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## Alkaline-Activated Materials for CO<sub>2</sub> Capture – Literature Review, Own Observations, and Future Perspectives

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Abstract: Alkaline-activated materials and geopolymers are materials derived from the alkaline activation of aluminosilicate precursors. Their production is based on the reaction of raw materials rich in silica (SiO<sub>2</sub>) and aluminum (Al<sub>2</sub>O<sub>3</sub>) with alkaline activators. They have been known for many years and are regarded as a viable alternative to traditional binders. One crucial aspect of these materials is their potential to capture and permanently immobilize CO<sub>2</sub> molecules within their structure, owing to their chemical composition and inherent porosity. This feature can bring significant environmental benefits and contribute to reducing atmospheric CO<sub>2</sub> levels, especially in the context of large-scale industrial use. Alkali-activated materials rank among the most promising systems for CO<sub>2</sub> reduction and may form a crucial component of future strategies aimed at combating climate change. At least three main mechanisms of CO<sub>2</sub> sequestration in such materials have been identified, including physical and chemical adsorption, as well as carbonate formation. However, this topic still faces substantial challenges that need to be addressed by the scientific community. This paper presents the current state of knowledge regarding CO<sub>2</sub> sequestration mechanisms in geopolymer and alkali-activated materials, while also outlining key future directions and research priorities.

**Keywords:** alkaline-activated materials; carbon dioxide adsorption; CO2 capture; geopolymer structure; zeolites

#### 1. Introduction

The intensifying greenhouse effect is a consequence of increasing atmospheric concentrations of greenhouse gases. It is estimated that, despite the optimization of fossil fuel combustion processes and the use of renewable energy sources, climate change will be largely caused by an increase in the concentration of greenhouse gases. Carbon dioxide (CO<sub>2</sub>) is widely recognized as the primary contributor to anthropogenic global warming due to its high atmospheric concentration and long residence time <sup>1), (2), (3)</sup>. While scientific perspectives may differ regarding the relative contributions of natural and human-related drivers of climate change, the steady rise in atmospheric CO<sub>2</sub> is clear, as is the urgent need to reduce its buildup. Recent

data from the *Global Carbon Budget 2024* indicate that global CO<sub>2</sub> emissions from fossil fuel combustion and industrial processes reached a record high of approximately 37.4 billion tonnes in 2024, continuing an upward trend despite efforts to increase renewable energy use <sup>4</sup>). This persistent growth highlights the challenge of mitigating climate change solely through emission reductions and underscores the critical need for effective carbon sequestration and removal technologies to complement decarbonization efforts <sup>5</sup>). The hope for a substantial shift in this trajectory lies in innovative Carbon Capture and Storage (CCS) technologies. Experts say that current CCS technology has the potential to contribute to the mitigation of approximately 14–20% of total anthropogenic CO<sub>2</sub> emissions <sup>6</sup>). Although numerous

methods for CO2 sequestration are available, current efforts remain insufficient. To stay on track with the 2°C scenario — that is, to limit global temperature rise to 1.5-2°C above pre-industrial levels in accordance with the Paris Agreement — the carbon capture and storage (CCS) sector would need to expand by 2050 to a scale 2-4 times larger than today's oil industry <sup>6),7)</sup>. While such a projection may appear unlikely, the commitments made under the Paris Agreement demand urgent mobilization and the pursuit of more effective sequestration solutions and materials. A responsible approach, when paired with the support of artificial intelligence in energy management and organizational processes, can produce tangible results even in the short term—outcomes that may currently seem unrealistic to many 8). The material challenges faced by researchers and innovators are to tailor and optimize the structural sorbents for their working CO<sub>2</sub> capacity, selectivity to other components, effects of pollutants and moisture, mass and heat transfer kinetics, and mechanical and chemical strength, all of which are specific to exhaust systems and exhaust composition 9).

A particularly promising direction involves development of materials capable of simultaneously reducing CO<sub>2</sub> emissions and enabling its long-term sequestration. CO<sub>2</sub> capture and conversion using structured porous sorbents and catalysts is a solution that can help decarbonize carbon-intensive industries. The building sector, in particular, offers substantial potential for emission reductions, as it currently accounts for over 30% of global energy consumption and approximately 26% of energy-related CO<sub>2</sub> emissions <sup>10</sup>. In this context, geopolymers — also referred to as inorganic polymers represent a low-emission alternative to conventional cement-based materials, particularly those relying on Portland cement. More importantly, they offer the potential to serve as the most effective media for permanent CO<sub>2</sub> sequestration.

Geopolymer materials have attracted increasing scientific attention due to their unique structural features and versatility in sorption-related applications 11). Their inherently high porosity, tunable pore structure, and welldeveloped specific surface area make them highly promising as solid adsorbents for various substances, including heavy metals and organic pollutants <sup>12),13)</sup>. These same features—combined with their alkaline composition and reactive surface—also make geopolymers particularly interesting candidates for CO<sub>2</sub> capture <sup>14-18)</sup>. Experimental studies have shown that CO2 adsorption can be enhanced through structural modification, such as the incorporation of zeolites or hydrotalcite phases into the geopolymer matrix, further improving capacity and selectivity 14-17). Numerous recent works have demonstrated the potential of geopolymer-based materials to effectively fix CO2 via

physical and chemical interactions with their surface and

internal microstructure <sup>16),17),19),20)</sup>. These interactions are

closely linked to their pore size distribution and the presence of active adsorption sites, which can be tailored during synthesis <sup>21),22)</sup>.

