

Editorial for Special Issue “Alkali Activation of Clay-Based Materials”

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1. Introduction

Low-carbon binders are the cornerstone of a sustainable society as they drastically reduce the carbon footprint of the construction industry. Among them, one of the most promising alternatives is alkali-activated materials (AAMs) [1]. Their synthesis requires combining solid reactive aluminosilicate precursors with alkaline-activating solutions [2,3]. Metakaolin is one of the most widely used precursors; however, its production relies on the calcination of kaolinitic clays at elevated temperatures (approximately 650–850 °C), making it a relatively energy-intensive and limited resource [4]. In addition, alkaline activators can represent a significant cost factor in AAM systems [1].

To address these limitations, increasing attention has been given to the use of lower-quality clay-based materials, which have recently successfully replaced metakaolin in AAMs [5], for example, claystones and shales [6,7], gravel wash mud [8], dredged river sediments [9,10] and carbonate-high illitic clays [11,12]. AAMs can be prepared in a highly sustainable way, using various aluminum- and silicate-rich industrial waste-based precursors, such as ashes from incineration plants [13,14], steel slags [15,16], red mud [17], paper sludge [18], mine tailings [19], and waste ceramics [13]. Moreover, alkali activators can be produced from industrial wastes [20]. Even when the energy used for producing alkaline activators and elevated-temperature curing is taken into account, the CO₂ emission is still 9% lower than for OPC binders [21].

The products that form in alkali-activated mixtures primarily depend on the molar ratio of Si:Al in the system. Specifically, at a molar ratio of Si:Al > 1.5, alkali-activated gels form, while at lower molar ratios, zeolites may crystallise. In alkali-activated binders, clay minerals can be combined with Ca sources (such as slags, limestone, etc.), resulting in the formation of ordinary Portland cement (OPC) and hybrid blended cement phases [22]. The reactivity of clay minerals in alkali-activated binders primarily depends on their layer structure and morphology and can be enhanced by different approaches, such as calcination, acid washing, or mechanical treatment. Optimal calcination temperatures differ among clay minerals and within individual mineral categories, depending on interlayer cations and structural order, which creates difficulties in optimising the calcination temperature of multi-mineral clays at an industrial scale [23]. Moreover, the broader alkali activation of clay-rich materials requires further in-depth investigation of several important aspects, such as workability, volume stability, mix design, and admixtures [23].

This Special Issue, “Alkali Activation of Clay-Based Materials”, presents four original scientific papers on the conventional alkali activation of clay-based materials and two dealing with the alternative chemical activation of clays. Together, these studies present



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recent advances in alkali activation and alternative activation strategies for clay-based and aluminosilicate materials, with a strong focus on sustainability and the valorisation of industrial by-products. The contributions cover a wide range of precursors, including clayey soils, brick waste, mining tailings, and industrial residues such as slag, fly ash, glass wool, rock wool, and metakaolin, demonstrating the versatility of alkali activation in producing functional materials from secondary resources. Several studies further explore strategies to enhance performance, including microwave irradiation for rapid reaction control, the optimisation of precursor particle size and calcium hydroxide content, and the use of alkaline additives and surface modification techniques to improve mechanical properties and durability. In addition, the scope extends beyond conventional alkali activation to include the chemical activation of clay minerals in drilling fluids and the thermochemical disintegration of clay-rich materials, thereby broadening potential applications in geotechnical and industrial fields. Collectively, the reported research highlights how tailored activation approaches can effectively control microstructural development and macroscopic performance, enabling the design of more durable, resource-efficient, and application-specific clay-based materials that support circular economy principles and sustainable engineering practices.

2. An Overview of the Published Articles

The alkali activation of clay-based materials was studied from two main perspectives: precursor type and methods for improving the properties of clay-based alkali-activated materials. From the precursor perspective, this Special Issue presents novel formulations based on clayey soils [24], brick waste [25], gold mine tailings [26], slag, glass wool, rock wool, fly ash, and metakaolin [27]. Regarding property enhancement, the contributions explore several advanced approaches, including (i) microwave irradiation [27], (ii) alkaline chemical additives [24], and (iii) the optimisation of precursor particle size and calcium hydroxide content [25].

Hoch et al. [26] (contribution 1) reported the stabilisation of gold mine tailings using alkali-activated binders based on sugar cane bagasse ash and hydrated eggshell lime precursors, with sodium hydroxide as the activator. The alkali-activated systems exhibited significantly improved compressive strength compared to untreated material, reaching 2.3 MPa, while stabilisation with OPC achieved 5.3 MPa. The study demonstrates a sustainable alternative binder system with potential for large-scale applications.

