CO2 capture on biomass-derived activated carbon: prolonged cyclic operation

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Abstract. Cyclic carbon dioxide CO2 adsorption-desorption experiments were conducted at ~295 K and a maximum pressure of 10 bar using laboratory-synthesized activated carbon pellets derived from almond shells. Over 20 cycles, the CO2 adsorption equilibrium on these CA-Am-Pas pellets demonstrated stability from the 9th cycle onward, with adsorption efficiencies ranging from 1 to 0.8 and desorption yields around 40%. Equilibrium modelling across all cycles consistently showed the Freundlich isotherm as the best fit, achieving regression coefficients very close to 1.

1 Introduction

Rising atmospheric carbon dioxide CO_2 levels from fossil fuel combustion necessitate efficient capture technologies to mitigate climate change and meet net-zero goals. CO_2 capture via solid adsorbents offers a low-energy alternative to solvent processes plagued by corrosion and high costs. Biomass-derived activated carbons, especially from almond shells, excel due to high surface area (>1,500 m^2/g), microporosity, and renewability.[1, 2, 3]

Key properties like narrow pore sizes (~0.4–0.7 nm) and surface chemistry drive capacity and cyclic stability. Precursors such as almond shells, date pits, and coconut husks yield superior CO_2 uptake (up to 5.5 mmol/g) via KOH/CO_2 activation. Recent 2025 studies highlight optimized date-palm AC and enhanced almond shell microporosity (32–36% gain).[1, 2, 4, 5, 6].

The present work is aimed to control the behaviour of a prolonged 20 adsorption – desorption cycles of CO2 at 10 bars under low temperature on almond shell activated carbon pellets chemically activated (CA-Am-Pas). So that desorption yield, adsorption efficiency and cumulated amounts will be examinate during this cyclic operation.

2 Materials and methods

2.1 Preparation of Activated Carbon Pellets

The manufacturing process of activated charcoal adopted in this study consists of three steps: firstly, the raw material is impregnated in a solution of phosphoric acid, then it is transformed

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into pellet form; subsequently, these pellets undergo a high-temperature heat treatment under a nitrogen atmosphere. In this section, we present the main characteristics of the precursor, the pre-treatment of the raw material, a description of the experimental process, and the various techniques used to determine the properties of the final product.

Throughout this study, almond shells precursors are used to produce activated charcoal in pellet form:

The almond shells, used as raw material for the production of activated charcoal, are harvested from almond trees in the Sfax region during the summer. They have a beige color and are in the form of small pieces of variable size, up to 3 to 4 cm.

Table 1. Manufacturing conditions of Almond shells

Raw materials	**Almond shells**
Nomination	CA-Am-Pas
Form	pellets
Manufacturing process	Chemical
Activation agent	H_3PO_4
step1 impregnation (Temperature, time & rate)	110 °C 9 h 1/3
Step2 carbonization (temperature & time)	410 °C 2h30min

2.2 Porosity and surface area

Textural analysis was performed by nitrogen adsorption/desorption at -196 °C over a relative pressure ranging from 9*10-6 to 0.99 P/P0. This characterization was carried out using an automated manometric apparatus

Table 2 .Textural properties

Activated carbon pellets	**SpBET (m^2/g)**	**Vpore (cc/g)**	**Vmicro(cc/g)**	**Vméso(cc/g)**	**D pore (Å)**	**% vmicro**	**%vméso**
CA-Am-P	1184	0,5097	0,389	0,120	21,563	76,341	23,658

This table confirms that about three-quarters of the pore volume is microporous and one-quarter mesoporous,

So that, such a micro/mesoporous balance is advantageous: micropores give high capacity for small adsorbates, while mesopores (and the narrow peak near 2.5–3 nm) facilitate diffusion and access to the microporous network.

3 Experimental setup for volumetric technique

Throughout this work, a detailed analysis of the testing of vegetable-based pellets as adsorbents for carbon dioxide was conducted using a volumetric measurement setup (Figure 1) located at the Research Laboratory of Process Engineering and Industrial Systems (GPSI) at the National School of Engineers of Gabes [7]. This setup consists of two cylindrical cells:

Cell A: This is a stainless steel chamber, referred to as chamber VA and with a volume VA, connected to the feed bottle via valve V-1, serving as a volumetric reservoir to control the amount adsorbed.
Cell B: This is a measuring cell, with a volume VB, with a diameter of 6.3 cm and a height of 11 cm. inside this cell is a stainless steel mesh basket containing the activated carbon bed. The basket has a diameter of 5 cm and a height of 6 cm, and it is attached to a vertical axis that is rotated by an electric motor equipped with a speed reducer.

