Review on Reactor Configurations for Adsorption-Based CO_2 Capture

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ABSTRACT: Adsorption-based CO_2 capture has enjoyed considerable research attention in recent years. Most of the research efforts focused on sorbent development to reduce the energy penalty. However, the use of suitable gas−solid contacting systems is key for extracting the full potential from the sorbent to minimize operating and capital costs and accelerate the commercial deployment of the technology. This paper reviews several reactor configurations that were proposed for adsorption-based CO_2 capture. The fundamental behavior of adsorption in different gas−solid contactors (fixed, fluidized, moving, or rotating beds) and regeneration under different modes (pressure, temperature, or combined swings) is discussed, highlighting the strengths and limitations of different combinations of gas−solid contactor and regeneration mode. In addition, the estimated energy duties in published studies and current technology readiness level of the different reactor configurations are reported. Other aspects, such as the reactor footprint, the operation strategy, suitability to retrofits, and the ability to operate under flexible loads are also discussed. In terms of future work, the key research need is a standardized techno-economic benchmarking study to calculate CO_2 avoidance costs for different adsorption technologies under standardized assumptions. Qualitatively, each technology presents several strengths and weaknesses that make it impossible to identify a clear optimal solution. Such a standardized quantitative comparison is therefore needed to focus on future technology development efforts.
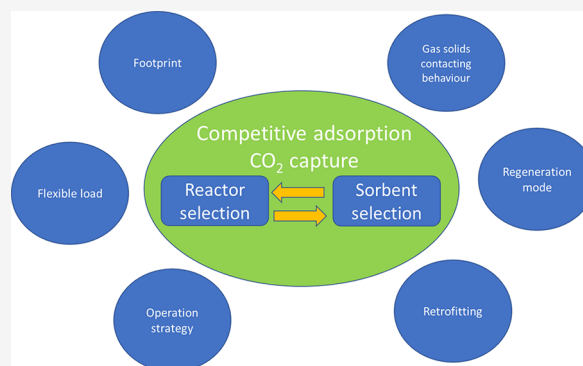
1. INTRODUCTION

The growing global warming threats caused by anthropogenic CO_2 emissions are increasingly dictating the need for a radical shift to a more sustainable energy system and environmentally friendly industrial production practices. In this context, the Paris Climate agreement, with the goal to limit global warming to well below 2 °C, recommended implementation of stringent policy measures to incentivize cutting CO_2 emissions. CO_2 capture and storage (CCS) is considered a vital technology to include in CO_2 emission reduction pathways for cost-effective mitigation of global warming threats.[1] Among other CCS technologies, there is a growing interest in adsorption-based postcombustion CO_2 capture due to its combined potential of reducing energy penalty and easy retrofitting with minimal integration with existing plants.[2,3] More importantly, this technology offers the flexibility of capturing CO_2 from different industrial CO_2 sources owing to its different sorbent regeneration modes (temperature/pressure swings) and reactor types. To date, research in this field has focused mainly on sorbent development to reduce the energy penalty through minimizing the heat of adsorption (sorption) and maximizing the adsorption capacity, but also improving tolerance to impurities such as SOx and NOx.[2,4] Sorbents could be classified in two categories depending on the heat of CO_2 sorption. The mechanism by which physisorption CO_2

adsorption occurs is driven by van der Waals and/or electrostatic interactions between the CO_2 molecule and adsorbent surface.[2,4,5] As for chemisorption, a chemical reaction takes place between CO_2 and the active sites introduced to the sorbent through functional groups that usually include alkaline carbonates or various amine groups.[2,4] The different adsorption mechanisms taking place in each category make the physisorption based process less sensitive to temperature and associated with low reaction enthalpy, being suitable to high CO_2 partial pressure gas streams, while the chemisorption based process is more sensitive to temperature swing and can handle low CO_2 partial pressure gas streams. Recent research on physisorption focused on metal organic framework (MOF) based sorbents that possess high specific surface area, thus maximizing the absolute adsorption capacity.[2,6−11] As for the chemisorption-based sorbents, the largest focus is on those that are polyethyleneimine (PEI)
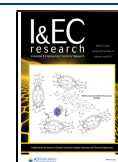
based, given their relatively high adsorption capacity, good kinetics, and insensitivity to water.[2,6,12−14]

On the other hand, the energy penalty was the main driving factor behind the rapidly growing research in this field. It was commonly stated that adsorption-based CO_2 capture can achieve a lower energy penalty due to the lower specific heat capacity of solid sorbents in addition to avoiding evaporation of a large amount of water in the regeneration as compared to solvent based technologies. This statement was however debated given the scattered range of the energy penalty data that were reported in the literature,[15] creating confusions about adsorption-based CO_2 capture competitiveness with benchmarking technologies. Therefore, there is an urgent need to identify the many factors (beyond the sorbent) that affect the performance and overall cost of adsorption-based CO_2 capture, and discuss the nonlinear interaction between them that affects the technology behavior, performance, and prospects for scale up and ultimate industrial implementation.

Adsorption CO_2 Capture beyond Sorbent Development. A suitable contacting system is a key factor for efficient utilization of each sorbent category, as it affects both the process efficiency, footprint and overall capture costs.[6] In other words, material development should be tightly linked to the reactor configuration and regeneration mode.[2,4] To this end, different types of reactors were applied to adsorption-based CO_2 capture, including fixed,[16,17] rotating,[18] moving,[19−21] and fluidized beds.[22,23] Substantial research has been conducted on the fixed bed configuration, due to the simplicity of its basic design, testing hundreds of sorbents under different regeneration modes,[16,17,24] but the interest to the other reactor configurations has steadily grown in recent years.[25−27]

Other key factors that affect the technology competitiveness are the total footprint, the ease of retrofitting to existing plants (e.g., level of integration with the existing plant and possible need for steam), and operability (some reactor configurations require only two reactors with sorbent circulating between them, while others operate using trains of dozens of reactors requiring advanced operating strategies). Additional aspects such as the potential for flexible operation and performance under partial capture scenarios are becoming increasingly important in a future energy system dominated by renewable energy. In this respect, similar to what was proposed for solvent-based CO_2 capture technology,[28,29] adsorption technology can make CO_2 capture cost-effective if it can use the excess of cheap renewable electricity in peak periods, combined with partial CO_2 capture when electricity price is high.[30]

All these aspects will be discussed in this review paper for the different reactor configurations proposed for low temperature postcombustion CO_2 capture by putting together key published research on those systems, discussing their working principles, nature of sorbents suitable for each configuration and suitable regeneration modes. The review will also highlight the pros and cons of each configuration, the energy penalty, the level of technological development, the total footprint, the ease of retrofitting into existing plants, operation strategy, the best suited CO_2 sources (industrial, power, waste, etc.), in addition to their potential for flexible operation and partial capture. This review will also shed light on the recent research trends and discuss the technical challenges and future research needs for further scale up of each configuration.

First, the various reactor configurations and different regeneration modes are discussed. Second, the process

integration and the technology readiness level (TRL) of these technologies are discussed. Finally, a discussion highlighting the role of all other factors affecting the overall competitiveness of adsorption technology and conclusions are presented. It should be noted that other published reviews partially touched on this topic,[4,15] but the focus was to a large extent on the fixed bed configuration, given the large number of studies existing in the literature for this configuration, but also due to the rapid advances made especially on the material development side. These reviews have paid little attention to the various other emerging reactor configurations and their sensitivity to the regeneration modes and other factors affecting their further scale up and commercialization.

2. REACTOR CONFIGURATIONS

The different reactor configurations previously proposed for low temperature adsorption-based CO_2 capture are shown in Figure 1. These configurations are classified into three main
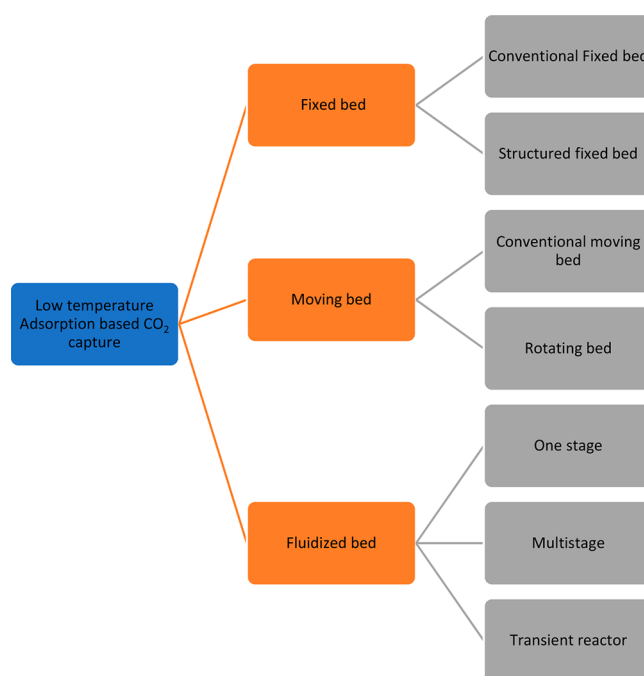

Figure 1. Reactor configurations used in low temperature adsorption-based CO_2 capture.

categories, that is, fixed, moving, and fluidized bed reactors. In a fixed bed reactor, adsorbent particles are stationary, while they are moving in the moving and fluidized reactor configurations. Each of these three categories are further divided into the different configurations. For fixed bed, it includes conventional fixed bed and structured reactor, for moving bed there is conventional moving bed and rotating bed, while for fluidized bed there is one stage, multistage, and transient reactor configuration. The details about these reactors, working principles, and their pros and cons are discussed in respective sections.

