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Review

Dealuminated Metakaolin in Supplementary Cementitious Material and Alkali-Activated Systems: A Review

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Abstract

This paper presents a comprehensive review of dealuminated metakaolin (DK), a hazardous industrial by-product generated by the aluminium sulphate (alum) industry and evaluates its potential as a component in cementitious systems for the partial or full replacement of Portland cement (PC). Positioned within the context of waste valorisation in concrete, the review aims to establish a critical understanding of DK formation, properties, and reactivity, particularly its pozzolanic potential, to assess its suitability for use as a supplementary cementitious material (SCM), or as a precursor in alkali-activated cement (AAC) systems for concrete. A systematic methodology is used to extract and synthesise relevant data from existing literature concerning DK and its potential applications in cement and concrete. The collected information is organised into thematic sections exploring key aspects of DK, beginning with its formation from kaolinite ores, followed by studies on its pozzolanic reactivity. Applications of DK are then reviewed, focusing on its integration into SCMs and alkali-activated cement (AAC) systems. The review consolidates existing knowledge related to DK, identifying scientific gaps and practical challenges that limit its broader adoption for cement and concrete applications, and outlines future research directions to provide a solid foundation for future studies. Overall, this review highlights the potential of DK as a low-carbon, circular-economy material and promotes its integration into efforts to enhance the sustainability of construction practices. The findings aim to support researchers' and industry stakeholders' strategies to reduce cement clinker content and mitigate the environmental footprint of concrete in a circular-economy context.

Keywords: dealuminated metakaolin; aluminium sulphate waste; cement; concrete; supplementary cementitious materials; alkali-activated cements/geopolymers

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1. Introduction

Concrete is the second most used material and the most manufactured material in the world; for this reason, its production is taxing to the environment not only in terms of water usage and mining/extraction of non-renewable raw materials and the related CO_2 emissions, but also due to the large quantities of cement binder used in its composition. According to studies on sustainable economics, the cement industry is considered one of the major culprits in terms of release of CO_2 and other gases associated with the burning of fossil fuels. Namely, the global cement industry was responsible for emitting 0.576 billion tonnes of CO_2 in 1990 but in the subsequent 24 years, these emissions experienced a significant increase, surpassing a threefold rise to reach 2.083 billion tonnes in 2014 [1]. By 2050, global CO_2 emissions from cement plants alone are expected to reach approximately 2.34 billion tonnes if no mitigation strategies are implemented [2]. Currently, the cement industry contributes approximately to 8% of the global CO_2 emissions [3], which play a critical role in global warming and climate change. The cement industry also affects soil, air, water, and human health. Hazardous heavy metals accumulate in the soil near cement plants, while CO_2, SO_2, NO_X, and fine dust particles cause health risks [4]. Additionally, cement production increases water contamination, leading to elevated pH, phosphate, nitrate, as well as total dissolved and suspended solids [5].

Given the environmental impact of the cement industry, there is a growing interest in finding adequate alternative materials that can either partially replace cement or complete substitute it, as is the case of alkali-activated or geopolymers cements (binders formed by the activation of aluminosilicate materials using various alkalis) [6]. The aim of this effort is to provide effective eco-friendly Portland cement (PC) alternatives with a low carbon footprint, while improving concrete properties in comparison to PC. Improvements could be linked to a rapid strength development, increase in compressive strength, reduced permeability, and high durability [3,6–9]. For example, studies have demonstrated that geopolymers can reduce embodied carbon by up to 80% when utilised as cement replacement [8], with the additional advantage that they can incorporate the use of waste aluminosilicate materials originating from other manufacturing processes (e.g., metakaolin (MK), water treatment sludge (WTS), silica fume (SF), ground granulated blast-furnace slag (GGBS), and electric arc furnace slag (EAFS), making them a promising solution for reducing the environmental impact of construction materials.

In this context, this paper studies dealuminated metakaolin (DK), as a potential material used as a precursor in alkali-activated cements (AAC)/geopolymers or as a supplementary cementitious material (SCM) for the partial replacement of PC. DK is a by-product of the aluminium sulphate (alum) industry, considered one of the most important industries, as alum is used in a wide range of applications like water treatment, manufacturing of paper, dyeing cloths, and others [10,11]. DK is formed during the process of acid leaching of metakaolin to produce the alum and is considered a hazardous material due to its low pH, which increases environmental risks when disposed in landfills, as well as due to its fine particles, which contribute to air pollution [5].

This paper conducts a thorough review of all relevant scientific literature on DK to establish a comprehensive understanding of its characteristics, formation, and potential applications with focus on its use in cement and concrete. Employing a structured and systematic methodology, the collected data has been organised into distinct thematic sections addressing key topics, including the definition and origin of DK, its formation process, hazardous properties, its pozzolanic activity, its performance as SCM in PC-based cement systems, and its integration with different AAC/geopolymer cements.

The innovation of this review lies in its dedicated focus on DK as a distinct industrial waste, rather than treating it generically alongside metakaolin or other aluminosilicates (which have received considerable attention), and its focus on the suitability and challenges of DK as a SCM or AAC precursor for mortar and concrete applications. These remain largely absent from scholarly discussions, despite the widespread production of DK. Importantly, the review critically synthesises the physicochemical characteristics, environmental hazards, pozzolanic potential, and specific cement-based applications of DK in a unified framework, which is currently lacking. Furthermore, the review integrates global data on DK generation and disposal, highlighting its underexplored potential in sustainable construction.

2. Methods

The protocol adopted in this study aligned closely with established methodologies, such as the one used by Elsebaei et al. [12]. The first step in the protocol was conducting a comprehensive search using carefully selected keywords across multiple academic databases, including Google Scholar, ScienceDirect, ASCE Library, Emerald Insight, SpringerLink, and others. The main keywords used in the search process included, though not limited to, the following: "Dealuminated Metakaolin", "Partially Dealuminated Metakaolin", "DK", DMK", "aluminium sulphate waste", "aluminium sulphate residue". Identified publications were initially screened for relevance based on their titles and abstracts. Subsequently, this was followed by a thorough review and in-depth analysis of the selected publications. To further strengthen the review, both backward and forward citation tracking was performed to identify additional relevant studies that may not have appeared in the initial search. This step was particularly important given the limited amount of existing research on DK. All findings were then systematically compiled and analysed to construct the foundation of the literature review. Finally, the gathered data was organised and categorised into well-defined sections and subsections, each addressing a specific topic. Figure 1 shows an outline of the literature review protocol used in this study.

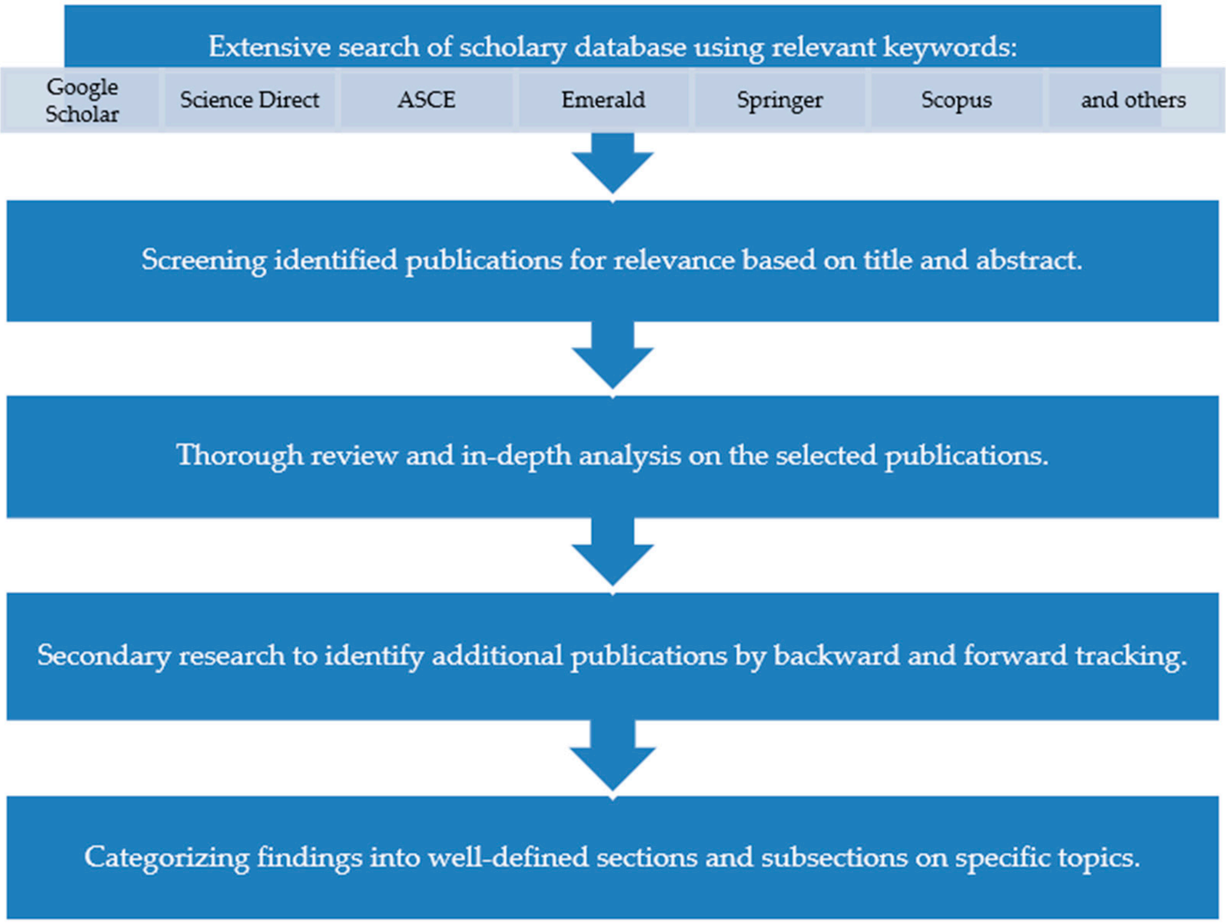


Figure 1. Literature review protocol.