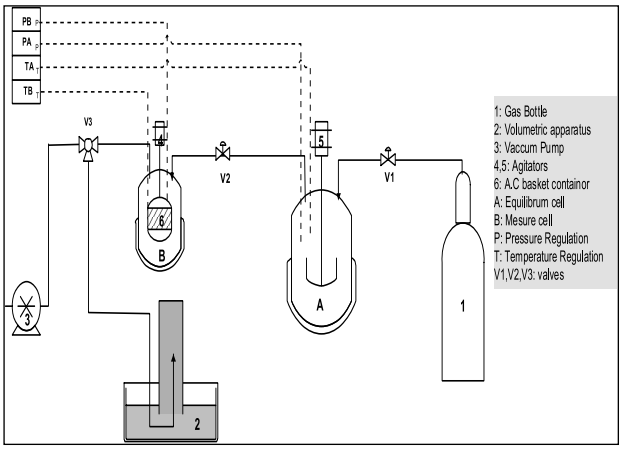

Fig. 1. Exper*i*mental setup

Carbon dioxide is stored on activated carbon pellets under specific operating conditions, which are summarized in the following table:

Table 3. Cyclic operation conditions

AC pellets	Cycle Numbers	T (°C)	Injections Numbers	P (bar)	m_{CA} (g)
CACAm-Pas	20	22	6	10	20

Experimental Protocol and Method for Calculating Adsorbed/Desorbed Quantities:
Before adsorption step, the activated carbon amount has to be degassed under vacuum at high temperature during 2 hours. Adsorption step is consisted of 6 injections, each injection lead to an intermediate equilibrium state with an adsorbed amount na,i.
Desorption step is done after the finish of adsorption one by gradual depression the desorption amount of each depression is nd,i
In desorption curves, nc is the cumulated amount and nrc,i is the residual cumulated amount as shown in Figure 2.

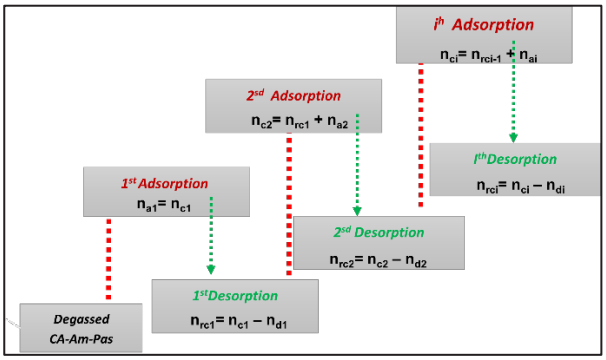

Fig. 2 : adsorption- desorption steps explication

4 Results and Discussion: Cyclic CO_2 capture on activated carbon pellets

4.1 CO_2 adsorption-desorption isotherms

In this section, CO_2 gas adsorption-desorption was performed at a maximum pressure of 10 bar at 22°C.

The curves for several adsorption-desorption cycles: cycle 1, cycle 5 and cycle 18 are grouped together in the following figure.

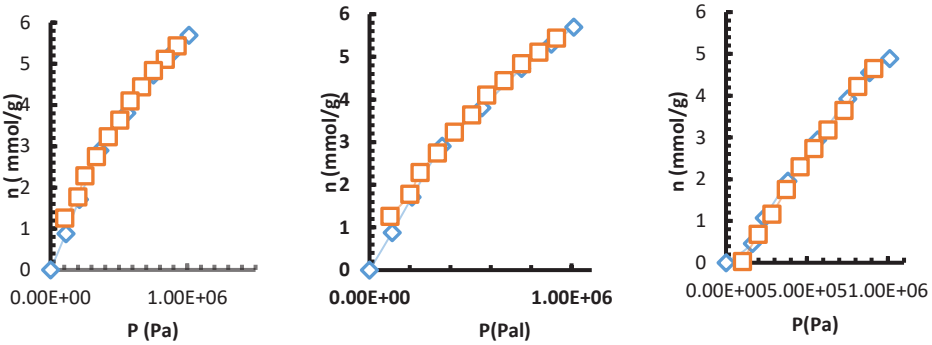

Fig. 3. Adsorption ◊ -desorption □ isotherms for the 1st , 5th and the 18 th cycles under 22°C up to 10 bars

Examination of Figure 3 shows almost total reversibility for the different cycles, with a pattern similar to type I, which must be confirmed at a later stage by modeling the adsorption equilibrium throughout the cyclic operation.

