# CARBON DIOXIDE ADSORPTION IN A SOUND-ASSISTED FLUIDIZED BED OF FINE POWDERS

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**Abstract**
An experimental assessment of the possibility to utilize sound-assisted fluidized beds to promote $CO_2$ capture, in post- and pre-combustion situations, by means of physical adsorption on fine porous solid particles is presented. The utilization of sound-assisted fluidized beds of activated carbon resulted to have a number of advantages with respect to the same process carried out in fixed bed or conventional fluidized bed systems. In particular, the main process performance parameters, the capture capacity and the breakthrough time were both remarkably improved moving from fixed bed condition to that of fluidized bed and, above all, to that of sound-assisted fluidized bed.

## Introduction

Several studies of the past three decades indicated the growing level of greenhouse gases (GHGs) in atmosphere as the main cause of global warming. Among all the GHGs, carbon dioxide is the largest contributor to the global warming effects, for about 60%. It is then generally recognized that massive reductions in $CO_2$ emissions must be achieved in order to avoid permanent damage to the environment [1].
There are three options to reduce total $CO_2$ emission into the atmosphere [2]:

i. to reduce energy consumptions, mainly by means of a more efficient utilization of energy;
ii. to reduce carbon intensity, by switching to non-fossil energy sources such as renewable energy and hydrogen-based systems;
iii. to enhance the sequestration of $CO_2$, by developing reliable and economical sustainable technologies to capture and sequester $CO_2$.

Carbon capture and storage (CCS) is a group of technologies that may reduce emissions into the atmosphere from large fixed industrial sources. The stage of $CO_2$ capture, in particular, is the most costly part of a CCS process (it has been estimated that it implies between half and two thirds of the total CCS cost) and also that entailing the largest (and significant) amount of utility consumption [3].
This promoted a worldwide research effort on new techniques able to reduce the economic and energetic costs, while achieving significant levels of $CO_2$ capture. It

is possible to distinguish between two different typologies of carbon dioxide capture processes in (large) power plants (Fig. 1):

- post-combustion capture, which is the situation typical of existing power plants, where $CO_2$ needs to be removed from a diluted (<15% by volume) flue gas stream.
- pre-combustion capture, which is instead typical of alternative power plant designs that include integrated gasification combined cycle (IGCC) plants, where the $CO_2$ can be selectively separated from the shifted-syngas ($CO_2/H_2$) prior to the generation of electricity and where the gas stream is under pressure and contains a high concentration of $CO_2$.

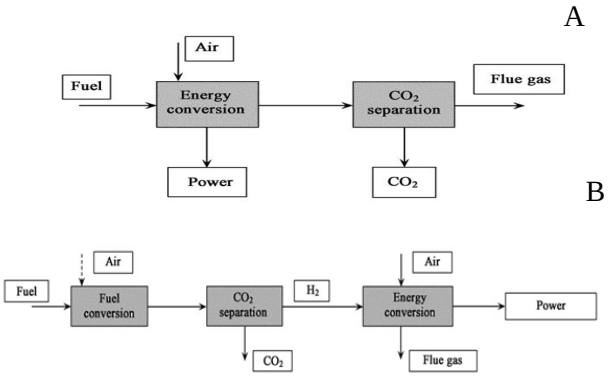


**Figure 1.** Simplified flow diagrams of a post-combustion (A) and a pre-combustion (B) capture process (redrawn from [2]).

Adsorption using solid sorbents is a promising alternative technology that so far has only been utilized with fixed bed reactor and appears to offer potential energy savings together with lower capital and operating costs [4]. Different types of solid sorbents are currently under study to separate $CO_2$ from energy-generating coal-fired power plants, such as zeolites, activated carbons, calcium oxides, supported amines and metal-organic framework (MOF) materials. An additional viable route is the chemical modification of the adsorbent materials by the introduction of functional groups with a great chemical affinity towards $CO_2$. In this respect, ultra-fine particles are particularly suitable to be easily functionalized [5]. Then, in order to use such materials without being previously pelletized, the development of a proper processing technology is needed. Sound assisted fluidization has been indicated as a valid technological option to smoothly fluidize fine powders.

This paper is an assessment of the possibility to utilize sound-assisted fluidized beds to promote $CO_2$ capture, in post- and pre-combustion situations, by means of physical adsorption on fine porous solid particles of activated carbon and zeolites. Firstly, the materials have been fluid-dynamically characterized by performing ordinary and sound assisted fluidization tests. Then, $CO_2$ adsorption tests have been

performed varying the $CO_2$ content in the gas stream, the gas velocity and the sound intensity and frequency.

