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► To cite this version:

Abdelkader Labidi, Haitao Ren, Qiuhui Zhu, Xinxin Liang, Jiangyushan Liang, et al.. Coal fly ash and bottom ash low-cost feedstocks for CO₂ reduction using the adsorption and catalysis processes. *Science of the Total Environment*, 2024, 912, pp.169179. 10.1016/j.scitotenv.2023.169179 . hal-04528393

HAL Id: hal-04528393

<https://hal.science/hal-04528393v1>

Submitted on 1 Apr 2024

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Review

Coal fly ash and bottom ash low-cost feedstocks for CO₂ reduction using the adsorption and catalysis processes

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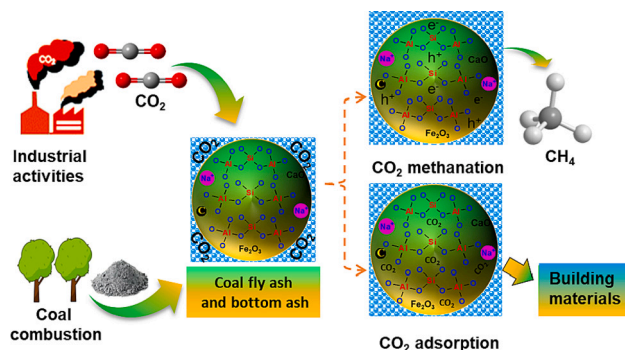
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- CO₂ emission in the atmosphere is the essential driver of climate change.
- The coal ashes usages for CO₂ adsorption and catalytic reduction are evaluated.
- Mechanistic insights of CO₂ capture and its selective reduction into CH₄ are described.
- Practical aspects for the real-world applications are discussed.
- Adsorption and catalysis are attractive approaches for CO₂ remediation technologies.



ABSTRACT

Combustion of fossil fuels, industry and agriculture sectors are considered as the largest emitters of carbon dioxide. In fact, the emission of CO₂ greenhouse gas has been considerably intensified during the last two decades, resulting in global warming and inducing variety of adverse health effects on human and environment. Calling for effective and green feedstocks to remove CO₂, low-cost materials such as coal ashes “wastes-to-materials”, have been considered among the interesting candidates of CO₂ capture technologies. On the other hand, several techniques employing coal ashes as inorganic supports (e.g., catalytic reduction, photocatalysis, gas conversion, ceramic filter, gas scrubbing, adsorption, etc.) have been widely applied to reduce CO₂. These processes are among the most efficient solutions utilized by industrialists and scientists to produce clean energy from CO₂ and limit its continuous emission into the atmosphere. Herein, we review the recent trends and advancements in the applications of coal ashes including coal fly ash and bottom ash as low-cost wastes to reduce CO₂ concentration through adsorption and catalysis processes. The chemical routes of structural modification and characterization of coal ash-based feedstocks are discussed in details. The adsorption and catalytic performance of the coal ashes

Keywords:

Adsorption
Bottom ash
CH₄ production
Catalytic conversion
Coal fly ash
CO₂ emission

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derivatives towards CO₂ selective reduction to CH₄ are also described. The main objective of this review is to highlight the excellent capacity of coal fly ash and bottom ash to capture and selective conversion of CO₂ to methane, with the aim of minimizing coal ashes disposal and their storage costs. From a practical view of point, the needs of developing new advanced technologies and recycling strategies might be urgent in the near future to efficient make use of coal ashes as new cleaner materials for CO₂ remediation purposes, which favourably affects the rate of global warming.

1. Introduction

Nowadays, air pollution and global warming have attracted great attention as the two important issues that threaten the human health and whole environment seriously (Khan et al., 2023; Li et al., 2022; Lim et al., 2023; Yan et al., 2023; Yoo et al., 2022; Yue et al., 2023; Zhou et al., 2021). Over the past few years, the population growth has increased the greenhouse gases demand (predominantly carbon dioxide, CO₂) through vehicles and industrial activities, which led to the huge emissions of gases in the atmosphere (Bao et al., 2023; Hu et al., 2022; Li et al., 2022; Mustafa et al., 2023; Sun et al., 2022; Zeng et al., 2022). Gaseous compounds such as CO, CO₂, NO_x (NO and NO₂), SO₂, CH₄, H₂S, fluorinated gases and volatile organic compounds (VOCs), have been identified by the World Health Organization (WHO) as the highest risk around the world and caused 4.2 million deaths per year due the harmful direct and indirect impacts on human health (Duguid et al., 2022; Hu et al., 2022; Leroutier and Quirion, 2022). According to the United States Environmental Protection Agency (USEPA), CO₂ is classified as the top list of the emitted gases (79 %) succeeded by methane (11 %), nitrous oxide (7.0 %) and fluorinated gases (3.0 %) (Emissions and Change, 2017). Consequently, government policies as well as social and industrial mobilisation have been developed to resolve this environmental problem and mitigate air pollution (Han et al., 2022; Kamkeng and Wang, 2023; Ma et al., 2022; Mei et al., 2022; Saravanan et al., 2022).

Regarding CO₂ pollution, human activities (i.e., transportation, electricity generation, agriculture, etc.) and industry sectors are the main sources of its emissions which have considerably risen over the past few years (An et al., 2023; An et al., 2022; Emissions and Change, 2017; Zhou et al., 2023). CO₂ in the atmosphere is, nowadays, considered as the principal cause of climate change (Ritchie et al., 2020). No doubt that oil-manufacturing countries are the major producers of carbon dioxide. According to the reports, countries with low population, such as Qatar, Trinidad and Tobago, Brunei, United Arab Emirates, Bahrain, Kuwait and Saudi Arabia are classified as the countries causing the greatest CO₂ emissions (>20 t/person) (Ritchie et al., 2020). Moreover, due to the population number, countries such as Australia (17 t/person), United States (16.2 t/person) and Canada (15.6 t/person) represent the highest total emissions of CO₂ in the atmosphere (Ritchie et al., 2020). On the other hand, Asian continent releases more CO₂ gas (53 %), compared to America, Europe and Africa, due to the high number of people in this continent. Interestingly, China is classified in the top list of CO₂ release with 10 billion tonnes of CO₂ emissions (Ritchie et al., 2020). As far as known, high CO₂ concentrations in the atmosphere can increase the level of ozone pollution and thereby the increase of earth temperature, resulting in the heat waves, hurricanes and fires, plague of ticks and mosquitoes as well as the harmful diseases such as Lyme disease, Zika, Dengue, and West Nile, respiratory illness, cardiovascular diseases, lung cancer, allergies and asthma (Apergis et al., 2018; Chaabouni and Saidi, 2017; Shindell et al., 2018). Notably, CO₂ is extensively emitted in China because of the rapid development of agriculture and industrial sectors, fuel combustion, residential sources and transportation (Ai et al., 2022; Cao et al., 2022; Miao et al., 2023b; Truong and Mishra, 2021; Zhu et al., 2023). From the ideas above statements, one may deduce that air pollution caused by CO₂ emissions is a serious problem which should be dealt with urgently. In this case, various protocols and strategies are developed by the governments to

control the emissions of this gas that present a real challenge for ozone mitigation and life existing (Cao et al., 2022). To reach the brilliant spots, several issues must be dealt with in regard with the CO₂ capture technologies.

For years, industrial companies and researchers have been mobilized to resolve this issue employing various methods of CO₂ reduction such as photocatalysis, oxidative adsorption methods, electrolysis, photo-electrochemical techniques, etc. (Cheng et al., 2021; Chu et al., 2018; Fu et al., 2022; Gao et al., 2020; Jiang et al., 2022; Masel et al., 2021; Ochedi et al., 2021). So far, some excellent efforts have especially focused on the preparation of highly reactive catalysts using metal oxides (i.e., VO_x, MnO_x, CeO₂, Fe₂O₃, CuO, etc.) (Jiang et al., 2022). Other inorganic and organic materials like mesoporous silica, metal-organic frameworks (MOFs), porous organic polymers (POP_s), covalent-organic frameworks (COF_s), zeolites, activated carbon and clay, have been also utilized for CO₂ reduction purposes (Kumar et al., 2020; Mandal et al., 2020; Ochedi et al., 2021; Sun et al., 2021; Zhang et al., 2022). It is worthy to note that these materials are widely applied for CO₂ reduction owing to their high specific surface area, high efficiency and excellent porosity. Until recently, eco-friendly, biodegradable and sustainable candidates have become widely utilized for CO₂ sensors development. Moreover, in order to develop new low-cost materials, coal ashes including coal fly ash and bottom ash obtained from coal combustion exhibited high efficiency and cost-effective materials due to their availability as wastes that can be reused in CO₂ reduction technologies. Till now, great progress has been made in coal ash-based materials for CO₂ adsorption and catalytic conversion processes. Google scholar search results show that during the last decade (2009–2019) (Fig. 1), there have been 17.700 articles with CO₂ adsorption on coal fly ash as the keyword and 17.800 articles with CO₂ conversion by coal fly ash as the keyword, but the number of reviews is <20 % for both processes. Regarding coal bottom ash, there have been 17.600 articles with CO₂ adsorption on coal bottom ash as the keyword and 17.700 articles with CO₂ conversion by coal bottom ash as the keyword and similarly to coal fly ash, the number of review papers is <20 %. Besides, most review papers only introduce coal fly ash and bottom ash as low-cost wastes for CO₂ mineralization and their applications in construction sector. Additionally, preparation methods and properties of coal ash-based materials has been discussed in some literature. It is worthy to note that during this period the number of publications focusing on fly ash uses are higher than coal bottom ash. Furthermore, this number of publications has increased from <2200 publications in 2009 to >5000 publications in 2019 for both inorganic feedstocks according to google scholar (Fig. 1). From the above-observation, coal fly ash and bottom ash inorganic feedstocks could afford a sustainable platform to produce highly efficient cleaner materials for CO₂ reduction technologies.

Interestingly, the functionalization of coal ashes by introducing organic and inorganic entities on their surfaces also improves their performances in CO₂ reduction reactions. In this review, the use of coal ashes as cost-effective inorganic feedstocks to capture and the catalytic reduction of CO₂ are addressed in particular. The design, preparation methods and characterization of porous materials based on coal ashes including coal fly ash and bottom ash are outlined. The benefits and drawbacks of coal ash derivatives for capturing and the catalytic CO₂ conversion into CH₄ are also highlighted. An overview of the plausible mechanisms reported by the previous researchers is also provided to further clarify the future perspective at the end (Fig. 2).

2. Coal fly ash and bottom ash

2.1. Coal fly ash

Coal fly ash (also named fly ash) is an industrial fine powder produced from the burning of pulverized coal and captured by electrostatic precipitators (Lv et al., 2022; Opiso et al., 2019). Since 2017, the top producers of coal ash are China and India with annual production of 780 million tonnes per year (Luhar et al., 2021). Coal fly ash contains SiO_2 , Al_2O_3 , Fe_2O_3 , CaO , Na_2O , SO_3 , MgO , K_2O , unburned carbon (10 wt %) and trace metals such as mercury, arsenic and lead which occur the existence of various crystal varieties e.g., reyerite, srebrodolskite, larnite, dolomite, and brownmillerite in its structure (Hong et al., 2022; Peng et al., 2023). Fly ash is chemically stable and does not leach harmful chemicals into the environment. This material has thermal insulation properties which arise from the specific inorganic crystalline phases inside the bulk structure. Fly ash can be sorted as two classes based on SiO_2 , Al_2O_3 , Fe_2O_3 contents: class (C), in which the amount of Fe_2O_3 , Al_2O_3 , and SiO_2 , is $>50\%$ and class (F) in which the total amount of SiO_2 , Al_2O_3 and Fe_2O_3 exceeds 70%. In general, the size distribution, specific surface area, surface morphology and crystal structure make fly ash a potential material with multiple advantages for the environmental and industrial applications (Hong et al., 2022). Taking a look at the recent field studies, fly ash has been especially utilized in cement/concrete as construction materials, road base, flowable, mining applications and soil modification (Makgabutlane et al., 2022; Yang et al., 2019). Also, this material showed various applications in agricultural sector as an amendment in soil (Table 1). As a strategy to limit its disposal into the environment, it should be recycled and reused since it has low-cost and sustainability properties. Indeed, coal fly ash waste possesses several useful properties that make it a versatile material for numerous applications considering its environmental benefits.

