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Evolution of Cementitious Binders: Overview of History, Environmental Impacts, and Emerging Low-Carbon Alternatives

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Abstract

Cementitious binders have long been a keystone of construction, evolving from ancient lime mortars in Neolithic structures to the widespread use of Portland cement in the 19th century, which remains critical in modern construction. This review traces the historical development of cementitious binders and highlights how their widespread adoption has also brought significant environmental challenges, particularly carbon dioxide emissions and intensive energy consumption. To mitigate these impacts, supplementary cementitious materials (SCMs), such as fly ash, slag, and silica fume, have been adopted to reduce clinker consumption and improve sustainability. Despite these advancements, cement continues to be one of the largest industrial contributors to global emissions. In response, alternative binders have been explored. Alkali-activated binders (AABs) demonstrate considerable potential to reduce emissions while offering enhanced durability and performance. These emerging technologies provide a pathway toward more sustainable construction practices. This review is based on a structured survey of the peer-reviewed literature, conference proceedings, and technical reports up to 2025, synthesizing key themes related to historical evolution, environmental impacts, and emerging low-carbon alternatives. The findings aim to inform the development of sustainable building materials for the future.

Keywords: cementitious binder; Portland cement; alkali-activated binders (AABs); environmental impact; supplementary cementitious materials (SCMs)



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1. Introduction

Cementitious binder plays a major role in shaping societies and improving lives through its diverse applications. Its significance lies in its ability to bind materials, forming the backbone of construction projects. The research is dynamic, reflecting its diverse applications and the continuous quest for enhanced performance [1]. Cementitious binder has been a cornerstone of various civilizations, adapting and evolving with changing needs. To comprehend the evolution of cement, a journey through its historical applications becomes essential. Over time, this essential binder has undergone updates, reflecting advancements in technology, construction methods, and societal needs [2–4]. The dynamic nature of research underscores its constant adaptation to meet the challenges of contemporary con-

struction, infrastructure, and environmental considerations. From ancient structures to modern skyscrapers, cementitious binders have been an integral component, transforming landscapes and improving the quality of life for communities worldwide [5]. Its versatility and enduring impact on architecture and infrastructure make binder a subject of continual exploration and innovation, ensuring its relevance in shaping the future of construction and development [6,7]. Modern construction demands not only the durability of the structures through sustainable materials but also the desirable structural performance under different loading conditions, such as improving the strength, ductility, drift capacity, and seismic performance [8–16].

While previous research, such as Pacheco-Torgal et al. [17] and Pacheco-Torgal et al. [18], provides critical insight on alkali-activated binders (AABs) from a historical and reaction mechanism perspective, this research further expands the performance of AABs to compare them alongside SCMs, LC3, and other emerging alternatives and to integrate discussions of structural performance requirements with environmental and economic trade-offs. Moreover, highlights on digital construction methods, life-cycle frameworks, and policy drivers for the adoption of emerging alternatives are also the original contributions to the literature. The present study has five objectives: (1) to synthesize the historical development of binders from Neolithic lime mortars to modern Portland cement; (2) to provide an up-to-date, evidence-based accounting of global production and CO₂ emissions through 2025; (3) to critically evaluate the technical and supply constraints of commonly used SCMs; (4) to assess the potential and practical barriers of alkali-activated binders (AABs), including environmental trade-offs; and (5) to identify priority research, standardization, and policy actions needed to enable large-scale, low-carbon adoption. This focus, combining the 2025 literature update with explicit analysis of scalability, LCA gaps, and regulatory barriers, distinguishes our review from other reviews.

2. Historical Development and Production of Cementitious Binders

2.1. Early Binders and Neolithic Innovations

In the monolithic ages, structures comprised stone blocks without cementing aids. Ancient buildings were often constructed using compacted earth shaped into walls or domes, with large stones stacked carefully in the Cyclopean masonry style, and smaller stones inserted to reduce porosity and tighten joints [19–21]. During the Neolithic period, natural binders began to be employed to assemble bricks or stones for floors, walls, and foundations. Archaeological findings indicate that builders experimented with lime-based concretes as early as 7000 B.C. [22,23]. They possessed the knowledge to calcine limestone (CaCO₃) above 825 °C to produce quicklime (CaO), slake it with water to form Ca(OH)₂, and then combine it with aggregates and water to create a durable concrete [24,25]. Such advancements demonstrate an early understanding of chemical transformations and laid the foundation for later construction technologies.

Around 6500 BC, Nabataea traders built the first concrete-like structures in southern Syria and northern Jordan (Figure 1). They developed waterproof mortars using lime mixed with locally available pozzolanic sands and volcanic ash, a practice that enabled durable floors and cisterns [26–30]. Around 3000 BCE, Mesopotamia saw the development of the first fired bricks, produced in ceramic kilns. Although costly due to fuel and labor requirements, these bricks were used where resistance to weathering was critical, such as pavements and upper wall courses [31]. In Egypt, mortars made from impure gypsum and sand were applied in pyramid construction, while Nile mud bricks were commonly used in arid climates [2,6,20,30,32]. Other early examples include terrazzo floors of burnt lime and clay at Göbekli Tepe (~12,000–10,000 BCE) and gypsum plasters at Çatalhöyük (~9000 BCE) [6]. In Galilee, a lime-bound concrete floor from 7000 BCE displayed strength

and durability, while at Lepenski Vir in Serbia, thick quicklime-based floors were discovered, demonstrating the wide geographic use of early lime binders [2,6,33,34].

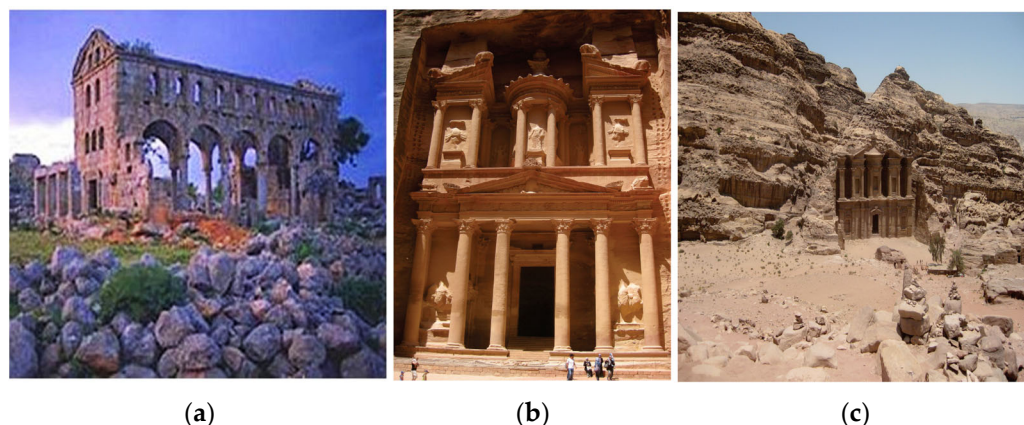


Figure 1. Examples of Nabataean buildings: (a) Kharab Shams, (b) Khasneh al-Fahroun, and (c) Jabal ad-Deir [30,35].

2.2. Roman Advances in Pozzolan Binders

The legacy of Roman construction techniques continued in Western Europe, where construction during this period primarily relied on timber [36]. British Roman culture showed no evidence of lime usage for 400 years after the fall of the Roman Empire. By the eighth century, mortar mills had been discovered in Northamptonshire, but mortar quality fell short due to insufficient kiln temperatures, which prevented the complete burning of the raw materials [2,6]. In the 12th century, technological advancements, such as improved grinding, burning, and sieving, enhanced mortar quality [37]. By the 13th century, mortar comprised non-hydraulic lime, disintegrated easily when exposed to the elements, and had lower bond strength. Post-14th century, significant improvements in mortar were achieved as dirt and clay were washed out of the sand. In the 17th century, pozzolanic trass was commonly added to mortar, marking a development in early modern construction [2,6]. Trass or pozzolana found application in English engineering projects around the 1660s, with major constructions between 1663 and 1683. Several studies aimed to enhance mortar properties. In 1729, Bernard Forest de Belidor published a brief work, followed by a comprehensive four-volume treatise from 1737 to 1753. He recommended a mixture of 12 parts pozzolana or trass, 9 parts quicklime, and 6 parts sand mixed with seawater for mortar preparation. By 1778, in Toulon, France, Barthelemy-Faujas de Saint Fond used pozzolana in mortar mixes [6]. Moropoulou et al. [38] presented thermal and XRD analysis results from ancient Byzantine, post-Byzantine, and later historic mortars from Greece. From the XRD analysis, it was found that the mortars consist mainly of calcite (N 80%) and quartz, whereas the binding material is finely crystallized calcite. The aggregates are mainly calcitic, consisting of microfossils, fossil fragments, and coarse elastic quartz grains. Andreotti et al. [39] analyzed the bonding mortars used to hold the ancient patches in place using a multi-analytical protocol, coupling mineralogical and petrographic investigation (XRPD and OM) along with FT-IR and chromatographic and mass spectrometric techniques to characterize the inorganic and organic components. It was found that medium- and fine-grained calcite crystals are used as aggregates for the inorganic part, whereas egg, beeswax, and Pinaceae resin were used for the organic fractions.

The 18th century marked a significant evolution in cementitious materials [40]. John Smeaton led research into why some types of lime were set underwater and the hardening rate. In 1756, Smeaton received the task of replacing Rudyard's lighthouse, originally constructed with wood. That same year, suitable materials capable of withstanding tides

and moisture were investigated. Contemporary beliefs suggested that the hardest stones produced the finest lime, but it was discovered that the hardest stones did not solidify underwater [41]. The best materials for underwater settings were impure lime, typically containing 82.6% calcium carbonate and 11.2% clay. Lime that failed to set underwater had a naturally occurring clay proportion between 6% and 20% [42]. While working extensively on the Eddystone Lighthouse, as depicted in Figure 2a, lime that could be set underwater was sought. Although cement was not found, the research contributed significantly to its development. Findings published in 1791 directly influenced the advancement of natural and artificial cements [2,24]. Furthermore, some of the bridges made of bricks and stones John Smeaton built are still standing today as shown in Figure 2b,c.

The Romans were pioneers in the use of cementing materials, particularly excelling in the development and application of hydraulic binders, which could set underwater and provided remarkable durability [43]. Building upon this legacy, Reverend Parker invented Roman cement in 1796 [44]. Unlike lime, Parker's cement did not require slaking but had to be ground before use. It exhibited a brown color and typically set within 15 min [34]. The composition of Parker's cement consisted of approximately 55% lime carbonate, 38% alumina, and 7% iron oxide [45], reflecting a careful balance of components to achieve rapid setting and structural strength.

