Recent progress of geopolymers for carbon dioxide capture, storage and conversion

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**A B S T R A C T**

Geopolymer materials have garnered considerable interest as one of the most promising eco-friendly inorganic options over the last decade, thanks to their remarkable properties, including mechanical, chemical, and thermal stability, cost-effectiveness, and sustainable synthesis process, enabling their use in numerous high-value applications. Meanwhile, the ever-increasing concentration of CO₂ in the atmosphere is an urgent concern for the environment and human health, forcing the development of effective CO₂ capture, storage, and transformation strategies. Various potential solutions for reducing carbon emissions have emerged with the advancement of novel materials and technologies for CO₂ capture from exhaust streams and air. Concurrently, significant studies have been conducted on utilizing geopolymers as a sustainable material for CO₂ capture. This review provides a comprehensive overview of geopolymers’ recent advances and obstacles for CO₂ management. We focus on state-of-the-art geopolymer foams and their composites, highlighting their potential for capturing CO₂. In addition, we review the use of geopolymers as catalysts or precursors for converting CO₂ into value-added chemicals and their potential for geological CO₂ sequestration. Moreover, we analyze the current limitations and opportunities for further development of geopolymers in CO₂ management. The review provides a perspective on the role of geopolymers in mitigating CO₂ effects on the environment and advancing a sustainable future.

1. Introduction

Continued population growth needs social development that consumes large amounts of energy and fossil fuels, which are still the world’s key energy resource [1,2]. With increasing fossil fuel consumption in transport, energy production, and industrial activities, the volume of CO₂ emission continues to rise and contribute to progressively severe climate change [3,4]. The average global atmospheric CO₂ concentration reached ~418 ppm in September 2023, whereas in 1958, it was 315 ppm, according to CO₂•earth data [5]. Excessive CO₂ emissions have intensified the greenhouse effect, which has negative impacts on the Earth’s environment, such as rising atmospheric temperatures, melting glaciers, and rising sea levels, and it has raised human concerns about the alterations in the ecological environment [6]. In this context, the international community has paid great attention to the concept of “carbon neutrality” in the last two decades. Therefore, scientific research has focused on reducing CO₂ emissions, CO₂ capture and sequestration, and CO₂ recycling by its conversion to chemicals and fuels while exploring and developing alternative energy sources (e.g., solar power, photovoltaic cells, wind power, and fuels from biomass) to replace conventional fossil fuels [7]. Concurrently, many studies have proposed that CO₂ capture and storage (CCS) is a sustainable approach to mitigate CO₂ emissions [8–10]. CCS involves capturing fossil CO₂ from energy-related and industrial sources and transporting it to storage locations for sequestering, or where it can be utilized in different ways like growing algae and plants, producing chemicals, fuels, or building materials, and improving oil recovery efficiency [11,12]. Researchers proposed post-combustion processes such as cryogenic distillation, membrane purification, absorption, and adsorption for CO₂ capture from point sources (Fig. 1) [13,14].

Cryogenic distillation is efficient for small volumes of CO₂ but requires high energy, preventing its use on a larger scale [15]. Membrane-based processes consume relatively little energy but are inefficient when CO₂ is present as a minor content [16]. Amine-based absorption is a mature technology for CO₂ capture, although the regeneration step of this process has high energy consumption, and...
amine species show high corrosivity [17]. CO₂ capture by adsorbents is an emerging technology due to its low energy consumption, simple operation, and no corrosion, as well as its low cost and high efficiency in the regeneration process [18–21]. Most efforts are focused on developing adsorbents with high adsorption capacity (AC) of CO₂, easy and economical regeneration process, and other parameters such as CO₂ selectivity from emission streams, good mechanical properties, and chemical and hydrothermal stability [14,22–25]. Considering these premises, the research community is designing, synthesizing, and characterizing porous sorbent materials capable of selectively adsorbing high volumes of CO₂. The porous structures of solid sorbents utilize electrostatic force or the van der Waals force between the adsorbent (solid foam) and adsorbate (CO₂) to capture CO₂ and act as a molecular sieve [26,27]. Recently, several foam solids, including zeolite, activated carbon and their composites, mesoporous silica, metal-organic frameworks (MOFs), alkali-metal-based materials, graphene, MgO-based sorbents, and covalent organic frameworks have been evaluated as potential sorbents for CO₂ capture (Fig. 2) [28,29]. They exhibited unique morphology, internal structures, and chemical nature, containing enormous narrow and homogenous pores and free surface-active sites where the CO₂ molecules are adsorbed. Thus, numerous excellent review articles have been published in past years, dealing with various strategies for CCS [30–35]. Considering the current industrial growth and economic conditions, researchers are still looking for inexpensive adsorbents with a high volume of adsorption to implement CCS approaches on a large scale. Consequently, the research community is diligently working to develop low-cost and sustainable adsorbents for CCS systems. Chouikhi et al. (2019) [22] and Tao et al. (2022) [30] reviewed the utilization of clay minerals for environmental CO₂ capture materials, including their advantages, disadvantages, challenges, and prospects in future CCS technologies. The utilization of solid wastes (e.g., steel slag, fly ash, concrete waste, calcium carbide residue, red mud, and biomass) for the development of sustainable CO₂ adsorbents is concluded by Xie et al. (2022) [31]. While various materials, such as foam solids, clay minerals, and solid waste utilization, have been examined for their potential in CO₂ capture, addressing the need for efficient CO₂ management and cost-effective production remains a priority.

One way to address the above issue is through using functional materials that should serve a multi-fold purpose, including good CO₂ management capabilities, maintaining stable properties for extended use, and being cost-effective to fabricate. Recently, a type of inorganic polymer known as geopolymer, typically formed by the reaction of aluminosilicate materials with alkaline activator solutions, has garnered attention from both industry and academia as adsorbents. This interest arises from its low cost, simple fabrication process, and unique properties such as thermal and chemical stability, long-term durability, and excellent mechanical properties [36–38]. Xu et al. (2022) [32] summarize the progress in the adsorption capability of geopolymers for metal cations, dyes, anions, and gases and their challenges and future opportunities. The potential uses of geopolymers for wastewater treatment, slag management, and energy generation are discussed by Rasaki et al. (2019) [33]. Liang et al. (2022) [34] provide an overview of current advancements in heavy metal ions removal by geopolymers, including different influencing factors (e.g., the composition of geopolymers, temperature, pH value, ions concentration, and adsorbent dosage). Concurrently, significant research has been conducted in the last few years to utilize geopolymers for CO₂ management. Freire et al. (2022) [35] recently evaluated the advantages of geopolymeric materials used for the separation or purification of CO₂ and a replacement of Portland cement for CO₂ storage wells in the gas and oil sectors. However, there is a critical gap in the existing literature regarding a review exclusively focused on recent advances in using geopolymers for CO₂ management strategies. Thus, a comprehensive and intensive discussion of the various influencing key factors, prospects, and challenges for utilizing geopolymers as a CCSU (carbon capture, sequestration, and utilization) material is essential for further progress. Concurrently, it is crucial to discuss how geopolymer promotes sustainability by offering environmentally friendly alternatives in the context of CCS strategies.

