



Article The Effect of Physicochemical Properties and Surface Chemistry on CO₂ Adsorption Capacity of Potassium Acetate-Treated Carbon Pellets

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Abstract: The aim of this study is to prepare a carbon pellet using low-cost material and a green process with excellent surface properties for carbon dioxide (CO₂) capture application. To enhance the surface properties of the carbon pellet, a chemical activation method was introduced by modifying the pellet with potassium acetate. Then, the carbon pellet was tested in a packed-bed adsorption column to evaluate their performance for breakthrough time and CO₂ adsorption. The effect of the physicochemical and surface chemistry of the carbon pellet on CO₂ adsorption was also studied. The SEM image showed remarkable changes in the surface morphology of the carbon pellet after modification with potassium acetate. In addition, the presence of oxygen-containing functional groups such as hydroxyl and carbonyl groups in the modified carbon pellet could effectively enhance the CO₂ adsorption capacity. Thus, it is proven that the carbon pellet modified with potassium acetate is suitable for CO₂ adsorption. The results revealed that the CAC-PA 2M obtained the longest breakthrough time (19.4 min), higher adsorption capacity (0.685 mmol/g), and good recyclability (the regenerated sample can be reused for more than five cycles). The comprehensive characterization study and CO₂ adsorption experimental data on new carbon pellets can provide a direction for new researchers that are venturing into the CO₂ capture field.

Keywords: activated carbon; carbon pellet; cellulose; CO2 adsorption; potassium acetate; regeneration

1. Introduction

The increase in atmospheric CO₂ concentration is a serious global issue that requires urgent scientific attention due to its negative environmental impact on ecosystems and human health. CO₂ emissions have been widely proven to be the main driver of global warming and climate change [1]. The current concentration of CO₂ in the atmosphere surpasses ~419.47 ppm in January 2023 and this value has increased to ~1.3 ppm compared to ~418.19 ppm in January 2022, which showed an increased trend every year [2,3]. Recent reports by the Intergovernmental Panel on Climate Change have stated that there is a need for strong efforts to limit global atmospheric temperature to 1.5 °C and it must reach a net-zero by 2050 [4]. Since the industrial revolution, the percentage concentration of greenhouse gas emissions in the atmosphere has increased drastically every year, thus raising environment and the planet. CO₂, methane, and nitrous oxide are the three major greenhouse gases that can cause global warming and climate change impact [5]. Among them, CO₂ is the largest contributor, which accounted for 79% of the total greenhouse gas emission is produced by human activities [6]. Therefore, the alternative



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Copyright: © 2023 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). solution to reduce CO_2 concentration is to use carbon capture technology, which is a promising technology to reduce CO_2 concentration in the environment [7]. Chemical absorption using aqueous alkanolamine is the benchmark of carbon capture technology and it has become the first commercial large-scale process in coal-fired power plants, located near Estevan, Saskatchewan, Canada, to capture CO_2 [8]. However, this absorption method has some limitations such as high energy consumption during the regeneration stage and some solvents used in the absorption method could cause corrosion to the equipment [9].

Adsorption by solid materials has been known as a very promising carbon capture technology with the potential to overcome the existing limitations of the absorption process using aqueous amine [7]. In terms of energy consumption, the adsorption process is lower compared to the absorption process. As reported by Anselmi et al. [10], the energy consumption of the absorption and adsorption process is 3.85 GJ/t. CO₂, respectively, for production capacity up to 1000 kg/h. This shows that the adsorption process a chieves a lower energy consumption than the absorption process.

The adsorption method has several advantages, including low energy consumption during regeneration [11], ease of scale-up [12], mild operating conditions requirement [11], and excellent regeneration performance [13], and it also does not produce by-products or harmful materials that can negatively impact the environment [12]. Many solid materials have been used as an adsorbent to capture CO_2 , including commercial activated carbon [14], biomass-based activated carbon [15], graphene [16], molecular sieve [17], mesoporous silica [18], and metal–organic frameworks [19]. The most ideal adsorbents for CO_2 capture should have low-cost material [7], low-pressure drops, fast kinetics, high adsorption capacity [11], high thermal stability, and the ability to be used in a large-scale process [12]. Among the adsorbents, biomass-based activated carbon is the most extensively used adsorbent and has become the ideal candidate material for CO_2 capture application [5].

The conversion of waste biomasses such as agricultural crops and residues, forestry crops and residues, animal manure, by-products of municipal solid waste, and industrial waste products into activated carbon provides an alternative solution to reducing waste disposal problems and environmental pollution [20]. Biomass is mainly composed of carbon, hydrogen, oxygen, nitrogen, and other elements [21]. Heteroatoms such as oxygen and nitrogen play an important role in enhancing CO_2 adsorption capacity [22]. Different types of biomass resources have different elemental compositions. For example, waste biomasses from peanut shells, coconut shells, palm kernel shells, algae, almond shells, and sea mango have a high carbon content (approximately 46–58 wt.%). A high carbon content helps in the creation of pores in the activated carbon and increases the formation of microporous structures. Almond shells, coconut shells, palm kernel shells, and olive stones have a relatively low ash content (<0.6 wt.%) [23]. High ash content will decrease the effectiveness of the activated carbon, reduce the mechanical strength, and thus affect the adsorption capacity [24]. In addition, algae, crab shells, walnut shells, and waste palm shells have a high nitrogen content of 7.2 wt.%, 3.9 wt.%, 3.2 wt.%, and 3.0 wt.%, respectively [23]. A high nitrogen content helps increase the CO_2 adsorption capacity. In Malaysia, the palm oil industry generates a huge amount of biomass waste [25]. These waste products, such as palm shells or palm kernel shells, have the potential to be converted into valuable products such as activated carbon. It has been proven that activated carbon derived from palm shells has a high carbon content, low ash content, large surface area, and the ability to produce excellent adsorption capacity that is suitable to be used as an adsorbent for CO₂ capture [26]. Therefore, in this study, the activated carbon is produced from waste palm shells.