The evolution of cement continued into the 19th century, with traces of Portland cement appearing between 1820 and 1850, conceptually linked to Roman construction practices [46]. Some historical records identify 1824 as the definitive year of Portland cement's invention [1,22,30]. Although there were several attempts to manufacture Portland cement before this, the credit is widely given to Joseph Aspdin (1778–1855) of Leeds, England, who successfully patented "Portland Cement" [1,46,47]. While the Portland cement used today differs in composition and performance from Aspdin's original formulation, it represents a direct lineage from his pioneering work.

By the mid-19th century, roughly 150 years ago, the modern form of cement began to take shape [47]. In 1845, Isaac Johnson advanced the technology by increasing the firing temperature of the chalk-clay mixture to approximately 1400–1500 °C. This higher-temperature process produced a partially fused material known as clinker, which, when ground, became the basis for modern cement [46]. Johnson's innovation significantly enhanced the strength and durability of cement, laying the groundwork for the widespread adoption of Portland cement in industrial-scale construction. Over time, improvements in raw material selection, kiln design, and chemical control have refined cement production, enabling it to meet the structural and environmental demands of modern engineering while preserving its historical roots in Roman and early 19th-century practices.

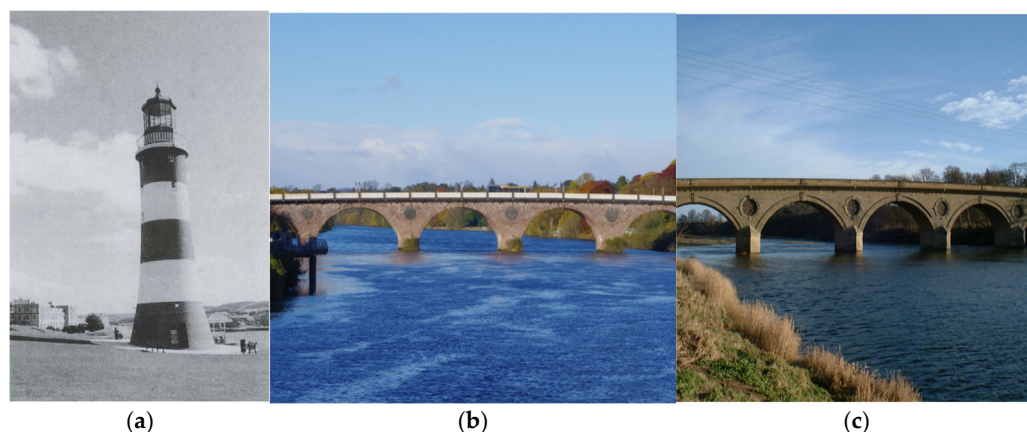


Figure 2. Some structures built by John Smeaton: (a) Eddystone lighthouse tower, UK [31]; (b) Perth Bridge, Scotland [48]; (c) Coldstream Bridge, Scotland [49].

2.3. Industrial Innovations and the Birth of Portland Cement

The late 18th and early 19th centuries marked rapid innovation. Reverend James Parker patented Roman cement in 1796 [43], while Lesage in France (1796), Canvas White in the U.S. (1817), and James Frost in England (1822) developed similar binders. Joseph Aspdin of Leeds patented “Portland Cement” in 1824, produced by calcining a mixture of limestone and clay [1,22,30,46,47,50]. Although his cement differed from the modern material, it represents the origin of today’s Portland cement industry. William Aspdin later discovered that over-burning mixtures improved strength, producing cements superior to both Roman and early Portland varieties [50,51]. Isaac Johnson’s innovation in 1845—firing at 1400–1500 °C to produce true clinker—marks the advent of modern Portland cement, whose composition and durability still underpin global construction [46,47].

2.4. Global Expansion of the Cement Production

By the mid-19th century, cement production spread across Europe. Denmark established its first plant in 1868, followed by Sweden in 1883 and Finland in 1869, with major industrial growth during the early 20th century [34,52–55]. Swedish cement was strongly export-oriented, supplying neighbouring Baltic countries and even contributing to post-earthquake reconstruction in San Francisco [34]. The 20th century saw exponential growth in cement production. Global output increased from modest quantities in the early 1900s to millions of tons by the mid-century [34,56,57]. Packaging also evolved from barrels and hemp bags to standardized paper bags and later bulk transport systems [52]. These technological and industrial expansions solidified cement as a cornerstone of modern construction worldwide but also set the stage for the environmental challenges discussed in later sections. Finally, Figure 3 presents the key milestones in the development of cementitious binders from the ancient age to the modern age.

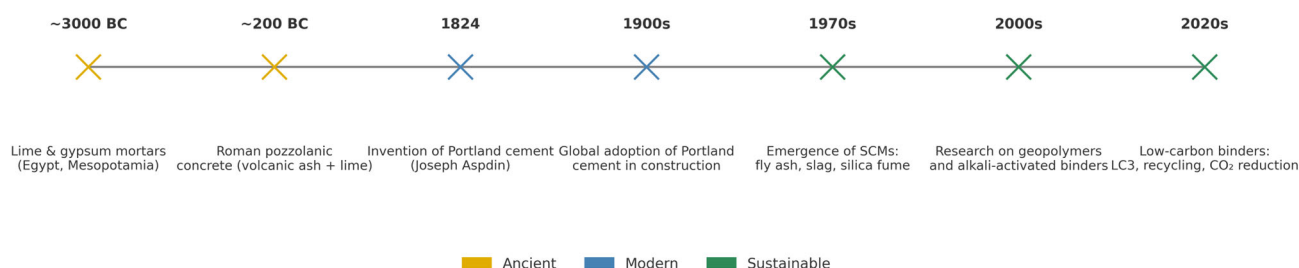


Figure 3. Timelines of the evolution of the cementitious binders.

3. Environmental Challenges of Cement Manufacturing

3.1. CO₂ Emissions and Climate Impact

While contributing to development and economic growth, the cement industry caused significant environmental damage. Some studies indicated that cement manufacturing contributed from about 7% to 8% of global carbon dioxide emissions [58–60]. Cement production releases approximately 1 ton of carbon dioxide for every ton of Portland cement produced [61]. Carbon dioxide alone was responsible for 65% of global warming among all greenhouse gases [62,63]. The production process also involved extensive energy use and posed additional challenges, including severe dust pollution that persisted in the atmosphere, creating critical conditions [52,64]. Moreover, the dust partially obstructed sunlight [65]. The CO₂ emissions increase with each kilogram of cement produced to meet the growing demand for cement. Neither cement manufacturing plants nor countries have significantly reduced these emissions [66]. In the early decades, this contributed to environmental harm. Carbon dioxide is a major contributor to the greenhouse effect and is efficiently released by the cement industry. Records indicate that the cement indus-

try contributes from approximately 5% to 7% of global carbon dioxide emissions [67,68]. The major cement-producing countries are responsible for 63% of global carbon dioxide emissions [69,70].

In manufacturing cement, 1 ton of carbon dioxide was produced for each ton of cement until 2001 [64,71]. By 2010, the carbon dioxide emissions had decreased to 0.87 tons per ton of cement [72]. The total carbon dioxide emissions from cement production amounted to 1.56 billion metric tons of carbon dioxide in 2023, which is double that of 2000 [73]. Such a large amount of carbon dioxide emissions throughout cement production is detrimental to the environment and the ecosystem. Over the past 200 years, the atmospheric concentration of carbon dioxide has risen from 280 ppm to 368 ppm [74]. Such a rapid increase in concentration suggests significant human influence. This change contributes to human-induced climate change. According to the scientific community, if current levels of carbon dioxide emissions persist, global temperatures could rise by 1.4 °C to 5.8 °C over the next 100 years [64,75]. Given these environmental challenges, the cement industry faces moral responsibility and global pressure to reduce carbon dioxide emissions.

As the challenges increase, the cement industry tries to cut carbon dioxide emissions through technological advancements and by adding supplementary cementitious materials (SCMs). As a result, clinker consumption was also slashed, which is also in environmental conservation. Some SCMs are fly ash, ground granulated blast furnace slag, silica fume, rice husk ash, palm oil fuel ash, bagasse ash, wood waste ash, bamboo leaf ash, and corn cob ash [67]. SCMs are considered waste and dumped directly into landfills. Utilization of these materials as SCMs will also lead to the consumption of industrial and agricultural waste. It also helps to reduce carbon dioxide emissions, as these materials will reduce the demand for clinker for cement production.

3.2. Energy Demand and Resource Consumption

Cement is an adhesive material capable of binding particles or pieces of solid matter together [6,76]. As people began to adopt cement for construction, it gradually became an essential component. Over time, it evolved into an exclusively recognized binder, with Portland cement penetrating the commercial market. Cement soon became a commonly used construction material due to its key property of being easily molded into various structural forms. Additionally, concrete gained widespread acceptance because of its physical and mechanical properties. Worldwide concrete consumption is listed second, followed by water [62]. Energy consumption for producing ordinary Portland cement (OPC), followed closely behind the steel and aluminum industries [65,77]. Concrete was important in infrastructure and building projects globally, primarily supplied by OPC [78]. In recent years, annual cement production increased from 1 billion tons to 3.6 billion tons [79,80], with projections to reach 5.8 billion tons by 2050, establishing the cement industry as a high-growth sector over this period [79,81]. This rising demand in the cement industry correlated with a population boom, as the global population was expected to reach 9 billion by 2050 (Table 1) [40,53]. On average, one cubic meter of concrete was consumed per person annually [82]. The cement industry accounted for 5% of the total energy used by all industries combined [62].

The global cement production was estimated at around 4.1 billion metric tons [83], with predictions indicating it could rise to 18 billion tons by 2050 [67]. The popularity of cement concrete is attributed to its favorable mechanical behavior, ease of design, relatively low cost, high serviceability, durability, and minimal chemical reactivity [84]. These advantageous properties have driven a surge in demand. This increase in cement consumption consequently leads to a substantial rise in energy demand. Today, energy needs are met primarily by electricity, whereas in the past, coal was the main source [85,86]. Considering

the environmental impact and energy implications due to the rise of cement production, there is growing interest in exploring sustainable alternatives to substitute ordinary Portland cement partially through supplementary cementitious materials (SCMs).

Table 1. CO₂ emissions and energy-use metrics for cement production.

Emission	Value/Statement	References
Global cement production (2023)	≈4.1 billion metric tons (2023)	[87]
Total CO ₂ emissions from cement production (2023)	1.56 billion metric tons CO ₂ (2023)—stated as double that of 2000	
CO ₂ intensity (per ton cement)	~1.0 t CO ₂ /t cement (until 2001); 0.87 t CO ₂ /t cement (by 2010)	[72]
Share of global CO ₂ emissions (industry)	Two ranges reported: 7–8% in some studies; ≈5–7% in others → overall 5–8%	[58–60]
Energy consumption (global/industrial share)	~2% of total global energy; ~5% of industrial energy consumption; also reported as 5% of total industrial energy	[62,69]
Historical/projected production trend (by 2050)	Production rose ~1.0 → ~3.6 billion t; projected up to ~5.8 billion t by 2050; another estimate cites 18 billion t by 2050	[80]

Different studies report a range for the cement sector's share of global CO₂ because of methodological differences. Key reasons are: (a) differing system boundaries—'cradle-to-gate' (manufacturing emissions only) vs. 'cradle-to-grave' (including use and end-of-life); (b) treatment of upstream fuel and electricity emissions; (c) year of reporting and rapidly changing production patterns; and (d) data source differences (national inventories vs. industry databases). For transparency, we present individual source values and their boundaries in Table 1; synthesizing these yields a conservative range of 5–8% for the sector's share of global anthropogenic CO₂ in the recent literature.

