

# CO<sub>2</sub> Capture via Adsorption Using Silica Gel

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## ABSTRACT:

This paper investigates the potential of silica gel as an effective adsorbent for CO<sub>2</sub> capture. The study explores the adsorption mechanisms, the efficiency of CO<sub>2</sub> uptake, and some factors influencing the adsorption capacity of silica gel. The experimental results demonstrate the significant potential of silica gel for CO<sub>2</sub> adsorption under various conditions. The adsorption capacity was found to be highly dependent on parameters such as gas flow rate and the particle size of silica gel. The findings suggest that with optimized conditions, silica gel could be a viable material for reducing atmospheric CO<sub>2</sub> levels. This research contributes to the development of sustainable and efficient technologies for mitigating climate change through carbon dioxide capture and storage.

*Keywords: CO<sub>2</sub> capture, adsorption, efficiency, silica gel*

## 1. Introduction

In the global effort to combat climate change, the reduction of carbon dioxide (CO<sub>2</sub>) emissions has become increasingly urgent on a global scale. With atmospheric CO<sub>2</sub> levels rising due to anthropogenic activities such as industrial processes and the combustion of fossil fuels, innovative approaches for CO<sub>2</sub> capture and storage are required (Rogelj et al., 2018). Among the many ways to capture CO<sub>2</sub>, adsorption has emerged as a promising technique, characterized by its versatility and efficacy (Buckingham et al., 2022). Unlike some other methods that may require extensive infrastructure and high energy input, adsorption involves the physical or chemical adherence of CO<sub>2</sub> molecules to a solid adsorbent material (Petrovic et al., 2021). This technique is widely employed for hydrogen purification in gas streams through CO<sub>2</sub> capture, while also offering adaptability to various CO<sub>2</sub> sources in industrial facilities, making it a versatile option for emissions reduction.

Among various materials, silica gel has emerged as a promising adsorbent for CO<sub>2</sub> due to its high surface area, pore structure, and thermal stability (Buckingham. et al., 2022). The adsorption properties of silica gel can be fine-tuned through various modifications, making it a versatile material for gas capture applications. These modifications include the incorporation of functional groups, alteration of pore size distribution, and the creation of composite materials, which have been shown to significantly improve the adsorption capacity of silica gel (Todorova et al., 2015; Ravutsov et al., 2021). Recent advancements in nanotechnology have also led to the development of nano-structured silica gels, offering

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enhanced performance due to their increased surface area and optimized pore structures (Amaraweera *et al.*, 2023).

Furthermore, studies have explored the synthesis of composite materials that combine silica gel with other adsorbents, such as activated carbon and metal-organic frameworks (MOFs), to further enhance its CO<sub>2</sub> capture efficiency (Younes *et al.*, 2020; Mahajan & Lahtinen, 2022). These composites leverage the synergistic effects of different materials, leading to improved adsorption kinetics and selectivity under varying environmental conditions, including changes in temperature and pressure (Amaraweera *et al.*, 2023). Despite these significant advancements, there is a growing interest in exploring the impact of physical properties, such as particle size and thermal pre-treatment, on CO<sub>2</sub> adsorption efficiency. These factors remain less studied but hold substantial potential for optimizing the material's performance in industrial applications (Dziejarski *et al.*, 2023).

This study aims to explore the CO<sub>2</sub> adsorption capacity of silica gel, examining both granular and powdered forms under two gas flow rates. Through investigation of the kinetics of CO<sub>2</sub> adsorption, the authors aim to clarify the mechanisms that govern this process. The purpose is to identify the optimal conditions to maximize the efficiency of CO<sub>2</sub> capture. The findings contribute to the development of more effective and sustainable CO<sub>2</sub> adsorption technologies, addressing the urgent need for improved carbon capture solutions in the fight against climate change.

## 2. Materials and methods

### 2.1 Adsorbent used

Silica gel was supplied by PanReac AppliChem (Barcelona, Spain) and used as an adsorbent in two different particle sizes: granular and powder (named as S-G and S-P, respectively). Particle sizes of 2.5-6 mm constituted the granular silica gel, while the powder silica gel, obtained by crushing the former, included particles of 0.025-0.5 mm. The main specifications of the silica gel used are listed in Table 1. These materials were dried in an oven at 105 °C for 12 hours before their use in adsorption process.