2.1. Fixed Bed Reactor. The fixed bed is the simplest reactor configuration where a flue gas is passed through a fixed bed of sorbents pellets (mm size) or through specially designed structured packings. Structure packings are used to optimize the surface areas and void space to achieve a high adsorption rate and low pressure drop. The plug-flow nature in this

Table 1. Physical Resistance Dominating in Bigger Pellets of Fixed Bed Reactors

Mass transfer	Heat transfer
Mass transfer of CO_2 from gas phase to the surface of the particle (film mass transfer)	Convective heat transfer from the gas phase to the surface of the particle (h_i = 20−50 W/(m² K))[32]
Diffusion of CO_2 inside the pores of the particle (k_{LDF} = 0.06 s⁻¹; D_e = 5.35 × 10⁻⁵ m²/s at 301 K and 20% CO_2 in N_2 for 13X Zeolite)[33]	Heat transfer from surface to the inside pores of the adsorbent particle (k_f = 0.259 W/(m K) for ion-exchange resin with a primary benzy sorbent)[34]
	Heat transfer from the heat transfer fluid to the gas phase (applicable in indirect heating cases (h_i = ∼10 W/(m² K))[32]

reactor configuration remains the main advantage, keeping the sorbent toward the end of the reactor in a highly regenerated state to ensure maximum CO_2 capture until almost the entire bed is saturated with CO_2. However, fixed beds are known to impose high pressure drops at even moderate gas flowrates, resulting in very large footprints.[31] Overcoming this key drawback requires the use of large particles or structured packings that greatly reduce the pressure drop, while striving to maintain high adsorption rates to allow for much higher gas throughput rates.[4] In addition, fixed beds have inherently poor heat transfer properties, making them best suitable to pressure swing adsorption using physical sorbents with low reaction enthalpy and low temperature sensitivity.

2.1.1. Conventional Fixed Bed Reactor. Generally, large adsorbent pellets are used in this configuration to minimize the pressure drop. This however comes at the expense of increased mass and heat transfer resistance as specified in Table 1, undermining the potential of the plug flow regime to maximize the working adsorption capacity. In such cases, these mass and heat transfer limitations create a dispersed reaction front traveling through the reactor. To illustrate this behavior, typical axial instantaneous plots (along the length of the bed) of the normalized CO_2 concentration in the gas phase and normalized vacant site in the solid phase are shown in Figure 2.
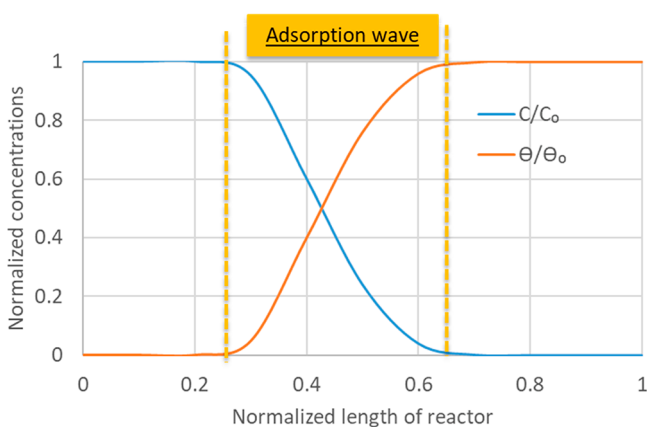
Figure 2. Concentration profile of CO_2 in gas and normalized vacant sites in the solid phase along the length of the reactor at particular time.

Before the adsorption wave, no CO_2 is adsorbed on the solids given that the sorbent is saturated upstream of the wave $\left(\frac{C}{C_0} = 1 \text{ and } \frac{\Theta}{\Theta_0} = 0\right)$. In the adsorption wave, part of CO_2 adsorbs while the rest slips to the next section $\left(0 < \frac{C}{C_0} < 1\right)$ leading to the formation of a dispersing adsorption wave with a decreasing CO_2 concentration that in turn creates an increasing gradient of vacant sites in the adsorption wave.

A wide adsorption wave will cause CO_2 to break through at the reactor outlet while a large portion of the bed is still not fully saturated.[35] Other parameters that affect the dispersion of the adsorption wave are the sorbent properties (i.e., reaction kinetics, heat of adsorption, specific heat, size of the pellet, porosity, nature of the support) and the initial CO_2 in the flue gas and the process conditions.

One main factor that enhances the creation of the adsorption wave is the heat generation associated with CO_2 adsorption, leading to the creation of a heat front that travels along the bed similarly to the reaction front. Its amplitude depends on the heat of reaction (a range of 25−100 kJ/mol-of-CO_2 was reported for different sorbents), the sorbent specific heat capacity, the sorbent active content, reaction kinetics and the initial CO_2 partial pressure in the flue gas.[4,36,37] The resulting increase in the bed temperature adversely affects the reaction equilibrium, thus decreasing the adsorption capacity. This requires more frequent switching of the inlet and outlet valves as less CO_2 can be captured in each cycle. Such a loss in the adsorption capacity is illustrated in Figure 3 (using the
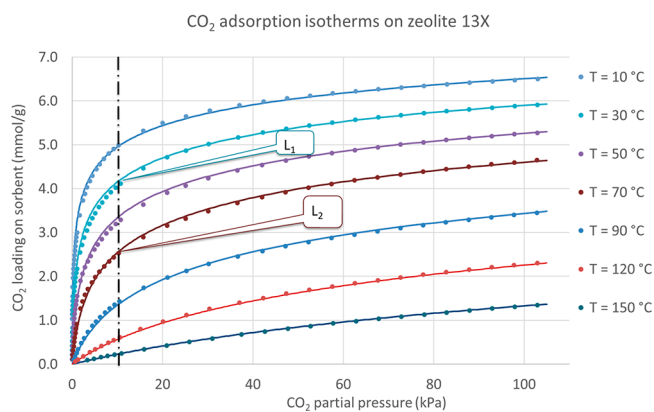
Figure 3. CO_2 isotherm model prediction for zeolite 13X illustrating the decrease in the adsorption capacity caused by the increase in temperature from 30 to 70 °C. Reprinted with permission from ref 38. Copyright 2019 Elsevier.

Virial isotherm model for zeolite 13X),[38] visualizing the theoretically predicted adsorption capacity L_1 at the target operating temperature and the achieved one L_2 due to the temperature rise caused by the heat generation when CO_2 is adsorbed. In the figure, L_1 is the CO_2 loading on zeolite 13X sorbent at 30 °C, the lower temperature, and L_2 is the CO_2 loading on zeolite 13X sorbent at 70 °C, the higher temperature.

Various efforts have been made to tackle the heat wave issue encountered in a fixed bed reactor configuration using two different approaches namely external and internal thermal management. The first approach is based on the use of smartly designed adsorbent particles that can absorb the released heat with minimal temperature change by embedding phase change

materials (PCMs) into the adsorbent particle.[39] The main challenge associated with this approach is the dilution of the active adsorbent material with the PCMs which leads to a decrease in the absolute capacity (moles of CO_2 adsorbed/m^3 of the reactor). The second approach uses a heat exchanger inserted in the reactor to remove the heat of adsorption, but it suffers from a long heating/cooling time because of the poor heat transfer properties of fixed bed reactors and is therefore not considered a viable option.[40] Both approaches lead to the increase in the volume and cost of the reactor.

2.1.2. Structured Fixed Bed Reactor. Structured adsorbent reactors are considered a good alternative to conventional fixed beds. Generally, these systems can accommodate sophisticated packings that can maximize the surface area per volume of sorbent in addition to minimizing gas–particle drag and heat transfer resistance within the packing, thus enabling high gas throughput rates at minimal pressure drop.[41,40–42] With such advantages, this configuration could theoretically reduce the cycle time and the productivity by orders of magnitude, thus accommodating the use of expensive high adsorption capacity sorbents while maintaining competitive CO_2 capture costs.[4] Tested reactors within this configuration consisted predominately of monolithic structures where the adsorbent material is coated with a thin film on the reactor wall. The thin film coating enables the use of higher flowrates at lower pressure drop thus improving the throughput by 3–10 times as compared to fixed bed configurations.[43,44] Some of the expected benefits could already be seen, as the pressure drop can be reduced by 50% as compared to pellets at a superficial velocity of 1 m/s.[43] Such high throughout would require sorbents with fast kinetics to avoid CO_2 slippage. Furthermore, good heat conductivity in the material and reactor wall helps maintain better control on the temperature. These benefits remain however limited due to the low effective sorbent bulk density achieved by using thin film (high percentage of the dead volume occupied by the support monolith). On the other hand, a benefit of having a high dead mass is that it acts as a heat sink and avoids the temperature rise in the adsorption step, which delays the CO_2 breakthrough resulting in better CO_2 capture efficiency. The different structured packings tested so far[40–43,45] remain unable to bring about the targeted considerable reduction in the footprint of fixed bed reactor configuration, given that all of them have less bulk density and use an inert body that occupies a large fraction of the reactor volume. Laminates and foamy structures were also evaluated for CO_2 capture purposes.[40,43,46,47]

2.2. Moving Bed. *2.2.1. Conventional Moving Bed Reactor.* Moving bed reactors are another widely explored reactor type for CO_2 capture as an alternative to fixed bed reactors. Fundamentally, moving beds behave similarly to fixed beds, with the primary benefit being that the moving particle bed allows for steady state operation. The plug-flow behavior of fixed beds is preserved with the additional benefit that the reaction front stays in the same location if the bed moves down at the same rate as the reaction front moves up. This allows for the use of a shorter reactor relative to conventional fixed bed concepts, which helps to reduce the pressure drop. However, the main drawbacks are the complexity of moving relatively large particles between different reactors and the interconnected nature of these reactors that exclude the possibility of a pressure swing. Temperature swing is more complex since the mixing in moving beds is much less than that in fluidized beds,

making indirect heating via heat exchange surfaces relatively inefficient.[48]

The first work on this configuration was proposed by Clyde Berg in 1946 and was known as the "Hypersorption" process[49] and applied for the refinery off gases to recover propane, ethane and ethylene. Lately, SRI International and Advanced Technology Materials, Inc. (ATMI) proposed a new moving bed reactor design with prospects for reducing the energy penalty of postcombustion CO_2 capture technology. The reactor design comprises a circulating system consisting of falling microbeads of an advanced carbon sorbent (ACS) to make the contact with the rising flue gas in counter-current mode.[50,51] The reactor consists of several sections that the sorbent goes through to complete the CO_2 capture cycle (an adsorber, a transition, a desorber, a dehydrator, a cooler, and a lift).[52] The sorbent microbeads enter the reactor from the top starting with the adsorber where the 12.5% CO_2 from the flue gas adsorbs on the sorbent at low temperature (50–60 °C) and atmospheric pressure. While moving downward it passes by a transition section where a purge of steam is used to preheat the sorbent before it enters the desorber for regeneration. In the regenerator, high temperature steam is purged for direct heating at 120 °C. This leads to an additional step in the dehydrator (drying step) followed by a cooling step before the sorbent is lifted to start a new cycle. It should be noted that the used sorbent should be unsensitive to water in this direct heating by steam, while the resulting accumulated water in the CO_2 stream should be removed by condensation to avoid the corrosion issues caused by water during transportation. Structural packings were embedded in different sections to improve the contact between the rising stream with the falling beads.

