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Performance and Surface Characteristics of CO_2 Adsorption by Adsorbent Derived from Unutilized Coal at Various Temperatures

(Prestasi dan Ciri Permukaan Penjerapan CO_2 oleh Penjerap yang Diperoleh daripada Arang Batu yang Tidak Digunakan pada Pelbagai Suhu)

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ABSTRACT

Surface characteristics and performance of adsorbent materials are vital for understanding CO_2 sorption. This study utilizes unexploited coal from a power plant, demonstrating its potential as an effective CO_2 adsorbent through proximate, ultimate analyses and iodine number evaluations. BET analysis showed that mesopores consistently exhibited higher volumes than micropores in all samples, with the 'CMC' sample showing the greatest mesopore volume at over 0.3 cm^3/g. Surface chemical functional groups and pore structure significantly influence the performance of activated carbon in adsorption processes. In this research, coal-based activated carbon served as a precursor, highlighting the effects of oxygen incorporation into the precursor structure to create new oxygen-containing sites on the surface. These groups were further examined by calcination in a nitrogen atmosphere. The oxygenation process was conducted at varying temperatures - 100, 200, 300, 400, and 500 °C - with subsequent calcination at 900 °C. The highest functional group activity occurred at 500 °C, transitioning from hydroxyl to carboxyl groups. High-temperature calcination at 900 °C facilitated the formation of nitrogen-carboxyl bonds, enhancing the micropore volume and CO_2 adsorption capacity. The calcined samples demonstrated extended breakthrough times, indicating improved adsorption efficacy. Overall, this study confirms that targeted oxygenation and calcination enhance the functional group composition and pore structure of coal-based activated carbon, albeit with a modest increase in adsorption capacity due to low volatile content.

Keywords: Adsorbent; calcination; coal-based activated carbon; CO_2 sorption; oxygen functional group

ABSTRAK

Ciri permukaan dan prestasi bahan penjerap adalah penting untuk memahami penyerapan CO_2. Kajian ini menggunakan arang batu yang belum dieksploitasi daripada loji janakuasa, menunjukkan potensinya sebagai penjerap CO_2 yang berkesan melalui analisis proksimat, analisis ultim dan penilaian nombor iodin. Analisis BET mendedahkan bahawa isi padu mesopori secara tekal lebih tinggi berbanding mikropori dalam semua sampel dengan sampel 'CMC' menunjukkan isi padu mesopori tertinggi melebihi 0.3 cm^3/g. Kumpulan fungsi kimia permukaan dan struktur liang memainkan peranan penting dalam prestasi karbon aktif dalam proses penjerapan. Dalam kajian ini, karbon aktif berasaskan arang batu digunakan sebagai bahan asas, menonjolkan kesan penggabungan oksigen ke dalam struktur bahan asas untuk menghasilkan tapak baharu yang mengandungi oksigen di permukaan. Kumpulan fungsi ini diperiksa lebih lanjut melalui proses kalsinasi dalam atmosfera nitrogen. Proses pengoksigenan dijalankan pada suhu berbeza - 100, 200, 300, 400 dan 500 °C - diikuti dengan kalsinasi pada suhu 900 °C. Aktiviti kumpulan fungsi tertinggi berlaku pada suhu 500 °C, dengan peralihan daripada kumpulan hidroksil kepada kumpulan karboksil. Kalsinasi suhu tinggi pada 900 °C memudahkan pembentukan ikatan nitrogen-karboksil, meningkatkan isi padu mikropori dan kapasiti penjerapan CO_2. Sampel yang dikalsinasi menunjukkan masa terobosan yang lebih panjang, menunjukkan keberkesanan penjerapan yang lebih baik. Secara keseluruhan, kajian ini mengesahkan bahawa pengoksigenan dan kalsinasi yang disasarkan meningkatkan komposisi kumpulan fungsi dan struktur liang karbon aktif berasaskan arang batu, walaupun dengan peningkatan kapasiti penjerapan yang sederhana disebabkan kandungan bahan meruap yang rendah.

Kata kunci: Kalsinasi; karbon aktif berasaskan arang batu; kumpulan fungsi oksigen; penjerap; penyerapan CO_2

INTRODUCTION

Carbon dioxide (CO_2), chiefly produced from the combustion of fossil fuels, plays a significant role in driving anomalous climate change and global warming (Florides & Christodoulides 2009). Projections suggest that emissions from fuel-based power plants are expected to surge to 50% by 2030 (Wickramaratne & Jaroniec 2013). Additionally, studies have shown that atmospheric CO_2 levels have risen by 50% over the past two centuries, contributing to significant increases in surface temperatures and alterations in climate patterns (Yuan et al. 2022). Coal-fired power plants are particularly notable contributors, having released over 2 billion tons of CO_2 since 2010 (Brunetti et al. 2010). The escalation of atmospheric CO_2 from 280 ppm in the pre-industrial era to 379 ppm in 2005 underscores the urgent need for effective strategies to reduce CO_2 emissions (Mondal, Balsora & Varshney 2012). The Intergovernmental Panel on Climate Change (IPCC) predicts that atmospheric CO_2 could reach 570 ppm, potentially causing a 3.8 m rise in sea levels and a 1.9 °C increase in average global temperatures (Mondal, Balsora & Varshney 2012).

Consequently, effective carbon capture and storage (CCS) methods mitigate environmental impacts. CO_2 capture technologies are broadly divided into five main categories: cryogenic distillation (Park et al. 2022), membrane separation (Powell & Qiao 2006), absorption (Khan et al. 2023), and adsorption (Taher et al. 2023a, 2023b). Each of these methods has its respective challenges and benefits. For instance, the adoption of membrane, cryogenic, and absorption technologies has been limited due to high operational costs, environmental implications, and significant energy demands. On the other hand, adsorption is heralded for its environmental sustainability and low energy consumption, which enhances its efficiency and selectivity in separating CO_2. The focus on CCS has recently intensified, recognizing its potential in various applications.

Common adsorbents utilized in CO_2 capture include porous carbon (Singh et al. 2019), zeolites (Rzepka et al. 2018), and metal-organic frameworks (MOFs) (Ghanbari, Abnisa & Wan Daud 2020). While zeolites and MOFs exhibit high CO_2 adsorption capacities, their performance can significantly decline in humid conditions due to their hydrophilic nature. However, advancements have led to developing new MOFs that maintain stability in the presence of water. In contrast, porous carbon is notable for its hydrophobic properties, expansive surface area, and excellent thermal and chemical stability. It is also relatively straightforward to produce, offers adjustable pore sizes, and is cost-effective. Porous carbon's structural characteristics and porosity are primarily influenced by the choice of carbon precursor and the preparation method.