**Table 1:** Specifications of silica gel

Specification	
Product name	Silica Gel 2,5-6 mm with indicator (without cobalt chloride) for analysis, ACS
Loss on drying (150°C)	max. 2.0%
Adsorption capacity (24 h, r.H. = 80 %)	min. 27%
Amber colour	activated
Light yellow colour	worn out (regenerate)

The following properties of the silica gel used were assessed to characterize the material.

Scanning electron microscopy (SEM) analyses of S-G were performed using a JEOL JSM-6700F field emission SEM (Tokyo, Japan). The sample powder was fixed on a carbon adhesive tape and coated with a thin layer of gold of approximately 15 nm. Images were taken with different acceleration voltages in the range 5-15 kV and at different magnifications. Energy-dispersive X-ray spectrometry (EDS) microanalyses were performed using an Oxford Instruments X-act EDS (Oxford Instruments, Abingdon, UK) coupled to the field emission SEM and data was treated with Oxford AZtecOne software.

The mineralogical composition of S-G was investigated by X-ray diffraction (XRD) using a Siemens D-5000 diffractometer (Karlsruhe, Germany) with  $\text{CuK}\alpha$ -radiation in the  $2\theta$  range from 10 to 70°.

Finally, the specific surface area, pore size, and pore volume of both type of samples, i.e. S-G and S-P, were characterized by a Micromeritics ASAP 2020 analyser (Quantachrome, USA) using nitrogen physisorption and applying the Brunauer–Emmett–Teller (BET) theory. Approximately 0.05 g of material was initially degassed at 90 °C under vacuum for 60 min. Then, the temperature was raised to 150 °C and held for 240 min.

## 2.2 CO<sub>2</sub> adsorption experiments

Adsorption tests with silica gel were conducted in a cylindrical PVC fixed-bed reactor with dimensions of 34 cm in height and 6.35 cm in diameter. The reactor was loaded with  $242\pm 17$  g of silica gel in granules or  $210\pm 2$  g in tests conducted with silica gel in powder form and fed with a synthetic syngas. The composition of the syngas was 50% hydrogen, 15% carbon dioxide, 20% carbon monoxide, and 15% methane. Experiments at 1 bar pressure and two different gas flow rates, 0.5 L/min and 1 L/min, were carried out. The composition of the outlet gas was continuously analysed using a maMoS Madur gas analyser (Zgierz, Poland). Measurements were recorded every second for greater accuracy. A schematic of the experimental setup used is presented in Figure 1. Breakthrough curves for CO<sub>2</sub>, adsorption capacity, and kinetic model under different working conditions were obtained from the recorded data.

The tests were considered complete when the composition of the outlet gases matched that of the inlet gases. The experiments were conducted in triplicate for the same pressure and flow rate, replacing the adsorbent with a new one after each test.

The CO<sub>2</sub> adsorption capacity (mg/kg) of silica gel in granules and powder was determined for the two flow rates studied from the breakthrough curves using the following equation:

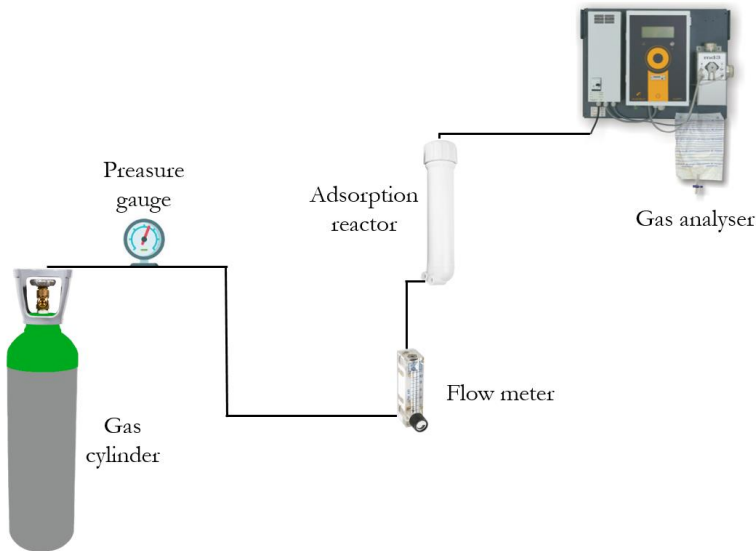
$$\left(\frac{x}{M}\right) t_{exh} = \frac{Q \cdot M \cdot P}{w \cdot R \cdot T} \cdot \left(C_0 \cdot t_{exh} - \int_0^{t_{exh}} C(t) dt\right) \quad (1)$$