2.2. Bottom ash

During the coal combustion, two kinds of powder can be obtained: coal fly ash and coal bottom ash (also called bottom ash) with dark grey colour (about 10–20%) collected from the bottom of furnaces (Singh, 2018; Zhou et al., 2022). This granular, sandy material waste is composed principally of silica, alumina, sulphates oxides with smaller percentages of trace metals such as calcium, magnesium and iron, unburnt carbon (7.5 wt%) and other compounds like trace metals (Alam et al., 2019). Coal bottom ash can be classified into two major categories

belonging to class F (SiO_2 , Al_2O_3 and Fe_2O_3 ($>70\%$)) and class C (CaO and a specific amount of SiO_2 , Al_2O_3 and Fe_2O_3 ranging from 50 to 70 wt%), respectively (Wang et al., 2022). Like coal fly ash, bottom ash has some physicochemical and mechanical proprieties that make it an excellent material act like sand in building sector (Chen et al., 2023; Ge et al., 2019; Muthusamy et al., 2020). Bottom ash has a unique drainage, porosity, and permeability (Table 1). It possesses good filtration properties due to its high porosity and permeability. These properties make it intensively employed in various construction, drainage and filtration applications. Moreover, it has shear strength, which makes it ideal for use in civil and geotechnical engineering applications such as sub-base material for highways, embankments and levies. Consequently, the high demand of cement, concrete and brick manufacture encourages the exploitation of bottom ash and reduces its disposal and storage cost. Moreover, the textural properties of bottom ash make it a versatile material for various applications in construction, engineering and industries (Table 1). Its wide uses and unique properties can provide environmental and economic benefits, especially for CO_2 reduction (Fernando et al., 2022; Muthusamy et al., 2020; Orozco et al., 2023).

3. Removal of CO_2 by coal ash-based adsorbents

3.1. Examples of adsorbents

Adsorption process using natural materials is considered as an interesting topic of recent researches and a cost-effective process for CO_2 removal (Giraldo et al., 2020). In the past few years, several works investigated the adsorption of CO_2 gas using sustainable and low-cost solid adsorbents (Heidari et al., 2022; Li et al., 2020; Pardakhti et al., 2019). Moreover, low-cost adsorbents, such as coal ash and their derivatives, have been given great attention and classified as potential materials for CO_2 removal through adsorption pathway. It has been also proven, in some studies, that coal ashes modification by introducing inorganic and organic groups on their surfaces enhances their potential for CO_2 capture. In this section, the efficient coal ash-based adsorbents for CO_2 reduction are described.

In a recent work, silica-alumina aerogels were fabricated from coal fly ash using sol-gel method for CO_2 adsorption (Guo et al., 2023). The authors applied X-ray diffraction (XRD), scanning electron microscopy (SEM) and X-ray fluorescence (XRF) to demonstrate the new structure, surface area and mesopores of the adsorbent. They concluded that good microscopic properties of the as-synthesized adsorbent with the specific pore volume ranging of 1.9 to 400 $\text{m}^2 \cdot \text{g}^{-1}$ of coal fly ash could be

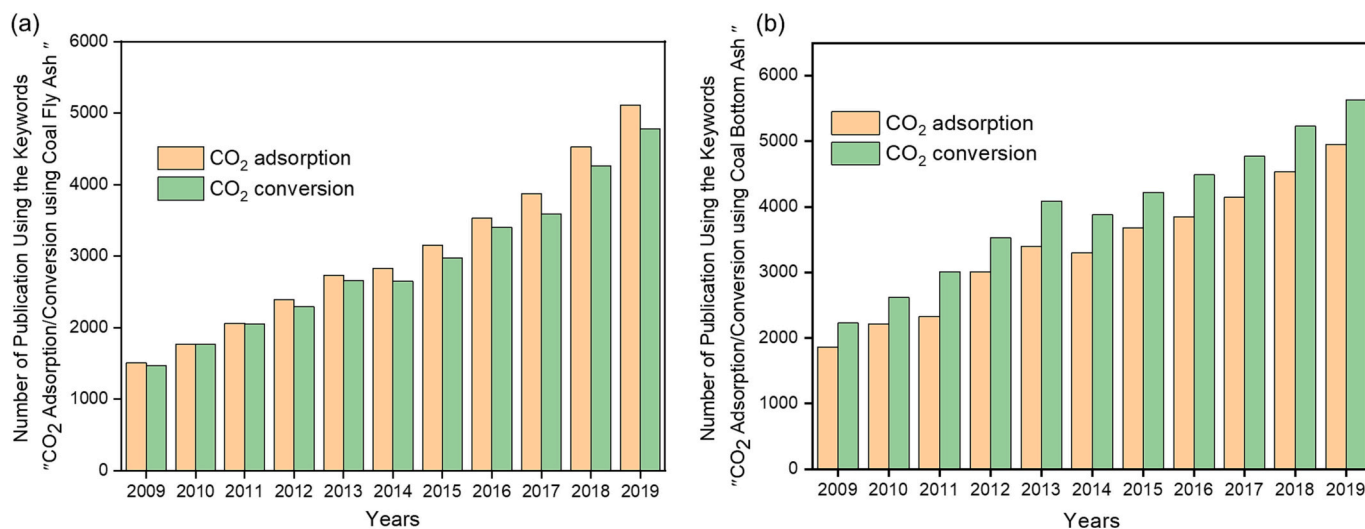


Fig. 1. Illustration of the number of publications using the keywords (a) “ CO_2 adsorption/conversion using coal fly ash” and (b) “ CO_2 adsorption/conversion using coal bottom ash” published from 2009 to 2019 according to Google Scholar.

obtained after alkali melting and acid leaching and the product was an excellent material for CO₂ adsorption. In their work, adsorption kinetic models, such as mass transfer kinetics and deactivation models were applied to predict the adsorption mechanism of CO₂ gas under the following condition: 60 °C of reaction temperature, 15 % of CO₂ concentration, 15 % water vapor concentration, and total gas flow rate of 500 mL. min⁻¹. Overall, the authors revealed that the surface modification of coal fly ash plays a crucial role to enhance CO₂ removal. Guo's co-workers deduced that, in a fixed reactor, the synthesized adsorbents display a good removal capacity of 2.02 mmol. g⁻¹ towards CO₂ gas based on the following equations (Eqs. (1)–(2)) (Guo et al., 2023).

$$\eta = \frac{C_{out}}{C_{in}} * 100\% \quad (1)$$

$$q = \int_0^t \frac{C_{in} - C_{out}}{1 - C_{out}} Q dt * \frac{1}{22400m} * \frac{T}{T_0} \quad (2)$$

where η denotes CO₂ breakthrough rate of the utilized fly ash-based materials (%), q presents the CO₂ removal amount in mmol. g⁻¹, C_{in} is the concentration of CO₂ entered into the fixed bed reactor (%) and C_{out} refers to CO₂ concentration obtained after adsorption process (%).

Researchers also revealed that the preparation of coal ash-based adsorbent by sol-gel method can diminish CO₂ emissions and its capture costs while making efficient uses of coal fly ash. Furthermore, by correlating the experimental data and kinetic models, they indicated that Avrami fractional kinetic model can be efficiently utilized to depict the adsorption mechanism of CO₂ removal.

In another study, Liu et al. prepared some porous nanosilica materials employing coal gasification fly ash (CGFA) via a two-step strategy, microwave-assisted alkaline extraction followed by hydrothermal process. The obtained products were found to be highly effective for CO₂ adsorption. During this study, the authors utilized SEM, high-resolution transmission electron microscope (HR-TEM), XRD and Brunauer-Emmett-Teller (BET) to prove the successful preparation the porous nanosilica (Fig. 3). They indicated that the obtained adsorbent has a surface area ranging from 0.7–1.0 cm³. g⁻¹ and crystallinity of hexagonal mesoporous structure. Besides, the silicon and aluminium in CGFA, consisting of sphere particles in the form of vitreous body with particular sizes ranging from hundreds of nanometers to a few microns. Liu's group revealed that the microwave synthesis presents an energy-efficient, rapid and eco-friendly heat source for fly ash conversion into CGFA which contains active silanol groups.

In this study, the batch adsorption experiments were conducted at 30 °C, the dosage of the adsorbent was 100 mg and adsorption pressure ranged from 1.0 to 1000 mbar. Moreover, in order to predict the mechanism of adsorption and different factors affecting the adsorption process, some isothermal models such as Langmuir (Eq. (3)), Freundlich

Table 1

Diverse applications of coal fly ash and bottom ash-based feedstocks.

Materials	Applications	Refs.
Coal fly ash	Preparation of carbon nanotubes, silica aerogels and cement building material.	(Gollakota et al., 2019; Reddy et al., 2023)
Coal fly ash	Production of ceramic tiles, application of fly ash in agriculture, production of concretes and wastewater remediation.	(Labidi et al., 2023; Luo et al., 2021)
Coal fly ash in Russia	Alumina production using coal fly ash in acidic conditions (hydrochloric acid and ammonium bisulfate/sulfuric acid).	(Valeev et al., 2022)
Coal fly ash	Production of coal fly ash with excellent texture, bulk density and water holding capacity and its application for the improvement of nutrient deficiency in agriculture sector.	(Singh et al., 2010)
Coal fly ash-based adsorbents	Suitable adsorbents for heavy metals removal (i.e., mercury and arsenic) from wastewater.	(Fernando et al., 2022)
Coal fly ash	Preparation strategies of zeolites and porous materials from crystalline fly ash.	(Ju et al., 2021; Lin et al., 2022)
Coal bottom ash	Synthesis of concrete containing coal bottom ash with mechanical and durability properties.	(Ankur and Singh, 2021; Cuenca-Moyano et al., 2023)
Coal-based bottom ash	Physiochemical characteristics and suitability of the bottom ash and its utilization as adsorbent material for inorganic and organic pollutants remediation.	(Adilla and Suzana, 2016; Merzouki et al., 2021)
Coal bottom ash	Synthesis of coal bottom ash as good resistant material for the replacement of fine aggregates or cement in concretes.	(Chindasiriphan et al., 2023; Gooi et al., 2020)

(Eq. (4)) and Sips (Eq. (5)) in the non-linear forms were applied to get an insight about the mechanism of CO₂ adsorption process (Liu et al., 2023b):

$$q = \frac{q_m K_L P}{1 + K_L P} \quad (3)$$

$$q = K_F P^{1/n} \quad (4)$$

$$q = \frac{q_m K_S P}{1 + K_S P^n} \quad (5)$$

where q presents the adsorbed quantity of CO₂, q_m denotes the monolayer CO₂ adsorption, K_L , K_F and K_S are the Langmuir, Freundlich and

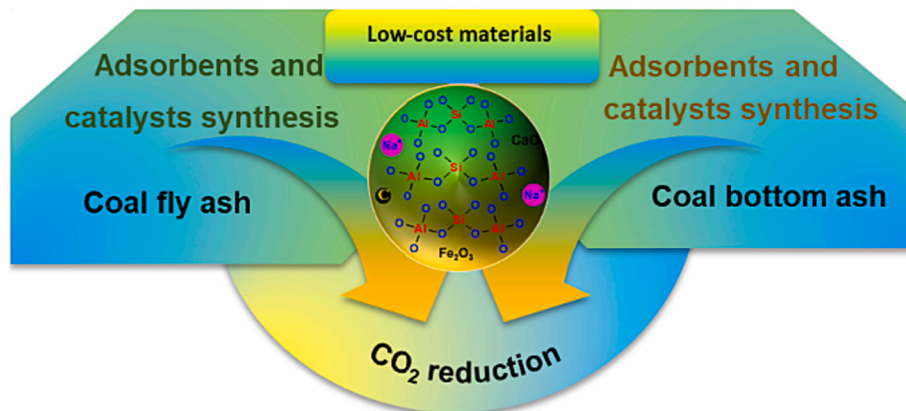


Fig. 2. Coal fly ash and bottom ash low-cost feedstocks for CO₂ reduction.

