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Utilisation of microwave energy for CO_2 desorption in post-combustion carbon capture using solid sorbents

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Abstract

One of the most promising technologies for CO_2 capture is adsorption using solid sorbents, with the most important advantage being the energy penalty reduction during capture and regeneration of the material compared to liquid absorption. Nevertheless, the challenge in this application remains the same, namely to intensify the production of a high purity CO_2 stream in terms of adsorption/desorption rates and energy use while preserving the textural characteristics of the sorbents. Towards this target, a new adsorption system is proposed in this research, namely Microwave Swing Adsorption (MSA), which takes advantage of the different heating procedure that electromagnetic energy can provide. Results showed that microwaves are able to enhance the rate of CO_2 desorption from the sorbent, contributing to a four times faster overall desorption process, compared to conventional heating desorption. It was also proved that the solid sorbent's CO_2 adsorption capacity is also well preserved, without any significant reduction even after 20 adsorption/desorption cycles.

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1. Introduction

Carbon dioxide capture and storage (CCS) has been recognised as one of the most important technologies towards climate change mitigation, mostly due to the fact that coal is still the dominant feedstock for energy production [1]. In coal post-combustion capture technology, CO_2 is separated from a diluted flue gas stream that contains typically around 12-15% CO_2, at ambient pressure and at temperatures of 40-100°C [2].

CO_2 capture process represents typically about 70% of the total cost of the CCS chain, and therefore, novel adsorption technologies that can offer various advantages over conventional absorption using alkanolamines, such as high operating flexibility and low maintenance costs, are gaining support nowadays [3]. However, efficient regeneration systems ensuring multiple re-use of adsorbent materials, while minimising energy consumption, are required.

Two desorption methods are typically applied in laboratory and industrial applications, namely Pressure Swing Adsorption (PSA) and Temperature Swing Adsorption (TSA). However, PSA and TSA are not considered to be the optimum pathways for CO_2 adsorption, as discussed in the authors' published work [4].

This study presents and analyses a relatively new approach for CO_2 capture, namely Microwave Swing Adsorption (MSA) technology, a process that can decrease the overall energy penalty by faster and more efficient heating of the sorbents. Another significant advantage of the MSA is that microwaves are able to preserve the textural characteristics of the sorbent material after heating, leading to re-use of the material for many cycles, contributing to the overall techno-economic feasibility of CO_2 capture.

The effects of microwave regeneration of sorbent materials can vary depending on various process parameters, such as applied power, mass of saturated adsorbent, purge gas flow rate, and irradiation. In this work, experimental studies of fixed bed CO_2 adsorption on microporous granular activated carbon (AC) with regeneration via MSA and TSA, investigating four different desorption temperatures, have been conducted.

Nomenclature

CH_{70}	conventional heating at 70°C
MW_{70}	microwave heating at 70°C
CH_{90}	conventional heating at 90°C
MW_{90}	microwave heating at 90°C
CH_{110}	conventional heating at 110°C
MW_{110}	microwave heating at 110°C
CH_{130}	conventional heating at 130°C
MW_{130}	microwave heating at 130°C
$[CO_2]$	CO_2 concentration
μ	location parameter of the log-normal distribution
σ	scale parameter of the log-normal distribution
t_h	time needed for the AC bed to reach the desired temperature
Q_{des}	amount desorbed,
t_{50}	time needed for 50% of q_{des},
t_{90}	reverse breakthrough time
dq/dt_{av}	average desorption rate,
t_{tot}	total desorption time ($t_{tot} = t_h+t_{90}$)

2. Experimental section

2.1. Sorbent characterization

A commercial AC provided by NORIT (product name: NORIT GCN 3070) was used. This particular AC has been produced from coconut shell using steam physical activation. The particle size of the AC measured by the supplier was 210-595μm (30-70 mesh, 93%). N_2 isotherm adsorption and pore size tests were conducted using Gemini VII 2390, provided by Micromeritics, calculating a surface area of 1514 m²/g, while the pore diameter was 1.2 nm, as seen in Figures 1a and 1b.