As shown in previous curves (fig.3), it is clear that the phenomenon studied is quasi-reversible, with a very small difference between the two curves, confirming that physical adsorption is very different from chemisorption, with very low hysteresis.

The steep initial slope at low pressure and continuous increase with pressure are typical of CO_2 adsorption on predominantly microporous activated carbons with additional mesopores. [8]

A maximum capacity near 5-5.5 mmol g^{-1} at 22 °C and 1 MPa is consistent with high-surface-area, micropore-rich carbons and indicates that CA-Am-P is well suited for pressure-swing CO_2 capture or storage.

The nearly linear increase in loading at higher pressures implies that further capacity gains are possible at slightly higher pressures, while the relatively narrow hysteresis loop should facilitate regeneration during desorption cycle.

4.2 Evaluation of adsorption-desorption capacity during cyclic adsorption-desorption of carbon dioxide on activated carbon pellets based on almond shells CA-Am-Pas

4.2.1 Adsorption capacities

The adsorption capacity of an adsorbent defines the amount of gas that the material can adsorb under average pressure and temperature conditions. The most effective material is the one with the highest maximum adsorption capacity.

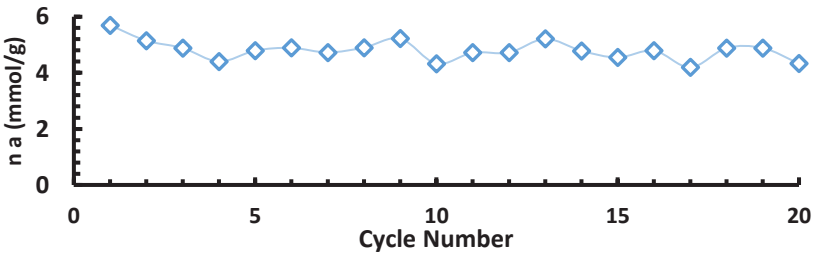

Fig. 4**:** CO_2 adsorption capacity on CA-Am-Pas undergoing cyclic testing.

High reproducibility was observed throughout the cyclic operation (fig4), with average adsorption around 4.8-5 mmol/g.

4.2.2 Efficiencies throughout the cyclic operation performed on activated carbon pellets made from almond shells

The adsorption efficiency of activated carbon is defined as the ratio of the quantity adsorbed in the i-th cycle to the quantity initially adsorbed by the virgin activated carbon in the first cycle.

$$e = \frac{n_{ai}}{n_{a1}} \quad (1)$$

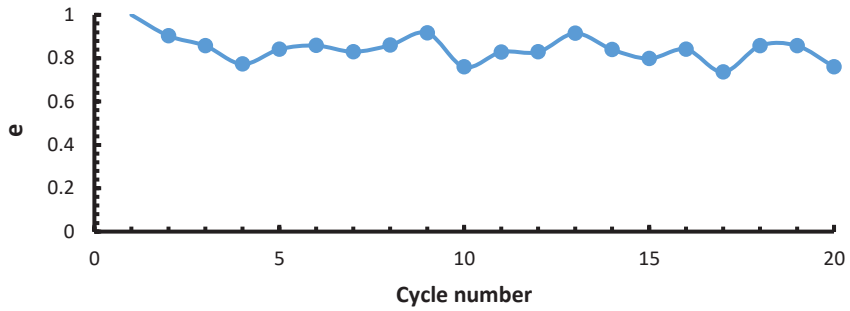

Fig.5 : Adsorption efficiencies throughout the cyclic adsorption-desorption process on activated carbon pellets made from almond shells.

Throughout the cyclic adsorption-desorption process, high efficiency is recorded, varying between 1 and 0.8, which proves the high quality of the activated carbon pellets and the almost total reversibility of the phenomenon.

4.2.3 Desorption efficiency throughout the cyclic operation performed on activated carbon pellets made from almond shells

The desorption efficiency of the ith cycle is defined as the ratio of the quantity desorbed in the ith cycle to the sum of the quantity adsorbed in the ith cycle na,i and the cumulative quantity from the previous cycle (i-1) nrc,i-1

$$\alpha, i\ (\%) = \frac{n_{d,i}}{n_{a,i}+n_{rc,i-1}} * 100 \qquad (2)$$

Where:
$n_{a,i}$: adsorbed amount per gram of Activated carbon (mmol/g) for the ith cycle.
$n_{d,i}$: desorbed amount per gram of Activated carbon (mmol/g) for the ith cycle.
$n_{rc,i-1}$: residual cumulated amount (mmol/g) for the ith cycle as explained in Figure 2.