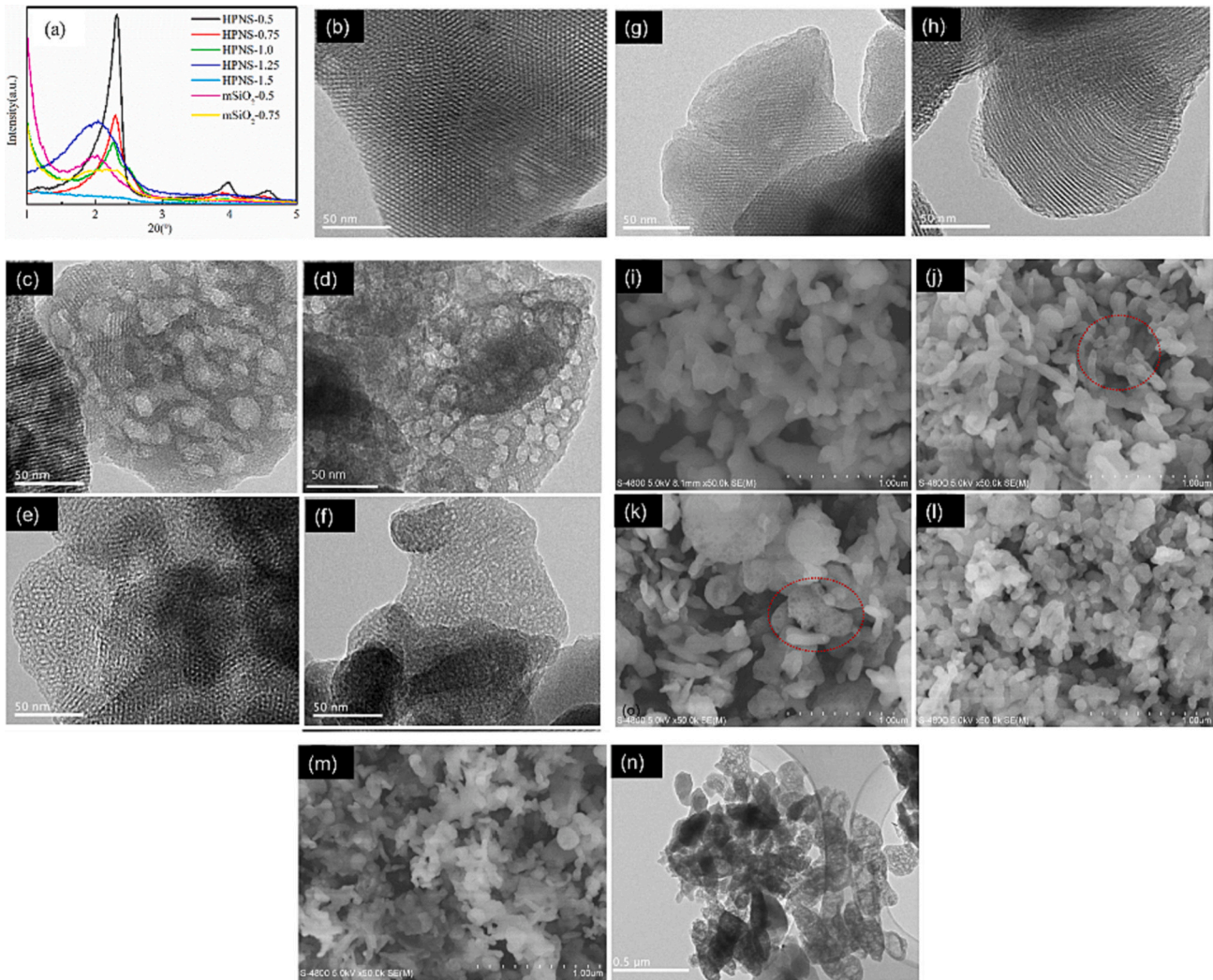


Fig. 3. (a) XRD patterns and TEM images of (b) HPNS: 0.5, (c) HPNS: 0.75, (d) HPNS: 1.0, (e) HPNS: 1.25, (f) HPNS: 1.5, (g) mSiO₂: 0.5, (h) mSiO₂: 0.75, SEM micrographs of (i) HPNS: 0.5, (j) HPNS: 0.75, (k) HPNS: 1.0, (l) HPNS: 1.25, (m) HPNS: 1.5 and (n) TEM illustration of HPNS: 0.75. XRD: X-ray diffraction, TEM: Brunauer–Emmett–Teller, SEM: scanning electron microscope, FTIR: Fourier transform infrared spectroscopy, HPNS: materials prepared from microwave-assisted alkali-dissolution and hydrothermal processes. (Reprinted with permission of Elsevier from Liu et al. (2023a).)

Sips adsorption isotherm constants, respectively, while n corresponds to the measure of heterogeneity of adsorbents in Sips model.

Liu's co-workers concluded that utilized Sips model fits best for all

the prepared adsorbents, signifying that the surface of the obtained adsorbents was heterogeneous and the adsorption process was a mixture of physisorption and chemisorption mechanism. Furthermore, the CO₂

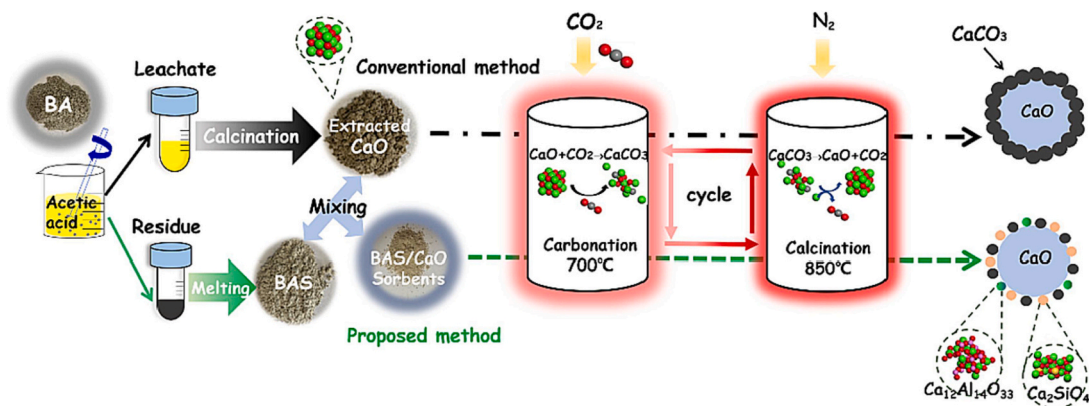


Fig. 4. CO₂ adsorption employing 100 % bottom ash derived CaO-based adsorbent via the proposed method. BA: bottom Ash. (Reprinted with permission of Elsevier from Zhang et al. (2023).)

uptakes of the as-prepared nanosilicas were between 0.65 and 1.63 mg/g at 30 °C and pressure of 1.0 bar (Liu et al., 2023a).

Like fly ash, the bottom ash is often utilized for CO₂ adsorption. In this regard, Zhang et al. (2023) reported the fabrication of robust CaO-based adsorbent through the acid extraction and thermal methods from bottom ash as feedstock (Fig. 4). In this facile route, BA-residue-derived stabilizer (BAS (20 wt%)/CaO) was successfully obtained from bottom ash as an inexpensive inorganic support.

The authors applied XRF, thermogravimetry-differential scanning calorimetry (TG-DSC), XRD, SEM and N₂ adsorption-desorption analyser to examine the chemical and phase compositions, texture and morphological properties of the obtained adsorbents (Fig. 5).

Zhang's co-workers utilized the as-synthesized adsorbent (BAS (20 wt%)/CaO) for the cyclic CO₂ adsorption experiments conducted in a vertical fixed-bed reactor, with an outer diameter of 35 mm and the length of 70 cm at different carbonation temperatures ranging from 600 to 750 °C. The CO₂ uptake was evaluated using the following equations (Eqs. (6)–(8)).

$$C_{CO_2} = \frac{\int_0^t Q_n (\varphi_{K,1} - \varphi_{N,1}) dt}{22,4} \quad (6)$$

$$m_{CO_2} = \frac{C_{CO_2} M_{CO_2}}{m_{adsorbent}} \quad (7)$$

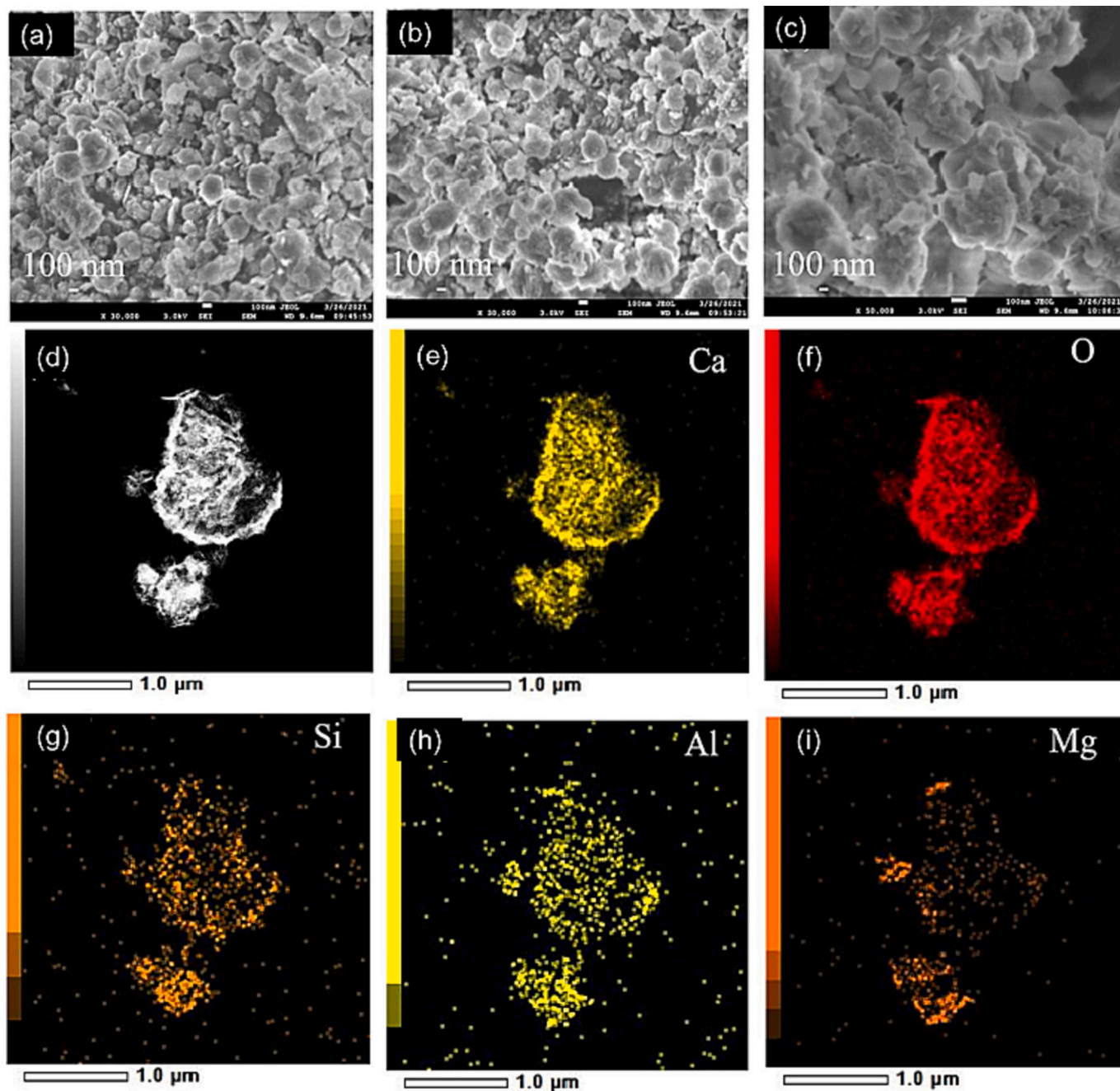


Fig. 5. SEM images of the as-obtained BAS/CaO adsorbents. (a) BAS: 10 wt%/CaO, (b) BAS: 20 wt%/CaO, (c) BAS: 30 wt%/CaO and (d) HAADF-STEM image of BAS: 20 wt%/CaO and elemental EDX mapping of Ca, O, Si, Al and Mg in BAS: 20 wt%/CaO adsorbent. SEM: scanning electron microscope, HAADF-STEM: high-angle annular dark-field scanning transmission electron microscopy, EDX: energy dispersive X-ray spectroscopy, BAS: bottom ash-derived stabilizer. (Reprinted with permission of Elsevier from Zhang et al. (2023).)

$$D = \frac{C_{1st} - C_{Nth}}{C_{1st}} \quad (8)$$

where C_{CO_2} is the molar quantity of the adsorbed CO_2 (mol), Q_n designates the flow rate of the outlet gases ($L \cdot \text{min}^{-1}$), $\varphi_{K,t}$ represents the volume fraction of CO_2 at time 't' in the absence of any adsorbent (vol %), $\varphi_{N,t}$ refers to the volume fraction of CO_2 at time 't' in the presence of BAS/CaO adsorbent (vol%), M_{CO_2} corresponds to the relative molecular mass of CO_2 , $m_{\text{adsorbent}}$ is the mass of the utilized adsorbents, D denotes the decay rate of CaO-based adsorbents (%), while C_{1st} and C_{Nth} are, respectively, the first CO_2 removal of the adsorbents (gCO_2/g adsorbent) and the N_{th} CO_2 removal of the adsorbents (gCO_2/g adsorbent).

The researchers have also stated that bottom ash residue-derived stabilizer (BAS) addition greatly improved the homogeneous dispersion of calcium oxide (CaO). Thus, the combination of the two materials elevated the CO_2 removal by ~30 % over the synthesized BAS/CaO adsorbent, reduced by only 48.15 % after 20 cycles of adsorption/desorption process. The later tended to be stable after 10 cycles under more harsh calcination atmospheres by replacing the pure N_2 with pure CO_2 or 85 % steam/ CO_2 for the desorption experiments (Fig. 6). Moreover, the small quantity of Al_2O_3 and SiO_2 in the Ca-Si-Al-O species of BAS reacted with CaO to acquire $Ca_{12}Al_{14}O_{33}$ and Ca_2SiO_4 materials during the cyclic carbonation/calcination process. Furthermore, CO_2 removal by (BAS (20 wt%)/CaO) adsorbent attained its lowest value at 600 °C and reached its highest value at 700 °C. These findings proved that the reuse of bottom ash as waste is a promising approach for CO_2 remediation technologies (Zhang et al., 2023).