The topics addressed in this review have been categorised into the following sections:

- Formation of DK
- Hazardous Nature of DK
- Statistics on DK
- Pozzolanic Effect of DK

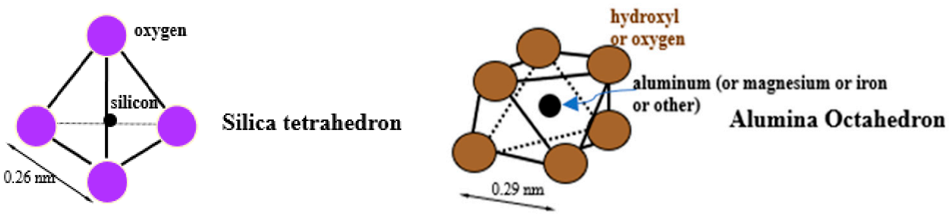
- Incorporation of DK as SCM in Portland Cement systems
- Incorporation of DK in Alkali activated cement systems:
    a. Incorporation of DK with Metakaolin
    b. Incorporation of DK with Water Treatment Sludge WTS
    c. Incorporation of DK with Slag (GGBS and EAFS)

3. Literature Review

3.1. Formation of DK

This section overviews the formation process of DK, starting with kaolinite mineral and covering the production process and relevant chemical reactions, and ending with the by-product DK. Kaolinite, with the chemical formula $Al_2Si_2O_5(OH)_4$, is a clay mineral consisting mainly of layered octahedral alumina (AlO_6) and tetrahedral silica (SiO_4) sheets (1:1 lattice). These are linked through hydrogen bonding between hydroxyl groups from the octahedral Al–OH sheet and basal oxygen atoms of the adjacent tetrahedral Si–O sheet, as well as covalent oxygen sharing between the tetrahedral and octahedral sheet within each 1:1 layer unit [13]. Figure 2 shows the structure unit of the sheets, and Figure 3 shows schematically the 1:1 lattice structure of kaolinite. During calcination at temperatures ranging at 550–900 °C dehydroxylation of kaolinite occurs [14,15]. As a result, chemically bonded water is released, and hydroxyl groups (-OH) in the octahedral alumina sheet are broken [16,17]. The crystalline structure collapses, transforming into a new disordered phase called "Metakaolin", a highly reactive, amorphous pozzolan with the chemical formula $Al_2Si_2O_7$ [5,15]. Equation (1) describes the chemical reaction of metakaolin formation:

$$Al_2O_3\cdot 2SiO_2\cdot 2H_2O(\text{kaolinite}) \overset{\Delta, 550\text{–}900\ ^\circ C}{\rightarrow} Al_2O_3\cdot 2SiO_2(\text{metakaolin}) + 2H_2O(g) \tag{1}$$



(**a**) (**b**)

Figure 2. (**a**) Tetrahedral silica and (**b**) octahedral alumina structural units.

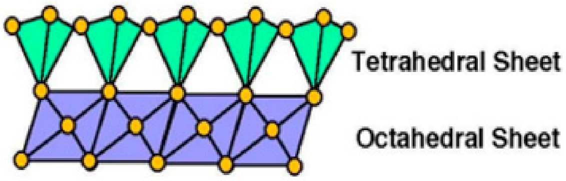


Figure 3. 1:1 lattice structure of the kaolinite mineral.

At temperatures above 900 °C, a different molecular arrangement occurs, leading to the formation of an intermediate aluminosilicate spinel-type phase with the approximate composition $2Al_2O_3\cdot 3SiO_2$, which precedes the crystallisation of mullite ($3Al_2O_3\cdot 2SiO_2$) [14,15].

When used in the alum industry, the metakaolin thus formed undergoes acid leaching with sulphuric acid to produce aluminium sulphate (alum), a process that also generates a solid by-product material rich in silica (SiO_2) known as dealuminated metakaolin

(DK) [18,19]. Equation (2) describes the chemical reaction of the dealuminated metakaolin using sulphuric acid [20]:

$$Al_2O_3{\cdot}2SiO_2 + 3H_2SO_4 + 48H_2O \overset{\text{yields}}{\to} Al_2\,(SO_4)_3{\cdot}51H_2O\ (\text{liquid alum}) + 2SiO_2 \quad (2)$$

It is important to note that the efficiency of the alum extraction reaction is not 100%; as a result, some alumina stays unreacted. That is why the X-Ray Fluorescence of any dealuminated metakaolin shows around 6% to 8% of alumina [20].

3.2. Statistics on DK

DK is produced wherever alum is manufactured, spanning Asia, Africa, Europe, and North America. The aluminium sulphate market was totalling ~USD 1082 million globally in 2024 and is projected to grow to ~USD 1370.6 million by 2032, with the primary driver for the growth of this market being the increasing global need for clean water, followed by its applications in the growing paper and pulp as well as textile industries, with some emerging niche applications being soil stabilisation, advanced paper coatings, and fire-retardant uses. For context, the global aluminium sulphate demand in these sectors, indicates its scale across the Asia-Pacific (42%—leading the market due to rapid industrialisation and infrastructure development, especially in China and India, countries that have expanded textile output, increasing demand for auxiliary chemicals), North America (24%—linked to industrial demand in the U.S.A. and Canada and advanced water treatment systems), and Europe (19%—demand linked to strict environmental regulations and paper and chemical industries). The remaining 15% is shared by Latin America, the Middle East, and Africa (demand linked to sanitation infrastructure improvement and urban growth) [21] (see Figure 4). This underscores the geographic spread of DK.

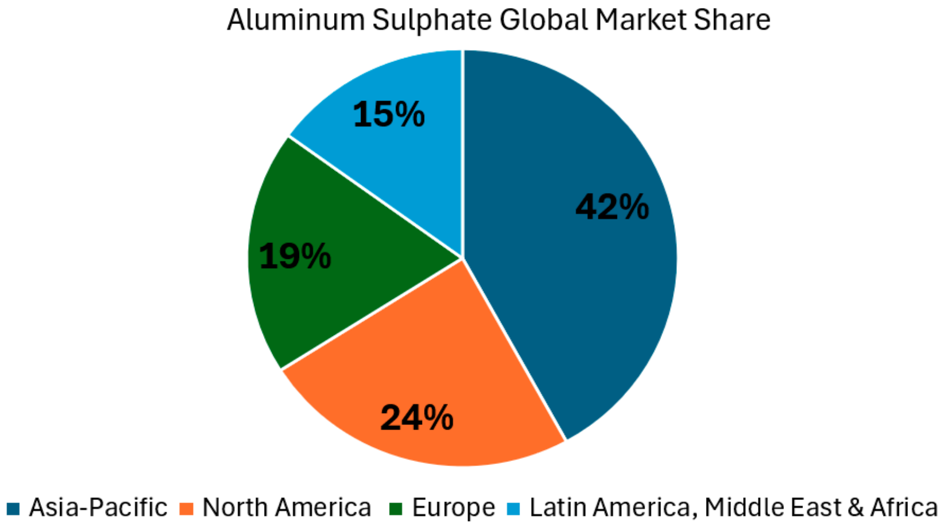


Figure 4. 1:1 Alum global market share data, reported in [21].

Although specific data on DK production are limited, substantial generation can be inferred in these regions based on alum output, as demonstrated by existing figures on DK production from China, Egypt, and Ethiopia. Namely, statistics on DK in China recorded around 770,000 tonnes of DK produced from the alum industry in 2019 [22]. Another recent study in China reported one factory in Hengyang alone disposing of 50,000 tonnes annually [23]. In Ethiopia, one alum plant was reported to generate approximately 5000 tonnes of DK per year [24]. Egypt possesses extensive kaolin deposits, concentrated in three key regions: Aswan, The Red Sea, and South Sinai, making it a primary resource for the ceramic and alum industries [25]. Studies regarding DK production in Egypt estimated the annual production of alum to be 240,000 tonnes, which generates around 80,000 tonnes of DK as a by-product [26]. Of this, only 1–2% is recycled, and the rest is dumped into the desert [27,28].

