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High pressure – high temperature carbon dioxide adsorption predictive model for a volcanic fly ash in Egypt based on experimental and mathematical analysis

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Abstract
This research develops two predictive mathematical models to predict the carbon dioxide (CO_2) adsorption capacity of a volcanic fly ash from Egypt. This type of fly ash is extremely common in Egypt, which makes the development of such mathematical models extremely useful and significant. The models are developed based on experimental results conducted in the Lab under high pressure high temperature conditions for CO_2 adsorption isotherm development. The lab experiments developed isotherms at 23, 40, 60, and 80 °C based on the saturation pressure of CO_2 at these conditions. Using the experimentally developed adsorption isotherms, mathematical models were developed and tested using external data for validation. The first mathematical model was developed to predict CO_2 adsorption capacity in moles as a function of CO_2 pressure and the temperature at which adsorption takes place. The second model predicts the CO_2 adsorption mass per unit mass of fly ash used, at different pressure and temperature conditions. This is extremely important since it can help determine the required fly ash mass in the application to capture a specific amount of CO_2, based on the CO_2 capture facility capacity. The mathematical models were validated using experimental results that were not used in the model development. The models had an average prediction accuracy of 91%, which was extremely high.

1 Introduction

Carbon capture is one of the most effective means of greenhouse gas reduction from the atmosphere. It focuses on capturing CO_2 from high intensity emission sources, or directly from the atmosphere, and then utilizing or storing CO_2 rather than emitting it to the atmosphere. One of the main limitations of CO_2 capture is its high cost [1–10]. Although multiple technologies have been introduced, they remain extremely costly, and thus not feasible to apply. Development of methods and utilization of material that can reduce the overall cost of CO_2 capture is therefore imperative for the widespread commercialization of CO_2 capture [10–15]. This can help boost the market for carbon capture, utilization and storage which will assist not just the environment, but also the local and global economies significantly. CO_2 storage can be enhanced via rapid CO_2 desorption and storage in underground reservoirs under high pressure.

One of the most cost-effective methods for CO_2 capture is via adsorption. Adsorption can either be chemical or physical depending on the bond between the adsorbent and the adsorbate [15–18]. Chemical adsorption

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is classified as monolayer high energy adsorption. The bond between the adsorbate and the adsorbent requires high energy to form and also to dissociate. The adsorbent forms a single layer on the surface of the adsorbate, which limits the availability of adsorption sites, and thus the adsorption capacity is reduced significantly [18–20]. Physical adsorption, on the other hand, is multilayer low energy adsorption [20–23]. Low energy is required to create and dissociate the bond between the adsorbent and the adsorbate. The adsorbent can form multiple layers on the adsorbate, with each layer having a weaker bond strength than the one before it. CO_2 adsorption onto fly ash, slag, and zeolites occurs in the form of physical adsorption. Adsorption behavior is usually modeled using adsorption isotherms [23–30]. For chemical adsorption, several isotherms are present that can model its behavior accurately, most notably the Langmuir Isotherm [30–33]. For physical adsorption, the multilayer adsorption behavior makes modeling challenging [33–36]. This can be overcome through development of individual isotherms for different material [36–38]. Based on these individual isotherms, mathematical models can be developed to further predict the adsorption behavior of the specific material beyond the extent of the experiments conducted.

Many researchers studied the use of material such as slag, shale, fly ash, and zeolites for CO_2 capture via adsorption. Several studies investigated the adsorption behavior of CO_2 in unconventional shale oil formations for CO_2 storage potential with a focus on adsorption for the purpose of CO_2 storage and not CO_2 capture [33–39]. Some studies explored the potential for CO_2 storage in coal seams. CO_2 is collected from flue gas in power plants and then injected into the coal seams. The studies did not focus on the collection process of CO_2 for carbon capture. A comprehensive review of CO_2 capture through both absorption and adsorption. The study reviewed several materials including ash and slag in multiple organic frameworks for CO_2 adsorption. Experimental work showed that CO_2 adsorption can be done with high efficiency when covalent organic frameworks are used along with direct air adsorption techniques. This method is extremely promising since it allows for the direct capture of CO_2 from air [25–39].

