EFFECT OF OPERATING CONDITIONS ON CO_2 ADSORPTION IN A SOUND ASSISTED FLUIDIZED BED

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Abstract
The present work is focused on the CO_2 capture by sound-assisted fluidization of fine activated particles with high specific surface particles (1060m^2/g). The powder fluidization quality has been preliminarily characterized by performing ordinary and sound assisted fluidization tests. Then, CO_2 adsorption tests have been performed pointing out the effect of the sound intensities (125 to 140dB) and frequencies (20 to 300Hz) and the fluidization velocity (0.1 to 2cm/s). In particular, sound intensities higher or equal to 125dB and frequency in the range 50-120Hz provide the best performances. Under sound assisted the breakthrough time undergoes an exponential increase by decreasing the fluidization velocity.

Introduction

Among all the approaches for CCS, post-combustion capture provides a means for near-term CO_2 capture from new and existing stationary power plants [1]. In this respect, adsorption is very promising, having the potential, in terms of energy saving, to substitute the present absorption technology. However, enhancement of solid sorbents efficiency is necessary [2]. In this respect, nanometric and micronic materials can be very easily functionalized on the surface by means of the introduction of functional groups with great affinity towards CO_2 molecules. Among gas-solid contact technologies, fluidization is one of the best available techniques to process large quantities of powders.

Nevertheless, fluidization of fine materials is particularly difficult due to cohesive forces existing between particles [3]. The use of sound assisted fluidization has been indicated to improve fluidization quality of fine powders [4,5]. In the present work CO_2 adsorption by sound assisted fluidized beds of fine activated carbon particles (mean size 0.49μm) has been investigated. Adsorption tests have been performed at ambient temperature and pressure in a laboratory scale reactor in ordinary conditions and under the effect of different acoustic fields (sound intensities, SPL, 125-140dB and sound frequencies, f, 20-300Hz). In particular, effectiveness of CO_2 adsorption has been assessed in terms of the moles of CO_2 adsorbed per unit mass of adsorbent, the breakthrough time and the fraction of bed utilized at breakpoint. Then, the effects of sound parameters (frequency and SPL) and superficial gas velocity, u, (0.1-2cm/s) have been investigated.

Experimental apparatus

CO_2 adsorption experiments have been carried out in a laboratory scale sound assisted fluidized bed apparatus (Fig. 1). The fluidized bed consists of a Plexiglas column of 40mm ID and 1000mm high, equipped with a porous plate gas distributor located at the bottom of the column. N_2 and CO_2 flowrates have been set by means of mass flow controllers (Bronkhorst), and subsequently mixed before entering the bed. Details about the sound-generation system are reported elsewhere [4,5]. The CO_2 concentration in the inlet and outlet gas streams has been measured by an ABB infrared gas analyzer (AO2020).

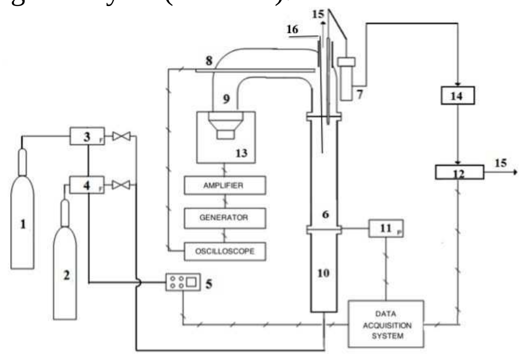

Figure 1. Experimental apparatus: (1) nitrogen cylinder; (2) CO_2 cylinder (3) N_2 flow meter; (4) CO_2 flow meter; (5) controller; (6) 40mm ID fluidization column; (7) filter; (8) microphone; (9) sound guide; (10) wind-box; (11) pressure transducer; (12) CO_2 analyzer; (13) loudspeaker; (14) pump; (15) stack.

