

Alkali Activation of Glass for Sustainable Upcycling: An Overview

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Abstract

The recycling of glass presently poses several challenges, predominantly to the heterogeneous chemical compositions of various glass types, along with the waste glass particle size distribution, both of which critically influence the efficiency and feasibility of recycling operations. Numerous studies have elucidated the potential of converting non-recyclable glass waste into valuable materials thanks to the up-cycling strategies, including stoneware, glass wool fibres, glass foams, glass-ceramics, and geopolymers. Among the promising alternatives for improving waste valorisation of glass, alkali-activated materials (AAMs) emerge as a solution. Waste glasses can be employed both as aggregates and as precursors, with a focus on its application as the sole raw material for synthesis. This overview systematically explores the optimisation of precursor selection from a sustainability standpoint, specifically addressing the mild alkali activation process (<3 mol/L) of waste glasses. The molecular mechanisms governing the hardening process associated with this emerging class of materials are elucidated. Formulating sustainable approaches for the valorisation of glass waste is becoming increasingly critical in response to the rising quantities of non-recyclable glass and growing priority on circular economy principles. In addition, the paper highlights the innovative prospects of alkali-activated materials derived from waste glass, emphasising their emerging roles beyond conventional structural applications. Environmentally relevant applications for alkali-activated materials are reported, including the adsorption of dyes and heavy metals, immobilisation of nuclear waste, and an innovative technique for hardening as microwave-assisted processing.

Keywords: glass recycling; up-cycling; mild alkali activation; molecular mechanism



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1. Introduction

Glass has been a prevalent material in human history since approximately 3000 BC [1,2]. Its exceptional versatility can be attributed to several key properties, including optical transparency, chemical inertness, high intrinsic strength, and low permeability [3,4]. These characteristics enable a wide range of applications across various fields, from packaging to construction and technological innovation.

The glass manufacturing is a continuous process and can be classified into four macro-phases: batch preparation, fusion and refining, conditioning and moulding, and finishing [5]. Among these phases, the melting process is a particularly energy-intensive part, as it requires substantial energy input to achieve the elevated temperatures (up to 1500 °C) necessary for turning raw materials into glass. This energy is typically supplied through the combustion of natural gas [6].

The glass manufacturing process, as well as the glass industry, is characterised by high energy consumption, driven by the energy demand per tonne and the large-scale

production. Glass container production was recorded at 65 million tonnes [7,8], reflecting a broader trend of growth in global glass production, which doubled until 300 Mt from 2014 [9] to 2021 [10].

Glass manufacturing contributes to greenhouse gas emissions, with approximately 95 million tonnes of CO₂ released annually [11]. These emissions include those directly resulting from combustion of raw materials, the decomposition of carbonates, and the indirect emissions associated with electricity production [12–15]. To meet the emission reduction targets established by the Paris Climate Convention in 2015 [16], it is crucial to consider the entire recycling process.

Currently, the recycling rate of waste glass is not uniformly distributed, showing substantial variation at the international level [17,18]. The estimated worldwide glass recycling rate has been estimated at around 20% [19]. However, Belgium exemplifies best practices with near-total glass recycling, in contrast to Turkey, with a recycling rate of below 10% [20,21].

To comprehend the complexities of glass recycling, it must be realised that, despite its theoretical recyclability, glass cannot be fully recycled in practical scenarios. The implementation of a true ‘closed-loop’ recycling system faces several limitations. A ‘closed-loop’ system, in fact, is intended to convert raw materials into equivalent starting products with comparable quality and function [22].

The standard practice for glass recycling is remelting. In terms of energy efficiency, the addition of glass cullet, a granular material produced from recycled glass, results in a reduction of approximately 3% in energy required for melting. This reduction in energy consumption is primarily due to cullet melts at significantly lower temperatures than unprocessed raw materials [23].

Despite its prevalence in industrial practice, ‘closed-loop’ recycling of glass continues to be limited by several obstacles, which are generally divided into supply-chain barriers and technical limitations. A key limitation in the recycled glass supply chain is the absence of consistent global regulations that guide the glass recycling process. Furthermore, transportation costs associated with moving cullet to treatment and recycling plants, due to the dense nature of glass, may represent up to 33% of total recycling cost [9]. Therefore, despite the prevalence of glass collection worldwide, a significant amount of glass is still disposed of in landfills due to the inadequate facilities capable of carrying out advanced sorting and decontamination processes.

Technically, the integration of glass with diverse chemical compositions into a unified recycling stream presents notable difficulties. In fact, the thermal and processing properties of glass [24] vary considerably depending on chemical formulation. Nevertheless, the remelting of individual glass with the same chemical composition entails certain limitations that prevent total recyclability. Focusing specifically on boro–alumino–silicate glass, trace levels of polymeric and metallic contamination are sufficient to compromise its suitability for pharmaceutical applications. Similarly, the production of cathode ray tube glass (CRT) is discontinued, primarily due to the hazardous emission of lead oxide during the remelting processes. In the context of hazardous emissions, opal glass, used in tableware manufacturing, raises environmental health concerns due to fluorine emissions in high-temperature processing. The main commercial uses and disadvantages of different glass chemical compositions are reported in Table 1.

Table 1. Chemical composition, main commercial use, and limitation in recycling of the most common type of commercialised glass, reprinted from [25].

Glass Type	Composition [wt%]	Commercial Use	Limitations in Recycling
Sodalime (SLG)	73% SiO ₂ 17% Na ₂ O 5% CaO 4% MgO 1% Al ₂ O ₃	- Functional or decorative surface coating on plates (screen printing) - Containers	- Permanent fusion of oxides renders coating removal unfeasible - By-products from forming equipment (glass, graphite, carbon, and oil)
Boro–alumino–silicate (BASG)	80% SiO ₂ 13% B ₂ O ₃ 4% Na ₂ O 2.3% Al ₂ O ₃ 0.1% K ₂ O	Pharmaceutical packaging	- Presence of polymeric and metallic contaminations in the finest fraction
Cathodic Ray Tube (CRT)	63% SiO ₂ 21% PbO 7.6% Na ₂ O 6% K ₂ O 0.6% Al ₂ O ₃ 0.3% CaO 0.2% MgO 0.2% B ₂ O ₃	TV screen	- CRTs no longer manufactured - PbO emission occurs in remelting process
Opal	72% SiO ₂ 12% Na ₂ O 8% Al ₂ O ₃ 5% F ₂ 2% CaO 2% BaO 1.5% K ₂ O	Kitchenware	- F emission occurs in remelting process
Liquid-crystal display (LCD)	55% SiO ₂ 15% Al ₂ O ₃ 10% B ₂ O ₃ 10% CaO 5% SrO 1% BaO	Electronic device	- Incorporation of functional coatings and optical filters

Overall, an effective incorporation of glass scraps into a ‘closed-loop’ recycling framework [26] requires strict colour-based sorting and removal of residual impurities.