**Experimental activity**

The fluidization curves and $CO_2$ adsorption capacity have been obtained by using a laboratory scale fluidization column, schematized in Fig. 2. The fluidized bed consists of a Plexiglas column of 40mm ID and 500mm high, with a porous plate gas distributor located at the bottom of the column.

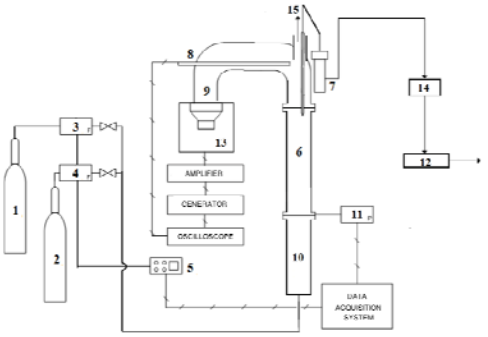


**Figure 2.** Experimental apparatus: (1) nitrogen cylinder; (3) $CO_2$ cylinder (3) $N_2$ flow meter; (4) $CO_2$ flow meter; (5) controller; (6) 40mm ID fluidization column; (7) filter; (8) microphone; (9) sound guide; (10) wind-box; (11) pressure transducer; (12) $CO_2$ analyzer; (13) loudspeaker;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

The temperature of the bed has been monitored by means of a thermocouple placed at the center of the bed. The bed pressure has been controlled by means of a pressure transducer installed at 5mm above the gas distributor. An infrared gas analyzer has measured the $CO_2$ composition of the outlet gas stream. The sound-generation system is made of a digital signal generator whose signal is amplified by means of a power audio amplifier. The signal is then sent to a loudspeaker. All fluidization and adsorption tests have been carried out at ambient temperature and pressure. H-ZSM-5 Zeolite (Zeolite Int.) and activated carbon DARCO FGD (Norit) have been used as adsorption materials. Both the powders have been characterized by a laser granulometer (Master-sizer 2000 Malvern Instruments). The properties of the powders are reported in Table 1.

**Table 1.** Properties of the adsorbent materials.

| Materials | $D_{SAUTER}$, μm | BET surface area, $m^2/g$ | Bulk density, $kg/m^3$ |
|---|---|---|---|
| **Activated Carbon** | 0.39 | 1060 | 510 |
| **Zeolite** | 12.6 | 400 | 450 |

In $CO_2$ adsorption tests materials have been preliminary heated up to 393K, in order to remove any trace of moisture. In a typical experiment, the sorbent (85g of zeolite and 100g of activated carbon) 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 operating conditions. This is followed by the adsorption step in which a $CO_2/N_2$ gas mixture at a fixed $CO_2$ concentration is fed through the column. The $CO_2$ composition in the column effluent gas is continuously monitored as a function of time (breakthrough curve) until the composition approaches the inlet gas composition value, i.e., until saturation is reached. Each adsorption test has been performed both in ordinary and sound assisted conditions. Table 2 reports the operating conditions selected for the adsorption experiments carried out in this work.

**Table 2.** Operating conditions of the adsorption tests.

| | ZEOLITE | ACTIVATED CARBON |
|---|---|---|
| **Fluidization velocity, cm/s** | 2, 5 | 2 |
| **$CO_2$ inlet concentration, %vol.** | 10 | 5, 10, 15 |
| **SPL, dB** | 140 | 125, 140 |
| **Frequency, Hz** | 80 | 80 |

**Results and discussion**

Both the powders have been characterized before the adsorption tests, from a fluid-dynamic point of view, by performing ordinary and sound assisted fluidization tests. A beneficial effect of the sound application in terms of more regular pressure drops and bed expansion curves has been observed.

Fig. 3 reports the breakthrough curves obtained with the activated carbon powder in ordinary, sound assisted fluidization and fixed bed conditions for the tests performed with a $CO_2$ concentration in the inlet stream of 10%vol. Analogous trends have been obtained for the other operating conditions. The analysis of the curves and the results reported in Table 3 show that adsorption efficiency is remarkably improved moving from fixed bed condition to that of fluidized bed and, above all, to that of sound-assisted fluidized bed as a consequence of better gas-solid contact. In particular, as a result of sound application the global adsorption capacity is strongly improved: the percentage increase moves from 13% for the test performed at 140dB and 10%vol. of $CO_2$ in the inlet stream up to 27% of the test performed at 125dB and 10%vol. $CO_2$.