Despite extensive research on various porous carbon materials, studies utilizing unburned coal from coal-fired power plants are limited. This study uses unutilized coal as a carbon precursor to address several critical issues. First, the vast amounts of unutilized coal represent a significant waste of resources and a missed opportunity for energy recovery. Utilizing this coal helps manage industrial waste and aligns with sustainable practice principles by converting a by-product into a valuable adsorbent. Furthermore, there is a lack of comprehensive analysis concerning the surface characteristics of porous carbon derived from such unutilized coal at different temperatures. This research aims to fill this gap by examining these materials' performance and surface properties across various temperature conditions, offering a holistic understanding of their potential applications in environmental remediation.

MATERIALS AND METHOD

MATERIALS PREPARATION

Activated carbon was derived from unutilized coal sourced from a local company in Indonesia. The coal was initially crushed to achieve a particle size of -10+24 mesh. The preparation of the material involved a two-stage modification process to introduce oxygen-containing functional groups. The initial stage involved thermal oxidation, exposing the material to controlled oxygen flow. This was conducted at varying temperatures of 100 °C, 200 °C, 300 °C, 400 °C, and 500 °C. Subsequently, the oxidized samples were subjected to a calcination process at 900 °C under an inert nitrogen atmosphere. Both methods were carried out in a rotary kiln measuring 200 cm long with an internal diameter of 30 cm, as depicted in Figure 1.

Nitrogen gas was purged into the kiln for approximately 10 min during the first oxidation stage to establish an oxygen-free environment. Approximately 50 g of the carbonaceous sample was then introduced into the kiln and heated under an oxygen-rich atmosphere at a flow rate of 1 L/min. The temperature was increased at a rate of 15 °C/min until the target temperature was reached, maintained for 2 h, and then allowed to cool with the oxygen supply turned off. Nitrogen gas was reintroduced post-oxidation to prevent further oxidative reactions as the samples cooled. The samples post-oxidation were labeled as O-100, O-200, O-300, O-400, and O-500.

In the calcination phase, each oxidized sample was treated at 900 °C under a nitrogen atmosphere, maintaining a flow rate of 1 L/min and a heating rate of 15 °C/min for 2 h. Post-calcination samples were labeled as C-O100, C-O200, C-O300, C-O400, and C-O500.

MATERIAL CHARACTERIZATIONS

Post-treatment, the materials were characterized using a Shimadzu Fourier Transform Infrared (FTIR) Spectrometer, model IRPrestige21, to identify the functional groups introduced during the thermal treatments. This method is instrumental in observing the transformations in chemical surface structure through the variation in peak spectra. Additionally, the samples' surface area and pore structure were quantitatively analyzed using the Brunauer-Emmett-Teller (BET) technique via a Quantachrome Nova 4200e apparatus. This technique assessed the porosity and specific surface area based on the principles of nitrogen adsorption at varying pressures and temperatures.

CO_2 ADSORPTION TEST

The CO_2 adsorption efficacy of the prepared activated carbon was evaluated using an adsorption column setup, as illustrated in Figure 2. Each test involved 20 g of the oxidized or calcinated sample placed within the column. CO_2 gas, constituting 15% of the gas mixture with nitrogen

FIGURE 1. Material preparation using rotary kiln

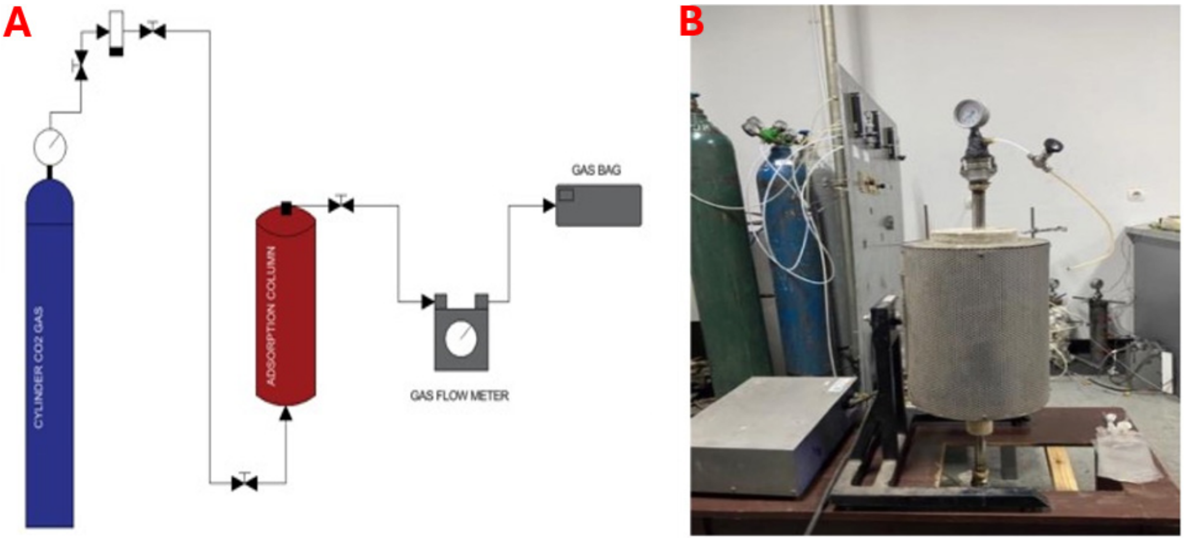

FIGURE 2. Schematic flow diagram of adsorption test (a) and adsorption column of CO_2 gas test (b)

making up the remaining 85%, was directed through the column at a flow rate of 0.1 L/min at atmospheric pressure. The adsorption capacity was determined by analyzing the exit gas every 5 min using gas chromatography until the concentration of CO_2 at the outlet equaled that of the inlet, indicating saturation of the adsorbent.