Figure 6 shows the desorption efficiency of activated carbon pellets throughout the 20 cycles studied:

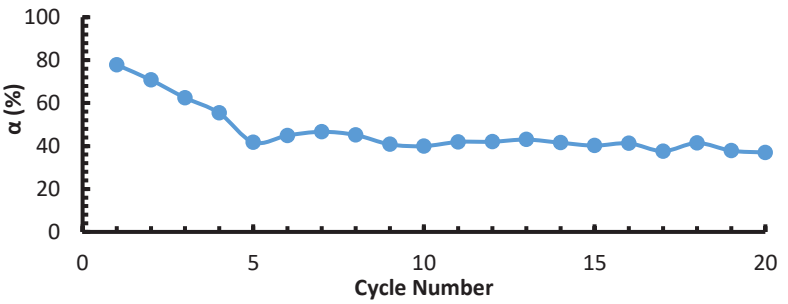

Fig. 6. Desorption yield throughout the cyclic adsorption-desorption process on activated carbon pellets made from almond shells.

This desorption efficiency reaches reproducible stability from the 9th cycle onwards, when the CA-Am-Pas pellets begin to exhibit stable and consistent behavior in desorbing the same amount of gas.

4.3 Modeling of CO2 adsorption-desorption isotherms on CA-Am-P

In this section, three theoretical models Langmuir, BET and Freundlich of adsorption-desorption equilibrium were tested for the 20 adsorption isotherms obtained. Figure 7 summarizes the results of the regression factor R^2 obtained.

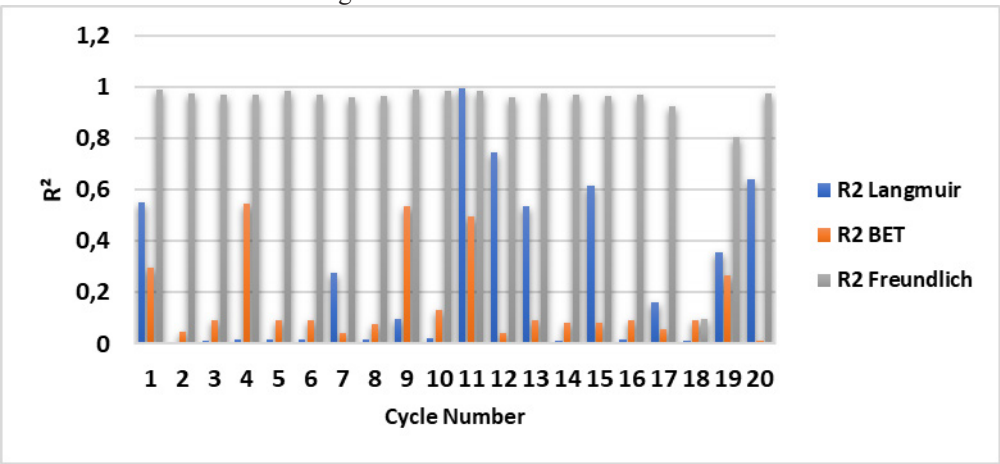

Fig. 7. Regression Factor for cyclic operation for the three models

Thus, the Freundlich model is the most appropriate model for these adsorption equilibria, confirming that these isotherms are type I and that there are no interactions between the adsorbed molecules and that each block of molecules is adsorbed at a single adsorption site. This model is adequate for all 20 cycles without exception and without modification at the beginning or end of this cyclic operation.[9]

5 Conclusion

This work has brought together a whole series of successive experiments on the same sample of activated carbon pellets made from almond shells under a maximum pressure of 10 bar relative and at T = 22°C.

This operation showed stability from the 9th cycle onwards, where the cumulative quantity and desorption stabilized at an efficiency of α = 40%, an adsorption capacity of around 5 mmol/g, and a desorption capacity of around 4.2 mmol/g.

Modeling of the adsorption equilibrium confirmed the suitability of the Freundlich model from the 1st to the 20th cycle, confirming type I isotherms and the physisorption of CO_2 molecules in CA-Am-Pas pellets.

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