These figures support the conclusion that DK is a considerably large and geographically widespread industrial residue, warranting further attention for reuse and valorisation in cement systems.

3.3. Hazardous Nature of DK

DK is considered a hazardous material and presents multiple environmental hazards, when dumped or discarded in landfills in large quantities.

The first hazard is linked to its fine particle size, attributing it hazards linked to fine respirable dust. The fineness of DK is due to its exposure to acid leaching process. It is lightweight and its surface area can typically reach up to 90.5 m^2/g [5,29]. These characteristics make it easily airborne, posing a risk of respirable particulate pollution near disposal sites. This dust can also settle on leaves and soil, disrupting photosynthesis, interfering with nutrient cycling, and affecting microbial communities in the rhizosphere, thereby reducing plant growth and impairing ecosystem function. DK dust may also introduce aluminium and other trace metals to plant surfaces and soils, further stressing ecosystems. Particulate matter deposition into rivers, lakes, or wetlands contributes to increased turbidity, reduced light penetration, and accumulation of pollutants (including aluminium and trace metals), affecting aquatic life, especially sensitive planktonic organisms [30].

Additionally, the pH value of DK typically measures around 4.3 [22] and can even reach as low as 3 [24]; due to its high acidity, uncontrolled DK disposal can cause severe damage to soil and groundwater due to acid leaching, increasing the solubility and mobility of potentially toxic metallic species, thus raising toxicity levels. It is therefore posing a major threat to agricultural soils and terrestrial ecosystems, while the mobilisation of toxic metallic species also results into their transport from the soil to aquatic environments. The toxic effects of Al on multiple aspects of freshwater and marine animal life (embryogenesis, reproduction, respiration, etc.) and toxicity evolution in response to acidification have been extensively researched and documented [31].

As noted earlier, although DK undergoes controlled leaching to remove some aluminium, which increases its silica-to-aluminium ratio, it still retains significant residual aluminium, which can become less stable in acidic environments. Thus, upon acid exposure DK is subject to further dealumination, releasing aluminous species from its structure to the environment. This includes higher, hazardous concentrations of bioavailable Al^{3+} and $Al(OH)_2{}^+$ as the pH values of DK align with pH ranges (<5.5) where aluminium solubility increases. Unfortunately, in many countries worldwide, the majority of DK is disposed in uncontrolled dump fields, which has led to problems such as farmland encroachment and soil and groundwater pollution, as reported in studies from Egypt [27,28] reporting that dumping DK has caused soil degradation, crop damage, and surface/subsurface acidification, illustrating its real-world hazards.

Finally, acidic leachate from DK can degrade nearby underground infrastructure, a well-known hazard related to acid-driven corrosion. This shows the urgent need for finding useful alternative outlets for this waste.

The potential for dust dispersion, acidification, heavy metal mobilisation, and exceedance of toxicity thresholds explain the classification of DK as a hazardous industrial residue. The related risks emphasise the need to divert DK into valorisation pathways, such as incorporation in cement systems, to protect soil, water, and human health.

3.4. DK in Cements for Mortars and Concrete

3.4.1. Pozzolanic Activity

Several studies performed on DK assessed its pozzolanic activity towards its use in cement and concrete. This was reported to be higher than that of silica fume (SF) but also metakaolin (MK) in supplementary cementitious material (SCM) systems [32–35]. This performance stems from the high content of hydrated amorphous silica in the DK, which reacts swiftly with calcium hydroxide ($Ca(OH)_2$) produced during cement hydration to form calcium silicate hydrate (C-S-H) gel and the modified microstructure of DK compared to MK, as acid attack significantly increases porosity, hence increasing DK specific surface area (e.g., BET values of 40–90.5 m^2/g, vs. 18.8 m^2/g for silica fume, were reported in [29,34]), although there is no considerable decrease in the average pore size [29]. These features facilitate rapid dissolution and release of silica into pore solution and enhance reactivity with portlandite (pozzolanic reaction), driving high activity indices, e.g., 120% and 127% relative to SF (ASTM C618 method) were reported, respectively, in [34,35].

These findings confirm that the performance of DK in SCM systems, is likely to be influenced by both its enhanced microstructure (surface area, porosity) and its chemical composition (amorphous silica), driving its rapid and effective reaction with calcium hydroxide to accelerate calcium silicate hydrate (C-S-H) formation, which is linked to strength development.

Based on these findings, numerous studies have explored the potential use of DK in the production of concrete with supplementary cementitious material (SCM) systems or alkali-activated cements (AACs)/geopolymers. The following sections provide a comprehensive review of such applications. The most essential research papers alongside with mix designs and key findings are also summarised in the comparative overview in Tables 1 and 2, respectively, for DK as SCM in Portland cement-based systems or precursor in AAC-based systems.

3.4.2. DK as SCM in Portland Cement-Based Concrete

As noted earlier, DK is chemically characterised by a significant reduction in alumina content compared to metakaolin (MK) and an increase in reactive amorphous silica. This transformation affects its behaviour as pozzolanic material. Most studies (15 publications) focus on DK as partial replacement of Portland cement (PC). An early comprehensive investigation [36] used two different Egyptian DK samples combined with CEM I and II (according to EN 197-1 [37]). Tests studied the setting time, flow, and slump of cement and fresh concrete as well as the compressive strength of mortars and hardened concrete. Samples were prepared using two approaches: partial replacement of cement with DK and DK added to PC. The results showed that 10–15% DK replacement enhanced mortar strength by up to ~14% (except for one mix where the strength was not affected) and concrete strength by ~10%. The lower strength improvement in concrete mixes was attributed to the difficulty of dispersion of DK particles in concrete mixes compared to mortar mixes. This observation is likely due to surface adsorption effects; DK immobilises water because of its high specific surface area, increasing the water demand and necessitating the use of superplasticisers. The reduction in flow (~50%) was also attributed to the fine particles of DK and the increased porosity, increasing water demand and necessitating superplasticiser use to maintain consistent slump (S4) (170–200 mm), compared to the control mix. The investigation also reported a high specific surface area for both DKs used, i.e., 20.1 m^2/g and 43.2 m^2/g, respectively, and high strength activity indices (SAI) at 28 days for both DKs, i.e., 0.94 (94%) and 1.13 (113%), respectively, for a 10% cement replacement by DK.

Setting time remained broadly unchanged. The authors do not explain this observation, but it is possible that DK, containing reactive alumina and sulphate, and its interaction with cement aluminates facilitated early ettringite (AFt) formation, thus compensating for potential retardation effects. Moreover, preliminary durability testing on selected concrete samples showed that the carbonation depth was almost 2 mm after exposure for 70 days at 25 °C using 4% CO_2 [36].

A study by Abdelalim et al. [34] compared acid DK (pH ~5, DKut) and lime-treated DK (pH ~8, DKt) used as partial PC replacement in mortars. They had, respectively, 42 m^2/g and 40.1 m^2/g BET specific surface area; DKut showed ~127% pozzolanic activity and DKt ~117% pozzolanic activity vs. SF. The mineralogical composition of both DKs consisted mainly of quartz (SiO_2) with small traces of anatase (TiO_2). The only difference was that DKt had a significantly higher amount of calcium sulphate hemihydrate ($CaSO_4 \cdot ½H_2O$) compared to DKut.

Both DK types required a higher water-to-binder (w/b) ratio to achieve a standard consistency as DK dosage increased; this was expected due to the high specific surface area of DK and is consistent with [36].

DKut accelerated setting by ~25% at 10% replacement, while DKt initially slightly reduced setting time at 5% replacement but retarded it above 5% replacement. The authors explained that DKut contains a higher amount of alumina (13%) versus only 5.6% in cement, which can promote the formation of crystalline hydration products when used with cement, thereby accelerating both hydration and setting. This is consistent with [29], which confirms the presence of significant reactive alumina content in DK, available in a soluble form, and observing that DK rapidly consumes lime and forms aluminous hydration products, including ettringite at high $Ca(OH)_2$ availability and allophone-type phases at low $Ca(OH)_2$ availability. Indeed, in PC, the presence of Al^{3+} and $SO_4{}^{2-}$ encourages early ettringite (AFt) formation, increasing rapidly the stiffness of the mix (thus quick setting). Conversely, the lime-treated DKt contains increased amounts of calcium sulphate hemihydrate ($CaSO_4 \cdot ½H_2O$), and likely some excess lime ($Ca(OH)_2$), which appear to shift the hydration pathway, potentially reducing the formation of ettringite (AFt) and favouring monosulphate (AFm) as a dominant sulphate-bearing phase. Although the authors do not directly attribute the delayed setting to AFm formation, the observed compositional differences suggest an altered early hydration pathway that may contribute to slower matrix stiffening, as AFm (which is less expansive than Aft) forms more slowly than Aft and is less associated with rapid early crystallisation; this could explain the observed delay in setting. These contrasting behaviours of the two DK types in fresh mixes show how small compositional shifts in DK (alumina form, sulphate) can alter early hydration kinetics and pathways, ultimately determining whether setting is accelerated or delayed.