Foaming techniques have been shown to further enhance the porosity and specific surface area of geopolymers, thereby improving their performance as adsorbent materials <sup>23-25</sup>).

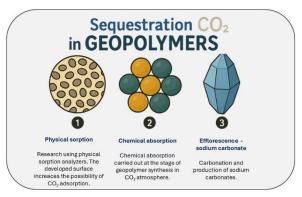
As a result, geopolymers are now being extensively investigated not only for their mechanical performance and durability, but also as functional materials in the context of carbon capture technologies <sup>26),27)</sup>.

This paper presents a literature review on the potential for CO<sub>2</sub> sequestration in geopolymers and alkali-activated materials. Several mechanisms and methods of CO2 sequestration are discussed, with particular focus on solidstate geopolymers due to their highly favorable composition and microstructure. Although briefly, the review also addresses CO2 fixation occurring during the synthesis and setting stages of geopolymer formation. Following the discussion of sequestration mechanisms, the key advantages of geopolymers as CO2-adsorbing materials are examined, particularly in comparison to other commonly used construction materials. A dedicated section discusses zeolites formed in situ within the geopolymer matrix, which have shown particularly high efficiency in capturing CO2. An important and original part of this work incorporates the authors' own research and applied observations from ongoing collaboration with the industrial sector. These insights highlight not only what the sector is currently pursuing but also where its focus should shift in the near future. The authors identify critical areas where the scientific community should direct its efforts to unlock the full potential of geopolymers. Given their sorption capabilities, geopolymers may eventually be recognized not merely as alternatives or competitors to traditional binders, but as active contributors to reducing the carbon footprint of the cement industry.

### 2. Mechanisms of CO<sub>2</sub> fixation in geopolymers and alkali-activated materials

As in other types of sorption materials or porous ceramics, the mechanisms that can be useful in  $CO_2$  sorption are several in geopolymers. Geopolymers, however, are among the most favorable materials in this regard, as they have a number of features that distinguish them from other similar material structures  $^{28-31}$ ).

Sequestration can occur both on hardened geopolymer surfaces and during the synthesis and curing stages of geopolymer formation. A distinction should be made between surface adsorption (reversible), including physical and chemical mechanisms, and structural and permanent reactions (irreversible), such as mineralization or the incorporation of CO<sub>2</sub> into binding phases during curing <sup>32-35)</sup>.



**Fig. 1:** Main mechanisms of CO<sub>2</sub> fixation in solid-state geopolymers

This paper focuses primarily on CO<sub>2</sub> sequestration in solidstate geopolymers. Three main sequestration mechanisms are identified here: physical adsorption, chemical adsorption, and carbonation/efflorescence. This classification is illustrated graphically in Figure 1. Additionally, to provide a more comprehensive overview, a brief review of selected sequestration methods occurring during the production stage is also included.

These mechanisms can interpenetrate and complement one another, often occurring simultaneously and collectively enhancing the overall capacity for CO<sub>2</sub> sequestration <sup>36-40)</sup>.

### 2.1. Mechanisms of CO<sub>2</sub> Interaction with Solid-State Geopolymer

#### 2.1.1. Physical and Chemical Adsorption

Physical adsorption involves the attachment of CO2 molecules to the surface of the geopolymer by means of weak van der Waals forces. This process depends on such parameters as the porous structure of the material (microand mesopores), the specific surface area of the geopolymer, and the process conditions (pressure, temperature, humidity) 41-47). Physical adsorption is a reversible process that allows regeneration of the material. Chemical adsorption involves chemical reactions between CO<sub>2</sub> and active chemical groups present on the surface or in the structure of the geopolymer. Chemical reactions that can occur include carbonation, which is the reaction of CO2 with active metal oxides (e.g., CaO, MgO), leading to the formation of carbonates (e.g., CaCO<sub>3</sub>, MgCO<sub>3</sub>), or reactions with hydroxyl groups (-OH) in the geopolymer matrix. Chemical adsorption is more durable than physical adsorption, but the process of regenerating the material is more difficult <sup>48-55)</sup>. Factors affecting CO<sub>2</sub> sequestration efficiency are primarily chemical composition. A high content of alkali oxides (e.g., Na2O, K2O) increases the capacity for carbonation, as well as the presence of reactive silicates and aluminates promotes CO<sub>2</sub> capture <sup>56)</sup>.

Another factor is the porous structure. Larger specific surface area and pore volume improve adsorption efficiency. The last factor is environmental conditions. Lower temperatures favor physical adsorption, while higher temperatures can initiate chemical reactions <sup>57)</sup>. CO<sub>2</sub> pressure and relative humidity have a significant impact on the adsorption process <sup>58-64)</sup>. Due to their large specific surface area, geopolymers can act as filter materials in CO<sub>2</sub> capture processes from industrial gases <sup>65-68)</sup>. Faqir et al. have shown that metakaolin-based geopolymers exhibit significant CO<sub>2</sub> adsorption capacity, with some formulations reaching a maximum adsorption capacity of up to 800 mg/g under certain conditions <sup>69)</sup>. The incorporation of zeolites and other additives can further improve adsorption properties by increasing surface area and introducing reactive sites for chemical interactions <sup>24),70)</sup>. This topic will be discussed in more detail in Subsection 3.2.