In addition, pre-treatment of activated carbon using a thermochemical process (physical treatment) through carbonization at high temperatures in the absence of oxygen and chemical treatments (activation using activating agent) helps enhance the surface properties of activated carbon [27]. Suitable operating conditions need to be applied during the thermochemical process to produce carbon-based material known as porous carbon. The combination of physical and chemical treatments can improve the physical and chemical properties of the activated carbon, such as surface area and pore volume [28], and increase elemental composition, thus increasing the adsorption capacity [29]. The product produced after the thermochemical process (porous carbon) can be in powder or granular form, depending on the temperature condition during the heating process. The powder-activated carbon can be applied only on the laboratory scale and is not suitable to be used in big reactors, particularly in large-scale or industrial-scale production. This is because the powder form can cause several difficulties such as high-pressure loss and excessive maintenance and additional costs for the separation steps if the powdered material becomes trapped (clogging) in the valve, pipeline, or reactor [30]. An alternative way to solve this issue is to formulate or convert the activated carbon powder into a pellet or bead. Activated carbon pellets can be fabricated using a pellet mill machine, the conventional method (hand-pressed machine), and the simple pelletized method by mixing the powder-activated carbon with a binder to form a pellet [31]. One of the methods implemented in the laboratory for pellet formation is the pelletized process. Binders are used in pellet formation to increase the density and strength, to protect the materials from external damage, and to improve the quality of the pellet [32]. There are various types of binding agents or binders, including both organics (starch, cellulose, protein, lignin) and inorganics (calcium hydroxide, sodium hydroxide, bentonite, and others), which have been used to improve the quality of the pellet in terms of its physical properties [33].

In this study, cellulose gum or carboxymethyl cellulose (CMC) is used as a binding agent to make strong pellets and improve the pellet's performance. CMC is an efficient organic binder [34]. The advantages of CMC binder include its being easily available, cheap, biodegradable, non-toxic (environmentally friendly binder), thermodynamically stable, posing good mechanical properties, and the presence of active functional groups (such as carboxyl and hydroxyl groups) that are suitable for CO_2 adsorption and have had their performance as a strong pellet tested [35,36]. However, to the best of the authors' knowledge, based on a literature survey, there are limited studies investigating the performance of carbon pellets as an adsorbent in CO_2 capture applications. For example, Wadi et al. [18] investigated the effect of binder ratio on mesoporous silicas pellets in CO_2 capture application. They used bentonite and LUDOX (Colloidal silica) as binders. The study indicated that the addition of bentonite and LUDOX as binders improved the CO_2 adsorption performance. Nguyen et al. [37] examined the effect of bagasse pellets as an adsorbent to capture CO_2 on a semi-industrial scale. It was found that the bagasse pellets have a large surface area, high pore volume, and the quality of the pellet is similar to that of the commercial pellet products. The results revealed that bagasse pellets have high CO₂ adsorption capacity at high temperatures and are suitable for use in a large-scale application. Oliveira et al. [38] fabricated an efficient pellet adsorbent from calcium oxide in a CO_2 capture application. They mixed the calcium oxide with carbon nanotubes to enhance the surface porosity of the pellet adsorbent. The study showed that after modification, the CO_2 capture capacity of the modified pellet was 98.2% higher compared to the unmodified pellet (64.1%). They found that the addition of a small amount of carbon nanotubes (0.05%)can significantly improve the CO_2 adsorption performance. They reported that converting powder into pellets shows an excellent performance in CO₂ adsorption capacity and can be used in large-scale production. According to Hong et al. [39], modification of the carbon pellet with activating agents such as potassium hydroxide can improve the textural and chemical properties of carbon pellet, thus increasing the CO_2 adsorption performance.

The aim of this study is to fabricate carbon pellets using a low-cost organic binder with excellent surface properties for CO₂ capture application. To enhance the surface properties of the carbon pellet, a chemical activation method was introduced by modifying the pellet with potassium acetate. The advantages of potassium acetate are low toxicity and corrosivity compared to potassium hydroxide [40]. The characteristic of potassium hydroxide is similar to potassium acetate in that it can create more pores on the pellet surface, thus improving pore development for adsorption capacity. The powder palm shell-activated carbon was mixed with CMS as a binder and potassium acetate solution at different concen-

trations to form a carbon pellet. The carbon pellet was tested in a packed-bed adsorption column to evaluate its performance for the breakthrough time, CO_2 adsorption capacity, and regeneration performance. The physical and chemical properties of carbon pellets were analyzed using scanning electron microscopy (SEM), elemental composition (CHNS), Fourier transform infrared (FTIR) spectroscopy, and X-ray photoelectron spectroscopy (XPS). There was no study conducted to evaluate the performance of potassium-treated carbon pellets in CO_2 capture application. Therefore, the conversion of biomass waste into products such as pellets with high performance has gained increasing attention among researchers because these pellet products can be used in large-scale applications.