Consistently with [36], the study also reported a considerable drop in the relative flowability (RF) (i.e., the ratio of the flow directly after mixing to the flow after an elapsed period) with the incorporation of either type of DK. After 60 min, the reported RF was 63% and 76% at 15% replacement levels of DKut and DKt, respectively. Finally, the study showed an enhancement in compressive and tensile strengths by 12% and 10%, respectively, at 10% DK replacement.

Other studies confirm up to ~20% strength gain in concrete with $\leq$15% DK substitution of PC [28]. Conversely, Moselhy [38] observed a 3% improvement in compressive strength of concrete with $\leq$15% DK substitution of PC and a 25% reduction in slump at 15% DK replacement, unlike [36]. However, [38] did not use any superplasticiser or water reducers, which may explain the poor workability and potentially the limited strength gain, as the mix could have been too stiff and porous, with insufficient water for effective hydration. Two subsequent studies incorporated DK in cementitious mortars [39,40], where DK was introduced as a sand replacement rather than cement replacement. Study [39] found that the setting time was broadly unaffected, consistently with [36]; an 18% drop in flowability at 10% DK content and a 46% increase in compressive strength with 17.5% DK content were observed. Higher DK contents led to compressive strength reduction. [40] mainly focused on the effect of incorporating different DK particle sizes ranging from under 75 µm to over 300 µm—a particle size range larger than expected for DK given the high specific surface areas reported in other studies. No considerable effect of DK particle size on compressive strength was found. At a replacement percentage of 2.5%, DK significantly delayed the setting time, acting as a clear retarder. Higher replacement percentages slightly reduced the setting time. Overall, the presence of DK shifted the setting time curve towards retardation, unlike in [34], where DK acted as a strong accelerator in both low and high DK percentages. However, the particle size range in [40] (75–300 µm) is unusually coarse for DK (considering particle size ranges reported in most other studies), which may partially explain the observed retardation.

A comprehensive study was carried out to assess the durability of concrete incorporating DK in aggressive environments [41]. Cement was partially replaced by DK at 10% and 15%, and concrete samples were tested at 7, 28, 60, 90, and 180 days under various curing regimes. Initially, compressive strength tests were performed using submerged curing, serving as a baseline for strength development. Additional curing methods were then applied, each addressing specific durability aspects. At 28 days, submerged-cured specimens with 10% and 15% DK showed compressive strengths of 52.6 MPa and 47.5 MPa, corresponding to increases of 27.7% and 15.3% over the control mix (41.2 MPa). Compressive strength was used as the primary indicator of durability, complemented by tests on resistance to chloride and sulphate attacks, seawater exposure, and elevated temperatures. Concrete specimens were cured in 5% NaCl solution and tested at the aforementioned ages. Strength continued to rise in DK-modified mixes, peaking at 65 MPa (10% DK) and 57.7 MPa (15% DK) at 90 days. This increase was attributed to the active amorphous silica in DK, which reduces chloride permeability and enhances Cl^- resistance. A slight strength drop (~7%) was observed at 180 days, likely due to Friedel's salt formation (calcium chloroaluminate) from $CaCl_2$ and calcium aluminate reactions. A similar trend was observed for specimens cured in seawater (salinity = 4.3%), with comparable strength values. Concrete exposed to 5% $MgSO_4$ solution showed strength gains, albeit lower than those seen under chloride or seawater exposure. The strength increase was attributed to the formation of gypsum, resulting from the reaction between the sulphate ions and calcium ions. Gypsum reacts with calcium aluminate to form ettringite mineral which may increase expansion.

Regarding heat resistance, the same study [41] reported increased compressive strengths at 300 °C, followed by reductions at higher temperatures. At 300 °C, 10% and 15% DK mixes reached 75.5 MPa and 73.8 MPa, respectively, then declined to 33.4 MPa and 25.7 MPa at 700 °C. The control mix reached 63.6 MPa at 300 °C and 17.8 MPa at 700 °C. Thus, the 10% DK mix showed the best thermal resistance at 700 °C, with a 36.5% strength reduction from its room temperature value (52.6 MPa). Total porosity also decreased notably in DK mixes: at 700 °C, 10% and 15% DK mixes had porosities of 12% and 13.4%, respectively, compared to 23.4% in the control. This reduces permeability and thus enhances chemical resistance, contributing to improved durability.

A related study with a narrower scope [42] assessed the durability of cement mortars with 7.5% and 10% DK exposed to seawater (salinity = 4.3%). Compressive strength was again used as the primary durability indicator, along with expansion and weight change. As in [39], strength increased up to 90 days then declined. At 28 days, compressive strengths of 0%, 7.5%, and 10% DK mixes were 37 MPa, 46 MPa, and 42 MPa, respectively. At 90 days, they were 44 MPa, 52 MPa, and 48 MPa, respectively; by 365 days, they had dropped to 31 MPa, 41 MPa, and 37 MPa. Mortar bar expansion was lowest in the 7.5% DK mix (0.078%) compared to the control (0.12%), indicating enhanced chemical resistance. Weight gain was also monitored: a slight increase was observed in all mixes, likely due to moisture absorption or calcium compound formation. The 7.5% DK mix had the lowest gain (0.62% at 365 days) versus 1.07% in the control.

Some studies also investigated the performance of DK-containing cement systems with further chemical additives such as titania (TiO_2) [43,44]. Three different ratios of cement/DK were tested (90/10, 70/30, 50/50) and an additional mix incorporating 5% TiO_2 (cement 50%, DK 45%, TiO_2 5%) was specially prepared. The 28-day compressive strength results showed that the inclusion of 5% TiO_2 raised the strength value from 56 MPa (sample with 50% DK) to 88 MPa as compared to the control mix (80 MPa). The study found that a high replacement level of up to 50% DK is feasible in cement systems and can even result in better compressive strength performance with the addition of a small percentage of commercial TiO_2. The gamma-ray shielding power was also investigated, and results showed a significant improvement, as the linear attenuation coefficient (μ) increased from 0.127 cm^{-1} to 0.199 cm^{-1} at 661.6 KeV for the DK mix with TiO_2 and control (0% DK) mix, respectively. The authors explained that the TiO_2 may act as a nano-filler and active seeds in the formation of gels CSHs, CAHs, and CASHs as indicated in the XRD, TGA, and SEM tests. The N_2 adsorption–desorption isotherms showed texture characteristic changes from macro to mesopores when TiO_2 was added. The increase in compressive strength was also attributed to structure redistribution. The innovative idea of incorporating a small amount of titania could potentially lead to a breakthrough in achieving higher DK contents in the concrete mixes.

DK as SCM with the addition of nano-carbon black (NC) was also studied for mortar mixes [35]. Cement mortar samples with up to 20% DK were tested; they all showed an enhancement in compressive strength, with 28-day strength values of 46.2 MPa and 50.7 MPa, respectively, for 5% and 10% DK. This corresponded to a 4.5% and 14.4% improvement, respectively, compared to the control sample (44.3 MPa). The FTiR results indicated the formation of more C-S-H gel when DK was introduced. By adding small percentages of NC (from 1% to 3% by DK weight), a marginal gain in compressive strength of approximately 1.5% was observed in all mortar samples. Regardless of the inclusion of NC, all samples with DK generally suffered a dramatic reduction of over 50% in setting times. Namely, the initial setting time was 58.3 min and 79.5 min for the 20% DK for the control and 20% mix, respectively; the final setting times were 118.6 min and 158 min for the control and 20% mix, respectively.

Silica fume (SF) was also incorporated into DK/cement mixes [45]. In this detailed experimental study, DK was used as a partial cement replacement at 20%, 25%, and 30% by weight to produce pozzolanic cement pastes with and without the addition of 5% SF. The experimental programme included evaluations of early and late compressive strengths, as well as hydration kinetic parameters such as chemically combined water (Wn%) and free lime content. The hydration behaviour and performance of these systems were evaluated over a 90-day period using compressive strength testing, Differential scanning calorimetry (DSC), X-ray diffraction (XRD), Fourier-transform infrared spectroscopy (FTIR), and scanning electron microscopy (SEM). The results showed that early-age compressive strengths (1–28 days) were generally lower in DK-modified pastes compared to the control (100% PC), which the authors attributed to unreacted DK, acting as a filler in the matrix until its pozzolanic activity becomes dominant. However, at 90 days, pastes with 20% and 25% DK demonstrated improved or comparable strength to the control, attributed by the authors to the gradual pozzolanic consumption of calcium hydroxide (CH) and the consequent formation of C-S-H. Conversely, the 30% DK blend exhibited the lowest strength across all hydration ages; the authors attributed this to excessive cement replacement resulting in insufficient portlandite (CH) generation to sustain further pozzolanic reactions, and hence strength gain, due to excessive unreacted material. In this case, the unreacted DK is likely to have acted more as an inert filler than as a reactive material. The sharp performance drop at 30% DK suggests a critical threshold for useful replacement, reinforcing the need for cautious mix optimisation.

