

Application of Alkali-Activated Sustainable Materials: A Step towards Net Zero Binder

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Abstract: Economic growth and rapid urbanization have resulted in the increase in demand for infrastructure development. To meet this ever increasing demand, conventional construction materials such as concrete are used, which requires an energy intensive process that in turn impacts the environment adversely. Ordinary Portland Cement, being the dominant binder in the industry, contributes around 8% of worldwide annual carbon emissions, and this is expected to reach around 20% by 2050. Population growth has resulted in the significant increase in agro-industrial waste generation during recent years. Inadequate waste management raises a number of environmental concerns. With the growing economy and rising living standards, global raw material consumption is expected to double by 2060. The reutilization of waste materials will aid in their management, while conserving the available resources. Alkali-activated materials (AAM) have recently been introduced as an eco-friendly alternative to conventional binders with fewer environmental impacts. AAM reduce the need for Ordinary Portland Cement (OPC) by substituting it with supplementary cementitious materials (SCM), and therefore, reducing the amount of subsequent carbon emissions. Alkali activation is a complex chemical process between the precursors (alumino-silicate materials) and their dissolution in the activators. Different materials react to alkali activators in different ways depending on their properties. The current study aims to provide a critical review of potential agro-industrial wastes on the fresh and hardened properties of alkali-activated concrete (AAC). To understand the design and development of AAC, influencing the parameters such as the molarity of NaOH, alkali activators, and the ratio of the activators have been discussed in detail. The curing regime and its effect on the behavior of alkali-activated concrete are mentioned. The different admixtures used to regulate the properties of AAC are highlighted. AAC exhibited optimized embodied energy, operational energy, life cycle cost, CO₂ emission, and raw material consumption rates than the conventional concrete did. However, these results varied based on the precursors used in them. This paper focuses on the design and development of AAC, and it should be viewed as an important contribution towards the adoption of AAC in practical applications. The study presents the potential of AAM as a net zero binder in the making of sustainable concrete with enhanced properties.



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1. Introduction

The term “sustainability” has gained global attention in the construction sector as a result of rising carbon emissions [1]. As per the 2022 global status report on buildings and construction, the amount of CO₂ emissions from building operations reached an all-time high at 10 GtCO₂, which is around a 5% increase from that in 2020 [2]. Ordinary Portland Cement, being the dominant binder in the industry [3–5], contributes around 8% of the worldwide annual carbon emissions, and this is expected to reach around 20% by 2050 [6–10]. For every ton of cement that is produced, approximately 0.7 to 1 ton of CO₂ is emitted into the atmosphere [11]. High levels of embodied energy are consumed throughout the production of OPC [12]. In terms of operational and embodied emissions, all of the new and existing assets must be net zero by 2050 as per the Marrakech Partnership

for Global Climate Action (MPGCA) from 2021 [2]. In addition to the carbon emission problem, the environmental challenges associated with increasing waste generation due to rapid population growth are real [13–16]. To address these challenges, researchers in the field are working hard to reduce the demand for OPC [17,18] and increase the use of alternate raw materials [19–22].

The construction industry has already recognized the benefits of using agro-industrial waste as a partial replacement for cement [23–25]. The alkali activation of materials is an emerging approach towards sustainable concrete production [26–29]. This is the most promising solution as it substitutes OPC with agro-industrial waste that contains alumino-silicates [30,31]. Replacing OPC with alkali-activated materials (AAM) has reported to reduce the carbon emissions by 9% [32]. AAMs act as a binding agent, and they are made by activating the source materials containing alumina and silica, which are known as precursors [33]. Alkali activation is a chemical process consisting of precursors and their dissolution in alkali activators. Agro-industrial wastes such as fly ash (FA), silica fume (SF), metakaolin (MK), ground granulated blast furnace slag (GGBS), rice husk ash (RHA), sugarcane bagasse ash (SBA), and palm oil fuel ash (POFA) are the most common precursors [6,16,32]. The most common alkali activators used in the construction industry are sodium hydroxide and sodium silicate [32]. Potassium-based activators have also been used in a few studies, but the dissolution rate of alumina and silica was found to be higher in sodium-based activators [28,34]. Previous studies have shown that alkali-activated concrete (AAC) offered better properties as compared to those of conventional concrete such as an early strength gain, resistance to high temperatures, and sulphate attack resistance. Patel et al. studied the physico-mechanical properties of self-compacting concrete. The effect of GGBS plus RHA on the mechanical properties were observed [35]. Metakaolin- and limestone-powder-based AAC were investigated by Ahmad et al. [36]. The durability properties such as chloride ion penetration, resistance to corrosion, and drying shrinkage of alkali-activated binders containing slag, FA, POFA, and RHA were investigated by Hossain et al. [37]. Different curing methods were attempted by researchers such as ambient environment curing, oven curing, water curing, steam curing, and sealing. Moraes et al. cured alkali-activated sugarcane straw ash (SCSA) specimens at 65 °C for 3 days, and after that, the specimens were kept at room temperature [38]. Aliabdo et al. [39] investigated the influence of varying the curing temperature (30–90 °C) and curing period (1–3 days). The optimal results were observed with a 30 °C temperature curing for 2 days [39].

Previous studies have concluded that the application of alkali-activated materials for making sustainable concrete has resulted in improved properties compared to those of conventional concrete. To provide further insight on alkali activation, this study aims to review the potential SCMs that are used in the alkali activation process and their effects on the properties of concrete. The influencing parameters such as the molarity of NaOH, alkali activators, the ratio of the activators, and the curing regime in order to design and develop AAC are discussed. This paper also includes a discussion about the environmental footprint of AAC in terms of embodied energy and operational energy. This study should be viewed as an important contribution towards the production and adoption of AAC in practical applications.

2. Supplementary Cementitious Materials (SCM)

The research community has focused on finding potential agro-industrial waste material with cementitious properties. Blended concrete has offered improved properties compared to those of conventional concrete [40,41]. Different types of agro-industrial waste were explored in previous studies as a full or partial substitution of the cement (Table 1). V. Charitha et al. [42] studied different ashes for utilization in concrete as an SCM. They reported that the material should contain the oxides SiO_2 , Al_2O_3 , and Fe_2O_3 , and the sum should be more than 70% as per ASTM C618. The chemical composition of the previously studied SCM is presented in Table 2. The inclusion of agro-industrial waste was investigated for its potential to worsen the shrinkage and leaching performances [42]. Jittin et al. [43] examined the influence of RHA and SCBA on concrete. They focused on

the rheological, mechanical, and durability properties of the concrete. The optimal results were observed with 10% SCBA in addition to 5% RHA. The effect of the calcined clay and limestone powder on the micro structure and mechanical properties of concrete were evaluated by Ruan et al. [44]. They found that 50–80% substitution is useful in densifying the microstructure and improving the toughness, but the compressive strength decreased gradually with the increase in the percentage substitution of cement. Chen et al. [45] carried out an experimental study to evaluate the potential of recycled dolomite powder (DP) waste utilization in the glass-fiber-reinforced mortar. The addition of 5–20% DP improved the long-term strength and durability. Similarly, Banu et al. [46] investigated agro wastes i.e., RHA, de-oiled earth (DOE), and spent bleach earth (SBE) to make binary, ternary, and quaternary concrete. The blended concrete showed great resistance to chloride ions penetration. Chinnu et al. [47] compared the influence of POFA on concrete with FA and slag. The addition of POFA and FA to the concrete resulted in an improved workability, whereas the addition of slag decreased the workability. Less chloride penetration and water absorption occurred for the blended concrete in comparison with the controlled concrete mix. Tong et al. [48] evaluated the potential of bamboo sawdust (BS) and found that the incorporation of BS content from 1% to 7% improved the setting time, however it decreased the workability, compressive strength, and flexural strength as compared to those of the control mix. The porous microstructure is generally observed in agro-waste ashes, which therefore decreases the slump value and increases the water demand [42].