2. Experimental Section

2.1. Materials

Raw activated carbon was purchased from Biotek Abadi Sdn. Bhd., Malaysia. The particle size of the powder-activated carbon was 1930 nm (~1.9 microns). Potassium acetate (\geq 99%) and CMC with analytical reagent grade were purchased from Sigma Aldrich. The gases (nitrogen and carbon dioxide) were purchased from Alpha Gas Solution Sdn. Bhd., Malaysia, and the purity of the gas was above 99.99%.

2.2. Preparation of Carbon Pellet

The carbon pellet was prepared with and without the chemical activation method. In this study, potassium acetate was used as an activating agent for chemical activation treatment to enhance the textural and chemical properties of the carbon pellet.

For sample preparation without chemical activation, the carbon pellet preparation involved the following steps: (1) mixing, (2) molding, and (3) drying and heating. During the mixing step, activated carbon powder was added into the binder solution (composed of CMC and water as solvent) at different mass ratios until it became slurry or a viscous paste. The ratio of the binder and the solvent is varied at different proportions (0.01 g–1 g). Meanwhile, the activated carbon powder was 1:0.1:1 (activated carbon powder: cellulose: water). Then, the prepared sample was shaped into small pellets in a pellet mold. The pellet was placed in an oven at 120 °C overnight for drying. After drying, the pellets were kept at 300 °C in a furnace to allow a complete drying process and to enhance the pellet hardness. This sample was labeled as CAC-water.

The pellet produced in this study is a spherical shape. The aspect ratio was measured using Image J software [41]. The calculated aspect ratio was found to be 1.01. These findings are consistent with the reported study by Chopra et al. [42] that recommended the aspect ratio limits for spherical pellets be less than 1.2. This confirmed the spherical shape of the pellet produced in this study.

In chemical activation using potassium acetate, the preparation process involved five steps (Figure 1): (1) preparation of the stock solution, (2) impregnation, (3) mixing, (4) molding, and (5) drying and heating. Firstly, the stock solution of 1 M, 2 M, and 3 M of potassium acetate was prepared using the dilution method. Then, the activated carbon powder was mixed with potassium acetate solution at different mass ratios using the impregnation method. After the impregnation process, the modified activated carbon powder was mixed with a binder at different mass ratios until it formed slurry or a viscous paste. The ratio of the binder and the activating agent varied at different proportions (0.01 g–1 g). Meanwhile, the activated carbon powder is fixed at 1 g. The optimum mass ratio of the binder solution to the modified activated carbon powder was 1:0.1:1 (activated carbon powder: cellulose: potassium acetate solution). The prepared sample was molded into a small pellet with a diameter of 3 mm in a pellet mold. Finally, the carbon pellet was dried in an oven at 120 °C overnight. Then, the carbon pellets were dried in a furnace at 300 °C for 1 h. The obtained carbon pellet sample was denoted as CAC-PA 1M, CAC-PA 2M, and CAC-PA 3M. Using this procedure, approximately 40 g of carbon pellets were prepared for the CO₂ adsorption study.



Figure 1. The schematic diagram for the preparation of potassium acetate-treated carbon pellets.

Since this experiment is the first attempt to make carbon pellets, the heating temperature was kept at a low temperature of 300 °C. For future studies, the effect of temperature during heating or carbonization will be investigated in detail.

2.3. CO₂ Adsorption Experiment

The preparation of CO_2 adsorption experiment was conducted using a semi-pilot plant packed-bed adsorption column. The schematic diagram of the experimental setup for packed-bed CO_2 adsorption has been described in detail in our previous study [43]. In order to evaluate the CO_2 adsorption capacity performance, carbon pellet samples were inserted into an adsorption column at fixed CO_2 inlet concentration (15%) and flow rate (200 mL/min), while the adsorption temperature was measured at varying operating temperatures of 25 °C to 55 °C. The experiment was performed at atmospheric pressure (low-pressure condition). The data logger system was used to collect breakthrough time results and the data generated in excel were used to plot a breakthrough curve. Results from the breakthrough curve were then used to calculate the adsorption capacity. The experimental data were collected every minute using a CO₂ analyzer instrument (Alpha Omega Instrument Series 9610, Lincoln). The breakthrough curve was obtained by plotting the normalized concentration (Ct/Co) versus time (t), where the Co and Ct are CO_2 concentrations at inlet and outlet streams. The breakthrough time (tb) was obtained at the point of 0.05 (5 vol.% of CO_2 inlet concentration) (Ct/Co = 0.05), which refers to the breakthrough curve [44].

Regeneration studies were performed after the adsorption process had completed or reached the saturation stage. The sample was heated at 120 °C and purged with nitrogen gas for 10 min. Then, the adsorption–desorption experiment was performed at 25 °C with similar steps as the above procedure. The same step was repeated for five cycles for the regeneration study.