A comparative study of coal ash-based adsorbents with their maximum adsorption capacity is summarised in (Table 2) to highlight the contribution of coal ash derivatives in CO_2 adsorption along with the optimal conditions (Chen et al., 2010; Cheng et al., 2020; Feroso and Sanna, 2022; Freire et al., 2020; Muriithi et al., 2020; Park et al., 2012; Rendeck et al., 2006; Verrecchia et al., 2020; Wang et al., 2018; Zhang et al., 2018; Zhao et al., 2019).

3.2. Mechanistic aspects

It is worthy to note that the adsorption mechanism of CO_2 on coal ash and bottom ash depends on various factors like temperature, pressure,

contact time, pH and the interactions between the CO_2 polar molecule and the chemical ingredients of coal ashes surfaces. Generally, to get an insight about the mechanism of CO_2 adsorption (physisorption, chemisorption, ions exchange, etc.) and study of the adsorption process, theoretical kinetics and isothermal models were widely applied. In addition to the Langmuir, Freundlich and Sips models reported by Liu et al. (2023b), other kinetic and isotherm models were utilized in the literature to predict the CO_2 adsorption mechanism (Fig. 7).

For example, in 2018, Guo et al. conducted a deep theoretical analysis about CO_2 adsorption in a fixed-bed column reactor using different kinetic models. (Guo et al., 2019). Guo's co-workers indicated that the pseudo-first-order (Eq. (9)), pseudo-second-order (Eq. (10)), Avrami fractional (Eq. (11)) and the modified Avrami fractional kinetic models (Eq. (12)) in their non-linear forms well described CO_2 adsorption kinetic behaviours over porous solid adsorbents such as co-firing fly ash and bottom ash (Guo et al., 2019; Guo et al., 2018).

The pseudo-first-order kinetic model can be written as follows:

$$q_t = q_e (1 - e^{-K_1 t}) \quad (9)$$

The pseudo-second-order kinetic model can be described as follows:

$$q_t = \frac{q_e^2 K_2 t}{1 + q_e K_2 t} \quad (10)$$

The Avrami-fractional kinetic model is formulated below:

$$q_t = q_e [1 - e^{-(K_a t)^n}] \quad (11)$$

The modified-Avrami fractional kinetic model can be described as follows:

$$q_t = q_e - \frac{1}{\left[\left((n-1) K_m / m \right) t^m + \left(1/q_e^{n-1} \right) \right]^{1/(n-1)}} \quad (12)$$

where, q_t and q_e are, respectively, the adsorbed amount of CO_2 at a selected time t and the adsorption capacity at equilibrium time ($mmol CO_2/g$); K_1 , K_2 , K_a and K_m present the kinetic constants in these models, while m and n denote the exponents in the Avrami fractional and the modified Avrami fractional kinetic models, respectively. In this study, the authors demonstrated that the efficiency of the adsorption process in

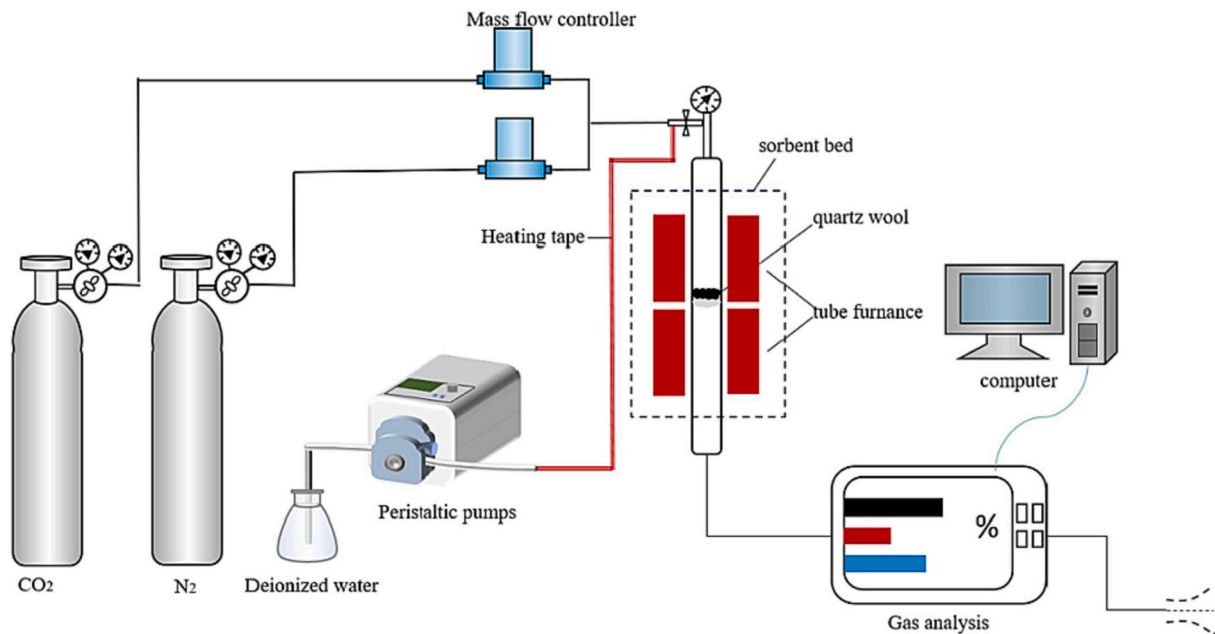


Fig. 6. Schematic presentation of the experimental system for CO_2 removal. (Reprinted with permission of Elsevier from Zhang et al. (2023).)

Table 2Examples of coal fly ash and coal bottom ash for CO₂ removal by adsorption process.

Materials	Preparation methods	Characterization techniques	Conditions and methods of CO ₂ adsorption	Mechanism of adsorption and CO ₂ rate removal	Refs.
Zeolites and hydrotalcite obtained from coal fly ash	Hydrothermal	XRD, SEM, HR-TEM, SAED, TGA, FTIR and BET	-Adsorption/desorption of CO ₂ under a helium atmosphere with a flow rate equal to 20 mL/min and heating the sample ranging from room temperature to 900 °C -Temperature-controlled desorption experiments (CO ₂ -TPD)	-100 % -Physisorption and chemisorption	(Muriithi et al., 2020)
Zeolites from coal fly ash	Fusion/hydrothermal	FESEM, FESEM/EDXS, BET and XRD	Flow rate of CO ₂ equal to 35 mL/min for 30 min at different temperatures ranging from 25 to 400 °C	-60 % of CO ₂ removal using commercial 13× zeolite -Heterogeneous adsorption	(Verrecchia et al., 2020)
PEI-impregnated pure SBA-15 from bottom ash	Impregnation	XRF, XRD, N ₂ adsorption-desorption, EDXS and TEM	15 % CO ₂ , desorption under N ₂ flow with feed flow rate equal to 30 mL/min and T = 75 °C	-169 mg CO ₂ /g adsorbent using 60 wt% PEI impregnated bottom ash	(Chen et al., 2010)
Bottom ash	Water quenching and magnetic separation	TGA and DSC	CO ₂ pressure ranging from 1.0 to 20 bar, 100 g of bottom ash, humidity = 25 % (w/w), T = 105 °C	-12.5 L/kg DM of MSWI bottom ash -Carbonation reaction	(Rendek et al., 2006)
Zeolite 13× form bottom ash	Alkali fusion, extraction and chemical precipitation	XRF, XRD, BET, TEM, SEM, TGA and Raman spectroscopy	CO ₂ adsorption-desorption, 0.01 g of adsorbent, flow rate = 20 mL/min, temperature activation = 100 °C, Reaction temperature = 75 °C	-132 mg/g of adsorbent	(Park et al., 2012)
Fly ash-activated NaOH	Alkaline activation	XRD, BET, SEM, ²⁹ Si, ²⁷ Al MAS NMR and XPS	10 mg of adsorbent, CO ₂ flow = 100 mL min ⁻¹ , T = 35 °C, t = 1.0 h, pressure = 1.0 bar	-5.2 gCO ₂ /g total ash	(Freire et al., 2020)
Potassium-aluminium silicates sorbents (K-FAs) from fly ash	Calcination and mortar grinder	XRD, FTIR, FE-SEM-EDX, (FE-SEM, BET and TGA	1.0 mg to 100 kg of Li ₂ CO ₃ and Na ₂ CO ₃ incorporation at 10–20 wt% on the adsorbents for pure (100 vol% of CO ₂) or diluted (14 vol%) CO ₂ adsorption, T = 700 °C	-CO ₂ uptake of 2.82 mmol CO ₂ /g adsorbent	(Fermoso and Sanna, 2022)
Coal fly ash-synthesized NH ₂ -MCM-41	Alkali fusion method and calcination	XRD, FTIR, SEM, N ₂ adsorption-desorption and TGA	CO ₂ (10 % vol.), gravity factor = 48.09, liquid-gas ratio = 0.07 and NH ₂ -MCM-41 dosage = 0.1 wt%	-CO ₂ uptake up to 95.66 % -High gravity rotating packed bed	(Cheng et al., 2020)
Tetraethylenepentamine (TEPA) loaded fly ash	Impregnation	XRF, XRD, BET, FTIR, SEM and EDS	T = 50–90 °C, 5–20 % CO ₂ with a total flow rate ranging from 200 to 1200 mL min ⁻¹ ,	-CO ₂ uptake of 1.19 mmol CO ₂ /g _{adsorbent} with 15 % CO ₂ flow	(Zhao et al., 2019)
Fly ash derived Li ₄ SiO ₄	Calcination and acid-leaching pre-treatment	XRF, XRD, SEM, and N ₂ adsorption-desorption,	12 vol% and different CO ₂ partial pressures (from 0. to 100 vol%) with balance with N ₂ at 600 °C	-27.5 wt% CO ₂ capture capacity at 600 °C in 100 % CO ₂	(Zhang et al., 2018)
Polyethylenimine-rice husk ash	Hydrothermal	XRF, XRD, TGA/DTG, N ₂ adsorption-desorption, FTIR, SEM and EDX	CO ₂ gas flow of 30 mL/min, temperature ranging from 90 to 135 °C under atmospheric pressure	-1.96 mmol/g _{adsorbent} at 120 °C	(Wang et al., 2018)

XRD: X-ray diffraction, SEM: scanning electron microscope, SEM-EDX: scanning electron microscope-energy dispersive X-ray spectroscopy, TEM: transmission electron microscopy, FTIR: Fourier-transform infrared spectroscopy, XPS: X-ray photoelectron spectroscopy, TGA: differential thermogravimetric analysis, XRF: X-ray fluorescence spectroscopy, NMR: solid-state nuclear magnetic resonance, FESEM: field emission scanning electron microscope, BET: Brunauer-Emmett-eller spectroscopy, DSC: differential scanning calorimetry, SAED: selected area (electron) diffraction, HR-TEM: high resolution transmission electron microscopy, CO₂-TPD: CO₂-temperature-controlled desorption, DM: dry matter. PEI: polyethyleneimine.

a fixed bed system depends on various factors including the physico-chemical characteristics of the coal ash-based adsorbents, the mode of operation, the flow rate of CO₂ and temperature. Moreover, the design and operation of the fixed-bed adsorption systems require the consideration of these factors to achieve the optimal performance of coal ash-based materials and minimize the operational costs of the experiments. The authors reported that CO₂ adsorption on coal ash-based materials in the fixed-bed column can be expressed by different mathematical models (Eqs. (13)–(19)) based on several assumptions given below (Guo et al., 2019; Lin et al., 2017):

- ✓ The dynamic adsorption experiments are performed under isothermal and isobaric conditions.
- ✓ The CO₂ gas phase is considered to be the ideal gas.
- ✓ The flow pattern in the fixed-bed reactor is considered to be plug flow.
- ✓ The gradients of concentration and temperature in the radial direction can be ignored.

- ✓ CO₂ amount in the feed compositions is low, the pressure drop is insignificant, and the linear velocity stills constant.