The efficiency of CO_2 adsorption was quantitatively analyzed by integrating the adsorption curve over time, allowing for the calculation of the cumulative percentage of CO_2 adsorbed as follows:

$$\text{Adsorption \%, t} = \int_0^t 1\, dt - \int_0^t \frac{1}{1+K\, x\, exp(A\, x\, t))} dt \quad (1)$$

where t is the residence time in minutes; and K and A are constants derived from fitting the experimental data to the Boltzmann equation. This equation represents the integral of the CO_2 not adsorbed over the total time, subtracted from the baseline integral, essentially quantifying the area above the adsorption curve relative to the total under-curve area. Subsequently, a mole of CO_2 gas is calculated using the ideal Equation (2).

$$\text{N, mole} = \frac{P \times V}{R \times T} \quad (2)$$

where P is the pressure (1 atm); V is the volume of CO_2 gas (0.015 L); RR is the ideal gas constant (0.082 atm L/mol K); and T is the temperature (298 K). This equation utilizes the ideal gas law to compute the molar amount of CO_2 involved in the adsorption process.

$$CO_2 \text{ mass} = n \times Mr \times t \quad (3)$$

where n is the number of moles of CO_2, Mr is the molecular weight of CO_2 (44 g/mole), and t is the time over which adsorption is calculated.

$$CO_2 \text{ adsorption (mg/g)} = \text{Adsorption (\%)} \times CO_2 \text{ mass} \quad (4)$$

This equation determines the adsorption capacity, indicating the mass of CO_2 adsorbed per gram of the adsorbent material, scaled to reflect the percentage of CO_2 adsorbed over the experimental duration.

RESULTS AND DISCUSSION

INITIAL MATERIALS PROPERTIES

Table 1 presents the material's proximate, ultimate analyses, and iodine numbers. The proximate analysis reveals a moisture content of 7.2%, which is advantageous for adsorption processes, as lower moisture levels facilitate a more efficient adsorption environment. This moisture content is significantly lower than the maximum limit of 15% permitted by the Indonesian National Standard 06 - 3730 – 1995 for adsorbents, highlighting the material's suitability for effective CO_2 adsorption.

The ash content of 16.0% in the studied material is relatively high, presenting challenges and opportunities for its use as an adsorbent. High ash indicates a lower available carbon content, which could diminish the material's overall adsorption capacity. However, the presence of ash might also confer catalytic advantages, as it often contains metal oxides that could enhance interactions with CO_2, potentially improving the adsorption efficiency for specific applications. Comparatively, the ash content of this material, while significant, is still lower than that found in several other commonly used materials. For example, olive peat exhibits an ash content of 21.2%, peach peat has 19.8%, coconut shell features 22.1%, groundnut shell contains 19.48%, loblolly pine presents at 33.9%, and Douglas fir bark is at 32.79% (Parikh, Channiwala & Ghosal 2007). This comparison contextualizes the ash content found in this study. It highlights its relative advantage, suggesting that despite its high ash content, the material might still be competitively viable for specific adsorptive applications where the catalytic properties of ash are beneficial.

The sample exhibits a notably low volatile matter at 3.4%, alongside a substantial fixed carbon content of 73.5%. High fixed carbon is essential as it predominantly comprises carbonaceous material, which is pivotal for adsorption efficacy. Its stable and porous structure is particularly effective for trapping CO_2. In contrast, other materials display significantly higher volatile matter: olive peat has an ash content of 75.6%, peach peat at 79.1%, coconut shell at 77.19%, groundnut shell at 73.72%, loblolly pine at 65.7%, and Douglas fir bark at 65.46% (Parikh, Channiwala & Ghosal 2007).

TABLE 1. Material properties

Proximate analysis				Ultimate analysis					Iodine number (mg/g)
M	A	VM	FC	C	H	N	S	O	
7.2	16.0	3.4	73.5	72.4	1.47	0.47	0.53	9.1	685

M: moisture, A: ash, VM: volatile matter, FC: fixed carbon, C: carbon, H: hydrogen, N: nitrogen, S: sulfur, and O: oxygen

From an elemental perspective, the ultimate analysis shows a carbon content of 72.4%, underscoring the material's potential for high adsorption capacity. Carbon structures provide essential sites for CO_2 capture. Minor elements such as hydrogen (1.47%), nitrogen (0.47%), and sulfur (0.53%) may enhance adsorption through chemical interactions, with nitrogen and sulfur potentially increasing the material's affinity for CO_2. Oxygen content at 9.1% suggests the presence of functional groups like carboxyls and hydroxyls, which can facilitate chemical sorption by forming bonds with CO_2 molecules.

The iodine number of 685 mg/g indicates a substantial microporous structure, significantly higher than polyurethane foam, exhibiting an iodine number of 436 mg/g (Saeed, Ahmed & Ghaffar 2003). This elevated value suggests that the adsorbent possesses an extensive surface area, which is critical for effective CO_2 adsorption. Micropores are particularly advantageous for trapping CO_2 due to their size, which is suitable for the physical adsorption of CO_2 molecules through van der Waals forces (Sun et al. 2022). Additionally, these pores provide sites for potential chemical interactions that further enhance adsorption efficiency.

Thus, the coal-based adsorbent's properties suggest a strong potential for CO_2 sorption, primarily due to its high fixed carbon content and significant surface area. The combination of physical and chemical properties, including the elemental composition with beneficial functional groups, positions this adsorbent as a promising candidate for CO_2 capture. Future work should focus on experimental validation of these properties through CO_2 adsorption tests to better quantify the adsorbent's capacity and understand the dynamics of the adsorption process, thus refining its application in industrial settings.

ANALYSIS OF PORE STRUCTURE MATERIAL

The data presented in Figure 3, encompassing nitrogen adsorption isotherms and pore distribution analyses, provides crucial insights into the textural properties of adsorbent materials and their implications for CO_2 sorption. We can better understand the material's porosity and potential efficiency in adsorption applications by examining these characteristics in detail. Figure 3(a) and 3(c) displays nitrogen adsorption isotherms for various samples before and after CO_2 activation. The isotherms typically exhibit Type IV characteristics, indicating mesoporous structures within the materials (Al-Ghouti & Da'ana 2020).

This type is consistent with the properties observed in activated carbon derived from corncob (Sarwar et al. 2021a), suggesting a similar adsorption behavior. Type IV isotherms are indicative of capillary condensation within the mesopores and are usually associated with materials that possess both high porosity and surface area, which are essential for effective adsorption processes (Schlumberger & Thommes 2021). These isotherms, characterized by hysteresis loops, suggest capillary condensation within the mesopores, a hallmark of materials suited to dynamic adsorption processes.

Pre-activation Isotherms (Figure 3(a)) show that none of the samples reach a plateau at higher relative pressures, suggesting an incomplete filling of pores, which can be attributed to larger mesopores or macropores. All lines show an increasing trend in % adsorption as the concentration increases. Each line reaches different levels at the highest concentration, ranging from approximately 80% to nearly 100% adsorption. The gradual increase in adsorption capacity at higher pressures indicates that these materials could be more effective in applications where larger pore volumes are beneficial, such as in the adsorption of large molecules. Post-activation Isotherms (Figure 3(c)) demonstrate increased adsorption capacities, which indicates that CO_2 activation may enhance the porosity of these materials. The sharp rise in adsorption at lower pressures points to a significant increase in microporosity, critical for increasing the surface area available for adsorption, thereby enhancing the material's capacity to capture CO_2.