One significant reason why glass cannot be fully recycled is the issue of glass scrap dimensions (see Figure 1). At the recycling centre, glass is sorted into categories: glass cullet, also known as the coarse glass fraction, and glass fines. Glass cullet is suitable for remelting following the sorting operation. On the other hand, the smallest size fraction, defined as particles smaller than 100 µm, typically generated during the crushing process, contains a considerable level of impurities, which complicates traditional recycling methods [27].

However, even within the finest glass fraction, it is possible to identify two sub-fractions: a purified fraction and one with a higher concentration of impurities. The purified fraction of glass can be suitable for recycling, often through an ‘open-loop’ recycling process [28]. Conversely, the only viable option for managing the contaminated fraction is the disposal in a landfill [19], as the presence of impurities and contaminants can adversely affect the properties of the newly produced glass [29].

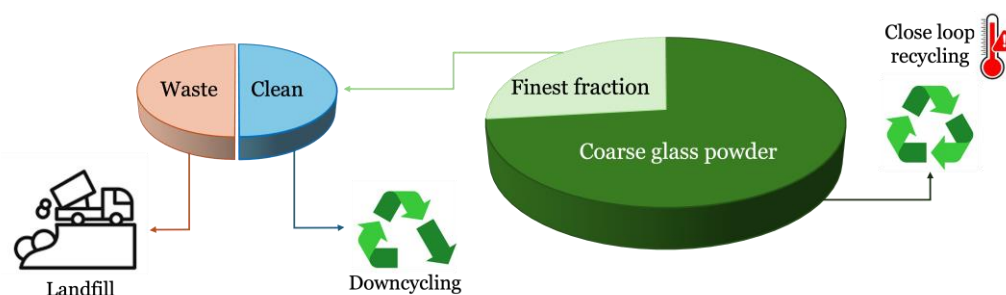


Figure 1. Current condition of glass recycling.

However, innovative recycling strategies have recently demonstrated the feasibility of processing glass materials categories that were considered incompatible with standard recycling practices. For example, to facilitate the development of a circular economy, SCHOTT company has initiated pilot-scale operation to explore the potential for reclaiming used glass-ceramic cooktop panel [Ceran®] and pharmaceutical packaging aimed at generating new products [30]. Ceran® glass is classified as lithium–alumino–silicate (LAS) glass-ceramics [31]. In fact, LAS glass-ceramics require higher processing temperature than typical glass bottles ($T_{\text{melt}} \approx 1500\text{ }^{\circ}\text{C}$) due to the melting point of neat $1650\text{ }^{\circ}\text{C}$ [32], making them incompatible with the standard glass recycling process. Moreover, the inclusion of LAS glass in the conventional glass recycling process contributes to highly viscous inclusions that compromise the quality of the recycled product [33]. For these reasons, waste LAS glass-ceramics are classified as non-recyclable materials, and typically landfilled [34]. Despite the advancement in recycling, the primary constraints for the material feedstock entering SHOTT pilot plants are the quality of the materials; in fact, the scraps must be free of contaminants and impurities. This requirement creates opportunities for technological innovation in the recycling of more heavily contaminated material fractions.

This work aims to provide an overview of the current state of the art concerning the use of glass fraction that cannot be directly recycled through the remelting process. Particular attention is given to the valorisation of glass waste in the field of alkali activation, initially examining its role as an additive in conventional binders (such as metakaolin) and subsequently exploring its potential as the sole precursor for the synthesis of AAMs. Emphasis is placed on mild alkali activation, molarities below 3 mol/L , and the molecular mechanism that governs this class of materials. Finally, the overview proposes alternative application areas that extend beyond the classical role of replacing conventional construction material, highlighting the versatility of alkali-activated glass-based materials.

Differences Between Waste Glass and Waste-Derived Glass

The discarded glass mentioned in earlier sections can be referred to as ‘waste glass’. Vitrification is considered a safe and efficient solution for hazardous waste treatment in the context of alternative waste management strategies [35]. Actually, various industrial waste streams can be vitrified individually or in combination with others through thermal treatment, ultimately yielding glass. During thermal treatment, waste is decomposed into a homogeneous melt, which must be quickly cooled to prevent crystalline phase formation [36].

To achieve a proper glass phase, the process fundamentally requires that initial waste materials include a minimum amount of SiO_2 . The final product is termed ‘waste-derived’ glass. Transforming waste into ‘waste-derived’ glass is beneficial, primarily due to high chemical resilience, which permits long-term containment and significantly reduces the volume of residual waste [37]. Despite the immobilisation of hazardous inorganic substances and the potential for disposal in landfills, vitrification has disadvantages, as it is associated

with financial and energy requirements. The potential release of hazardous gases, including chlorine, resulting from incomplete encapsulation of certain compounds in the glass structures, is a critical issue for this application [38]. An alternative approach to mitigate energy consumption is the conversion of ‘waste-derived’ glass into useful components that exhibit multifunctional properties. For example, waste-derived glass can serve as a raw material for glass fibres [39] or glass-ceramic tiles [40]. In addition to producing dense materials, researchers have also reported the transformation of ‘waste-derived’ glass into porous glass-ceramics [38].

The issue of recycling materials derived from the ‘waste-derived’ waste remains largely unresolved. The lack of a single raw material makes a ‘closed-loop’ recycling process unfeasible. In this context, alkali activation emerges as an innovative strategy for reusing ‘waste-derived’ glass precursors to synthesise materials under low-temperature conditions, bypassing conventional high-energy manufacturing steps [41,42].

2. ‘Up-Cycling’ of Glass: Current Applications

In recent years, there has been growing interest in recycling, accompanied by numerous efforts to enhance the value of scrap materials. Despite these advancements, many technologies and processes lead to materials degradation during recycling, a phenomenon known as ‘down-cycling’. While this approach contributes to mitigating waste buildup, it does not maximise the functional capabilities of the product. For glass specifically, the predominant form of downcycling consists of incorporating it as aggregate in road base layers or cementitious construction materials [43].

From a sustainability standpoint, ‘up-cycling’ emerges as a more attractive process. ‘Up-cycling’, as defined by Gunter Pauli, is a value-retention strategy whereby post-consumer waste or industrial by-products are upgraded into functionally valuable items [44–46].

A comprehensive summary of the principal products via ‘up-cycling’ of glasses is provided in the following subparagraphs. Each final product derived from ‘up-cycling’ exhibits a dual nature, comprising both beneficial aspects, such as the utilisation of materials that would otherwise be sent to a landfill, and negative attributes, primarily concerning energy requirements and potential environmental impacts.