3.3. Sources of Emissions in Cement Manufacture

Cement production is associated with substantial CO₂ emissions, which originate from both chemical and energy-related sources. The largest share arises from the calcination process, in which limestone (CaCO₃) is decomposed into lime (CaO) at high temperatures, releasing CO₂ as a by-product. This process-related emission is unavoidable and typically represents the single greatest contributor to the sector's carbon footprint [64,66].

A second major source is fuel combustion for generating the high kiln temperatures (≈1400–1500 °C) required to produce clinker. Historically, coal has been the dominant fuel, but in some regions, natural gas, petroleum coke, and alternative fuels such as biomass or waste-derived fuels are increasingly used. The choice of fuel significantly influences both direct CO₂ emissions and overall energy efficiency [71,72].

In addition to these, electricity consumption and ancillary processes, including the grinding of raw materials and clinker, operation of fans and conveyors, and transportation of raw and finished products, account for a smaller but non-negligible share of emissions. Although proportionally less significant than calcination and fuel combustion, these electricity-related emissions gain importance in the context of decarbonization, as strategies such as process electrification and renewable energy integration directly target this share [76].

Understanding these distinct emission sources is crucial because each requires different mitigation strategies. Process emissions from calcination can only be reduced through clinker substitution with SCMs, novel low-lime binders, or through carbon capture and storage (CCS). Emissions from fuel combustion may be mitigated by improving kiln efficiency, adopting low-carbon or renewable fuels, and enhancing waste heat recovery [79]. Finally, electricity-related emissions can be addressed through energy efficiency measures

and the integration of renewable power sources. Together, these insights provide a more granular perspective on the drivers of cement's carbon footprint and the technological pathways available for its decarbonization.

Based on the life-cycle analysis, it was found that Portland cement produces around 800 kg CO₂ per ton of binder [88]. Research carried out by Yang et al. [89] showed that supplementary cementitious materials (SCMs), such as GGBS and fly ash can reduce CO₂ emissions by 22% and 14%, respectively. On the other hand, the application of AABs further reduces the CO₂ emissions by as much as 80% as reported by Duxson [90]. Furthermore, life-cycle assessment on various sustainable materials also highlights that the energy demand, durability, and recyclability of concrete with different binders are influenced by the choice of binders. Nonetheless, the sustainability due to the adaptation of SCMs should be properly investigated and justified.

4. Supplementary Cementitious Materials (SCMs)

4.1. SCMs: Definitions, Background, and Historical Development

Supplementary cementitious materials (SCMs) encompass a wide range of materials commonly incorporated into concrete alongside Portland cement. They contribute to concrete properties through pozzolanic or hydraulic behaviors [86]. SCMs can be blended with Portland cement before packaging or used as partial replacements in concrete production [91]. The adoption of SCMs has increased globally, reducing the average clinker content in cement from 85% in 2003 to 77% in 2011, with projections to further decrease to 71% [57,92]. Currently, more than 60% of ready-mix concrete incorporates SCMs [81]. Their use not only reduces carbon dioxide emissions and energy consumption but also enhances concrete strength, durability, and economic efficiency while mitigating land waste and environmental issues [93,94]. SCMs improve mechanical properties through chemical reactions. Hydraulic additives participate directly in the hydration process, while pozzolanic additives react with calcium hydroxide to form additional C-S-H gel, enhancing concrete strength [95]. Additionally, the fine particle size of SCMs allows them to act as fillers, reducing permeability and increasing durability [67,96]. The use of SCMs has a long history, dating back to ancient civilizations. Greeks mixed lime with volcanic ash as early as 600 B.C., a technology later adopted by the Romans around 150 B.C. During their 600 years of dominance, the Romans developed various SCMs, such as Rhenish trass and volcanic ash in Germany and Italy, respectively. Many Roman structures, including the Pantheon in Rome (constructed 120 A.D.), still stand today [2,24,64,97,98]. With the emergence of Portland cement, natural pozzolana was blended with cement, replacing lime-pozzolana binders. Early uses include the Los Angeles waterway (1910–1912) in North America. Experimental combinations of slag and lime began with Loiriot in 1774, and commercial applications emerged in Germany (1865) and later in the Paris metro (1889). Slag–Portland cement blends were used in Germany in 1892 and the United States in 1896, with masonry mortar in the Empire State Building (1930) also incorporating this blend [97]. Commercialization of modern SCMs occurred later. Fly ash, a byproduct of coal-fired power plants, became widely available in the 1930s, with notable use in the Hungry Horse Dam project (1948–1952) in Montana, USA [99]. Silica fume, discovered in 1948, saw its first commercial application in Pennsylvania, USA, in 1983 for Kinzua Dam repair due to its abrasion resistance, and its use in Canadian ready-mix concrete also began in 1983 [100,101]. Later, combinations of slag and silica fume facilitated high-strength concrete production, exemplified by the 68-story Scotia Plaza in Toronto, leading to widespread adoption of high-strength SCM-based concrete [97].

4.2. Research Status of the Emerging SCMs

Cement manufacturing is energy-intensive, consuming about 2% of total global energy and 5% of industrial energy consumption [69]. This energy demand is largely met by carbon-intensive fuels such as coal and its derivatives, making the cement industry a significant emitter of carbon dioxide. In addition to the energy demand, the industry releases substantial amounts of carbon dioxide during clinker production and calcination [102]. Consequently, the cement industry has become a major source of carbon dioxide emissions, drawing global attention to the need for emission reduction. Due to the environmental implications, scientific societies are under immense stress to reduce carbon dioxide emissions. To achieve such a desired target, the industry uses several SCMs, and it brings the change, but the desired output is miles away [103,104]. To reach that landmark, the researchers, apart from the field of Portland cement, sought to develop a new binder that works similarly to Portland cement, and these binders were classified as alkali-activated binders. Purdon [105] first used blast furnace slag with a sodium hydroxide solution, which acts as a binder. As per Purdon [105], the reaction occurs in two parts. Firstly, silica, aluminum and calcium hydroxide liberation occurred, followed by silica and alumina hydrate formation and alkali solution regeneration [17]. These binders are entirely developed from industrial byproducts and do not contribute to carbon dioxide emissions [61].

In addition to the SCMs previously discussed, several other materials exist, but their potential has not been fully explored. So far, these SCMs have primarily been used for experimental purposes in laboratories and have not yet been widely accepted by industry or government bodies. These SCMs include rice husk ash [106–110], palm oil fuel ash [111–117], bagasse ash [118–123], wood waste ash [97,108–113,124–128], bamboo leaf ash [87,127,129,130], corn cob ash [128,131–133], and rock dust [63,72,134]. All the SCMs mentioned above are derived from agricultural waste. These wastes are generated in large quantities, prompting researchers to explore their potential uses. However, since agricultural wastes are naturally occurring and not manufactured, there is a significant possibility that they may be alkali-unstable, with variations in chemical composition depending on their geographic origin [96]. In addition, large-scale utilization is constrained by challenges such as seasonal availability, logistical collection, and competing applications in energy or agriculture. The lack of standardization and quality control further complicates their consistent use in cementitious systems. Moreover, issues such as the presence of unburned carbon, heavy metals, or other impurities raise concerns regarding long-term durability, safety, and environmental performance. Thus, while promising in small-scale research, the scalability and industrial adoption of these agricultural waste-derived SCMs remain limited. Despite the widespread use of SCMs, the cement industry has not been able to significantly reduce overall carbon dioxide emissions or energy consumption in production. While incorporating SCMs and implementing technological changes have lowered the energy demand and carbon dioxide emissions per kilogram of cement produced, the continuous rise in total cement production has negated these improvements. Therefore, it is essential to identify entirely new alternatives with much lower energy demands. Since the aforementioned SCMs alone cannot sufficiently curb cement's carbon footprint or have only a limited impact, researchers developed alkali-activated binders (AABs) as a sustainable alternative to ordinary Portland cement.

Wei et al. [135] examined the effects of ultra-fine recycled fine powder and thermally activated ultra fine powder on the mechanical properties of recycled fine powder-based binder. Furthermore, Zhao et al. [136] investigated the thermal debinding process of the binder jetting green part using a diffusion-controlled kinetic model. Glass powder and fine powder have been examined as a sustainable SCM [137–140]. The fine glass powder exhibits pozzolanic activity that is suitable as a clinker replacement. Research shows that

the replacement of traditional clinker with fine glass powder of up to 20–30% has a very insignificant effect on their compressive strength or durability performance. However, beyond that, the early-strength of concrete and durability is significantly affected.

Dai et al. [141] proposed waste glass powder (GP) as a high-temperature stabilizer in blended oil well cement pastes. Based on the tests on hydration, microstructure, and mechanical properties, it was reported that the application of GP and silica fume (SF) causes the reduction of the peak heat flow and cumulative heat release of the blended pastes. Hassan et al. [142] reviewed the progress in 3D printed concrete, emphasizing the application of sustainable materials as binders. Huang et al. [143] developed a heat-insulating and heat-resistant cement-based material by including expanded perlite (EP), SiO₂ aerogel (SA), and basalt fiber (BF). Defoaming powder (DP) was also added to improve the pore structure. The authors of [143] examined the effects of various temperatures on the physical, thermal, mechanical, and microstructures of the developed material. Shi et al. [144] examined the influences of recycled carbon fibers on the fracture toughness of the mortar. Test results show that the fracture energy and ductility index of fiber-reinforced mortar are significantly higher than those of conventional mortar. The transition of binders from historical lime- and gypsum-based mortars to Portland cement and other advanced sustainable binders must also be understood in the context of modern structural performance demands. The current construction industry emphasizes that sustainable binders must not only address durability issues of structures from a material perspective but also address key performance criteria of structures, such as improving seismic and lateral load-resisting applications [15,16].

4.3. Limitations and Risks of Using SCMs

Despite environmental and performance benefits, SCMs face challenges: (i) chemical variability of agricultural ashes and coal fly ash, (ii) declining supply in regions with reduced coal power, (iii) potential contaminants (unburnt carbon, heavy metals), (iv) durability trade-offs such as delayed strength gain, and (v) logistical and seasonal supply issues for agricultural residues. These risks emphasize the need for standardized quality protocols, robust life-cycle assessments, and secure supply chains before widescale adoption.