Table 1. Supplementary cementitious materials used in concrete.

Agro-Industrial Waste Materials	Binder	Substitution Percentage	Compressive Strength (MPa)	Remarks	Source
Marble Powder waste (MP) + Calcined Bentonite (CB)	MP + Cement	5–20% Marble Powder waste + 5–20% Calcined Bentonite	40	Water absorption increases with in percentage of marble powder and calcined bentonite, 15–20% MP as a partial substitution of cement, and 15–20% of CB as partial substitution of fine aggregate is feasible	[41]
RHA+SBA	SBA + RHA + Cement	10–30% SBA + 5–15% RHA	67	10% SBA with 5% RHA substitution gave the desired results	[9]
Calcined Clay + Limestone Powder	Calcined Clay + Limestone Powder + Cement	50–80% Calcined Clay + Limestone Powder	54	50–70% substitution is recommended to increase the toughness of cementitious materials, but there was no effect on the compressive strength	[44]
Dolomite powder waste (DP)	DP + Cement	0–20%	56	Flexural strength and compressive strength increased, and water absorption rate and drying shrinkage decreased with 10% substitution of DP	[45]
Waste foundry sand	Cement	0–50%	29.06	20% substitution of foundry sand offered improved strength compared to that of control mix	[49]

Table 1. Cont.

Agro-Industrial Waste Materials	Binder	Substitution Percentage	Compressive Strength (MPa)	Remarks	Source
Crushed water glass aggregate (WGA) + demolished bricks (DB)	Cement	0%, 25%, 50%, and 100%	26	Workability of concrete decreases with increasing amounts of WGA and demolished brick as substitution of coarse aggregate, 50% replacement of WGA and DB achieved desired results for hardened properties	[50]
Ground granulated blast furnace slag (GGBS) + waste foundry sand	GGBS + Cement	5–20% GGBS + 10–40% WFS	58	M30 grade concrete with 15% GGBS as partial substitution for cement plus 30% waste foundry sand as partial substitution for sand gave satisfactory results	[51]
Sugarcane bagasse ash (SBA) + Stone Dust (SD)	SBA+ Cement	10% SBA + 0–50% SD	-	M25 grade concrete prepared with 10% substitution of SBA as cementitious material and 40% substitution of SD as a filler material showed a better resistance to both hydrochloric and sulphuric acids attack	[52]
Sugarcane bagasse ash (SBA)	SBA+ Cement	0–25%	35	Compressive strength improved at 10% substitution of SBA for M25 grade concrete	[53]

Table 2. Chemical composition of SCM.

Element (%)	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	Na ₂ O	K ₂ O	TiO ₂	P ₂ O ₅	SO ₃	LOI	Source
FA	53	30.6	3.8	4.6	1.3	0.5	1.4	1.1	-	-	2.3	[54]
	57.2	28.8	3.67	5.16	1.48	0.08	0.94	-	-	0.1	0.12	[55]
	49.46	34.53	6.34	3.57	0.52	0.42	1.38	1.92	0.4	1.14	6.1	[56]
GGBS	30.8	10.9	0.64	51.8	4.57	0.45	0.36	-	-	0.06	0.22	[55]
	36.5	11.6	0.43	39.46	8.14	0.39	0.68	0.84	0.15	0.73	0.03	[57]
SF	96.81	0.25	0.45	0.16	0.26	0.14	0.28	-	-	0.14	1.3	[58]
SBA	71.4	3.39	3.51	6.74	-	-	0.74	-	-	2.44	-	[9]
	96.7	0.3	1.26	0.57	0.27	0.05	0.45	0.02	0.31	-	-	[59]
	85.79	0.67	0.97	2.94	-	-	4.65	-	-	2.89	-	[60]
RHA	95.6	-	0.24	0.7	-	-	2.666	0.02	0.52	0.15	-	[61]
	70.57	2.48	0.28	5.59	3.05	14.49	1.35	-	-	0.19	0.95	[55]
WBG	72.8	12.2	0.54	0.01	1	13.5	-	-	-	-	-	[55]
MK	64.86	20.56	0.31	1.24	0.22	0.11	0.5	0.37	-	1.01	10.82	[62]
HSA	12.36	3.83	5.34	48.52	4.67	2.17	16.42	-	-	0.4	6.29	[62]
MP	0.51	0.13	0.06	56.24	0.1	0.45	0.01	-	-	0.01	42.49	[41]
CB	63.04	17.15	3.17	1.93	4.12	2.64	1.47	-	-	0.38	-	[41]

Note: WBG = waste bottle glass, WC = waste ceramic, HSA = hazelnut shell ash.

3. Alkali-Activated Materials (AAM)

The activator and the precursor are two essential components of AAMs [63]. The first step towards the new generation binder is understanding the precursors, alkali activators, alkali activation solution, and alkali activation mechanism.

3.1. Precursors

Abhishek et al. [28] mentioned that material containing high alumino-silicate and calcium contents can be used as a precursor in the alkali activation process. GGBS, RHA, silica fume, fly ash, metakaolin, bottom ash, corn cob ash, and sugarcane bagasse ash are widely studied precursors [43,64,65] (Table 3). Mostly GGBS and fly ash or a combination of both of them are used as an alumino-silicate-based precursor for alkali-activated concrete [19,28] (Figures 1 and 2). Marvila et al. [63] reported the two types of precursor, one that is rich in alumino-silicates (without calcium oxide), and a second one that is rich in calcium oxide that may or may not contain aluminum oxide ($\text{Ca}/(\text{Si} + \text{Al})$ ratio higher than one). Nodehi et al. [32] studied the three systems of AAM i.e., high calcium, low calcium (geopolymer), and hybrid (calcium and OPC). Low-calcium-based AAMs generally require oven curing to increase their chemical reactivity, while high-calcium-based AAMs show poor durability properties [32].