Figure 3(b) and 3(d) illustrates the pore size distributions of the samples before and after CO_2 activation, respectively. In Figure 3(b), mesopores consistently exhibit higher volumes than micropores across all samples. Notably, the first sample, likely designated as 'CMC', exhibits the highest mesopore volume at over 0.3 cm^3/g. This result in the current study shows a significantly higher total volume than a previous study, where modified zeolite samples showed mesopore volumes of 0.038 cm^3/g and 0.022 cm^3/g, respectively (Indira & Abhitha 2023).

This finding aligns with research since 1998, demonstrating that a larger volume of adsorbent pores can enhance gas sorption. This increase in pore volume is crucial for improving the adsorption capacity, making the material more effective for applications requiring efficient gas capture (Mangun et al. 1998). This substantial increase underscores the effectiveness of the materials used in the current study for adsorption applications. The larger mesopore volumes suggest an enhanced capacity for accommodating adsorbate molecules.

This analysis highlights significant differences in pore architecture and its evolution due to treatment. The initial pore distribution (Figure 3(b)) indicates a predominance of mesopores in all samples, accompanied by varying levels of micropores. This mesoporous structure is advantageous for applications requiring rapid adsorption kinetics, as mesopores facilitate faster transport of adsorbate molecules into the core of the adsorbent, enhancing overall adsorption efficiency. Supporting this, a previous study identified zeolite as a mesoporous material with promising capabilities for CO_2 capture (Kumar, Srivastava & Koh 2020), highlighting its suitability for efficient gas adsorption applications. This alignment of mesoporous characteristics with high adsorption efficiency is crucial for developing more effective CO_2 capture technologies.

The Post-CO_2 Activation Pore Distribution (Figure 3(d)) demonstrates a noticeable increase in the volume of micropores in certain samples, signifying effective pore development post-activation. Despite this increase, a consistent pattern emerges where mesopores exhibit higher volumes than micropores across all samples. The third sample, likely identified as 'Co40', notably shows the highest mesopore volume at around 0.24 cm³/g. This enhancement in microporosity after activation indicates the successful creation of smaller pore structures, which are particularly advantageous for CO_2 adsorption. Micropores provide a high surface area crucial for the efficient capture of CO_2 at a molecular level, thus, maximizing the adsorption capacity per unit volume of the material (Mohd Firdaus et al. 2021; Raganati, Miccio & Ammendola 2021; Singh et al. 2020). This configuration is optimal for applications targeting high-efficiency CO_2 capture and storage.

The detailed analysis of the nitrogen adsorption isotherms and pore distributions before and after CO_2 activation elucidates the potential of these materials for CO_2 sorption applications. This insight is enhanced by a previous study, which indicated that carbon-based materials enhanced with nitrogen or oxygen could promote CO_2 uptake due to stronger specific interactions with these heteroatoms (Canevesi et al. 2022). The presence of these elements typically enhances the adsorptive properties of the materials by providing additional active sites that interact more effectively with CO_2 molecules, thereby improving the sorption capacity. This finding is crucial for the development of advanced adsorbents that are not only efficient but also specifically tailored to maximize CO_2 capture.

The increase in microporosity post-activation enhances the surface area. It suggests that CO_2 activation is an effective method for tailoring the pore structure of adsorbents to optimize them for specific adsorption needs (Shen 2022). With their optimized meso- and microporous structures, such materials are ideal candidates for industrial applications where high efficiency and capacity for CO_2 capture are required. Further empirical studies focusing on the adsorption behavior under real-world conditions and long-term stability tests are essential to validate these