2.1. Stoneware

Traditionally, stoneware is produced using more refractory clays and undergoes firing at high temperatures that can vary from 1100 to 1300 °C, contingent upon flux content [47]. The resulting bodies that are vitrified reach low porosity [48]. Due to its material flexibility, the ceramic industry represents an effective platform for converting waste into secondary resources, such as supplementary additives or auxiliary compounds [49]. By serving as alternative raw materials, glass scraps can be effectively recycled within the production of stoneware tiles, offering a sustainable waste management solution [50]. A wide variety of glass waste materials has been investigated [51]. As a part of a typical porcelain stoneware mixture, feldspar sand and traditional fluxing agents can be substituted by finely ground glass powder [52]. Glass powder, in fact, acts as an effective fluxing agent when incorporated into ceramic mixtures.

The incorporation of glass into ceramic formulations may notably reduce the temperature required for synthesis, reducing the temperature requirement below 1000 °C [50]. A fluxing agent, also known as a softening agent, is an additive used to reduce the sintering temperature in powder-based sintering processes, thereby contributing to energy saving. Moreover, during firing, glass powder enhances the densification process, resulting in a beneficial effect such as reduced open porosity and lower water absorption.

From an industrial point of view, large-scale glass recycling remains challenging: exceeding 10 wt% of waste glass typically results in reduced performance [53]. In fact, the use of glass powder waste may introduce drawbacks, such as increased dimensional shrinkage and elevated closed porosity. The high total porosity (total porosity > 13%) of the samples prevents them from being classified as stoneware [54].

2.2. Glass Wool Fibres

Due to their superior insulation characteristics, flame resistance, low production costs, and simple installation procedures, glass wool and stone wool are among the most widely utilised fibrous materials in building applications [55]. Glass wool consists of thin glass fibres arranged in a random and non-uniform pattern. During the formation process, these fibres enclose numerous voids, which reduces the material's density and enhances the thermal insulation capability by preventing natural convection [56].

The manufacture of glass wools typically utilises a mixture containing up to 85% waste glass (referred to as 'cullet'), while the remainder consists of mineral additives such as sand, limestone, soda, borax, and/or dolomite. However, in certain facilities, glass wool is produced using 100% waste glass, without the incorporation of other virgin raw materials [57,58]. During the melting stage, the raw materials blend must reach temperatures nearing 1400 °C for complete fusion to occur [59]. The glass wool melting process offers improved flexibility over traditional methods by accommodating larger volumes of impure recycled glass, including mixed cullet colours and organic materials, significantly advancing material recycling efforts [60]. The industry is continuously exploring technologies to reduce glass melting temperatures. Notably, the introduction of recycling glass waste helps decrease the energy required for the production of liquid melt [61].

2.3. Glass Foams

Foam glass represents an alternative method for recycling glass waste. Glass foams are cellular glass materials that fulfil strict requirements for fire resistance and structural durability [62]; the materials can be fully derived from glass waste, eliminating the need for virgin raw materials [63,64]. Although glass foams are predominantly utilised for thermal or acoustic insulation [65,66], recent studies highlight their applicability in environmental fields, particularly in filtering dyes and hazardous substances [67].

A gas-releasing additive, commonly referred to as a foaming agent, is frequently derived from finely milled carbon or carbon-based compounds [64]. The thermal treatment of glass powder and foaming agent mixture at temperatures ranging from 850 °C to 1000 °C, approaching the softening threshold of the glass, promotes the viscous flow and bubble expansion [68,69]. During the cooling stage that follows thermal processing, the gas bubbles stabilise and solidify into pores, leading to the development of the foam's characteristic porous architecture [70]. The formation of a uniform structure is governed by a delicate equilibrium between viscous flow and gas release. In the literature, foaming agents are usually classified into two types: redox agents and neutralisation agents [71].

Over 30% of studies related to redox agents focus on carbon-based materials [72]. Optimised foaming performance driven by oxidation of carbon-based agents necessitates sufficient oxygen from both the atmosphere and inter-particle spaces [73]. The available oxygen in the glass is influenced by several factors, including glass composition, presence and concentration of polyvalent ions, and their respective oxidation states. These ions are commonly employed to promote oxidation, with manganese oxide (MnO₂ or Mn₃O₄), iron oxide (Fe₂O₃), or cobalt oxide (Co₃O₄) [74] being among the most effective agents. However, incorporating these additives into glass-based foams increases the cost of production and requires careful control of glass release.

Silicon carbide (SiC) is assessed as an effective foaming agent for foam glass production, capable of producing homogeneous foams with controlled and precise cell sizes [75]. SiC exhibits a higher oxidation temperature than pure carbon, ranging from 950 °C to 1150 °C, which influences the melt viscosity. At elevated temperature, the melt viscosity is low, leading to rapid gas formation that can result in foam collapse [76]. For foams to be stable, it is essential to balance their viscosity with a uniform internal cell structure, ensuring uniform pore size and thin wall [77]. Incorporation of oxidation agents results in a decrease in foaming temperature while promoting greater gas evolution.

Carbonates decompose under thermal treatment, leading to the emission of carbon dioxide. Dolomite ($\text{CaCO}_3 \cdot \text{MgCO}_3$) and calcite (CaCO_3) are commonly employed as foaming additives in industrial processes. The involved reactions promote the decomposition of minerals, resulting in gas release [78]. The primary advantage of using carbonates is their role as fusion agents, reducing the sintering and foaming temperature [62].

In addition to conventional foaming agents, alternative compounds have been explored for their foaming capabilities. One such compound is aluminium nitride (AlN) powder. At elevated temperatures, the glass powder transitions to a viscous liquid state, during which AlN decomposes to produce gas that rapidly expands, resulting in the formation of bubbles within the glass melt. It is crucial for the glass to maintain an appropriate viscosity to facilitate this process [68]. AlN can function on its own or together with other additives, including TiO_2 or Fe_2O_3 [79].

The potential of a range of organic compounds as redox foaming agents has been assessed, showing their ability to create a consistent porous arrangement [80]. One notable advantage of utilising organic compounds is their lower decomposition temperature, which allows for earlier reactions that achieve the desired viscosity. Among these organic materials, glycerol is one of the most used foaming agents; however, other classes of compounds, such as fatty acids, surfactants, and polymers, are also utilised [81].

2.4. Glass-Ceramics

At first, the term 'glass-ceramics' referred to materials with more than 50% crystalline content, but over the years, materials with much lower crystal fraction have also been developed [82]. Materials first formed as glass and later crystallised into glass-ceramics using nucleating agents. Glass-ceramics are high-performance composite materials characterised by their superior mechanical properties, high thermal stability, excellent chemical resistance, and exceptional efficiency in heavy metal immobilisation [83].

The formation of glass-ceramic necessitates a supplementary thermal treatment. In fact, the nucleation stage is performed between 500 and 750 °C, while the crystallisation stage happens between 750 °C and 1000 °C, depending on the chemical composition of the starting glass [84].