Due to the variation in fly ash properties from one location to the other due to the origin of the fly ash, it is important to investigate the adsorption properties of each fly ash separately. Based on this, this research builds upon the experimental results of Fakher et al. [1] that developed multiple isotherms for an extremely common volcanic fly ash present in Egypt, while focusing on high pressure applications. This research constructs two mathematical models with high prediction accuracy to determine, mathematically, the CO_2 adsorption capacity of the experimentally tested fly ash as a function of pressure, temperature, and fly ash mass. This can allow for the direct prediction of CO_2 capture volume, which is an imperative parameter when designing any CO_2 capture facility.

2 Mathematical model development

The mathematical models developed in this research are based on experimental results conducted on the same fly ash. Several studies have indeed investigated the ability of fly ash to adsorb CO_2. The main difference between this research and previous studies is the type of fly ash used to conduct experiments. Different types of fly ash have different adsorption capacities and capabilities depending on the origin of fly ash. It is difficult to compare different fly ash samples together due to their different origins, and therefore, their different structures and compositions. The details of the experimental material are mentioned in Fakher et al. [1]. The model was developed based on three main steps that were conducted to be able to create a common link between all the experimentally developed isotherms. The steps followed to construct the mathematical models are as follows:

a. After conducting the CO_2 adsorption experiments, the data was plotted for each adsorption isotherm generated. This step involved determining the behavior of each adsorption isotherm in order to determine the appropriate function to use. This was done based on the R^2 values for each isotherm, and in correlation to previous research that was conducted to develop isotherms for CO_2 with fly ash, zeolites, and furnace slag.
b. Once the appropriate function was determined, the trendlines were generated, and the R^2 value for each trendline was determined. If the R^2 value for any trendline was less than 0.9, the experiments were repeated to ensure that there was no experimental error. After all plots had an acceptable R^2 value, the plan for the mathematical models was developed based on the most significant parameters required for modeling. Based on this, fitting parameters were obtained for each mathematical correlation based on the experimental data. The fitting parameters were used to create functions of more than two variables, which is imperative in the CO_2 adsorption equations since they are impacted by multiple variables.
c. Based on the fitting parameters obtained from the trends, mathematical models were developed. Following this, a validation of mathematical models was conducted. This was done by testing the prediction ability of the mathematical model and comparing the results to those of actual experimental results that were not used in the development of

Table 1 Pressure and saturation pressure values

Temperature (°C)	Saturation Pressure (MPa)
23	6.35
40	9.60
60	15.48
80	24.30

the mathematical models. Both models resulted in an average accuracy of 91%, which is considered reasonably high.

The temperatures and pressure conditions were selected based on the most common application for CO_2 capture using adsorption-based methods. The relative humidity for all experiments was controlled at 42%. The pressure values used were 1.46, 2.81, 4.21, 5.53 MPa. The saturation pressures at different temperature conditions are presented in Table 1. The saturation pressures were calculated using Clausius Clapeyron equation.

The first mathematical model can be used to predict adsorption of CO_2 in mmol g^{-1} fly ash used, as a function of pressure and temperature. The second mathematical model can be used to predict the mass of CO_2 adsorbed instead of moles, which is easier and more realistic for real life applications. The first mathematical model is more applicable in lab-based applications. Both models are in total agreement, with only a variation in terms of reporting the volume of CO_2 adsorbed.