#### 2.1.2. Carbonation and Associated Efflorescence

The formation of efflorescence is related to leaching of sodium or potassium ions from the structure of geopolymers, and is explained similarly to hydraulic binders by the fact that when the products are in contact with moisture, water, due to capillary forces, leaches the alkali ions and "carries" them to the surface. After the water evaporates, these ions react with CO2 in the atmosphere and hydrated sodium or potassium carbonates are formed, e.g. Na<sub>2</sub>CO<sub>3</sub>.nH<sub>2</sub>O, NaHCO<sub>3</sub>.nH<sub>2</sub>O, K<sub>2</sub>CO<sub>3</sub>. This happens especially when there is an excess of alkaline substances in the material and the material has high porosity. This is especially true for geopolymers with a high Na<sub>2</sub>O/Al<sub>2</sub>O<sub>3</sub> ratio. Studies conducted so far on efflorescence on geopolymers have shown that white efflorescence on the surface of geopolymer samples is Na<sub>2</sub>CO<sub>3</sub>-7H<sub>2</sub>O. The identified carbonate crystallizes from inside the pores and "grows" on the outside of the sample. Studies of the 23Na NMR MAS spectrum indicate that alkali metals are bound by Al in the Si-O-Al chain structure in the Na,K(H<sub>2</sub>O)n+ form rather than in the Na+, K+ po-form 71). The binding of alkali metals in the form of Na,K(H2O)n+ complexes is weaker than direct Na+ binding, which is the reason for the ease of leaching of alkali metals from alkali-activated structures and geopolymers <sup>72),73)</sup>.

Examples of efflorescence on the surface of geopolymers are shown in Figure 2 and Figure 3. Efflorescence on the surface of geopolymers is sodium carbonates formed by CO2 found in the surrounding atmosphere and sodium ions found on the surface of geopolymers.

The formation of carbonates on geopolymer surfaces is a gradual process. Initially, only small amounts of efflorescence appear, which increase over time until the surface becomes completely covered.

This efflorescence phenomenon is typically regarded as a degradation feature, particularly when these materials are used as building components, since it adversely affects their aesthetic appearance and poses a barrier to wider implementation of geopolymers <sup>74-76</sup>). However, when

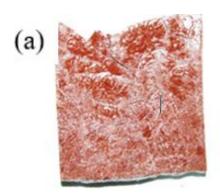




Fig. 2: Examples of geopolymer materials covered with efflorescence in the form of sodium carbonate: (a) shows the beginning of efflorescence formation, (b) the advanced stage. (Source: authors' own unpublished research; illustrative purposes only)

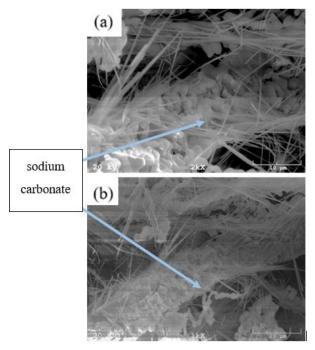


Fig. 3: SEM micrographs showing sodium carbonate formations on the surface of geopolymer samples: close-up views corresponding to the materials presented in Fig. 2 (a) and (b) (Source: authors' own unpublished research; illustrative purposes only)

managed appropriately, this process may also offer opportunities for atmospheric CO<sub>2</sub> reduction. By creating sufficiently large geopolymer elements with highly developed surface areas, these materials can serve as effective CO<sub>2</sub> capture systems <sup>77-79</sup>. The emerging sodium carbonates formed on their surfaces can be systematically harvested and used as raw materials in various chemical and industrial applications. Notably, sodium carbonate has well-established uses in the glass and paper industries, as well as in the formulation of chemical cleaning agents <sup>80),81</sup>. Harnessing this potential requires a comprehensive and systematic research approach to develop suitable technological solutions and optimize efflorescence management.

### 2.2. Bulk CO<sub>2</sub> incorporation during synthesis and curing

Another strategy for enhancing the carbon sequestration potential of geopolymers involves the direct incorporation of CO<sub>2</sub> during material synthesis and curing. Unlike conventional post-synthesis adsorption processes, this approach enables permanent immobilization of carbon dioxide within the bulk matrix by taking advantage of carbonation reactions occurring concurrently with geopolymerization. The presence of CO2 in the reaction environment can lead to the formation of metastable carbonate phases that evolve during curing, contributing not only to carbon capture but also to the mechanical performance of the hardened material. An example is provided by the study of Matalkah and Soroushian on geopolymers formulated with fluidized-bed combustion (FBC) coal ash, synthesized in a CO<sub>2</sub>-rich environment <sup>82)</sup>. Characterization using XRD, SEM/EDS, TGA, and FTIR confirmed the presence of non-crystalline (metastable) carbonates formed in situ during curing. These metastable carbonates, upon subsequent crystallization, enhanced the hydration-like binding processes within the geopolymer matrix. Thus, enhanced binding mechanisms resulted in significant improvements in mechanical strength compared to samples cured in air. The optimized geopolymer formulation achieved CO2 uptake of approximately 7-8 wt%. Additionally, the material exhibited good fresh-state workability and reached compressive strength values suitable for structural applications after 28 days of curing.