To mitigate energy demand, a potential strategy is the utilisation of waste-derived glass. The conversion of waste-derived glass into glass-ceramics has revealed its potential for value-added applications [35,85]. Dense glass-ceramics suitable for building applications such as tiles [86,87] or face bricks [88] can be produced through this method. Additionally, porous glass-ceramics foams have garnered scientific interest for the application in thermal and acoustic insulation [65,89,90], as well as catalyst support [91]. These glass-ceramic foams exhibit excellent physical characteristics, which make them appropriate in the construction field [92]. The transformation of waste-derived glass with correct composition into glass-ceramics can be achieved thanks to the sinter-crystallisation technique [38]. The procedure involves the co-occurrence of glass particles melting, due to the viscous flow sintering, and the development of a crystalline phase.

2.5. Geopolymers

Inorganic polymers, also known as ‘geopolymers’, have demonstrated their potential in the civil engineering and building sector due to their environmental sustainability [93,94]. In fact, this class of materials acts as substitutes for cements, offering significantly lower CO₂ emissions and energy consumption, reducing consumption by 60–80% [95].

Since the 1970s, Davidovits [96] defined geopolymers as three-dimensional aluminosilicate structures formed through inorganic polycondensation, defined by the formula $M_n[-(SiO_2)_z-AlO_2]_n \cdot wH_2O$, where M is the alkali metal cation; n indicates the degree of polycondensation; and z can be 1, 2, or 3. However, Kriven et al. [97] revised the definition of geopolymers as inorganic poly silicate–alumino polymers or chemically bonded ceramics.

The two main components of geopolymers are a powdered alumino–silicate-based material and an alkaline activator. The geopolymerisation process consists of converting a significant amount of the amorphous alumino–silicate phase into a dense structure through polycondensation [98].

The properties of geopolymers are greatly influenced by the choice of raw materials. Metakaolin, the anhydrous calcinated form of the clay mineral kaolinite, is commonly used for its high reactivity. Nevertheless, a fundamental conflict exists between the growing need for sustainability in geopolymers and the energy-intensive thermal treatment required to produce them. As a response to this challenge, alternative aluminosilicate precursors, such as ashes and ground granulated blast furnace slag, have been explored. A novel approach to geopolymer production involves using industrial residues and waste materials.

Similar to the conventional concrete production, scrap glass is employed as an aggregate [99]. However, incorporating additional materials, as metakaolin, is essential to fulfil the specific structural needs of geopolymers. Particle size, chemical composition, and temperature play a key role in influencing the reactivity of glass in geopolymeric mixtures [100].

Despite these advancements, the current scientific literature lacks examples of entirely glass-based geopolymers. This indicates that glass recycling is still incomplete, as the presence of glass powder impacts phase development and the following curing times in geopolymer manufacturing. Thus, while waste glass powder offers potential benefits, its integration into geopolymer systems necessitates careful consideration of a complementary position to achieve optimal performance.

The use of glass as an alternative to natural limestone is investigated in the production of geopolymers [101]. However, glass replacement can reduce mechanical properties because of the poor bonding between the matrix and glass. Research has indicated that the optimal inclusion of waste glass should be around 25% [102]. Initially, glass powder acts mainly as a filler, but over time, it is involved in the development of calcium/sodium aluminate silicate hydrate gel (C/N(A)-S-H), which enhances the overall strength of the geopolymer matrix.

Finally, Torres-Carrasco et al. [103] investigated the potential application of urban-derived waste glass as an alkaline activator in geopolymers, specifically in the form of sodium silicate hydrate, or waterglass. Activators are essential in geopolymer production, providing high molarity that results in extreme causticity, high corrosiveness, and notable viscosity. These characteristics require careful handling and safety protocols. Additionally, the significant energy demand for producing these activators increases the cost of geopolymer technology, which may limit its application to small-scale projects [104].

To date, there are no reported examples of geopolymer systems in which the final products are composed solely of glasses, as these formulations consistently require the addition of other synthetic or natural components. The following section, which examines alkali-activated materials, closely related to geopolymers, highlights examples of systems

produced entirely from glass, whether specifically formulated or sourced from waste glass by-products.

3. Alkali Activation: Definitions, Mechanisms, and the Role of Waste Glass

Alkali-activated materials (AAMs) are becoming increasingly important in reducing CO₂ emissions, alongside geopolymers. While AAMs and geopolymers share similarities, they are distinguished by their aluminosilicate precursor composition and gelation mechanisms. The terminology surrounding AAMs and geopolymers is still evolving, and several studies have proposed different classification schemes. Alkali-activated materials, alongside inorganic polymers, are considered a broader category within which geopolymers are identified as a distinct subclass. From a chemistry-focused perspective, Van Deventer et al. [105] define alkali-activated materials as the result of a reaction between a calcium-rich aluminosilicate precursor and an alkaline source.

Provis, similarly, categorised alkali-activated material as high-calcium binders [106]. The calcium content influences not only the kinetics of the reaction but also the resulting gel chemistry and microstructure. In calcium-enriched systems, Ca(OH)₂ and other calcium species may interact with silicate and aluminate ions in solution. In the early stages, calcium silicate hydrate (C-S-H) forms, and further reaction with Al³⁺, Si⁴⁺, and Ca²⁺ ions with alkalis leads to the development of tobermorite-like calcium–aluminium–silicate hydrate (CaO·Al₂O₃·SiO₂·*n*H₂O, C-A-S-H) gel. In alkali-activated materials, these gel networks are characterised by tetrahedrally coordinated silicate chains with a characteristic pattern, comprising (3*n* – 1) tetrahedra. In composition with moderate calcium concentration, a hybrid gel structure may form, where both C-A-S-H gel and sodium–aluminosilicate hydrate (N-A-S-H) phases [107] have been identified. The variability in gel chemistry highlights the intricate structural nature of AAMs and demonstrates how their properties are strongly influenced by both the composition of the precursor and the conditions under which the activation occurs.

One of the most promising approaches in alkali-activated research involves the incorporation of waste materials or industrial by-products, notably waste glass. Waste glass can function as a supplementary filler or aggregate in alkali-activated materials, often in combination with primary precursors such as fly ash or ground granulated blast furnace slag.

Chemically tailored glass acts as a suitable feedstock for alkali activation and subsequent inorganic polymer formation.

An alternative strategy [108] focuses on converting aluminosilicate sources into reactive binders adapted for alkali activation, offering a further alternative to mere vitrification methods. This process, involving components as SiO₂, Al₂O₃, and Na₂CO₃, leads primarily to the formation of glass thanks to thermal treatment at 1250 °C. The obtained glass is activated thanks to a highly concentrated alkaline solution. The final materials reveal the presence of sodium aluminosilicate hydrate (N-A-S-H) gel, along with the development of secondary zeolitic phases. Despite their technical potential, such a method faces several critical challenges, including the reliance on high-purity reagents, the energy consumption associated with the melting and rapid cooling, and the environmental impact of using highly concentrated alkaline solutions.