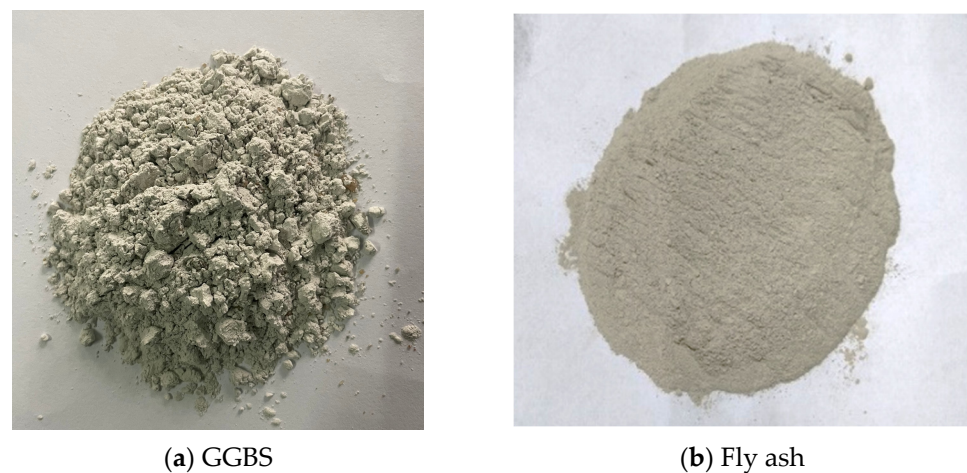


Figure 1. Precursors used in AAC.

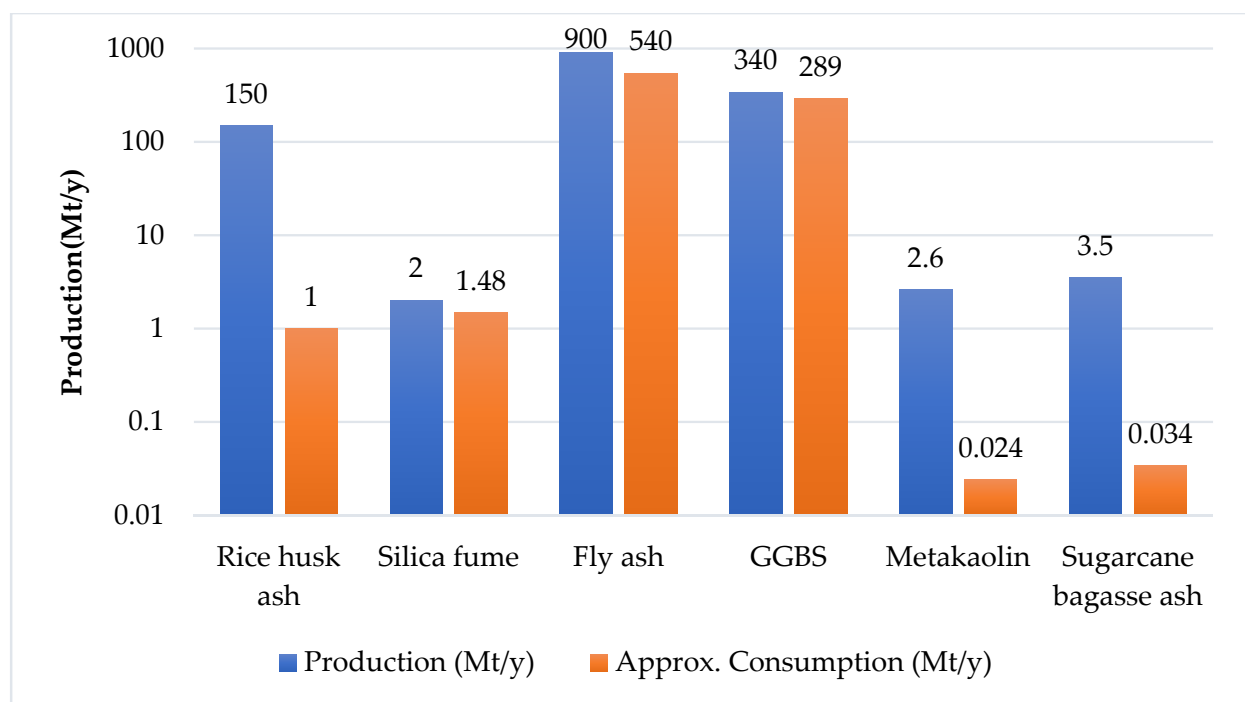


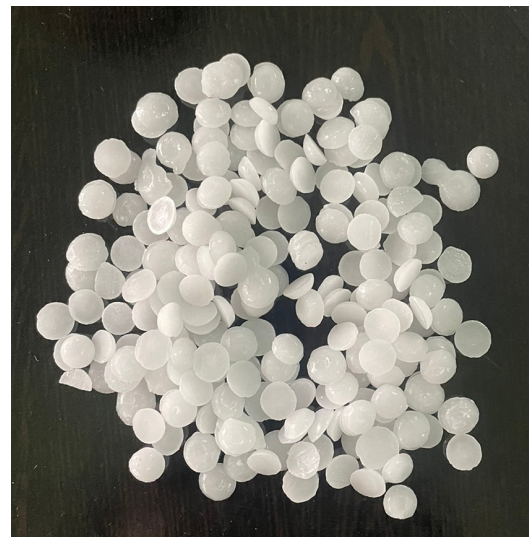
Figure 2. Production and consumption of different precursors [32].

Table 3. Alkali-activated materials.

Precursors	Molarity of NaOH	Na ₂ SiO ₃ /NaOH	Curing Condition	Compressive Strength (MPa)	Tensile Strength (Mpa)	Flexural Strength (Mpa)	Remark	Source
WBG, FA, GBFS, and waste ceramic	2 M–16 M	3	Ambient curing	54	-	-	-	[55]
Blast Furnace Slag (BFS) + RHA	12 M	2	Ambient curing	55	-	6.9	-	[59]
	16 M	2.5	Thermal Curing	58	-	7	-	[66]
	16 M	2.5	-	71	4.8	7.1	-	[66]
GGBS+FA	16 M	2.5	-	71	4.8	7.1	-	[66]
RHA, GGBS, M-Sand, and Copper Slag	8 M	1:2.5	Ambient	31.1	1.95	-	Inclusion of copper slag of up to 40% is beneficial in chloride aggressive environment	[67]
GGBS+FA	14 M	1:2.5	Ambient temperature (27 ± 2 °C)	54	3.8	4.9	Addition of 0.5% of graphene can improve the compressive strength	[68]
FA	8 M–16 M	-	Oven curing	21.5	-	5	Mechanical properties increased with increase in molarity and decreased with higher Na ₂ SiO ₃ /NaOH ratio	[69]
FA	11.5–13.5 M	-	Ambient	92.86	-	-	Strength decreases linearly with the increase in water to alkali activators ratio	[70]
GGBS+SF	13 M	-	-	32.4	-	-	M25 grade concrete prepared with 13 Molar NaOH, 40% Na ₂ SiO ₃	[71]
GGBS+FA (40:60)	8 M	1.5	Ambient temperature (27 ± 2 °C)	60	-	-	-	[72]
FA	14 M	2	23 ± 2 °C and Relative humidity 80 ± 5%	60	-	-	Replaced conventional aggregate by palm oil clinker aggregate	[73]
Metakaolin + Bottom Ash	8 M	2	Ambient curing	58.95	5.96	6.94	-	[74]

3.2. Alkali Activators

Depending on the state that is being used, there are two different sorts of activators: liquid activators and solid activators. Alkali-activated materials can be made using one-part alkali activation (dry powder and water) or two-parts alkali activation (dry powder and liquid activator) methods. There are several activators available on the market such as potassium hydroxide, potassium silicate, sodium hydroxide, sodium silicate, sodium carbonate, and sodium sulfate [75–81] (Figure 3). Sodium metasilicate is the most commonly used solid activator, while sodium hydroxide and sodium silicate are widely used liquid activators (Table 3). Compared to potassium-based activators, sodium-based activators are easily accessible and less expensive. Skin inflammation and other harmful effects have been observed while using potassium hydroxide [32]. The two-part alkali activation is the primary approach used to develop alkali-activated materials, and the majority of research work has been conducted on this only. It can be used for precast work on a large scale, where the chemical handling and temperature for curing can be monitored closely. Researchers are now concentrating on one-part alkali activation, where only the water needs to be added because the transportation part with liquid activators is the major problem. Once the one-part mix method overcomes its limitation, it has the potential for mass production, and it can be distributed as a bagged material.



