of PMs.

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