These findings align with other research that highlights the beneficial influence of carbonation during curing, including enhanced geopolymerization and polymerization effects in CO<sub>2</sub>-cured alkali-activated materials <sup>34)</sup>. The patent by Khalifeh and Hodne detailed specific parameters, such as the concentration of alkaline activators, temperature, and curing atmosphere, that promote simultaneous geopolymerization and carbonation reactions. This co-occurrence should lead to enhanced densification of the microstructure and improved

mechanical properties due to the filling effect of carbonate minerals. Additionally, the method enabled the utilization of various industrial by-products (e.g., fly ash, slag) as precursors, thereby contributing to sustainability and waste valorization <sup>83)</sup>.

In the specific case of geopolymers based on fluidized-bed combustion (FBC) ash cured in CO<sub>2</sub>-rich environments, a study by Ridha et al. has shown CO<sub>2</sub> uptake in the range of 7–8 wt%, along with 20–35% improvements in compressive strength over conventional air curing at various time points <sup>84</sup>).

Another promising approach involves using sodium carbonate (Na<sub>2</sub>CO<sub>3</sub>), commonly known as soda ash, synthesized through CO2 capture, as an alkali activator in geopolymer production. While the formation of soda ash as a product of carbonation was discussed earlier in Section 2.1.1 (Carbonation and Associated Efflorescence), it is important to note that Na<sub>2</sub>CO<sub>3</sub> can also be deliberately used as an alkali activator during the synthesis of geopolymer binders. This dual functionality opens new opportunities for closing the carbon loop—converting captured CO<sub>2</sub> not only into a stable product but also into a reactive precursor that facilitates further CO2 fixation. Studies have demonstrated that carbonate-based activators, such as Na<sub>2</sub>CO<sub>3</sub>, can replace or complement conventional activators like sodium hydroxide and sodium silicate. Ma et al. showed that partial replacement of sodium silicate with Na<sub>2</sub>CO<sub>3</sub> in slag-fly ash geopolymers yielded materials with mechanical strength comparable to traditional systems, while reducing the carbon footprint of the process 85).

Similarly, hybrid one-part geopolymers activated entirely with Na<sub>2</sub>CO<sub>3</sub> have demonstrated excellent compressive strength and workability, while embedding carbonate into the matrix

Expanding on this idea, Gupta et al. introduced a CO<sub>2</sub>-assisted geopolymerization method in which CO<sub>2</sub> is used to both synthesize soda ash and initiate aluminosilicate polymerization in fly ash systems <sup>86</sup>). His approach yielded materials suitable for construction applications. Notably, the process consumed a substantial amount of CO<sub>2</sub>, providing a meaningful pathway for direct atmospheric carbon utilization.

A somewhat comparable strategy was demonstrated by Zheng et al., who utilized calcium oxide (CaO) as both the alkaline activator and an in situ CO<sub>2</sub> sorbent in one-part alkali-activated slag systems. In this dual-function role, CaO underwent carbonation during the reaction, forming a surface layer of CaCO<sub>3</sub> that influenced the microstructure and reactivity of the material. The carbonation process was facilitated by physically adsorbed water and governed by CO<sub>2</sub> diffusion under ambient conditions. While the formation of CaCO<sub>3</sub> contributed to microstructural densification, it also led to increased setting times due to the development of a product layer on the activator surface,

which hindered further reaction by increasing diffusion resistance <sup>87)</sup>.

### 3. Geopolymer-Based Strategies for Enhancing CO<sub>2</sub> Sequestration

### 3.1. Advantages of Geopolymers over Conventional Materials for CO<sub>2</sub> Capture

The synthesis of geopolymers for CO2 capture offers several advantages over conventional materials, particularly in terms of environmental impact and functional performance. One key advantage is the possibility of utilizing industrial by-products and mining waste as precursors. Commonly used materials in this context include fly ash and, to a lesser extent, blast furnace slag due to their high aluminosilicate content and favorable reactivity. A less typical yet promising example is presented by Schneider et al. 88). They demonstrated that geopolymer-zeolite composites produced using phosphate mining tailings form hierarchically porous structures that are effective in capturing CO2. Vasconcelos et al. developed a process for synthesizing geopolymers using raw materials such as metakaolin, slag, and fly ash, emphasizing the importance of removing impurities that could hinder material performance. The resulting geopolymers demonstrate enhanced mechanical and chemical stability, making them suitable candidates for long-term CO2 capture applications. Their tailored composition allows for tunable porosity and surface chemistry, which can be optimized to maximize CO2 uptake while maintaining structural integrity <sup>89)</sup>.

The intrinsic microstructure of geopolymers plays a critical role in their gas adsorption capacity. As shown by Zarębska et al. <sup>90)</sup>, the presence of irregular pores and a highly interconnected aluminosilicate network enhances the physical sorption of CO<sub>2</sub> molecules, while also enabling the removal of other gaseous pollutants such as SO<sub>2</sub> and H<sub>2</sub>O vapor.