To mitigate these limitations on synthetic precursor, the synthesis of compositionally tailored glasses has been refined by integrating naturally sourced materials, such as clay [109]. Nonetheless, the success of these systems remains dependent on careful selection and dosage of pure activators. Poorly chosen combinations may lead to the development of heterogeneous gel structures, where N-A-S-H gel is dispersed in C-A-S-H gel matrix.

Recent studies have proposed using waste glass as the sole precursor in AAM synthesis [110]. To the best of the authors' knowledge, in fact, Cyr and colleagues were the first to report the use of soda–lime waste glass as the sole precursor for the synthesis of geopolymer-like materials activated with diluted alkaline solution. Remarkably, these materials developed sufficient mechanical performance at moderate curing temperatures (40–60 °C) without requiring additional thermal treatment, offering a more energy-efficient and sustainable process. Nevertheless, the activation of soda–lime glass has a significant limitation due to its chemical composition, which hampers the formation of an appropriate zeolitic gel. Therefore, the term 'geopolymer' is also deemed inappropriate for these materials. Despite the variation in chemical composition, further studies conducted by Idir et al. [111] reveal that the optimal concentration of the activating solution is consistently found to be 3 mol/L, giving rise to the concept of weak or mild alkali activation. Despite these advancements, the molecular mechanism underlying hardening in mild alkali-activated materials remains poorly described. For this reason, the next section is intended to explore and address the existing gap in the current understanding and development of the molecular mechanism of mild alkali-activated glass. Briefly, weak alkali activation promotes the development of a gel layer on the particle interface, instead of total dissolution.

In parallel, Rincon et al. [112] proposed a dual approach that combines alkali activation, followed by sintering, to minimise the overall cost of glass-ceramic foams. The hardening of the suspensions is attributed to the formation of calcium silicate hydrate phases. Nonetheless, the produced porous materials exhibit chemical instability, necessitating thermal treatment. While this methodology has been extended to various glass types [113], the gel compounds are often difficult to identify. It is noteworthy that the requirements for thermal treatment, even at low temperatures, constitute a major drawback in terms of environmental sustainability [46]. Moreover, the use of highly concentrated alkaline solutions also represents a limitation in terms of ecological compatibility, in addition to increasing production cost for alkali-activated materials.

These considerations highlight the relevance of mild alkali activation, in which the alkaline solution remains below 3 mol/L, as a more sustainable path.

4. Weak Alkali Activation of Pharmaceutical Glass: Reaction Mechanism

The emergence of COVID-19 has led to a global pandemic. The global healthcare system faced major challenges, but coordinated action among industries and the scientific community enabled accelerated vaccine development and the implementation of an effective distribution framework [114]. The escalation in production has inevitably resulted in a corresponding increase in waste, highlighting the need for careful management of these by-products. Notably, the closed-loop recycling of pharmaceutical boro–alumino–silicate glass (BASG) containers requires exceptionally pure cullet for effective recycling processes.

The potential of employing BASG glass as a raw material for geopolymer synthesis was introduced by Taveri et al. [115]. Their selection is based on the chemical composition of glass residue, which contains critical species, as amorphous silica and alumina, essential for zeolitic network development. Additionally, the presence of boron oxide in the glass chemical composition can partially replace alumina, thereby promoting polycondensation reaction. Boron appears to serve an essential function in the geopolymerisation of BASG activated with a strongly alkaline NaOH solution (13 mol/L). In fact, the ^{11}B NMR spectra of the geopolymers show a notable shift in the relative peaks' intensity: the signal of BO_4 , at 1 ppm, persists, whereas the signal for the trigonal boron at 11 ppm has nearly disappeared, suggesting that trigonal boron dissolves and reorganises into a tetrahedral configuration, characteristic of zeolite gels. Unlike soda–lime glass systems, in boro–alumino–silicate glass,

the negligible CaO amount prevents the synthesis of calcium silicate hydrate compounds. In contrast to alkali activation of glass containing CaO [112,116], the calcium oxide facilitates the formation of binding products.

Rincon et al. [117] explored the alkali activation of BASG using mild chemical attack with alkali hydroxide activators not exceeding 2.5 mol/L, without incorporating any other silicate or aluminosilicate feedstock. The result was the formation of a complex hydrated gel, predominantly amorphous with a zeolite-like framework, incorporating alkali ions to balance charge.

Further experiments conducted by Metha et al. [118] focused on reducing the firing temperature of boroaluminosilicate glass to 550–650 °C and exploring complete avoidance of the thermal stage. The incorporation of surfactants significantly affected the nature of the gel phase. Denser pieces from the hardening of suspensions without surfactants remained intact after the boiling test; however, the foams disintegrated. This suggests that alkaline ions in the foams contributed beyond the stabilisation of the boron and aluminium tetrahedral species in the gel matrix, but actively contribute to the development of a highly depolymerised network, constraining the formation of strong Si–O–Si bonds. On the other hand, in compact cold-consolidated materials, the reduced incorporation of alkali ions, caused by hydrated alkali carbonates development, favours a more cross-linked network, strengthening glass particles' bonding and improving the integrity of the final product.

Tameni et al. [119] provided significant advancements in understanding the molecular mechanism of alkali activation of boroaluminosilicate glass with mild alkaline solution (NaOH/KOH, 2.5 mol/L). The NMR analyses [Figure 2] indicate that, despite the process of alkali activation and potential boiling, there are no significant alterations in the ^{29}Si , ^{27}Al , and ^{11}B MAS NMR spectra. In fact, the alkali activation of BASG using diluted solutions should not be equated with the geopolymerisation process. Boron and aluminium oxides in the glass inhibit the formation of a zeolite-like gel. According to Taveri et al. [115], a stable gel network with a notable increase in BO_4 units can only be achieved under specific activation conditions, such as highly concentrated alkali solution and additives containing a high level of Al_2O_3 .

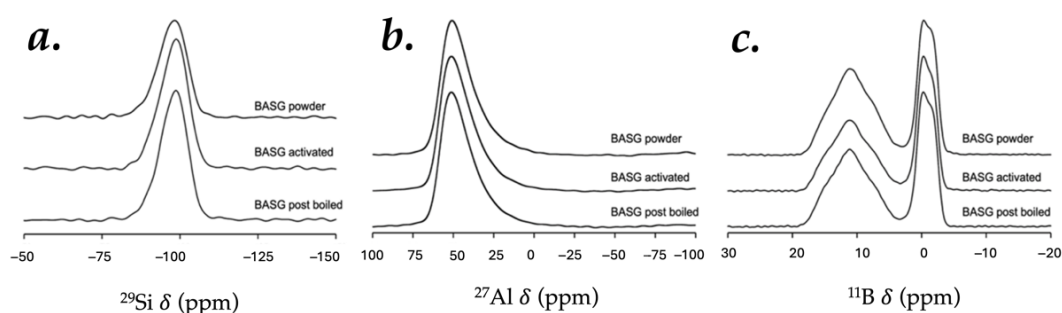


Figure 2. NMR spectra of (a) ^{29}Si , (b) ^{27}Al , and (c) ^{11}B of BASG in as-received, alkali-activated, and boiled conditions. Adapted from [119], Elsevier, 2025.