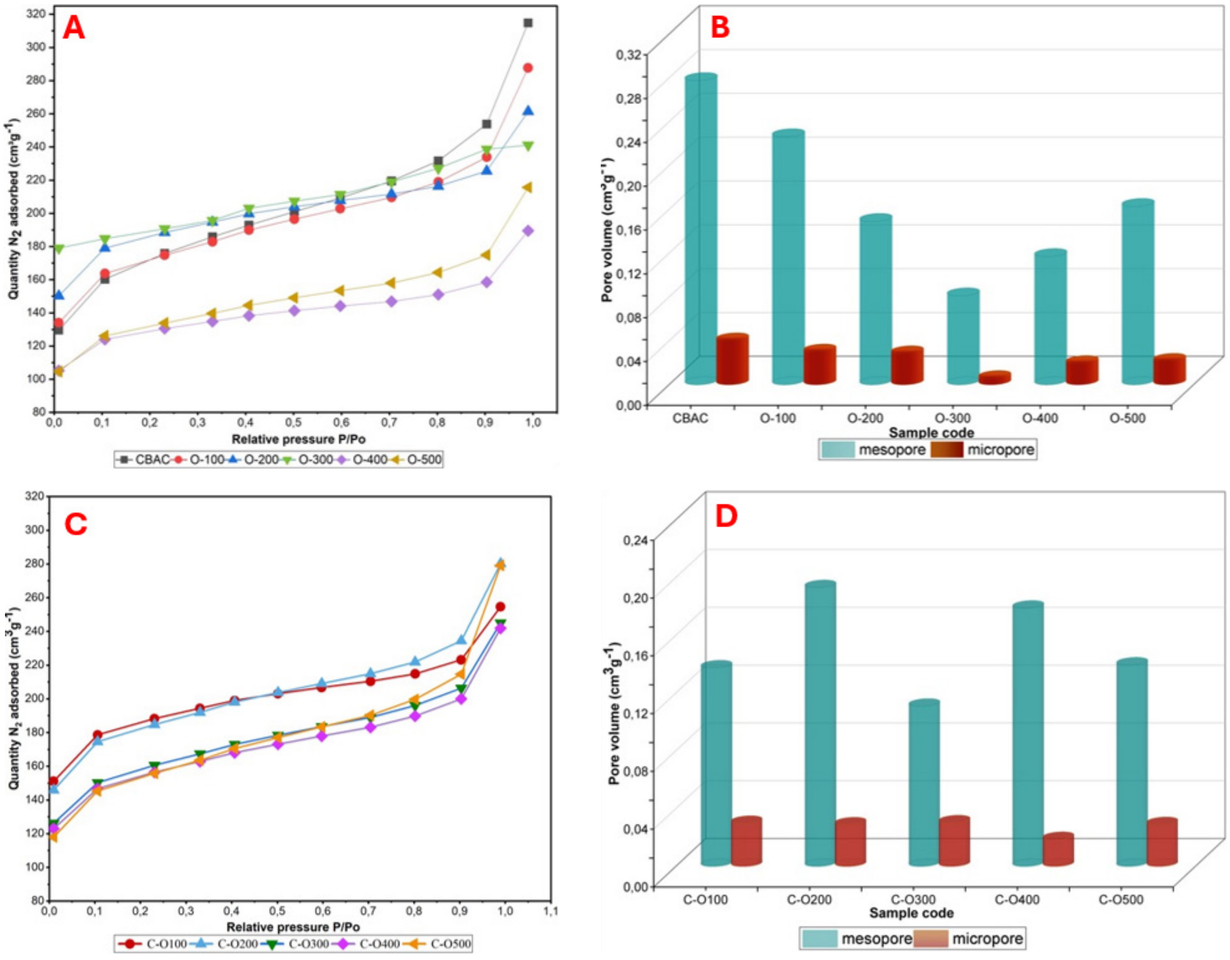

FIGURE 3. N_2 adsorption isotherm and pores distribution

findings and facilitate the scaling-up of these materials for commercial use. This will also help understand the adsorbents' kinetic behaviors and regeneration capacities, paving the way for more sustainable and economically viable CO_2 capture technologies.

PERFORMANCE ANALYSIS OF CO_2 SORPTION

The performance analysis of CO_2 sorption through breakthrough curves, as depicted in Figure 4, shows significant insights into the dynamic adsorption characteristics of both oxidized and calcinated oxidized samples. In the case of the oxidized samples, the rapid initial adsorption phase illustrated by the steep ascent of each curve suggests a high availability of active sites quickly utilized as the CO_2 gas passes through the adsorbent bed. This rapid uptake indicates strong interactions between the CO_2 molecules and the adsorbent's surface, particularly through oxygen-containing functional groups like carboxyls, phenols, and lactones introduced during oxidation (Dai et al. 2021; Jalilian et al. 2024; Petrovic, Gorbounov & Masoudi Soltani 2022). These functional groups increase the surface polarity of the adsorbents, enhancing their ability to capture polar molecules such as CO_2 (Jiang et al. 2023; Zhang et al. 2024).

On the other hand, the calcinated oxidized samples demonstrate a different behavior. The breakthrough curves post-calcination show a delayed time, indicating an enhanced adsorption capacity and efficiency. This improvement can be attributed to structural and chemical modifications induced by the calcination process, such as increased porosity and the development of new active sites, which are crucial for prolonged and effective adsorption. The steeper and more prolonged adsorption phase seen in these samples points to an increased effectiveness, likely due to enhanced thermal stability and an increase in microporosity, which supports the creation of additional adsorption sites (Petrovic, Gorbounov & Masoudi Soltani 2022; Singh et al. 2020).

Figure 4(a) demonstrates that all lines exhibit a rapid increase in adsorption capacity within the first 20 min, followed by a gradual ascent until reaching an approximate plateau around 35 min. Notably, Sample Co500 achieves the highest adsorption capacity, nearing a Cd value of 0.9 mg/g. Conversely, Sample Co10 records the lowest adsorption capacity, slightly above 0.5 mg/g, by the end of the timeframe. Similarly, Figure 4(b) depicts a sharp increase in adsorption capacity during the initial 20 min for all samples, which then tapers off toward the period's end. The curve labeled 0.5a-D attains the highest adsorption capacity, approaching 0.8 mg/g. The lowest capacity is noted in the curve labeled 0a-C, barely surpassing 0.4 mg/g after the observed period. In both panels, the adsorption rates display a characteristic pattern of rapid initial uptake followed by a leveling off, indicating that equilibrium conditions are being approached. The variation in adsorption capacities among the samples or conditions likely reflects differences in the composition or treatment of the adsorbents.

The findings from this analysis underscore the critical role of surface treatments such as oxidation and calcination in enhancing the performance of carbon-based adsorbents for industrial applications like CO_2 capture. The extended breakthrough times and heightened adsorption capabilities following these treatments suggest the potential for tailoring these materials to meet specific operational demands in gas separation processes. Further research should delve into these treated adsorbents' long-term stability and regeneration efficiency, which are vital for assessing their practicality and economic viability in

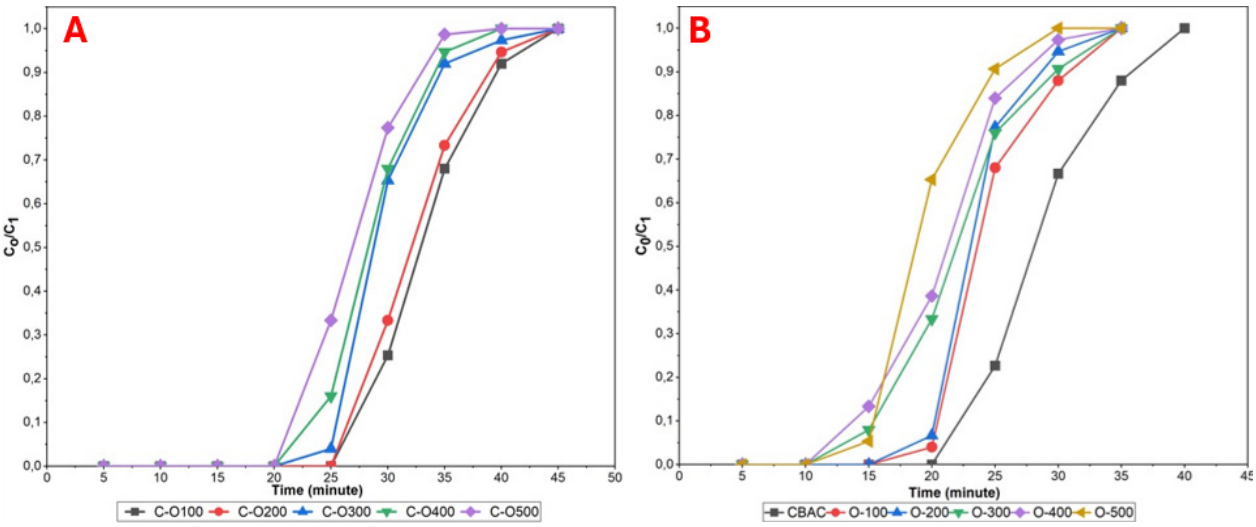

FIGURE 4. Breakthrough curve of the oxidized sample (a) and breakthrough calcinated of the oxidized sample (b)

industrial scenarios. Additionally, a deeper examination of the molecular interactions at the adsorbent surface, possibly through advanced spectroscopic techniques, could shed further light on the mechanisms underpinning the observed enhancements in adsorption performance, offering pathways to optimize these materials for even greater efficiency and sustainability in environmental applications.