Where:  $Q$  is the inlet flow rate (L/min),  $M$  is the molecular weight of CO<sub>2</sub> (g/mol),  $P$  is the pressure (atm),  $w$  is the mass of the adsorbent (kg),  $R$  is the ideal gas constant,  $T$  is the temperature (K),  $C_0$  is the inlet concentration of CO<sub>2</sub> and  $t$  represents the adsorbent saturation time (min).

Furthermore, the adsorption efficiency (%) was analyzed in each case using the following equation:

$$\text{Efficiency [\%]} = \frac{C_{f_{H_2}} - C_{i_{H_2}}}{C_{i_{H_2}}} \cdot 100 \quad (2)$$

Where:  $C_{f_{H_2}}$  is the maximum hydrogen concentration recorded during the test, occurring at breakthrough time, and  $C_{i_{H_2}}$  is the percentage of hydrogen present in the feed syngas (50%).



**Figure 1:** Experimental setup diagram adsorption

### 2.3 Adsorption kinetic models

Silica gel is an inorganic solid whose adsorption capacity is determined by its ability to trap gases in its pores through the action of intermolecular forces such as Van der Waals forces. Understanding the adsorption kinetics of this material is crucial for achieving a highly efficient adsorption process. Information on the adsorption rate, the mechanisms involved in the process, or the time at which material saturation occurs is provided by kinetic models.

The adsorption process occurs in different stages (Song et al., 2016): i) external transport of the solute from the bulk gas phase to the gas film surrounding the adsorbent, ii) the gas film must be traversed by the adsorbate to reach the surface of the adsorbent, iii) interparticle diffusion, where the adsorbate reaches the surface of the adsorbent and moves between the particles of the adsorbent before reaching the pores of the solid, iv) intraparticle diffusion within the pores of the adsorbent, and finally, v) adsorption of the molecules on the surface of the internal pores of the adsorbent.

In this research, a kinetic study of silica gel was conducted. Different kinetic models were evaluated (Avrami, Ritchie or Elovich model, among others), selecting those that provided the best results for the analysed adsorbent, silica gel, in its two granulometries (granular and powder). These models allow for the optimization of adsorption mechanisms by determining the dependence of adsorption process outcomes on the surface properties of the adsorbent. The adequacy of the studied models was determined by the regression coefficient ( $R^2$ ). This coefficient ranges from 0 to 1 and indicates how well the data fit a linear regression.

$$R^2 = \frac{\Sigma(q_{mean}-q_{cal})^2}{\Sigma(q_{cal}-q_{mean})^2+\Sigma(q_{cal}-q_{ex})^2} \quad (3)$$

### 2.3.1 Avrami model

The model is represented by the following equation:

$$\frac{dq}{dt} = k_A^{n_A} \cdot t_A^{n_A-1} \cdot (q_e - q_t) \quad (4)$$

Where  $k_A$  is the kinetic constant and  $n_A$  the Avrami exponent. Changes that may occur in the adsorption process are represented by this exponent. It also represents the dimensional growth of the sites where adsorption takes place as follows (Cestari et al., 2005):

- $n_A = 1 \rightarrow$  It implies homogeneous adsorption where there is an equal probability of adsorption occurring in any region at a given time.
- $n_A = 2 \rightarrow$  Represents one-dimensional growth.
- $n_A = 3 \rightarrow$  Assumes two-dimensional growth.
- $n_A = 4 \rightarrow$  Represents three-dimensional growth.