An activated carbon DARCO FGD (Norit) has been used as adsorbent material. Its particles size distribution has been characterized using a laser granulometer (Master-sizer 2000 Malvern Instruments). The Sauter diameter is 0.39 and 2μm with and without ultrasound, respectively. Superficial area measurements have been carried out according to the BET method using N_2 at 77K with a QUANTACHROM 1-C analyzer. The activated carbon is characterized by a pore size ranging from the mesoporous (2nm $< d <$ 50nm) to the microporous ($d <$ 2nm) and by a relatively large surface area (1060m^2/g).

All adsorption tests have been carried out at ambient temperature and pressure. The sorbent material has been pre-treated at 393K, in order to remove any trace of moisture. In a typical experiment, the sorbent (110g) is loaded in the column in order to obtain a bed height of 15cm. Then, in a pre-conditioning step of about 10min, N_2 is fluxed in the column in order to stabilize a fluidization regime at fixed superficial gas velocity and sound parameters. This is followed by the adsorption step in which a CO_2/N_2 gas mixture is fed through the column. The CO_2 composition in the effluent gas is continuously monitored as a function of time (breakthrough curve) until the bed saturation. Table 1 reports the operating conditions selected for the adsorption experiments carried out in this work.

Table 1. Operating conditions of the adsorption tests.

Fluidization velocity, cm/s	0.1, 0.25, 0.5, 0.75, 1, 1.5, 2
CO_2 inlet concentration, %vol.	10
SPL, dB	120, 130, 135, 140
Frequency, Hz	20, 50, 80, 120, 300

Results and discussion

Fluidization tests. In Fig. 2 the pressure drop (a and c) and bed expansion (b and d) curves obtained in ordinary and sound assisted conditions are reported. The fluidization quality in ordinary conditions (Fig. 2a-b) is particularly poor (channeling), as clearly confirmed by the fact that asymptotic value reached by the pressure drops is lower than 1. Therefore, the application of the sound (Fig. 2c-d) is required to achieve a proper fluidization regime, which is closely related to an efficient break-up of the large aggregates yielded by cohesive forces into smaller structures easily to be fluidized [4,5]. In particular, an in-depth study has been carried out in order to evaluate the most effective acoustic conditions, namely whether it is possible or not to find optimal values of SPL and frequency. Figure 2e reports the effect of SPL (at fixed frequency, 80Hz) and frequency (at fixed SPL, 140dB) on the fluidization quality in terms of minimum fluidization velocity, u_{mf}.

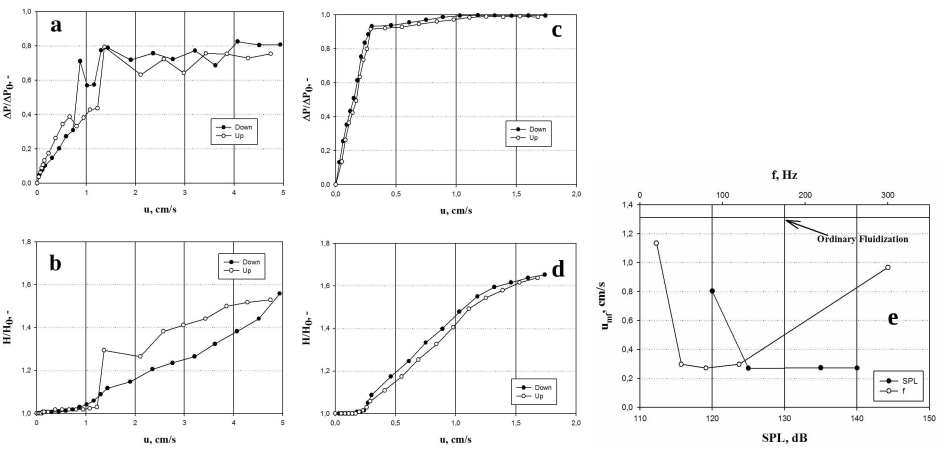

Figure 2. Ordinary pressure drops (a) and expansion (b) curves; sound assisted pressure drops (c) and expansion (d) curves (140dB-80Hz); d) effect of SPL and f on u_{mf}.