This comprehensive review thoroughly explored the potential uses of geopolymers in all stages of the carbon capture, conversion, and storage process, considering their physicochemical, textural, and mechanical properties for the first time. The influencing parameters, such as the chemical compositions of starting raw materials, solvents, reagents, and inclusion of other sorbents (e.g., zeolite, carbon) etc., on the AC have also been discussed in detail. Moreover, the potential use of geopolymers as catalyst supports for CO₂ conversion and as replacements for conventional CO₂ storage materials is also presented in detail. Finally, the possibilities and future research challenges of the geopolymers for CCS have been briefly summarized. The use of geopolymers and their composites for CO₂ capture, storage, and conversion, along with their key parameters for each, are given in Fig. 3.

![Fig. 1. Schematic illustration of carbon emissions, capture, and storage.](https://example.com/fig1)

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2. Preparation of geopolymers

Geopolymers are an inorganic polymeric system composed of amorphous or semi-crystalline 3D networks of a large number of aluminosilicates, formed by alternately units of aluminates \( [\text{AlO}_4]^{4-} \) and silicates \( [\text{SiO}_4]^{5-} \) tetrahedra [39,40]. The composition of geopolymers is usually shown as \( n\text{M}_2\text{O} \cdot \text{Al}_2\text{O}_3 \cdot x\text{SiO}_2 \cdot y\text{H}_2\text{O} \), where \( \text{M} \) is an alkali metal (Na\(^+\) or K\(^+\)) [40]. It is fabricated by activation of aluminosilicate sources using alkali solutions at ambient or elevated temperatures (< 120 \(^\circ\)C) [41,42], polycondensation reaction is expressed as [43]:

The geopolymerization mechanism of metakaolinite-alkali consists of three steps: (i) deconstruction of aluminosilicate particles, (ii) initial polymerization or gelation of dissolved silicate and alumina species into \( \text{Al-O-Si} \) and (iii) further polymerization or stabilization into large amorphous gels or growth into semi-crystalline structures, as illustrated in Fig. 4(a) [39,44]. Based on the Si/Al ratio, geopolymers form three basic units as poly(sialate) (-Si-O-Al-O-, S-type), poly(sialate-siloxo) (-Si-O-Al-O-Si-O-, SSS-type), and poly(sialate-disiloxo) (-Si-O-Al-O-Si-O-Si-O-, PSDS-type) (Fig. 4(b)) [45]. Depending on the composition of the mix, the geopolymer matrix may retain Na–Al–Si–H, Na–Si–H, and/or Ca–Al–Si–H gels. The properties of the final geopolymer product depend on the amount and characteristics of the polymeric gel formed, affected by the ratios of Al/Si, Na/Si, and Na/(Si+Al) [46]. In the early stage of this technology, the most commonly used ingredients are kaolin clay, metakaolin (MK), and zeolites with alkaline solutions (e.g., NaOH/KOH, \( \text{Na}_2\text{SiO}_3 \)) for the synthesis of geopolymers [39,43]. Later, researchers are adding numerous wastes to synthesize sustainable geopolymers, and this number continues to expand. Generally, fly ash (FA) [47,48], slag [49,50], rice husk ash (RHA) [51,52], red mud (RM) [53,54], mine tailings [55], waste glass [56], and other solid wastes are now used to fabricate geopolymers.

Porous geopolymers are fabricated by self-forming, direct foaming, the addition of porous fillers, and particle stacking or other methods, categorized by Yu et al. (2021) [36]. Direct foaming is a simple, low-cost, and quick method to create a large volume of pores inside the geopolymer using foaming agents or surfactants [38]. The size and volume of pores can be easily tailored by controlling the processing parameters, e.g., amount of foaming and stabilizing agent, slurry viscosity, curing process, etc. [57]. Generally, geopolymer slurry is prepared by mixing solid ingredients (e.g., MK, FA, or Si source) and blowing agents (e.g., Al or Si metal powders and \( \text{H}_2\text{O}_2 \)) with alkali activating solutions (e.g., NaOH, KOH, or \( \text{Na}_2\text{SiO}_3 \)) and stabilizing agents (e.g., sodium dodecyl sulfate, calcium stearate, TEXAPON® K12 anionic surfactant), as shown in Fig. 5(a) [38,57,58]. Subsequently, the curing or solidification process is performed after a slight increase in temperature (40–100 \(^\circ\)C) [36,38,57]. \( \text{H}_2\text{O}_2 \) decomposes in alkaline environments (Eq. 2) and generates

\[
(Si_2O_3 \cdot Al_2O_3)_n + nH_2O \xrightarrow{\text{MOH}} (nOH)_3 - Si - O - Al^{-1} - OH_3 + nH_2O \xrightarrow{\text{MOH}} (M) \left( - Si - O - Al^{-1} - O - \right)_n + 3nH_2O
\]
bubbles (O₂) that create voids in the geopolymers matrix [59,60]. Meanwhile, metal powders (e.g., Al or Zn) release H₂ in the NaOH solution through an oxidizing reaction, generating pores in geopolymers [61]. Recently, additive manufacturing (AM) methods have also been gaining attention for developing porous geopolymer architectures [62]. Hierarchically porous 3D structures are typically created through layer-by-layer deposition of pseudoplastic geopolymer paste in pre-planned paths from a computer-aided design (CAD). The pore size, shape, size distribution, and pore connectivity of the 3D-printed shape can be easily controlled by changing the CAD parameters [63]. Direct ink writing is an extrusion-based AM technique that is an ideal process for geopolymer slurries, and recently, a few studies have used it to develop porous geopolymer shapes, as shown Fig. 5(b) [64,65].

\[ 2H_2O_2 (aq) \rightarrow 2H_2O (aq) + O_2 (g) \uparrow \]  

(2)

3. Geopolymer based materials for CO₂ capture

Geopolymer-based adsorbents are a promising option for capturing CO₂ owing to their remarkable chemical stability, strong resistance to compression, affordable cost, adjustable textural characteristics, and low energy needs for regeneration. Thus, numerous investigations have targeted using geopolymers as adsorbents for post-combustion CO₂ capture in various CO₂-generating sectors. Recently, some patents have also been invented regarding using geopolymers for CO₂ capture [66–68]. Concurrently, several authors have analysed the integration of zeolite and activated carbon to improve the adsorption properties of the geopolymer matrix. Here, we discussed the CO₂ AC of geopolymers and various factors that influence it.

**Fig. 3.** Key parameters for capture, storage, and conversion of CO₂ using geopolymers.
Fig. 4. (a) Schematic illustration of the geopolymerization process: (i) deconstruction of MK by the activation of alkaline solution, (ii) polymerization of produced alumina/silica-hydroxy species, and (iii) stabilization of freshly generated structures. (b) Classification of geopolymer systems based on the number of siloxo Si-O units (Open access ref. [45]).
(a) Copyright with permission from Ref. [39].