Figure 5 offers a comprehensive overview of the CO_2 adsorption capacities of various materials, each uniquely identified by sample codes and visually distinguished through different colors representing distinct series or categories. The adsorption capacities range widely, with C-BAC showing 40.24 mg/g, 10-C-010 at 44.58 mg/g, 20-C-020 at 43.41 mg/g, 30-C-030 at 39.83 mg/g, 0.3%-C-0.3% at 38.43 mg/g, 40-C-040 at 36.29 mg/g, and 0.5%-C-0.5% at the lowest with 25.74 mg/g. This range highlights that the highest adsorption capacity was achieved by the sample coded '1A0-C-0100' at 44.58 mg/g, while the lowest was recorded for '5A0-C-0500' at 25.74 mg/g.

The variation in these figures underscores the significant impact of the materials' characteristics and their preparation methods on adsorption efficiency. Notably, samples with specific concentration percentages, such as 0.3% and 0.5%, typically exhibit lower adsorption capacities than those marked with an increasing sequence (10-C to 40-C). This trend suggests that certain treatments or concentrations might reduce the effectiveness of CO_2 adsorption, potentially due to alterations in the materials' pore structure or surface chemistry (Igalavithana et al. 2020a).

Materials demonstrating high adsorption capacities, such as those depicted by the orange and dark green bars, suggest an optimal combination of porosity and the presence of functional groups that facilitate enhanced CO_2 interaction. This high performance could indicate superior activation methods or unique precursor materials tailored explicitly for efficient CO_2 capture. Similarly, other high-performing samples hint at effective sorption traits, potentially due to robust pore structures or specialized chemical modifications that boost their adsorption capabilities. Conversely, materials represented by lighter green bars with moderate to low adsorption capacities suggest differences in material quality or preparation. These samples may possess compromised structural integrity, reduced surface areas, or fewer functional groups conducive to CO_2 adsorption, possibly due to less effective activation processes or the precursor materials' inherent properties.

The diversity in adsorption capacities points to significant differences in material characteristics, such as pore size distribution, overall surface area, and the types of chemical functionalizations employed (Costa et al. 2021; Li et al. 2021). Materials engineered with enhanced pore development techniques or those incorporating functional groups like amines, which are known to interact with CO_2, naturally exhibit higher adsorption capacities (Nguyen et al. 2023; Qian et al. 2021; Sai Bhargava Reddy et al. 2021; Zhao et al. 2021). Additionally, the activation method-whether thermal or chemical-plays a crucial role in shaping the material's properties, influencing both the porous structure and the presence of functional groups (Leng et al.

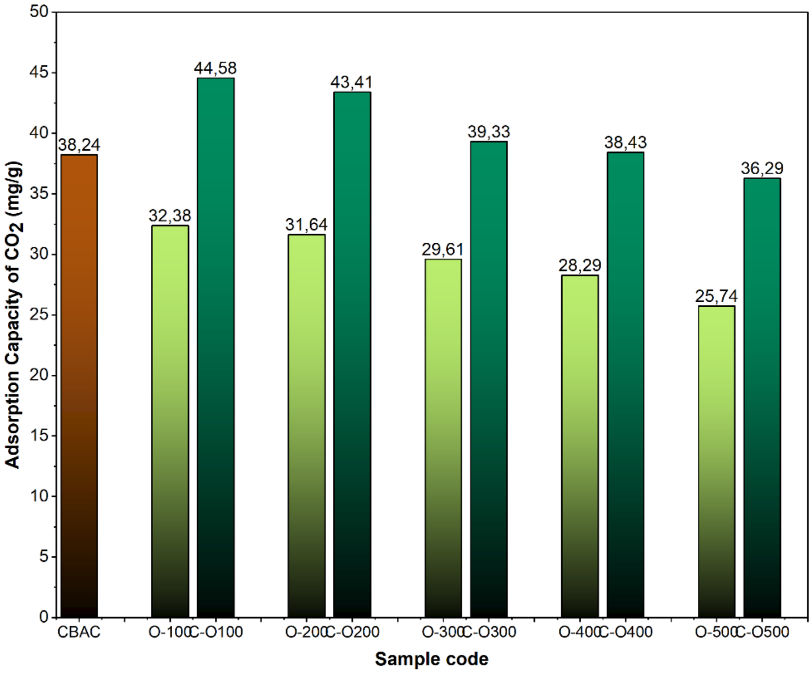

FIGURE 5. Sorption capacity of CO_2 adsorption for each material

2021; Qiu et al. 2023). For applications like carbon capture and sequestration (CCS), where high efficiency and capacity are paramount, selecting materials with superior adsorption capacities is essential. The insights obtained from this analysis inform the further development of adsorbent materials and assist in the scaling up of effective materials for broader industrial use. Thus, the performance data illustrated in Figure 5 highlights the critical role of material properties and processing techniques in defining the CO_2 adsorption capacities of different adsorbents. By continuing to explore and optimize these factors, it is possible to advance the effectiveness of adsorbent materials for key environmental applications, including CCS. Future research should establish clear correlations between specific material properties and adsorption performance, which will aid in developing targeted and highly efficient adsorbent materials for combating climate change.

To contextualize the CO_2 adsorption performance of the synthesized coal-based activated carbon, it is crucial to compare it with other conventional and biomass-derived adsorbents reported in recent literature. The highest adsorption capacity achieved in this study was 44.58 mg/g (sample 10-C-010), which is comparable to or exceeds several biomass-based adsorbents under similar conditions.

For instance, activated biochar derived from pine sawdust (ABC-700) showed a CO_2 adsorption capacity of 4.21 mmol/g (approximately 185.3 mg/g) at 25 °C and 1 bar, indicating a high-performance material for CO_2 capture (Quan et al. 2020). Similarly, steam-activated biochar made from pine sawdust and paper mill sludge achieved capacities between 0.67-0.73 mmol/g (approximately 29.5-32.2 mg/g) under post-combustion conditions with 15% CO_2, highlighting the role of microporosity and surface oxygen groups in boosting performance (Igalavithana et al. 2020b).