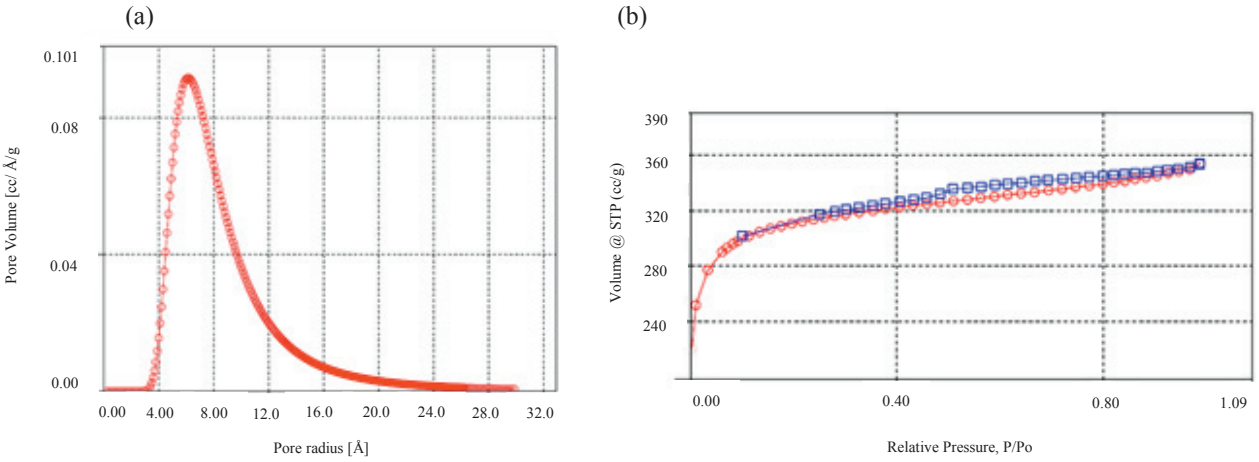

Fig. 1. (a) pore radius (Å) of the granular AC; (b) N_2 adsorption isotherm of the granular AC

2.2. Experimental setup

Figure 2 schematically shows the lab-scale experimental setup used during TSA and MSA tests, as discussed in the authors' prior published work [4].

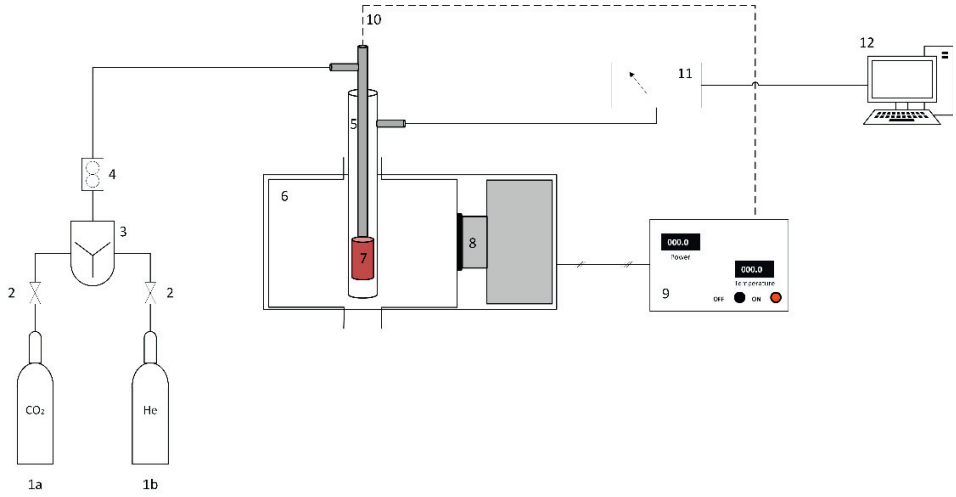

Fig. 2. Layout of the experimental rig, 1a. Gas cylinder – CO2, 1b. Gas cylinder – He, 2. Valve, 3. Gas mixer, 4. Mass flow controller, 5. Reactor column, 6. Microwave cavity, 7. AC packed bed, 8. Magnetron, 9. PID temperature controller, 10. IR Pyrometer, 11. Mass Spectrometer (MS), 12. PC

The temperature of the AC packed bed was controlled by an InfraRed (IR) pyrometer (Optris CTfast CT LT 25F) which measured the AC bed temperature from the top of the reactor through a zinc selenide (ZnSe) window (diameter = 20 mm, thickness = 3 mm), as depicted in Figure 2. For the MSA experiments, the IR pyrometer was also connected to a PID controller (Eurotherm 3504), which was able to control the microwave power transmitted from the magnetron to reach the specific target temperature. TSA experiments were conducted using a commercial tubular furnace.

CO_2 and He concentrations were continuously measured by a mass spectrometer provided by Hiden analytical (HPR-20 QIC) and the data were acquired and analysed using Hiden QGA Pro software.

2.3. Experimental procedure

Two different categories of experiments were conducted using the above setup, namely CO_2 desorption via TSA and MSA and AC recycling tests.