Geopolymers are also distinguished by their excellent thermal stability and chemical durability. Nasvi, Ranjith, and Sanjayan <sup>19)</sup> demonstrated that properly formulated geopolymers exhibit low CO<sub>2</sub> permeability and strong resistance to leaching, making them suitable for long-term carbon storage, including applications such as well cementing in CO<sub>2</sub> sequestration reservoirs.

In terms of practical applications, geopolymers have been extensively researched for use in CO<sub>2</sub> sequestration wells, demonstrating performance that surpasses traditional Portland cement. Adjei et al. <sup>91)</sup> conclude that geopolymers exhibit excellent resistance to carbonation, eliminating the degradation risks associated with cement-induced efflorescence. Moreover, geopolymers maintain mechanical integrity under CO<sub>2</sub>-rich and high-pressure downhole conditions, showing negligible changes in

compressive strength and Young's modulus even after six months of exposure <sup>92)</sup>. They also perform significantly better in acidic and high-salinity environments, remaining compatible with drilling fluids and maintaining seal integrity <sup>93)</sup>.

Further insight into the adsorption capabilities of geopolymers was provided by Ishwarya et al. 74), who investigated CO<sub>2</sub> uptake in metakaolin-based geopolymers of the (K)poly(sialate-siloxo) type, synthesized using potassium silicate solutions without the use of poreforming agents. Despite the absence of artificial porosity enhancers, the resulting materials exhibited relatively high total porosity (30–60%) and a specific surface area ranging from 22 to 50 m<sup>2</sup>/g, as determined by Brunauer-Emmett-Teller (BET) analysis. The measured CO2 adsorption capacity was approximately 0.6 mmol/g at atmospheric pressure. Although this value is lower than those reported for advanced sorbents such as metal-organic frameworks or synthetic zeolites, the geopolymer demonstrated markedly higher selectivity for CO2 over lighter gases like methane and nitrogen. This behavior was attributed to competitive sorption mechanisms, molecular size exclusion, and preferential interaction with CO2 molecules.

These properties collectively position geopolymers as a multifunctional and low-impact alternative to traditional materials used in carbon capture and storage systems. However, even greater CO<sub>2</sub> capture performance can be achieved through targeted modifications of the geopolymer matrix. These include the formation of geopolymer–zeolite composites, which combine the structural advantages of geopolymers with the high surface area and microporosity of zeolites; the enhancement of porosity through physical or chemical foaming techniques; and chemical functionalization of the surface, for instance by introducing amine groups that promote selective chemisorption of CO<sub>2</sub>. These strategies are discussed in detail in the following subsections.

### 3.2. In-Situ Zeolite Formation and its role in CO<sub>2</sub> capture in alkali-activated systems

One of the distinguishing features of alkali-activated materials, particularly geopolymers, is their ability to form zeolites under certain synthesis conditions. This phenomenon, known as in-situ zeolite formation, has gained increasing attention due to its significant impact on the structure, porosity, and sorption properties of the resulting materials <sup>94-98</sup>).

Zeolites themselves are well known for their excellent adsorption and separation capabilities, particularly in gas separation applications. Scientists placed particular emphasis on zeolite frameworks with micropores whose dimensions are closely aligned to the kinetic diameter of light gases, such as CO<sub>2</sub>. Nanoporous zeolites offer distinctive opportunities for the development of high-

performance zeolite and zeotype membranes intended for CO<sub>2</sub> removal and capture applications <sup>99-103)</sup>.

A study worth mentioning in this context is the work by Hayakawa and Himeno  $^{104)},$  who presented the synthesis of silica-based ZSM-58 zeolite membranes on tubular  $\alpha$ -alumina supports. By optimizing the synthesis parameters, they obtained high-purity ZSM-58 membranes with a high efficiency of CO<sub>2</sub> separation from CH<sub>4</sub> and N<sub>2</sub>, confirming the potential of such materials for selective gas adsorption and separation.

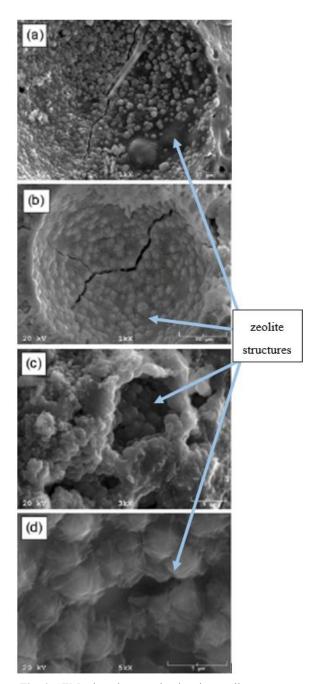
The integration of the mechanical strength of the geopolymer with the adsorptive properties of zeolite crystals within the geopolymer matrix enhances its suitability for membrane applications <sup>94</sup>).

The synthesis of zeolites from materials/raw materials of anthropogenic origin is most often carried out in a similar manner to the manufacture of geopolymer materials, namely by alkaline activation. For this reason, zeolites of various types are very common in geopolymer structures <sup>17),105-110)</sup>. The high reactivity and homogeneity of metakaolin-based geopolymers promote extensive zeolitization during hydrothermal treatment <sup>111)</sup>.