Kawasaki has also developed a moving-bed process known as the KCC process for CO_2 capture.[20] The process comprises an adsorption reactor, a desorption reactor, and an adsorbent dryer as shown in Figure 4. The sorbent (porous material
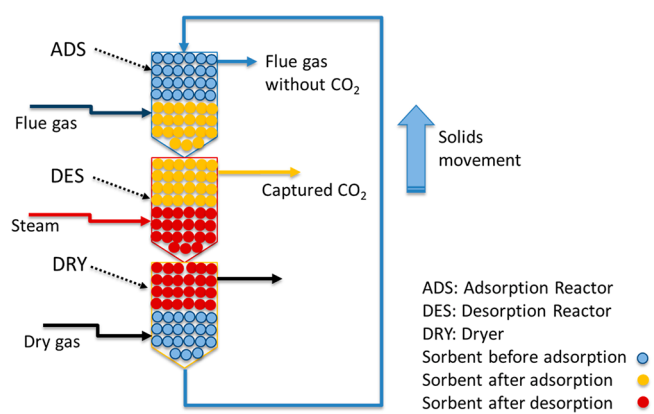

Figure 4. Moving bed process (KCC process) developed by Kawasaki. Adapted with permission from ref 53. Copyright 2017 Elsevier.

impregnated by an amine) material enters the adsorption reactor from the top where the exhaust combustion gas is contacted with the fresh sorbent at low temperature (~30 °C) in counter-current mode. The saturated sorbent with the CO_2 moves then to the desorption reactor where low pressure steam (~60 °C) is contacted in counter current mode to desorb the CO_2 from the sorbent. During this step, steam condenses in the sorbent material, and highly concentrated

CO_2 is recovered at the outlet of the desorption reactor. To remove the accumulated water and control the water content, the sorbent is fed to the adsorbent dryer, where dry gas (i.e., warm air) is contacted with the sorbent in counter-current mode. At last, the sorbent material is discharged from the adsorbent dryer and refed to the top of the adsorption reactor to separate the CO_2 present in the exhaust gas. This process uses low grade steam (<100 °C) for the regeneration.[53] It is not clear how the temperature of the sorbent is decreased after being at the temperature of the dryer to the adsorber inlet.

To avoid the direct steam contact, another version of the MBTSA process was proposed by Knaebel in which hot flue gas is used to indirectly heat the adsorbent in the regeneration.[54] A modeling study of such indirect heating in a moving bed reactor was recently conducted by Mondino et al.,[38] and included some heat integration to recover part of the heat from the hot sorbent leaving the desorption section for preheating the beads leaving the adsorption section (Figure 5). However, the main uncertainty in this version remains the efficiency of heat transfer in the moving bed for the indirect heating and the envisaged heat integration.
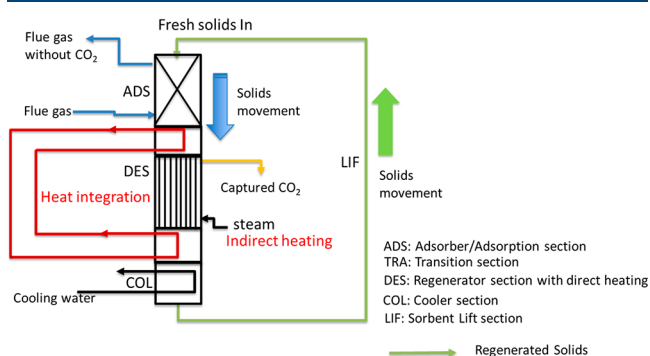

Figure 5. Moving bed with indirect heating. Adapted with permission from ref 19. Copyright 2017 Elsevier.

2.2.2. Rotating Bed. As an alternative to traditional moving beds, Svante Inc. (formerly Inventys, Inc.) has developed a process using a rotating reactor that can efficiently separate CO_2 from industrial flue gas (not much information available in the open literature).[55,56] A moving bed comprising a rotating bed adsorber (RBA) with combined pressure and temperature swing regeneration mode was proposed and simulated by Gupta et al.[18] The RBA consists of disc-shaped adsorbent sheets with parallel passages that are divided into four sections as shown in Figure 6. At any instant, two of the
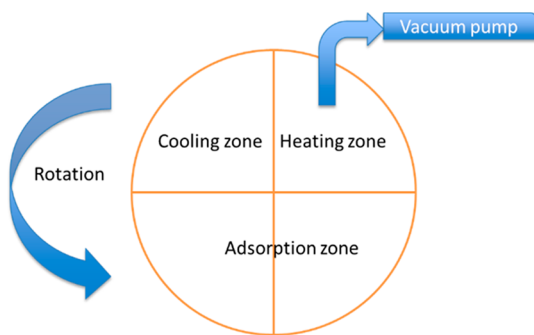

Figure 6. Rotating bed adsorber. Adapted with permission from ref 18. Copyright 2015 Elsevier.

sections are exposed to flue gas for CO_2 adsorption, while the other two remain in the desorption chamber which consists of a heating section using steam that is maintained under vacuum and a cooling section to cool the adsorbent for the next cycle. The separation efficiency and CO_2 purity in this concept depends on the efficiency of the sealing mechanism used to prevent the leakage between the sections operating under different pressures while the solid is rotating. Additionally, a long cycle time should be expected in this configuration given that heating and cooling of the sorbent may be slow.

2.3. Fluidized Bed. In a fluidized bed, the adsorbent particles behave like a fluid with a high mixing rate resulting in excellent heat transfer rate within the bed. This inherently removes the heat and adsorption waves drawback encountered in the fixed bed configuration. A better temperature control could though be established using heat transfer surfaces (heat transfer coefficient in the range of $h_i = 300-600$ W/(m^2 K))[57,58] embedded in the bed to recover or add heat, depending on the need, making the fluidized bed configuration particularly attractive for temperature swing adsorption (TSA) applications. However, the good mixing in fluidized beds also presents its primary drawback: the degree of sorbent adsorption happens uniformly within the bed in equilibrium with CO_2 in the flue gas, leading to early breakthrough of CO_2 in the transient reactor and CO_2 slippage in the continuous reactor configurations.

The adsorption-based CO_2 capture in a fluidized bed reactor predominantly focused on the use of two interconnected reactors[59,60] (Figure 7), namely the adsorber and regenerator
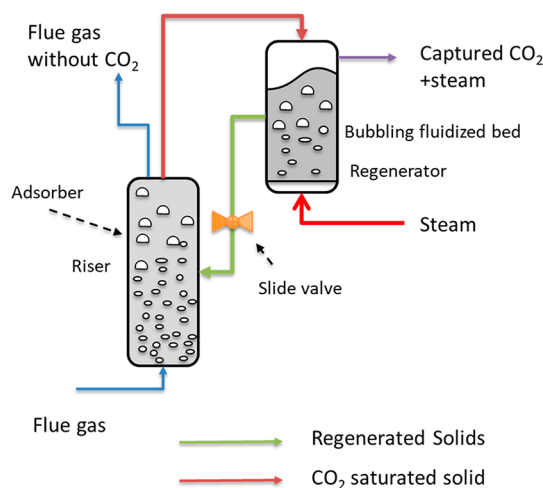
Figure 7. Interconnected fluidized bed configuration.

with the adsorbent particles circulating between them. In early works, the reactors were mainly operated at a cocurrent mode at regimes covering bubbling to fast fluidization running,[61,62] with mainly chemisorption based dry sorbents such as potassium and sodium carbonate. Such a configuration was tested both at the lab and pilot scales with the largest being the one by the Korea Institute of Energy Research (KIER) and the Korea Electric Power Research Institute (KEPRI), using dry sorbents (potassium carbonate) at real flue gas conditions from two MW coal fired plants. The adsorption (carbonation) was carried out in the range of 70−90 °C while the regeneration was completed at 160 °C. The average CO_2 removal was however low (∼70%) despite the very long riser used in the adsorber as a direct result of the good mixing as discussed

above.[59] Application of an acoustic field has resulted in better adsorption capacity and adsorption rate as compared to a conventional fluidized bed adsorption reactor.[63] However, the practicality, scalability, and performance of such a concept in a bigger scale remain questionable. Recently, chemisorbed sorbents such as monoethanolamine impregnated activated carbon, diethanol amine, impregnated activated carbon, and PEI (polyethyleneimine) supported on silica (SiO_2) have received major attention for use in this configuration.[64,65]

To minimize the energy penalty in TSA applications with this reactor configuration, an important challenge is the need for a lean/rich heat exchanger (heat exchange between the hot sorbent from the regenerator and the cold sorbent from the adsorber). Such heat exchange becomes particularly important when the sorbent working capacity reduces, requiring a larger sorbent circulation rate. A solid−solid heat exchanger is considerably more complex, bulkier, and less efficient than the similar liquid−liquid heat exchanger typically used in absorption processes. In addition, effective solids circulation between reactors needs cyclones for gas−particle separation and loop seals to prevent gas mixing between the adsorber and regenerator. Fluidized bed adsorption applications will also require mechanically strong particles to minimize attrition.

Another reactor configuration based on a fluidized bed is the toroidal fluidized bed (also known as vortexing fluidized bed (VFB)[66] with potential for improved gas−particles contact due to the swirling. This is however counteracted by the good mixing that reduces the CO_2 capture rate as emphasized earlier. The major challenges with this configuration remain the high adsorbent attrition rate and the lack of a robust scale up methodology. This design remains however conceptual with no reported experimental demonstration activities to confirm the benefits and identify the drawbacks.

2.3.1. Multistage Fluidized Bed. An important milestone in the use of fluidized bed-based reactor configuration in adsorption-based CO_2 capture was marked by the introduction of the multistage fluidized bed (Figure 8a) where the solid
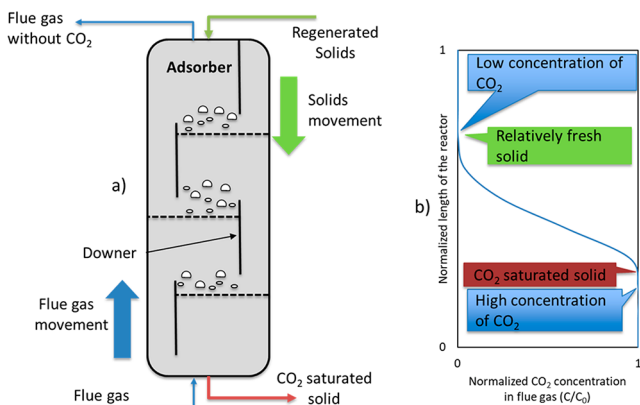

Figure 8. (a) Multistage fluidized bed and (b) gas phase CO_2 concentration variation across the reactor length.

flows counter-current to the gas, driven by the need to enhance the working adsorption capacity in these systems. Unlike the single stage fluidized bed reactor, the multistage stage reduces the overall internal back mixing to introduce a degree of plug flow behavior, bringing the same enhanced CO_2 capture benefit as packed beds.