The integrated form of this kinetic model is represented by:

$$q_t = q_e \cdot \left(1 - e^{-\frac{(k_A \cdot t)^{n_A}}{n_A}}\right) \quad (5)$$

### 2.3.2 Ritchie kinetic model

The Ritchie equation is based on the adsorption of gas in the active sites of the adsorbent, where the adsorbate can occupy  $n$  active sites. This equation does not consider the desorption process. The Ritchie equation is represented by:

$$\frac{\partial \theta}{\partial t} = \alpha \cdot (1 - \theta)^n \quad (6)$$

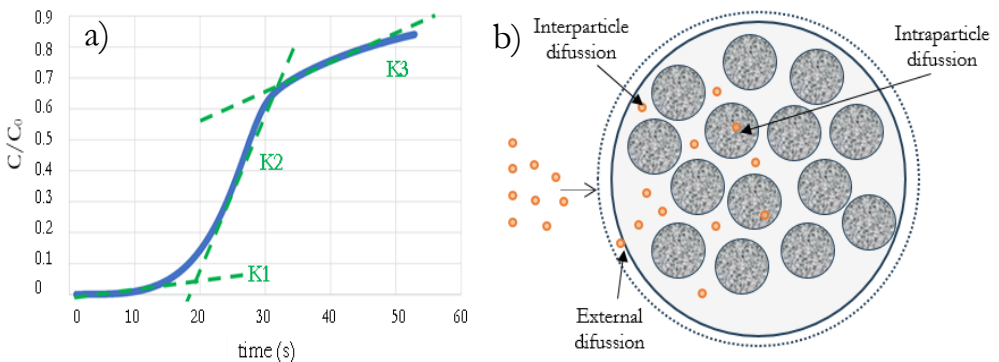
The integrated form of this kinetic model is represented by:

$$q_t = q_e \cdot \left((n - 1) \cdot k_n \cdot t + q_e^{1-n}\right)^{\frac{1}{1-n}} \quad (7)$$

Where  $k_n$  is Ritchie's constant,  $q_e$  is the maximum amount adsorbed and  $n$  is the order. The parameters of this kinetic model were fitted by nonlinear regression methods using MatLab.

## 2.4 Adsorption diffusion study

The different granulometry of the silica used in the adsorption tests, granular (2.5-6 mm) and powder (0.025-0.5 mm), can lead to different behavior during adsorption tests. To further elucidate the mechanism of CO<sub>2</sub> adsorption, an intraparticle diffusion model is employed. Therefore, a check by means of Weber-Morris slopes was conducted in order to corroborate where the diffusion process is carried out. The Weber-Morris kinetic model generates a curve with three slopes (Saleh, 2022). Each of these slopes identifies the different stages of the adsorption process (Figure 2): i) diffusion through the film where the mass transfer of each of the species from the fluid phase to the outermost solid phase takes place ii) the inter-particle diffusion that generally occurs through the pores of the adsorbent surface and iii) the surface diffusion, once equilibrium is reached, produced in the active zones of the adsorbent. For each CO<sub>2</sub> molecule, it can be deduced that three processes influence the rate of adsorption; however, only one step is rate-controlling in the overall CO<sub>2</sub> adsorption process, the slowest stage (the stage with the lowest slope) (Amaraweera *et al.*, 2023).