The mass conservation formula of the breakthrough model is written as follows (Eq. (13)):

$$\varepsilon_B \frac{\partial C}{\partial t} + U \frac{\partial C}{\partial z} + \rho_B \frac{\partial q}{\partial t} = D_Z \frac{\partial^2 C}{\partial z^2} \quad (13)$$

where, D_Z presents the axial diffusion coefficient, while the axial dispersion is ignored, the later equation (Eq. (13)) can be formulated as follows:

$$\varepsilon_B \frac{\partial C}{\partial t} + U \frac{\partial C}{\partial z} + \rho_B \frac{\partial q}{\partial t} = 0 \quad (14)$$

where, ε_B presents the bed voidage, Z refers to the bed height, ρ_B corresponds to the bed density that can be written as follows: $\rho_B = (1 - \varepsilon_B) \rho_S$ where ρ_S presents the particle apparent density, while q denotes the CO₂ removal amount. Besides, the linear driving force equation was,

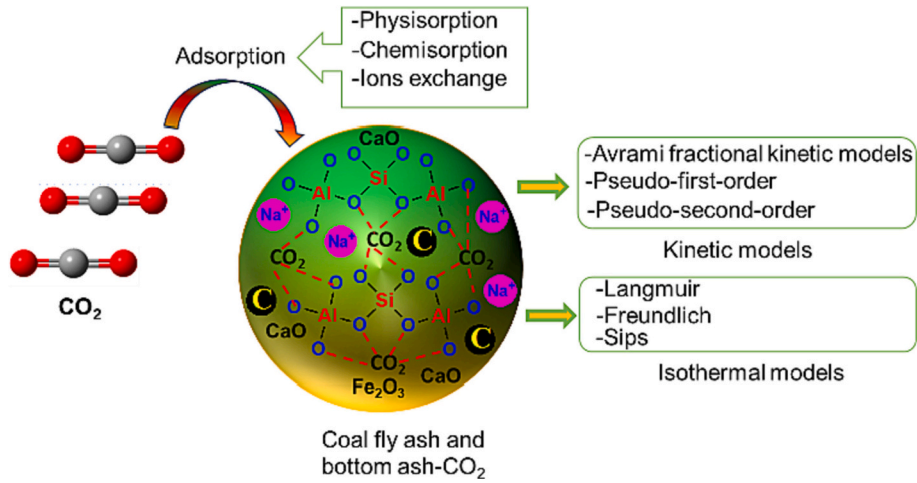


Fig. 7. Examples of adsorption mechanism including kinetic and isothermal models for CO₂ adsorption on coal fly ash and bottom ash.

then, applied to study the adsorbed CO₂ on the surface of adsorbents (Eq. (15)):

$$\rho_B \frac{\partial q}{\partial t} = K_F a_V (C - C^*) \quad (15)$$

where, C and C^* are, respectively, the concentration of CO₂ at any time (t) and at the equilibrium time, K_F denotes the overall mass transfer coefficient and a_V is the surface area of the adsorbent. The overall mass transfer coefficient can be written as shown in (Eq. (16)) (Ye et al., 2008; Zhao et al., 2012):

$$\frac{1}{K_F} = \frac{1}{K_F} + \frac{1}{\beta K_s} \quad (16)$$

where, K_F presents the external mass transfer coefficient and βK_s denotes the internal mass transfer coefficient. The breakthrough adsorption of CO₂ in an ideal plug-flow fixed-bed reactor can be expressed by Eq. (17) (Lin et al., 2017):

$$\frac{C}{C_0} = e^{-X-Y} I_0(2\sqrt{XY}) + \int_0^Y e^{-X-Y} I_0(2\sqrt{XY}) dY \quad (17)$$

where, C_0 and C correspond to the CO₂ amounts at the inlet and outlet of the fixed-bed reactor, I_0 denotes the zero-order Bessel function, while X and Y present the model coefficients.

$$I_0(X) = \sum_{k=0}^{\infty} \frac{1}{k\Gamma(k+1)} \left(\frac{X}{2}\right)^{2k} \quad (18)$$

$$X = \frac{K_F a_V Z}{u}, Y = \frac{K_F a_V t}{\beta \rho_B} \quad (19)$$

where, u is the superficial velocity and β designates the equilibrium constant. By correlating the CO₂ adsorption experiments to the different kinetic equations and mathematical analysis, Guo's group indicated that the modified Avrami fractional kinetic model can be successfully applied to estimate the equilibrium CO₂ adsorption amount employing different adsorbents. Moreover, they assumed that the proposed mathematical model can be utilized to logically predict the CO₂ adsorption breakthrough plots in fixed-bed reactor. They indicated that the overall mass transfer coefficient increase slightly with the rising temperature as well as with the augmentation of the initial CO₂ concentration and gas flow rate (Guo et al., 2019).

In another experimental and computational study, Zgureva and Boycheva (2020), investigated CO₂ adsorption by simulation modelling of the CO₂ dynamic adsorption on the zeolites extracted from fly ash. They indicated that the input data, which are set while building the

model and the findings provided by the utilized characterization of the as-obtained low-cost materials with accordance to their physicochemical characteristics, the equilibrium CO₂ adsorption amount, heat of adsorption, etc. The authors demonstrated that the resulting dynamic CO₂ adsorption capacity on zeolite materials is higher than that of the commercial zeolite, which indicates the technical feasibility of the prepared materials as cost-effective adsorbents for CO₂ capture process. From the high CO₂ adsorption capacity of zeolite material in dynamic studies and the low difference in the breakthrough times at 5.0 vol% CO₂ in the exhaust stream at the studied regeneration temperatures, they concluded that the desorption step should be performed at the temperature of 50 °C (Zgureva and Boycheva, 2020).

3.3. Benefits and drawbacks

By now, huge exploitation of low-cost supports (e.g., coal fly ash and bottom ash) has been reported for the synthesis of various adsorbents. Moreover, the remarkable advances have been made in the application of eco-friendly adsorbents for CO₂ removal due to their nontoxic and high availability for the synthesis of efficient adsorbents. In fact, coal ash-based materials have exhibited an excellent CO₂ adsorption due to their good physical and chemical properties. The functionalization of these materials for reusing them presents an interesting research topic in the field. In this context, numerous studies have explored CO₂ adsorption on coal ash-based materials (Fan et al., 2023; Feng et al., 2021; Miao et al., 2021; Soares and Castro-Gomes, 2022; Yang et al., 2023), employing several processes including commercially liquid amines absorption approach, Ionic liquids capture, calcium looping technology, molten carbonate fuel cells (MCFCs), etc. In these research works, it was stated that coal ash based-materials are beneficial in term of CO₂ adsorption. Furthermore, CO₂ removal by coal ashes and their derivatives present other advantages listed below:

- ❖ Coal ash and bottom ash are characterized by high CO₂ absorptivity and, they can, consequently, be utilized to produce mineral materials employed in the construction sector.
- ❖ Coal ashes of abundant resources present the most appropriate and low-cost materials for CO₂ absorption. They can be selected based on the actual demand for highly efficient adsorbents synthesis.
- ❖ Coal fly ash has several benefits for CO₂ adsorption such as: high CO₂ adsorption capacity ranging from 3.0 to 4.0 mmol.g⁻¹, high renderability (>1000 cycles), CO₂ gas selectivity removal over other gases (>1.0 mmol.g⁻¹.min⁻¹) and low adsorbent cost (\$5-15/kg adsorbent).

- ❖ The adsorbed CO₂ molecules onto the coal fly ash and bottom ash surface can be regenerated through some commonly used methods such as pressure swing adsorption (PSA), pressure and vacuum swing adsorption (PVSA), electric swing adsorption processes, temperature swing adsorption (TSA) and vacuum swing adsorption.
- ❖ Great research efforts were made to modify raw coal fly ash and bottom ash led to an excellent adsorption capacity of CO₂. Moreover, coal ashes including coal fly ash and bottom ash, present high CO₂ uptake, CO₂ selectivity, adsorbent regeneration, excellent kinetics and sustainability. Consequently, coal ash-based adsorbents and catalysts form a new trend in CO₂ remediation technologies employing the adsorption process.

On the contrary, numerous studies indicated that it is crucial to find the optimal removal conditions for CO₂ adsorption on coal ash-based materials which result in low diffusion resistance of CO₂ and facile access to the active sites in coal ashes and their derivatives (Lai et al., 2021). Thus, CO₂ removal by adsorption over the aforementioned materials present some limitations stated below:

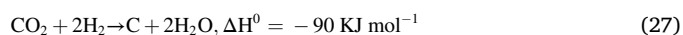
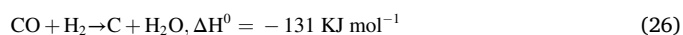
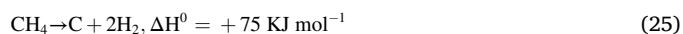
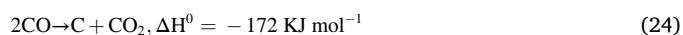
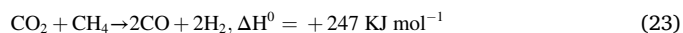
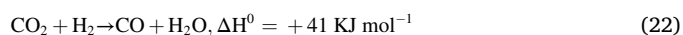
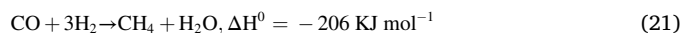
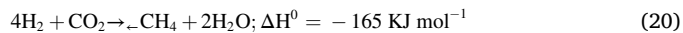
- The main drawback of employing absorption technology for carbon dioxide reduction is, in some cases, the high operation cost due to the use of the expensive coal-based materials which make adsorption an expensive process.
- Even though the CO₂ adsorption systems using coal ash and bottom ash have relatively low efficiency rates, meaning that only a small percentage of the captured CO₂ is successfully removed by coal ash-based adsorbents. The efficiency of some coal ash-based adsorbents is still unsatisfactory in CO₂ adsorption.
- In order to be effective, a CO₂ adsorption strategy by coal ash derivatives must be able to uptake carbon dioxide in large amounts of air. Consequently, adsorption process can take up a lot of space and may not be practical in coal ashes uses.
- The operation of adsorption system utilizing coal ashes requires a safe and high-performing system to ensure CO₂ adsorption and control its measurement.
- Most importantly, once the CO₂ was adsorbed by coal ash-based materials, it needs to be safely disposed which can be challenging and expensive. In addition, some concerns about the long-term CO₂ storage into coal ash derivatives have been raised.

4. Catalytic conversion of CO₂

4.1. Coal ash-based catalysts

Thanks to their availability and biodegradability, coal fly ash and coal bottom ash have the potential to emerge as effectual, cost-effective, sustainable and eco-friendly catalysts for CO₂ conversion into CH₄. On the one hand, injecting CO₂, by carbonation process, into fresh concrete mixtures based on fly ash and bottom ash can produce eco-friendly CO₂-sequestering construction materials with high mechanical, microstructural performances and satisfactory workability (Luo et al., 2022; Suescum-Morales et al., 2022). Consequently, CO₂ mineral sequestration by fly ash and bottom ash presents an exciting research option for CO₂ curing and the construction of new building materials (Miao et al., 2023a; Song et al., 2022). The CO₂ methanation is also applied as a new approach for this gas conversion within the catalytic process through chemical modification of fly ash (Czuma et al., 2022; Lee et al., 2021). In a recent study, Strucks et al. (2021) have demonstrated that various methods of forming methane were developed in the literature. Indeed, the reaction mechanisms proposed for CO₂ methanation can be classified into two steps: the first step examines the conversion of CO₂ to the intermediate product CO and consequently CO methanation. The next step presents the direct hydrogenation of CO₂ to methane without the initial production of CO. The authors have indicated that there are no general rules on the reaction kinetics and

mechanism for the methanation of CO₂, even though the first step suggesting that CO as an intermediate is more expected. Different pathways for CO₂ conversion into CH₄ are discussed with more details employing the following equations (Eqs. (20)–(27)) (Lee et al., 2021; Strucks et al., 2021):



The idea of CO₂ methanation seems interesting. It is used to produce high value-added chemicals such as methanol, methane, polymeric materials, formic acid, etc. (Czuma et al., 2022). Furthermore, several techniques, such as catalytic CO₂ methanation (Cabrerro-Antonino et al., 2023; Lee et al., 2021; Shirsath et al., 2023), electrochemical CO₂ reduction (Chung et al., 2023; Kim et al., 2023), photosynthesis and photocatalysis (Feng et al., 2023; Pan et al., 2021; Zewdie et al., 2023), thermal CO₂ reduction (Hossain et al., 2019; Xu et al., 2023) and biological CO₂ conversion (Fenske et al., 2023; Nisar et al., 2021) were proposed to reduce CO₂. Moreover, the physicochemical transformations of coal ashes by organic and inorganic entities and their applications present an exciting trend to reduce and convert CO₂ to valuable products. Catalysts based on coal ash are often deployed to control the CO₂ emissions. Moreover, organic and inorganic entities incorporation in coal ashes structures is an exciting solution adopted to enhance the catalytic activities of coal fly ash and bottom ash. In this sub-section, the catalytic materials based on coal ashes including coal fly ash and bottom ash as inorganic supports, the characterization techniques and their efficiency of catalytic conversion of CO₂ to syngas such as CH₄, are reviewed. In 2020, Czuma's group synthesized Ni/zeolite X catalyst synthesized from fly ash for CO₂ conversion to CH₄. The authors utilized XRD, XRF, and many other techniques to assess the chemical and structural properties of the as-obtained catalysts. Then, they employed temperature-programmed reduction (H₂-TPR) and temperature-programmed desorption of CO₂ (CO₂-TPD) in order to evaluate the catalytic CO₂ conversion. The batch experiments were performed utilizing a tubular quartz fixed-bed column heated by an electric furnace from 250 to 470 °C and the CO₂ methanation and CH₄ selectivity were estimated using the following equations (Eqs. (28) and (29)) (Czuma et al., 2020):

$$\text{CO}_2 \text{ conversion (\%)} = \frac{(F_{\text{CO}_2, \text{in}} - F_{\text{CO}_2, \text{out}})}{F_{\text{CO}_2, \text{in}}} * 100 \quad (28)$$

$$\text{CH}_4 \text{ selectivity (\%)} = \frac{F_{\text{CH}_4, \text{out}}}{F_{\text{CH}_4, \text{out}} - F_{\text{CO}_2, \text{out}}} * 100 \quad (29)$$

It was demonstrated the zeolite structure was successfully conserved upon the calcination of the catalyst and the formation of large crystallites due to the loading of nickel on fly ash-based materials. Moreover, the Ni/zeolite X support obtained from fly ash material contained a micropore volume of 0.086 cm³·g⁻¹. It was noticed that the catalytic product was efficient for CO₂ conversion which was not very high (~50%) for the as prepared Ni-catalysts employing the fly ash derived zeolite X as inorganic support. The authors demonstrated that the CO₂

methanation increased with the rise of the reaction temperature in the fixed bed reactor and the addition of Ni in catalyst surface led to efficient active sites for CO₂ conversion which was evidenced by CO₂-TPD analysis results.