As regards the effect of the SPL, sound intensities higher than or equal to 125dB are enough to obtain a good fluidization quality, which means that any additional increase of sound intensity does not succeed in further enhancing the fluidization quality. As regards the sound frequency, the results reported in Fig. 2e show that it has a not monotone effect on the fluidization quality. Actually, it is possible to find an optimum range of frequency (50-120Hz) giving the best fluidization quality. Either too low or too high frequencies, which fall out of this range (20, 300Hz), correspond to worse fluidization qualities.

Adsorption tests. Figure 3a reports the typical breakthrough curves (i.e. C/C_0 versus time, C and C_0 being the CO_2 concentration in the effluent and feed stream, respectively) obtained in ordinary and sound assisted conditions (140dB-80Hz), with C_0=10% and u=1.5cm/s. These curves have been worked out to evaluate: i) the moles of CO_2 adsorbed per unit mass of adsorbent, n_{ads}, integrating the breakthrough curves; ii) the breakthrough time, t_b, or breakpoint, which is the time it takes for CO_2 to be detected at the outlet (5% of the inlet concentration); iii) the fraction of bed utilized at breakpoint, W, namely the ratio between the CO_2 adsorbed until the breakpoint and that adsorbed until saturation. In order to highlight the section before and soon after t_b, the same graph has also been reported in logarithmic scale (Fig. 3b). The analysis of the curves suggests that the application of the sound greatly increases the breakthrough time, which, in sound assisted tests (63s) is more than four times the value obtained in ordinary conditions (12s). The application of the sound affects also the global adsorption capacity: n_{ads}, moves from 0.31mol/kg, in ordinary conditions, to 0.37mol/kg, in sound assisted conditions. The fraction of bed utilized at break point (W) is also greatly enhanced by sound, moving from values lower than 3%, in the tests performed in ordinary conditions, up to values higher than 10%, in the sound assisted tests. The beneficial effect shown by the sound is probably due to the enhancement of the fluidization quality (better gas-solid contact and mass transfer coefficients) with respect to the tests performed in ordinary conditions. In particular, the application of the sound greatly enhances the break-up mechanism and re-aggregation of fluidizing aggregates [4,5], thus constantly renewing and making more readily available the exposed surface.

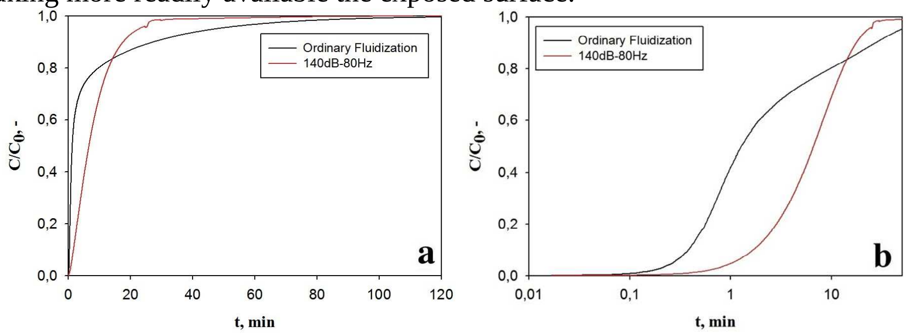

Figure 3. Breakthrough curves in ordinary and sound assisted tests, (a) linear and (b) logarithmic scale. u=1.5cm/s; C_0=10%.

The effect of SPL on CO_2 adsorption efficiency has been evaluated by carrying out tests at fixed frequency (80Hz) and different sound intensities (120-140dB). The comparison in terms of n_{ads}, t_b and W are reported in Fig. 4a. The adsorption process undergoes a significant enhancement only when SPLs higher or equal to 125dB are applied. Indeed, 125dB is a sort of threshold intensity beyond which any further increase of SPL is ineffective. Whereas, the behaviour observed at 120dB is intermediate. Then, in order to point out the effect of sound frequency, tests have been performed at fixed SPL (140dB) and varying the sound frequency (from 20 to 300Hz) (Fig. 4b). The best results in terms of CO_2 adsorption efficiency can be achieved when sound frequencies falling in the range 50-120Hz are applied. Whereas, the adsorption tests carried out at 20 and 300Hz are remarkably worse.