The staging of a fluidized bed with horizontal screens was initially introduced by Varma and it was shown experimentally to reduce the axial mixing of the emulsion and bubble phases in addition to limiting the formation and growth of large bubbles.[67] This promising result prompted several studies combining the multistage fluidized bed with a counter-current adsorber[27,34,64,65,68−71] where the adsorbent particles move downward through a series of bubbling fluidized bed stages while the gas is fed at the bottom serving as a fluidizing agent in the stages. Such an arrangement enables contacting fresher adsorbent particles (lower loading of CO_2) flowing downward with the decreasing CO_2 partial pressure as the gas stream rises through the bed (see the illustration in Figure 8b). This creates a high driving force for adsorption and therefore results in improved CO_2 capture efficiency at each stage. A thermodynamic study on such a gas−solids contactor with 25 wt % of polyethyleneimine (PEI) and 25 wt % of 3-(aminopropyl)-triethoxysilane (APTES) on a porous silica support has shown that this configuration leads to significant reduction in the sorbents recirculation (indication of improved adsorption working capacity) compared to a single stage fluidized bed contactor, thus achieving a higher energy efficiency.[72] Additional higher energy saving (20%) could be achieved in these reactors by providing a lean−rich heat exchanger to recover the sensible heat from the hot regenerated rich sorbent for use to preheat the sorbent from the adsorption reactor.[70] Such solid−solid heat exchange is considerably more complex than the liquid−liquid heat exchange typical in absorption systems though. It should also be noted that the additional pressure drop caused by the distributor plate in each stage may reduce the energy saving enabled by the reduced circulation rate. Another important component in this counter-current configuration is that each stage has a down comer to allow the downward transfer of the adsorbent particles between the stages. Different down comer configurations were explored with the aim of maximizing the achievable solid flux through them.[69] All these additional internal components will increase the cost of the multistage bed relative to a conventional single stage fluidized bed.

The increased complexity of the counter-current multistage fluidized bed in the adsorber makes the hydrodynamics difficult to predict, thus imposing incremental development of such configurations requiring thorough testing for refinement and validation of the preliminary design in a cold flow model before implementation of the reactive case.[73] Such an exercise should be repeated each time the design parameters (e.g., sorbent physical size and density) are changed. This may impose large changes to the design in order to control the solid circulation rate, for example, when adsorbent materials were changed from 180 μm to 115 μm.[74] Additionally, this also brings difficulties in estimating the tube-to-bed heat transfer coefficient when heat addition or removal in the bed is required.[75] Such a challenge was encountered in the bench scale unit shown in Figure 9 designed as a TSA capture system using indirect heating through the tubes inserted in the regenerator. This study concluded that the CO_2 capture performance (especially for high feed concentration of CO_2) of the unit was limited by available heat transfer surfaces.[68]

Another multistage based configuration was studied by Veneman, where the solid (supported amine sorbents) concentration in the adsorber is maintianed very low (high void fraction greater than 90%).[76] The diluted bed allows operation of the reactor at high velocity with a low pressure
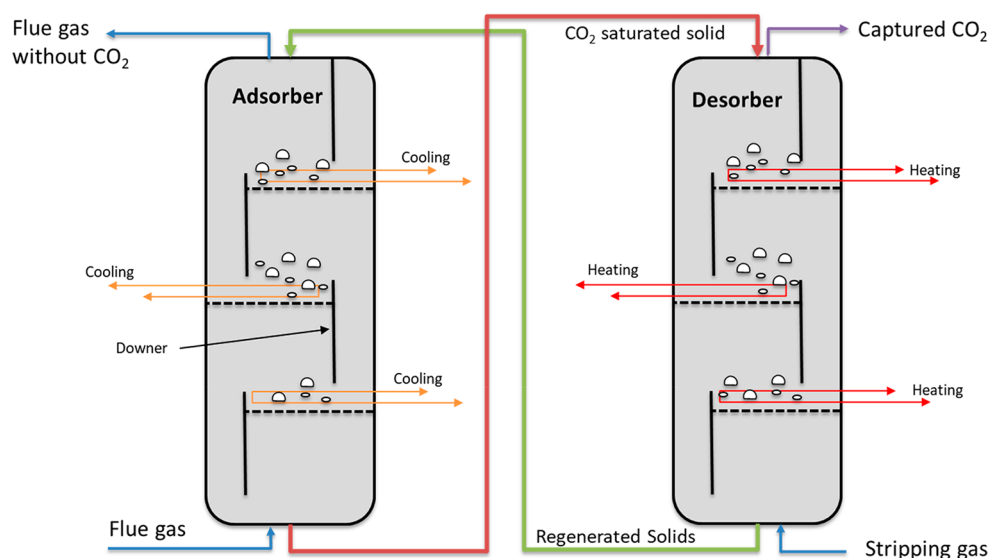
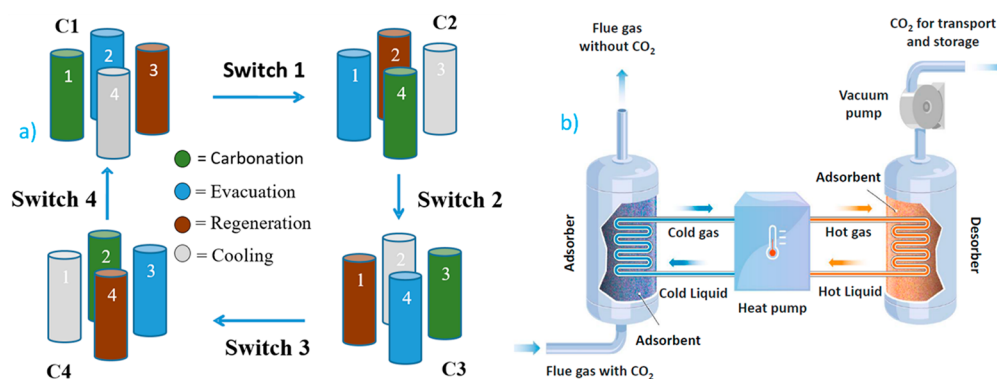**Figure 9.** Multistage fluidized bed with downer and heat exchanger.

Figure 10. SARC conceptual design: (a) a cluster of SARC reactors for continuous gas stream processing; (b) SARC working principle showing heat transfer from a reactor under adsorption to one under regeneration using a heat pump. Reprinted with permission from ref 79. Copyright 2019 Elsevier.

drop. It is also referred to as a trickle flow reactor where the contact between gas and solid is maintained in counter current mode. This configuration provides the plug flow contacting pattern between both the gas and solid phase which is desired in the adsorption process.

Despite the aforementioned challenges, the multistage fluidized bed configuration remains promising for adsorption-based CO_2 capture and a number of pilot scale studies have already been completed with such configuration.[13,34,77]

Another approach to reduce the back mixing uses multiple isolated circulating fluidized bed reactors as proposed by Zanco et al.[78] The modeling work suggested that this configuration is close to the counter-current multistage adsorption as discussed earlier. However, to achieve the desired separation and minimized energy penalty, the multistage counter current reactor configuration with a single regenerator is still preferred.

2.3.2. Transient Fluidized Bed. A transient fluidized bed reactor known as the swing adsorption reactor cluster (SARC) concept was proposed by Zaabout et al.[26] As shown in Figure 10a the SARC concept comprises a cluster of multistage fluidized bed reactors operated in bubbling/turbulent mode, where each reactor is exposed to four different process conditions (i.e., adsorption, evaluation, regeneration, and

cooling). No solids circulation is involved in this concept enabling application of a vacuum swing in the regeneration stage. An additional temperature swing is applied using a heat pump transferring heat from the exothermic adsorption (referred to as carbonation carried out at 60−80 °C) to the endothermic regeneration (carried out at 80−100 °C) as shown in Figure 10b. This is advantageous because the vacuum swing substantially reduces the extent of the temperature swing required, allowing for highly efficient heat transfer via the heat pump.

Such an arrangement brought significant reduction in the energy penalty in comparison to benchmarking technologies, specifically when the reactors were operated under the multistage configuration to reduce the extent of back mixing.[80,81] This work also provided a quantitative example of the benefits of a higher degree of plug-flow behavior on the CO_2 breakthrough curve. As shown in Figure 11, inserting three perforated plate separators in the fluidized bed greatly delayed the breakthrough of CO_2. Without separators, significant CO_2 concentrations were observed at the outlet after only one minute, but the inclusion of separators delayed this breakthrough to around 8 min, allowing a much larger fraction of the CO_2 adsorption capacity to be utilized before excessive CO_2 slippage occurs. As discussed in Figure 2, this is
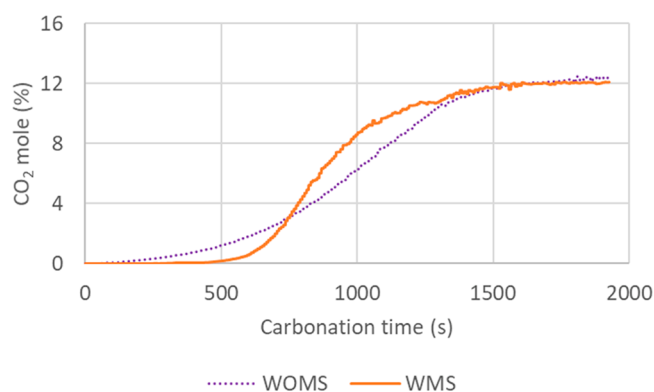
Figure 11. CO_2 breakthrough curve in the fluidized bed with separators (WMS) and without separators (WOMS). Reprinted from ref 57. Copyright 2020 American Chemical Society.

the result of the reaction front moving through the bed, first loading the bottom sorbent with CO_2, while leaving the top sorbent in a highly regenerated state to ensure complete CO_2 capture. However, in a well-mixed fluidized bed, the sorbent is uniformly loaded with CO_2, leading to CO_2 breakthrough as soon as the equilibrium CO_2 partial pressure at this uniform loading rises significantly above zero.