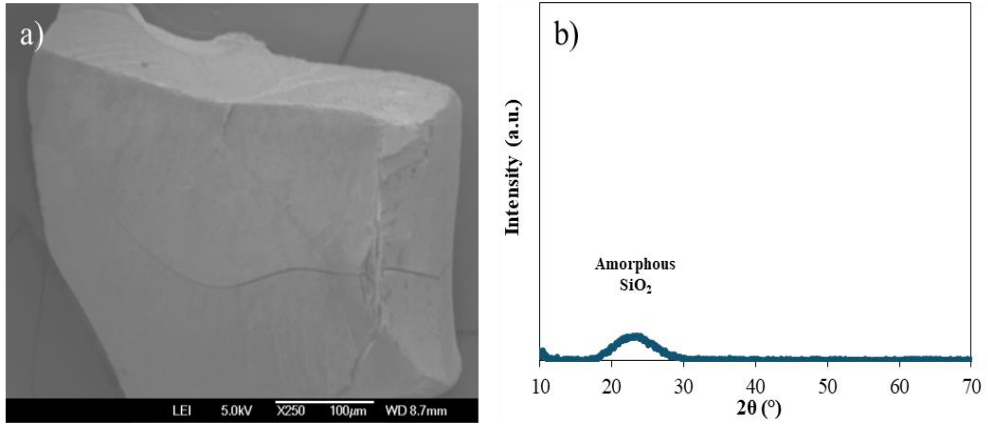


**Figure 2:** a) Slopes on a curve of the Weber-Morris model, b) Adsorption process from a kinetic point of view.

## 3. Results and discussion

### 3.1 Adsorbents characterization

Figure 3 shows in detail the tabular morphology of the silica gel particles used in the study. The chemical analysis obtained by EDX indicates that it is composed exclusively of Si and the diffractogram obtained by XRD shows a single peak associated with the presence of amorphous nature of silica (Ni'mah *et al.*, 2022) (Khan *et al.*, 2021) (Nallathambi *et al.*, 2011), without the presence of other impurities.

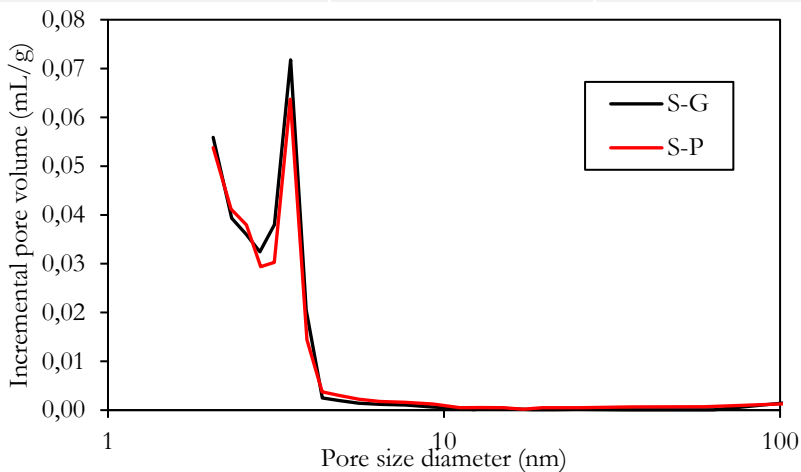


**Figure 3:** SEM image of the studied silica gel (a) and XRD patterns of the studied silica gel.

Figure 4 shows that the milling process hardly affects the micro and mesoporous structure of the silica gel used, observing that both maintain a similar microporous volume (see Table 2). In both cases, almost 98% of the pores are in the pore size range between 5-2 nm, which correspond according to the IUPAC classification to mesopores.

**Table 2:** BET surface area, total pore volume, and average pore diameter of the studied silica gel.

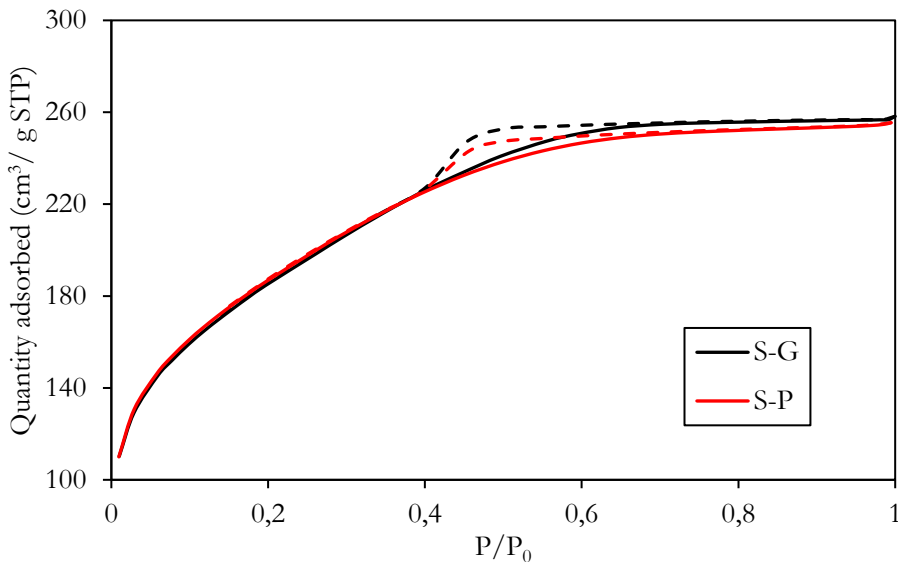
	S-G	S-P
BET surface area ( $\text{m}^2 \cdot \text{g}^{-1}$ )	666.90	671.85
Total Pore volume ( $\text{cm}^3 \cdot \text{g}^{-1}$ )	0.397	0.393
Total Micropore volume ( $\text{cm}^3 \cdot \text{g}^{-1}$ )	0.046	0.046
Average pore diameter (nm)	2.807	2.806