Fig. 5. Schematic illustration of the fabrication of geopolymer foam by direct foaming method. (b) Flowchart of preparation of porous geopolymer lattices by direct ink writing.
(a) Copyright with permission from Ref. [58]. (b) Copyright with permission from Ref [64,65].
3.1. Geopolymers

MK-potassium silicate ((K)poly(sialate-siloxo), K-PSS) based geopolymers were synthesized without using pore forming agents, and CO₂ adsorption performance was studied by Minelli et al. (2016) \([69]\). Geopolymer samples exhibited a porosity (\(p_t\)) and Brunauer–Emmett–Teller (BET) surface area (\(s_a\)) of 30–60% and 22–50 m²/g, respectively (Table 1). It shows that an AC for CO₂ is significantly higher than other lighter gases such as CH₄ and N₂ (CO₂ >> CH₄ > N₂) due to the energetic exclusion or size exclusion or competition effects when the favour specie is present \([70]\). Therefore, the selectivity of geopolymers is considerably higher than that of many other solid adsorbents commonly employed for CCS. However, the CO₂ AC of this geopolymer is ~0.6 mmol/g at atmospheric pressure, which is lower than best performing MOFs or zeolites.

Incorporating RHA into MK-based geopolymers increases microporosity in the system, which slightly improves the CO₂ AC \([71]\). Significant higher AC (2.69 mmol/g at 1 atm) was observed in the alkali-activated FA when Al powder and propylene glycol were used as foaming agents and surfactants, respectively \([72]\). Al powder creates pores in the system through the formation of H₂ gas via the following reactions \([74,75]\):

\[
\begin{align*}
2\text{Al} (s) + 6\text{H}_2\text{O}(l) & \rightarrow 2\text{Al} (\text{OH})_3 (s) + 3\text{H}_2 (g) \quad (3) \\
2\text{Al} (s) + 2\text{NaOH} (aq) + 2\text{H}_2\text{O}(l) & \rightarrow 2\text{NaAlO}_2 (s) + 3\text{H}_2 (g) \quad (4) \\
2\text{Al} (s) + 3\text{Ca} (\text{OH})_2 (aq) + 2\text{H}_2\text{O}(l) & \rightarrow 3\text{CaO} \cdot \text{Al}_2\text{O}_3 \quad (5)
\end{align*}
\]

However, these reactions depend on the composition of FA. Propylene glycol helps generate fine gas bubbles. CO₂ absorption is slightly increased with increasing NaOH content that helps to produce carbonate ions (CO₃²⁻) by reacting with CO₂ \([72]\). The presence of a high content of calcium in FA leads to a higher CO₂ AC in the system through the following reactions \([76]\):

\[
\begin{align*}
\text{CaO} (a) + \text{H}_2\text{O}(l) & \rightarrow \text{Ca} (\text{OH})_2 (aq) \quad (6) \\
\text{Ca} (\text{OH})_2 (aq) + \text{CO}_2 (g) & \rightarrow \text{CaCO}_3 (s) + \text{H}_2\text{O} \quad (7)
\end{align*}
\]

AC of geopolymer mix increases with the geopolymerization reaction due to the increase of polysialate-PS (Si-O-Al-O) units on the surface, as observed by Freire et al. (2020) \([71]\). Concurrently, CO₂ AC increases

### Table 1

The composition, properties (\(p_t\): porosity, \(s_a\): surface area, and \(p_v\): pore volume), and CO₂ adsorption capacity of geopolymers.

<table>
<thead>
<tr>
<th>Composition</th>
<th>Properties</th>
<th>CO₂ adsorption capacity at 1 bar (mmol/g)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Solid powder</td>
<td>Alkali solution</td>
<td>Pore former</td>
<td>(p_t) (%)</td>
</tr>
<tr>
<td>MK, KOH, K₂SiO₃</td>
<td>-</td>
<td>-</td>
<td>30–60</td>
</tr>
<tr>
<td>MK, RHA, NaOH, Na₂SiO₃</td>
<td>-</td>
<td>Al</td>
<td>-</td>
</tr>
<tr>
<td>FA, NaOH, Na₂SiO₃</td>
<td>Al</td>
<td>-</td>
<td>35.9</td>
</tr>
<tr>
<td>MK, NaOH</td>
<td>Acid treatment</td>
<td>-</td>
<td>412</td>
</tr>
</tbody>
</table>

**Fig. 6.** (a) CO₂ adsorption kinetics of FA-MK and RHA-MK based geopolymers and corresponding fitting curves and parameters for pseudo-first-order kinetic model. (b) CO₂ adsorption kinetics of amine loaded geopolymers and corresponding fitting curves and parameters for different kinetic models. (a) Copyright with permission from Ref. \([71]\); (b) Copyright with permission from Ref \([73]\).
with reduced Si/Al atomic ratio, which may enhance the basic Lewis sites on the geopolymer surface. Therefore, acidic CO$_2$ can adsorb readily on the basic sites available on the geopolymer surface [77]. Geopolymers show good regeneration capacity, which implies that CO$_2$ binds to the surface due to physical adsorption [71]. CO$_2$ adsorption experimental data for FA-MK and RHA-MK based geopolymers were fitted with the pseudo-first-order kinetic model as illustrated in Eq. (8) [78]:

$$q_t = q_e [1 - e^{-k_1 t}]$$

(8)

where $q_t$ is the adsorption capacity (mmol/g) at time $t$ (min), $q_e$ is the equilibrium capacity (mmol/g), and $k_1$ (min$^{-1}$) is pseudo-first-order rate.

Fig. 7. (a) CO$_2$ AC in K$^+$-based geopolymer, zeolite Na13X, and Na$^+$-based geopolymer/zeolite composites at 35 °C. (b) Thermogravimetric data for CO$_2$ adsorption and desorption cycles at 35 °C at 1 bar on the different samples. DSC/TG curves of CO$_2$ adsorption and desorption cycles on geopolymer-zeolite composite and zeolite sorbents in isothermal conditions (35 °C) at 1 bar and with yCO$_2$ = 20%: (c) DSC curves; (d) normalized weight change. (a) Copyright with permission from Ref. [86] (b) Copyright with permission from Ref [89].
constant. CO₂ adsorption data closely align with the pseudo-first-order kinetic model shown in Fig. 6(a), suggesting that the CO₂ connection to the surface is likely due to physical adsorption [71]. Balachandra et al. (2021) [79] observed that the adsorption performance of FA-based geopolymers depends on the age of the used FA because the pH value, mineralogical compositions, and reactivity of FA changed with time. Harirchi and Yang (2022) [80] found that the CO₂ can be adsorbed during FA activation. The maximum CO₂ uptake after three days of curing was recorded at 4.8 wt% of FA-alkali precursor concentration, with a carbon sequestration efficiency of ~22.6%. The carbonation of the species in the system causes this CO₂ adsorption phenomenon.