2.4. Characterization of the Carbon Pellet

The morphology images were analyzed using a SEM workstation (Tescan VEGA-3, Brno, Czech Republic) to determine the surface morphology. The particle size distribution was examined using a particle size analyzer (Mastersizer 2000, Malvern Panalytical Ltd., Malvern, UK). CHNS/O elemental analysis to determine the percentage of Carbon (C), Hydrogen (H), Nitrogen (N), Sulfur (S), and Oxygen (O) in the carbon pellet sample was analyzed using CHNSO Elemental Analyzer (Thermo Scientific, FlashSmart, Waltham, MA, USA). The functional group of the carbon pellet samples was determined using an in situ electrochemical FTIR spectra instrument (Bruker, VERTEX 70v, Rosenheim, Germany) at a wavenumber range of 4000–400 cm⁻¹. Micromeritics Accelerated Surface Area and Porosity (BET ASAP 2020, Norcross, GA, USA) adsorption analyzer was used to evaluate BET surface area and micropore volume. XPS was used to analyze surface elemental species and possible molecular bonding of carbon and oxygen atoms for carbon pellets. XPS analysis was characterized by Axis Ultra delay-line detector (DLD) (Kratos/Shimadzu Manchester, UK) using Al K α (1486 eV) monochromatic X-ray as the excitation source (spot size < 15 um). The high-resolution scans were deconvoluted to their constituent peaks using CasaXPS Version 2.3.14 (Casa Software Ltd., Cheshire, UK) based on the characteristic binding energies of the functional groups. Aspect ratio was measured using image analysis software (Image J 1.53t version).

2.5. Adsorption Isotherms

To understand the adsorption mechanism and the interaction between the adsorbate and the adsorbent, three isotherm models (Langmuir, Freundlich, and Temkin) were fitted with experimental data of CO₂ adsorption capacity at different temperatures (25, 35, 45, and 55 °C) and as a function of CO₂ partial pressure. The parameters of these models were estimated by the non-linear equation using OriginPro 8.1 software (Origin Lab, USA). The Langmuir, Freundlich, and Temkin isotherm models are given by Equations (1), (2), and (3), respectively [45].

$$q = \frac{q_e K_L P}{1 + K_L P} \tag{1}$$

$$q = K_F P^{1/n} \tag{2}$$

$$q = \left(\frac{RT}{b}\right)(lnK_TP) \tag{3}$$

where q is amount of CO₂ adsorbed (mmol/g); qe is the maximum CO₂ adsorption capacity (mmol/g); P is CO₂ partial pressure (atm); K_L is Langmuir constant (1/atm); K_F is Freundlich constant related to the adsorption capacity (mmol/g (1/atm)1/n); 1/n is the dimensionless adsorption intensity constant; and K_T is Temkin constant.

3. Results

3.1. Performance Comparison of Carbon-Based Pellets

In this study, four carbon pellet samples were prepared to investigate the breakthrough time, CO₂ adsorption capacity, and the effect of the physicochemical properties of carbon pellets on the CO₂ adsorption performance. Firstly, carbon pellet samples of CAC-water, CAC-PA 1M, CAC-PA 2M, and CAC-PA 3M were compared in terms of their breakthrough time and CO_2 adsorption capacity. The breakthrough experiment was performed at a fixed adsorption temperature of 25 °C and a flow rate of 200 mL/min to compare their adsorption performance. The breakthrough adsorption experiment was completed until the saturation stage is reached, whereby the outlet CO_2 concentration is equal to the inlet concentration ($C_{out} = C_{in}$). The results from the breakthrough adsorption experiment will be used to calculate the maximum adsorption capacity of the carbon pellet samples. The breakthrough adsorption experiment for carbon pellet samples is depicted in Figures 2 and 3. As seen in Figure 2, the highest breakthrough time of 19.4 min was observed in the modified carbon pellet of CAC-PA 2M. Meanwhile, CAC-water (unmodified carbon pellet) recorded the lowest breakthrough time (13 min). By increasing potassium acetate concentration from 1M to 2M, the breakthrough time and CO₂ adsorption capacity increased from 17 min (0.622 mmol/g) to 19.4 min (0.685 mmol/g) at 25 °C (Figures 2 and 3). It is obvious that the breakthrough time and the adsorption capacity increase with increasing activating agent concentration and then decrease. CAC-PA 2M showed the best adsorption performance. Activating agent improves the adsorption properties. As expected, the

breakthrough time and adsorption capacity increase in the following order: CAC-water > CAC-PA 3M > CAC-PA 1M > CAC-PA 2M. However, the breakthrough time and adsorption capacity decreased with a further increase in the activating agent concentration. For example, the breakthrough time and adsorption capacity of CAC-PA 3M were 15.2 min and 0.5351 mmol/g, respectively, which are lower compared to CAC-PA 2M. Similar findings were also reported by Singh et al. [46] and Ghosh and Barron [47], who stated that by increasing activating agent concentration, adsorption capacity decreases. The breakthrough time and adsorption capacity increased from 1M to 2M and then decreased at higher activating agent concentrations (more than 3M). This is due to pore blocking, whereby CO_2 molecules cannot penetrate into the pores of the carbon pellets. The number of available active sites also decreases, and this reduces the porosity which results in decreased adsorption capacity [48].



