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A multi-stage fluidized bed system for continuous CO₂ capture by means of temperature swing adsorption – First results from bench scale experiments

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ABSTRACT

Most recently, the authors of this work introduced a reactor design for a new post-combustion CO₂ capture process that is based on continuous temperature swing adsorption. The system comprises two interconnected multi-stage fluidized bed columns that enable counter-current contact of adsorbent and gas streams in the adsorption and desorption step. The system further allows for effective heat management through indirect heat exchange with the fluidized adsorbent material in each stage. Based on the proposed reactor design, a fully integrated bench scale unit (BSU) has been constructed and put into operation to study the process experimentally. The BSU is designed to capture 90% of the CO₂ present in synthetic flue gas mixtures at a maximum capture rate of 35 kg_{CO₂,capt} per day using amine-functionalized adsorbent materials. First results obtained from continuous CO₂ capture experiments showed that the design capture targets can be reached with this unit. Despite the fact that only five fluidized bed adsorption stages were implemented, CO₂ was not detected at the adsorber outlet during the initial minutes of the experiments, indicating that gas-solids contact is sufficient and adsorption kinetics are fast. However, in the initial CO₂ capture experiments it was not possible to achieve the desired operating temperatures within the desorber and the CO₂ capture capacity of the unit seemed to be limited by heat transfer. Installation of heat exchangers with a higher surface area entailed an improved capture performance. It was thus concluded that performance limitations of the TSA BSU lie within its heat transfer properties rather than within the prevailing adsorption conditions.

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1. Introduction

Global warming is unequivocal and anthropogenic greenhouse gas emissions have been identified as its predominant cause. Continued emission of greenhouse gases (GHGs) would cause further warming of the climate and thus amplify already existing risks for natural and human systems. Consequently, substantial and sustained reductions of greenhouse gas emissions are required [1].

Among the anthropogenic GHGs, CO₂ emissions that arise from combustion of fossil fuels have the greatest impact on climate change. Nevertheless, the contribution of fossil fuels to the global primary energy demand will remain or most likely even increase in the next decades to come [2]. Thus, a portfolio of different strategies and technologies is required to achieve the required CO₂ emission reductions.

According to the climate mitigation technology portfolio proposed by the International Energy Agency, carbon capture and storage (CCS) needs to play a significant role on a mid-term basis as this technology allows for continued utilization of fossil fuels and existing assets while CO₂ emissions into the atmosphere are reduced significantly. CCS provides that CO₂ from large point emission sources such as power plants or industrial facilities is separated from the flue gas stream and subsequently transported to a suitable storage site (e.g. depleted oil or gas field) where the CO₂ can be stored and kept from the atmosphere on a long term. However, for successful deployment of CCS technologies it is essential to develop energy and cost efficient CO₂ capture technologies since over 75% of the costs related to the complete CCS chain arise from the gas separation step [3].

Different CO₂ capture routes have been proposed [4], whereas post-combustion CO₂ capture (PCC) technologies seem to exhibit unique selling points such as applicability as simple end-of-pipe solution and a great retrofitting potential for existing CO₂ sources in both power production and industry [5]. Among the PCC technologies discussed today, absorption in aqueous amine solvents is considered as the most mature technology. However, amine scrubbing technologies exhibit

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several disadvantages that typically result in large process heat demands and CO₂ capture costs. The large process heat demands arise from high heat capacities of the aqueous amine solutions, from the heat of reaction that needs to be supplied during regeneration of the loaded solvent and from the fact that this regeneration step is accompanied by water evaporation. Due to rather low absorption rates, sizeable, packed absorption columns are required to achieve high CO₂ capture rates. Furthermore, solvent degradation products are potentially corrosive, which sets high demands on construction materials as well as on off-gas treatment facilities. The corresponding capital costs are thus high and add to the comparably large overall CO₂ capture costs [5]. In the past decades, the development of novel amine solvents together with the elaboration of optimized heat integration strategies led to process improvements and reduced CO₂ capture costs [5–8]. However, the aforementioned drawbacks of amine scrubbing remain as they are intrinsic to the process. Consequently, breakthroughs in the reduction of CO₂ capture costs require the development of alternative capture technologies. A great effort has thus been put into the identification and development of alternative CO₂ capture processes that are capable of capturing CO₂ with lower parasitic energy demand and at significantly reduced costs.

The application of adsorption based processes has been proposed as a viable route to achieve this goal [9–12]. Solid sorbents usually exhibit higher CO₂ working capacities, lower material heat capacities and lower heat of reaction compared to chemical solvents. Furthermore, evaporation of water in the regeneration step can be completely avoided. Thus, the process energy demand of an adsorption based CO₂ capture process is expected to be markedly smaller compared to an amine scrubbing process. The large intrinsic areas of the porous adsorbent materials may also allow for fast adsorption kinetics which in turn should allow for reduced plant sizes and capital costs.