Chang et al. (2021) [81] utilized geopolymer microspheres to immobilize the carbonic anhydrase and improve the CO₂ capture capacity of enzyme. The stable properties, high surface area, and consider-able porous structure of geopolymer microspheres behave as a promising carbonic anhydrase immobilizer that enhances the CO₂ capture potential ~5 times than the free enzyme. Functionalized amine loaded geopolymers were used as a solid sorbent for post-combustion CO₂ capture by Chen et al. (2022) [73] and Mirkovic et al. (2023) [82]. Nitric acid treated MK-based geopolymer exhibits a larger sa with a great silanol density, facilitating grafting a high amount of amine functional groups. This functionalized solid sorbents possess a CO₂ AC of 1.80 mmol/g at 60 °C and 1 atm (Table 1). The pseudo-second order kinetic and Avrami kinetic models, as illustrated in Eqs. 9 and 10, respectively [78,83]:

\[
q_t = \frac{k_2 q_e^2 t}{1 + k_2 q_e} \quad (9)
\]

\[
q_t = q_e(1 - e^{-k_3 t})^n \quad (10)
\]

where, k₂, kₙ and nₙ are the pseudo-second-order rate constant (g mmol⁻¹·min⁻¹), Avrami rate constant (min⁻¹), and the order of the Avrami model, respectively. The fitting curves (Fig. 6(b)) of the pseudo-first order (Eq. 8) and pseudo-second order kinetic (Eq. 9) models with experimental CO₂ adsorption data of amine loaded geopolymers are incongruous and show the fitting correlation coefficients of these models are 0.984 and 0.973, respectively [73]. It reveals that single adsorption kinetic models cannot accurately explain the CO₂ adsorption process because the adsorption of CO₂ on the amine loaded geopolymers is neither single physisorption nor chemisorption. However, the Avrami kinetic model (Eq. 10) exhibits a perfect fit to the experimental adsorption data with a correlation coefficient (R²) of 0.999 shown in Fig. 6(b), indicating that both physisorption and chemisorption play a role in adsorption phenomenon on amine loaded geopolymers.

The substantial pore structure, ample surface area, stable properties, and the presence of basic Lewis sites make geopolymers suitable candidates for CO₂ adsorption. Adsorption of CO₂ onto geopolymers is a surface-bound, reversible, and predominantly physical phenomenon [67,70]. The desorption process can be carried out by reducing pressure or raising temperature, which is cost-effective as it demands less energy. Concurrently, the AC of CO₂ depends on a combination of temperature-swing, pressure-swing, and surface functionalization strategies. Geopolymer materials possess attractive characteristics to selectively capture CO₂, which combined with ease of processability, good mechanical resistance, and cost-effectiveness, makes the geopolymers emerge as compelling and promising solid adsorbents.

### 3.2. Geopolymer-zeolite composites

Geopolymer-zeolite composites were fabricated by mixing synthetic zeolite as a microporous filler into mesoporous MK-based geopolymer matrices for CO₂ adsorption by Papa et al. (2018) [84]. Composite monolith based on a mixture of Na-geopolymer (PSS-type) with 27.3 wt % zeolite NaX that exhibits a compressive strength (CS) of about 3 MPa, a hierarchical porosity consisting of a wide range of porosity about 5 Å to 2 μm and a sₐ of about 211 m²/g. The inclusion of zeolite NaX significantly increased the sₐ of the composites, as 80% of the sₐ is due to the micropores. The formation of zeolite NaA phase in the geopolymer matrix increases the CO₂ AC of geopolymer by more than 50%. In alkali hydrothermal conditions, geopolymers containing Si/Al molar ratios of ~1-1.2 tend to form zeolite NaA phase [85]. Additionally, the adsorption selectivity of zeolite towards CO₂ gas is improved in the presence of an amorphous geopolymer matrix [69]. Geopolymer-zeolite composites exhibit characteristics of Type I isotherm, which is evidence of micropores existing within the solid. K⁺-based geopolymer shows considerably lower CO₂ AC than Na⁺-based geopolymer and zeolite NaX, as shown in Fig. 7(a) [86,87]. Typically, the sorption curve of CO₂ displays stepwise increases at near-zero pressure, probably due to chemical adsorption, while the gradual increase observed at higher pressures is generally attributed to physical adsorption [88]. Na⁺-based composites also exhibit larger AC than K⁺-based sorbents. The formed zeolite NaA phase has the most possible CO₂ connection sites on the surface, further promoting AC in geopolymer containing the zeolite NaA phase (Table 2).

Subsequently, Na⁺-based sorbents exhibit adsorption/desorption cycle close to zeolite NaX (Fig. 7(b)). The adsorption/desorption cycles rely on the thermal effects that can be utilized to reduce sorbent regeneration time [89]. The CO₂ adsorption enthalpies of geopolymer-zeolite composite and zeolite NaX are −29.5 ± 2 kJ/mol and −39.5 ± 2 kJ/mol, respectively. These values were determined from DSC signals, as illustrated in Fig. 7(c) [89]. The composite shows lower enthalpy than the zeolite due to the amorphous geopolymer phase, which has an adsorption enthalpy −16 kJ/mol. The adsorption and desorption kinetics in the geopolymer-zeolite composites exhibit a slightly different behaviour, with the regeneration step being slower than the adsorption step, as shown in Fig. 7(d). It indicates that the mass transport is partly impeded during the regeneration step, leading to reduced CO₂ removal from the composite adsorbent.

The CO₂ adsorption isotherms are analyzed using the dual-site Langmuir approach, as shown in Eq. (11) [86,96], which can interpret the experimental data for gas adsorption across the entire pressure range:

\[
q = q_1 \frac{b_p}{1 + b_p} + q_2 \frac{b_p}{1 + b_p} \quad (11)
\]

where, q signifies the adsorption capacity (per unit mass of adsorbent), q₁ and q₂ are the maximum adsorption capabilities of the available two

### Table 2

<table>
<thead>
<tr>
<th>Composite</th>
<th>Geopolymer</th>
<th>Other</th>
<th>Properties</th>
<th>CO₂ adsorption capacity at 1 bar (mmol/g)</th>
<th>Reference</th>
</tr>
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<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>ps (%)</td>
<td>sa (m²/g)</td>
<td></td>
</tr>
<tr>
<td>Na-MK</td>
<td>Zeolite NaX</td>
<td>47</td>
<td>211</td>
<td>0.47</td>
<td>2.5</td>
</tr>
<tr>
<td>K-MK</td>
<td>Zeolite, activated carbon</td>
<td>-</td>
<td>256</td>
<td>0.33</td>
<td>2.68</td>
</tr>
<tr>
<td>FA</td>
<td>Zeolite X, H₂O₂</td>
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<td>579.69</td>
<td>-</td>
<td>7.91</td>
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<tr>
<td>Na-MK</td>
<td>Zeolite (Na/ZSM-5)</td>
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<td>Na-MK</td>
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<td>-</td>
<td>693</td>
<td>0.358</td>
<td>2.7</td>
</tr>
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</table>

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sites, $b_1$ and $b_2$ represents the affinity constants for the two sites, and $p$ denotes the adsorption pressure. The selectivity ($s$) of the sorbents for CO$_2$ (e.g., relative to N$_2$) can be evaluated using the ideal adsorbed solution theory, which is defined as follows [90,97]:

$$s = \frac{q_C}{q_N} \frac{p_C}{p_N}$$

(12)

where, $q_C$ and $q_N$ define the CO$_2$ and N$_2$ adsorption capacities at their respective partial pressures ($p_C$ and $p_N$), as determined by Eq. (11). The specific CO$_2$ AC ($\eta$) can be calculated by dividing the weight change of the sorbent, as evaluated by TGA, by the initial weight of the sorbent ($m_0$) and the molecular weight of CO$_2$ (Eq. 13) [86].