For these reasons, imposing a larger number of stages resulted in greater plug flow behavior, which improved the CO_2 capture efficiency and sorbent working capacity.[82] It should be noted that the number of stages to adopt should take into consideration the practicality of implementation at the industrial scale and the increase in the pressure drop associated with the additional distributor plates in each stage.[82] However, the desired reduced back mixing could be achieved by smart arrangements of the heat transfer surfaces embedded in the bed for the heat pump, without the need for placing additional distributor plates between the stages. The low energy penalty and cost of CO_2 avoidance of this concept[30] and its ease of retrofitting into existing plants, due to the use of heat and vacuum pumps that require only electricity for operation, makes this concept advantageous over the interconnected configuration with pure TSA reliant on steam for regeneration.

The merits and limitations of the various reactor configurations discussed above for CO_2 adsorption processes are summarized in Table S1 (Supporting Information).

3. MODE OF REGENERATION

Most of the discussion in the previous section was about the behavior of the different contacting systems in the adsorption. A second important segment that influences the choice of the reactor configuration is the mode of regeneration. In adsorption-based CO_2 capture, the sorbent regeneration is carried out either by changing the temperature (temperature swing; TSA), changing the pressure (vacuum/pressure swing; VSA/PSA), or changing both temperature and pressure resulting in a hybrid regeneration (VTSA/PTSA). The different regeneration modes adopted in published studies are specified in Figure 12.

It should however be noted that the choice of the regeneration mode depends mainly on the adsorbent material physical properties (specific heat, thermal conductivity, diffusivity in the material) and nature of adsorption (physi- or chemi-sorption based) that determines the shape of CO_2 isotherms returned by the adsorbent and its heat of reaction.
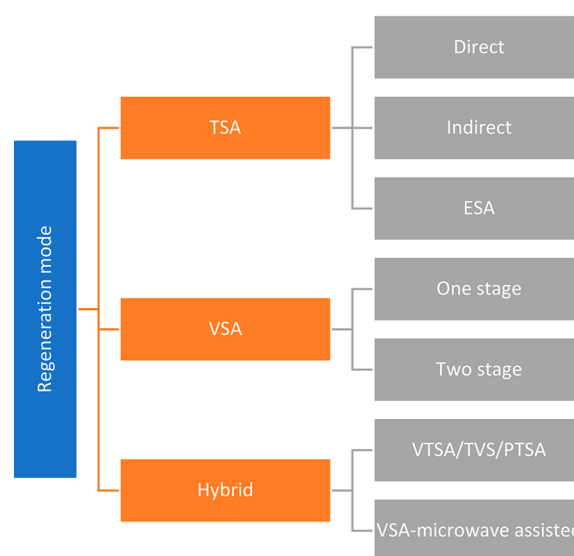

Figure 12. Various regeneration modes: TSA, temperature swing adsorption; VSA, vacuum swing adsorption; VTSA/TVS/PTSA, vacuum combine temperature swing; ESA, electric swing adsorption.

Conventional sorbents are generally classified into physisorption based sorbents that are better suited to pressure swing, and chemisorption based sorbents that are more suited to temperature swing. In this respect, the CO_2 concentration in feed flue gas from the source is also an influential parameter on the choice of the regeneration mode. CO_2 concentration varies widely over different flue gas sources (4−6% in a natural gas power plant, 12−15% in a coal power plant, and higher in cement and biogas plants in the range of 20 and 45%, respectively). When a high CO_2 capture rate and maximal working capacity are targeted, the chemisorption-based sorbents (with sharper isotherms) combined with a TSA regeneration mode are more suited to sources with low CO_2 concentration, while the physisorption-based sorbents combined with a VSA regeneration mode are more suited to flue gases with high CO_2 concentration.[83] As highlighted earlier, the sorbent physical properties such as the specific heat and thermal conductivity can have a big influence on the different regeneration modes; TSA, VTSA, and ESA (electrothermal swing adsorption). The pros and cons of each regeneration mode, their optimal reactor configuration, and operating conditions will be presented and discussed in this section.

3.1. Temperature Swing Adsorption (TSA). TSA is either carried out either in direct mode, in which a hot stream (steam, CO_2) is used as a purge gas in direct contact with the adsorbent particles, or in indirect mode, in which a heat exchanger is used to provide the required heat for regeneration. The regeneration energy requirement in TSA comprises sensible heat required to heat the adsorbent to the target regeneration temperature, reaction heat to drive the endothermic desorption reaction, and latent heat of vaporization if the sorbent has adsorbed water as well.[84] This regeneration mode has been applied in the different reactor configurations with pros and cons as will be discussed in what follows. The various research works have focused on reducing the overall heat requirement in this TSA mode through sorbent development with improved adsorption capacity and lower specific heat capacity, but more importantly, with lower heat of reaction. Others focused on reducing the energy requirement through heat integration in the process.[85,86]

3.1.1. Indirect Temperature Swing Adsorption (Indirect TSA).
Indirect TSA is done by heating or cooling the adsorbent by means of a heat exchanger in the reactor as illustrated in Figure 13. This heating mode was widely used in early works in
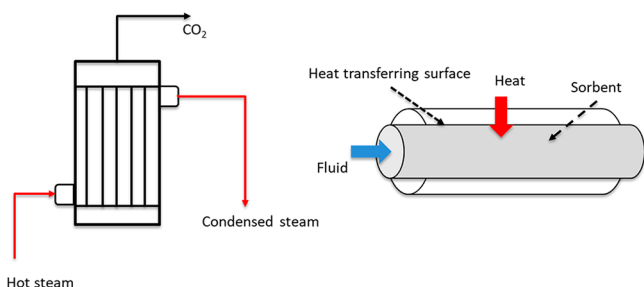

Figure 13. Indirect TSA.

a fixed bed configuration due to its simplicity and the achieved high CO_2 purity if no purging is applied through the bed.[87] Its major drawback remains the very low tube-to-bed heat transfer coefficient in a fixed bed configuration making such a heating mode inherently unfeasible in a conventional fixed bed. This is well illustrated in Figure 14 showing the very low heat transfer
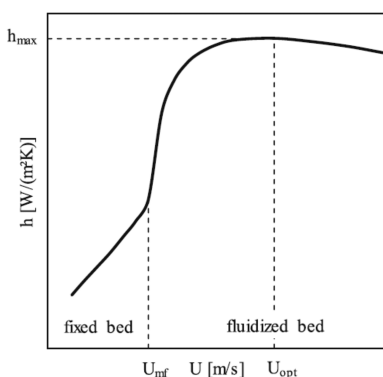

Figure 14. Heat transfer coefficient with velocity for Geldart Type B particles. Reprinted with permission from ref 58. Copyright 2018 Elsevier.

coefficient in the fixed bed, although some gas purge could improve the convective heat transfer,[88,89] but remains limited though against the values achieved in a fluidized bed that could be up to an order of magnitude larger (Figure 14). This advantage makes the fluidized bed favored for indirect TSA, not only for supplying heat in the regeneration, but also for improving the control of temperature in the adsorption to maximize the working adsorption capacity and improve the capture efficiency.[26,38,68,70] Nevertheless, a good example of a successful experience of indirect heating in a fixed bed was demonstrated in a hollow polymeric fiber that enabled cycling fast TSA in the reactor by pumping hot/cold fluid through the hollow structure[45] discussed earlier. With the rapid advances made in 3D printing, making a smartly structured bed with embedded heat transfer surfaces could become a feasible and viable option to implement a TSA regeneration mode in a fixed bed.

3.1.2. Direct Temperature Swing Adsorption (Direct TSA).
Direct TSA is the mode of regeneration in which the adsorbent is heated directly by means of a hot gas stream, preferably steam (Figure 15). This TSA mode has a much better heating
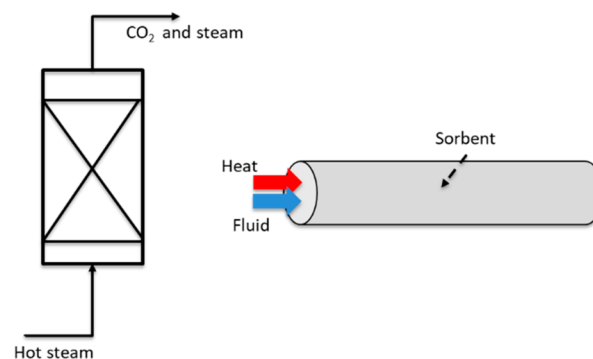

Figure 15. Direct TSA.

rate than the indirect one, thus decreasing the heating time. The main advantage remains the capability of achieving sufficiently high purity of CO_2 with this mode explaining the widespread application of such an option to the different reactor configurations discussed in the previous section. The addition of steam reduces the CO_2 concentration which further increases the driving force for desorption, reducing the size of the temperature swing required. However, this also has some drawbacks such as (i) the need for an additional unit operation for water removal from the captured CO_2 before being compressed for transportation and storage; (ii) the used sorbent should tolerate the presence of water and in many cases a drying step is needed before starting a new cycle (this depends on the nature of sorbent and operating conditions in the regeneration step). Such additional steps increase the process complexity, the energy penalty, and costs.

To address the aforementioned challenges, direct heating of a fixed bed reactor by a recovered hot CO_2 product gas (purge) was studied by Ntiamoah et al.[88] The process comprises a basic three-step cycle of (i) adsorption, (ii) hot gas purge where the regeneration takes place, and (iii) cooling by N_2/ air. Their study with zeolite NaUSY adsorbent indicated a specific (thermal) energy consumption as high as 4.5 MJ/kg of CO_2 at a temperature of 250 °C to yield CO_2 purities greater than 91% and CO_2 recoveries of only 83.6%. This purging with hot CO_2 gas removed the need for additional process units associated with the use of steam but it reduced the driving force for desorption, thus imposing the need for higher regeneration temperature and resulting in higher thermal energy demand. The source of this hot CO_2 stream is also an important consideration.

3.1.3. Electrothermal Swing Adsorption (ESA).
Electrothermal swing adsorption (ESA) is considered as another potential mode to reduce the energy penalty of adsorption-based CO_2 capture. The ESA mode is conducted by heating the adsorbent beds by means of Joule heating inside the particles by passing an electric current.[90] This in-situ heating of the particles enables a fast heat transfer rate in comparison to a conventional TSA mode and also provides better desorption kinetics.[91] The essential feature for the adsorbent to work under an ESA mode is its electrical conductivity. Activated carbon fiber was considered as a potential sorbent that work in the ESA mode of regeneration.[25] The ESA mode is however applicable only in a fixed bed configuration where the long cooling time counteracts the advantage of the in-situ fast heating, thus hindering the prospects of such a mode in implementation at the industrial scale. From an economic point of view, the main drawback is that electrical energy is

many times more costly than the low-grade heat typically used for regeneration in the TSA processes.