Commercially available physisorbents like activated carbons or zeolites are known to be capable of adsorbing CO₂. However, physisorbents typically show weak CO₂ selectivity especially in the presence of water and low adsorption capacities at small CO₂ partial pressures; i.e. at conditions relevant for CO₂ capture from stack flue gas. This circumstance promoted a world-wide effort in the development of novel CO₂ adsorbents. Inspired by the excellent CO₂ absorption properties of amine-based solvents, researchers immobilized amines onto solid support materials and thereby developed a new class of highly selective CO₂ adsorbent materials [13–19]. The high selectivity is reached through chemical reactions (i.e. chemisorption) that take place between active amine sites on the adsorbent's surface and CO₂ from the bulk gas. However, this desirable property comes at the cost of comparably large amounts of heat released in the chemisorption step. This heat needs to be withdrawn during CO₂ adsorption and subsequently supplied during CO₂ desorption. This in turn sets high demands on the heat management capabilities of the adsorption process. On the other hand, it has been shown that larger adsorption enthalpies lead to stronger temperature dependencies of the adsorbent's adsorption capacity [11]. This in turn is beneficial for efficient operation in temperature swing adsorption (TSA) based processes as shown in Fig. 1.

Compared to the effort put into the development of novel adsorbent materials, rather little work has been attributed to the development of suitable reactor designs or to experimental investigations of the TSA process under practical conditions.

Zhao et al. [20] studied a continuous TSA process for CO₂ capture using a single-stage fluidized bed in the adsorber and the desorber. Although a fluidized bed system provides excellent heat and mass transfer properties, which are advantageous for the gas separation task, Pröll et al. [21] showed that for efficient operation of the TSA process in a fluidized bed system, counter-current flow of sorbent and gas needs to be realized. This will allow for high working capacities of the sorbent material together with low CO₂ concentrations in the adsorber outlet and high CO₂ concentrations in the desorber off-gas. Veneman et al. [22] implemented counter-current flow in their TSA process and

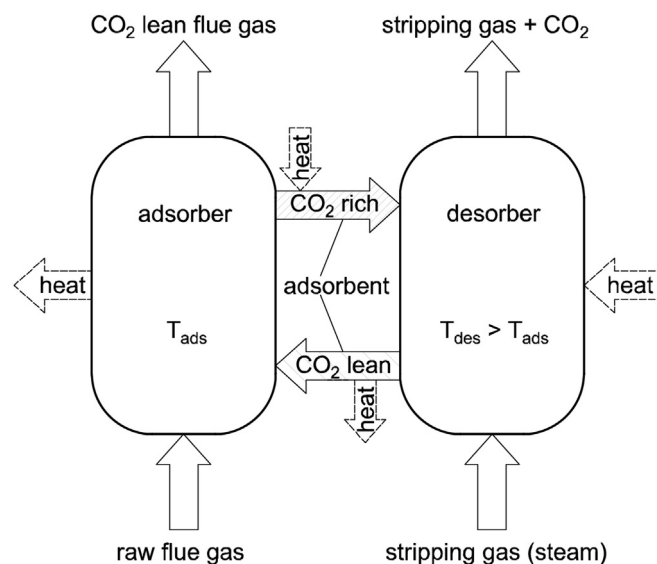


Fig. 1. Principle of a continuously operated temperature swing adsorption process.

investigated a setup comprising a gas-solid trickle flow adsorber and a multi-stage fluidized bed desorber. Instead of using steam as stripping gas, Krutka et al. [23] fluidized the sorbent material in the desorber with pure CO₂ which makes counter-current flow unnecessary from a thermodynamic point of view.

In the TSA system introduced herein, both the adsorber and the desorber feature a multi-stage fluidized bed design. The reactors are interconnected by a solids transport system consisting of screw conveyors, pneumatic transport risers and gravitational gas-solids separators. Furthermore, a stripping gas in the form of nitrogen or steam can be used to fluidize the sorbent material in the desorber and to promote the regeneration of the loaded sorbent material coming from the adsorber. This setup allows for reaching high dynamic CO₂ loadings of the circulating adsorbent stream at low stripping gas demands. Such a system has been recently introduced by the authors of this work and theoretically evaluated by using a thermodynamic equilibrium adsorption model [21]. Furthermore, experiments within a multi-stage fluidized bed cold flow model have been conducted to study the operating behavior of the proposed system from a fluid-dynamic point of view [24]. Basing on these studies, a fully integrated TSA bench scale unit (BSU) for continuous CO₂ separation from synthetic flue gas mixtures has been designed, constructed and put into operation. This work introduces the design and set-up of the BSU and further presents first results obtained from continuous CO₂ capture tests.

2. Experimental setup and operation

2.1. The TSA bench scale unit for continuous CO₂ capture from flue gas streams

Fig. 2 shows a basic scheme of the TSA bench scale unit (BSU) for continuous CO₂ capture from flue gas streams together with a picture of the fully commissioned unit. The BSU has been constructed in order to deliver a proof of concept for the proposed continuous TSA CO₂ capture process and to further experimentally study said process and the effect of the relevant operating parameters on the process performance. As one can see from Fig. 2, both the adsorber and the desorber column consist of five individual fluidized bed stages that are stacked on top of each other. The columns are, however, built modularly, thus allowing for experiments with varying stage numbers in each column. Each of the individual stages features a perforated plate type gas distributor, a downcomer that enables controlled flow of adsorbent from stage to stage, an exchangeable weir plate and an immersed heat exchanger

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