Tesovnik and Horvat [27] (contribution 2) demonstrated that low-power microwave irradiation can rapidly and effectively immobilise ongoing chemical reactions in AAMs through complete dehydration, offering a fast, solvent-free alternative to conventional methods for stopping reactions. Using various precursors, including slag, fly ash, glass wool, rock wool, and metakaolin, microwave treatment successfully removed water without significantly altering the mineralogical structure while also suppressing efflorescence development during storage. Microwave-induced pore morphology was further identified as a potential indicator of curing completion, distinguishing between fully and insufficiently cured AAM systems. Although porosity increased and compressive strength decreased due to crack formation, the approach provides important opportunities for post-characterisation preservation and the development of porous alkali-activated structures for filtration and adsorption applications.

Medeiros-Junior et al. [25] (contribution 3) investigated the alkali activation of clay brick waste, focusing on the influence of precursor particle size and calcium hydroxide content on material properties. The microstructure, compressive strength, and rheological properties of the synthesised material were evaluated. Reducing particle size improved mechanical performance and reduced yield stress, plastic viscosity, and hysteresis area.

In contrast, the addition of calcium hydroxide accelerated geopolymerisation, enhanced compressive strength, and reduced workability over time. The study provides practical guidance for optimising the processing and performance of alkali-activated systems.

Karozou et al. [24] (contribution 4) addressed the high absorbency of clay-based mortars, which can lead to matrix degradation. To mitigate this, mechanical properties were enhanced using the highly alkaline potassium metasilicate activator, while durability against water ingress was improved through hydrophobic treatments. Two different clayey soils were investigated. The alkali activator improved mechanical performance and stability in both cases, while nano-clay significantly reduced capillary absorption, particularly in the soil with higher SiO₂ content. Acrylic emulsion further enhanced overall durability for both materials.

Besides conventional alkali activation, this Special Issue also covers the chemical activation of clay materials in drilling fluids used during borehole construction [28] and the thermochemical disintegration of clay-based materials in alkali solution [29], providing fundamental insights into the transformation of aluminosilicate materials. Papadimitriou et al. [28] (contribution 5) demonstrated that Greek palygorskite, when chemically activated with alkaline MgO, can effectively improve the rheological and filtration properties of water-based drilling fluids under high-salinity and high-hardness conditions, highlighting its potential as a sustainable alternative to conventional bentonite systems. In parallel, Akhmediyeva et al. [29] (contribution 6) explored solvothermal disintegration in alkali solution at elevated pressure as an activation strategy for clay- and mineral-based materials, leading to structural decomposition, phase transformation, and increased material reactivity. Together, these contributions illustrate how different activation approaches involving alkali addition can be used to tailor the physicochemical and functional properties of clay-based materials for a wide range of industrial and engineering applications.

3. Conclusions

This Special Issue presents recent developments in clay-based materials and their activation through chemical, thermal, and physico-chemical methods, highlighting their increasing significance in sustainable materials engineering. The contributions demonstrate the versatility of clay-rich and aluminosilicate systems, which, with appropriate activation, can be transformed into functional materials with tailored properties for construction, geotechnical, and industrial applications. Particular emphasis is placed on the use of secondary raw materials and industrial by-products as precursors, supporting the transition towards more resource-efficient and circular material flows.

The studies in this Special Issue also show how material performance can be significantly influenced by processing strategies such as particle size optimisation, alkaline activation chemistry, and innovative techniques including microwave irradiation and thermochemical treatment. These methods enable improved control over reaction pathways, microstructural evolution, and macroscopic behaviour, thereby expanding the practical applicability of clay-based systems beyond traditional uses. Moreover, the extension of activation concepts to drilling fluids demonstrates their applicability both as support materials for construction processes and as functional products used in construction applications.

Overall, this Special Issue underscores the importance of continued research into the fundamental mechanisms and practical implementation of activation processes. By bridging materials science, environmental considerations, and engineering practice, the works presented contribute to the development of more adaptable and sustainable mineral-based materials while opening new directions for their use in both established and emerging technological fields.

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List of Contributions

1. Zakharia Hoch, B.; Tonini de Araújo, M.; Festugato, L.; Consoli, N.C.; Reddy, K.R. Mechanical and Microstructural Behavior of Mine Gold Tailings Stabilized with Non-Conventional Binders. *Minerals* **2025**, *15*, 995. <https://doi.org/10.3390/min15090995>.
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