5. Alkali-Activated Binders (AABs)

5.1. AAB Technology and Background

Generally, AABs comprise a concentrated aqueous solution of carbonate, silicate, and alkali hydroxide with solid aluminosilicate powder [6]. These binders were manufactured from industrial waste, such as fly ash and slag [64]. Fly ash, obtained from coal-fired power plants, and slag, derived from the iron and steel industry, formed the primary components. Although AABs represented a potential binder for the future, their use remained limited historically. Despite their long history, AABs have not been extensively implemented in research or commercial applications like Portland cement. Because AABs were produced from industrial waste, they required minimal energy, primarily to alter their physical form, leading to a reduction in energy demand by up to 60% [17] and a 90% reduction in carbon dioxide emissions [61,145]. Historically, the first use of slag in concrete occurred in 1908, initiated by Hermann Passow, and officially recognized by German regulations in 1916. Similarly, the United Kingdom issued standards for blast furnace slag in 1923 [7]. This prompted the scientific community's interest in using slag as a binder.

5.2. Properties and Advantages of AABs

Research and development in alkali-activated binders (AABs) gained momentum during the 1980s and 1990s, evolving into a highly active research area [57]. Table 2 presents

a chronological overview of the evolution of alkali-activated binders, highlighting key historical developments. This table outlines significant milestones, discoveries, and advancements in the field, offering a structured reference for understanding the progression of these binders over time. It aids researchers in tracking the historical development of this important construction material.

Table 2. Research on alkali-activated binder.

References	Year	Significance
Feret [146]	1939	Slags used for cement.
Purdon [105]	1940	Alkali–slag combinations.
Glukhovsky [147]	1959	Theoretical basis and development of alkaline cement.
Glukhovsky et al. [148]	1979	First called “alkaline cements.”
Davidovits [149]	1989	“Geopolymer term.”
Malinowski et al. [22]	1991	Ancient aqueducts characterized.
Forss [150]	1983	F-cement (slag–alkali–superplasticizer).
Barnes et al. [151]	1984	Ancient building materials characterized.
Krivenko [152]	1986	DSc thesis, $R_2O-RO-SiO_2-H_2O$.
Malek et al. [153]	1986	Slag cement-low level radioactive waste forms.
Davidovits [154]	1987	Ancient and modern concretes compared.
Roy and Langton [155]	1989	Ancient concrete analogs.
Talling and Brandstetr [156]	1989	Alkali-activated slag.
Wu et al. [157]	1990	Activation of slag cement.
Roy and Silsbee [158]	1991	Alkali-activated cements: an overview.
Palomo and Glasser [159]	1992	CBC with metakaolin
Roy and Malek [160]	1993	Slag cement.
Glukhovsky [161]	1994	Ancient, modern, and future concrete.
Krivenko [162]	1992	Alkaline cements.
Wang and Scivener [163]	1995	Slag and alkali-activated microstructure.
Shi [164]	1996	Strength, pore structure, and permeability of alkali-activated slag.
Fernández-Jiménez et al. [165]	1997	Studies of alkali-activated slag cements.
Katz [166]	1998	Microstructure of alkali-activated fly ash.
Davidovits [167]	1999	Chemistry of geopolymetric systems, technology.
Roy [59]	1999	Opportunities and challenges of alkali-activated cement.
Palomo et al. [168]	1998	Alkali-activated fly ash is a cement for the future.
Gong and Yang [169]	2000	Alkali-activated red mud–slag cement.
Puertas et al. [170]	2000	Alkali-activated fly ash/slag cement.
Bakharev et al. [171]	2000	Alkali-activated slag concrete.
Palomo and Palacios [172]	2003	Immobilization of hazardous wastes.
Grutzeck et al. [173]	2004	Zeolite formation.
Sun et al. [174]	2006	Sialite technology.
Duxson et al. [175]	2007	Geopolymer technology: the current state of the art.
Hajimohammadi et al. [176]	2008	One-part geopolymer.
Provis and Devente [177]	2009	Geopolymers: structure, processing, properties, and industrial applications.
Haha et al. [94]	2011	Durability of alkali-activated slag and fly ash concrete
Junior et al. [178]	2013	Properties of alkali-activated concrete with varying mix design.
Provis et al. [179]	2015	Geopolymers: structure, properties, and applications.
Liu et al. [180]	2016	Utilization of waste materials in alkali-activated binders.
Provis, J. L. [181]	2018	Development of low-carbon alkali-activated materials.
Khalifa et al. [182]	2020	Advances in the use of metakaolin in alkali-activated binders.
Labianca et al. [134]	2022	Incorporation of sustainable materials in alkali-activated binders.
Chen et al. [183]	2023	Incorporation of sustainable materials in alkali-activated binders.
Manjunath et al. [184]	2024	Incorporation of sustainable materials in alkali-activated binders.
Murali et al. [185]	2024	Incorporation of sustainable materials in alkali-activated binders.
Park et al. [186]	2024	Incorporation of sustainable materials in alkali-activated binders.
Singh et al. [187]	2025	Incorporation of sustainable materials in alkali-activated binders.
Lemougna et al. [188]	2025	Incorporation of sustainable materials in alkali-activated binders.
Amin et al. [189]	2025	Durability study of concrete incorporating sustainable materials in alkali-activated binders.
Amin et al. [190]	2025	Durability study of concrete incorporating sustainable materials in alkali-activated binders.

Table 2. Cont.

References	Year	Significance
Zhang et al. [191]	2025	Incorporation of sustainable materials in alkali-activated binders.
Xiang et al. [192]	2025	Incorporation of sustainable materials in alkali-activated binders.

The first possibility of creating a binder from blast furnace slag, caustic soda, and slaked lime emerged in 1895. Kuehl [193] reviewed the behavior of blast furnace slag with caustic potash. The first alkali-activated binder was registered in 1908, created by the reaction between solid aluminosilicate and an alkali source, although this discovery did not gain much attention. Purdon [105], who used sodium hydroxide to activate blast furnace slag, made significant advancements in alkali-activated binders. Purdon [105] and Feret [146] had also contributed to this field, but their research primarily focused on using slag as a partial replacement for Portland cement. Remarkable progress followed with the work of Glukhovskiy [194], Krivenko [195], and Davidovits [149]. Additionally, between 1979 and 1981, several studies identified the presence of analcime zeolite in the mortar composition of ancient buildings dating back to 7000 B.C.

Purdon [105] found that adding sodium hydroxide to slag cement significantly increased early and later-day strength compared to conventional cement manufactured from clinker. He also found that incorporating sodium hydroxide during concrete mixing offered advantages in minimizing various issues. When solid sodium hydroxide was mixed with slag in powdered form, it absorbed moisture and tended to form carbonate. Purdon [105] concluded that the activated specimen showed low water permeability, minimal heat of hydration, and nominal shrinkage. He cast specimens for compressive and tensile strength evaluations, summarized in Figure 4, and determined that generating sodium hydroxide in situ through the reaction of calcium hydroxide with sodium sulphate or sodium carbonate was the most effective binary activator. Adding sodium thiosulfate to the activator further enhanced strength, but this approach was not pursued due to its high cost. His work did not consider the potential of alkali silicate activators, which Glukhovskiy et al. [148] later explored.

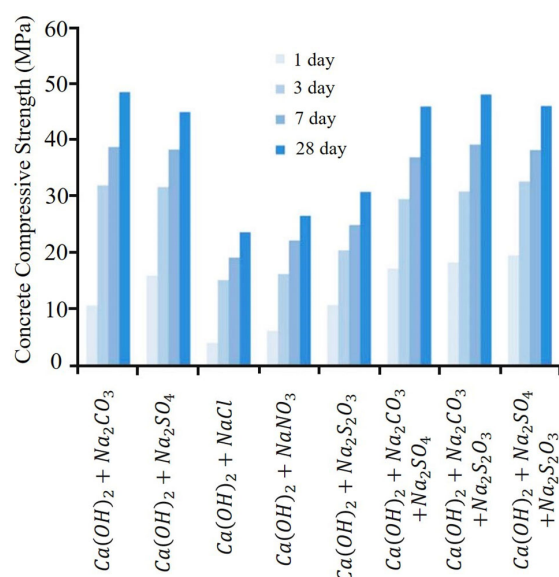


Figure 4. Compressive strength of AABs vs. different activators at different curing days [174].

Glukhovskiy [194] put an effort into the investigation of the binder used by Romans and Egyptians in the construction; from the investigation, he concluded that the binder is composed of aluminosilicate calcium hydrate, which is the same as that of Portland cement

and the crystalline phase of analcite. This also gives the reason for the durability of the Roman and Egyptian binder. Based on the previous research, Glukhovskiy [194] developed a new binder named “soil cement.” The reason behind such a name, Glukhovskiy [194] explained that the binder looks like ground rock and the material has cementitious properties. The binder is prepared with a grounded aluminosilicate blend and rich alkali industrial waste. However, Glukhovskiy et al. [196] made a pivotal fact-finding about the activation of blast furnace slag: (a) hydration compound consists of calcium silicate hydrate, calcium aluminosilicate hydrate, and sodium aluminosilicate hydrate; (b) when clay mineral is incorporated to alkali activation form aluminate silicate hydrate. Glukhovskiy [197] classified the six activator groups: alkalis (MOH), weak acid salts (M_2CO_3 , M_2SO_3 , M_3PO_4 , MF), strong acid salts (M_2SO_4), silicates ($M_2O \cdot nSiO_3$), aluminates ($M_2O \cdot Al_2O_3$), and aluminosilicates ($M_2O \cdot Al_2O_3 \cdot (2-6) SiO_2$).

In 1978, a French scientific investigator, Davidovits et al. [198] coined the term “geopolymer,” which was obtained from the alkali activation of metakaolin. As per Davidovits et al. [198], the binder is similar to Roman and Egyptian binders, with certain modifications and adjustments in the binder. Even Davidovits et al. [198] suggest that the pyramid is not prepared with natural stone and binder; depending on the chemical and mineral study, it was prepared by a mix of limestone sand, sodium carbonate, calcium hydroxide, and water. Davidovits et al. [198] studied the pyramids and concluded that the pyramid stones were not made of calcium. The pyramid stones contain an amorphous aluminosilicate and zeolite-like substances [77]. As per Davidovits et al. [198], the geopolymer is called a polymer because it transforms, becomes hard, and polymerizes at low temperatures, and even at high temperatures, the geopolymers are inorganic, hard, and stable [17]. During the chemical decomposition of geopolymer, it was named “polysialates” in which Sialates denote the compound aluminosilicate oxide, and it comprises of anions $[SiO_4]^{4-}$ and $[AlO_4]^{4-}$ combined with oxygen with much-needed cation (K, Na, Ca) to balance the charge of Al^{3+} in tetrahedral coordinates [199], different types of polysialates in Figure 5. The empirical formula of polysialate is given as follows:

$$M_n \{ -(SiO_2)_z - AlO_2 \}_n \cdot wH_2O$$

where n is the degree of polymerization, z is 1, 2, 3 or > 3 , and M is alkali cation, such as sodium and potassium.