For the purposes of the CO_2 desorption tests, 5g of AC were placed inside the fixed bed reactor that were firstly purged using a 20 ml/min flow of He for 30 minutes. Next, CO_2 adsorption took place, separating it from a 40 ml/min He/CO_2 (50:50) gas stream at room temperature and atmospheric pressure until a plateau in the breakthrough curve was observed. Then, CO_2 desorption experiments with the same 40 ml/min He/CO_2 (50:50) gas stream at 70, 90, 110, 130°C under microwave or conventional heating were conducted. When desorption was completed, the reactor was cooled down with a 20 ml/min flow of He for 30 minutes and the AC bed was replaced with new material.

For the AC recycling tests, the procedure was almost similar, with the main difference being that 10g of AC were used and were not replaced after each adsorption/desorption cycle, and only one desorption temperature (70°C) was tested. Samples of 2g after 10 and 20 cycles were obtained and evaluated by thermogravimetry (TG) using a thermogravimetric analyzer TGA500.

For easier comparison of the experimental data, the expression reverse breakthrough time (t_{90}) was introduced and was defined as the time needed to achieve 90% desorption after reaching the desired temperature. The amount desorbed (Q_{des}) was calculated by equation 1:

$$Q_{des} = \int_0^{t_{90}} Q(t)dt \qquad (1)$$

3. Results and discussion

3.1. CO_2 desorption via TSA and MSA

Figures 3a-3d represent the time-dependent CO_2 outlet concentration profiles during CO_2 desorption by conventional heating (CH) and by microwave heating (MW) using four different temperatures - 70°C, 90°C, 110°C and 130°C. It is clear that there are two different outlet concentration patterns, both of which follow the log-normal distribution. The following formula can be applied to these profiles:

$$[CO_2] = \frac{1}{\sigma t\sqrt{2\pi}}\, e^{-(lnt-\mu)^2/2\sigma^2} \qquad (2)$$

where for TSA σ=1, μ=0 and for MSA σ=0.25, μ=0.

This distribution suggests that the maximum [CO_2] is reached relatively fast, meaning that most of the adsorbed CO_2 is quickly desorbed, due to the rapid diffusion of the gas in the fine porous structure of the AC [3].

One notable difference is that the maximum desorption (curve peaks) and the duration of the process are different for both temperatures, depending on the desorption method followed; in the case of MW the peak is reached clearly faster, approaching ~55% of CO_2 in less than 30 seconds for all four desorption temperatures. On the other hand, in the case of CH, the peak only reaches a maximum of 20%-30% of CO_2 after 2 minutes of heating the AC bed.

Another clear difference between the two patterns is the long tail that can be observed in the CH process; suggesting that the residual CO_2 desorption takes place slowly while, in the case of MW, the curve reaches an asymptote more

rapidly, indicating that almost all the CO_2 is desorbed immediately after irradiating the AC bed with electromagnetic energy.