**Figure 4:** Pore size distribution of both silica gel used as adsorbents.

The only variation observed is that the milling process increases the specific surface area, which is logical as the particle size decreases, and slightly reduces the total pore volume (within the measurement range of the equipment that corresponds to the micro and mesopores). The SSA of the silica gel is around 15 times higher than shown by other adsorbents also used in CO<sub>2</sub> capture such as activated carbon (Encinar *et al.*, 2023).

Figure 5 shows the nitrogen adsorption–desorption isotherms obtained results. The amount of N<sub>2</sub> adsorbed by both adsorbents is similar although slightly higher in S-G. According to IUPAC, the adsorption curve of both silica gels is mainly a type I adsorption isotherm, since both exhibit asymptotic behaviour as the P/P<sub>0</sub> ratio increases. This isotherm is typical of a monolayer adsorption and is consistent with the Langmuir equation. The isotherm I may be due to the existence of chemisorption, adsorption in micropores, or physical adsorption on very homogeneous surfaces. Both adsorbents also show a certain hysteresis typical of type IV isotherms, indicative of the presence of mesopores, where adsorption occurs in the form of multilayer. The combination of type I and type IV isotherms demonstrate the co-existence of micro and mesopores in the structure of both silica gels as occurs in other studies (Mojoudi *et al.*, 2019).

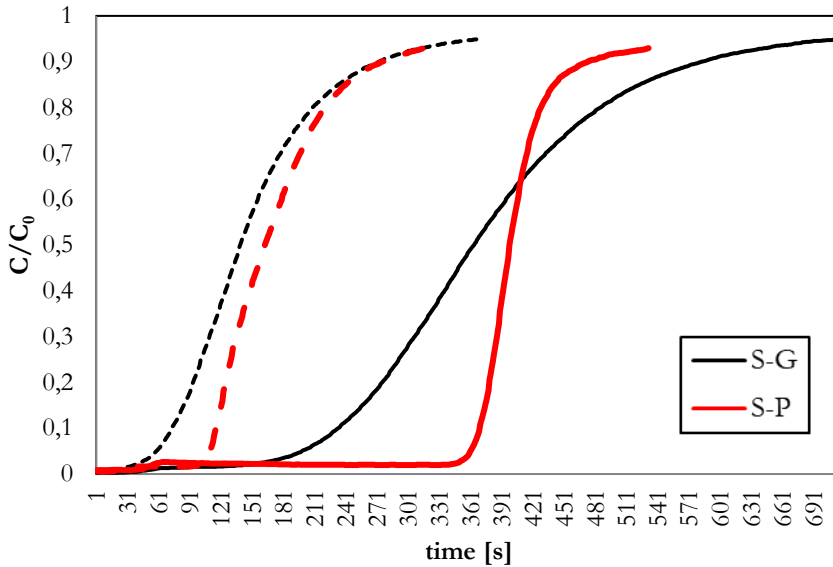


**Figure 5:** N<sub>2</sub> adsorption/desorption isotherms at 77.35 K for the selected adsorbents (solid line-adsorption; dashed line-desorption).