3 Mathematical model 1: CO_2 adsorption in moles as a function of temperature and pressure

The first mathematical model developed in this research was aimed at creating a correlation between the CO_2 adsorption capacity in mmol g^{-1} as a function of the operating temperature and pressure of the CO_2. The fitting of all the data points obtained from the experiments conducted at 23, 40, 60, and 80 °C is presented in Fig. 1. As the temperature of the experiment increased, the R^2 also increased. This indicates that the developed mathematical model will be able to predict the adsorption values at elevated temperatures better. This was verified after the models were developed and was true, with the lowest error percentages being at the highest tested temperature. For all cases, the fitting was forced to pass through the coordinate (0,0). This was imperative since at a pressure of zero, no adsorption took place on the sample. With the increase in temperature, the slope of the trendline also increased. This is an indication of the increase in the adsorption potential with the increase in temperature. If the slope decreases, this indicates an almost constant change in adsorption with pressure, while a steeper slope shows potential for more adsorption at higher pressure values. This is due to the expansion of

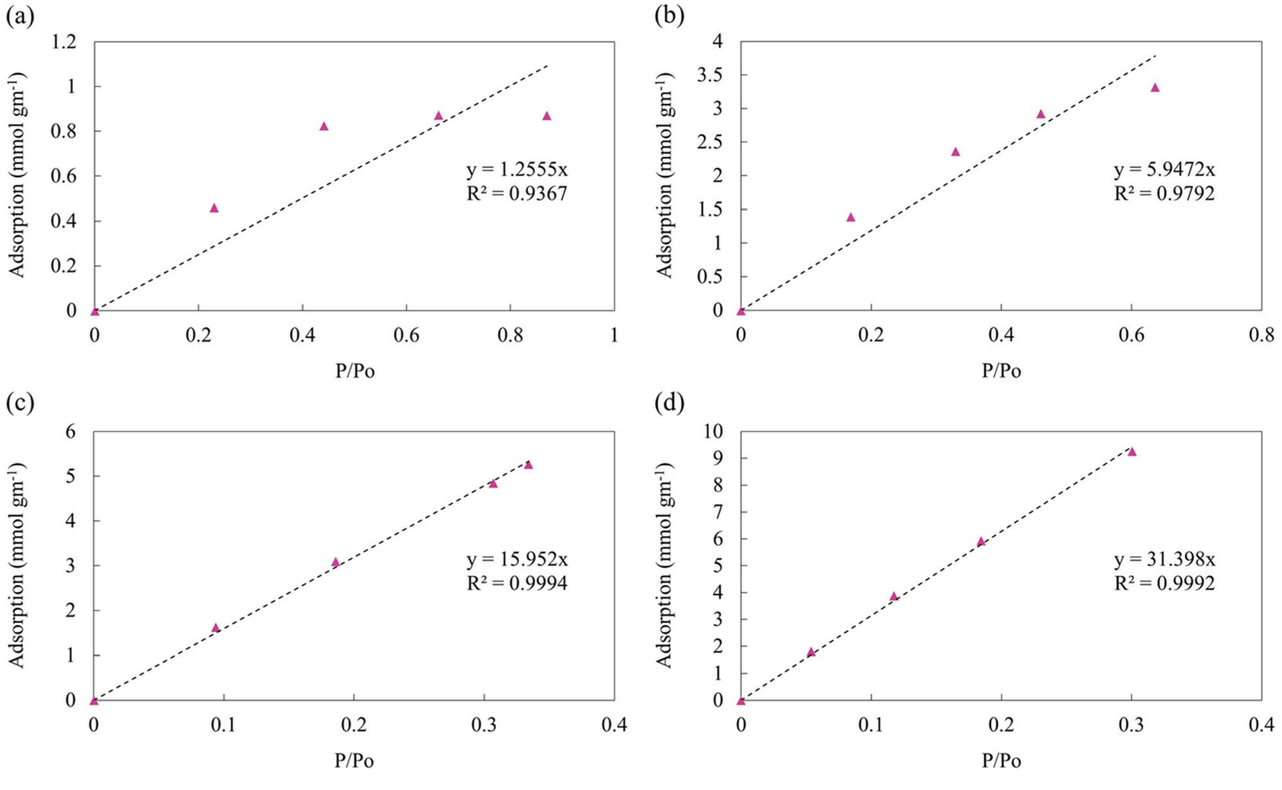

Fig. 1 CO_2 Adsorption isotherms for fly ash (**a**) 23 °C, (**b**) 40 °C, (**c**) 60 °C, (**d**) 80 °C

