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Effect of acoustic field on $CO₂$ desorption in a fluidized bed of fine activated carbon

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A B S T R A C T

Adsorption using solid sorbents has the potential to complement or replace current absorption technology, because of its low energy requirements. Among the commercially available adsorbent materials, attention is focused on activated carbons because they are easily regenerable by reason of their low heat of adsorption. These sorbents are generally available in the form of fine powders. Sound-assisted fluidization can process large amounts of fine powders, promoting and enhancing CO₂ capture on fine sorbents, because it maximizes gas–solid contact. Temperature swing adsorption (TSA), consisting of inducing sorbent regeneration and CO₂ recovery by appropriate temperature increase and gas purge, is one of the most promising techniques. This study investigates the $CO₂$ desorption process by TSA in a sound-assisted fluidized bed of fine activated carbon. Desorption tests were performed under ordinary and sound-assisted fluidization conditions to assess the capability of sound to promote and enhance the desorption efficiency in terms of CO_2 recovery, CO_2 purity, and desorption time. The results show that the application of sound results in higher desorption rates, $CO₂$ recovery and purity. Regular and stable desorption profiles can be obtained under sound-assisted fluidization conditions. This stability makes it possible to successfully realize a cyclic adsorption/desorption process.

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Introduction

Global climate change is a widespread public concern, with annual global emissions of $CO₂$ increasing by approximately 80% between 1970 and 2004 ([Steeneveldt,](#page--1-0) [Berger,](#page--1-0) [&](#page--1-0) [Torp,](#page--1-0) [2006\).](#page--1-0) This drastic rise has been attributed to an increasing dependence on the combustion of fossil fuels (coal, petroleum, and natural gas), which accounts for 86% of anthropogenic greenhouse gas emissions. One possible way to mitigate this trend is to remove $CO₂$ from the gaseous streams in which it is contained to safely dispose of it in deep underground areas ([Steeneveldt](#page--1-0) et [al.,](#page--1-0) [2006;](#page--1-0) [Yang,](#page--1-0) [2010\).](#page--1-0) Post-combustion $CO₂$ capture processes have the greatest nearterm potential for reducing greenhouse gas emissions because these processes can be retrofitted to existing units, providing a quicker solution to mitigate $CO₂$ environmental impacts. Among all the post-combustion technologies, adsorption processes are attrac-tive because of their low energy requirements ([Pirngruber,](#page--1-0) [Guillou,](#page--1-0)

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[Gomez,](#page--1-0) [&](#page--1-0) [Clausse,](#page--1-0) [2013\).](#page--1-0) However, for adsorption to be used as a $CO₂$ capture technique three major points need to be addressed: the choice of adsorbent materials, regeneration strategy, and reactor configuration.

The regeneration energy, followed by the capital cost of capturespecific equipment, are the two variables that most significantly contribute to the cost of $CO₂$ capture. In particular, a significant contributor to the regeneration energy is the maximum separation efficiency that can be achieved by a given capture material. Enhancing this efficiency has the greatest potential to lower the overall cost of capture systems in the near-term, with improvements in the capture capacity of new materials representing one of the foremost challenges. Great interest is now focused on the use of fine materials for this process. Because the adsorption efficiency of a given sorbent is the result of a complex combination of physical and chemical characteristics, the development of $CO₂$ specific adsorbents is necessary. There is a need to design new materials whose physical and chemical properties can be tuned at the molecular level ([D'Alessandro,](#page--1-0) [Smit,](#page--1-0) [&](#page--1-0) [Long,](#page--1-0) [2010\).](#page--1-0) Fine powders are particularly versatile and simple to adjust according to the application requirements. Because of their special size and shape,

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ultra-fine particles can be easily tailored and/or functionalized on the surface with different ligands to induce significant changes in their physical and chemical properties ([Dawson,](#page--1-0) [Cooper,](#page--1-0) [&](#page--1-0) [Adams,](#page--1-0) [2013;](#page--1-0) [Keramati](#page--1-0) [&](#page--1-0) [Ghoreyshi,](#page--1-0) [2014;](#page--1-0) [Wang,](#page--1-0) [Liu,](#page--1-0) [Ramström,](#page--1-0) [&](#page--1-0) [Yan,](#page--1-0) [2009\).](#page--1-0) Moreover, many common adsorbent materials (such as activated carbon, zeolites, etc.) are generally commercially available in the form of fine powders.