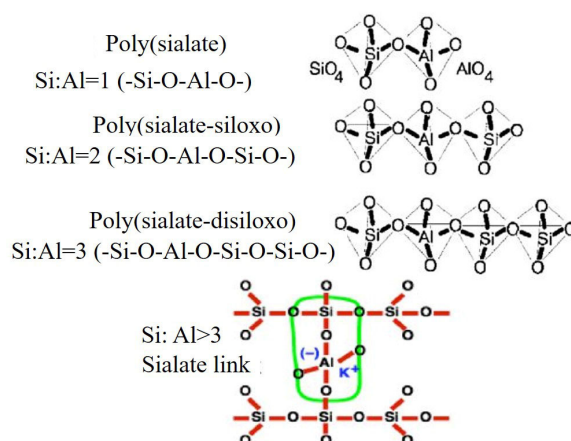


Figure 5. The chemical composition of geopolymer [200].

The simplest way to understand the reaction process of geopolymer is illustrated in Figure 6. The process consists of three stages: the initial one is the dissolution and restructuring of particles, the second is condensation, and the last is polymerization [201].

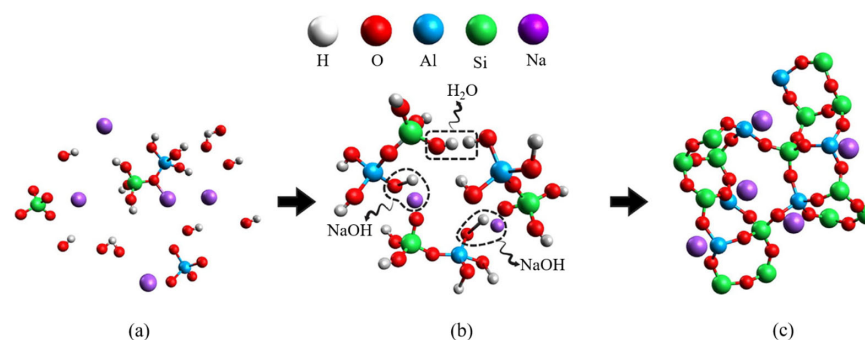


Figure 6. Geopolymerization process: (a) reorganize aluminosilicate, (b) formation of gel from oligomers condensation, and (c) polymerization [201].

Metakaolin, as mentioned above, is a pozzolanic material obtained from kaolin. It is derived from the Chinese term ‘kao-ling,’ which means high hill, and this Kaolin was found near Jachau Fu and mined for centuries. The typical kaolin is shown in Figure 7a. Kaolin was mined for ceramics due to its component Kaolinite, which is known as a hydrous silicate. Kaolinite is structurally composed of alumina octahedral sheets and other sheets of silica tetrahedral attached alternately with a theoretical composition of SiO_2 , Al_2O_3 , and H_2O , which are 46.54%, 39.5%, and 13.96%, respectively, as shown in Figure 7b. Kaolin, after suitable heat treatment, is converted to pozzolana and is named as metakaolin [202].

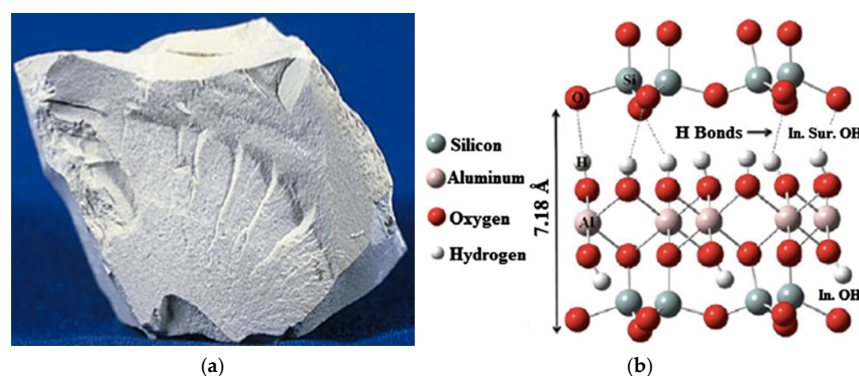


Figure 7. Images of (a) Kaolin [203] and (b) Kaolinite structure [204].

In 1986, Krivenko [162] published his conclusion on the principle of physico-mechanical property and regulation and technology of alkali-activated concrete. Further, Krivenko [162] showed that alkali metal salts and alkalis behave the same as aluminates, silicates, and aluminosilicates while reacting in an aqueous medium, which is alkaline in nature under the condition of high alkali concentration. Such types of occurrences take place with clay minerals, aluminosilicate glasses of artificial and natural, in which calcium compounds are absent, as well as the calcium-based cementitious system in ambient conditions with the water reluctant alkaline or alkaline earth aluminosilicate hydrate, which are the analogues of naturally occurring zeolites and naturally occurring mica. Krivenko [162] divides the binding network into two main groups: (a) $\text{Me}_2\text{O}-\text{Me}_2\text{O}_3-\text{SiO}_2-\text{H}_2\text{O}$; (b) $\text{Me}_2\text{O}-\text{MeO}-\text{Me}_2\text{O}_3-\text{SiO}_2-\text{H}_2\text{O}$. Alkaline aluminosilicate minerals are the possible product of the first one, whereas varied alkali-alkaline earth aluminosilicate and calcium hydroxide and carbonate are the final products [59].

5.3. Reaction Mechanism

The activation of slag is completely dependent on the chemical or activator used. The exact reaction and the mechanism of reactions were not fully explained or understood, although the whole thing depends on the chemical activators and the primary raw material.

The method of activation and reaction mechanism that occurs when the cement is partially replaced by another material. When water is added to the mix, it reacts with slag and then breaks down to form a firm protective layer of Ca^+ , coating the slag particle and slowing further reactions. The reaction will induce three elements when cement is mixed with slag. The first is the cement hydration reaction, followed by the slag hydration reaction, and the third and final one is the slag pozzolanic reaction [205]. Later, further research was carried out on the binder, which contains 100% fly ash and slag used to develop the mortar with chemical activators or alkali activators such as silicate salts, silicate salts, and non-silicate salt of weak acids [194,195,206]. Depending on the chemicals, there are typically two models for activating fly ash or slag. The first activation is through low to mild alkali chemicals, and the second is via a high-alkaline solution. The activation through mild to low alkali chemicals is usually used for slag where the ultimate product is calcium silicate hydrate, similar to the Portland cement [207], with a highly alkaline solution. The material used contains aluminate and silicate as a result. It creates an inorganic binder via the polymerization process [208].

Blast furnace slag contains a decent amount of calcium and little aluminum inside the glassy phase. The hydration of slag via activators includes several steps, which comprise initial destruction to polycondensation of the reaction products [209]. The glassy phase inherently contains high calcium and low aluminum. A study demonstrated the mechanism of splitting of glass containing cations of valencies 1 and 2 as shown in Figure 8 [210]. The primary difference between sodium ions and calcium ions shows much bigger damage due to the removal of divalent cations than monovalent cations from the glass structure [209,211]. It is observed that some glasses exhibit the release of Si and Al. The leaching of the component was discussed to be dependent on the specific glass used and the leaching environment. Krizan et al. [212] analyzed the heat liberation in alkali-activated blast furnace slag release and noticed that higher sodium oxide and silica modulus are related to higher hydration levels. Meanwhile, the initiation begins with the splitting of slag bond Ca-O , Mg-O , Si-O-Si , Al-O-Al , and Al-O-Si . Afterwards, a layer of Si-Al formed over the face of the slag particle, and at last, the final product formed, as shown in Figure 9. Results showed that water glass-activated slag exhibited two heat evolution peaks. Furthermore, it can be observed that increasing the dosages of the water glass increases the combined and accelerated hydration peaks.

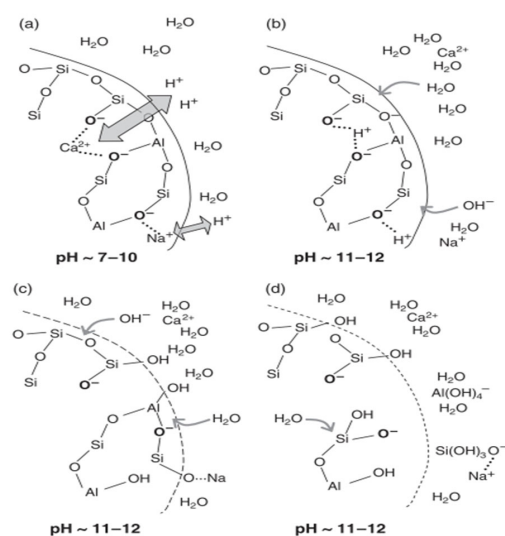


Figure 8. Dissolution mechanism of aluminosilicate glass while premature reaction of single-part geopolymer mix. (a) Interchange of H^+ for Ca^{2+} and Na^+ (b) hydrolysis of Al-O-Si bond, (c) breakage of depolymerized glass network, (d) release of Al and Si [210].