(a) NaOH Pellets



(b) Liquid Sodium Silicate

Figure 3. Common alkali activators used in AAC.

3.3. Activator Solution

The molarity or concentration of the alkali activator has major influence on the alkali activation process. The relationship between the moles of a solute and the volume of a solution is described by the molarity.

$$M = \frac{\text{Number of moles of solute (n)}}{\text{Volume of the solution (v)}}$$

$$M = \frac{\text{Mol}}{L}$$

The rate of the dissolution of aluminosilicates depends on the concentration of the alkali solution. Previous studies have shown that a higher molarity increases the dissolution rate. However, a high concentration also reduces the strength, which may be due to the congestion of hydroxide ions (OH^-) as a result of the high dissolution rate. Researchers have studied molarity in the range from 4 M to 16 M [82,83], and 8 M–12 M molarity solutions showed the optimal results [39,66,84–86]. When they were using two separate activators to

achieve the desired results, the ratio of the activators was another key influencing factor. Das et al. [87] investigated the influence of molarity (from 6 M to 16 M) and the alkali activators ratio (1.5, 2.5, and 3.5) on ambient cured concrete. The optimum results were observed with 10 M alkali solution and a 2.5 activators ratio. The further increase in the ratio reduced the flowability and setting time of the mixes by increasing their cohesiveness. The rapid hardening of concrete was observed with an increased ratio of the activators.

3.4. Alkali Activation Mechanism

3.4.1. Alkali Activation Mechanism for Low Calcium Precursors

For alkali activation, two essential elements are needed: an activator solution and a solid precursor, which has high silica and alumina contents [20,26,34,88–90]. The alkali activation process is the synthesis of the alumino-silicates in strong alkaline media [19]. The alkali activation reaction was divided into four prime steps by Duxson et al. i.e., dissolving, condensation, polycondensation, and gel crystallization [91] (Figure 4). Marvila et al. [63] further explained the Duxson's alkali activation steps. The dissolution of alumino-silicate materials occurs when the covalent bonds Si-O-Si and Al-O-Al break down in a strong alkaline medium (pH of higher than 14). In other words, a high pH solution breaks the bond between alumino-silicates, resulting in the formation of a colloidal phase [63]. After the dissolution process, the colloidal phase initiates the water elimination process [63,91]. Condensation, a chemical equilibrium process, continues to release water molecules and initiates the gel formation. The gels subsequently undergo a polycondensation process, which increases the number of stable gels that are present, which may or may not lead to crystallization, thereby forming amorphous gels (N-A-S-H) i.e., sodium alumino-silicate hydrate and crystalline or semi-crystalline phases called zeolites [63,92]. Zeolites are made of many interconnected (TO₄: T = Si or Al) tetrahedra that have defined pore sizes at the molecular level [63,93,94]. Zeolites can be formed only under specific pressure and temperature conditions (from 25 °C to 300 °C) [63]. The hardening process of the material starts developing the mechanical properties of alkali-activated material.

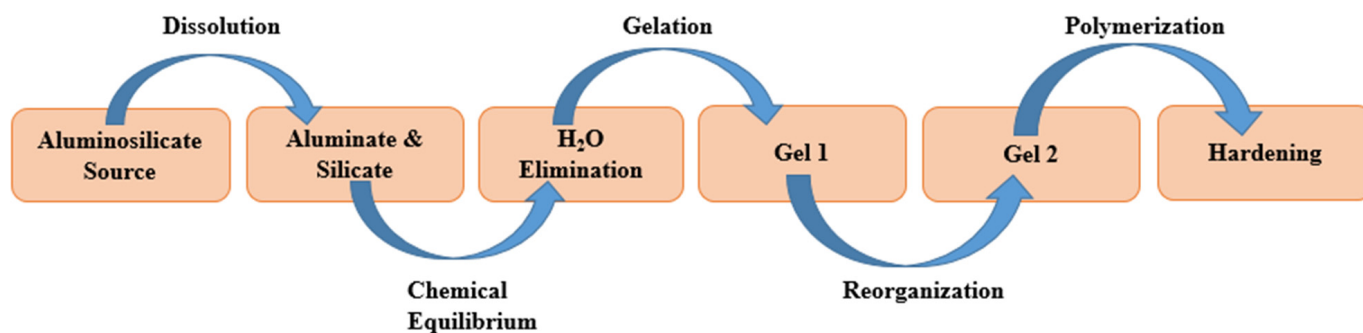


Figure 4. Alkali activation mechanism for low calcium precursors (Duxson's model) [95].

3.4.2. Alkali Activation Mechanism for High Calcium Precursors

The mechanism of alkali activation for high calcium precursors differs from that of the low calcium precursors. They are more reactive at a moderate alkaline pH than the low calcium precursors are, allowing the use of other activators besides sodium or potassium hydroxides and silicates, such as alkali metal carbonate or sulphate solutions [63]. Hydrated calcium silicate gels are predominantly formed as hydration products, which are similar to the hydration products of OPC. The gels have a greater amount of Al present at the tetrahedral locations than Ca, which leads to a higher degree of polymerization. For example, the compound formed in cement hydration is C-S-H, while C-A-S-H gels are formed in the alkali activation of slag [63,96,97]. If excess Al remains present and the material composition contains Mg, then a secondary compound called hydrotalcite is formed. The precursor with low Mg content favors the formation of zeolites over hydrotalcites [63].

4. Properties of Alkali-Activated Materials

4.1. Physico-Mechanical Properties of AAM

The physical and chemical characterization of the raw materials majorly influence the physico-mechanical properties of alkali-activated materials. The molarity of NaOH, the Na_2SiO_3 -to-NaOH ratio, the binder-to-aggregate ratio, the curing method, and the curing period are all important parameters that determine the performance of the AAM [6] (Figures 5 and 6). Aliabdo et al. [39] studied the effect of NaOH molarity (10 M, 12 M, and 14 M) on the properties of alkali-activated GGBS concrete. The increased molarity resulted in the increase in the compressive strength, split tensile strength, and modulus of elasticity, and it had a significant effect on the porosity. Karim et al. [98] carried out the experimental investigation to create zero cement binder using slag, POFA, and RHA with NaOH as an activator. The developed binder was tested for its fresh and hardened properties [98]. The optimal mixture composition consists of 42% slag, 28% POFA, 30% RHA, and 5% NaOH. At 28 days, the zero cement binder had a maximum compressive strength of 40.68 MPa and a flexural strength of 6.57 MPa [98]. Hwang et al. [61] concluded that the sample with the NaOH concentration of 10M and a RHA content of 35% showed the highest strength, however, a further increase in these values decreased the compressive strength.