$$\eta = \frac{m(t) - m_0}{M_{CO_2} \cdot m_0}$$

(13)

Han et al. (2022) [91] fabricated FA-based geopolymer-zeolite composite foams through in-situ zeolite X phase formation in the geopolymer gels. The ingredients were mixed and poured into plastic molds for curing, including FA, water glass, NaOH, foaming agent (H$_2$O$_2$), and foam stabilizer (calcium stearate). The molds were sealed by polyethylene plastic film to prevent moisture transfer to create a saturated steam condition and cured at 353.15 K for 48 h. The zeolite X phase is successfully precipitated in the geopolymer matrix and shows an X-ray diffraction (XRD) pattern like commercial zeolite X, as shown in Fig. 8 (a). The adsorption properties of FA-based geopolymers depend on the extent of zeolite phase formation in the matrix [98]. The in-situ composite comprises shell-like pore structures with pores of various sizes in the micron range (Fig. 8(b)). The CO$_2$ AC of composite foam increased gradually with increasing relative pressure and achieved an AC of 7.91 mmol/g, when relative pressure reached 1 bar (Fig. 8(c)). The geopolymer-zeolite X composite foam exhibits greater AC than most data available in Tables 1 and 2 due to higher AC of the adsorbent (zeolite X) and large $s_a$ of the foam. The CO$_2$ AC of zeolites generally reduces by ~15–20% during granulation using common binder materials (e.g., bentonite or kaolin clay) because they are inert towards CO$_2$ adsorption [99]. The geopolymer can solve this problem by playing a dual role as a binding agent and a sorbent [100].

Crystalline ZSM-5 zeolite phase induced in amorphous MK-based geopolymer gels via hydrothermal crystallization conversion by Chen et al. (2019) [92]. The synthesised geopolymer-Na/ZSM-5 exhibited a CO$_2$ AC of 1.79 mmol/g. AC is significantly improved when Na/ZSM-5 is modified through ion-exchange with Ni$^{2+}$ and found an AC value of 2.38 mmol/g. It happened because Ni$^{2+}$ has a higher partial reactivity index than Na$^+$, as defined by the Fukui function [101,102]:

$$f_x^+ = q_x(N + 1) - q_x(N)$$

(14)

where, $x$ denotes an atom within the cluster, N refers to the total number of electrons in the cluster, and $q_x$ represents the electronic population of the atom $x$. Moreover, the adsorption energy of Ni/ZSM-5 is also lower than that of Na/ZSM-5 for CO$_2$. The nature of the zeolite significantly influences the AC and CS of the geopolymer-zeolite composites, as observed by Lu et al. (2022) [103]. NaX zeolite-geopolymer composite
exhibits more AC than the geopolymer monolith and other composites containing NaA, X zeolite or clinoptilolite phases (Fig. 9). It happened due to 13X zeolite retaining a higher s\textsubscript{a} and pore volume than other specimens; a larger s\textsubscript{a} provides more adsorption sites, enhancing AC. However, CS reduces with the incorporation of zeolite in the composite, as shown in Fig. 9. The performance of the AC is influenced by temperature, it increases as the temperature decreases and reaches maximum at lower temperatures [104].

3.3. Geopolymer-carbon composites

The adsorption performance of geopolymer was improved by incorporating activated carbon by Chen et al. (2021) [93]. The alkaline solution in the system facilitates polycondensation of geopolymer species and functionalization of activated carbon. Hence, geopolymer serves dual roles as a binder and a surface modifier for activated carbon to prepare a monolithic adsorbent. As the activated carbon increases, the specific s\textsubscript{a} and p\textsubscript{v} increase. Concurrently, the alkali-activator destroys the graphitized C=C bonds of the carbon and increases the C-O bonds in the system (Fig. 10(a)). Thus, CO\textsubscript{2} AC of the composite showed ~2.25 times greater than the predicted value of the mix constituents (physical mixture of activated carbon and geopolymer) (Fig. 10(b)). Activated carbon (30 wt%)/phosphoric acid-based geopolymer composite exhibits higher CO\textsubscript{2}/N\textsubscript{2} IAST selectivity than pure activated carbon [105]. The CS of the geopolymer improved significantly with the incorporation of activated carbon.

3.4. Geopolymer-hydrotalcite composites

Papa et al. (2019) [106] developed geopolymer-hydrotalcite composites for CO\textsubscript{2} adsorption at intermediate temperatures (200–400 °C). Hydrotalcite belongs to a class of anionic clay group, having a general formula of [M\textsubscript{I}\textsuperscript{2+}M\textsubscript{II}\textsuperscript{3+}(OH)\textsubscript{2}]\textsuperscript{x+}[A\textsuperscript{-}\textsubscript{x/2}]\textsubscript{m}H\textsubscript{2}O (M = metal ions and A = anion, commonly carbonate) consists of positively charged layers of trivalent and divalent cations [107]. CO\textsubscript{2} AC (at 200 °C) of geopolymers increased from 0.052 mmol\textsuperscript{-1}g\textsuperscript{-1} to 0.109–0.145 mmol\textsuperscript{-1}g\textsuperscript{-1} when different types of hydrotalcite (28 wt%) were incorporated into the composite. It happened due to an increase s\textsubscript{a} and available active basic sites in the system. At lower temperatures (35 °C), CO\textsubscript{2} AC increases slightly compared to higher temperatures due to greater involvement of physisorption with chemisorption mechanism [94]. Moreover, geopolymer acts as a binder that helps build a self-supporting and designable monolith of commercial hydrotalcite powders.

3.5. Geopolymer-zeolite-carbon composites

Hierarchical porosity containing geopolymer-zeolite (in-situ NaX)-activated carbon composites were fabricated, and their CO\textsubscript{2} adsorption performance was evaluated by Candamano et al. (2022) [95]. Foaming agent and one or two thermal curing steps were used to generate pores and geopolymerization with in-situ NaX zeolite conversion in the
system, respectively. The two thermal curing steps, i.e., low temperature curing (50°C/1 h) and hydrothermal (90°C/24 h) yielded more NaX crystalline phase than direct curing at high temperature (Fig. 11). Meanwhile, pore former is more effective in increasing the surface area of the composite than adding activated carbon to the composition. Geopolymer-zeolite (in-situ NaX) with foaming agent exhibited higher AC than active carbon-foaming containing geopolymer-zeolite (in-situ NaX) composite. The micropores larger than 8 Å in activated carbon are less effective in CO$_2$ adsorption at 25°C [108,109]. Meanwhile, Can-damano et al. (2022) [95] used activated carbon contained micropores larger than 10 Å. Thus, the characteristics of activated carbon used are also important parameters for developing carbon-based adsorbents.