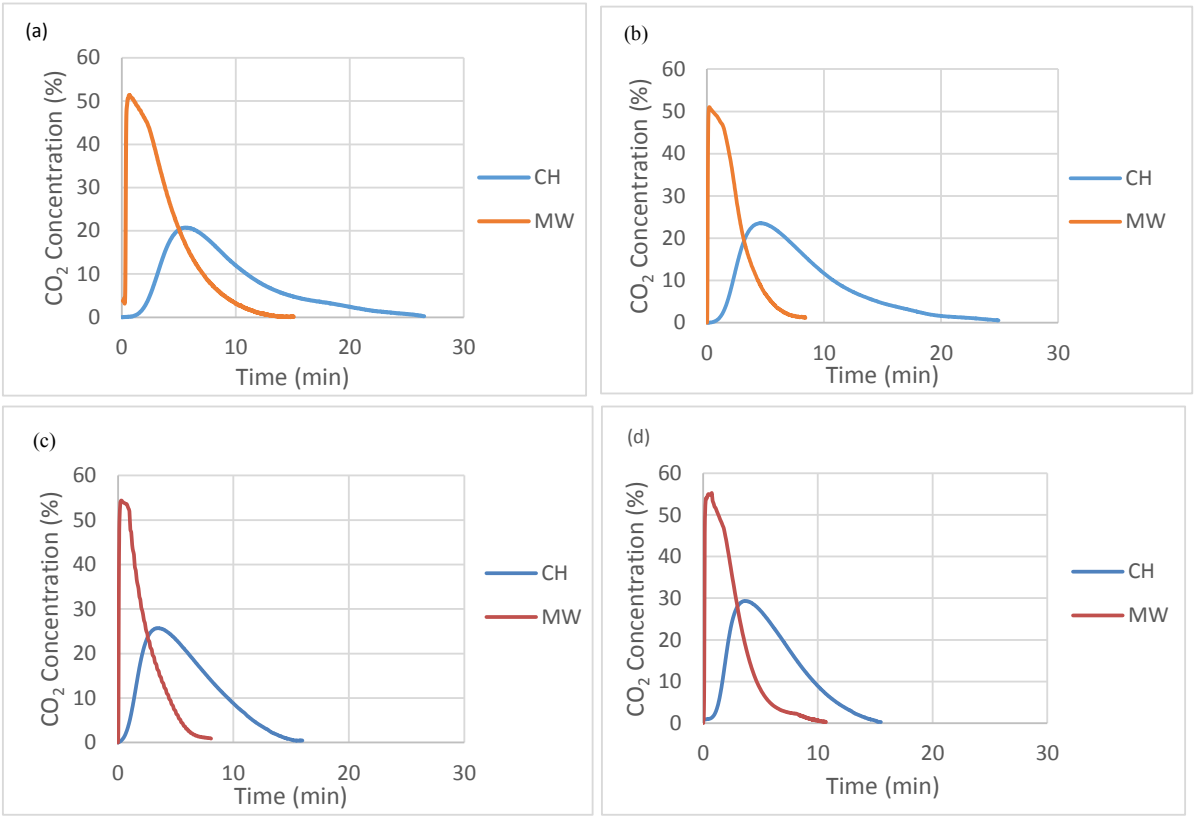

Fig. 3. Microwave and conventional heating CO_2 desorption at (a) 70°C; (b) 90°C; (c) 110°C; (d) 130°C

Table 1 summarises the experimental data collected during the comparison of TSA and MSA. One important observation is that for both CH and MW, as the desorption temperature rises, the time needed for the process to be completed decreases gradually. However, in the case of CH the time varies from 10 to 16 minutes for the four different temperatures, while in the case of MW only 4.5 to 7 minutes are needed for the completion of the desorption step. Moreover, there is also a clear decrease in % of pure CO_2 (Q_{des}) obtained after heating the AC bed. The aforementioned facts imply that there is an optimum desorption temperature between 70°C and 130°C, that combines fast desorption rates and sufficient generation of pure CO_2. Further research will be conducted towards this direction in the future.

Table 1: Main parameters obtained from AC regeneration experiments under conventional and microwave heating

	t_h (min)	Q_{des} (ml)	t_{50} (min)	t_{90} (min)	dq/dt_{av} (ml/min)	t_{tot} (min)
CH_{70}	6	28.2	8	16.5	1.54	22.5
MW_{70}	1.5	31.85	2.5	7	4.61	8.5
CH_{90}	7.8	26.92	7	14.8	1.9	22.6
MW_{90}	2.1	27.84	2.2	5.7	4.88	7.8
CH_{110}	11.4	26.37	5.4	11	2.35	22.4
MW_{110}	3.5	24.48	2	5.1	4.74	8.6
CH_{130}	15	25.85	5.5	10	2.66	25
MW_{130}	5.5	24.2	1.5	4.5	5.09	10

3.2. AC recycling tests

One very important parameter for the feasibility of the CO_2 adsorption technology is the re-utilisation of the sorbent material for various adsorption/desorption cycles. The ease of regeneration as well as the preservation of the material will help to reduce the cost of capture. The sorbent must demonstrate microstructure and morphological stability and retain the CO_2 capture capacity after multi-cycling between adsorption and regeneration steps. As a result, AC recycling tests were conducted in order to observe how the CO_2 adsorption capacity of the sorbent is affected after multiple - 10 and 20 - microwave heating cycles, as described in section 2.3.

The CO_2 adsorption capacity of the original and the regenerated AC were evaluated by thermogravimetry. Thermogravimetric analyses at 35°C isothermal conditions demonstrated that the CO_2 adsorption capacity of this AC is kept almost constant with a very slight decrease in the samples subjected to microwave cycles; the CO_2 uptake of the original AC was 8.55%, falling to 8.46% after 10 cycles, declining to 8.32% after 20 cycles. This means that, a decrease of only 0.23% was reported after 20 consecutive cycles of MSA, which is a very promising fact for the economics of the overall CO_2 adsorption process.

4. Conclusions

In this work, a microporous granular activated carbon (AC) was used to study CO_2 desorption following TSA and MSA procedures. Four desorption temperatures (70, 90, 110 and 130°C) as well as the material stability after microwave heating were investigated. Experimental results showed that even though both heating processes follow the log-normal distribution, there is a clear distinction between them; MW is responsible for faster CO_2 desorption rates, without the drawback of a slow desorption of the residual CO_2, compared to CH. Moreover it is possible to heat the AC bed 3 to 4 times faster when using MW. As a result, MW offers the possibility of a more accelerated overall CO_2 desorption process when using AC as adsorbent by increasing desorption rates significantly. Furthermore, it was proved that microwave irradiation did not affect the sorbent's morphology significantly, as CO_2 adsorption capacity decreased only 0.23% after 20 consecutive MSA cycles.

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