Table 2 Slope value for each temperature trendline

Temperature (ºC)	Trendline Slope (m)
23	1.26
40	5.95
60	16.0
80	31.4

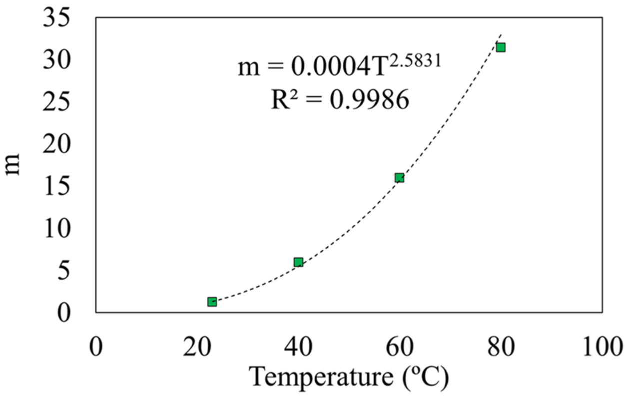

Fig. 2 Slope fitting parameter for CO_2 fly ash adsorption isotherm at different temperature values

the fly ash particles at higher temperatures, which creates more adsorption sites, due to the increase in surface area, as was explained by Fakher et al. [1].

Following the generation of equations for all adsorption isotherms, shown in Fig. 1, a fitting parameter was needed in order to combine all four equations together. Since all the equations generated for the isotherms were straight line equation with no y intercept, the slopes for all four lines were plotted versus temperature to generate a generalized equation for the slope. The slope values for each adsorption isotherm are shown in Table 2.

Figure 2 shows the plot for the fitting parameter based on the slope of the isotherm lines at each temperature. The best fit line followed a power law model. The function obtained for the slope "m" can then be used in the original linear equation as a fitting function for the initial equation. By substituting the power law function obtained from the relation between the slopes of the isotherms and the temperature, a general equation can be derived for all four isotherms.

Based on the fitting equation generated for the slope values of the isotherms and the general straight-line equation that was found to fit the behavior of all the adsorption isotherms, the mathematical model to predict the CO_2 adsorption capacity in mmol/gm as a function of temperature, pressure, and CO_2 saturation pressure is as follows:

$$Ads_{CO_2}\left(mmol\ g^{-1}\right) = \left(0.0004T^{2.58}\right)\frac{P}{P_o}$$

Where Ads_{CO_2} is the CO_2 adsorption capacity in mmol g^{-1}, T is the operational temperature of the adsorption facility in ºC, P is the operating pressure in MPa, and Po is the saturation pressure of the CO_2 at the operating pressure, in psi.

4 Mathematical model 2: CO_2 adsorption mass as a function of temperature and pressure

The second mathematical model developed in this research was used to predict the mass of CO_2 adsorbed per mass of fly ash used in the adsorption facility as a function of operating temperature, operating pressure, and CO_2 saturation pressure at the operating temperature. Initially, two attempts were made, one to determine the mass of the CO_2 as a function of fly ash mass, and the second was to determine the volume of the CO_2 as a function of fly mass. Based on the experimental results and the previous research, it was found that it would be much more practical to create a correlation for the CO_2 mass as a function of fly ash mass. If the volume of CO_2 is required, it can be easily obtained via the density of the CO_2 as the specified pressure and temperature conditions.