Fly ash-based geopolymers, despite their more heterogeneous nature, can also achieve a substantial degree of zeolite formation, especially when the type of activator, curing temperature, and solution composition are appropriately optimized <sup>112</sup>).

Examples of zeolite crystals formed within fly ash-based geopolymer matrices, as observed in the authors' experimental materials, are presented in Figure 4. These images are provided for illustrative purposes only, to show the typical morphology of such in-situ crystalline phases. The adsorption properties of fly ash-based geopolymers have been shown to depend strongly on the extent of zeolite phase formation within the matrix. Zheng et al. demonstrated that during the in-situ transformation of amorphous aluminosilicate gels in such systems, Na-P1 zeolite crystals can form, significantly enhancing the material's mechanical integrity and its ability to adsorb metal ions such as Cu<sup>2+</sup> and Ni<sup>2+</sup> <sup>113)</sup>. These results confirm the dual benefit of zeolitization in geopolymers, contributing both to structural development and functional performance in adsorption processes.

Furthermore, traditional methods of granulating zeolites for practical use often involve the addition of inert binder materials such as bentonite or kaolin clay, which tend to reduce the CO<sub>2</sub> adsorption capacity of the final product by approximately 15–20%. In this context, the use of geopolymers as a binder offers a promising alternative. As shown by Vegere et al., alkali-activated metakaolin can serve simultaneously as a zeolite-like binder and an active adsorbent phase, eliminating the need for inert additives and preserving or even enhancing CO<sub>2</sub> adsorption performance <sup>114</sup>). This highlights the potential of geopolymer-based systems not only as precursors for



**Fig. 4:** SEM microphotographs showing zeolite structures formed in fly ash-based geopolymers (source - authors' own research)

zeolite formation but also as functional sorbents in their own right. Expanding the understanding of zeolite performance for CO<sub>2</sub> capture, Shi et al. conducted grand canonical Monte Carlo (GCMC) simulations to evaluate the CO<sub>2</sub> sorption potential of 2625 aluminosilicate zeolite structures <sup>115</sup>. These were selected from a set of 262,500 hypothetical ABC-6 models with Si/Al ratios ranging from 1.0 to 10. Beyond conventional metrics such as CO<sub>2</sub> adsorption selectivity, the study also assessed working capacity, adsorbent performance score, and regenerability to offer a comprehensive evaluation of zeolite performance in temperature-swing adsorption (TSA), vacuum-swing

adsorption (VSA), and pressure-swing adsorption (PSA) processes. The results revealed hundreds of hypothetical structures with CO2 capture efficiencies exceeding that of Na-X zeolite, which is currently regarded as one of the most effective CO2 sorbents. These findings provide guiding experimental valuable insights for the development of new high-performance zeolite materials. Significantly, many of the hypothetical zeolite frameworks identified in the simulations may be synthesized through carefully controlled geopolymerization processes. This opens up opportunities for designing geopolymer-based materials that not only serve as structural components but also function as efficient CO2 sorbents. Given the temperature dependence of CO2 capture, such multifunctional materials could be applied in thermally exposed environments-such as chimney liners-where elevated temperatures favor the adsorption process. Properly engineered, these materials could contribute to insitu carbon capture from flue gases, combining structural utility with environmental functionality.

Several researchers investigating the CO<sub>2</sub> adsorption potential of zeolites have focused on structural and surface modifications to improve their sorption performance. Current trends aimed at improving the CO2 adsorption capacity of zeolites include amine, silica, and ionexchange modifications. Other promising efforts to improve the adsorption capacity of zeolites include the processing of zeolite nanopowders, nanofibers, and zeolite-based foams 116). Noteworthy results were reported by Panda, Kumar, and Singh 117, who introduced mesoporosity into zeolite 4A (Z4A) bodies by employing a urea-assisted method to synthesize hierarchical zeolite 4A (HZ4A) with tailored textural properties. As a result, a series of hierarchical zeolites—HZ4A-1-3, HZ4A-1-1, and HZ4A-3-1—was obtained. Among them, HZ4A-1-3 demonstrated a significantly enhanced CO2 adsorption capacity, even under humid conditions at 1 bar and 40 °C. Furthermore, this material exhibited excellent stability over ten consecutive adsorption-desorption cycles. The introduction of mesopores in HZ4A-1-3 facilitated rapid diffusion of CO2 within the pore system, leading to faster adsorption (by 29 seconds) and desorption (by 92 seconds) compared to conventional Z4A. It also resulted in reduced water uptake (20% lower), a lower desorption temperature (by 13 °C), and a 14% decrease in CO<sub>2</sub> heat of adsorption. These improvements translated into higher adsorption efficiency and more favorable regeneration energy requirements. Overall, the study demonstrates the potential of hierarchical zeolite 4A, particularly HZ4A-1-3, as a highly promising sorbent for efficient and stable CO2 capture applications.

### **3.3. Foamed Geopolymers: Increasing Surface Area and Porosity**

Foamed geopolymers are studied for their potential in CO<sub>2</sub>

capture, owing to their enhanced surface area and porous structure. Here, chemical foaming is the most common method compared to other foaming techniques. It involves the incorporation of a foaming agent into the geopolymer precursor mixture, which reacts or decomposes to release gases that form air voids within the material under alkaline conditions <sup>118</sup>).