In another study, Dong et al. (2020) studied the high catalytic efficiency of bimetallic Ni-Re/CCFA catalyst obtained from coal fly ash by co-impregnation method for CO₂ methanation using a fixed bed reactor (Fig. 8).

The authors employed XRF, N₂ isothermal adsorption-desorption, XRD, H₂ temperature-programmed reduction (H₂-TPR), SEM, TEM and thermogravimetric analysis (TG) to examine the physicochemical properties of the obtained catalyst (Fig. 9). In comparison with the coal combustion fly ash (CCFA), the surface area of the Ni-Re/CCFA increased specifically from 10.43 to 23.53 m². g⁻¹. It was concluded that the Ni and Re are relatively homogenous on the (CCFA) support, which proves that the Ni active species and Re promoter interacted with the support in a dispersive way and the Ni dispersion of Ni-Re/CCFA was 14.8 %. In this study, the CO₂ methanation was investigated on a continuous fixed-bed reactor in the range of 350–450 °C using the reactant gas flow with a composition of H₂: CO₂: N₂ = 4.0:1.0:0.5 and a total gas hourly space velocity of 2000 h⁻¹.

Gas composition was quantified by the gas chromatography instrument. The authors indicated that, at the whole temperature range from 350 to 450 °C, CO₂ conversion and CH₄ selectivity for the Ni-Re/CCFA was higher than 94 and 55 %, respectively. They utilized in-situ diffuse reflection infrared Fourier transform spectrum characterization to clarify the CO₂ methanation process. The intermediates of CO₂ methanation over Ni-Re/CCFA catalyst were detected and a possible loop

reaction mechanism was proposed involving the surface hydroxyl group, bicarbonate, bidentate formate and methoxyl groups (Fig. 10) (Dong et al., 2020).

In 2022, Chernyak et al. synthesized Ni-based catalysts supported on amorphous silica and purified rice husk (RH) ash. They utilized scanning electron microscopy-X-ray microanalysis (SEM-EDX), TEM, thermogravimetric analysis (TGA), XRD, TPR and XPS to characterize the prepared catalysts. Chernyak's group performed CO₂ methanation tests by the as-synthesized catalyst in a quartz tubular vertical reactor equipped with mass-flow controllers. CO₂ conversion (XCO₂), product selectivity (S) and the activity of the catalyst in the methanation reaction were estimated as exposed below (Eqs. (30)–(33)) (Chernyak et al., 2022):

$$X_{CO_2} = \left(1 - \frac{N(\text{out}CO_2) * N(\text{in}N_2)}{N(\text{in}CO_2) * N(\text{out}N_2)} \right) * 100\% \quad (30)$$

$$S(\text{CH}_4) = \frac{N(\text{CH}_4)}{N(\text{conv}CO_2)} * 100\% \quad (31)$$

$$S(\text{CO}) = \frac{N(\text{CO})}{N(\text{conv}CO_2)} * 100\% \quad (32)$$

$$A = \frac{N\text{CH}_4}{m_{Ni} * t} \quad (33)$$

The researchers indicated that Ni particles were well encapsulated into the support matrix composed of amorphous silica and rice husk ash with small 5.0–10nm nanoparticles and the huge ones of 50–100nm in diameter. Meanwhile, the catalyst showed irregular globular structure

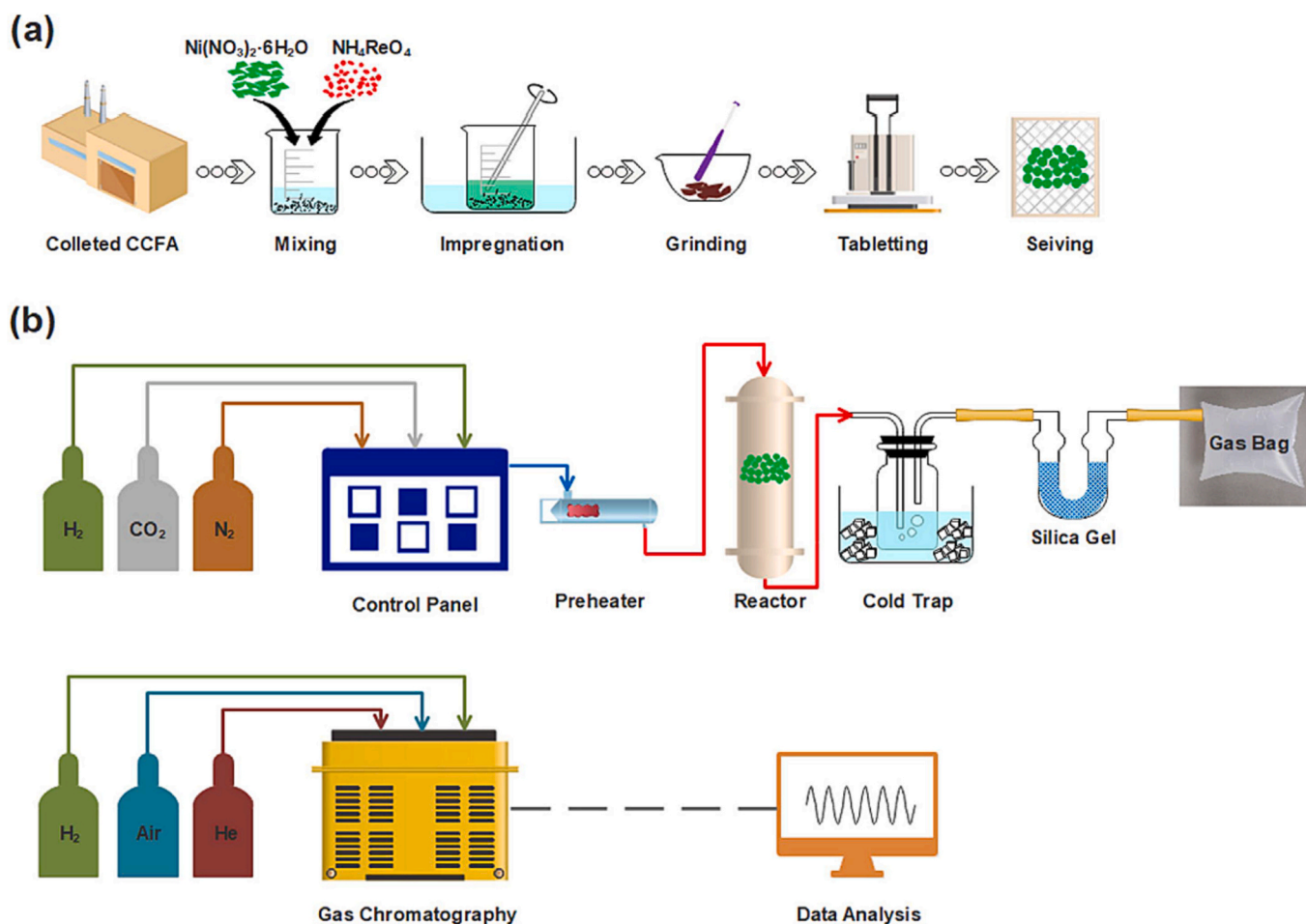


Fig. 8. Schematic illustration of catalyst preparation and its efficiency testing: (a) catalyst preparation process and (b) CO₂ methanation process. (Reprinted with permission of Elsevier from Dong et al. (2020).)

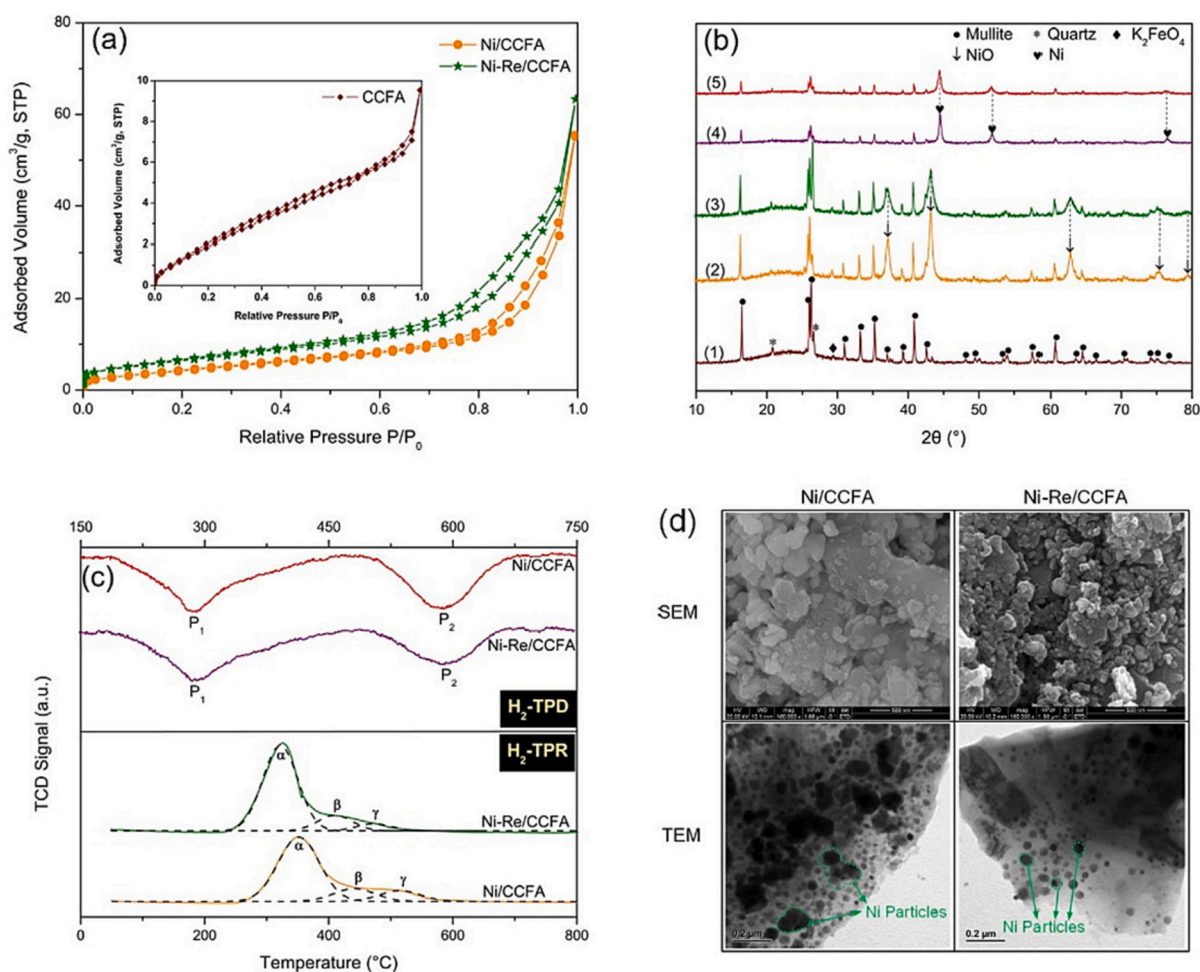


Fig. 9. Characterization results: (a) N_2 adsorption isotherms of as-prepared catalysts, (b) XRD patterns, (c) H_2 -TPR plus H_2 -TPD curves and (d) SEM plus TEM morphologies of reduced catalysts. The sequence number (1)–(5) in XRD patterns represents CCFA, as-prepared Ni/CCFA, as-prepared Ni-Re/CCFA, reduced Ni/CCFA and reduced Ni-Re/CCFA, respectively. XRD: X-ray diffraction, H_2 -TPR: H_2 temperature-programmed reduction, H_2 -TPD: H_2 temperature programmed desorption, SEM: scanning electron microscope, TEM: Brunauer–Emmett–Teller, CCFA: coal combustion fly ash. (Reprinted with permission of Elsevier from [Dong et al. \(2020\)](#).)