Figure 2. Breakthrough adsorption performance of carbon pellet samples at a fixed flow rate of 200 mL/min, 15% CO₂ concentration, and 25 °C.



Figure 3. Adsorption capacity of carbon pellet samples at a fixed flow rate of 200 mL/min, $15\% \text{ CO}_2$ concentration, and $25 \degree$ C.

In addition, the surface area of the modified carbon pellets showed a significant effect on adsorption capacity. An increase in potassium acetate concentration shows a decrease in the surface area. The highest surface area of $883.76 \text{ m}^2/\text{g}$ (Table 1) was obtained with CAC-PA 2M, and this has resulted in a high adsorption capacity of 0.685 mmol/g. Meanwhile, a slight decrease in the surface area from $883.76 \text{ m}^2/\text{g}$ to $825.08 \text{ m}^2/\text{g}$ was observed with CAC-PA 3M (Table 1). This could be caused by the blocking and collapse of pores at high concentrations, which thus decreases the surface area [46].

Samples	BET Surface Area, (m²/g)	Micropore Volume, (cm ³ /g)	Elemental Composition (%)		
			С	Н	0
CAC-water	843.92	0.27	73.98	1.78	6.65
CAC-PA 1M	856.44	0.31	69.92	1.65	10.52
CAC-PA 2M	883.76	0.36	66.36	1.95	14.72
CAC-PA 3M	825.08	0.21	67.53	2.62	12.04

Table 1. BET surface area, micropore volume, and elemental compositions of carbon pellets.

Considering the ease of preparation, fewer environmental impacts (low toxicity and corrosiveness), and high adsorption performance compared to potassium hydroxide, the resulting modified carbon pellet with potassium acetate can be used as an alternative activating agent in carbon capture applications.

3.2. Surface Morphology of Carbon-Based Pellets

The morphological changes in the surface of the carbon pellets before and after potassium acetate modification were analyzed using SEM images. As shown in Figure 4, the SEM micrographs of all samples show a closely packed surface texture with the presence of variable pore sizes. The CAC-PA 2M presents a well-developed spherical pore compared to other samples. Compared to the unmodified sample (CAC-water), the modified samples (CAC-PA 1M and CAC-PA 2M) showed the presence of many smaller pores. This is due to the surface modification with potassium acetate which resulted in more developed pore structures, hence providing more adsorption sites for the removal of CO₂ [40]. A similar observation was reported by Singh et al. [49], whereby, by adding a potassium-based activating agent, the development of small and large pores can be observed on the carbon surface, which makes the carbon pellets an attractive material for CO_2 adsorption. As seen in Figure 4, potassium acetate concentration significantly affects the pore structure, thus causing the formation of different sizes of pores as observed at CAC-PA 1M and CAC-PA 2M carbon pellets. The changes in pore structure (porosity) at low concentration of potassium acetate is non-significant. However, as the potassium acetate concentration increased at CAC-PA 3M, some pores have undergone a reduction in size until they become smaller (fewer pores on the carbon surface) and disappear. A similar trend was observed in Li et al. [50] and Khalid et al.'s [51] studies, in which increases in the concentration of the activating agent resulted in a decreased number of pores (only a few pores on the carbon surface), which is due to the destruction or blockage of the carbon pore structure. The decrease in the pore structure is because of the reaction between the carbon and the activating agent at high concentrations during the carbonization process (heating process), which destroys the pores on the carbon surface [52]. The addition of the activating agent (potassium acetate) is effective in creating more pores to enhance the adsorption site for CO₂ capturing. A highly porous structure and more micropores are easily observed in Figure 4; thus, it is proven that the modified carbon pellet of CAC-PA 2M is suitable for CO_2 adsorption.



Figure 4. SEM images of (a) CAC-PA 1M; (b) CAC-PA 2M; (c) CAC-PA 3M; (d) CAC-water at a magnification of $1000 \times$ and $2000 \times$.

3.3. Elemental Analysis of Carbon-Based Pellets

Elemental analysis of carbon pellets showed that all samples have high carbon and oxygen contents (Table 1). Oxygen content increased from 10.52% to 14.72% with an increase in the potassium acetate concentration from 1M to 2M and then decreased to 12.04% at 3M. This confirms that by adding potassium acetate (1M–2M), the enhancement of oxygen content was observed. A similar trend was observed by Singh et al. [46] and Kaur et al. [53]. The highest oxygen content was obtained at CAC-PA 2M, which is good for enhanced CO₂ adsorption. The oxygen heteroatom is important to increase the surface properties of a carbon pellet, thus increasing the adsorption capacity.

3.4. Surface Area of Carbon-Based Pellets

The highest BET surface area (883.76 m²/g) and pore volume (micropore) (0.36 cm³/g) were obtained at CAC-PA 2M. With increasing potassium acetate concentration from 1M to 2M, the BET surface area was increased from 856.44 m²/g to 883.76 m²/g, then decreased to 825.08 m²/g at a higher potassium acetate concentration of 3M. This is due to the collapsing of pores at high activating agent concentration, as was also observed by Kaur et al. [53].