Structural characterisation using nuclear magnetic resonance, in conjunction with X-ray diffraction (XRD) and Fourier Infrared Spectroscopy (FT-IR) analyses, elucidated that the transition from the 'as-received' glass to alkali-activated glass consists of a cold-consolidated glass network, a gel phase, and alkali carbonates. Notably, the boiling test effectively removed the soluble phases, leading to the reversion to the original phase structure. NMR spectroscopy shows that alkali activation of BASG induces chemical changes only at the particle surface, while the interior of the glass remains unchanged, even after exposure to boiling conditions.

In other words, mild alkali activation of glass suggests a behaviour more closely aligned with corrosion phenomena, a concept that has been extensively documented in the context of nuclear waste [120–122]. The mild alkali activation primarily affects the surface layer of the particles, avoiding substantial dissolution experienced in geopolymers. However, in mild alkaline solutions, glass components released are considered minimal compared to the surface hydration of powdered particles, a phenomenon that leads to the formation of hydroxyl groups through bond cleavage of strong bonds (Si–O–Si, Si–O–Al, and Si–O–B). The condensation of reactive surface groups on adjacent particles facilitates the rebounding. This progressive polycondensation process ends with the formation of a ‘skeleton’ network of glass particles, with interstitial regions occupied by the secondary phase.

The secondary phase consists of gel, carbonates, and eventually other crystalline phases. The amorphous or semicrystalline gel is generated for partial dismantling of the glass network, comprising borates, silicates, and aluminates. The stabilisation of this gel is contingent upon the activation conditions and the interplay between the chemistry of the glass and the activating agents. Simultaneously, carbonates result from the interaction between alkali ions and CO₂ from the atmosphere. However, only strong bonds forming the primary phase are responsible for the stability of mild alkali-activated glass, as shown in Figure 3.

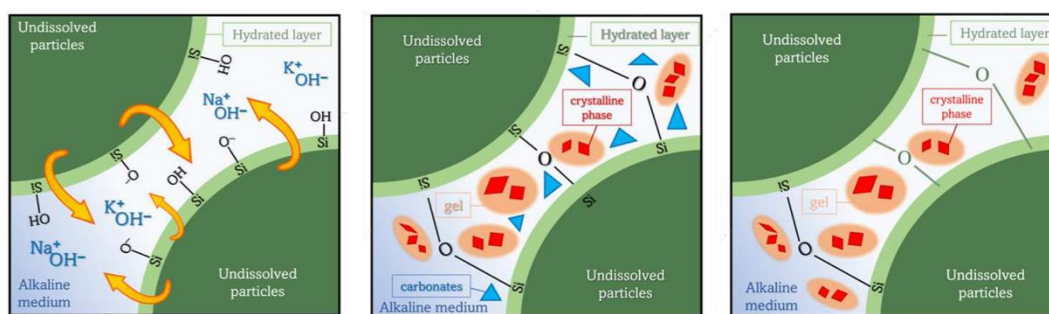


Figure 3. Scheme of alkali activation and cold consolidation.

5. Perspective of Glass Mild Alkali Activation

Beyond presenting the chemical molecular mechanism governing these materials, this section aims to offer new insights into the potential application of mild alkali-activated materials. Progress in using alkali-activated materials (AAMs) to replace traditional cement-based materials in construction and other applications has experienced significant growth in recent years. This section emphasises the innovative applications that are derived from the alkali activation of waste glass.

5.1. Alkali Activation of Glass as Building Materials

Waste glass has been utilised as a partial substitute for other aluminosilicate precursors, such as metakaolin, fly ashes, and ground granulated furnace slag [123–125] in the production of alkali-activated mortars in the building materials context. Nevertheless, an increasing number of studies have focused on the activation of glass as a standalone material.

Redden et al. [126] elucidated the potential for the alkaline activation of soda–lime glass, using an NaOH activating solution (4–8 mol/L), with compressive strength values exceeding 20 MPa regardless of the molarity. Interestingly, prolonged curing durations and high temperatures adversely affect the compressive strength of the final products.

Also, Torres-Carrasco et al. [127] explored the use of soda–lime glass produced by industrial and urban waste treatment plants as a precursor for alkali-activated materials, using various highly concentrated alkaline solutions, including NaOH, NaOH/Na₂CO₃,

sodium silicate hydrate, and KOH. Their studies demonstrated that the compressive strength varied significantly, achieving values between 17 MPa and 88 MPa, based on the selected activator and the applied curing methods, indicating the complexity and tunability of glass activation chemistry.

As previously reported, Cyr et al. [110] developed alkali-activated mortars using soda-lime glass powder as the primary binder, employing NaOH and KOH as alkali activators at different concentrations (1, 5, and 10 mol/L). Their findings demonstrated that waterglass was not required for the setting and hardening of the AAMs, and the optimal alkali content is fixed at 5 mol/L. In a subsequent investigation [111], the same authors demonstrated that the maximum compressive strength, which reaches up to 50 MPa, was achieved with alkali-activated materials using KOH at a concentration of 3 mol/L, reinforcing the viability of ‘mild’ alkali activation.

More recently, boro–alumino–silicate glass is also eligible as a raw material for construction and building alkali-activated materials. Tameni et al. [128] demonstrated that boro–alumino–silicate glass (BASG) can undergo alkali activation (NaOH:KOH, 2.5 mol/L) and cold consolidation by maintaining a temperature of 40 °C for 7 days, yielding compressive strength higher than 22 MPa and a geometrical density of approximately 1.7 g/cm³. These values place these materials in the range of commercial Plaster of Paris or concrete (structural lightweight).

In an effort to evaluate the robustness of this approach, the same authors also applied it to glass fractions characterised by varying degrees of contamination, including those that have come into contact with oils (BASG_dirty) or grinding machinery (BASG_waste). Through the use of a commercial materials selection software (Ansys Granta EduPack version Ansys 2024 R1, Ansys Granta, Canonsburg, PA, USA), Figure 4 presents a comparative evaluation of new alkali-activated materials and conventional building materials. In the context of construction, Ashby’s plot compares the density and the compressive strength to obtain the optimal trade-off [129]. BASG_waste (represented by ‘blue item’) overlaps with structural lightweight concrete. Moreover, the BASG_clean and BASG_dirty (‘green item’ and ‘orange item’, respectively) exhibit lower density and a comparable range of compressive strength of structural lightweight concrete, making them particularly promising solutions as building materials.