By determining the quantity of CO_2 adsorbed per mass of fly ash, the amount of fly ash needed for a CO_2 adsorption facility or setup can be estimated. This shows the importance of developing this particular mathematical model, since it can directly assist in the design of a CO_2 capture system that relies on the volcanic fly ash used in this study. Figure 3a shows the CO_2 mass and equivalent volume adsorbed g^{-1} fly ash at 23 °C. As the CO_2 pressure increases, the CO_2 adsorption mass also increases. This is due to the increase in the available adsorption sites due to the physical adsorption stacking effect. It can be noted however, that at the pressure ratio 0.66 and beyond, the points begin to normalize. This is an indication that the maximum adsorption potential is being reached. The CO_2 volume behavior is the opposite of the mass behavior. With the increase in CO_2 pressure, the volume decreases. This is due to the compression of the CO_2 molecules at elevated pressures, which results in a reduction in volume. The reduction in volume behavior will be impacted by both the pressure and temperature and can easily be calculated using the mass and the density of the CO_2.

Figure 3b shows the CO_2 adsorbed mass and the equivalent volume at 40 °C. The same trend is observed as in the 23 °C experiment, with two distinctions. Firstly, the slope of the CO_2 mass line increased. This is an indication of the increase in CO_2 adsorption, especially at elevated temperatures. This can also be observed at the highest pressure ratio where the pressure point shows a slightly higher increasing trend compared to the lower temperature experiment. The slope for the CO_2 volume