This approach is particularly attractive because it can be performed under ambient hygro-thermal conditions, resulting in lower energy consumption compared to alternative methods <sup>119</sup>. The chemically induced porosity not only reduces material density but also significantly increases the specific surface area, enhancing the material's ability to adsorb and fix CO<sub>2</sub> molecules.

Recent studies have demonstrated that tailoring the composition and processing parameters of foamed geopolymers can significantly enhance their CO<sub>2</sub> adsorption performance. Chindaprasirt and Rattanasak showed that the use of chemical foaming agents such as aluminum powder, in combination with surfactants, leads to the formation of well-developed porous structures in alkali-activated fly ash-based materials. These materials exhibit a range of interconnected pores that facilitate gas diffusion and improve CO<sub>2</sub> capture efficiency <sup>120</sup>.

Han et al. reported the successful in-situ formation of zeolite X within foamed geopolymer matrices, achieved through the inclusion of hydrogen peroxide as a foaming agent and calcium stearate as a stabilizer. The crystallization of zeolite phases within the foam structure contributed not only to increased surface area but also to enhanced chemisorption of CO2 due to the presence of microporous zeolitic domains <sup>25)</sup>. This study builds upon the earlier work of Han et al., in which the authors synthesized self-supported zeolite foam geopolymers from fly ash under saturated steam conditions. The combination of foam structure and zeolite crystallization yielded materials with hierarchical porosity, ranging from microto macro-scale. The earlier study also confirmed that the alkali activator modulus significantly affects the formation and distribution of zeolitic phases, such as Na-X and Na-P, thereby influencing sorption properties. Although the 2020 work focused on heavy metal adsorption (e.g., Pb2+), the results clearly demonstrated the potential of these materials as efficient solid adsorbents, with implications for broader gas capture applications <sup>97)</sup>.

Similarly, Candamano et al. developed hierarchical geopolymer-zeolite composites by combining chemical foaming with a two-step curing process. Their results indicated that hydrothermal treatment promoted the formation of NaX crystalline phases. At the same time, the addition of pore formers had a more pronounced effect on increasing specific surface area than the incorporation of activated carbon. These findings highlight the relevance of optimizing both porosity and mineralogical composition to maximize adsorption capacity <sup>24</sup>).

A chemo-physical foaming technique was explored by Monjezi and Javanbakht, who combined chemical gas-releasing reactions with emulsion templating to fabricate geopolymer foams enriched with ZSM-5 zeolite <sup>121)</sup>. Their results confirmed that hybrid foaming strategies improve textural properties and adsorption performance. Although their application targeted dye removal, the structural optimization—high porosity, interconnected pores, and presence of zeolitic phases—is equally beneficial for CO<sub>2</sub> capture.

The integration of in-situ zeolite formation with chemical foaming thus represents a promising strategy for producing geopolymer-based materials with enhanced CO<sub>2</sub> capture capabilities.

### **3.4.** Chemical Functionalization with Amine Groups

Chemical functionalization with amine groups is a wellestablished strategy to enhance CO2 chemisorption in porous materials. The approach relies on the formation of chemical bonds between CO2 and surface-bound amine groups, resulting in stable carbamate or bicarbonate species and significantly increasing the adsorption capacity low CO<sub>2</sub> partial pressures. Amine functionalization has been applied mainly to silica, MOFs, porous polymers, and carbon-based sorbents 122-125). However, its extension to geopolymers is also emerging. Wang et al. synthesized porous geopolymer spheres from municipal solid waste incineration fly ash and subsequently functionalized them with amine groups <sup>30)</sup>. The resulting material exhibited a significantly increased CO<sub>2</sub> adsorption capacity, attributed to both the high surface area and the successful grafting of amine functionalities. The amine-functionalized geopolymer spheres showed improved performance under low CO2 partial pressures, highlighting their potential for post-combustion capture applications.

Another promising strategy was proposed by Mirković et al., who developed hybrid metakaolin–brushite composite materials modified with polyethylenimine (PEI) and other amines <sup>126)</sup>. Their study revealed that the addition of PEI increased the number of accessible amine sites and improved chemisorption without severely compromising the structural integrity of the geopolymer matrix. The materials maintained stability under cyclic adsorption–desorption testing and demonstrated considerable selectivity toward CO<sub>2</sub> over N<sub>2</sub>.

Chen et al. introduced a structural regulation approach, tailoring the geopolymer network to better accommodate amine loading and diffusion <sup>127)</sup>. By optimizing the porosity and controlling the polymerization degree of the aluminosilicate gel, they enhanced both the dispersion and the stability of the grafted amine groups. The resulting sorbents showed high CO<sub>2</sub> uptake, strong thermal and chemical resistance, and favorable regeneration

performance over multiple cycles.

Although the presented examples of amine-functionalized geopolymers demonstrate promising performance, research in this area is currently limited, opening a rich opportunity for further exploration of optimal amine types, loadings, and integration with porous geopolymer matrices.

#### 4. Future prospects and critical goals

Despite strong scientific interest, geopolymers—alkaliactivated inorganic polymers known for over a century—have yet to achieve widespread commercial adoption. While they were once considered a promising alternative to conventional Portland cement-based concretes, recent developments have challenged this optimism. Today, more scientists view geopolymers as a "dead end", primarily due to increasing economic and logistical barriers that undermine their scalability <sup>128)</sup>.