3.5. CO₂ Adsorption Performance of CAC-PA 2M

3.5.1. Effect of Adsorption Temperature

Since CAC-PA 2M showed the highest breakthrough time and CO₂ adsorption capacity among all the carbon pellets, the effect of temperature on CO_2 adsorption was performed at CAC-PA 2M. Adsorption temperature is known to be the most significant factor affecting CO₂ adsorption performance. Investigating the effect of temperature is important and could help provide a clear understanding of the theory of adsorption whether it is a physical or chemical adsorption reaction [54]. The experimental temperature range was selected at 25 °C to 55 °C. As shown in Figures 5 and 6, as the temperature increased from 25 °C to 55 °C, the breakthrough time and the adsorption capacity of CAC-PA 2M decreased. For example, at a low temperature (25 °C), the longest breakthrough time of 19.4 min and the high adsorption capacity of 0.685 mmol/g were obtained. When the temperature increases up to 55 °C, the breakthrough time and the adsorption capacity decreased to 13 min and 0.404 mmol/g, respectively. This might be due to the exothermic process during CO₂ adsorption whereby this is confirmed by the physical adsorption reaction. Based on the adsorption theory, adsorption capacity decreases with increasing temperature, and it can be concluded that physical adsorption is unable to withstand high temperatures. This is due to the physical adsorption interaction between the adsorbate (CO_2 molecule) and the adsorbents (carbon pellets) via a weak van der Waals force in which the chemical bonds tend to break at high temperatures [55]. Therefore, physical adsorption processes preferably at a low temperature. Similar observations have been reported by Rashidi et al. [56].



Figure 5. Breakthrough time of CAC-PA 2M pellet sample at a varying adsorption temperature (25 $^{\circ}$ C to 55 $^{\circ}$ C).



Figure 6. Adsorption capacity of CAC-PA 2M pellet sample at a varying adsorption temperature (25 $^{\circ}$ C to 55 $^{\circ}$ C).

3.5.2. Adsorption Isotherm Study

Adsorption isotherms are important to better understand the adsorption mechanisms of carbon pellets; that is, whether it occurs through physical or chemical adsorption, depending on the interactions between the adsorbate (CO_2 molecule) and the adsorbent (carbon pellet) [55]. In the present study, three adsorption isotherm models, namely, Freundlich, Langmuir, and Temkin, were used to fit the experimental data. The isotherm models were plotted at different adsorption temperatures and CO₂ partial pressure using OriginPro 8.1 software (Origin Lab, USA). The non-linear isotherm plots for all models are shown in Figure 7. As seen in Figure 7, all isotherm models are well-fitted with the experimental data. The isotherm models were compared based on the coefficient of determination (R^2) [57]. The highest R^2 indicates a good agreement of isotherm models with the experimental data. Among the isotherm models, the Freundlich isotherm model gave a high R^2 value indicating a good fit of the experimental data (Table 2). Moreover, the Freundlich isotherm model is the most appropriate isotherm to describe the adsorption mechanism of a carbon pellet at different adsorption temperatures (Figure 8). The results are consistent with the work by Zaini et al. [26], in which the study reported that activated carbon showed the best fit with the Freundlich isotherm model. The parameters of the Freundlich isotherm model were calculated using the non-linear regression method and the results are reported in Table 3. The Freundlich constant (K_F) is an indicator of adsorption capacity while n and 1/n values are a function of the adsorption strength in the adsorption process [58]. To determine favorable adsorption, the n values in the Freundlich isotherm should be around 1–10 [59]. A larger value of n (n > 1) and a smaller value of 1/n indicates stronger interaction between the CO_2 molecules and the carbon pellet [56]. In this study, the n value is greater than 1, which indicates that the adsorption of CO_2 on the carbon pellet is favorable. Table 3 shows that a decrease in the K_F with an increase in the adsorption temperature indicates a physical adsorption process. It is observed that the adsorption on carbon pellets is more favorable at a lower temperature due to the carbon pellet surface being energetically heterogeneous [55,60]. Figure 8 shows the decrease in the values of adsorption capacity with an increase in the adsorption temperature, thus confirming that the adsorption phenomenon on carbon pellets is an exothermic process.



Figure 7. Adsorption isotherm models predicted of CO₂ adsorption on CAC-PA 2M at a varying adsorption temperature (25 °C to 55 °C).



Figure 8. Freundlich isotherm model predicted CO₂ adsorption on CAC-PA 2M at a varying adsorption temperature (25 °C to 55 °C).

Isotherm Models			
Freundlich	n	K _F *	R ²
	1.38	3.84	0.988
Langmuir	qe*	K _L *	\mathbb{R}^2
0	11.65	0.405	0.987
Temkin	B*	K _T *	\mathbb{R}^2
	0.779	15.577	0.966

Table 2. Adsorption isotherm parameters at 25 °C.

 K_{F}^{*} (mmol/g (1/atm)1/n); K_{L}^{*} (1/atm); K_{T}^{*} (1/atm); q_{e}^{*} (mmol/g); $B^{*} = RT/b$; b (J/mol).

Temperatures (°C)	n	K _F	R ²
25	1.38	3.81	0.989
35	1.29	2.59	0.963
45	1.26	2.17	0.998
55	1.19	1.48	0.861

Table 3. Freundlich isotherm parameters at different temperatures.