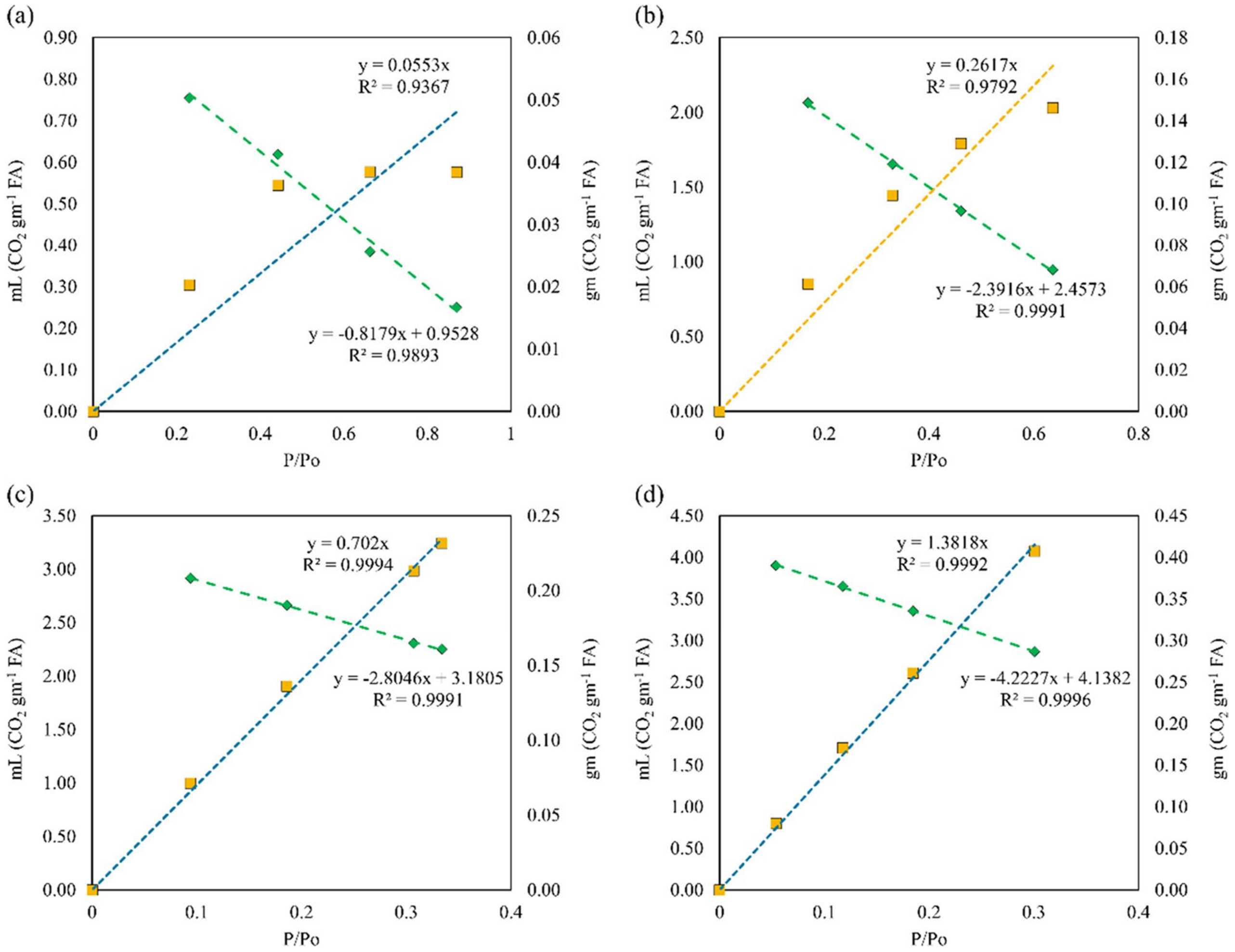

Fig. 3 CO_2 Adsorption mass and volume per mass of fly ash isotherm at (**a**) 23 °C, (**b**) 40 °C, (**c**) 60 °C, (**d**) 80 °C

also increased in the negative direction. This is due to the expansion of the CO_2 at elevated temperature, hence the increase in the volume. This can also be observed in the values of the CO_2 volume, which are higher than those of the 23 °C experiment.

Figure 3c shows the CO_2 adsorbed mass and the equivalent volume at 60 °C. At this temperature, the CO_2 mass line shows an increasing trend. This indicates that the maximum adsorption potential is yet to be reached. Also, the CO_2 volume increased due to gas expansion at higher temperatures.

Figure 3d shows the CO_2 adsorbed mass and the equivalent volume at 80 °C, which is the highest temperature experimentally evaluated in this research. At this temperature, the highest CO_2 adsorption capacity and the highest CO_2 volumes were reached. This was due to the increase in available adsorption sites due to the fly ash expansion under high temperature. Also, gas expansion at elevated temperatures resulted in a decrease in the CO_2 compressibility, and an increase in CO_2 volume. This is a strong indication of increase in CO_2 adsorption to this particular type of fly ash with the increase in temperature.

Following the generation of the trendline for all four CO_2 mass/FA mass behaviors and their equivalent equations, the slope was used to generate a fitting equation for all four temperatures. This is similar to what was done in the first mathematical model. Figure 4 shows the fitting of the slope value, denoted "k" for all four experiments, and the equivalent fitting equation. A power law model was found to have the best fit for the data, as was the case in the initial mathematical model. This was then used to develop the generalized mathematical model for CO_2 mass per FA mass.

Table 3 shows the slope value for each experiment conduted at different temperatures for the CO_2 mass caluclations. These values were used to generate the generalzied mathematical model which can be used to determine the CO_2 mass for a given mass of fly ash.

Based on the fitting equation generated for the slope values of the isotherms and the general straight-line equation that was found to fit the behavior of all the

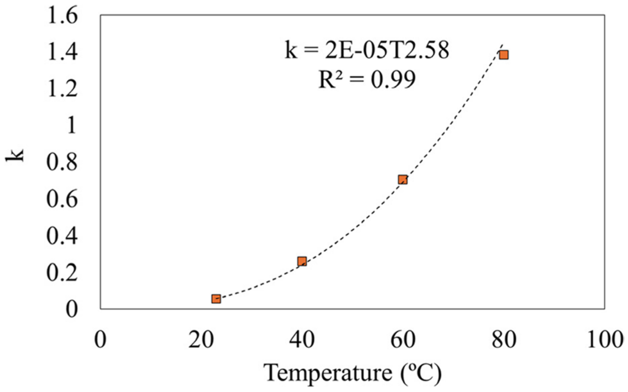

Fig. 4 Slope fitting parameter for CO_2 mass fly ash adsorption isotherm at different temperature values