One critical issue is the reliance on industrial by-products such as fly ash and blast furnace slag. With the ongoing closure of coal-fired power plants across Europe and North America, fly ash availability has significantly declined, resulting in volatile supply and rising costs. In Germany, fly ash prices increased by 40% in 2022 alone, driven by energy transition policies <sup>129</sup>. This scarcity is compounded by competition from the cement and concrete industries, which also consume these secondary raw materials.

Additionally, geopolymer production depends on alkali activators such as sodium silicate, which are substantially more expensive than cement clinker, costing approximately three to five times more per ton. Because most geopolymer production remains limited to pilot or small-scale facilities, the lack of economies of scale results in 25–35% higher unit costs compared to conventional cement production <sup>129</sup>. These economic disadvantages increasingly affect the feasibility of geopolymers as mainstream construction materials.

At the same time, updated life cycle assessment (LCA) results have become less favorable, particularly when accounting for the environmental footprint of activator production and transport <sup>130</sup>. Although geopolymers continue to offer scientific promise—especially for carbon sequestration and high-temperature applications—their market entry remains hampered by practical constraints. Since the cement industry remains the dominant force in construction and a major source of CO<sub>2</sub> emissions, geopolymers have the potential to play a transformative role in decarbonizing the sector in the coming years. The perception of geopolymers could undergo a paradigm shift if their dual function—as both durable structural materials and highly effective carbon sequestration media—is fully recognized and harnessed to its full potential.

To fully harness these opportunities, the following critical goals and tasks must be prioritized and addressed in the near term:

Increase research on the capacity of geopolymers for  $CO_2$  adsorption by thoroughly examining the processes of adsorption and desorption under various temperature and pressure conditions.

Conduct detailed research on the reversibility of sorption processes (CO<sub>2</sub> adsorption and desorption).

Search for alternative sources of anthropogenic raw materials to fly ash and slag, which would ensure a continuous supply and support the large-scale industrial production of geopolymers. Focus on locally occurring raw materials (e.g., <sup>131-133)</sup>)

Intensify research on substances that catalyze CO<sub>2</sub> adsorption in geopolymers (chemical sorption).

Develop methods to synthesize highly porous geopolymers while simultaneously producing zeolite structures (geopolymer production conditions must allow for the simultaneous nucleation of zeolites).

Develop a solution that leverages the efflorescence formation on geopolymers' surfaces as sodium carbonates and employs a process to recover these carbonates. This process should expose the surface, allowing new carbonate structures to form using sodium ions from the geopolymers. Numerous practical challenges still hinder the widespread application of geopolymers for CO2 capture and storage. Crucially, only solutions that are logistically feasible and scalable will have a meaningful impact. Continued support is therefore essential for efforts aimed at overcoming the implementation barriers within the construction industry. The absence of standardized procedures, construction and regulatory frameworks tailored geopolymers—unlike in the case of conventional building materials such as Portland cement—combined with limited public acceptance, significantly hinders their broader adoption. Nonetheless, the research conducted to date including the examples discussed in this article—clearly indicates that geopolymers remain one of the most promising material-based strategies for addressing excess atmospheric CO<sub>2</sub>.

#### 5. Conclusions

This review highlights recent research and developments demonstrating the feasibility of carbon dioxide sequestration within geopolymer matrices. The emerging body of literature and growing interest in this field suggest a strong scientific basis for further exploration.

Compared to Portland cement-based systems, geopolymers offer several advantages that make them particularly suitable for CO<sub>2</sub> sequestration applications, including higher porosity, thermal stability, resistance to leaching, and the potential for producing energy-efficient foamed or composite insulation materials. These features, combined with the ability to tailor their microstructure and chemistry, position geopolymers as a promising platform for multifunctional, environmentally responsive

construction materials.

Several CO<sub>2</sub> sequestration mechanisms can occur simultaneously in geopolymers, such as physical and chemical adsorption, mineralization, and carbonation. In addition to intrinsic sorption capabilities, materials can be further enhanced through modifications such as amine functionalization or controlled foaming to increase porosity and surface reactivity. Zeolites, which can form in situ during geopolymerization or be intentionally introduced, are well-known for their CO<sub>2</sub> adsorption capacity. Their integration into geopolymer systems further improves sorption performance. Additionally, novel approaches such as using carbonic acid-based or soda ash activators offer routes for integrating captured atmospheric CO<sub>2</sub> directly into the synthesis process.

Although large-scale implementation remains limited and various technical and economic challenges persist, the potential of geopolymers and alkali-activated materials for CO<sub>2</sub> sequestration may serve as a catalyst for broader industrial adoption. Continued research and interdisciplinary collaboration are essential to advancing these materials from laboratory concepts to viable large-scale solutions for climate mitigation.

#### **Author Contributions**

Research concept and design, A.P. and M.Ł.; Collection and/or assembly of data, A.P. and M.Ł.; Data analysis and interpretation, M.Ł.; A.P.; K.S.; J.P. and K.R.; Writing the article, A.P. and M.Ł.; Critical revision of the article, A.P. and M.Ł.; Final approval of the article, M.Ł.; P.R. and J.M. Funding

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