Four main options are available as regeneration techniques: pressure swing adsorption (PSA) ([Kikkinides,](#page--1-0) [Yang,](#page--1-0) [&](#page--1-0) [Cho,](#page--1-0) [1993;](#page--1-0) [Thiruvenkatachari,](#page--1-0) [Su,](#page--1-0) [Yu,](#page--1-0) [&](#page--1-0) [Bae,](#page--1-0) [2013\),](#page--1-0) vacuum swing adsorption (VSA)([Dutcher,](#page--1-0) [Adidharma,](#page--1-0) [&](#page--1-0) [Radosz,](#page--1-0) [2011;](#page--1-0) [Xiao](#page--1-0) et [al.,](#page--1-0) [2008\),](#page--1-0) temperature swing adsorption (TSA) [\(Merel,](#page--1-0) [Clausse,](#page--1-0) [&](#page--1-0) [Meunier,](#page--1-0) [2008;](#page--1-0) [Plaza,](#page--1-0) [García,](#page--1-0) [Rubiera,](#page--1-0) [Pis,](#page--1-0) [&](#page--1-0) [Pevida,](#page--1-0) [2010;](#page--1-0) [Yu,](#page--1-0) [Huang,](#page--1-0) [&](#page--1-0) [Tan,](#page--1-0) [2012\),](#page--1-0) and electric swing adsorption (ESA) [\(Grande,](#page--1-0) [Ribeiro,](#page--1-0) [Oliveira,](#page--1-0) [&](#page--1-0) [Rodrigues,](#page--1-0) [2009;](#page--1-0) [Yu](#page--1-0) et [al.,](#page--1-0) [2012\).](#page--1-0) Among these, TSA is one of the best technological alternatives, presenting several advantages over the others. In a conventional TSA regeneration process, the adsorbent temperature is increased by purging the bed with a preheated gas (e.g., steam or N_2) ([Merel](#page--1-0) et [al.,](#page--1-0) [2008\).](#page--1-0) Direct heating using a carrier gas usually results in dilution of the desorbed $CO₂$ by the carrier gas. Also, the use of large quantities of heated $N₂$ for adsorbent bed regeneration would not be suitable for large scale applications. However, the heat for $CO₂$ desorption (to regenerate the solid) can be provided in a different way: for instance, the adsorbent can be indirectly heated by heat exchanger tubes. In this way, $CO₂$ is desorbed without the use of any carrier gas and the desorbed gas could be recovered by thermal expansion, thus overcoming the dilution problem (resulting in a pure $CO₂$ stream as the desorption gas) ([Merel](#page--1-0) et [al.,](#page--1-0) [2008\).](#page--1-0) Indirect heating can provide much more energy than direct heating because the heating fluid (which is water vapor by default) can be allowed to condense. The heat of condensation releases much more energy than the simple cooling of gas.

Finally, for the choice of reactor configuration, although certain attributes of solid sorbents prove to be promising, they must be integrated into a viable process that can take full advantage of the sorbent properties and maximize the separation efficiency. If a solid-based $CO₂$ capture technology is to be commercialized in the near future, processes and equipment that are already commercially available for other applications must be evaluated and employed to the greatest extent possible ([Sjostrom,](#page--1-0) [Krutka,](#page--1-0) [Starns,](#page--1-0) [&](#page--1-0) [Campbell,](#page--1-0) [2011;](#page--1-0) [Yang](#page--1-0) [&](#page--1-0) [Hoffman,](#page--1-0) [2009\).](#page--1-0)

Common adsorption operations are generally performed in fixed bed reactors. However, this technology does not appear suitable to fully exploit the potential of an ad hoc fine powder adsorbent material. For fine materials to be used in fixed bed operations, a previous pelletization step is needed to overcome the prohibitively high pressure drops related to fine particles beds. Therefore, a reliable processing technology that directly uses these free-flowing fine powders can provide serious benefits overfixed bed adsorption methods. Moreover, the temperature swing in fixed bed reactors that serve as both adsorber and regenerator, is naturally inefficient. More specifically, fixed beds are not recommended for exothermic reactions ($CO₂$ adsorption) or endothermic reactions ($CO₂$ regeneration). Poor local heat dissipation for the exothermic adsorption reactions produces local hot spots, and poor heat transfer to the solid sorbents for the endothermic regeneration reactions produces local cold spots, preventing efficient conversions. In light of these considerations, a fluidized bed could be a viable alternative to the fixed bed configuration. The problem of $CO₂$ dilution could also be easily overcome in the case of a fluidized bed reactor, by preheating the sorbent with tube heat exchangers immersed in the bed (i.e., without any hot gas purged inside the bed during heating of the sorbent). After this pre-heating step (similar to the case of a fixed bed, with no gas flowing inside the bed), the high heat transfer coefficients typical of fluidized bed reactors (because of the large interfacial particle-gas surface area) would maintain the temperature uniformly at the desired value. This would avoid the presence of low temperature zones inside the reactor (because of the endothermicity of the desorption process). The high heat transfer rate means easier temperature control compared with fixed bed operations, during both the adsorption and regeneration steps. Also, the unfortunate case of a particularly large exothermic heat of adsorption, causing the solid sorbent particles to be heated beyond the desired $CO₂$ adsorption temperature, could be resolved by embedding heat transfer surfaces inside the fluidized bed adsorber to remove the heat generated during $CO₂$ capture ([Yang](#page--1-0) [&](#page--1-0) [Hoffman,](#page--1-0) [2009\).](#page--1-0) Finally, because of the small sizes of the solid sorbents employed, the operating velocity tends to be low and thus it can be expected that attrition, which is one of the main drawbacks of fluidization, may not be excessive.