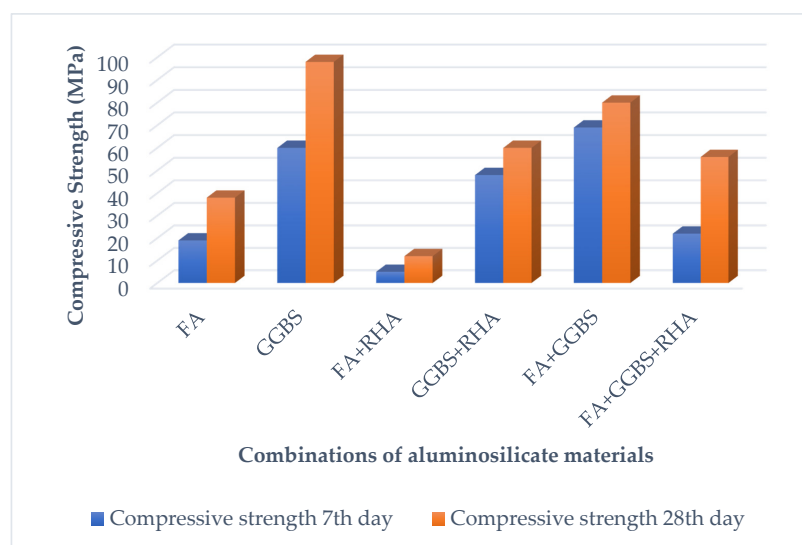


Figure 5. Influence of different precursors on compressive strength of AAC [99].

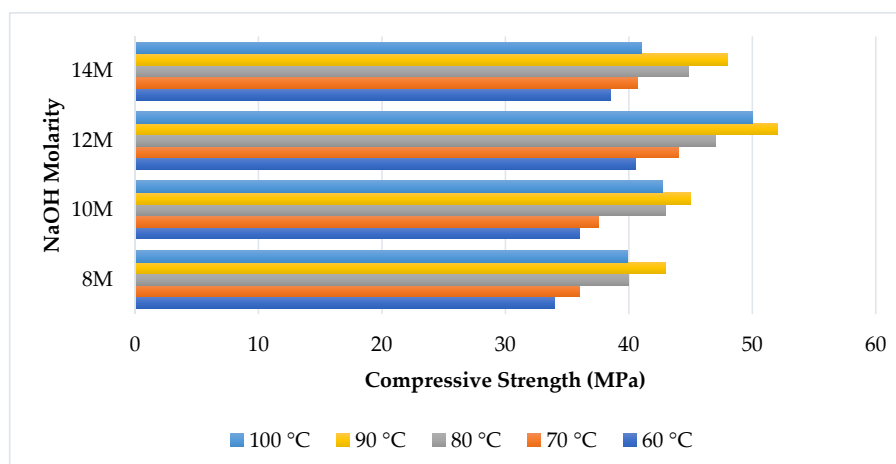


Figure 6. Effect of NaOH molarity and curing temperature on 28th day compressive strength of FA/slag-based AAM [100,101].

The combination of GGBS and FA was investigated by Xie et al. [102], and it was found that the compressive strength increased with an increasing GGBS content. Kishore et al. [103] also mentioned a similar observation while investigating the partial substitution of GGBS by metakaolin. There was a significant reduction of the strength with an increase in the percentage of metakaolin. Even though the amount of GGBS positively affects the compressive strength, a decrease in the GGBS content results in an improved workability and setting time as observed by Hadi et al. [83]. In comparison to OPC mortar, Wu et al. [104] achieved a 70% higher compressive strength for the alkali-activated eco-binder by using circulating fluidized bed co-fired fly ash (CFFA) (30 wt%) and GGBS (70 wt%). The improvement in the strength of the concrete was due to the increase in calcium aluminosilicate gel (C-A-S-H) and sodium aluminosilicate gel (N-A-S-H) formation as a result of an increase in alkalinity. The dissolution of aluminates and silicates increases with increasing molarity, creating a dense and strong structure. Ghafoor et al. [69] prepared the cement-free binder with varying NaOH concentrations (8 M–16 M) and achieved the optimum compressive strength with the 14 M solution. The further increase in the molarity caused a reduction of the strength, which could be due to the congestion of hydroxide ions (OH⁻). Thunuguntla et al. [105] mentioned that the NaOH concentration has the most significant effect on the mechanical strength, followed by alkaline solution/binder ratio. Nagaraj et al. [106] found that the increase in the molarity from 2 M to 12 M increased the compressive strength from 8 to 59 MPa. They achieved the desired results with a 12 M NaOH solution with an alkali activator ratio of two. Sadangi et al. [93], Reddy et al. [70], and Murugesan et al. [80] found that a 10 M NaOH concentration resulted in the highest compressive strength, while Aliabdo et al. [38] and Kumar et al. [49] concluded that a 14 M concentration can give the desired compressive strength. However, Saini et al. [66] concluded that the higher the molarity is, the better the compressive strength is. They observed a similar trend for the split tensile and flexural strengths by obtaining values of 6.398 MPa and 7.875 MPa after 90 days for the 16 M concentration, while Bernardo et al. [107] investigated the alkali activation of volcanic ash from Mt. Etna at a low NaOH molarity (3 M) in order to demonstrate sustainability. Thomas et al. [108] compared the alkali-activated concrete made using FA and GGBS with the conventional OPC concrete and found it to be better in terms of the compressive and tensile strengths.

4.2. Durability Properties of AAM

The durability properties of AAM are mostly influenced by the type of precursor, the activator concentration, and the curing regime. The incorporation of fly ash and a high percentage of slag are observed to improve the durability properties of alkali-activated products. Thunuguntla et al. [105] investigated the influence of the NaOH concentration (1 M–8 M) and the alkali-to-binder ratio (0.4–0.55) on the durability properties of alkali-activated slag concrete (AASC). The novel concrete may offer a high resistance to water absorption as its capillary sorptivity was found out to be low. The mix trials with higher molarity showed a better resistance to acid attacks as compared to the lower molarity ones, which could be due to the high alkalinity and strong porous structure of the concrete. Cai et al. [109] examined the alkali-activated ultra-high strength concrete against water absorption, permeability, and chloride resistance as compared to those of OPC ultra-high strength concrete. They found a lower rate of water absorption and better resistance to chloride diffusion for the alkali-activated concrete than they did for the OPC concrete. Furthermore, they reported that adding silica fume had a negligible effect on the durability properties of the concrete. Zhang et al. [110] mentioned that the substitution of metakaolin in a certain range in alkali-activated slag/metakaolin blended concrete can reduce the water absorption and permeability. Huynh et al. [7] studied the influence of the rice husk ash and fly ash inclusion percentage on the water absorption and porosity of eco-friendly mortar. The water absorption percentage increased with an increasing rice husk ash content (Figure 7). A similar trend was observed for porosity: the highest porosity value was witnessed for the 45% rice husk ash content sample (Figure 8).