### 3.2 Breakthrough curves

Breakthrough curves for carbon dioxide were obtained when granular silica gel (S-G) and powdered silica gel (S-P) were used as adsorbents in the adsorption process (Figure 6). Experiments at a constant pressure of 1 bar and 2 different gas flow rates, 0.5 L/min and 1 L/min, were conducted. In these curves C represents the outlet CO<sub>2</sub> concentration and C<sub>0</sub> is the inlet CO<sub>2</sub> concentration.





**Figure 6:** Effect of flow rate at 1 bar on the adsorption breakthrough curves (solid line 0.5 L/min; dashed line 1 L/min).

Longer adsorption times were recorded when flow rates of 0.5 L/min were applied for both granular and powdered silica gel. For S-G, breakthrough times close to 200 seconds were measured, reaching saturation of the material at times around 700 seconds (Table 3). Breakthrough times five times lower were detected when the feed gas flow rate was increased to 1 L/min. Thus, lower CO<sub>2</sub> adsorption capacity was observed and 5% less H<sub>2</sub> purification efficiency was obtained in the latter case (Table 3).

**Table 3:** Granular silica gel (S-G) and powdered silica gel (S-P) results.

Adsorbent	Flow rate (L/min)	Breakthrough time (s)	Time (s)	*Q <sub>tb</sub> (mg/kg)	Q <sub>max</sub> (mg/kg)	Efficiency (%)
S-G	0.5	198	717	0.222	0.460	22.38
	1.0	58	371	0.202	0.374	17.45
S-P	0.5	360	531	0.342	0.391	32.44
	1.0	109	315	0.236	0.392	19.30

\*Q<sub>tb</sub>: Q at breakthrough time

The best results in terms of total CO<sub>2</sub> adsorption capacity were recorded in the case of S-G with 0.460 mg/kg when a flow rate of 0.5 L/min was applied. However, from an industrial point of view, maximum CO<sub>2</sub> adsorption capacity should be achieved at breakthrough time, at which time the adsorbent begins to saturate. In this sense, S-P recorded the maximum value of all with 0.342 mg/kg for a time of 360 seconds, leading

to an efficiency slightly higher than 32%. Extended breakthrough times result in enhanced CO<sub>2</sub> adsorption (Danish *et al.*, 2021).

Better results associated with a lower gas inlet flow rate were also presented by other authors (Al Mesfer *et al.*, 2020), who recorded breakthrough times for silica gel between 185 seconds and 220 seconds when surface velocities of 0.052 m/s and 0.032 m/s were used. It can be due to lower gas flow turbulence and better gas diffusion through the pores of the adsorbent material (Boonchuay & Worathanakul, 2022).

### 3.3 Adsorption kinetics study

The parameters corresponding to the two kinetic models with the best fits among the different models studied are presented in Table 4. Where  $q_e$  represents the theoretical maximum absorbed amount calculated by the model and  $q_{exp}$  the maximum absorbed quantity experimentally reached.

**Table 4:** Kinetic parameters of granular silica gel (S-G) and powdered silica gel (S-P).

S-G				S-P			
Kinetic model		Q=1 L/min	Q=0.5 L/min	Kinetic model		Q=1 L/min	Q=0.5 L/min
Avrami	$n$	0.815	1.109	Ritchie	$n$	0.938	0.718
	$k_A \left( \frac{L}{mg \cdot h} \right)$	0.005	0.004		$k_n \left( kg^{n-1} \cdot \frac{mg^{1-n}}{min} \right)$	0.007	0.003
	$R^2$	0.986	0.989		$R^2$	0.998	0.985
	$q_e$ (mg/kg)	0.601	0.046		$q_e$ (mg/kg)	0.423	0.416
	$q_{exp}$ (mg/kg)	0.603	0.046		$q_{exp}$ (mg/kg)	0.435	0.429

For S-G, the Avrami model presents a good fit for the 2 flow rates studied, which is corroborated by  $R^2$  values higher than 0.98 in both cases. This means that adsorption is carried out in the active zones of the adsorbent thanks to the forces produced between CO<sub>2</sub> and silica gel. This is an internal diffusion model; therefore, it is taking place in the intraparticle or interparticle zone. Also, a good fit of the model can be observed in the maximum absorbed quantity for both flow rates of 0.5 L/min and gas flow rates of 1 L/min ( $q_e$  values very close to  $q_{exp}$ ). The Avrami model also presented good fits in the study conducted by Doan & Chiang (2023) when silica hybrid aerogel impregnated with polyethyleneimine was used in CO<sub>2</sub> adsorption. The excellent fits of the Avrami's kinetic model may be due to its ability to account for both chemical and physical adsorption of CO<sub>2</sub> (Serna-Guerrero & Sayari, 2010).