The inclusion of 5% SF in the DK blends led to considerable improvements in compressive strength at both early and late hydration ages, with the greatest enhancement observed in the 30% DK mix. The authors attributed this to a synergistic pozzolanic effect between DK and SF, both of which react with portlandite (CH) to form large quantities of amorphous and microcrystalline C-S-H. Evidence for this interpretation was based from the combination of multiple techniques: DSC and XRD analyses, which showed reduced CH peaks and stronger C-S-H signatures; FTIR spectra, which revealed stronger bands associated with silicate and aluminate phases; SEM micrographs, which illustrated denser microstructures in SF-containing pastes with fewer visible portlandite crystals and a more homogeneous distribution of hydration products. The microstructural evidence presented (especially from SEM) suggests that the fine particle size of SF may improve packing density and reduce porosity.

These observations support the authors' view that SF contributes not only chemically through extended pozzolanic activity but also physically by enhancing matrix densification, although it is unclear whether the observed microstructural densification is primarily due to filler effect of SF or a continuation of pozzolanic activity under limited CH, which are both possible. This dual chemical–physical role may be particularly beneficial in systems where DK has already consumed most of the available CH, allowing SF to continue contributing to long-term strength through slower pozzolanic reactions and microfiller effects.

Hydration kinetic measurements further corroborated the overall trends. Chemically combined water (Wn%) increased with both DK and SF content over time, reflecting the progressive formation of hydration products. However, as each mix used a different water content to achieve standard consistency direct comparisons of Wn% across different mixes are difficult to make. Free lime measurements revealed that CH content decreased over time in all DK-containing mixes, with the most pronounced reductions observed in mixes containing both DK and SF. This confirms the active pozzolanic consumption of portlandite to produce C-S-H and aligns with the observed strength development and phase changes.

Overall, the study provides a thorough analysis of how DK and SF interact in cement pastes. While some aspects, such as the exact contribution of SF relative to DK in high-replacement mixes, could benefit from further quantitative differentiation between chemical and physical effects, the authors' interpretation is well-supported by multi-technique evidence. The findings point to a potentially valuable synergy between a rapidly reacting pozzolan (DK) and a more slowly reacting one (SF), particularly in enhancing long-term mechanical and microstructural performance that could be explored for DK mix design optimisation.

To conclude the section and visualise key findings, Figure 5 presents compiled compressive strength results (for pastes, mortars, and concrete) from five available studies to facilitate a comparison between the results using DK as SCM. Each study is represented with a distinct colour, marking the respective control mixes and those incorporating DK. The figure clearly illustrates that incorporating DK in low percentages (typically between 5% and 15%) consistently enhances compressive strength by an average of 20%, regardless of the tested material (paste, mortar, or concrete), the type of cement used, or the source of DK. However, only one study [43] investigated higher replacement percentage (above 20%), and a notable decline in compressive strength was observed. This explains why most researchers have shifted focus towards multicomponent SCM-DK systems with lower DK content, as compressive strength remains the most important mechanical property in evaluating concrete performance. This study [43] specifically tested higher percentages of

DK reaching 50% replacement and reported a high drop in compressive strength of 30% compared to the control mix, unless a small amount of TiO_2 was used.

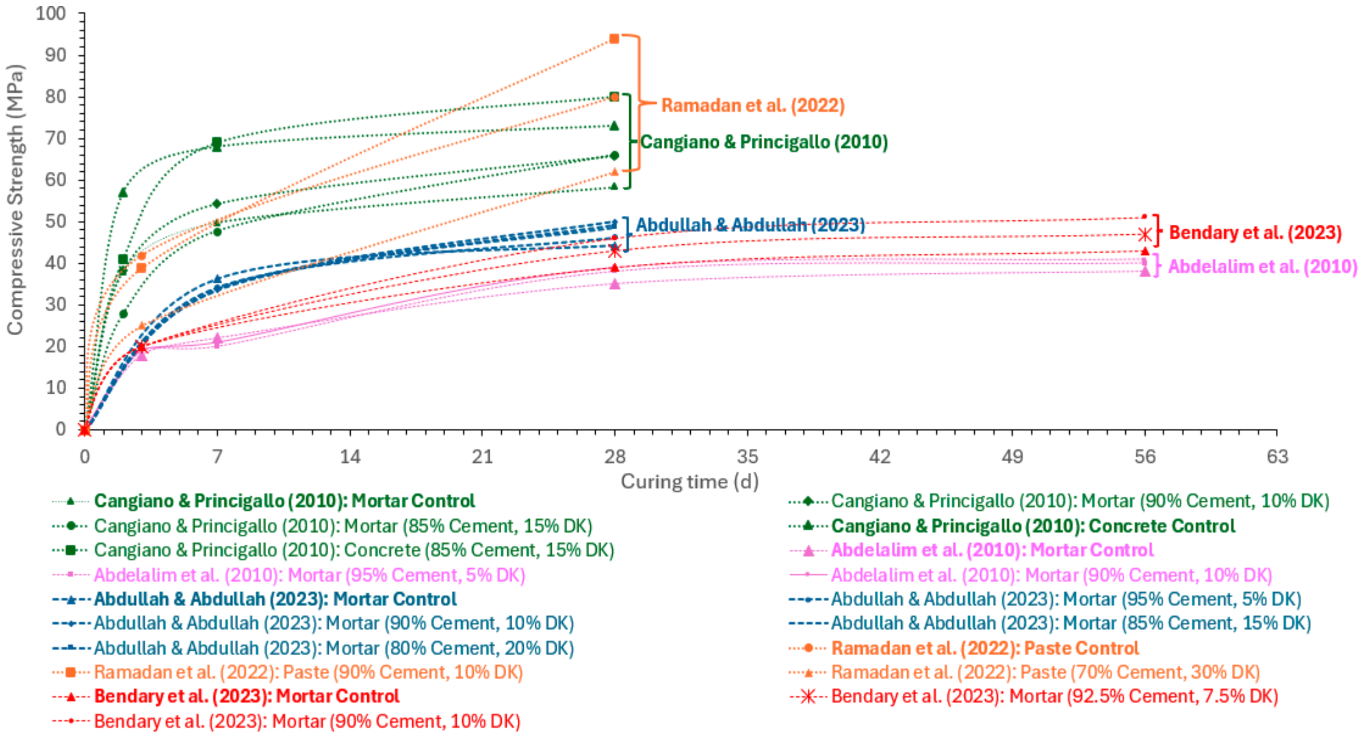


Figure 5. Compiled compressive strength results of DK used in SMS systems—based on Abdelalim et al (2010) [34], Abdullah & Abdullah (2023) [35], Bendary et al. (2023) [42], Cangiano & Principallo (2010) [36], and Ramadan et al (2023) [43].

To summarise, the above findings confirm that DK can be used as a SCM, primarily due to its high amorphous silica content and strong pozzolanic reactivity. In PC systems, the reactive silica in DK reacts with portlandite ($Ca(OH)_2$) to form additional calcium silicate hydrate (C-S-H), the principal phase contributing to long-term strength. This explains the strength enhancements observed in SCM mixes incorporating DK, particularly at moderate replacement levels (20–25%).

Additionally, the relatively low alumina (Al_2O_3) content in DK may help limit the formation of delayed ettringite ($C_6A(SO_4)_3{\cdot}H_{32}$), which typically occurs due to the reaction of calcium aluminates with sulphates in sulphate-rich environments. In systems with sufficient sulphate, ettringite can form early via the hydration of C_3A, as illustrated below:

$$C_3A + 3CaSO_4{\cdot}2H_2O + 26H_2O \rightarrow C_6A(SO_4)_3{\cdot}H_{32} \tag{3}$$

$$SiO_2\ (\text{amorphous}) + Ca(OH)_2 + H_2O \rightarrow \text{C-S-H} \tag{4}$$

where C = CaO, S = SiO_2, A = Al_2O_3, and H = H_2O.

The high surface area and amorphous structure of DK enhance its pozzolanic reactivity, contributing to improved long-term strength and reduced permeability. In some DK-containing systems, rapid setting has been observed, which may result from soluble sulphate species and residual reactive alumina promoting early hydration reactions and localised ettringite and C-S-H formation. However, the formation of ettringite is highly dependent on system chemistry and lime availability, as shown by the absence of ettringite in low-lime DK systems studied in [29].