The application of the sound greatly increases the percentage of bed utilized until break point and the breakthrough time, which in all the sound assisted tests is more than five times the value obtained in ordinary conditions (see Table 3). The beneficial effect shown by the sound is probably due to the enhancement of the

fluidization quality with respect to the tests performed in ordinary conditions, namely without the aid of any external force.

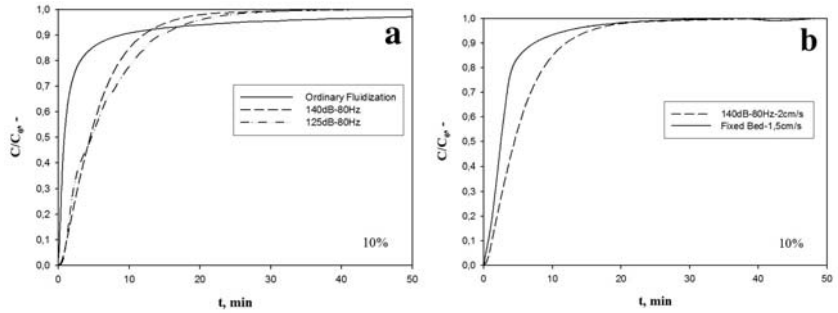


**Figure 3.** Activated carbon breakthrough curves in a) ordinary and sound assisted fluidization; b) sound assisted fluidization and fixed bed conditions.

**Table 3.** Experimental results obtained for activated carbon.

| **Inlet $CO_2$ %vol.** | **5** | | | **10** | | | **15** | | |
|---|---|---|---|---|---|---|---|---|---|
| | **$t_b$ s** | **$n_{ads}$ mol/kg** | **W %** | **$t_b$ s** | **$n_{ads}$ mol/kg** | **W %** | **$t_b$ s** | **$n_{ads}$ mol/kg** | **W %** |
| **Ordinary Fluidization** | 15 | 0.22 | 3.6 | 8 | 0.30 | 2.7 | 7 | 0.37 | 2.7 |
| **125dB** | 71 | 0.28 | 14 | 55 | 0.38 | 14 | 48 | 0.46 | 15 |
| **140dB** | 73 | 0.26 | 15 | 51 | 0.34 | 14 | 43 | 0.44 | 14 |
| **Fixed Bed** | | | | 6 | 0.18 | 2.5 | | | |

In particular, as already shown in our previous works [6], the fluidization of cohesive powders is based on a break-up and re-aggregation mechanism, which is greatly enhanced by the application of acoustic fields; so, the improved fluidization quality arising from the application of the sound is likely to be the main reason of the observed enhancement of the adsorption efficiency. In other words, the continuous aggregates break-up and re-aggregation mechanism makes the internal surface of the activated carbon more readily available for the adsorption process.

As regards the influence of $CO_2$ partial pressure, the analysis of the results reported in Table 3, shows, as expected, that the $CO_2$ capture capacity of the adsorbent at a fixed temperature increases with $CO_2$ partial pressure; this trend is absolutely consistent from a thermodynamic point of view; in fact, the $CO_2$ partial pressure represents the driving force of the adsorption process, so its increase corresponds to higher adsorption capacity.

The experimental results have been fitted by Langmuir equation in order to obtain the activated carbon adsorption isotherms, Fig. 4. From the analysis of the curves the above-mentioned beneficial effect played by the application of the acoustic field can be inferred. It is indeed clear that with sound assisted fluidization the adsorption isotherms move to more favorable adsorption conditions.

Finally, the increase of temperature due to the exothermicity of the process is very small (2-3°K) in all the experimental tests due to the high heat capacity of fluidized beds.
Similar results have been obtained with zeolite powder but the effect of the sound on the global $CO_2$ capture capacity is strongly reduced in this case.

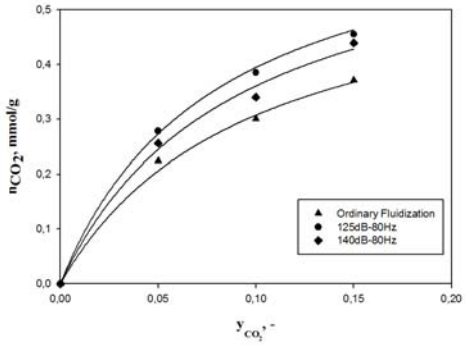


**Figure 4.** Activated carbon adsorption isotherms in ordinary and sound assisted fluidization.

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**References**

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