with dentate rectangular elements typical for rice hull with the presence of mesopores and micropores, C–O, C=O and O–C=O groups on the surface. However, the latest findings in the field of CO_2 methanation have proven that, when a huge amount of rice husk ash was burned, the pores were blocked with impurities. Thus, ash and SiO_2 should be controlled during the catalyst synthesis. Regarding CO_2 methanation, the obtained products were highly catalytic active with >80 % of CO_2 conversion and 100 % of CH_4 selectivity. Moreover, the authors revealed that the presence of Ni/ SiO_2 –OH groups on the catalyst surface was responsible for CO_2 methanation at 250 °C as an optimum temperature. Overall, the physicochemical transformation of coal ash and bottom ash were established as highly efficient materials to convert CO_2 to hydrocarbon like methane. Therefore, improvement and progress in the development of catalysts based on coal ashes make them good wastes to be reused in CO_2 reduction technologies. The efficiency of the different coal ash-based catalysts in CO_2 methanation is compared in (Table 3) to study the characteristics of coal ash derivatives for CO_2 methanation along with the optimal conditions ([Abdullah et al., 2018](#); [Czuma et al., 2020](#); [Dong et al., 2020](#); [Hoyos et al., 2020](#); [Huang et al., 2018](#); [Huang et al., 2021](#); [Lin et al., 2018](#); [Paviotti et al., 2020](#); [Samojedan et al., 2019](#)).

4.2. Reaction mechanism

CO_2 methanation involves several steps that lead to the conversion of carbon dioxide to methane through a series of intermediates. The mechanism of this process is an important aspect to understand the reaction kinetics and optimize the reaction conditions and, consequently, to ensure the efficient conversion. The major steps are described below:

- In the initial step, CO_2 is adsorbed on the surface of coal fly ash-based catalysts and activated by breaking the C=O bond. This activation of CO_2 is an important step as it makes the CO_2 molecule more reactive towards further reactions.
- The activated CO_2 on the surface of coal fly ashes reacts with hydrogen (H_2) to form intermediate species such as formate ($HCOO^-$), bicarbonate (HCO_3^-) and carbonate (CO_3^{2-}) ions.
- The intermediate species react with hydrogen to form surface species such as CO, H_2O and H_2CO_3 .
- The surface species react with hydrogen to form methane (CH_4), which is the final product of the CO_2 methanation process.
- These mechanistic insights can direct the rational design of catalyst active sites to boost the activity and selectivity of CO_2 reduction.
- Understanding the reaction mechanisms of CO_2 and CO methanation processes is critical to successfully develop heterogeneous catalysts based on coal ashes with high efficiency.

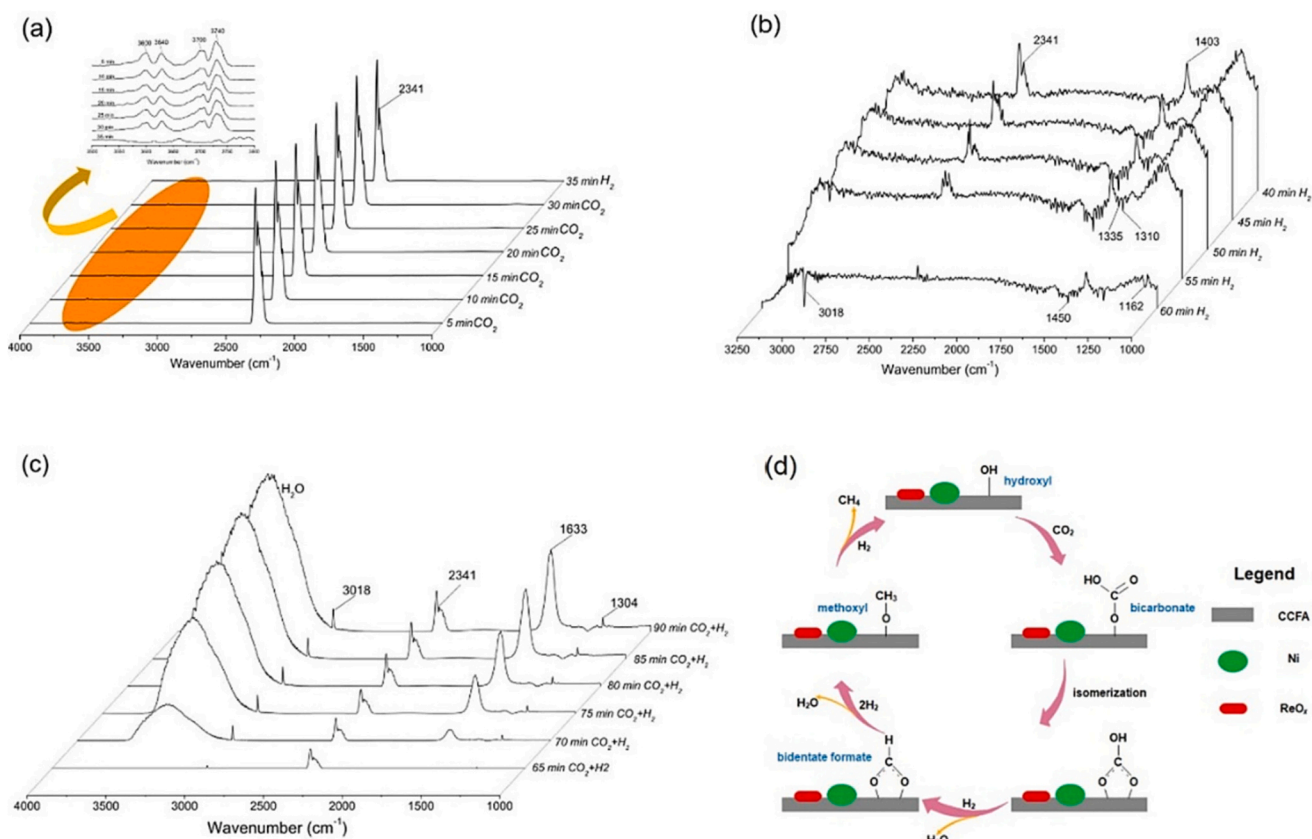


Fig. 10. In-situ DRIFTS spectra (a, b, c) of CO₂ adsorption and methanation over reduced Ni-Re/CCFA and proposed reaction mechanism (d). DRIFTS: Diffuse reflection infrared Fourier transform spectrum, CCFA: coal combustion fly ash. (Reprinted with permission of Elsevier from [Dong et al. \(2020\)](#).)

Table 3
Examples of coal ash-based materials for CO₂ methanation.

Materials	Preparation	GHSV (h ⁻¹)	Catalyst activation	Feed	X _{CO₂} max [%] (T [°C])	S _{CH₄} [%] (T [°C])	Refs.
Ni/Zeolite X synthesized from waste fly ashes	Hydrothermal/Fusion	12.000	Flow of pure H ₂ at 470 °C for 1.0 h	15 % CO ₂ , 60 % H ₂ and 25 % Ar	50 % 400–450 °C	>90 % 400–450 °C	(Czuma et al., 2020)
fly ash (CCFA) as Ni–Re bimetallic catalyst	Co-impregnation	2000	Pure H ₂ flow at 450° C for 4.0 h.	H ₂ : CO ₂ : N ₂ = 4.0:1.0:0.5	99.55 % 400 °C	70.27 % 400 °C	(Dong et al., 2020)
Coal fly ash as a support for oxygen carrier	Impregnation	NA	1000 °C at a heating rate of 10 °C/min in 10v. %CH ₄ -90v.%N ₂ stream	10v. %CH ₄ -90v. % N ₂ and 10v. % O ₂ -90v.%N ₂	91–94 % 800–850 °C	94–100 % 800–850 °C	(Huang et al., 2018)
Ni/Nafly ash–MgO	Sol-gel method	18.000	N ₂ for 10 min, and then a 5.0 vol% H ₂ –Ar mixed gas with a flow rate of 50 mL·min ⁻¹ , 800 °C for 2.0 h	CH ₄ :CO ₂ :Ar = 1.0/1.0/8.0	NA	84 % 750 °C	(Huang et al., 2021)
Cenospheres from fly ash	Wet impregnation	12.000	CH ₄ /CO ₂ /Ar = 1.0/1.0/8.0 550 to 750 °C	5.0 % (v/v) H ₂ /Ar	95 % 750 °C	95 % 750 °C	(Samojedjen et al., 2019)
Industrial fly ash	Hydrothermal	NA	300–400 °C under 1.0 atm. of hydrogen flow (0.08 L.min ⁻¹) for 3.0 h	He:H ₂ = 0.08	>95 % 400 °C	>95 % 400 °C	(Lin et al., 2018)
Ni/SBA-15 from palm oil fuel ash	Fusion	NA	50 mL/min of H ₂ flow at 700 °C for 1.0 h	CH ₄ : CO ₂ : N ₂ = 1.0:1.0:1.0	89 %	31.5 %	(Abdullah et al., 2018)
Mesoporous MCM-41 from rice husk ashes (RHA)	Impregnation	NA	50 % H ₂ /Ar at 873 K	CO ₂ :CH ₄ : Ar = 1.0:1.0:1.0	41 % 873 K	41 % 873 K	(Hoyos et al., 2020)
Ni-MCM-41from rice husk ashes	Impregnation	8600	50 % H ₂ /Ar stream at 600 °C for 3.0 h	H ₂ /CO ₂ = 4.0 diluted in 50 % of Ar	60 % 500 °C	79 % 400 °C	(Paviotti et al., 2020)

GHSV: gas hourly space velocity.
NA: Not available.

➤ The catalyst structure, i.e., particle size and exposed crystal faces, adsorption and desorption characteristics of the coal ash-based feedstocks, temperature, concentration of reactants, the presence of

functional groups on the surface, H₂/CO₂ ratio, and the presence of impurities in the feedstock gas play an important role in CO₂ methanation process.

For example, Schmider and his co-workers utilized a microkinetic model to get an insight about CO₂ methanation process. In this computational work, the authors indicated that this model is developed for methane oxidation and steam reforming over Ni, and later extended to also include CO₂ reforming reactions. As illustrated in (Fig. 11), there are several pathways of methane formation from both CO and CO₂. CO activation is represented by a direct dissociation of adsorbed CO(s) to a surface carbide species (I) and a hydrogen-assisted dissociation, both in a single reaction step and via a formyl intermediate (II). The conversion of CO₂ also features multiple pathways. The direct dissociation of CO₂(s) to CO(s) (III) is included in addition to the formation of a formate/carboxyl species COOH (IV), which can itself, further react to CO(s) or HCO(s). Through this work, the authors assumed that in this model, C(s) is an active intermediate species and does not block the Ni surface through coke formation (Schmider et al., 2021).

Then, the authors used several catalysts such as Ni/Al₂O₃, Ni/SiO₂, Ni/TiO₂ which present a principal compounds of coal fly ash and bottom ash to validate the used kinetic model for CO₂ methanation. Moreover, they utilized the DETCHEM^{CHANNEL} code and took in account thermodynamic considerations and experiments conditions to gain further information on the mechanism of the methanation reaction. As displayed in Fig. 12, they indicated that the CO₂ methanation is predicted to proceed significantly via the direct hydrogenation of CO₂(s) and the dissociation of the resulting formate intermediate to form CO(s), which consequently forms the surface carbide C(s) by the way of H-assisted dissociation. This last step is anticipated to proceed directly, without the formation of a formyl intermediate (Schmider et al., 2021).