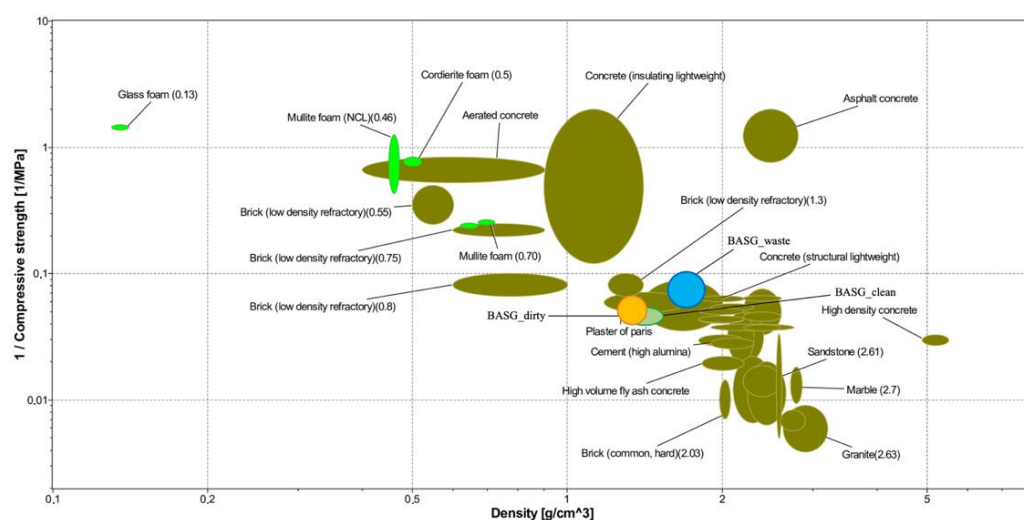


Figure 4. Graph illustrating the trade-off between compressive strength and density of lightweight products (Ansys Granta EduPack version Ansys 2024 R1) in comparison to BASG samples.

5.2. Alkali Activation of Glass as Sorbent for Dyes

Beyond the applications in the construction sector, alkali-activated materials are increasingly being recognised for their potential in water and wastewater treatment. Their intrinsic porosity, characterised by the presence of nano- and micropores, contributes to their effectiveness in adsorbing contaminants and facilitating various purification processes [130]. Thanks to these properties, AAMs are suitable for use as adsorbents, ion-exchangers, membranes, and filtration media [131].

Among the various purification techniques, adsorption constitutes an efficient and economically viable approach for the removal of pollutants in wastewater treatment. This method is characterised by simplicity, cost-effectiveness, and high efficacy, requiring minimal operational complexity [132,133]. In particular, the effectiveness of AAMs in adsorption processes is closely related to their microstructure, especially the surface and the porosity, together with the presence of surface functional groups [134]. These characteristics are particularly critical for dye removal, where both physical and chemical interactions influence adsorption capacity.

It has been demonstrated by Metha et al. [120] that cold-consolidated pharmaceutical glass powder enables the fabrication of innovative glass membranes. Additionally, these materials also function as dye-degrading sorbents when combined with TiO_2 .

Moreover, further innovation has been achieved by the integration of additive manufacturing technology. Mahmoud et al. [135] produce masked-stereolithography scaffolds functionalised through alkali activation, resulting in the formation of zeolites and sodium carbonate phases. These innovative filters exhibit a high performance in the filtration and removal of methylene blue, attaining an impressive removal efficiency of 81%. Building upon findings from prior studies, the same research group [136] successfully developed stable glass-based functional scaffolds that do not rely on any organic binders. The reaction rate of new scaffolds achieved 100% degradation of methylene blue in 75 min and retained their performance over five cycles without a significant loss of efficiency. The stability, potential for repeated use, and ease of scaling up mark a significant step toward the filtration technologies based on waste glass AAMs.

5.3. Alkali Activation of Glass as Sorbent for Heavy Metals

Heavy metal contamination in wastewater is a serious risk for both human health and the environment, thereby driving efforts to develop sustainable removal techniques [137]. Wastewater often contains trace metallic elements [138], which are considered persistent in the environment since they do not decompose [139].

Traditional adsorbents, including activated carbon, alumina, and silica, are known for their high efficiency in removing traces of heavy metals. However, their elevated production costs have restricted their widespread use in practical applications [140,141]. In recent years, alkali-activated materials have emerged as an alternative for adsorption and have attracted increasing attention [142] due to low-cost synthesis, variable chemistry, and the capacity to immobilise toxic species.

Boro–alumino–silicate glass activated with a mild alkaline solution (NaOH/KOH , 2.5 mol/L) has been demonstrated to be an excellent matrix for the stabilisation of copper ions [143]. In fact, alkali activation can minimise ion mobility by coupling them with the glass matrix. Specifically, after the optimisation of the pH of $\text{Cu}(\text{NO}_3)_2$ aqueous solution, adsorption tests indicate an adsorption efficiency of $62 \pm 3\%$, consistent with geopolymeric matrices [144]. The final stabilisation is achieved thanks to the firing process at 700°C . The efficiency of copper retention was assessed through leaching tests. This suggests that mild alkali activation combined with controlled heat treatment offers an effective strategy for immobilising hazardous ions.

5.4. Alkali Activation of Glass for Nuclear Waste Immobilisation

Waste management necessitates the implementation of effective protocols to diminish the concentrations of radioactive contaminants to levels permissible for the safe discharge of decontaminated liquids into the environment, as well as for the secure disposal of concentrated radionuclides [145,146].

Vitrification is widely regarded as the predominant treatment technology for the immobilisation of high-level radioactive waste, effectively converting hazardous materials into a stable glass matrix that enhances long-term storage and containment characteristics [147]. Boro–silicate glass is a commonly used glass composition for vitrification due to its high chemical and thermal stability [148].

On the other hand, cement solidification represents a well-established and advanced technology that is based on the hydration of cement and its ability to form gels for immobilising radioactive materials. This technology still encounters several technical limitations, [149] such as minimal volume reduction and the relatively high tendency to release contaminants, especially with cesium [150,151]. Indeed, due to the radiation emitted by Cs-137 and Sr-90, ordinary Portland cement-based materials are not appropriate for the immobilisation of these isotopes due to low thermal resistance, high content of free water, and inadequate immobilisation [152,153].

Furthermore, alkali-activated materials offer a viable alternative in encapsulating nuclear waste, due to their cationic binding sites and durability [154–156]. In the context of immobilising simulated nuclear waste through alkali activation, the common matrices are metakaolin [157], fly ash [158], and blast furnace slag [159].

Pioneering studies have been conducted by Lago et al. [160] for the immobilisation of CsOH using alkali-activated boro–alumino–silicate matrices. They successfully achieved an immobilisation of over 20 wt% cesium through a cold consolidation process. A portion of cesium, incorporated into boro–pollucite, is immobilised immediately upon the cold consolidation process. The resulting cold-consolidated blocks resist immersion in boiling water. The sintering process allows further strengthening. Moreover, the leaching tests performed on fired samples revealed minimum cesium release.