Table 3 Fitting parameter value for temperature and mass trendlines

Temperature (°C)	Trendline Slope (k)
23	0.055
40	0.26
60	0.70
80	1.38

adsorption isotherms, the mathematical model to predict the CO_2 mass per mass of fly ash used as a function of temperature, pressure, and CO_2 saturation pressure is as follows:

$$\frac{\mathrm{mass}_{\mathrm{CO_2}}}{\mathrm{mass}_{\mathrm{FA}}} = \left(2\times\ 10^{-5}\mathrm{T}^{2.58}\right)\frac{\mathrm{P}}{\mathrm{P_o}}$$

Where $mass_{CO_2}$ is the mass of the adsorbed CO_2 in grams, $mass_{FA}$ is the mass of the fly ash used in the setup in grams, T is the operational temperature of the adsorption facility in °C, P is the operating pressure in MPa, and Po is the saturation pressure of the CO_2 at the operating pressure, in MPa.

5 Limitations of mathematical models developed

The volume of CO_2 adsorbed is a strong function of the volume of fly ash used and the pressure and temperature conditions. As for the cost of the application, the cost for utilization of the fly ash is extremely low due to three reasons. Firstly, the fly ash itself is extremely low in cost due to it being a waste product that does not have a well developed market in many places in the world. Secondly, the fly ash is not consumed in the adsorption/desorption process, therefore it can be used indefinitely and does not require replacement. Finally, since the adsorption process for CO_2 and fly ash is physical adsorption, desorption can be done easily and does not require any chemical interference, therefore the structure of the fly ash remains unchanged.

The two mathematical models developed in this research managed to predict CO_2 adsorption with high accuracy. The mathematical models were validated using validated using more than 50 data points from 10 experimental sets that were not used to develop the models. The experiments conducted for adsorption included multiple fly ash samples and more than 120 experiments. The experiments conducted beyond the range of the mathematical models were in alignment of the models with an R^2 of more than 0.95. The developed models have several limitations that must be mentioned, however. These include the following:

- The models were developed for only one type of fly ash. Since different fly ash samples have different properties, this can impact the adsorption behavior of the CO_2 on the fly ash. It is therefore important to generate different models for different samples.
- The models are restricted to the conditions in which the data is used to develop. The experiments were conducted under specific temperature and pressure conditions. Also, the data used for validation was for the same fly ash sample. It is therefore unknown whether the developed models will be able to yield results with high accuracy under the different conditions. Also, the models were only developed for CO_2.
- The models were developed based on volumetric adsorption method experiments. If a different experimental setup is used to measure adsorption, this could lead to slightly different adsorption values.
- The models were developed based on pure CO_2 capture experiments. They did not take into account a mixture of gases or impure CO_2.

6 Conclusions

This research develops two mathematical models that can be used to predict the CO_2 adsorption to a volcanic fly ash sample from Egypt. The mathematical models were developed as a function temperature, pressure, CO_2 saturation pressure, and CO_2 adsorption in moles and grams. The main conclusions obtained from this research are as follows:

- For all adsorption isotherms for the volcanic fly ash used, a linear trend was observed. The lines had a changing slope, and a zero intercept due to lack of adsorption at zero pressure.
- When creating a fitting equation for each mathematical model, the best fit equation followed a power law model in both cases. The power law model was a function of temperature and the slope values.

- Increasing the temperature resulted in an increase in the CO_2 mass adsorbed to the experimental setup. This was due to the expansion of the fly ash particles at higher temperatures which resulted in an increase in the available adsorption sites.
- Increasing the pressure resulted in an increase in the mass of the CO_2 adsorbed. On the contrary, the volume of the CO_2 decreased due to increase in fluid compressibility.

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Authors' contribution
Sherif Fakher: Conceptualization; Funding; Experiments; Equipment; Writing and Revising Manuscript; Methodology.
Abdelaziz Khlaifat: Funding; Experiments.
Ali El-Sayed: Experiments.
Ann Maria Salib: Experiments.

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Data availability
The datasets during and/or analyzed during the current study are available from the corresponding author on reasonable request.

Declarations

Competing Interests
The authors declare that they have no competing interests.

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