The porosity of geopolymer-zeolite (in-situ by hydrothermal crystallization) composites was improved by incorporating activated carbon in the matrix by Chen et al. (2020) [90]. Zeolite yields in the geopolymer by hydrothermal also help to enhance the specific $s_\text{p}$ of composites. However, the CO$_2$ AC of composites is more related to the chemical nature of the exposed surface than the available surface area [110]. The activated carbon present in the composite forms the functional C-O bonds, in-situ zeolite, enhanced surface area, and porosity significantly improved the CO$_2$ AC of the composites (Fig. 12). Concurrently, the activated carbon containing composite monolith exhibits a CS of ~23 MPa, which is higher than geopolymer-zeolite based CO$_2$ adsorbents [84,86].

Geopolymer exhibit slightly lower CO$_2$ AC compared to some of the MOFs, zeolites, or amine-based sorbents, however, geopolymer sorbents have several advantages, including facile fabrication methods, good stability, high CS, and are more economical than many high CO$_2$ adsorption materials. Concurrently, the geopolymers exhibit good selectivity for CO$_2$/N$_2$. Incorporation of porous fillers or foaming agents into geopolymer systems can greatly increase CO$_2$ AC by creating microporosity. The formation of in-situ zeolite phases by hydrothermal processes or the incorporation of zeolite fillers into the geopolymer matrix highly increases the AC of geopolymer-zeolite composites. NaX zeolite is a more suitable choice for improving the CO$_2$ AC of geopolymers than other zeolite phases. Geopolymer can serve as an advanced binder system to produce activated carbon-based monolithic adsorbents, as it also aids in functionalizing carbon surfaces. Furthermore, geopolymer can be used to develop advanced CO$_2$ detection tools due to its unique properties, such as absorbing, storing CO$_2$ and selective uptake of CO$_2$ over other gases. Geopolymers can act as a substrate or sensor material for monitoring CO$_2$ levels in various environments. This can help in tracking the effectiveness of carbon capture and storage systems, as well as in developing new ways to detect and quantify CO$_2$ concentrations.

4. CO$_2$ conversion using geopolymers as catalyst supports

With the advancement of science and technology, the research community is trying to utilize CO$_2$ as a raw material to deliver carbon sources. The capture and conversion of stable CO$_2$ molecules into usable chemicals or fuels are the most inspiring carbon neutral options existing today [111,112]. Commonly, converting CO$_2$ into industrial products or fuels such as HCOOH, H$_2$COCH$_3$, C$_2$H$_4$, CH$_3$OH, CH$_4$, and C$_2$H$_4$OH has attracted the attention of governments and scientists worldwide [113-115]. CO$_2$ is thermodynamically passive, hence, there is an imperative need for efficient and economical carbon dioxide activation processes and catalysts. Thus, the current research in this field involves designing an efficient, low-cost, and stable catalyst for CO$_2$ hydrogenation or methanation [116,117]. In the past few years, the use of geopolymers in the domain of heterogeneous catalysis has started to draw the attention of researchers [118]. In geopolymers, the monovalent charges of alumina tetrahedra are balanced by extra-framework alkali cations, typically Na$^+$ or K$^+$, which can undergo ion exchange to build geopolymers with zeolite-like behaviours. Consequently, geopolymers can be evaluated as the amorphous phase of certain synthetic zeolites [119] and can provide catalyst support. Furthermore, geopolymers have good thermal stability, mechanical properties, and an inexpensive fabrication process. Thus, researchers study the potential of geopolymers as catalytic materials for wastewater treatments, biodiesel synthesis, hydrogen production, and gas purification [118,120-122].

Na-MK based geopolymer microsheres were fabricated using a one-pot method for CO$_2$ hydrogenation by Ye et al. (2022) [123]. The prepared geopolymer slurries were injected dropwise into a heated (60°C) silicone oil bath with continuous stirring (800 rpm) (Fig. 13(a)). The developed microsheres were cured in an oven (60°C) for 24 h and then washed in boiled water. The geopolymer microsheres were heated at 500°C and washed again with water. Dry microsheres were used for catalyst support. Geopolymer based catalysts were prepared by adsorbing the metal cations (Pt, Pd, and Ni) from the respective solutions (nitride or chloride) at 80°C with continuous stirring (200 rpm). Alkali metals (e.g., Na$^+$ or K$^+$) in geopolymers positively affect the dispersion of active metals, facilitating the development of small-sized metal clusters distribution. The hydroxyl groups of geopolymers also promote the homogeneous distribution of metal cations on the surface of the catalyst support. No CO$_2$ conversion activity was observed for geopolymer up to 550°C, but geopolymers containing active metals started conversion activity at 350°C (Fig. 13(b)). Ni-geopolymer exhibits higher CO$_2$ conversion rates than Pt and Pd due to low CO selectivity. Almost no loss of catalytic activity was observed over six consecutive cycles for this geopolymer supported catalyst (Fig. 13(c)).

Wan et al. (2022) [124] used synthesized slag to fabricate geopolymer microsheres in a one-pot method, and Ni-loaded microsheres were further used for CO$_2$ methanation. The microsheres retained wedge-shaped holes and a large specific $s_\text{p}$ which facilitated the dispersion of Ni species (Fig. 14(a)). CO$_2$ species are adsorbed by alkali line sites and OH groups on the geopolymer surface. Moreover, the presence of Ca$^{2+}$ and Mg$^{2+}$ in the geopolymer increases the alkaline sites on the catalyst surface, which enhances the adsorption of CO$_2$ [125]. The Ni-geopolymer catalyst showed no activity up to 200°C and exhibited an optimal reaction temperature of ~400°C (Fig. 14(b)). This catalyst exhibits CO$_2$ conversion efficiency and CH$_4$ selectivity of 80% and 99% at 400°C/0.1 MPa, respectively (Table 3). Generally, CO$_2$ methanation reactions proceed by two pathways, first is the dissociation of formate into CO$_2$, which is then hydrogenated to form CH$_4$, and the
other is direct hydrogenation of formate to generate CH₄ [126]. However, Ni-geopolymer catalyst produces CH₄ via direct hydrogenation of the formate pathway, as depicted in Fig. 14(c). At the interface of Ni-geopolymer, CO₂ reacts with OH groups to produce bicarbonate or carbonate. Typically, H₂ is adsorbed by the Ni species and decomposed to H⁺, which then reacts with the carbonate to create formate and further produces CH₄ via hydrogenation. Nitric acid treated blast furnace slag supported Ni-CeO₂ catalyst achieved a CO₂ methanation of ~81% at 350 °C with long-term stability (100 h) [127]. The presence of CeO₂ reduced the agglomeration, increased the dispersion of Ni particles at high loadings, and enhanced active catalytic basic sites, which improved the catalytic ability at low temperatures (350 °C). Schuster et al. (2023) [128] developed functionalized geopolymer-based electrodes with Sn for the electrochemical reduction of CO₂ to formate. This innovative approach yielded formate concentrations of ~22.7 mmolL⁻¹ and a corresponding current efficiency of 14% at a current density of
which is like the carboxylate pathway. The *COOH is an intermediate conversion over the Cu-geopolymer catalyst is illustrated in Fig. 15 (a), 20 mA cm$^{-2}$

Catalytic conversion of CO$_2$ using geopolymer support catalysts.