3.5.3. Regeneration Study

Regeneration is an important factor in determining the reusability and efficiency of an adsorbent (carbon pellet). Some adsorbents are expensive and inefficient for repeated use which will increase the operating cost due to the disposal of used adsorbents, create environmental problems, and may limit their applications on a large scale [61]. The adsorption capacity of the regenerated CAC-PA 2M carbon sample after adsorption–desorption equilibrium was conducted for five cycles with a fixed flow rate of 200 mL/min at 15% CO_2 concentration and at a temperature of 25 °C. As shown in Figure 9, the adsorption capacity of the regenerated CAC-PA 2M showed good adsorption capacity, stability, and reusability in several adsorption–desorption cycles (over five cycles). There are no significant changes in regeneration performance which can be seen since the CAC-PA 2M dropped to around 2% in the second cycle and remained constant until five cycles.



Figure 9. Regeneration study, adsorption-desorption cycle trends for CAC-PA 2M at 25 °C.

3.5.4. Surface Chemistry (Functional Groups)

The FTIR spectra for all carbon pellet samples are demonstrated in Figure 10. As seen in Figure 10, there are no obvious changes in the peak intensity for all of the modified samples (CAC-PA 1M, 2M, and 3M). Therefore, in order to compare the changes in the functional groups before and after CO₂ adsorption, the unmodified (CAC-water) and modified (CAC-PA 2M) samples were compared at different temperatures of 25 °C to 45 °C as illustrated in Figures 11 and 12, respectively. By comparing the CAC-water and CAC-PA 2M before and after CO₂ adsorption, the qualitative changes that occur in the material and the effect of the temperature can be observed by distinguishing the bands of interest, mainly, hydroxyl, carbonyl, and carboxyl groups.



Figure 10. The FTIR spectra of carbon pellet samples before CO₂ adsorption at 25 °C.

The broad band at approximately 3800 cm^{-1} corresponded to the -OH stretching vibration from the adsorbed water on the carbon surface. The peak at the range of $3610-3751 \text{ cm}^{-1}$ can be attributed to the free OH stretching of the hydroxyl functional group of alcohols and phenols, which are found in all samples. In addition, the region from 1994 to 2346 cm⁻¹ gives a clue for the presence of O-H bonds of carboxylic acids along with the presence of C-H bonds of aromatic rings [53]. The presence of -OH groups on the carbon surface confirms the presence of a large quantity of oxygen, which presents as surface functional groups on the carbon, hence leading to the preferential adsorption of CO₂. The strong bands at around 1739 cm⁻¹ and 1364 cm⁻¹ are probably associated with carbonyl and carboxylate (COO-) groups. Moreover, the adsorption peak at 1217 cm⁻¹ corresponds to the C-O-C stretching and could be related to the presence of alcohol and phenols, thus confirming the presence of OH groups on the materials and the existence of oxygen. The stretching vibration occurring between 1500 cm⁻¹ and 1600 cm⁻¹ indicated the presence of cyclic alkenes (C=C stretching) [46,62].



Figure 11. The FTIR spectra of CAC-water before and after CO₂ adsorption at 25 °C to 45 °C.



Figure 12. The FTIR spectra of CAC-PA 2M before and after CO₂ adsorption at 25 °C to 45 °C.

Figures 11 and 12 display the FTIR spectra of CAC-water and CAC-PA 2M before and after CO₂ adsorption at different temperatures. The important absorption band was observed at the double carbonyl O=C=O functional group, which can be noticed in the range of 2200–2400 cm⁻¹ [62]. The peak intensity of the CO₂ functional group increases as the temperature increases. However, increasing the regeneration temperature at 45 °C resulted in a progressive increase of the peak intensity for CAC-water but in the case of CAC-PA 2M, the peak intensity decreases. This occurrence might be due to the structural transition upon heating in the presence of potassium acetate as the activation agent. The higher intensity and the change in the position of this peak confirm that the potassium acetate activation helps in the formation of oxygen-containing functional groups on the surface of carbon pellets.

3.5.5. Surface Chemistry (XPS Analysis)

The elemental composition of the CAC-PA 2M carbon pellet using a CHNS/O analyzer showed 66.36%, 1.95%, and 14.72% of C, H, and O, elements, respectively. The result of the elemental composition exhibited the presence of high oxygen content of 14.72% for CAC-PA 2M. The presence of this oxygen can provide more active functional sites on the carbon pellet, thus helping to enhance CO₂ adsorption capacity. The effect of oxygen-containing functional groups such as hydroxyl, carboxyl, and carbonyl groups are investigated using XPS analysis [63].