However, because of their intrinsic cohesive character, fine particles cannot be fluidized under ordinary conditions [\(Shabanian,](#page--1-0) [Jafari,](#page--1-0) [&](#page--1-0) [Chaouki,](#page--1-0) [2012;](#page--1-0) [Wang,](#page--1-0) [Rahman,](#page--1-0) [&](#page--1-0) [Rhodes,](#page--1-0) [2007;](#page--1-0) [Zhu,](#page--1-0) [Yu,](#page--1-0) [Dave,](#page--1-0) [&](#page--1-0) [Pfeffer,](#page--1-0) [2005\);](#page--1-0) therefore, assistance methods are required to achieve a smooth fluidization regime. Several externally assisted fluidization techniques have been proposed in the literature, all of them involving the application of additional forces generated, for example, acoustically ([Ammendola,](#page--1-0) [Chirone,](#page--1-0) [&](#page--1-0) [Raganati,](#page--1-0) [2011a;](#page--1-0) [Ammendola,](#page--1-0) [Chirone,](#page--1-0) [&](#page--1-0) [Raganati,](#page--1-0) [2011b;](#page--1-0) [Ammendola](#page--1-0) [&](#page--1-0) [Chirone,](#page--1-0) [2010\),](#page--1-0) electrically [\(Kashyap,](#page--1-0) [Gidaspow,](#page--1-0) [&](#page--1-0) [Driscoll,](#page--1-0) [2008\),](#page--1-0) by magnetic fields ([Zang,](#page--1-0) [Zhou,](#page--1-0) [&](#page--1-0) [Yang,](#page--1-0) [2008\),](#page--1-0) or by mechanical vibrations ([Kaliyaperumal,](#page--1-0) [Barghi,](#page--1-0) [Briens,](#page--1-0) [Rohani,](#page--1-0) [&](#page--1-0) [Zhu,](#page--1-0) 2011) to enhance the powder dynamics in the fluidized bed. The application of acoustic vibrations does not require any material modification, it is relatively inexpensive and can easily be technically implemented using simple sound-generation devices. It is likely that soundassisted fluidization may also be competitive at the industrial level ([Raganati,](#page--1-0) [Ammendola,](#page--1-0) [&](#page--1-0) [Chirone,](#page--1-0) [2014b\).](#page--1-0) The scale-up of this technology would not be a problem because an array of loudspeakers can produce a plane wave if conveniently placed on the cross-sectional area of the column [\(Raganati](#page--1-0) et [al.,](#page--1-0) [2014b\).](#page--1-0) The use of an array of loudspeakers would also provide greater reliability for continuous operation, because replacement of one of the loudspeakers can easily be carried out without interruption of the sound-assisted process.

On the basis of these considerations, it appears that the use of fine sorbents (even functionalized) in a TSA process under sound-assisted fluidization conditions is a viable and promising technological solution. Sound-assisted fluidization has already been proven to maximize the $CO₂$ adsorption capacity of fine sorbents as compared with common technologies, because of the efficient gas–solid contact that maximizes the solid sorbent exposed surface [\(Alfe,](#page--1-0) [Ammendola,](#page--1-0) [Gargiulo,](#page--1-0) [Raganati,](#page--1-0) [&](#page--1-0) [Chirone,](#page--1-0) [2015;](#page--1-0) [Ammendola,](#page--1-0) [Raganati,](#page--1-0) [&](#page--1-0) [Chirone,](#page--1-0) [2015;](#page--1-0) [Raganati,](#page--1-0) [Ammendola,](#page--1-0) [&](#page--1-0) [Chirone,](#page--1-0) [2014a,](#page--1-0) [Raganati](#page--1-0) et [al.,](#page--1-0) [2014b,](#page--1-0) [Raganati,](#page--1-0) [Ammendola,](#page--1-0) [&](#page--1-0) [Chirone,](#page--1-0) [2015;](#page--1-0) [Raganati,](#page--1-0) [Gargiulo,](#page--1-0) [Ammendola,](#page--1-0) [Alfe,](#page--1-0) [&](#page--1-0) [Chirone,](#page--1-0) [2014;](#page--1-0) [Valverde](#page--1-0) et [al.,](#page--1-0) [2013\).](#page--1-0)

Therefore, this work is focused on the study of $CO₂$ desorption in a sound-assisted fluidized bed of commercial activated carbon. In particular, experimental tests were performed to assess the capability of the sound to promote and enhance the regeneration process in terms of $CO₂$ recovery, $CO₂$ purity, and desorption time.

Material and methods

Experimental apparatus

The experimental tests were performed in a laboratory scale sound-assisted fluidized bed experimental apparatus. The scheme of the plant is presented in [Fig.](#page--1-0) 1.

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