<table>
<thead>
<tr>
<th>Catalyst support</th>
<th>Catalyst</th>
<th>CO$_2$ conversion</th>
<th>Selectivity</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Na-MK geopolymer</td>
<td>Pt</td>
<td>55% 550°C CO$_2$</td>
<td>97%</td>
<td>[123]</td>
</tr>
<tr>
<td>Slag-based K-geopolymer</td>
<td>Ni</td>
<td>80% 400°C CH$_4$</td>
<td>99%</td>
<td>[124]</td>
</tr>
<tr>
<td>Blast furnace slag</td>
<td>Ni-CeO$_2$</td>
<td>81.6% 350°C CO$_2$</td>
<td>99.8%</td>
<td>[127]</td>
</tr>
<tr>
<td>Slag-based Na-geopolymer</td>
<td>Cu</td>
<td>48% 550°C CO$_2$</td>
<td>96%</td>
<td>[129]</td>
</tr>
</tbody>
</table>

where, $n_{\text{CO}_2}^\text{in}$ and $n_{\text{CO}_2}^\text{out}$ represent the flow rates of CO$_2$ in the feed and the outlet, respectively. The presence of Cu$^+$ species on the catalyst surface promotes CO$_2$ activation. A possible reaction pathway for CO$_2$ conversion over the Cu-geopolymer catalyst is illustrated in Fig. 15(a), which is like the carboxylate pathway. The *COOH is an intermediate meta-stable compound in converting CO$_2$ to CO. The carbonate species react with OH groups on the geopolymer surface to produce HCO$_3^-$ species. *COOH is formed via the reaction of HCO$_3^-$ species with Cu adsorbed H$_2$. H$^+$ reacts again with the developed “COOH on the Cu surface to generate CO” and OH$^-$, and further, CO and H$_2$O dissociate from the catalyst surface. After 100 h on steam, the Cu-geopolymer showed ~95% of its initial conversion capacity (Fig. 15(b)), suggesting that the catalyst is also stable for the long-term.

The presence of Na$^+$ and hydroxyl on the geopolymer surface facilitates the dispersion of active metal nanoparticles to achieve adjustable active sites containing single-atom catalysts. Geopolymer support can design atomically dispersed or highly nanoparticles dispersed catalysts for target substance fabrication. Geopolymers have excellent carrier performance with long-term stability, converting CO$_2$ into CO intermediates or directly into value-added raw materials or products. Geopolymers can be customized to support various catalysts, including those for CO$_2$ conversion. Geopolymers are chemically inert and can withstand various chemical conditions and temperatures, which is important in CO$_2$ conversion processes. Researchers can also modify the geopolymer’s chemical composition and structure to optimize its compatibility with specific catalysts, enhancing the overall performance of the catalytic system. Thus, geopolymers can play a significant role in developing a new generation catalytic system for CO$_2$ conversion by providing a stable and versatile support material for catalysts. It encourages the utilization of CO$_2$ greenhouse gas as a carbon source and environmental protection.

5. Geopolymers for CO$_2$ storage wells

Among the various CO$_2$ emission reduction processes, one of the best proposed is to capture CO$_2$ and injection it into deep underground formations (such as depleted oil, gas reservoirs, and saline aquifers), known as geological CO$_2$ sequestration (Fig. 16(a)), which seems especially attractive because it provides a long-term and environmentally satisfactory solution for the decrease of CO$_2$ emission into the atmosphere [130]. Moreover, this process significantly affects the efficiency of oil or gas recovery, where CO$_2$ is utilized to increase the pressure of depleted reservoirs, which helps to recover and increase gas or oil production [131]. However, materials selection for injection and production wells is challenging because of the high downhole force and corrosive environment (CO$_2$ in water forms carbonic acid), which causes cracking and corrosion in the wellbore [132]. Consequently, the integrity of the wellbore should be constructed using a material that is anti-corrosive, durable, chemically inert, and adaptable to temperature and pressure variations to prevent leakage of CO$_2$ [133]. Portland based oil cement is typically used for wellbore construction. There are still durability issues in wet CO$_2$ environments that create porosity, increase permeability, and reduce the strength of the cement structure [134]. The research community is trying to find alternative cement systems for wellbore applications.

Gouedard et al. (2008) [137] invented a pumpable geopolymer suspension using metakaolin and sodium disilicate for use in CO$_2$ storage wells. CO$_2$ permeability in various types of geopolymers at different injection pressures, like deep down-hole environments, was investigated by Nasi et al. (2014) [135]. The CO$_2$ permeability in the geopolymers is significantly lower than that of class G oil cement, and when slag is incorporated into the geopolymer mix, the permeability further reduces (Fig. 16(b)). The CO$_2$ permeability is directly related to the connectivity and structure of pores in the system. G cement possesses more courser pores, whereas geopolymer shows finer pore diameter, accounting for the lower permeability of geopolymers [138]. The permeability of
CO₂ permeability increases with increasing curing temperature for a given injection pressure condition (Fig. 16(c)). At low temperature curing for a long time, the geopolymer shows a low porosity and pore connectivity, and uniform microstructure, whereas geopolymers cured at high temperature exhibited spherical pores containing a poor geopolymer matrix due to rapid hardening, and the microstructure does not improve with aging [139]. Thus, pore connectivity in the low temperature curing samples reduces through further reaction, typically in additional aging time, leading to low permeability. Concurrently, permeability of geopolymers decreases with increasing inlet pressure due to slip flow, as suggested by Klinkenberg [140]. However, CO₂ permeability of geopolymers (0.0004–0.04 µD) is much lower than that of OPC-based wells (1–150 µD) under all test conditions (different curing temperature and inlet pressure) [136]. Geopolymers exhibit a minimal weight variation of up to 420 °C, indicating that geopolymers can use a wide range of temperatures [131]. In CO₂-rich environments, geopolymers possess significantly lower permeability, excellent thermal stability, and mechanical properties than OPC-based wells under down-hole stress positions [141,142].

The physisorption of CO₂ on the geopolymer surface depends on the temperature and CO₂ gas pressure [143]. In wet CO₂ injection conditions, FA-based geopolymers form calcium carbonate and zeolite on the surface, modifying the microstructure and enhancing strength [144–146]. Calcium carbonate polymorphs (e.g., calcite, aragonite, and vaterite) are precipitated all over the lime activated slag-based geopolymers in CO₂ environment at 100 °C [147]. Nasvi et al. [148] measured the CS of geopolymers in a CO₂ environment for up to 6 months and found no significant changes. Concurrently, geopolymers exhibit good stability in different hydraulic fracturing conditions (e.g., injection pressure, axial stress, confining pressure, and tube length). The geopolymers have low CO₂ permeability, high mechanical strength, and excellent durability in corrosive environment. Thus, alkali activated geopolymers can be used as a replacement for traditional sealant materials for a typical wellbore. Concurrently, geopolymers can also be a stable and durable material for long-term carbon storage. Once CO₂ is captured, it can be stored within the structure of the geopolymer. This creates the opportunity for secure and long-lasting carbon sequestration.

6. Challenges and future perspectives

With the increase of CCS projects in recent years, many new sorbent materials have been evaluated or developed. Over the last few decades, geopolymers have been considered a potential eco-friendly adsorbent for CCS projects. However, it is also clear that significant improvements to the state of the art of porous geopolymers are necessary in various directions for industrial implementation, as follows:

- The use of geopolymer as sorbents is still limited to the laboratory scale. Overcoming issues such as the significant leaching of sodium...
or potassium into the medium is crucial, as it can lead to equipment corrosion and encrustation [149].