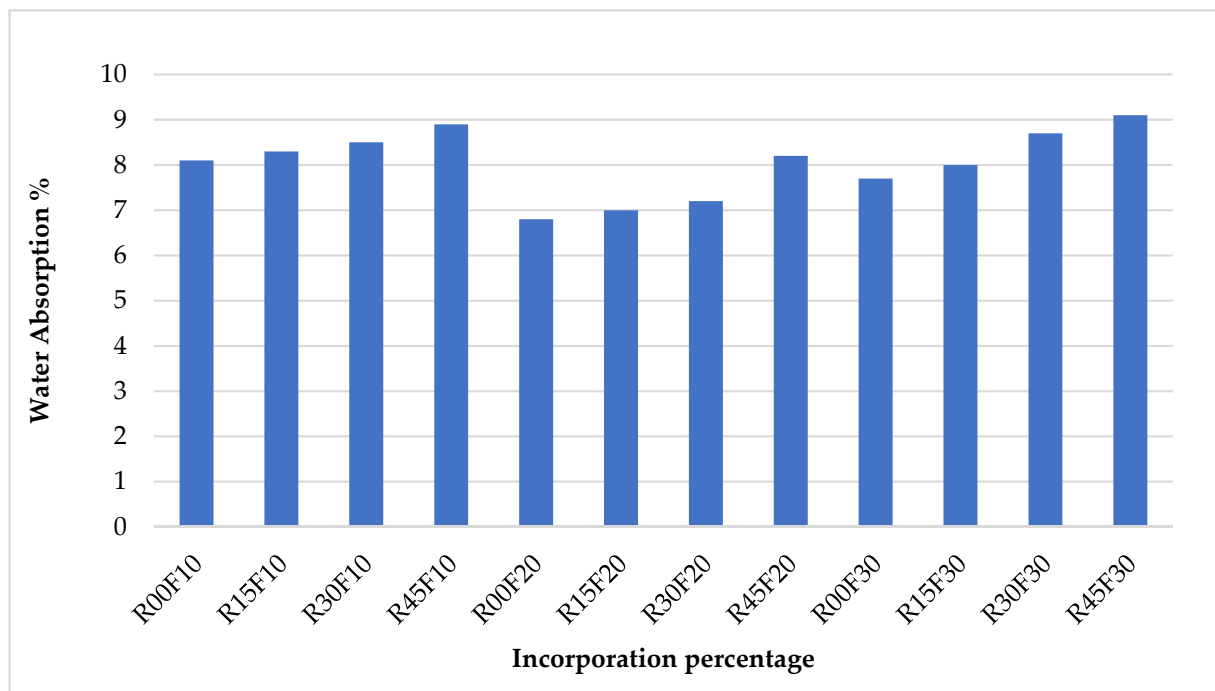


Figure 7. Effect of addition of agro-industrial waste on water absorption (R00F10 refers to 0% rice husk ash and 10% fly ash) [7].

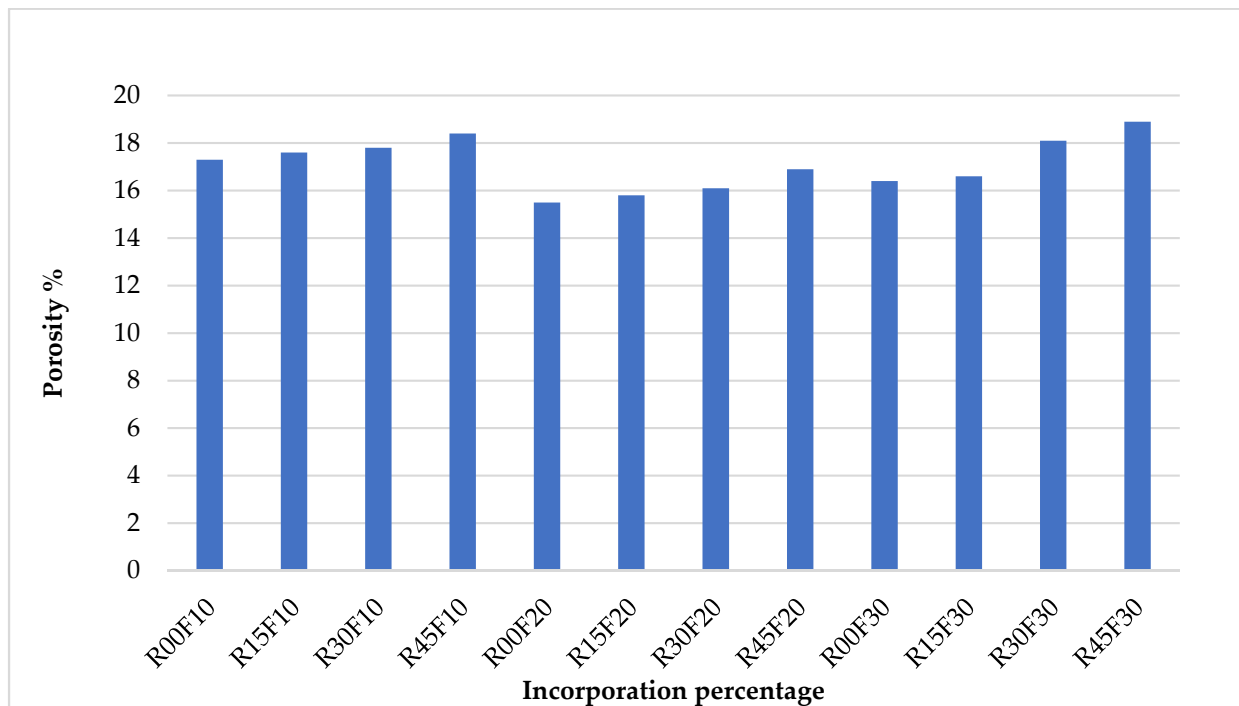


Figure 8. Effect of addition of agro-industrial waste on porosity (R00F10 refers to 0% rice husk and 10% fly ash) [7].