In contrast, many untreated or conventionally activated biomass carbons display lower adsorption capacities. For example, biochars from corn cob, wheat straw, and sugarcane bagasse generally exhibited capacities below 2 mmol/g (less than 88 mg/g) under ambient conditions (Maniarasu et al. 2024). Additionally, corncob-derived activated carbon modified via phosphoric acid activation demonstrated capacities around 198 mg/g at 1 bar and 15 °C, but only under high-pressure conditions (Sarwar et al. 2021b). Moreover, while some metal-organic frameworks (MOFs) exhibit theoretical capacities exceeding 1000 mg/g under ideal conditions, their performance often declines in humid environments. In contrast, biomass-derived carbons and coal-based adsorbents like the one synthesized in this study maintain performance in moist conditions due to hydrophobic surface properties (Durán et al. 2017).

Lastly, the highest CO_2 adsorption capacity observed in our study was 44.58 mg/g (1.01 mmol/g) achieved at 100 °C oxidation, followed by calcination under a nitrogen atmosphere at 900 °C. As Table 2 demonstrates, the adsorption capacity of our adsorbent (derived from coal) is competitive when compared to other biomass-based adsorbents, such as coconut shell and almond shell, and similar to values reported for anthracite coal.

MECHANISM OF ADSORPTION

The adsorption mechanism of CO_2 onto the coal-derived activated carbon involves both physisorption and chemisorption, as supported by structural and spectral analyses. Physisorption primarily occurs through van der Waals forces within the microporous framework, particularly due to the high surface area and well-developed microporosity of the calcinated samples. This is evident from the nitrogen adsorption isotherms showing increased surface area and micropore volume post-calcination, which facilitates physical entrapment of CO_2 molecules (Manmuanpom et al. 2018; Shi & Liu 2021).

In addition to physisorption, chemisorption plays a significant role, particularly in samples treated at higher oxidation temperatures and subjected to nitrogen

TABLE 2. Comparison of CO_2 adsorption performance and surface area of various carbon-based adsorbents

Precursor	Surface area (m^2/g)	Uptake (mmol/g)	Agent	Activation temp	Reference
Coal	639	1,01 at 298 K	Nitrogen atmosphere	900 °C	Our work
Coconut shell	371	1.8 at 298 K	CO_2	800 °C	Rashidi et al. (2014)
Almond shell	862	2.7 at 298 K	CO_2	750 °C	González et al. (2013)
Olive stone	1215	3.1 at 298 K	CO_2	800 °C	González et al. 2013)
Nutshell	573	4.5 at 298 K	CO_2	950 °C	Bae and Su (2013)
Oil palm empty fruit bunch	548	2.09		900 °C	Joseph et al. (2017)
Cladophora cellulose	500	2.64 at 273 K	N_2	800 °C	Xu et al. (2018)
Anthracite	607	1.5 at 298 K	Steam	800 °C	Maroto-Valer et al. (2005)

calcination. The FTIR analysis shows the presence of surface functional groups such as hydroxyl, carboxyl, amine, and carbonyl groups, which can form stronger, more specific interactions with CO_2 molecules (Fan et al. 2020; Manmuanpom et al. 2018). Hydroxyl and amine groups are known to interact chemically with acidic CO_2 through hydrogen bonding or by forming carbamate/bicarbonate species, enhancing adsorption performance (Hebert et al. 2015).

Furthermore, the emergence of nitrogen functionalities during high-temperature calcination under an inert atmosphere may contribute to basic sites that promote acid-base interactions with CO_2 (Ojeda-López et al. 2021; Yuan et al. 2023). This dual-mode interaction-combining pore-structure-driven physisorption and functional group-mediated chemisorption-explains the improved breakthrough times and higher adsorption capacities observed, particularly in the C-O100 and C-O500 samples.

Overall, the synergistic effect of surface chemistry and porosity modifications underpins the observed adsorption behavior. These mechanisms confirm the material's potential for use in CO_2 capture applications, especially in post-combustion conditions where a combination of physical and chemical sorption is advantageous.

FUNCTIONAL GROUPS EFFECT ON OXYGEN-BLOWING AND TEMPERATURES

The FTIR spectra depicted in Figure 6 for oxidized material (Figure 6(a)) and calcinated oxidized material (Figure 6(b)) provide valuable insights into the surface characteristics and functional groups of the adsorbent materials, which are critical for their performance in CO_2 sorption. In the spectra, the broad bands observed in the 3800-3500 cm^{-1} range signify the presence of hydroxy compounds, including alcohols and phenols, known for their ability to bond hydrogen with CO_2 molecules. This interaction is essential for enhancing CO_2 sorption through chemical mechanisms. Similarly, the amine groups identified in the 3500-3300 cm^{-1} range suggest potential for chemisorption, where primary and secondary amines can form stable carbamates or bicarbonates with CO_2, thus aiding in its capture. Supporting this result, a previous study also reported the same bands in wavenumbers 3500-3300 cm^{-1} (Akatsuka, Nakayama & Tamura 2024).

The spectra also show significant peaks within the 2300-2200 cm^{-1} range, corresponding to nitrogen compounds such as nitriles and isocyanates. Although these groups might interact with CO_2, their role in sorption is typically less direct than hydroxyls and amines. The presence of alkyne and alkenyl groups, indicated by the peaks between 2260-2190 cm^{-1} and 1700-1500 cm^{-1}, introduces unsaturated carbons into the structure. These unsaturated carbons facilitate π-π interactions with CO_2, enhancing the material's physical sorption capabilities.

Furthermore, the 1300-1100 cm^{-1} peaks, representing ethers and oxygen-containing compounds, highlight the presence of C-O stretches. These functional groups enhance the polarity of the adsorbent surface, making it more attractive to polar molecules like CO_2 through dipole interactions. Lastly, the 1000-600 cm^{-1} range includes inorganic ions and aromatic structures, which contribute to the stability and porosity of the adsorbent. These features are crucial as they indirectly affect the material's ability to adsorb CO_2 by influencing its physical properties.

Thus, the FTIR analysis of oxidized and calcinated materials showcases a rich array of functional groups that are pivotal for CO_2 sorption. Hydroxyl, amine, and oxygenated groups enhance the chemical sorption capacity, while unsaturated carbons improve physical adsorption throughπ-π interactions. The calcination process modifies these functional groups, potentially tailoring the material for optimized CO_2 capture. Experimental validation through CO_2 adsorption studies would be crucial to quantify these effects and adapt the material effectively for industrial applications.