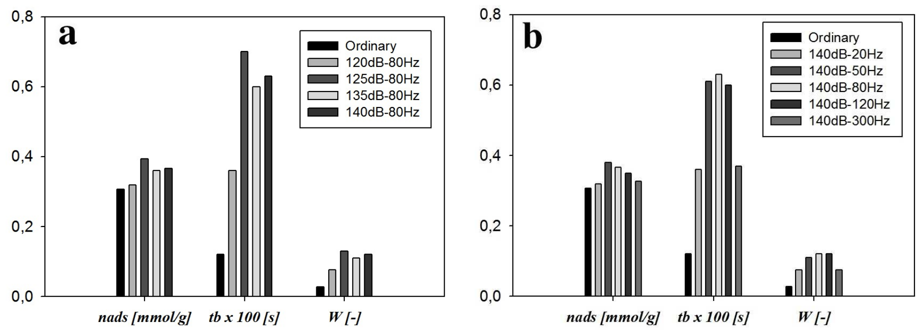

Figure 4. Effect of SPL (a) and frequency (b) on CO_2 adsorption. u=1.5cm/s; C_0=10%.

The effect of the fluidization velocity on the adsorption process has been pointed out by performing adsorption tests at different superficial gas velocities. In particular, the dependence of t_b on the contact time, t_c, defined as the ratio between the mass of adsorbent and the CO_2 volumetric flow, has been highlighted. The curves obtained are shown in Fig. 5. The dependence of t_b on t_c, i.e. the fluidization velocity, is linear, as one could expect, only for the tests performed in ordinary conditions. Whereas, t_b is found to exponentially increase with t_c, namely decreasing u from 2 to 1cm/s, for the sound assisted tests. This evidence is likely due to the fact that the decrease of the fluidization velocity results in a more homogeneous fluidization regime, which is characterized by a lower by-pass of gas through the bed with respect to the tests performed at higher fluidization velocity. On the contrary, in ordinary conditions the system is quite insensible to changes of fluidization velocity, being the fluidization quality always very poor: the observed linear increase of t_b with the decrease of u is only due the CO_2 taking more time to flow through the bed.

Further tests have been performed at superficial gas velocities lower than the minimum fluidization velocities (about 0.3cm/s in sound assisted and 1.3cm/s in ordinary conditions) in order to fully investigate the effect of u on t_b. The analysis of Fig. 5 suggests that in ordinary conditions t_b keeps linearly increasing even for u lower than 1cm/s.

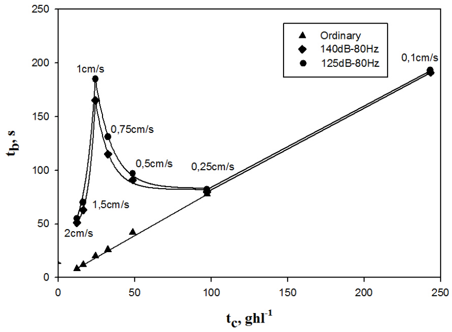

Figure 5. t_b as functions of t_c for ordinary and sound assisted tests, C_0=10%.

However, the trend obtained for the sound assisted tests is not monotone. In particular, a further increase of t_c (u passing from 1cm/s down to 0.25cm/s) results in an exponential decrease of the t_b; whereas, a linear increase of t_b is obtained by finally decreasing the fluidization velocity down to 0.1cm/s, similarly to that obtained in ordinary tests. The first exponential decreasing trend of t_b is due to the worsening of the fluidization quality as a result of the further decrease of the fluidization velocity. On the other hand, the linear increase of t_b, is due to the fact that the bed is not actually fluidized in the tests performed at the lowest velocities (0.25 and 0.1cm/s), the bed behaviour being qualitatively very similar to that of the ordinary tests.

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References

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