Higher drying shrinkage was observed in AAM as compared to OPC in several studies [111–113]. The Si/Al ratio and gel structure are the two main parameters which have a major influence on the drying shrinkage [95,112]. The effective capillary pressure induced during drying is the primary cause of higher drying shrinkage in AAM [114]. Zhang et al. [115] mentioned that the capillary pressure can be controlled using a polyether

shrinkage-reducing admixture. Previous studies show that the pore structure has a significant impact on the drying shrinkage in concrete [113,114,116]. Mastali et al. [112] reported that alkali-activated slag binder has high value of drying shrinkage, which can be reduced by replacing the slag with other alumino-silicate materials. A similar observation was noted by Hojati et al. [117], whereby a binder containing slag exhibited high time-dependent shrinkage under the internal stress induced by drying. The high visco-elastic/ visco-plastic compliance of alkali-enriched C-A-S-H is the main reason for higher drying shrinkage rates in alkali-activated slag binders [114]. Due to the alkali enrichment, C-A-S-H produced at an ambient temperature is thermodynamically unstable [114]. However, steam curing and high temperature curing can considerably reduce the rate of drying shrinkage [114,117]. The sodium hydroxide concentration can increase or decrease the shrinkage rate in fly-ash-based alkali-activated binders [112]. A lower ratio of alkaline solution to fly ash has resulted in lower drying shrinkages for Chi et al. [118]. Thomas et al. [116] identified that increasing the sodium oxide dose from 4% to 6% reduced the drying shrinkage by 500mm/m. Coppola et al. [113] investigated the combined effect of admixtures on AAM for shrinkage reduction. They used an ethylene-glycol-based shrinkage-reducing admixture (SRA), an calcium-oxide-based expansive agent (EA), methylcellulose (MC), and modified starch (MS) to evaluate the shrinkage behavior of one-part alkali-activated slag-based mortars and pastes. The MC and MS admixtures added to the mix to improve the water retention had no effect on the shrinkage. Both the EA and SRA reduced the shrinkage by 50% and 40%, respectively. Furthermore, the MC, MS, and SRA increased the setting time without affecting the consistency, while the EA reduced the initial workability and setting time [113]. Similarly, Xiao et al. [119] also found that an expanding agent composed of quick lime and anhydrite is useful for reducing the drying shrinkage.

Sulphate attack is a major issue in concrete, which leads to expansion, cracking, and the reduction of the strength of the concrete. Earlier studies have found that fly ash, slag, and metakaolin increase the sulphate attack resistance. Similar findings were reported by Duan et al. [120], wherein the partial replacement of fly ash by metakaolin can reduce the level of damage by sulphate attack. They further explained this phenomenon by noting that the addition of metakaolin leads to the formation of a denser microstructure which exhibits a reduction of the strength loss of concrete when it is exposed to a sulphate solution. Chi et al. [118] reported that alkali-activated slag concrete showed better sulphate attack resistance than conventional concrete. The addition of sugarcane bagasse ash can enhance the sulphate resistance of alkali-activated concrete, as claimed by Jha et al. [53]. Pacheco-Torgal et al. [121] reported that the alkali-activated binder shows high stability under elevated temperatures. Furthermore, they mentioned that alkali-activated binders are susceptible to the development of efflorescence. It can be prevented by adding calcium aluminate admixtures or performing hydrothermal curing treatments [121].

5. Admixtures Used in AAC

The major challenges with the practical application of AAC are high shrinkage rates, poor workability, and rapid setting behaviors [122]. The admixtures or additives developed for OPC concrete do not work in AAC. Raju et al. [122] investigated the effect of the chemical admixtures on AAC and mentioned that the selection of admixture depends on the type of activator solution, the type of binder, and the need for an admixture in concrete. Lignosulphonates (LS), sulphonated naphthalene formaldehyde (SNF), polycarboxylic ether (PCE), and air-entraining admixtures are commonly used in AAC for improving its rheological characteristics [122]. LS, PCE, and vinyl copolymer are effective retarders [122]. According to Tong et al. [123], boric acid and phosphoric acid can be used as retarders for alkali-activated slag-based concrete to enhance the setting time, but they may decrease the compressive strength. Similarly, Makhadmeh et al. [124] found that a glycol ether-based shrinkage-reducing admixture (SRA) is useful in extending the setting time for slag-based alkali-activated binders by retarding the hydration process and the dissolution of ions. Zhang et al. [115] used a polyether SRA and found that it had no effect on the type of

hydration products, but it significantly decreased the rate of hydration production. The extended setting time was initially obtained by them, but it dropped as the SRA content was increased. They further mentioned that an increase in the SRA content increased the mass loss, but it reduced the drying shrinkage, which may be related to the pore structure.

6. Curing Regime

The curing method and curing period have a major impact on the properties of AAM. Alkali activators and precursors react differently to the varying curing temperatures depending on their chemical composition. The curing method can influence the properties of the concrete such as an early strength gain, shrinkage, freeze–thaw cycles, and efflorescence [100]. Elevated temperature curing or ambient temperature curing are generally used for alkali-activated concrete (AAC). Mehrab Nodehi et al. [100] studied the curing methods such as thermal curing, immersion in water, ambient curing, sealing, and microwave curing and their effects on the properties of AAC. They found that water curing and prolonged thermal curing were the least favorable methods for AAC. Water curing causes the activator to be diluted, resulting in an increase in the amount of free water within the air-void pores. Heat curing helps the AAC gain strength early on, but if the heat curing continues during the ageing process, there will be rapid moisture loss, which would worsen the mechanical properties. Gómez-Casero et al. [89] studied the effect of the curing temperature on bottom-ash-based alkali-activated materials. The test specimens were cured at two different temperature values, i.e., 60 °C for 24 h and 20 °C (longer duration). Both of the temperature values resulted in samples with similar mechanical properties.

Aredes et al. [125] investigated the impact of an hour-long oven curing process using alkali-activated metakaolin at temperature of 55 °C, 65 °C, and 85 °C and discovered that the 65 °C-cured samples had the maximum compressive strength. Porosimetry measurements revealed that heat curing increases the open pore volume, while helium pycnometry revealed that the samples cured at 65 °C had the lowest closed pore volume, which may be the cause of the improved compressive strength [125]. Mo Bing-hui et al. [125] studied the effect of the curing temperature within the range from 20 °C to 100 °C for alkali-activated metakaolin using sodium hydroxide and sodium silicate. They observed that the rise in temperature increases the dissolution of metakaolin particles and the precursor reaction. The optimum curing temperature was found out to be 60 °C, with the compressive strength of 97.95 MPa on the 7th day. Higher curing temperature (from 80 °C to 100 °C) showed rapid setting speeds, which restrained the transformation of the specimens into having compact and tough structures. It was observed that the strength values of the oven-cured samples at 80° and 90 °C for a period of 1 day almost doubled as compared to those of the ambient cured specimens on the 28th day [88]. Based on the reviewed literature, it is observed that the elevated temperature for curing improves the properties of AAC, however, Aliabdo et al. [39] reported that increase in temperature from 30 °C to 90 °C decreased the mechanical properties of alkali-activated slag concrete. More water evaporates as a result of the rising temperature. It affects the formation of calcium silicate hydrate (C-S-H), and therefore, it reduces the subsequent compressive strength. AAC is susceptible to the development of efflorescence, but this limitation can be significantly reduced by applying a hydrothermal curing treatment or using calcium aluminate admixtures [121]. Compared to other curing techniques, GGBS resulted in a higher compressive strength when it was cured in an ambient atmosphere (Table 4). If the thermal curing option is not available, the addition of GGBS may be the efficient solution.

Table 4. Impact of curing method on alkali-activated GGBS.