3.2. Vacuum Swing Adsorption (VSA). Another widely used regeneration approach is by varying the pressure, commonly known as vacuum swing adsorption (VSA) or pressure swing adsorption (PSA). In postcombustion CO_2, only VSA makes sense as the PSA requires pressurizing the incoming large quantities of the flue gas from the source making the process uneconomical. VSA is predominantly applied to a fixed bed reactor configuration with physisorption based sorbents such as zeolites or activated carbon.[10,11] However, as emphasized in the previous section, the VSA regeneration mode can achieve a very short cycle, but a low pressure drop across the bed in adsorption is a prerequisite, thus favoring the structured advanced fixed bed. Additionally, a high CO_2 capture rate requires deep vacuum levels making this regeneration mode suitable only for industrial applications producing flue gases with high CO_2 partial pressure.[92,93]

The first proposal of vacuum swing adsorption (VSA) regeneration mode comprises four steps: pressurization with feed gas, adsorption, forward blowdown, and reverse evacuation. As shown in Figure 16, the flue gas is fed to the
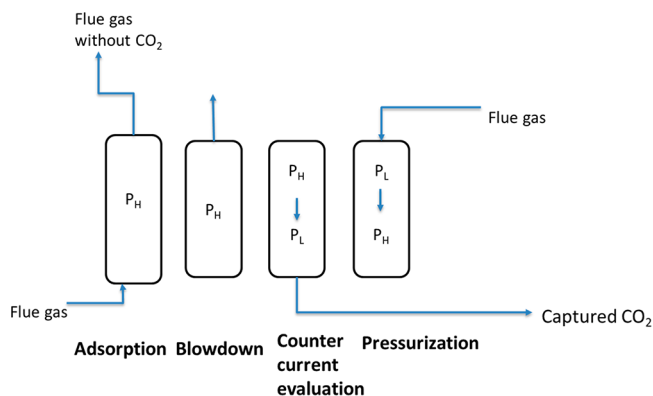

Figure 16. Basic VSA process (P_H, high pressure; P_L, low pressure).

adsorption step where CO_2 is adsorbed close to atmospheric pressure, then the following blowdown step is used to remove the accumulated N_2 from the reactor by using a slight vacuum before starting the following regeneration step at higher vacuum levels. The blowdown step improves the purity of CO_2 but can also reduce its recovery as some CO_2 can be lost during this step. Finally, the reactor is pressurized again by the flue gas preparing for a new cycle. These basic steps are simple in operations, but either suffers from low CO_2 purity or recovery. An experimental study indicated that the VSA mode can lead to high purity CO_2 (around 99% purity) but is limited with a low CO_2 recovery of 85%.[89] An interesting approach that was proposed to improve the CO_2 recovery was by recycling some of the CO_2 product to the blowdown step (known as heavy reflux or high-pressure rinse[94]). The presence of the recycled CO_2 improves the CO_2 purity as it displaces the accumulated nitrogen in the reactor, and it also increases the loading of the CO_2 in the column during blowdown. Such an approach has led to higher CO_2 recovery (98.7%) and high purity (98.7%) during the regeneration step. Another improvement in the conventional four-step cycle in terms of CO_2 recovery was demonstrated in a pilot scale VSA system using Zeochem zeolite 13X.[93] The authors proposed to purge the light product (flue gas without CO_2) for pressurization (LPP)

in the counter-current direction. This avoids the slippage of CO_2 present in the reactor after the evaluation step. The CO_2 recovery remained below 90%, though.

Another tactic to improve the recovery with high purity is to use a two-stage VPSA process.[95] This is a method to make pressure swing applicable to flue gases with lower CO_2 concentrations for which a single step VSA cannot concentrate the CO_2 sufficiently. Wang et al.[44] simulated this two-stage VPSA unit for a coal power plant flue gas, and the results indicated that CO_2 purity can be increased to 65% in first stage and then to 96% in the second stage while the achieved CO_2 recovery increases to 93.35%. Such a two-stage VPSA was experimentally demonstrated at a pilot-scale installed in an existing coal-fired power plant in China by Wang et al.[44] In addition to the two-step VPSA process units, a dehumidifying unit was used in the pilot plant to remove the water vapor in the desulfurized flue gas by alumina adsorbent before being fed to the CO_2 capture unit. This study shed light on the issues of the existence of water in the flue gas in the VSA CO_2 capture mode given that such a system operates at low temperatures that may result in substantial watering of the sorbent if it is not removed in a preprocessing step. An example of a VSA CO_2 capture process with a water removal preprocessing step is the dual-adsorbent, four-step vacuum swing adsorption (VSA) process with silica gel and zeolite 13X packed separately in two beds as illustrated in Figure 17.[96] Designing such a system
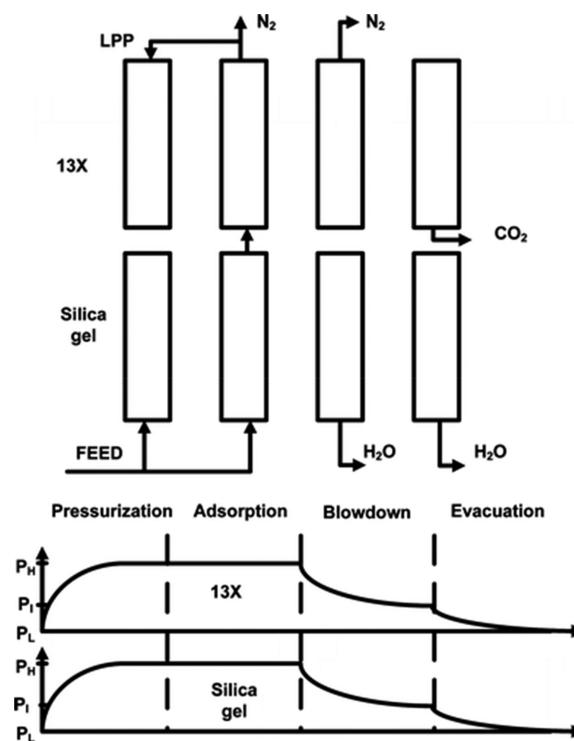
Figure 17. Dual-adsorbent, two-bed vacuum swing adsorption process for CO_2 capture from wet flue gas. Reprinted from ref 96. Copyright 2014 American Chemical Society.

requires careful sizing of each reactor taking into consideration the isotherms of water and CO_2 adsorption on their respective sorbents. An increase in the energy penalty should be expected due to the additional pressure drop imposed by the additional reactor and the vacuum pump consumption for water removal.[97]

Figure 18. A simplistic illustration of the extent of complexity of the operating strategy of the different reactor configurations and operating mode.

Overall, a VSA regeneration looks to be an attractive option for reducing the cycle time in adsorption-based CO_2 capture to maximize the productivity of the process. The main hurdle remains the high pressure drop encountered in a conventional fixed bed making it hard to achieve the low vacuum values required for achieving reasonable CO_2 recovery within sufficiently short cycle times. A design for a rapid vacuum pressure swing adsorption (RVPSA) based on a conventional fixed bed was recently proposed to capture CO_2 with 95%+ CO_2 purity and 90%+ CO_2 recovery from an existing 10 MWth biomass-fueled CHP plant.[17] They considered a two-stage VSA with two parallel beds in the first VSA stage in order to split the total flue gas feed and reduce the overall pressure drop. Achieving the vacuum pressure of 7.5 kPa in seconds (18 s was considered in the entire VSA cycle) will need a specially designed large vacuum pump that could be a practical challenge at the industrial scale. Moreover, the operation of a VSA based system on large scale CO_2 sources, such as a coal power plant, requires a large number of reactors (73 and 23 for first and second PSA stage, respectively) that have to operate in an automated manner to achieve continuous processing of the flue gas from the plant.[31] Such a case imposes designing a complex operation strategy to automate the different reactors cycling the different process steps.

3.3. Hybrid Regeneration Approaches. Several attempts were made to overcome the shortcoming of the unpracticality of reaching deep vacuums in large scale VSA to achieve the target high CO_2 recovery, by using hybrid regeneration modes such as VTSA and steam aided vacuum swing.[98−100] One of the first successful demonstration of 2000 h was completed at a pilot scale fixed bed reactor by Ishibashi et al.[98] for a two-stage process comprising a first stage PTSA and a second stage of PSA. Recently, Wurzbacher et al.[101] evaluated the effect of moisture in humid air on the working capacity of the diamine-functionalized commercial silica gel sorbent when operated under VTSA mode. Their study indicated that under dry conditions, desorption pressures above 100 mbar lead to working capacities below 0.03 mmol g^{-1}, while under humid conditions (40% relative humidity) during adsorption working capacities above 0.2 mmol g^{-1} at 150 mbar were achieved. This increase was the result of the dilution effect of steam during the regeneration mode creating a larger driving force for desorption of CO_2. Later, Fujiki et al.[102] proposed a low temperature steam-aided vacuum swing adsorption (SA-VSA) process for regeneration of an amine-based solid sorbent. This dilution with steam under vacuum resulted in a similar effect of a direct heating that improved the working capacity. The experimental study was conducted in a lab scale set up and it has shown a high CO_2 purity (>98%) and recovery rate (>93%) against a recovery of 45% with only vacuum (VSA). It should be noted, however, that the improvement in the desorption driving force created by steam does not lead to the expected large saving in the energy penalty because the vacuum pump has to extract a larger amount of gas that increases its electricity consumption.[103]

Another nonconventional hybrid VTSA approach used a microwave-assisted vacuum swing over 13X zeolite by Webley et al.[16] They showed that a brief exposure to microwave radiation improved the speed of CO_2 and water desorption at reasonably higher achievable vacuum levels. They suggested that this positive effect of microwave could lead to the reduction in overall lower energy penalty. The main challenge with this concept remains large-scale application and a safety consideration with the microwave. The cost of using electrical energy for regeneration is another important challenge.