The presence of the zeolite phase in the geopolymer matrix greatly increases the AC, and has also been evaluated. Synthetic zeolites are mostly expensive, which increases the production cost. Thus, further research is required to achieve the precise in-situ formation of zeolitic phases in the system during geopolymerization.

Additional studies are needed to determine the most appropriate processing techniques and synthesis conditions for designing geopolymer foams with enhanced properties, such as controlled pore size and volume, mechanical strength, stability, AC, and regeneration capacity. It is worth exploring innovative processing techniques such as additive manufacturing to create geopolymer monoliths with highly complex shapes and improve their performance by better controlling the pore size distribution. Despite the exciting opportunities offered by 3D printing, there are some engineering challenges that need to be overcome, such as the time-dependent viscosity of slurries and the setting times associated with geopolymerization reactions.

The research community is focused on using industrial waste for low-cost and environmentally friendly production of geopolymers. However, ensuring homogeneity in the physical and chemical properties of geopolymers remains a challenge, which restricts their large-scale processing and application. Thus, it is essential to develop standard manufacturing procedures to accommodate the diverse physical and chemical properties of various raw material streams.

The combination of multiple manufacturing techniques appears to be very promising, particularly in producing open cell porous geopolymer monoliths. The process can be adapted from fabrication processes used in other porous materials such as ceramics, cements, and polymer. Incorporating a wider range of lightweight, inherently micro-, and meso-porous fillers (e.g., micro, and nano porous silica and carbon), along with foaming or additive manufacturing techniques for geopolymer slurries, has the potential to create more intricate and efficient geopolymer monoliths that possess a multi-scale porous structure.

The performance of sorbents for adsorption and selection is influenced by temperature. However, the high temperature adsorption performances of geopolymers have not been extensively investigated, and more studies are necessary to obtain a comprehensive understanding of their capability for adsorption at elevated temperatures.

Regenerating geopolymer materials for CO₂ adsorption is crucial for sustainable use. Efficient regeneration processes must be developed to remove and capture CO₂ effectively, ensuring long-term utility. Scaling up geopolymer production and application can reduce costs through economies of scale, making the technology more accessible to various industries. Geopolymers show promise in mitigating CO₂ emissions but addressing regeneration, scale-up, and cost-efficiency is vital for widespread adoption.

Developing a fundamental comprehension of the interactions between CO₂ and sorbents and achieving mastery of surface binding activities that determine the selective uptake of CO₂ from flue gases will accelerate the progress of advanced materials with excellent sorption capabilities. The design of catalysts using earth abundant materials and understanding the basic mechanisms involved in reducing CO₂ into hydrocarbons and fuels on catalytic surfaces can facilitate using and recycling this carbon source to produce practical products.

As a sealant material for geological CO₂ storage, geopolymer may be a suitable replacement for Portland cement. However, new activators should be developed to overcome the limited geopolymerization conditions, i.e., high curing temperatures, and enhance the performance of geopolymer cement.

Molecular dynamics simulation studies are a relatively new approach to understanding the thermodynamic properties, bonding information, and structural formation mechanisms of materials. Furthermore, this approach has the potential to reduce the need for expensive experimental equipment and the time required for experiments. Consequently, there is a need for molecular dynamics simulation studies on geopolymers to design an accurate adsorption model and choose an appropriate forcefield.

Life Cycle Assessment (LCA) of geopolymers for CCS is necessary for a comprehensive understanding of the environmental and sustainability implications of this innovative technology. LCA will allow researchers and industries to assess the entire life cycle of geopolymer-based CO₂ adsorption and storage systems, considering factors such as raw material extraction, production processes, transportation, usage, and end-of-life disposal. LCA will assist in optimizing geopolymer technologies for a more effective and eco-friendly approach to CO₂ capture and mitigation.

Cost assessment is also crucial for the implementation of geopolymer-based materials for CO₂ management. It enables the comparison of geopolymer-based CO₂ capture with other existing or emerging carbon capture technologies, aiding in the selection of the most economically viable option. A detailed cost assessment can reveal opportunities for cost reduction through process optimization, technological advancements, and economies of scale. The environmentally friendly and sustainable nature of geopolymers can contribute to long-term cost savings by reducing the carbon footprint associated with CCS technologies.

Finally, the CO₂ AC of geopolymer is significantly lower than that of some nanomaterials, zeolites, and MOFs. Hence, further efforts should be made to tailor adsorptive properties of geopolymers and their composites to improve CO₂ adsorption and separation performance.

7. Conclusions and outlook

The geopolymer technologies could become more widely used if they are adopted for environmental remediation, renewable energy production, and other multifunctional applications. The review offers a thorough overview of how geopolymers can be used for CO₂ capture, storage, and conversion. Among all the options for adsorption, geopolymer-based sorbents derived from waste materials are economical, environmentally friendly, and comparatively easy to fabricate. Generally, geopolymer-based adsorbents exhibit a CO₂ AC lower than zeolites, MOFs, and amine-based sorbents. Thefoaming agents (e.g., H₂O₂ solution, Al, and Si powder) and porous fillers (e.g., rice husk ash) can improve the AC of geopolymer by increasing its specific surface area. Recently, there has been growing interest in using geopolymer-zeolite and geopolymer-carbon composites as CO₂ adsorbents. Zeolites and activated carbon are effective fillers that can modify the number of active functional sites and increase the s₅ of geopolymers, thereby expanding their adsorption capacities and potential applications in geopolymer-based composites. The in-situ formation of zeolite phases (preferably NaX zeolite) by hydrothermal processes and high levels of calcium and sodium in geopolymer compositions can lead to a high CO₂ AC. Meanwhile, geopolymers can be used as a binder system to produce structured adsorbents.

The presence of Na⁺/K⁺ and hydroxyl groups on the geopolymer surface makes it favourable to the dispersion of active metal nanoparticles, which can design atomically dispersed or highly nanoparticles dispersed heterogeneous catalysts for target substance fabrication. Geopolymer materials exhibit long-term stability with good carrier performance that can be used as low-cost catalyst supports for converting CO₂ to CO intermediates or directly to value-added products. Concurrently, geopolymers possess significantly low permeability, excellent thermal stability, and mechanical properties, making them a potential candidate for a substitute for OPC-based wells in geological CO₂ sequestration. FA and slag-based geopolymer modify the microstructure in wet CO₂ injection by forming calcium carbonate and zeolite
on the surface, enhancing stability in different hydraulic fracturing conditions.

The geopolymer technology is still immature for CCS applications with respect to processing techniques and adsorption capacity. However, a great number of studies are still required to realize the potential of geopolymer technology for adsorption applications in industries. We expect that future research efforts will uncover new opportunities for greener and porous geopolymer in the CCS applications and low-cost and durable geopolymer for geological CO₂ sequestration and high ion exchange-capable geopolymer for heterogeneous catalyst supporters.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data Availability

Data will be made available on request.

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