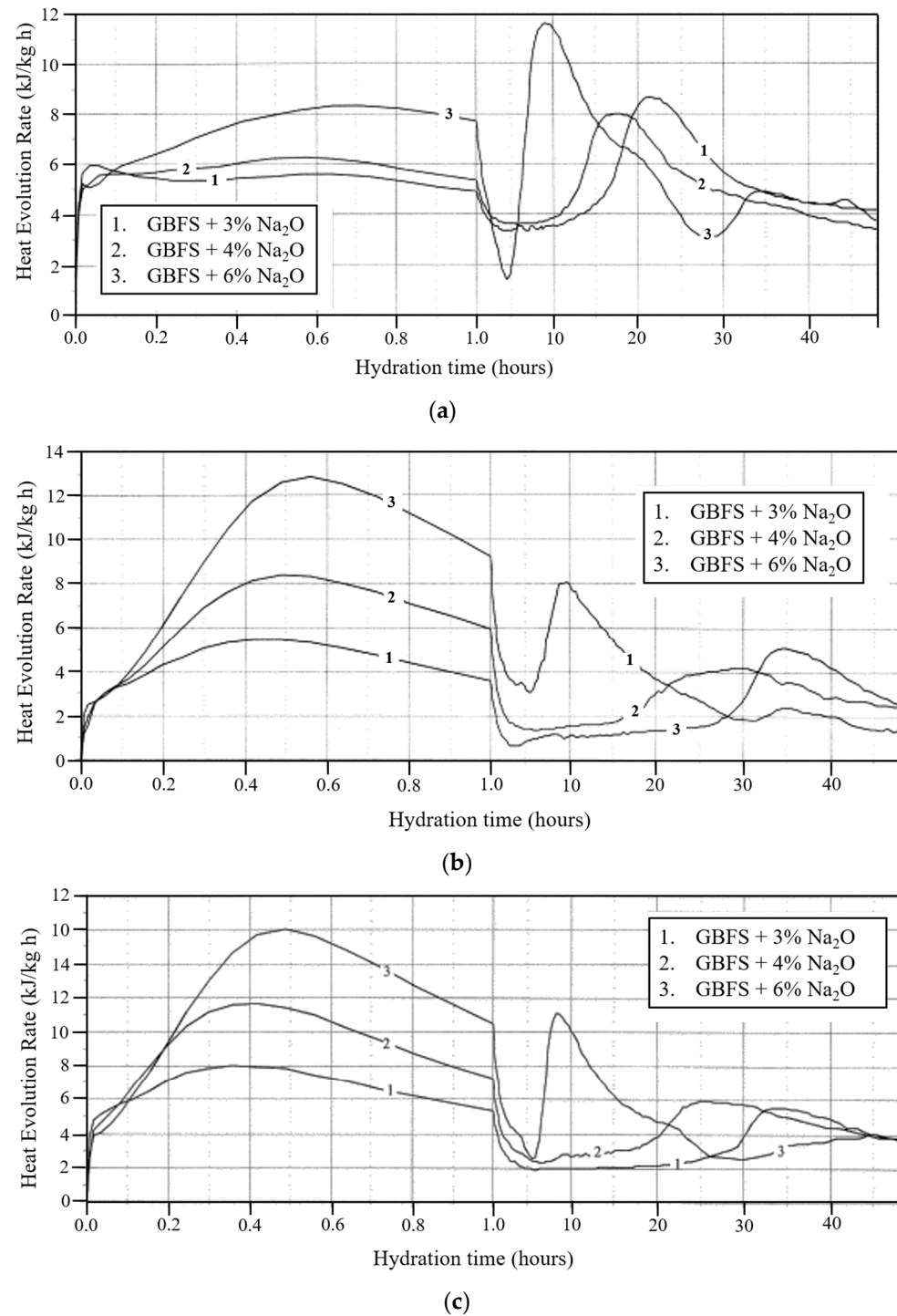


Figure 9. Alkali-activated slag heat evolution, (a) silica modulus (M_s) = 0.6, (b) M_s = 1.2, (c) M_s = 1.5 [212].

Some researchers believe the nucleation mechanism comprises ordering the water bits by the cation formed from the alkali [213,214]. The lower dimension cations are fitter than those in the higher dimension. However, advanced condensation levels have been observed in a less ordered system, as it can be the case that a mix with a higher water proportion and fewer cations is used to command it. As a result, a highly dense and complex structure is formed [213]. Other researchers [215,216] also performed tests to observe the heat of evolution on sodium hydroxide-activated metakaolin, which had several unidentified peaks. The first peak is due to the breaking of metakaolin, followed by orientation time where the release of heat is minimal, and finally, the third peak is when the final struc-

ture is formed, which is influenced by alkali activator concentration [215,216] as shown in Figure 10. However, when the concentration of sodium hydroxide was greater than 10 M, the curve profile for all the samples was very similar. The calorimetric signal for samples with 5 M NaOH was lower in intensity than that of other samples.

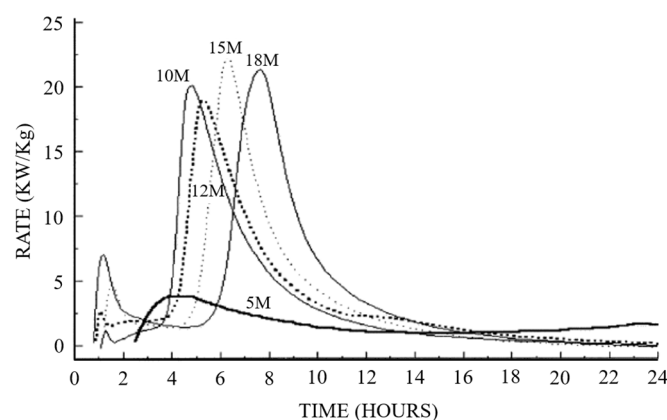


Figure 10. Heat evolution in the sample when activated by sodium hydroxide [215].

Lee et al. [217] find the major difference between ordinary Portland cement and alkali-activated binders. In ordinary Portland cement, the water used for mixing has a neutral pH that gradually turns alkaline as the hydration progresses. In the case of alkali-activated, a strong alkali is required to initiate the reaction. To achieve good chemical, physical, and mechanical properties, it is mandatory to add soluble sodium silicate, as the preliminary pH is high, which prevents the gelling and polymerization of silicate. As the pH drops because of the breaking of prime materials, condensation happens in no time. Then, a collection of reactions of polysialatization, jelling, colloidal establishment, and hardening occurs in the product. The NaOH carries the reaction at a faster pace than sodium water glass. However, later, a similar degree of reaction was observed with the activators. Danse hydrate rims were formed when the NaOH was used. This will lead to a large, coarse, porous structure and a lower-strength structure compared to the reaction with water glass.

From the durability perspective, AABs exhibited improved resistance to sulfate attack, chloride penetration, and freeze–thaw cycles compared with OPC [218,219]. However, the requirement for specific controlled operation and cost concerns associated with alkali activators hinder their large-scale adaptation. Furthermore, the lack of design codes and insufficient data from field validation are some of the additional barriers hindering their applications. Therefore, steps should be taken to overcome the aforementioned issues in order to increase the scalability of AABs and geopolymers.

5.4. Challenges and Practical Barriers for AABs

While AABs offer significant CO₂ savings, they face the following critical barriers:

- Safety and handling concerns with alkaline activators (NaOH, sodium silicate).
- High embodied carbon and cost of some activators, requiring careful LCA accounting.
- Feedstock variability and pre-treatment needs for slags, fly ash, or agricultural residues.
- Limited long-term durability evidence across diverse exposure conditions.
- Absence of standardized codes, test methods, and acceptance criteria.
- Supply-chain and economic uncertainties at an industrial scale.

Addressing these challenges requires coordinated R&D, development of one-part “just-add-water” AAB systems, robust LCA frameworks, and pilot demonstrations to build confidence in performance and scalability.

6. Comparison Between AABs and Portland Cement

6.1. Material Characterization

Ordinary Portland cement and Alkali-activated binders are completely different binding materials with their own property and characteristics. The typical composition of slag used as the binder and ordinary Portland cement is mentioned in Table 3.

Table 3. Typical chemical composition of OPC (%) and slag (%) [100,212].

Chemical Constituents	OPC (%)	Slag (%)
SiO ₂	19.9	37.7
Al ₂ O ₃	4.62	14.4
Fe ₂ O ₃	3.97	1.1
CaO	64.27	37.3
MgO	1.73	8.7
MnO	N. A	0.02
SO ₃	2.56	0.4
LOI	2.9	1.4
Glass Content	-	92

X-ray diffraction (XRD) patterns of Portland cement (PC), AAS, geopolymer alkali-activated slag (GP-AAS), and geopolymer (GP) are presented in presented in Figure 11, respectively (A–D). The Portland cement diffraction peak is centred at 30° on a 2θ scale, and it is only observed in the case of Portland cement. The XRD result shows several peaks due to ettringite, portlandite, quartz, and calcium silicate hydrate. The XRD pattern of alkali-activated slag, geopolymer alkali-activated slag, and geopolymer having a halo diffraction peak at 30°, as shown in Figure 11, is fully amorphous with a lack of long-range zone as visible in the XRD pattern [220]. C-S-H gel was also found in AABs [82,221].

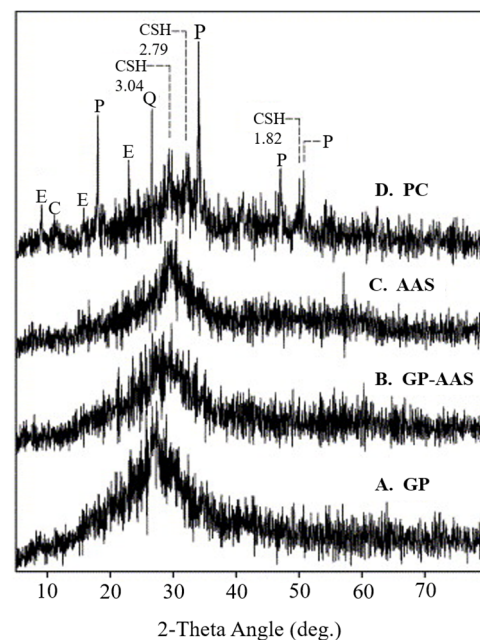


Figure 11. The XRD pattern of (A) geopolymer, (B) geopolymer alkali-activated slag, (C) alkali-activated slag, and (D) Portland cement [220].

Besides XRD infrared analysis of the Portland cement, alkali-activated slag, geopolymer alkali-activated slag, and geopolymer samples were processed. Infrared spectra of all the specimens fall in a similar wavenumber zone. The observed value between the range of 3440 λ and 1650 λ wavelength is due to the stretching and bending of water molecules

to O-H; a similar observation was reported for 1000 λ and 450 λ . All samples other than geopolymer contain carbonate peaks at 1450 λ and 870 λ (Figure 11). At 1450 λ , the peak is higher due to large absorption, and at 870 λ , it is vice versa. Two energy absorption bands at 720 λ and 590 λ appear for the geopolymer and geopolymer alkali-activated slag specimens displayed in Figure 12. The higher one is related to the symmetric stretching of Si-O-Si(Al) bridges, followed by the lower absorption at 590 λ due to the cyclosilicate vibration [220]. In the alkali-activated slag infrared spectrum, energy absorption is around 700 λ due to forming three-membered rings and a minus polymerized structure [222,223]. Finally, the geopolymer and geopolymer alkali-activated slag possess similar infrared spectra; the geopolymer alkali-activated slag has gone through a transition phase between the geopolymer and alkali-activated slag infrared spectra. The alkali-activated slag specimen is slightly different, with an intermediate shape between the Portland and geopolymer specimens. This indicates that the phase of alkali slag is in transition between the structure of geopolymer and Portland cement [220].

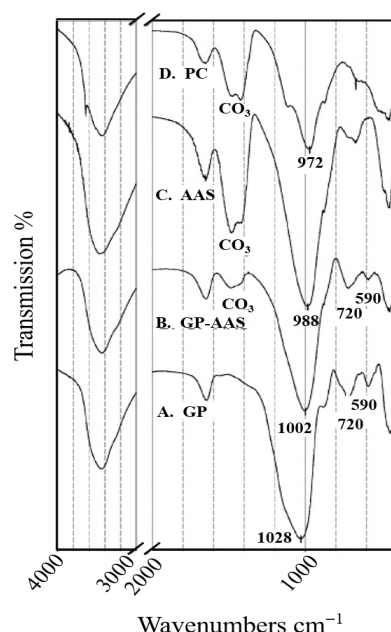


Figure 12. Infrared spectra of (A) geopolymer, (B) geopolymer alkali-activated slag, (C) alkali-activated slag, and (D) Portland cement [220].