XPS analysis was conducted on the CAC-PA 2M sample to understand the chemical bonding states on a carbon pellet's surface. The XPS spectra of the CAC-PA 2M carbon pellet are shown in Figures 13 and 14 and are summarized in Tables 4–6. The XPS survey spectra show the presence of carbon (C1s) and oxygen (O1s) as observed in Figure 13. The peak at 284.54 eV (C1s) is assigned to the graphitic carbon [64]. The largest peak at 532.54 eV binding energies is attributed to the different functional groups, such as the hydroxy and carboxyl groups [12,63]. The elemental composition is listed in Table 4 Based on the results in Figure 14b, the O1s have the highest peak, and it shows that oxygen has the highest composition and binding energy. It is observed that oxygen molecules are added to the carbon pellet due to the chemical activation process, which is consistent with the CHNS/O elemental composition results. Deconvolution was carried out to understand the chemical bonding states of the carbon and oxygen functional group of the CAC-PA 2M carbon pellet sample as shown in Figure 14a,b. High-resolution XPS spectra for C1s could be deconvoluted into five distinct peaks located at 283.15 eV, 285.52 eV, 284.77 eV, 286.46 eV, and 287.32 eV, indicating the presence of an aromatic ring, graphitic carbon, π - π * transitions in the aromatic rings, hydroxyl, or ether groups, carbonyl, and carboxyl groups, respectively (Table 5) [53,63]. Deconvolution of the O1s region can help to understand and provide complete information on the distribution of the surface oxygen functionalities [65]. Four distinct types of oxygen-containing functional groups of ester, hydroxyl, carboxyl, and carbonyl groups are related to the 533.45 eV, 531.91 eV, 534.23 eV, and 530.37 eV peaks, respectively, and are summarized in Table 6. The fraction of each carbon or oxygen species was obtained by determining the contribution of individual carbon or oxygen state curve regions to the total curve area. The abundance of carbon and oxygen on the surface of the carbon pellet with a high specific surface area offers a strong tendency to gas adsorption [66]. Based on the XPS analysis results, it can be concluded that the surface of the CAC-PA 2M carbon pellet sample contained rich oxygen species that would be beneficial for enhancing CO_2 adsorption capacity.



Figure 13. XPS survey spectra of CAC-PA 2M carbon pellet.



Figure 14. XPS spectra of CAC-PA 2M carbon pellet: (**a**) high resolution spectra of C1s and (**b**) high resolution spectra of O1s.

Name	Position	FWHM ^a	R.S.F ^b	Area	%Concentration
C1s	284.54	5.103	1	15,418.11	71.84
O1s	532.54	3.526	2.93	17,708.09	28.16

Table 4. XPS survey spectra of CAC-PA 2M carbon pellet.

^a FWHM: Full width at half-maximum intensity. ^b R.S.F: Relative sensitivity factors (RSF) to measure peak areas.

Table 5. XPS results of C1s spectra of CAC-PA 2M carbon pellet.

Name	Bond	Functional Group	Position	FWHM ^a	R.S.F ^b	Area	%Concentration
C1	C-C	Aromatic carbon	283.15	0.915	1	4260.82	41.28
C2	C-C/C-H	Alkane	285.52	1.085	1	3052.69	29.58
C3	C=C	Aromatic π - π *	284.77	0.941	1	1026.28	9.94
C4	C-OH/C-O-C	Hydroxyl	286.46	1.291	1	1156.67	11.21
C5	C=0 0-C=0	Carbonyl/Carboxyl/Ester	287.32	1.171	1	824.96	7.99

^a FWHM: Full width at half-maximum intensity. ^b R.S.F: Relative sensitivity factors (RSF) to measure peak areas.

Table 6. XPS results of O1s spectra of CAC-PA 2M carbon pellet.

Name	Bond	Functional Group	Position	FWHM ^a	R.S.F ^b	Area	%Concentration
01	O-C=O	Ester	533.45	1.365	2.93	6821.00	57.65
O2	C-OH/C-O-C	Hydroxyl/ether	531.91	1.574	2.93	4253.39	35.95
O3	C(=O)-OH	Carboxyl	534.23	0.849	2.93	454.45	3.84
O4	C=O	Carbonyl	530.37	1.096	2.93	302.68	2.56

^a FWHM: Full width at half-maximum intensity. ^b R.S.F: Relative sensitivity factors (RSF) to measure peak areas.

4. Conclusions

A carbon pellet with low-cost material and a green process method has been successfully developed for enhancing CO₂ adsorption capacity. The carbon pellet was prepared using chemical activation treatment by the addition of potassium acetate as an activating agent to improve the physical and chemical properties of the carbon pellet surface. The advantages of potassium acetate are low toxicity and corrosivity compared to potassium hydroxide. The carbon pellet products were characterized for their chemical and textural properties as well as CO₂ adsorption performance. This study is believed to be the first report on the modification of the surface properties of carbon pellets using potassium acetate for CO₂ capture application. The results revealed that CAC-PA 2M (modified carbon pellet) showed excellent physical and chemical properties as well as a high CO₂ adsorption capacity. Furthermore, the CAC-PA 2M achieved the highest breakthrough time of 19.4 min, and the lowest breakthrough time of 13 min was observed for CAC-water (unmodified carbon pellet).

The CO₂ adsorption capacities were attained at 0.4737 mmol/g, 0.622 mmol/g, 0.685 mmol/g, and 0.535 mmol/g, for the CAC-water, CAC-PA 1M, CAC-PA 2M, and CAC-PA 3M, respectively. This is due to the fact that potassium acetate creates more pores on the carbon pellet sample, thus increasing the breakthrough time and adsorption capacity. The FTIR spectra of the carbon pellet after CO₂ adsorption showed a significant change in the peak intensity at 2340 cm⁻¹, which is associated with the double carbonyl O=C=O functional group. This peak was only detected after the CO₂ adsorption process. In addition, the presence of high oxygen content on the carbon pellet can help to increase CO₂ adsorption capacity. It can be concluded that carbon pellets modified with potassium acetate showed excellent CO₂ adsorption performance and have the potential to be used in large-scale processes due to their low-cost material.

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