Material	Molarity	Curing Condition	Compressive Strength (MPa)	Source
SCBA + GGBS	6M	Ambient	68.22	[86]
SCBA + GGBS	8M	Ambient	71.21	[86]
SCBA + GGBS	10M	Ambient	76.17	[86]
SCBA + GGBS	6M	Heat- 65 °C (24 h)	48.93	[86]
SCBA + GGBS	8M	Heat- 65 °C (24 h)	61.04	[86]
SCBA + GGBS	10M	Heat- 65 °C (24 h)	61.78	[86]
GGBS + RHA	10M	Ambient	60	[99]
FA + GGBS + RHA	10M	Ambient	56	[99]
FA + GGBS + RHA	-	Steam curing	37.2	[126]

7. Environmental Footprint of AAC Compared to OPC

The building sector consumes around 30% of the total amount of global energy, i.e., 135 EJ, and it accounts for 27% of the global operational related carbon emissions. According to the Marrakech Partnership for Global Climate Action (MPGCA) in 2021, all of the new and existing assets must be net zero over their entire life cycle, including the operational and embodied emissions, by 2050 [2]. Previous literature related to the energy analysis of alkali-activated materials has shown a significant reduction of both the amount of embodied and operational energy [18,31,106,127,128]. The alkali-activated bricks by Gavali et al. [31] resulted in a reduction of the amount of embodied energy, operational energy, and the cost by 21%, 17%, and 7% respectively, when they were compared to those of a conventional brick making practice. Alsaman et al. [12] conducted an experimental study to compare the energy and carbon emissions of AAC with OPC concrete. According to their findings, OPC is the primary contributor, accounting for 80% of the energy and 91% of the CO₂ emissions, whereas, the activator solution is the primary contributor in AAC. For the 40 MPa strength concrete, AAC resulted in energy demands and emissions that decreased by 46% and 73%, respectively. Furthermore, they reported that the material used in concrete has a substantial impact on the energy consumption and CO₂ emissions (Table 5). FA- and slag-based AAC use less embodied energy as compared to metakaolin-based AAC [80].

Table 5. Energy and emissions from materials [12].

Materials	Energy (GJ/t)	Emission (t-CO ₂ /t)
OPC	4.53	0.84
FA	0.033	0.004
MK	2.5	0.33
GGBS	0.857	0.052
SF	0.036	0.014
Aggregates	0.083	0.0048

The operating energy and embodied energy for conventional and slag-based AACs were investigated by Gevaudan et al. [129]. A lifecycle assessment was used to calculate the embodied energy, and a whole-building energy simulation was used to calculate the operational energy. With no effect on operational energy performance, the AAC achieved reductions in the material consumption and embodied energy of 44.4% and 66.5%, respectively. Zhang et al. mentioned that the further reduction of operational energy can be achieved with alkali-activated foam concrete [130]. Life Cycle Analysis (LCA) and Life Cycle Cost (LCC) analyses of lightweight AAC have resulted in its decreased carbon footprint, embodied energy usage, and overall costs [131,132]. Alkali activation-related LCA studies have resulted in reduced carbon emissions of between 40% and 80% [30,133,134]. AAC exhibited optimized embodied energy, operational energy, life cycle cost, CO₂ emissions, and raw material consumption values than those of the conventional

concrete. A comparison between AAC and OPC concrete based on prior studies is presented in Table 6.

Table 6. Comparison of AAC and OPC Concrete.

Parameters	Remarks	Source
Setting Time	AAC exhibits lower setting time as compared to that of OPC concrete	[122,123]
Compressive Strength	Higher compressive strength achieved for AAC as compared to that of OPC concrete	[135]
Tensile Strength	Similar strength was achieved as compared to that of OPC concrete	[136]
Flexural Strength	Higher strength was observed as compared to that of OPC	[136]
Durability	AAC was found to be more durable than OPC concrete was (depends on raw materials used)	[137], [121]
Environmental Impact		
CO ₂ Emission	44.7% less emissions than OPC	[135]
Energy	43% less energy consumption than OPC	[12]

8. Discussion

Alkali-activated materials have been discovered to be an effective solution for sustainable development. They meet many requirements such as high strength, low carbon footprint, and high temperature resistance, etc., however, it depends on many factors, especially the molarity of NaOH, the type of precursors, and the curing regime. The compressive strength of AAC was observed to be similar to or even higher than that for OPC concrete. According to several authors, the maximum strength for AAC was achieved after 3 days of oven curing. The tensile and flexural strengths were satisfactory, thus meeting the desired requirements. Poor workability was observed for AAC, as compared to OPC, especially for slag-based AAC, but it can be improved by using an admixture. Despite the great efforts made in the development of AAC, formulating an admixture that can be used with any activator is necessary to promote the practical application of AAC. The durability properties such as acid attack resistance, high temperature resistance, sulfate attack resistance, and freeze and thaw resistance are good in AAC, except for drying shrinkage. It was found that the drying shrinkage rate is three to four times higher than it is for the OPC concrete, which can be reduced by using shrinkage-reducing admixtures (SRA). Curing regimes are found to be very influential in order to achieve the desired properties. The requirement of an elevated temperature is a major challenge for the onsite production of AAC.

The environmental impact of AAC is lower than that of OPC concrete in terms of waste utilization, carbon footprint, and energy consumption, however, 80–90% of the AAC emissions are from the activators. There is a need for alternative activators that have a lower impact. The available quantity of commonly used activators such as sodium hydroxide and sodium silicates is nowhere mentioned, which raises the issue with completely replacing OPC. Similarly, research on the one-part alkali activation technique is popular, but there has been no discussion on the shelf life of the activators used in two-part alkali activation, as the alkaline solution has to be made within 24 h only.

9. Conclusions

This paper presents a critical review of the supplementary cementitious materials, precursors, alkali activators, alkali activation mechanisms, and their properties. The following conclusions are drawn based on the literature available related to AAM and AAC.

The most common SCM used as a low calcium precursor for alkali activation includes fly ash and metakaolin, whereas GGBS is a high calcium precursor. The combination of fly ash with GGBS is very commonly used in the making of AAC. The two most widely utilized activators are sodium hydroxide and sodium silicate. They were used separately or in combination to achieve greater results. The ratio of sodium hydroxide to sodium silicate

is typically taken to be 1–3. Lower and higher molarities both indicated poor fresh and hardened properties, so the molarity of NaOH should be between 10 M and 12 M for the best results.

The recommended incorporation percentage for agro-industrial by-products was 10–35%. Any further increase in the percentage resulted in an unfavorable effect on the mechanical properties of the alkali-activated concrete. Heat curing is the most suitable method for the alkali-activated material's curing regime. Ambient curing can also result in the required mechanical properties when GGBS is added. Metakaolin has proven to be useful in enhancing the durability properties of alkali-activated materials. Ligno-sulphonates (LS), sulphonated naphthalene formaldehyde (SNF), polycarboxylic ether (PCE), and air-entraining admixtures are commonly used admixtures in AAC.

AAC has a lower impact on the environment, with around 40–80% fewer carbon emissions as compared to those of conventional concrete. Significant reductions in the embodied and operational energy are observed for AAC. The alkali-activated materials have the potential to be used as a net zero binder to replace OPC due to their eco-friendly and enhanced properties.

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