6.2. Resistance to Acid Attack

Those who work in alkali-activated binders reported that chemical resistivity is the major advantage over Portland cement. Concrete prepared by conventional Portland cement is usually not resistant to acids. Glukhovsky [197] reported that in the alkali-activated slag mortar specimen, when immersed in an acidic solution of lactic and hydrochloric acid of pH 3, there is an increase in tensile strength as compared to Portland cement. Li et al. [224] studied the durability in similar conditions for 6 months in 5% acidic solution. When the specimens were immersed in the citric acid solution, the change in strength was very low in AABs and a bit higher compared to Portland cement. Similarly, when immersed in the solution of nitric acid and hydrochloric acid, the fall in strength is moderate but still less than that of Portland cement. Severe deuteriation is recorded for sulphuric acid, but AABs are still less deuterated [224].

Davidovits et al. [225] reported mass loss when the AABs and Portland cement specimens were immersed in a 5% concentrate solution of sulphuric and hydrochloric acid for four weeks. In the finding, he concluded that the mass loss of AABs is between 6% and 7%, whereas in the case of Portland cement, it is between 78% and 95%. Palomo et al. [226]

investigated the behavior of metakaolin mixes activated with sodium hydroxide and water glass for different time frames till 90 days when submitted to sodium sulfate pH = 6, sea-water pH = 7, and sulphuric acid pH = 3. There is a minimal decrease in flexural strength for 7 to 28 days; between 28 and 56 days, there is an increase in flexural strength, even from 56 to 90 days. Shi et al. [227] compare the resistivity of different binders in acidic solutions: alkali-activated slag, Portland cement, fly ash binder, and high alumina binder in nitric acid and acetic acid, having pH 3 and 3.5, respectively. As per the report, the mass loss is maximum in case of Portland cement. This happens because of the type of hydration reaction in Portland cement, not porosity, as the alkali-activated slag is less porous, and fly ash and alumina binder are more porous.

Bakharev et al. [228] investigated the comparison of AABs and ordinary Portland cement when exposed to sulphate attack, as the AABs show lower strength reduction [229]. Song et al. [230] work also confirmed the resistivity of AABs when compared to ordinary Portland cement when exposed to 10% concentrated sulphuric acid for the time frame of 8 weeks, as the AABs only lose 3% of mass loss and 35% of strength loss while the ordinary Portland cement loses more. Fernando et al. [231] reported an average mass loss of 2.6% after being subject to sulphuric, hydrochloric, and nitric acids; meanwhile, the loss in the case of ordinary Portland cement is more than twice that.

Bernal et al. [232] studied the performance of alkali-activated slag and Portland cement. The samples used for the test were cured in water for 60 days. The latest specimens were exposed to sulfuric acid, hydrochloric acid, nitric acid with a pH of 3, and acetic acid at a pH of 4.5 for 150 days. After keeping the specimens in the acid environment, the samples were tested on the 30th, 90th, 150th, and 300th day, and the volume of the permeable pore of the specimen was checked at the end of 150 days. No change in compressive strength was recorded when the specimen was dumped into the water for 30 days, but after 90 and 150 days, there was an increase in strength for the alkali-activated slag specimen. Besides water, the compressive strength was identical for slag binder and Portland cement when the specimens were dipped in minerals. After a long exposure to hydrochloric acid and sulphuric acid, there was a slight increase in the mechanical strength of Portland cement. This is because the acid can not damage the core of the specimen and continues to gain its maturity. The interaction of Portland cement with sulphuric acid produces gypsum in its first 30 days. During this tenure, there has been a noticeable increase in the weight of the specimen, which has also been confirmed by different authors [231,232]. Similarly, when the specimen was exposed to hydrochloric acid, calcium-rich salt was produced because of the chemical reaction between CaCl_2 and unreacted C_3A [233]. Portland cement in acetic acid for 90 days recorded a slight gain in strength. Thereafter, at 150 days, a reduction in strength was recorded. For alkali-activated slag, a slight fall in strength was recorded for all the specimens in the initial days. Later, there was a substantial increase in strength thereafter. Alkali-activated slag performs better in sulphuric acid and acetic acid when compared to hydrochloric acid. It was also observed that the increase in strength post-30 days recorded for Portland cement was not similar to alkali-activated slag.

Bakharev [234] studied geopolymer material under different acidic conditions. The samples were prepared using fly ash. The material was tested by 5% sulphuric acid and acetic acid. For reference, the author used a conventional binder. Bakharev [234] noted a regular fall in strength for all the cases. The only difference recorded was the percentage fall in strength. The geopolymer strength falls 4.5% in the first month, 10% in the second month, and 36.3% in the tenure of 6 months in the acetic acid from the same specimen with a sulphuric acid solution. There was a recorded fall of 45% in the first 2 months, but it recovered to 40% in 6 months. A conventional binder with acetic and sulphuric acid also performed a similar test. The conventional binder loses 89% of its strength in the first

2 months; after 6 months, it is around 91%. A similar binder, when tested with sulphuric acid, has a negligible record of strength.

6.3. Alkali-Silica Reaction (ASR)

The chance of being affected by ASR is an unknown subject; however, in the case of ordinary Portland cement, knowledge about ASR is present in previous research. ASR was first introduced by Stanton [235]; as it was concluded that for the reaction, three things are required: (a) amorphous silica in good quantity, (b) alkaline cations, and (c) water Pacheco-Torgal et al. [18]. The ASR begins when the aggregate silica reacts with the cement hydrate's alkaline ions. After the reaction, alkali-silica gel forms, and then the gel attracts the nearby water and tends to expand, which causes cracking [236]. Davidovits [199] compared the AABs with ordinary Portland cement when submitted to ASTM C227 subjected to the mortar bar test, reported shrinkage in the first case and severe expansion in ordinary Portland cement. Fernández-Jiménez et al. [237] also reported the effect of ASR in AABs and OPC, and the expansion is more in the case of OPC. Moreover, Puertas [238] believes that ASR occurs in AABs containing reactive opera aggregate. Bakharev et al. [239] compare the expansion of OPC and AABs, and the case of OPC shows more expansion, which is clearly shown in Figure 13. Where gel accumulated, fewer cracks were observed. However, the cracks were not associated with alkali-silica gel formation but could have resulted from shrinkage.

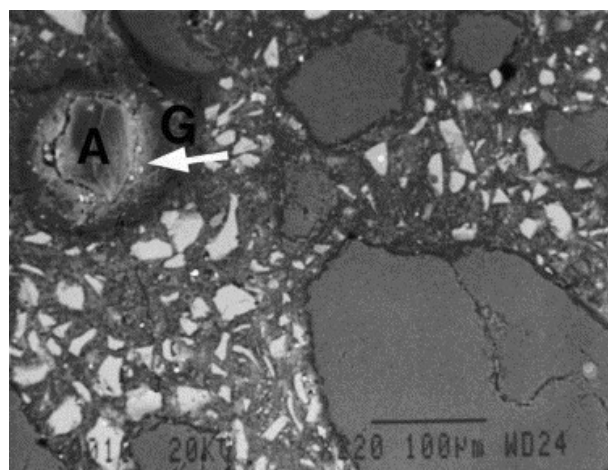


Figure 13. AABs concrete after 300 curing days: (A) reactive aggregate, (G) alkali-silica gel [239].

6.4. Resistance to High Temperature

Concrete prepared from Portland cement shows weak performance when subject to thermal loading above 300 °C. Above 300 °C, it begins to disintegrate, and the specimen prepared with AABs remains stable even at 1000 °C [240–242]. Other authors who study the behavior of metakaolin and shale waste at high temperatures between 600 °C and 1000 °C found minimal loss in strength; however, in a few cases, the strength will rise at 1200 °C [243]. Kong et al. [244] studied the behavior in high temperatures of alkali-activated metakaolin and detected that the Si/Al ratio influences the residual strength later in a thermal phase up to 800 °C. The mixes with a Si/Al ratio between 1.5 and 1.7 show a high residual strength. Krivenko et al. [245] tested the AABs under fire and suggested their use in tunnel and high-rise buildings, whereas the OPC could not match the level of resistance of AABs.

6.5. Strength Development

While comparing ordinary Portland cement and alkali-activated binder, Gruskovnjak et al. [246] concluded that the strength after 28 days is similar. However, the early strength evolution of the two systems is entirely different. The AABs (referred as AAS in that study) concrete was characterized by high initial strength, but after day one, the rate of gaining strength decreased. The strength-gaining phase for the OPC is longer than that for AABs. Based on the calorimetric measurements as shown in Figure 14, it was reported that the heat flow evolution for OPC has a very broad peak between 4 h and 25 h and a maximum from 12 to 17 h, whereas the AABs have a very narrow peak between 13 and 17 h and a maximum of 16 h. On the contrary, the system referred as ‘slag without activator’ exhibited a very low heat flow compared to OPC and AABs.

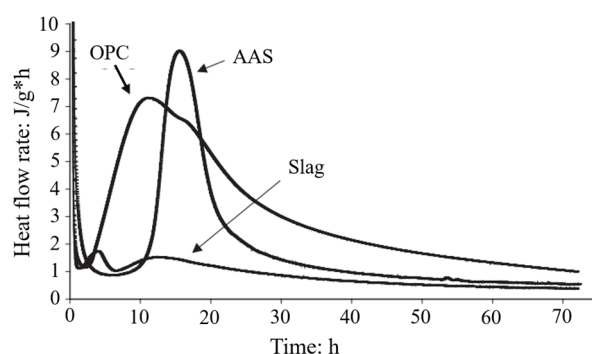


Figure 14. The heat of evolution for AABs (referred as AAS), OPC, and slag without activator [246].

The strength development mechanisms of AABs differ notably from those of Portland cement. While Portland cement gains strength through hydration reactions forming calcium-silicate-hydrate (C-S-H) gels, AABs achieve strength via the polymerization of aluminosilicate precursors activated by alkaline solutions, leading to the formation of binder phases such as sodium-alumino-silicate-hydrate (N-A-S-H) or potassium-alumino-silicate-hydrate (K-A-S-H), depending on the calcium content of the precursors [247]. In practical terms, AABs can exhibit rapid strength gain, achieving high compressive strengths within 24 h, which can be advantageous for applications requiring quick turnaround times. For instance, certain alkali-activated slag mortars have demonstrated a 52–89% increase in compressive strength compared to traditional cement mortars, suggesting their potential to replace conventional binders in various construction scenarios [248]. However, the unique properties of AABs necessitate adjustments in construction practices. The rapid setting times of some AABs formulations may require modifications in handling and placement techniques to ensure proper workability and finish. Additionally, a study focusing on the durability performance of AABs in chloride-bearing environments highlighted their potential in resisting chloride-induced corrosion, which is crucial for reinforced concrete structures [78,249]. A study exploring the mechanical and durability properties of slag-based AABs with varying proportions of granite dust indicated that these materials exhibit satisfactory performance under specific conditions. However, the study also noted that more research is necessary to fully understand their behavior in diverse environmental scenarios [250]. Furthermore, experimental investigations have indicated that AABs maintain their strength and durability properties even after soil burial exposure, suggesting their applicability in geotechnical applications [251].