The swing adsorption reactor cluster (SARC) that was proposed by Zaabout et al.[26] is another VTSA based hybrid system that has a high potential to bring substantial energy savings. This concept implements heat integration between adsorption (referred as carbonation) and regeneration reactors by means of a heat pump. This is also combined with a practically achievable vacuum swing to minimize the temperature difference between the adsorption and regeneration, thus maximizing the coefficient of performance of the heat pump and reducing the overall energy penalty. Such a configuration achieved competitive energy penalties against benchmarking technologies, both for coal power plants and cement[80,81] targeting a CO_2 recovery of 90% and a CO_2 purity of 96%. Experimental demonstration tests using polyethyleneimine sorbent have proven the feasibility of achieving a small temperature difference between adsorption and regeneration at a moderate vacuum of 100 mbar.[99]

Several other VTSA hybrid studies have followed. A heat-integrated vacuum and temperature swing adsorption process in a multibed reactor for a low temperature adsorption process was studied by Plaza et al. to capture at least 85% of the CO_2.[104] Waste heat from a power plant will be used to regenerate the sorbent, but the fixed bed configuration used may impose serious heat transfer limitations as discussed earlier. Zhao et al. also evaluated a monolith reactor using a hybrid process of ESA combined with vacuum swing adsorption (VESA). They reported that the total specific energy with this approach can be lower than ESA alone, but still higher than VSA.[25]

The advantages and limitations of various regenerations modes are summarized in Table S2 (Supporting Information).

4. REACTOR OPERATION STRATEGIES

The combination of reactor configuration and regeneration mode selected for the adsorption-based CO_2 capture technology determines the operation strategy of the adsorption plant. The following operation strategies could be foreseen for the different reactor configurations. The relative complexity of the operation strategies of different configurations is illustrated in Figure 18.

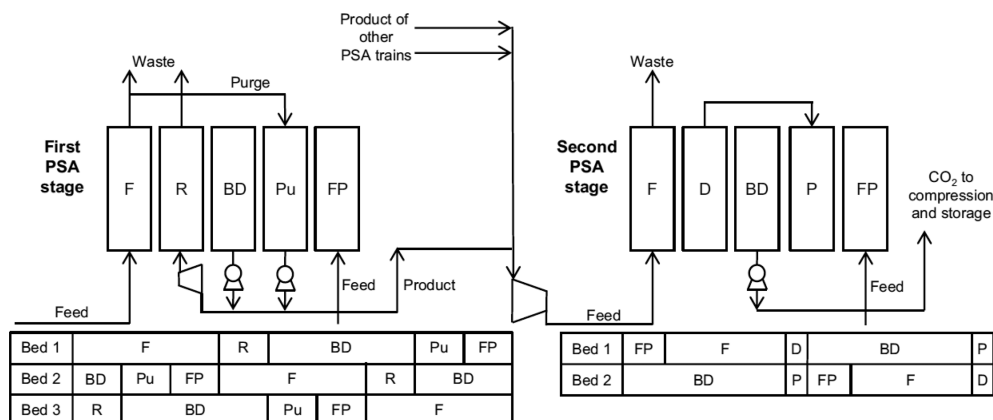**4.1. Fixed Bed.** A cluster of reactors is needed for continuous gas processing using the fixed bed reactor

Figure 19. Two-stage VPSA process for postcombustion CO_2 capture from a coal plant. Representation of the sequence of steps undergone by a single column in the first and second PSA stage. Feed (F), rinse (R), depressurization (D), blowdown (BD), purge (Pu), pressurization (P), null (N) where the column is left idle, (FP) feed pressurization. Reprinted with permission from ref 31. Copyright 2015 Elsevier.

configuration, regardless of the nature of the regeneration mode adopted. The number of reactors in the cluster depends on the number of steps in the cycle and the time of the steps. If a VSA regeneration mode is adopted, achieving high CO_2 recovery with most flue gases requires a second processing stage, thus increasing the number of reactors in the plant (Figure 19 shows an example of an operating strategy for continuous feed processing using a two-stage VSA fixed bed configuration). A total of 96 reactors were needed for continuous flue gas processing from a coal-based power plant using a two-stage VSA system.[31] This complicates the operation strategy, involving hundreds of valves and complex piping to accomplish the different process steps needed to complete the CO_2 capture cycle.

4.2. Rotary Bed. This reactor configuration can also have a simple operation strategy given that only the speed of rotation and the areas of the different process steps must be specified correctly (these two parameters are mainly affected by sorbent isotherms, physical properties, and reaction kinetics). TSA could be the simplest regeneration mode to adopt although the cooling and heating times would be long, imposing a large reactor footprint. Implementing additional steps to improve the separation efficiency in this reactor configuration brings additional complexities to the operation strategy. When a VTSA regeneration mode is adopted as proposed in ref 18, the operation strategy will not be affected given that the only difference is that, in the regeneration zone, a vacuum will be applied in addition to the temperature swing. If an efficient sealing system is implemented to minimize the gas leakage between the zones operating at different pressures, applying a vacuum in addition to the temperature swing could be beneficial for the process as it will reduce the heating and cooling times due to the reduced temperature swing. However, the extent of complexity in the operation strategy in this case will depend on the level of complexity involved in the sealing solution used for improving the separation efficiency.

4.3. Interconnected Fluidized Bed Reactors and Moving Bed. The only reported regeneration mode for these two configurations is TSA (Figure 4 and Figure 7) because a pressure swing would create substantial operational challenges in such an interconnected system. Such a system operates like conventional absorption-based CO_2 capture technologies, continuously feeding the flue gas to the adsorber operating at low temperature while the regeneration occurs in

a second reactor operating at higher temperature with continuous solids sorbent circulation between the two reactors.[23,60,105] Indirect heating using an inbuilt heat exchanger can be efficient in circulating fluidized beds, given the high tube-to-bed heat transfer coefficient.[58] This option can also ensure efficient use of waste heat that could be available in the host plant, thus further minimizing the energy penalty of CO_2 capture. As for the moving bed, direct heating with steam or CO_2 seems to be the most efficient option due to the low tube-to-bed heat transfer.[20,52]

4.4. Dense Fluidized Bed Using the Switching Concept. Similar to a fixed bed, a cluster of reactors are needed for continuous flue gas processing. A study on the SARC concept with PEI sorbent involved the use of 24 reactors for continuous flue gas processing, although this number could be reduced by increasing the reactor size. This is substantially lower than the fixed bed two-stage VSA case and requires no integration with a second processing stage, but it would still require dozens of valves and complex piping to complete the different steps.[26] Additional complexity should be expected when the heat pump is used for transferring heat from the reactors under adsorption to those under the regeneration and when heat is exchanged between the reactors under cooling and the others under evacuation/heating. The heat pump working fluid will have to be continuously redirected between different reactors cycling through the transient operating strategy. Applying an additional vacuum in the regeneration (jointly with the temperature swing) is not expected to add any complexities to the operation strategy.

5. PROCESS INTEGRATION AND ENERGY REQUIREMENT

One of the most important aspects in any CO_2 capture technology is the energy penalty involved. Various efforts have been made to reduce this energy penalty, mainly by sorbent development, but also via more efficient process integration. Process integration options include the use of solar heat in PTSA systems,[106] the combination of high and low temperature sorbents where heat released from the adsorption of the high temperature sorbent regenerates the low temperature sorbent,[107,108] the use of a heat pump to transfer heat from adsorption to regeneration,[26,109] and close heat integration between three moving bed reactors.[21] Others considered

Table 2. Energy Requirement for Various Modes of Regeneration and Reactor Configuration

Regeneration mode	Feed CO_2 partial pressure (%)	Reactor	CO_2 purity (%)	CO_2 recovery (%)	Regeneration energy (MJ/kg)	Productivity (kg/($m^3_{adsorbent}$ h))	ref
Two-stage VSA (Coal)	16.5	Fixed	95.6	90.2	2.44[b]	65.2[b]	Wang et al.[44]
VSA-two-stage	15	Fixed	96.5	93.4	2.64[a,c]	20.9[c]	Wang et al.[11]
TSA-direct- CO_2	15	Fixed	91.0	83.6	4.50[c]	32.9[c]	Ntiamoah et al.[88]
TSA- heat integration for sensible heat recovery	15	Simulated moving			2.53[c]		Jung et al.[110]
VPSA -three-bed seven-step	15	Fixed	85	79	2.37[b]	83.7[b]	Liu et al.[32]
TSA-indirect	12	Fixed	0.97	0.77	4.07[c]	46.5[c]	Joss et al.[111]
TSA- indirect with steam purge	14	Fixed-monolith	95.6	85.4	3.59[c]	228.4[c]	Plaza et al.[112]
TSA-steam + CO_2							
Heat integration in adsorber and desorber (Coal)	13.8	Circulating bubbling fluidized			2.49[c]	42[c]	Zhang et al.[113]
TSA-indirect with vacuum and heat pump- CO_2 purge	13.4	Multistage fluidized	96.0	90.0	2.8[a,c]	68.3[b]	Zaabout et al.[26,57]
Thermally couple column- TSA (coal)	13.2	Fast fluidized bed		85.0	1.73[c]		Vogtenhuber et al.[108]
VSA-membrane	12.6	Fixed bed membrane	95.0		4.1[b]	10.8[b]	Warmuzinski et al.[100]
TSA-indirect	12.5	Fixed	99.0	79.0			Tlili et al.[89]
VPSA-two-stage	10	Fixed	95.3	74.4	3.61[a,c]	26.8[c]	Shen et al.[10]
TSA-indirect with purge- optimized	10	Fixed	95.0	81.0	3.23[c]	43.1[c]	Clausse et al.[114]
TSA-indirect with steam (without heat integration)	5.15	Moving	95.1	96.0	2.21[c]		Mondino et al.[38]
TSA-indirect with steam (With heat integration)	5.15	Moving	95.1	96.0	1.46[c]		Mondino et al.[38]
TSA-steam + CO_2 Heat integration in adsorber and desorber (natural gas)	4.1	Circulating bubbling fluidized			2.54[c]	42[c]	Zhang et al.[113]

[a]Electric efficiency of 20% is assumed in the conversion of electrical energy to thermal energy for regeneration, excluding electricity required for compression and pressure drop. [b]Experimental measurement. [c]Model prediction.

combining a single stage VSA with a CO_2 membrane system to reduce the energy penalty.[100]

A summary of the thermal energy requirement for sorbent regeneration of various concepts is presented in Table 2. To allow for reasonable comparisons between different regeneration modes, electrical energy consumption is multiplied by a factor of 5 to account for the fact that low-grade heat would normally be used for sorbent regeneration. This heat is usually supplied by extracting low pressure steam from the steam cycle that would otherwise only be able to produce power at about 20% efficiency. In other words, if a VSA process used 0.5 MJ/kg of electrical energy for sorbent regeneration, a TSA process would be able to use about 2.5 MJ/kg of low grade heat extracted from the power cycle, resulting in the same 0.5 MJ/kg loss in electricity output. In addition, power consumption for CO_2 compression and flue gas blowers are subtracted where necessary to focus only on the energy requirement for capturing CO_2 (mainly regeneration enthalpy and sensible heat). It is worth noting that the comparison in Table 2 is rather for illustration from published data of the different reactor configurations and a more reliable comparison requires carrying out a standardized techno-economic assessment study.