3.4.3. DK in Alkali-Activated or Geopolymer Cement Systems

Alkali-activated cements (AACs) are PC-free binders originating from the reaction of an alkali metal source with an (alumino-) silicate. They are commonly under two main categories and respective activation models, namely (1) high-calcium (Ca+Si) model, such as the activation of blast furnace slag (with a $CaO–SiO_2 > 70\%$) to form C-(A)-S-H (calcium (alumino-) silicate hydrate) gel as a main reaction product, which is like the gel obtained during PC hydration (except here Al is also present), and (2) low-calcium (Si+Al) activation model, comprising activation of materials rich mainly in aluminium and silicon, e.g., type F fly ash (low in calcium), metakaolin or silica fume. Although DK has not been discussed in this context in the reviewed literature, it would belong to the latter category. In this category, the main reaction product is N-A-S-H (alkaline aluminosilicate) gel, a three-dimensional inorganic alkaline polymer that can be regarded as a zeolite precursor, known as a geopolymer [6].

The main process of geopolymer formation involves three main steps: (1) dissolution of aluminosilicate precursors, releasing silicate and aluminate monomers; (2) polycondensation and reorganisation of these monomers into semi-crystalline three-dimensional structures; (3) gel hardening, where the aluminosilicate gel condenses further into a rigid, cross-linked network. Monovalent alkali metal cations (e.g., Na^+, K^+), generally denoted as M^+, are required to balance the negative charge of $AlO_4{}^-$ units, and water is progressively released during the condensation process. The general chemical reactions representing each stage are as follows:

$$Al_2Si_2O_7\ (s) + 4OH^- + 7H_2O \rightarrow 2Al(OH)_4{}^- + 2Si(OH)_4 + 2H_2O\ \text{(Dissolution)} \quad (5)$$

$$Si–OH + HO–Si \rightarrow Si–O–Si + H_2O\ \text{(Polycondensation)} \quad (6)$$

$$Si–OH + HO–Al \rightarrow Si–O–Al + H_2O\ \text{(Hydrate gels)} \quad (7)$$

$$[–AlO_4{}^-] + M^+ \rightarrow [–AlO_4{}^- \cdot M^+]\ \text{(Hardening)} \quad (8)$$

where M^+ represents a monovalent alkali metal cation such as Na^+ or K^+.

The chemical composition of the aluminosilicate precursor, alkali combination used in Equation (5) and the gel concentration and hard composite geopolymer formed in Equation (8) depends on several variables such as particle size, dissolution and alkaline concentration, among others. The following section reviews research publications in DK used as precursor for geopolymers through the process described in Equations (5)–(8).

DK with Metakaolin

DK has also been explored in the development of AAC/geopolymer cements. As major constituents of DK are silica and alumina, DK is likely to be a good candidate as AAC/geopolymer precursor. Nine studies were identified in the literature, three of which investigate DK and metakaolin (MK) precursor system. Namely, in [16], mortars consisting of various blends of DK and MK were examined to evaluate their compressive strength performance. A total of 13 mortars with different DK/MK ratios (ranging from 20% DK to 70% at 5% intervals) were tested, with sodium hydroxide (NaOH) used as the alkali activator. Water content and NaOH were used to maintain a fixed water-to-sodium oxide (H_2O/Na_2O) molar ratio of 10 in all 13 mixes. Results showed that the mortars containing 35% and 40% DK exhibited the highest compressive strength values at both 7 and 28 days of curing. The values of compressive strength for the 40% DK mortar at 7 days and 28 days were 41.4 MPa and 51.3 MPa, respectively. SEM/EDX showed a more homogenous matrix and a smooth surface with lower cracks in mixes with highest strengths. Another study with a wider scope tested 26 mortars with different DK/MK ratios (ranging from 0% DK to 60% at 5% intervals) [15]. Water and NaOH were used to form two amounts of Na_2O (1.94 M and 2.32 M) and a unified H_2O/Na_2O molar ratio of 13.7. The mix with 25% DK (with the lower Na_2O = 1.94 M) showed the best results in terms of highest compressive strength, lowest water absorption, and lowest porosity. Compressive strength values of this mix at 7 days and 28 days of curing were 55 MPa and 64 MPa, respectively, compared to the 0% DK mix (11 MPa and 18 MPa, respectively). Porosity and water absorption of the 25% DK mix were 12.5% and 6.5%, respectively, compared to the 0% DK mix (16.5% and 8.5%, respectively). Similar results were also obtained by Hassan H. et al. [18] who tested nine DK/MK mortar mixes using three different activator concentrations. The 50% DK mix had the highest compressive strength at 7 days of 63 MPa. Flowability and setting time were also tested and it was shown that their values reduced with increasing DK contents.

DK with Water Treatment Sludge

Two papers [46,47] used DK with water treatment sludge (WTS) in geopolymer cements. In [46] mortars consisting of different WTS:DK ratios were studied using different concentration of activators and a curing regime under different temperatures. Results showed that the mix with WTS:DK ratio of 30:70 cured at 70 °C was the optimum mix, as the mortar reached a compressive strength of 7 MPa and 22 MPa, respectively, after 7 days and 28 days of curing. A further study by the same group investigated a different application of WTS/DK geopolymer systems, namely their heavy metal immobilisation efficiency based on leaching tests. The results showed a high immobilisation efficiency of Cd^{2+}, Pb^{2+} and Hg^{2+}; the highest immobilisation efficiency of 98% was found for Pb^{2+} [47].

DK with Slag (GGBS and EAFS)

The literature search identified four studies where DK was used as a precursor together with a slag. Two of these incorporated DK with GGBS in AAC mortars. The first study [27] investigated GGBS-DK precursor systems with varying proportions of DK ranging from 0% to 60%; it also used four different concentrations of alkali activators (sodium hydroxide, NaOH and sodium silicate, Na_2SiO_3). The results indicated that the maximum compressive strength achieved was 18.4 MPa at 40% DK content—considerably lower than the strength typically attainable with GGBS alone [6,9]. Additionally, the study also reported a drop in flowability and water absorption with increasing DK percentages. Microstructural analysis and morphology scans revealed the formation of C-S-H gel and geopolymer phases. Notably, incorporating up to 25% DK with the GGBS contributed to the development of a more intensified and homogeneous geopolymer matrix, characterised by the coexistence of amorphous N-A-S-H and C-(A)-S-H gels. However, further increase in the DK content beyond 25% led to greater structural heterogeneity, which therefore reduced the overall effectiveness of the geopolymerisation reaction [27].

In another study [26], GGBS was investigated with two different types of DKs: original DK (untreated) and a thermally modified DK subjected to an elevated temperature treatment of 1000 °C (FDK). Both DK types were used as GGBS replacement at percentages of 10%, 20%, and 30% to produce slag pastes. The results showed that the replacement of slag with DK caused a substantial reduction in compressive strength at 28 days, dropping from 42 MPa for the DK free mix to 26 MPa for the 10% DK sample. The strength degradation which occurred with the inclusion of 10% DK exceeded 60% of the 0% DK mix strength. Moreover, it also prolonged the initial setting time (from 58 min for DK free mix to 92 min) and increased the total porosity (from 23% to 35%). Conversely, the replacement of GGBS by 10% FDK (thermally treated DK) improved the compressive strength at 28 days by 9.5%, reaching 46 MPa. In addition, the setting time almost remained the same as the DK free mix, and the porosity was improved by about 3%. However, higher FDK percentages also showed an adverse effect on the compressive strength, setting time, and total porosity. These effects are likely attributed to the high silica content of DK, which comes at the expense of available calcium in the slag. Furthermore, the poor performance of the matrix with more DK or FDK in the system was attributed generally to the excess of Na_2O in the matrix (which comes from the activator) and the higher silica content, both of which delayed the condensation process and reduced the slag dissolution [26].

Two other studies [5,48], proceeded with the partial replacements of electric arc furnace slag (EAFS) by DK in AAC systems. In [5], four percentages of DK (10%, 20%, 30%, and 50%) were used as EAFS replacement to produce geopolymer pastes. Results of compressive strength at 90 days showed an increase in compressive strength with 30% DK replacement from 17.6 MPa (0% DK) to 22.9 MPa. Incorporation of DK in excess of 30% caused a dramatic reduction in compressive strength. An extensive material analysis study was performed (XRD, TGA/DTG, SEM-EDS), which showed that DK considerably improved the formation of gels such as CSH, CASH, and N(C)ASH, which are characteristic of alkali activation. This was attributed to the high content of reactive silica and the high reactivity of alumina present in DK. An extended study on DK and EAFS by the same research team showed better durability in terms of fire resistance of the same mixes [48]. The reported compressive strength at 1000 °C for the 30% DK mix increased to 44.7 MPa, indicating thermal stability. The same study also reported a good durability of this mix against seawater with a compressive strength improvement of about 3 MPa after 180 days of immersion in seawater. The authors explained that the reacted particles of DK act as synergetic agent filling the pores, which reduces the formation of microcracks in the matrix, thus preventing the penetration of chloride and sulphate ions. This contributed to the overall durability enhancement.