Collins et al. [252] investigated the mechanical properties of slag-based AABs referred as AAS concrete and OPC concrete for 91 days and found that the strength of AABs concrete is always higher when compared to OPC (Figure 15). The one-day strength of all concrete was very similar. However, AABs concrete exhibited the most rapid strength

development compared to other concretes. Interestingly, when the strength gain of OPC concrete became flat between 56 and 91 days, AABs concrete continues to gain strength.

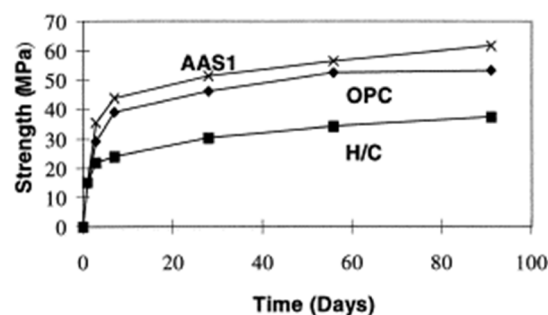


Figure 15. Compressive strength of slag-based AABs (referred as AAS) and OPC concrete [252].

Bernal et al. [253] tested AABs concrete and OPC concrete with different binder content, say 300 kg/m^3 , 400 kg/m^3 , and 500 kg/m^3 , for 91 days. The investigator found that the strength for the day is maximum in the case of AABs concrete. Bernal et al. [254] tested the AAS and OPC concrete for 28 days on 40 kg/m^3 and 120 kg/m^3 . For all the parameters, the AAS concrete has a better result in terms of strength. It is important here to compare the performance of PC and AABS for strength, durability, and environmental impact. Li et al. [255] conducted a comparative study of different OPC and AABs. The slag-based AABs showed higher compressive strength than fly ash-based AABs and OPC. A similar kind of observation is stated by Kumar et al. [56]. The AABS has better performance than OPC concrete [249,256–258].

Wang et al. [259] figured out that most of the work is on strength measurement under different conditions, so the researchers performed a study on factors affecting the strength of alkali-activated slag. The study reported that it was more fundamental and helped understand the factors responsible for strength development. The authors concluded that (a) the type of activators, (b) method through which the activator was introduced, (c) doses of the activator, (d) modulus of water glass solution, (e) type of slag, (f) fineness of slag, and (g) additive are the major factors that affect the strength development. Fernández-Jiménez et al. [260] conducted a similar study on alkali-activated slag mortars. The authors discuss the factors influencing the strength behavior. The researcher discusses the factors, including the specific surface of slag, curing temperature, activator concentration, and the nature of the alkaline activator. From the study, the author concluded that the most important factor is the alkaline activator, followed by activator concentration and curing temperature. The curing temperature and specific surface of the slag are almost side by side. Bakharev et al. [261] investigated the activation process of slag and analyzed the strength property of the slag. The activation was done with sodium silicate, sodium hydroxide, sodium carbonate, sodium phosphate, and any combination. Discussing the compressive strength of the paste after 28 days of hydration, liquid sodium silicate was proven to be the best one with respect to its competitor. The author also concluded that the liquid sodium silicate concentration below 4% Na was insufficient to activate the slag particle in the first 24 h. To accelerate the process, a 4% Na concentration is a must in the solution. Other than that, the best possible result came when the modulus ranged from 0.75 to 1.5.

Collins et al. [262] developed a new binder by alkali activation. Sodium hydroxide and sodium carbonate were used to form the activation. It was reported that the one-day compressive strength of OPC can be attained using 100% AABs. Atiş et al. [263] used sodium carbonate for activation and concluded that the sodium concentration is highly responsible for reducing the final setting time. Liquid sodium silicate, followed by sodium hydroxide and sodium carbonate, produces maximum compressive and flexural strength in mortar

specimens. The increase in sodium concentration also led to more compressive and flexural strength. Post-28-day strength analysis gives a detailed picture. The slag activated by liquid sodium silicate was more than that of conventional Portland cement. The sodium hydroxide-activated slag possesses more compressive and less flexural strength when compared to conventional Portland cement. The sodium carbonate activator exhibits almost similar results [263]. From the durability perspective, AABs and geopolymers exhibit better performance than Portland cement in aggressive environments. Slag and AABs have shown excellent resistance to chloride penetration, acid attack, freeze–thaw cycles, and sulfate attack due to their refined pore structure. Thus, AABs have been considered to be suitable for marine and sulfate-rich exposures. Ibrahim et al. [264] showed the level of corrosion of embedded steel for different binders (Figure 16). The corrosion in the steel embedded in concrete manifests the cracking of concrete followed by leaching. However, when the dosages of $n\text{SiO}_2$ incorporated in concrete, the resistance of concrete to cracking due to corrosion of reinforcing steel was significantly improved. Other research also found that the concrete with AABs, unlike ordinary Portland cement concrete, demonstrates reduced resistance to carbonation [264–266]. The factors that significantly influence the carbonation process in alkali-activated materials are generally acknowledged to be the type of precursor material used, the specific activators employed, the composition and concentration of ingredients utilized in its mixture design, along with the nature and abundance of alkali activators present.

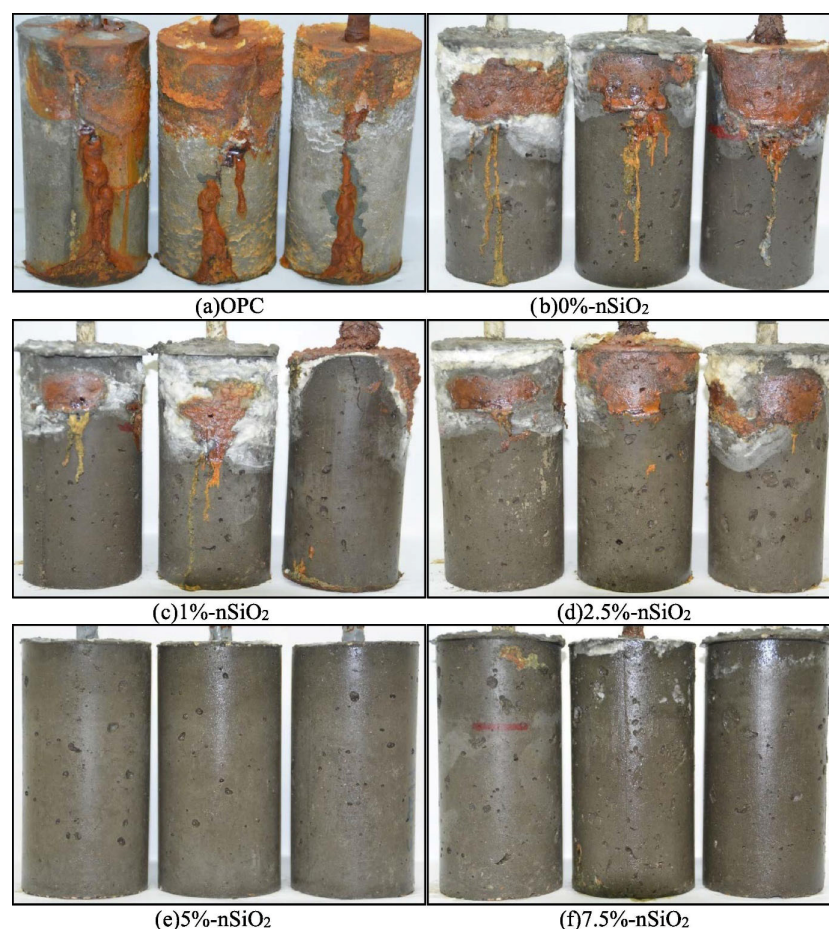


Figure 16. Condition of OPC and AABs subjected to accelerated corrosion [264].

7. Conclusions and Future Perspectives

7.1. Conclusions

In the history of construction, cementitious binders have played a central role, evolving from ancient lime mortars to the global dominance of Portland cement. This review paper presents the development of various binders across different eras, highlighting the challenges and reviewing alternative binders developed over time. The following conclusions can be drawn from this study:

1. The development and innovation of binders have evolved from lime mortars in ancient civilizations to Portland cement, driven by the availability of local resources and technological advancements. The rise of Portland cement paralleled rapid industrialization and infrastructure growth in the 19th and 20th centuries.
2. Although Portland cement is dominating as the main binder in the modern era, its large-scale production is a main source of CO₂ emissions and high energy consumption.
3. Rapid innovation in developing different supplementary cementitious materials (SCMs) from sustainable sources provides partial solutions to reducing clinker use and lowering emissions. However, their widespread adoption in the construction industry remains limited.
4. Alkali-activated binders (AABs) and geopolymer technologies represent promising alternatives to Portland cement. Although AABs outperform Portland cement in durability, acid resistance, high-temperature stability, and strength development, the complex handling of activators, the lack of standardized codes limit their applications. To increase the adoption of AABs further, usability, scalability, and regulatory hurdles need to be addressed.

7.2. Future Perspectives

To ensure a sustainable future for construction, future research aspects are suggested as follows:

1. Expand the applicability and scalability of alkali-activated binders (AABs) and geopolymers as viable alternatives to traditional Portland cement locally produced from diverse industrial and agricultural by-products to meet local construction demands.
2. Conduct extensive studies on the long-term durability of concrete with sustainable binders derived from industrial and agricultural wastes, including examining carbonation resistance, chloride penetration, and chemical attack to ensure their performance under aggressive environments.
3. Establish regulatory frameworks and industry standards that promote the application of AABs and geopolymers, enabling broader acceptance and integration into the construction industry.
4. Undertake life cycle assessments (LCA) to evaluate the effects of AABs and geopolymers on the embodied carbon, energy demand, and cost analyses of concrete to validate real-world feasibility.
5. Support large-scale construction trials to bridge the gap between laboratory research and field applications, providing critical data on constructability, performance, and scalability.
6. Strengthen interdisciplinary collaboration between material scientists, civil engineers, policymakers, and industry stakeholders to foster innovation in this field and adoption through proper policy management, standards and risk assessments.
7. To examine the sustainability trade-offs between mechanical performance, cost and emissions, life-cycle assessment (LCA), and life-cycle cost (LCC) analysis should be performed that captures embodied energy and CO₂ emissions. A combined LCA–

LCC approach will enable the adoption of rational decision-making in order to balance performance, affordability, and environmental benefits.

8. Digital construction methods such as 3D-printed concrete will play a significant role in adopting sustainable binders as such construction significantly uses cement. Three-dimensional-printed concrete has seen a significant rise in its adoption in the construction industry due to efficient utilization of materials, reducing formwork requirements, and enabling topology-optimized designs that reduce material waste as well as reduce binder consumption. Thus, research should focus on examining the effects of various sustainable binders in the extrusion and setting of 3D-printed concrete as well as their structural performance.

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