To minimise the production of soluble species, an engineered treatment using both CsOH and NH_4OH was employed to maximise the immobilisation of cesium [161]. Despite the absence of increased boro–pollucite formation, the observed enhancement in cesium retention is likely attributable to the formation of a less soluble gel associated with the interaction of Cs^+ ions with an elevated concentration of glass components released into the solution.

5.5. Alkali Activation of Glass and Microwaved-Assisted Process

Microwave heating represents an innovative technological advancement within the cement and concrete industries, attracting significant interest and ongoing enhancements attributable to its numerous advantages [162,163]. First of all, rapid heating rate allow to reduce energy and time: Li et al. [164] outlined that the concrete curing with microwave heating requires one-third of the duration and 10% of energy compared to the traditional process. Moreover, precise and immediate control through electronic systems, the formation of fine microstructure that enhances the properties of the resulting cement, and the absence of secondary waste production are highlighted as benefits [165].

Microwave heating has been applied to alkali-activated materials and geopolymers. Microwave curing has the potential to significantly expedite the hardening phase of these materials [166]. Aschoff et al. [167] reported that the use of microwave treatment leads to an accelerated setting and hardening process in metakaolin and aluminium orthophosphate-based geopolymers. In fact, microwave heating, unlike conventional methods, minimises

heat loss and eliminates the need for the heat to be transferred between the heating device and the samples: the energy is directly converted from electromagnetic to thermal energy, resulting in a faster hardening process.

Moreover, microwave treatment leads to a noticeable improvement in the compressive strength of the final product. Hong et al. [168] demonstrate that microwave treatment, applied to coal bottom ash-based geopolymers, after an initial curing phase of 36 h, allows for an increase in compressive strength from 13 to 40 MPa. Carollo et al. [169] performed microwave-assisted cold consolidation on glass waste slurries, revealing that the compressive strength increases from 10 MPa to 18 MPa thanks to 5 min microwave heating at 450 W.

Alkali activation of boro–alumino–silicate glass, combined with microwave curing, provides an opportunity for understanding the molecular mechanisms of ‘mild’ alkali activation [170]. The application of microwave irradiation facilitates the promotion of carbonate formation associated with potassium (K^+), thereby promoting the formation of less soluble gels, which substantially enhances the overall stability of the material, as shown in Figure 5. Moreover, the microwave treatment has demonstrated a beneficial impact on the mechanical properties of alkali-activated glass, with an enhancement of up to 100% together with an increase in total porosity.

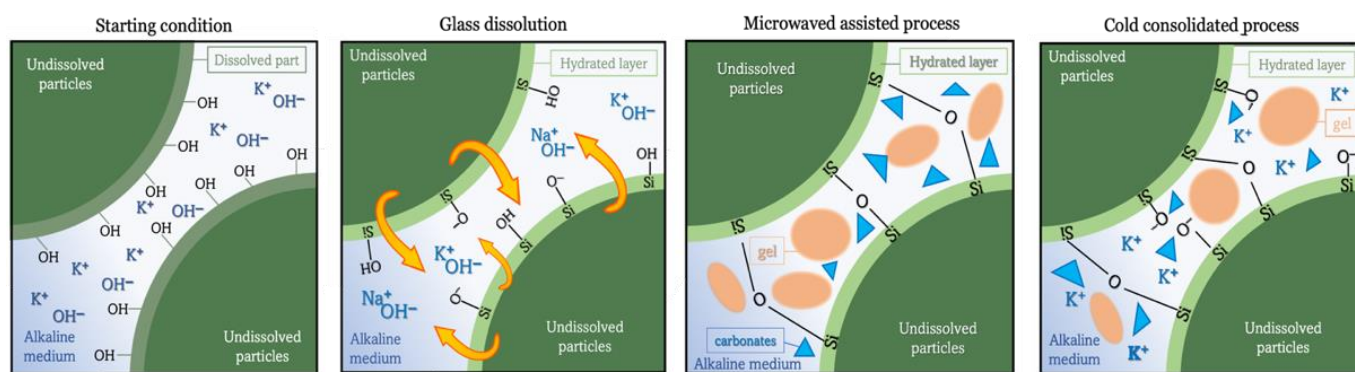


Figure 5. Graphical representation of the molecular mechanism of the microwave-assisted process and cold-consolidated process.

6. Conclusions

Despite glass being widely recognised as one of the most recyclable materials, significant challenges persist in the management of glass waste, particularly concerning the recovery of fine glass fractions. These glass fragments frequently evade efficient recovery through traditional recycling methods and are landfilled. However, emerging trends in recycling and upcycling present promising avenues for addressing this issue. Specifically, the utilisation of glass in the production of geopolymers and alkali-activated materials represents a significant opportunity to convert waste into high-value applications. This comprehensive overview results in the following conclusions:

- Glass has the potential to serve as the sole raw material for alkaline activation, functioning independently of its chemical composition and the molarity of the alkaline solution. This suggests that the molecular mechanism of mild alkali-activated glasses is similar to glass corrosion. Due to the limited molarity, the alkaline solution does not allow for the complete dissolution of the glass used as raw material, but only affects the surface of the glass particles. Moreover, the OH^- ions from the alkaline source led to the cleavage of strong bonds, such as siloxane bonds, resulting in the formation of a hydrated surface layer ($-OH$). During the hardening phase, there is the rearrangement of $Si-O-Si$, $Si-O-Al$, and $Si-O-B$ between the particles. The other constituents in the

solution reorganise into a secondary phase that may involve the formation of various compounds, such as carbonates, gel, and potential crystalline phases. The secondary phase does not contribute significantly to the stability of the final product.

- In contrast to their conventional application as substitutes for cements and concrete, alkali materials may be considered for environmental uses. Alkali-activated materials are used as membranes for dye adsorption, such as methylene blue. Moreover, the removal of heavy metals can be performed with an alkali-activated sorbent.
- Alkali-activated glass presents a promising avenue for the immobilisation of simulated nuclear waste, particularly in regard to cesium hydroxide (CsOH). The samples fired at 700 °C exhibited only trace levels of cesium release during leaching tests.
- The microwave-assisted process significantly contributes to reducing the hardening time of materials. Microwave techniques can also provide deeper insights into the molecular mechanism during the hardening process of alkali-activated glass. In fact, the formation of a less depolymerised gel, and consequently a more stable structure, is favoured by a more efficient allocation of potassium ions.

This overview offers a comprehensive overview of the current advancements in alkali activation, with particular focus on mild alkali activation systems. However, several important research gaps remain, most notably the absence of durability studies among the works cited, a key factor in evaluating their long-term viability and application. This is particularly evident in the lack of investigations into resistance to freeze–thaw cycles, chemical attack, and carbonation, which hinders direct comparison with traditional materials such as Portland cement.

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