The above findings confirm DK as a viable precursor in AAC. However, its performance is highly dependent on the nature of the co-precursor, alkali concentration, and curing conditions. In MK-based systems, DK contributes positively to strength development up to an optimum dosage, likely due to its high amorphous silica content, which promotes the formation of N-A-S-H-type geopolymer networks. The optimal mechanical performance observed in DK-MK blends (30–40%) suggests the formation of a compact and well-connected gel matrix. At this ratio, MK provides sufficient reactive alumina to balance the Si/Al ratio, while DK contributes additional amorphous silica. SEM and EDX analyses from [27] confirm that DK-MK precursor systems result in a homogeneous and dense matrix at moderate DK contents, characterised by well-developed binding gels and low porosity. Alkali concentration is another key factor: low dosages lead to incomplete precursor dissolution, whereas excessive alkali may destabilise the gel structure. Strength gains and reduced porosity were consistently reported at moderate activator levels.

In slag-based systems, DK showed beneficial effects when added at 25–30%, likely due to its filler role and contribution to gel development. However, higher DK contents appear to disrupt the Ca/Si balance necessary for robust C-(A)-S-H formation, leading to strength loss and delayed setting. Based on DTA, FTIR, and SEM, study [26] demonstrated that excessive untreated DK resulted in increased porosity and lower C-S-H/C-(A)-S-H gel development. Conversely, thermal activation (FDK) improved the performance of DK at lower dosages (e.g., 10%), enhancing strength and hydration, likely due to increased silica reactivity. Similarly, in EAFS-based binders, moderate DK additions improved matrix densification and durability, presumably through secondary gel formation and refined pore structures, a behaviour also observed in SCM systems. The compatibility of DK with co-precursors therefore depends strongly on precursor chemistry: [27] highlighted the importance of achieving balanced Na_2O/Al_2O_3 and SiO_2/Al_2O_3 ratios in DK-MK-slag blends to optimise strength and densification, while [26] found that excessive DK in calcium-rich matrices compromised C-S-H/C-(A)-S-H development. This indicates that DK performs more favourably in low-calcium systems (e.g., MK or WTS), where its silica integrates into aluminosilicate networks. In high-calcium systems such as GGBS or EAFS, its use must be more controlled, either by limiting dosage or using thermal pretreatment.

Overall, these findings highlight the need to balance reactive silica and calcium availability while tailoring activation regimes, ensuring efficient precursor dissolution and gel formation. DK is more compatible with low-calcium aluminosilicate systems, where excess silica promotes N-A-S-H formation, rather than in calcium-rich systems, where unreacted silica may interfere with C-(A)-S-H unless DK is carefully dosed or pretreated.

4. Discussion

This review has synthesised the current state of research on dealuminated metakaolin (DK) as both a supplementary cementitious material (SCM) and a precursor in alkali-activated cement (AAC) systems. While the number of studies remains limited, the evidence demonstrates that DK is a potentially viable material with unique characteristics. Its high amorphous silica content, moderate alumina, and high specific surface area make it a reactive material capable of contributing to both pozzolanic and geopolymerisation reactions. In terms of innovation, DK offers an opportunity to valorise a by-product of alum production that would otherwise be landfilled, thus contributing to circular-economy goals.

Despite these promising results, several challenges remain. First, the current number of studies is very limited to give confidence in the industrial deployment of this material. These studies often are not exhaustive, and therefore very considerable gaps remain regarding the behaviour of DK in mortar and concrete mixes and the optimal mixes designs for the industrial usage of cement systems with DK; for example, in the reviewed literature, a number of studies relied on single alkali activator dosages or curing regimes, limiting our understanding of the behaviour of DK under varied processing conditions. Moreover, few studies exist to validate long-term durability and performance under real-environment conditions. Furthermore, systematic comparisons between preparation methods and different DK sources are lacking, making it difficult to establish general design guidelines, a challenge commonly encountered when attempting to valorise waste materials for their use in concrete applications.

The body of research remains fragmented and lacks depth in key insights of underlying processes. For instance, the variability in the effects of DK on setting time, workability, and hydration reactions reflects an incomplete understanding of the interaction of DK with cementitious matrices. Few studies only (e.g., [26,29,45]) attempted to explore the underlying chemical processes, such as how DK alters gel formation, ion mobility, or pore connectivity, which are crucial for performance prediction and mix design. This review has tried to offer plausible interpretations, but dedicated material analysis of the cement systems with DK will be required to confirm the underlying processes and their effect on the concrete and mortar properties.

The above factors contribute to the current absence of DK from international cement standards such as EN 197-1, which currently excludes materials without robust performance data and standardised testing methods. This is a clear barrier to the industrial adoption of DK in cement systems for concrete and intensive work is required to overcome this obstacle to commercialisation. Additionally, available production and usage data for DK are regionally concentrated and often inconsistent, complicating global extrapolation and creating uncertainties about supply in quantities required for concrete, the second most used material in construction after water.

Moreover, an overlooked aspect in the literature and a potential limitation for the use of DK in structural concrete involves the residual acidity of DK, due to acid-washing during alum production. Thus, if used in high proportions, DK can lower pore solution pH, compromising the protective passive layer on steel reinforcement (requiring a pore solution pH > 12.5) and increasing corrosion risk. Therefore, DK may require neutralisation for use in concrete systems, including concrete with AAC.

From an environmental perspective, the potential for CO_2 reduction through partial cement replacement with DK remains underreported in the literature. Considering that DK is a by-product of the alum industry, its environmental burden would be small, as its footprint would be mostly linked to post-processing (e.g., drying, grinding, transport, etc.). For the sake of illustration, an estimated CO_2 emission of ca. 50 kg CO_2/tonne DK will be assumed, based on typical figures reported for the energy usage linked to the drying and grinding of other recycled cementitious materials [49] and road transport [50]. Assuming a 10–15% replacement of Portland cement with DK, which aligns with the optimal mechanical performance range observed in most reviewed papers in the literature, 10–15% lower PC contents could be used. Considering that the average CO_2 emission from manufacturing 1 tonne of Portland cement is about 850–900 kg CO_2, a 10–15% DK use would potentially translate to an estimated saving of 80–130 kg CO_2 for every tonne of cement replaced. However, if DK requires post-processing such as neutralisation with lime or NaOH to mitigate acidity before its use in reinforced concrete applications, additional emissions must be considered. Lime production, for instance, is reported to emit >1.0 per tonne of lime produced (1.0–1.8 tonnes of CO_2, depending on the kiln type [51]); thus, even small additions for pH adjustment (e.g., 5–10 wt.%) can increase embodied carbon of DK by >50–100 kg CO_2 per tonne. On the other hand, if washing is employed as an alternative treatment, it may not introduce additional environmental burden, since DK is typically washed prior to disposal in landfills [20]. In such cases, redirecting DK to concrete applications could offer a path towards more sustainable use without requiring extra water or processing beyond what is already standard practice. Nevertheless, a full life cycle assessment (LCA) is required to confirm the net environmental benefit of using DK in cement systems for concrete.

In summary, DK presents a promising, underutilised material with demonstrated reactivity and performance benefits in both SCMs for PC and AAC systems. However, to enable broader adoption, further research is needed to define optimum mix designs, understand long-term durability, establish reactivity benchmarks, and assess environmental impacts. Addressing these challenges through coordinated research will be key to unlocking the full potential of DK in SCM and AAC systems for mortar and concrete.

Table 1. Mix Design and key findings of DK utilised as SCM in Portland cement systems.

Ref.	Precursor	Mix Design	Key Findings
[36]	Cement + Two types of DK	Cement/Concrete + Two Types of DK: DK replacement by 10% and 15%; Cement content: 370 kg/m^3 and 415 kg/m^3; w/b: 0.43; Superplasticizer: 1% to 1.4% of binder.	- Improved compressive strengths (CS) or remaining unchanged at both 7 and 28 days. - The flow is reduced because of the DK's fine particle size. - 10% DK mortar complies with 52.5R strength requirement EN 197-1. - Penetration of water under pressure was less than 20 mm. - No workability loss was recorded.