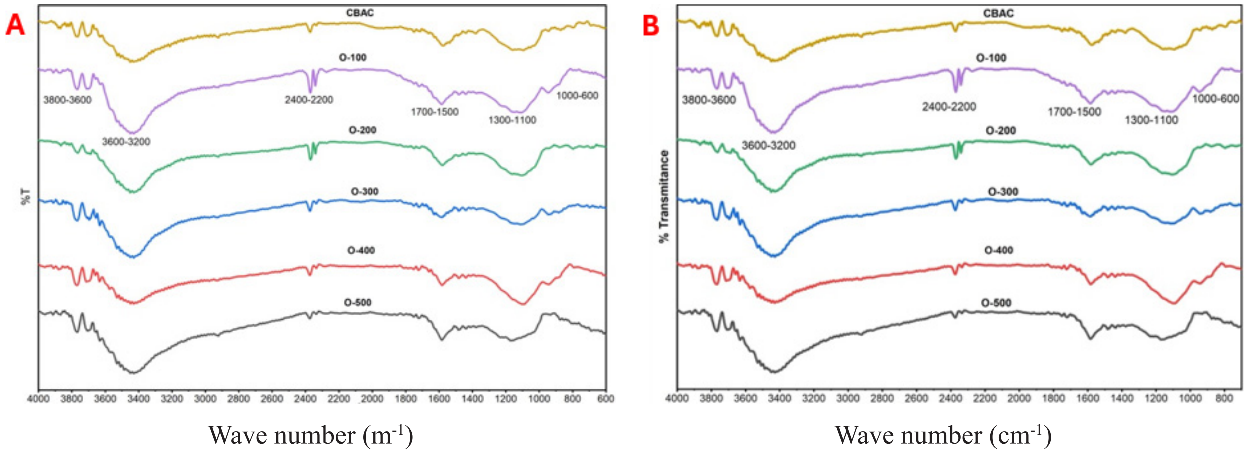

FIGURE 6. FTIR analysis of oxidized material (a) and calcinated oxidized material (b)

ENVIRONMENTAL BENEFITS AND RECOMMENDATIONS FOR THE FUTURE RESEARCH DIRECTION

The primary environmental benefit of enhancing CO_2 adsorption technology is the significant reduction of greenhouse gases in the atmosphere. Efficient capture of CO_2 from industrial emissions, such as those from power plants and manufacturing facilities, is critical in mitigating climate change. This is crucial given that CO_2 is a major contributor to global warming due to its voluminous emissions and long atmospheric lifetime. Furthermore, advanced adsorbents contribute to sustainable energy solutions by converting captured CO_2 into valuable products like fuels, chemicals, and building materials (Anwar et al. 2020; Li, Mo & Unluer 2022). This not only helps recycle CO_2 but also supports a move towards a more circular economy, reducing the demand for fossil fuel extraction and minimizing the overall carbon footprint of industrial activities.

In addition to their role in CO_2 capture, many adsorbent materials also significantly reduce other pollutants from industrial exhaust streams, such as sulfur dioxide, nitrogen oxides, and particulate matter. These capabilities extend the environmental benefits of such materials, as they contribute to overall air quality improvement. For instance, a previous study discusses using various adsorbents like carbon materials, zeolites, and metal-organic frameworks for capturing harmful gases, including CO_2, SO_2, and NO_x (Huang et al. 2019). These materials are specifically highlighted for their effectiveness in gas separation and purification processes, showcasing their broad applicability in industrial settings.

Further emphasizing the versatility of these adsorbents, a novel desulfurization concept outlined focuses on a regenerable adsorbent explicitly designed for capturing sulfur oxides from industrial emissions (Vierling et al. 2021). The study highlights the potential for repeated adsorption cycles, crucial for practical applications that reduce SO_2 emissions with CO_2 and other gases. This regenerative approach not only enhances the economic feasibility of the adsorption technology but also contributes to sustainable industrial practices by minimizing waste and reducing the frequency of adsorbent replacement. This multifunctionality significantly improves air quality, particularly in urban and industrial areas, contributing to better public health outcomes.

Future research should focus on synthesizing new materials with higher adsorption capacities, faster kinetics, and lower regeneration energies. Exploring hybrid materials that combine organic and inorganic components could be particularly promising. Improving the efficiency of existing materials through surface modification techniques, such as functionalization or doping with metals, can also increase their selectivity and capacity for CO_2. Optimizing the regeneration process of adsorbents to reduce energy consumption and operational costs is crucial. This includes the development of low-energy desorption techniques and the integration of renewable energy sources into the regeneration process.

Scaling up laboratory findings to pilot and industrial scales remains critical. Future studies should focus on designing and optimizing large-scale adsorption systems to ensure they are economically viable and technologically robust. Integrating CO_2 capture technologies with renewable energy sources can enhance the sustainability of these systems. Research into powering adsorption systems using solar, wind, or other renewable energies would be beneficial. Comprehensive lifecycle assessments of adsorbent materials and processes are needed to understand their environmental impacts, from production to disposal. This will help identify areas for ecological footprint reductions.

Further economic analysis is necessary to evaluate the cost-effectiveness of deploying advanced adsorbent technologies on a large scale. This includes analyzing capital and operational costs of carbon pricing and market conditions. Research should also consider the policy framework that supports adopting CO_2 capture technologies, focusing on developing incentives, regulations, and standards that promote sustainable and effective CO_2 capture solutions. By focusing on these aspects, research can continue to enhance the role of adsorbent materials in environmental protection, aiming to mitigate the effects of climate change and advance toward a more sustainable and economically viable future.

CONCLUSIONS

This study successfully synthesized coal-based activated carbon as an adsorbent for CO_2 capture, analyzing its surface characteristics and adsorption capacities. The adsorption capacities varied significantly, with sample 10-C-010 achieving the highest at 44.58 mg/g and sample 0.5%-C-0.5% recording the lowest at 25.74 mg/g. Notably, the oxygen enrichment of the coal-based activated carbon at various temperatures altered the oxygen functional groups, transitioning from hydroxyl to carbonyl compounds. This modification led to a decrease in micropore volume and enhanced the alkaline properties on the carbon surface. Further, the calcination of oxidized samples under a nitrogen atmosphere at 900 °C demonstrated an increase in micropore volume while maintaining the oxygen functional groups. Nitrogen augmented the nitrogen binding sites on the carbon surface, potentially enhancing its alkaline properties. The optimized conditions led to longer breakthrough times and higher CO_2 adsorption capacities, with the peak adsorption observed in sample C-O100 displaying distinct spectral peaks for hydroxyl, carbonyl, and nitrogen compounds. The findings underscore the critical roles of oxygen, nitrogen, and elevated temperatures in augmenting the CO_2 capture capacities of coal-based activated carbon. Future studies should focus on optimizing the flow rates of oxygen and nitrogen, as well as the contact time during the oxidation and calcination processes, to further enhance the adsorptive performance of these materials.

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