In the case of S-P the Ritchie model exhibited a very good fit with the experimental data over the entire adsorption range, especially when the flow rate used in the experiments was 1 L/min, obtaining a correlation coefficient higher than 0.99. These results are in agreement with those obtained by Koshraftar *et al.*, (2021). The results

obtained indicate that this model can be used to simulate the adsorption mechanism and that adsorption is carried out in active zones thanks to the pores of the adsorbent and the physical interactions in the form of forces that it establishes with the adsorbate.

### 3.4 Adsorption diffusion study

The three slopes of the Weber-Morris plot for granular silica gel and powdered silica gel have been calculated and are shown in Table 5. Thus, the overall step governing the kinetics of CO<sub>2</sub> adsorption in the case of silica gel can be deduced from the slope of the linear segments.

**Table 5:** Slopes of the Weber-Morris model curve for granular silica gel (S-G) and powdered silica gel (S-P).

Adsorbent	Flow rate	Curve slope 1 (10 <sup>-3</sup> ) mol/kg·s <sup>0.5</sup>	Curve slope 2 (10 <sup>-3</sup> ) mol/kg·s <sup>0.5</sup>	Curve slope 3 (10 <sup>-3</sup> ) mol/kg·s <sup>0.5</sup>
S-G	1.0 L/min	1.6	15.2	2.6
	0.5 L/min	3.9	30.3	4.5
S-P	1.0 L/min	2.4	16.9	1.7
	0.5 L/min	3.5	10.1	2.6

It can be observed that in the case of S-G the slope corresponding to the initial stage is the smallest. This means that the diffusion of CO<sub>2</sub> through the gas phase to the external surface of the silica gel is the slowest and, consequently, the limiting step. These results agree with those obtained by Loganathan et al., (2014) who have recorded similar behavior for mesoporous silica. Nonetheless, variations in behavior were observed when S-P was used. In this case, the dominant step was the third one, where intraparticle diffusion becomes slower due to the saturation of the active sites of the adsorbent. Differences in adsorption were also recorded by Gatti et al., (2017) when particles of different sizes of amino-functionalized MCM-41 silicas were used on CO<sub>2</sub> adsorption.

## 4. Conclusions

In this study, the potential of silica gel as an effective adsorbent for CO<sub>2</sub> capture was investigated. Adsorption experiments at 1 bar pressure and flow rates of 0.5 L/min and 1 L/min using powdered and granular silica gel were conducted. The results revealed that higher adsorption capacity was achieved when the lower gas flow rate was supplied.

The adsorption kinetics were analysed using kinetic models. Additionally, the dominant adsorption mechanism was studied through the Weber-Morris model. The Avrami kinetic model exhibited the best fit for granular silica gel, while the Ritchie model emerged as the best candidate for powdered silica gel. Moreover, it is found that film diffusion is highly involved in controlling the CO<sub>2</sub> uptake kinetics on granular silica gel.

However, intrapore diffusion, within the active sites, dominates when powdered silica gel is used.

This study demonstrates the versatility and efficacy of silica gel in adsorbing CO<sub>2</sub>, making it a promising candidate for environmental and industrial applications in carbon dioxide capture and sequestration. Future studies will delve deeper into the balance between flow rates and adsorption efficiency. These investigations will aim to refine the adsorption process across different industrial settings, ensuring that the variables are optimized for maximum effectiveness. Moreover, future research should also consider the environmental and economic impacts of CO<sub>2</sub> capture technologies. As the world moves toward more sustainable practices, it is imperative to not only focus on the technical efficiency of adsorption materials but also to assess their feasibility from a broader perspective.

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