Table 1. *Cont.*

Ref.	Precursor	Mix Design	Key Findings
[35]	Cement + DK + Nano Carbon referred to as NCDK	Cement Mortar with: *w*/*b* ratio: 0.43; Sand to binder ratio: 2.25; DK replacement by 5%, 10%, 15%, and 20%; Then DK is replaced by NC: 1% to 3%; Water-reducing agent is used.	- DK has a very high surface area 20 m^2/g. - NC improves CS for all mixes by approximately 3%. - 10% NCDK mortar increases CS by 14%. - Flow is reduced for all NCDK. - Setting time is decreased by up to 50% with more NCDK. - More CSH gel is formed when NCDK is used.
[43]	Cement + DK + TiO_2	Cement Pastes with: *w*/*b* ratio: 0.3; DK replacement by 10%, 30% and 50%; Another mix is prepared with 50% cement, 45% DK, and 5% TiO_2; Superplasticizer: 0.45% to 1%.	- DK samples showed lower early-age CS compared to the control. - 10% DK sample showed 16% enhancement in CS. - Other DK samples (30% and 50%) showed a reduction in CM. - Samples with 5% TiO_2 and 45% DK enhanced the CS by 50% compared to samples without the TiO_2. - 5% TiO_2 scans shows more CSH gel, which strengthens the microstructure of mix.
[28]	Concrete (cement + DK)	Concrete with: Two cement grades: 42.5 N and 52.5 N; Cement content: 350 Kg/m^3 and 400 Kg/m^3; *w*/*b*: 0.45; DK replacement/addition by 5% to 15%.	- All samples, in general, showed a significant increase in CS.
[39]	Cement + DK	Cement Mortar with: *w*/*b* ratio: 0.65; Sand-to-binder ratio: 2.75; DK replacing sand by 0% to 22.5% by 2.5% interval.	- Inclusion of DK up 17.5% enhanced the CS up to 46%. - Inclusion of more DK than 17.5% caused a drop in CS. - No significant effect of setting time is observed. - Inclusion of DK reduced the followability significantly.

Table 2. Mix Design and key findings of DK utilised in alkali activated cement/geopolymer systems.

Ref.	Precursor	Mix Design	Key Findings
[16]	MK + DK.	Metakaolin mortar characteristics: NaOH activator is used described as: $Na_2O/Al_2O_3 = 1.2$; $H_2O/Na_2O = 10$. DK is introduced as additive starting from 20% to 70% of the MK by 5% intervals.	- DK XRD shows amorphous peaks of quartz, anatase, and sillimanite. - The inclusion of DK causes an increase in CS up to 40%. - More DK than 40% causes a reduction in CS.
[18]	MK + DK	Metakaolin mortar characteristics: NaOH solution 50% concentration; Solution to precursor = 50%; Water Content: addition by 15%, 17.5%; DK is introduced as an added percentage starting from 0% to 60% of the MK by 10% intervals.	- DK reduced the flowability, and the initial setting time. - The addition of DK increased both 3 days, and 7 days CS up to addition percentage of 50%. - More DK to the matrix causes a significant drop in CS.

Table 2. *Cont.*

Ref.	Precursor	Mix Design	Key Findings
[26]	GGBS + DK GBFS + FDK	Slag paste characteristics: *w*/*b* ratio: 0.25; NaOH wt.%: 3%; DK and FDK replacement by 10%, 20% and 30%.	- Inclusion of DK caused a substantial reduction in CS reaching 60%. - 10% FDK sample showed an improvement in CS (9% enhancement). - Other FDK samples (20% and 30% reduced the CS. - DK and FDK prolong the setting time and cause a significant delay. - DK and FDK increase the porosity.
[27]	GGBS + DK	Slag mortar characteristics: Sand ratio: 2.75; a/B: 50% (including water in solution); Activator: 52% NaOH solution, waterglass; NaOH:Na_2SiO_3 = 1:0, 1:2, 1:2, and 1:3; DK replacement percentage starting from 0% to 60% by 10% intervals.	- More DK content causes an adverse effect on flow and setting time. - More DK content led to higher CS reaching maximum of 18 MPa compared to control mortar (4 MPa). - Scans show the formation of CSH and geopolymer in the mixes. - 25% DK mortar resulted in the formation of gels: NASH and CASH. - Increasing DK up to more than 25% increased the dissimilarity.
[5]	EAFS + DK	Slag paste characteristics: *w*/*b* ratio: 0.21 to 0.24; Activators: waterglass + pellets NaOH; SiO_2 = 0.5 mol/kg on all of mixes; Na_2O = 0.5, 0.75, 1, 1.5 mol/kg; DK replacement by 10% to 50%.	- Increasing the DK content has led to a significant increase the in the CS (more than 30% enhancement). - More DK than 30% causes a reduction in compressive strength.

5. Conclusions and Future Directions

This review consolidated the current understanding of DK, a poorly characterised hazardous by-product of alum manufacturing, and evaluated its potential for use in cementitious systems. While related materials like MK have received considerable attention, DK remains largely absent from scholarly discourse despite its likely widespread production. This paper addressed this gap by performing a systematic investigation of studies on DK valorisation, with a focus on its use in cement, mortar, and concrete, while identifying key knowledge gaps and barriers to adoption.

A structured protocol of filtering existing literature was followed to extract relevant data related to DK, analyse the extracted information, and categorise it. The review began by describing the formation of DK in the aluminium sulphate industry and its hazardous characteristics; namely, high acidity and ultrafine particles, highlighting its environmental and health impacts. It then presented a comprehensive analysis of DK used as an SCM in PC or as a precursor in AAC/geopolymer systems, including hybrid combinations with other aluminosilicate precursors such as metakaolin, slags, and water treatment sludge (WTS).

The key contribution of the paper lies in the comprehensive scope of the review and the first-of-its-kind synthesis, integrating DK origin, environmental hazards, physicochemical properties, pozzolanic behaviour, and performance within both SCM and AAC, towards potential commercial valorisation of this hazardous waste. By classifying performance trends across different SCM-PC and AAC binder systems, it provides a comparative framework previously missing in the literature and a foundation for future research.

The main findings on the feasibility of using DK in cements for mortars and concrete were that:

- 10–15% DK as SCM in cement systems enhances compressive strength by 15% to 30% but causes a dramatic reduction in flowability due to its fineness. Superplasticizers are therefore required in concrete mixes with DK used as SCM. Setting time was variably affected.
- DK used as SCM in cement systems led to enhanced concrete durability. However, further studies are still required to consolidate these findings.
- In AAC/geopolymer systems, DK performed best with MK or WTS but showed a poorer performance when partially replacing GGBS. With EAFS, late strength gains were observed, particularly after seawater exposure. In general, the available studies on DK lack diversity in mix designs, particularly with respect to AAC systems, and suitable AAC systems are yet to be developed for a consistent performance in terms of strength.
- AAC mix designs with modest DK contents were shown to enhance concrete durability, which concurs with findings using DK as SCM and aligns with microstructural observations.

Overall, the reviewed literature highlighted the promising technical performance particularly in terms mechanical strength and durability, when DK is incorporated at moderate levels. However, the current body of work remains fragmented, with incomplete understanding of chemical and physical mechanisms underpinning the behaviour of DK in cements for concrete, which is still subject to ongoing study. More detailed studies are needed to clarify reaction kinetics, gel formation, ion mobility, and pore structure evolution, and how these influence performance across different binder systems.

From an environmental perspective, the use of DK has the potential to significantly reduce cement-related CO_2 emissions. Replacing 10–15% of Portland cement with DK could reduce emissions by an estimated 80–130 kg CO_2 per tonne of cement. However, the actual benefit depends on processing assumptions and could be offset if DK requires neutralisation (e.g., with lime or NaOH) to mitigate acidity, particularly for use in reinforced concrete. Detailed life cycle assessments (LCA) are essential and need to be performed to determine the net environmental benefits.

DK is not currently included in cement standards (e.g., EN 197-1) likely due to its variable composition and limited performance data. Differences in DK particle fineness, amorphous content, and acidity across production sites present a major challenge for standardisation. However, these barriers could be overcome through better classification, much like the acceptance pathway for other SCMs or AAC precursors.

To support the broader adoption of DK in binder systems for mortars and concrete and translate laboratory demonstrations to industrial applications, future work should prioritise the development of standardised mix design protocols that can account for DK variability. More studies focusing on further mix design optimisation both in PC systems and AAC systems are required for further strength enhancement particularly towards structural concrete applications. This includes optimising dosage levels in both PC and AAC systems to ensure consistent strength and workability, particularly with respect to activator concentration, precursor combinations, and fresh-state properties. Importantly, the durability and long-term performance including reinforcement compatibility, chemical resistance, and dimensional stability of DK-cement systems in concrete applications must be thoroughly investigated under both accelerated and real-world conditions. For AAC systems, in particular, further investigation into reaction kinetics, pore refinement, and gel evolution (e.g., C-(A)-S-H, N-A-S-H) will be essential to clarify the link between mix chemistry and performance. These efforts should be guided by microstructural characterisation and benchmarked against relevant construction standards to support eventual qualification and code compliance.

Overall, the review indicated that DK offers a promising route for sustainable binder development, especially in combination with other SCM or precursors. Based on the quantities of concrete produced worldwide, the use of DK in concrete could outweigh the need for DK disposal, providing a useful outlet for this voluminous hazardous material, alternative to landfilling. However, realising this potential will require not just broader experimentation, but a shift in how such industrial by-products are classified, tested, and incorporated into construction norms. Robust life-cycle assessments, economic feasibility studies, and standardised performance validations are necessary to translate this potential into widespread commercial use.

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