In another investigation, Yang et al. (2020) utilized the density functional theory calculations (DFT) to predict the reaction mechanism and thermodynamic parameters, rational design of the catalyst active centers for CO₂ reduction using Rh/TiO₂, as catalyst. They demonstrated that the metal-support interface is the active site and CO₂ methanation on Rh/TiO₂ is dominated by the reverse water-gas shift (RWGS) + CO hydrogenation pathway and CO hydrogenation is much more thermodynamically and kinetically favorable for CO₂ methanation over Rh/TiO₂ catalyst. Moreover, they assumed that there is also evidence that CO₂ methanation via the reverse water-gas shift (RWGS) + CO hydrogenation pathway prefers to proceed through the following equation (Eq. (34)) (Yang et al., 2020):



4.3. Advantages and disadvantages

The conversion of carbon dioxide to hydrocarbon and non-hydrocarbon fuels has attracted particular attention of the scientific researches and industrialists in the recent few years, with a typical pattern utilized to convert CO₂ waste to valuable compounds (Garba et al., 2021). Additionally, the methane produced by CO₂ methanation can be stored or transported through the existing natural gas infrastructure. This approach offers high flexibility and the potential to stabilize electricity grids with a high share of renewable resources. Besides,

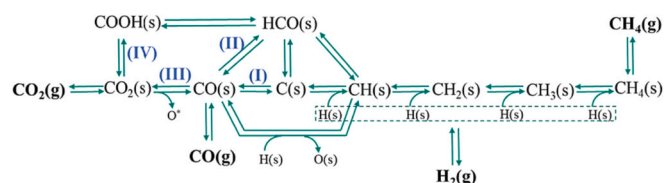


Fig. 11. Reaction scheme of the kinetic model developed in this work. Some reactions are omitted for clarity. Featured pathways: (I) carbide pathway; (II) H-assisted CO dissociation; (III) direct CO₂ dissociation; (IV) H-assisted CO₂ dissociation.

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the functional catalysts based on coal fly ash and bottom ash have shown obvious benefits in CO₂ conversion to methane hydrocarbon.

- The conversion of CO₂ using coal fly ash and bottom ash as inorganic supports into valuable products, such as CH₄, can help reduce greenhouse gas emissions and mitigate the impact of climate change, making CO₂ conversion an environmentally-sustainable process.
- Utilization of the waste products, including coal ashes for CO₂ transformation to hydrocarbon products such as CH₄, can reduce the amount of waste generated by the industrial activities and limit the high disposal of coal fly ash and bottom ash.
- CO₂ methanation using coal ash-based catalysts is considered as new sustainable technologies taking to account the high availability of coal fly ash and bottom ash. It can reduce the dependence on non-renewable resources, such as petroleum and natural gas, and provide more sustainable alternatives providing economic benefits to industries and society.
- The conversion of CO₂ into CH₄ using coal fly ash and bottom ash has numerous environmental and economic benefits, making it an exciting option to reduce their impact on the environment while making them economically valuable.

However, the conversion of CO₂ by coal ash-based catalysts to some useful products, such as methane, has some limitations stated belows:

- The benefits of high temperature methanation in the overall process efficiency are well understood, while the potential for low-temperature methanation is not deeply studied.
- Besides, CO₂ conversion to methane by coal ashes as inorganic supports involves a high energy process where, to produce compounds such as CH₄.
- Although catalytic conversion technology is one of the efficient methods currently in use to minimize CO₂ emissions, it is very expensive to maintain in some cases due to the high cost of some catalysts based on coal ashes.
- Technically, the conversion of CO₂ involves the development of new catalysts based on coal ashes and sophisticated processes that can be technically challenging and costly.
- Sources of CO₂ are not always readily available and their concentration in the atmosphere is relatively low, making their collection and concentration for use in conversion processes using coal ashes difficult and expensive.
- Many types of CO₂ conversion processes are still at the laboratory scale and have not been commercialized. This means that the scaling up of coal ashes uses from the laboratory to commercial scale can be a challenge and new manufacturing processes should be deeply investigated.
- The conversion of CO₂ to CH₄ has some limitations that need to be addressed in particular. However, several research works were conducted and new catalysts based on coal ashes and processes were developed to address these limitations, bringing CO₂ conversion using coal ashes closer to commercial viability.

5. Future challenges

As the release of the anthropogenic CO₂ in the atmosphere is vigorously increasing due to the power plants, chemical processing, overuse of fossil fuels and deforestation on-going effort should be made to reduce the adverse effects of global greenhouse emissions, causing climate change, by establishing an effective approach to capture CO₂. Thus, CO₂ reduction in the atmosphere has been recently considered as an urgently and important process that must be given greater attention in the future. For instance, technologies, such as adsorption and catalytic conversion, show promising solutions if fully implemented. Moreover, scientific investigations have already risen up with more alternative inventions of high-efficient materials. Nowadays, adopting coal ashes as

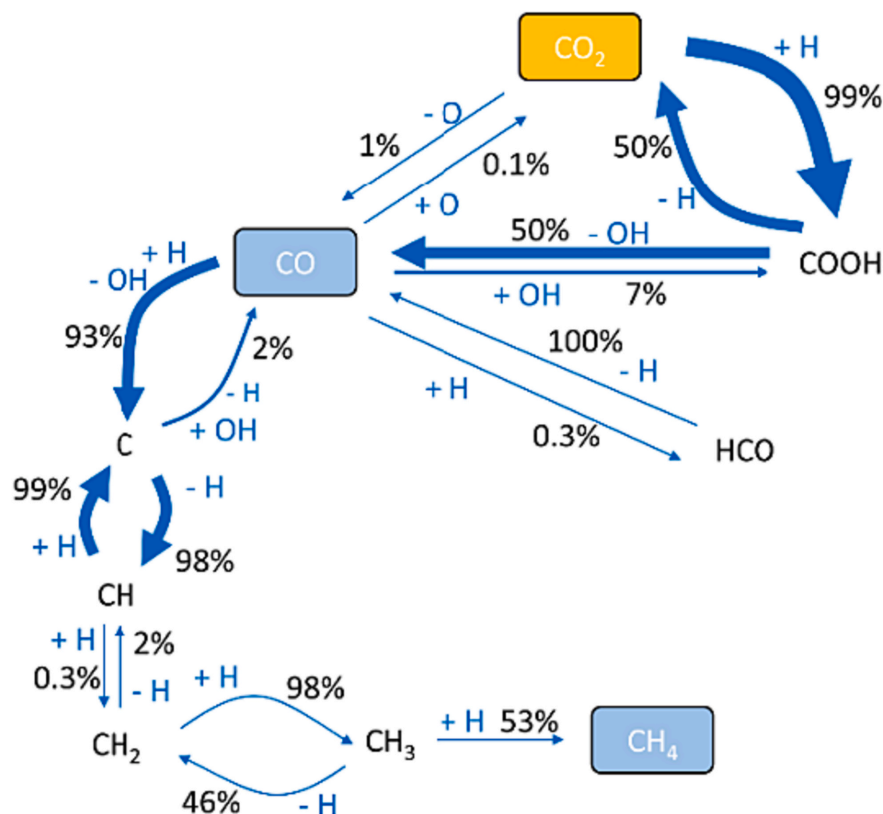


Fig. 12. Reaction flow analysis for a CO₂ methanation experiment with a gas mixture of 10 % CO₂ and 40 % H₂ with 50 % inert gas over a 40.8 % NiAlO_x catalyst at 600 K.

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porous materials for the adsorption and catalytic conversion of CO₂ has been considered as promising strategy that may lower the amount of regeneration energy necessary related to the commercially implemented liquid amines absorption approach for post-combustion carbon dioxide uptake. Moreover, the physiochemical stability of coal ash-based materials, their performance in the presence of moisture, their stability towards impurities, gas diffusion rates, reversibility and regeneration as well as their ability to improve the economic and practical usage of the porous adsorbents need to be addressed to effectively capture CO₂ when competing in a state-of-the-art scrubbing process are the most significant factors from the industrial viewpoint. In this way, expensive raw materials, precursors and the processing techniques are practically undesirable for CO₂ adsorption. In the context of CO₂ remediation by the means of the adsorption and catalysis processes, the high development of new materials based on coal fly ash and bottom ash to selectively separate and capture of CO₂ is one of the highest priorities which should be given more attention by researchers and industrialists. Particular consideration should be paid to the following aspects in future investigations:

- Exploitation of natural supports (e.g., coal fly ash, bottom ash as cost-effective feedstocks, sustainable, nontoxic and abundantly available resources for the scalable fabrication of new efficient porous materials) and their application in CO₂ adsorption/catalytic conversion processes.
- Conventionally, new greener processes should be developed to reduce the cost of the coal ash-based adsorbents synthesis under mild conditions and their removal prowess must be investigated in the future.
- Enhancing coal ash surface to produce more active sites by adding more organic or inorganic entities to ameliorate the coal ash efficiency for the adsorption and catalytic conversion of CO₂. Moreover, high-surface areas of adsorbents are generally more suitable for pressure swing adsorption or vacuum swing adsorption.
- Nano-sized materials based on coal ash provide a copious amount of adsorption sites and catalytic activities, but the issue of self-aggregation and operational difficulties arise more and more in coal ash application. To avoid this scenario, immobilized low-cost metal oxides with coal ashes should be used and yield an excellent removal

percentage in discontinuous adsorption and in fixed-bed column operation

- e) The CO₂ reduction by adsorption and catalytic conversion processes using coal ashes has experienced only a little progress over the last years due to some associated problems such as the low efficiency and stability of some materials based on coal ashes.
- f) The CO₂ conversion using coal ashes as inorganic supports should be deeply investigated in terms of active component, carrier, catalyst electronic effect, synergistic effect, size effect and catalytic conversion, especially at higher pressures of catalysts which are commonly utilized in larger-scale methanation processes.
- g) Easy alternative pathways and techniques should be developed to obtain more efficient adsorbents/or catalysts, compared to the traditional coal ash-based materials which have low elimination capacity slow adsorption kinetics and other limited applications.
- h) In the future, theoretical estimations, such as DFT calculations, thermodynamic consistency and experiments conditions, that favour the methanation of CO₂ should be well explored, especially in coal ash and bottom ash employed to remove or convert CO₂.
- i) Regarding the synthesis of coal ashes derivatives, the quality of fly ash and bottom ash can vary depending on the source, combustion conditions and post-combustion treatment. This variability of coal ashes can affect the synthesis process, and the variation in properties may impact the performance of the final materials obtained from coal fly ash and bottom ash for the synthesis of adsorbents and catalysts.
- j) In the future, the synthesis of fly ashes adsorbents and photocatalytic materials may require large energy inputs in some cases, especially when using high-temperature processing methods. This can offset the environmental benefits of using fly ash products over the traditional alternatives.

6. Conclusions

This review highlights the efficient use of coal ashes-based adsorbents and catalyst materials to reduce the emission of CO₂ greenhouse gas considering its negative impacts on the environment and human health. Over the last decades, with the aim of CO₂ emissions decrease, several coal ash-based materials have been investigated due to their multiple advantages in which make them suitable candidates for the selective CO₂-to-methane conversion technologies. Design strategies, fabrication steps, and the characterizations were discussed first. Then, the benefits and limitations of producing CH₄ directly from CO₂ and its adsorption on coal fly ash and bottom ash were listed. On the one hand, coal ash-based feedstocks are low-cost materials and more eco-friendly than some synthesized constructions utilized to reduce CO₂. Herein, several coal ashes derivatives were introduced in details in terms of preparation methods and their suitability for the industrial adsorption and conversion of CO₂ to methane. On the other hand, the improved reactivity of coal fly ash and bottom ash could, therefore, be explained by the incorporation of inorganic and organic entities on their surfaces. The catalytic activity and the adsorptive efficiency of the coal ash-based materials were quantified with the indices proposed in the literature such as the alkali index, the Al₂O₃/SiO₂ ratio and CaO amounts. The mechanism of CO₂ adsorption and selective reduction over the aforementioned cost-effective catalysts were then discussed. As a whole, this review suggests that the surface of coal ash and bottom ash can be easily modified with a variety of functional moieties to give efficient catalytic derivatives for being used and further developed within practical CO₂ reduction technologies in the near future.

CRediT authorship contribution statement

Abdelkader Labidi: Conceptualization, Methodology, Formal analysis, Software, Data curation, Visualization, Investigation, Writing – original draft. **Haitao Ren:** Conceptualization, Methodology, Data

curation, Writing – original draft, Software, Data curation, Visualization. **Qiuhui Zhu:** Data curation, Writing – original draft, Writing – review & editing. **XinXin Liang:** Conceptualization, Data curation, Writing – review & editing. **Jiangyushan Liang:** Conceptualization, Data curation, Writing – review & editing. **Hui Wang:** Conceptualization, Data curation, Writing – review & editing. **Atif Sial:** Conceptualization, Data curation, Writing – review & editing. **Mohsen Padervand:** Writing – original draft, Writing – review & editing. **Eric Lichtfouse:** Writing – original draft, Writing – review & editing. **Ahmed Rady:** Writing – original draft, Writing – review & editing. **Ahmed A. Allam:** Writing – original draft, Writing – review & editing. **Chuanyi Wang:** Conceptualization, Resources, Supervision, Project administration, Writing – review & editing.

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

No data was used for the research described in the article.

Acknowledgments

This work was supported by the Foreign Expert Program of SAFEA of China (Nos DL2023041004L and G2023041021L).

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