Table 2 shows considerable variation in the reported regeneration energy requirements for different adsorption-based CO_2 capture processes. Good process performance appears to fall in the 2−2.5 MJ/kg range for TSA processes requiring only heat as well as VSA and heat pump processes with electricity consumption converted to low-grade heat with a 5× multiplication factor. For perspective, this is in line with

advanced solvents for absorption-based CO_2 capture and well below conventional MEA (>4 MJ/kg).[115]

Two values below 2 MJ/kg are observed. For the thermally coupled process,[108] considerable savings are achieved by cascading the heat down three different adsorption processes operating at different temperatures. However, the heat must be introduced into the process at a higher temperature, requiring the extraction of higher-grade heat from the power plant. For this reason, the energy penalty of this concept was 9.5−11.5%-points, which is comparable to that for MEA. The moving bed TSA with heat integration[38] has a low thermal energy requirement, but includes a considerable additional energy penalty for water removal from the flue gas, resulting in an energy penalty similar to that of an MEA benchmark. The heat integration in the moving bed is also likely to be expensive, given the relatively low heat transfer coefficient.

Another important key performance indicator for bench-marking the different reactor configurations is the productivity that measures the amount of captured CO_2 per unit of sorbent volume and time. As can be seen in Table 2, the different studies reported a wide range of values with the best performance achieved by the monolithic reactor configuration.[112] This resulted from the short cycle time enabled by this configuration combined with the fast reaction kinetics of the chosen sorbent and the low mass transfer resistance. The TSA-indirect with vacuum and heat pump-CO_2 configuration has the potential to maximize the productivity despite the low value of 68.3 kg-CO_2/(m^3 h) found in the experimental study.[57] It was shown in this study that increasing the flow rate to achieve the 1 m/s superficial gas velocity used in the techno-

Table 3. Demonstration Status of Various Concepts

Reactor	Regeneration mode	Scale	Adsorbent	Sector	ref
Fluidized	TSA	Pilot, 200 t/d	Dry sorbent (K or Na)	Coal	Park et al.[119]
Moving bed (KCC process)	TSA	Pilot, 3.5 t/d	Amine functionalized sorbent	Coal	Okumura et al.[53]
Fixed	VSA (2 stage)	Pilot, 1.6 t/d	Stage 1, zeolite 13X APG; stage 2, activated carbon	Coal	Wang et al.[44]
Moving bed (SRI)	TSA	Pilot, 0.56 t/d	Carbon	Coal	Hornbostel et al.[51]
Fluidized bed (RTI)	TSA	Pilot, 0.15 t/d	PEI based sorbent	Cement	Nelson et al.[13]
Fixed	VSA - LPP	Pilot	Zeochem zeolite 13X	Coal	Krishnamurthy et al.[93]
Multistage fluidized	TSA-N$_2$	Bench scale,- 35 kg/d	PEI/SiO$_2$	Gas	Schöny, G. et al.[68]
Multistage fluidized	VTSA	Lab scale, 24 kg/d	PEI/SiO$_2$	Coal	Dhoke et al.[57]
Fixed + membrane	VSA-membrane	Lab scale	Zeolite molecular sieve 13X Grace	Coal	Warmuzinski et al.[100]
Sound assisted fluidized		Lab scale	Fine activated carbon	Gas/Coal	Raganati. et al.[63]
Fluidized bed	TSA	Pilot scale	Amine based	Coal	Sjostrom et al.[118]

economic study[26] (against 0.19 m/s in experiment) was restricted by the experimental setup (elutriation of particles), not by the kinetics of the reactions nor by the mass transfer. This implies that a productivity of ~360 kg-CO$_2$/(m^3 h) could be achieved by such a reactor configuration.

Finally, it should be noted that VSA and heat pump processes will appear considerably more attractive in industrial processes where an abundance of low-grade heat is not available. In this case, the conversion to thermal energy should be done assuming typical thermal power plant performance (40−60%), roughly halving the converted thermal requirements of these processes reported in Table 2. For example, the SARC process showed only marginal gains over typical MEA energy penalties in CO$_2$ capture from a coal power plant,[26,81] but strongly outperformed MEA in a cement plant where heat for the MEA process had to be generated in a dedicated boiler.[80] In such industrial applications, CO$_2$ capture concepts relying on electric energy show the largest potential relative to conventional TSA benchmarks.

6. DEMONSTRATION STATUS OF RESEARCH

ADA has been working on developing solid sorbents and fluidized bed technology to efficiently capture CO$_2$ from power plant flue gas for over a decade. Sorbent screened was completed at 1 kWe scale to demonstrate the ADA's solid sorbent CO$_2$ capture process.[116] Later they designed and completed the construction of a first-of-its-kind, 1 MWe scale, CO$_2$ capture pilot facility in 2014.[117] The reactor configuration comprises a three-stage fluidized-bed adsorber integrated with a single-stage fluidized-bed regenerator. A recent pilot plant study by ADA indicated 90% CO$_2$ capture with the TSA mode of regeneration and reported two main operational challenges with the pilot plant: the first associated with handling of the sorbent at operating high temperature and the second associated with the preloading of the sorbent in the transport line between the regenerator and adsorber that reduced the working capacity.[118]

A brief summary about the other low temperature adsorption concepts demonstrated at various scales in the past are presented in Table 3. Few of the concepts have been demonstrated at the pilot scale while the majority of the studies were conducted at lab scale. Capturing CO$_2$ from coal power plants remains the dominant sector that was targeted by the demonstration studies showing a clear need for extension to other CO$_2$ intensive sectors such as cement, metal industry, and biofuel.

7. DISCUSSION

The different sections presented above highlighted the clear fundamental difference in the behavior of the different reactor configurations in the adsorption that affect the performance in terms of CO$_2$ recovery efficiency and purity in addition to the overall footprint. Another important factor that affects the choice of the reactor configuration is the regeneration mode that is in general dictated by the nature of the sorbent selected and the initial CO$_2$ partial pressure in the flue gas. The combination of reactor configuration and regeneration mode are the key elements that impact the footprint, operation strategy, the flexibility for part load operation when needed, and the overall energy penalty imposed for CO$_2$ capture. All these factors will influence the techno-economic attractiveness of the different configurations for different applications. The scattered energy/heat duty values reported in the limited existing number of studies on techno-economic assessment of adsorption-based CO$_2$ capture (Table 2) are far from being systematic enough to provide any firm conclusions on the link of the reactor configuration and regeneration mode to the overall performance of the technology. It can also be observed that there is a big lack of economic assessment studies with this technology giving clear estimates on the cost of CO$_2$ avoidance imposed by such technology.

Retrofitting to existing plants is also an important factor when considering the costs of CO$_2$ capture technology. In principle, regeneration modes that require only electricity are best suited to retrofitting (e.g., VSA, ESA, microwave assisted and SARC using a combination of a heat and vacuum pumps), while the others that require heat for TSA (both direct and indirect) impose constraints to retrofitting, dictating sourcing heat from the plant through complex integration or involving building boilers and associated infrastructure. The interaction of the different factors and their impact on the different reactor configurations are qualitatively discussed and evaluated in Table 4.

Future Research Needs. From the above discussion, it can be clearly concluded that there is an urgent need for studies that standardize the assumptions for thermodynamic and economic assessments of the different configurations combined with the different regeneration modes. Such studies will bring clear and accurate comparisons of the cost of CO$_2$ avoidance involved with the different combinations. It can however be speculated that for a specific initial CO$_2$ partial pressure in the feed flue gas, the different reactor configurations and regeneration modes will perform differently

Table 4. A Qualitative Evaluation of the Different Factors Impacting the Choice of the Reactor Configuration for Adsorption-Based CO_2 Capture[a]

	Adsorption	Regeneration	CO_2 recovery and purity	Operation strategy	Footprint	Flexible load operation
Conventional fixed bed	(−) The high pressure drop results in a high energy penalty or footprint. Heat and reaction fronts limit the working adsorption capacity and apply thermal stresses on the sorbent.	(O) VSA is the most suited because low heat transfer rates make TSA impractical. However, the high pressure drop imposes a long regeneration time.	(−) Complex two-stage VPSA configurations are required for good CO_2 recovery and purity from most flue gases.	(−) Complex control of multiple transiently operated reactors, particularly when a two-stage process is involved.	(−) High pressure drop both in adsorption and regeneration limit gas velocities, increasing the footprint.	(+) Possible due to the stand-alone reactors. Beds could also be regenerated faster when electricity is cheap and carbonated later for greater flexibility.
Structured bed	(+) Smartly engineered structures can reduce heat and mass transfer limitations and pressure drop. Sorbents with fast kinetics should be used though.	(+) Similar VSA suitability to fixed beds, but indirect TSA is also possible if the structure enables flowing a fluid through the bores for heat addition or removal.	(−) Similar to the conventional fixed bed.	(O) Indirect TSA could result in simpler one-stage operation strategy, but given that the cluster would still be needed, the operation strategy will be relatively complex.	(O) A lower pressure drop can allow for substantially faster gas flows and thus smaller footprints than fixed beds.	(O) Similar to fixed bed, although the higher cost of the structure could make storing beds of regenerated sorbent during low electricity prices uneconomical.
Moving bed	(O) Counter-current operation maximizes the working adsorption capacity, but this is counteracted by the low heat transfer coefficient that may lead to heat build-up in the adsorption.	(O) Direct heating with steam or CO_2 remains the most feasible mode due to the low tube-to-bed heat transfer coefficient. A drying step should be foreseen when steam is used as a heating agent.	(+) High if leakage between the chambers can be minimized.	(+) Simple steady state operation strategy involving solids circulation to the different chambers of the reactor	(−) Counter-current operation involves a large footprint due to the limitation on the gas velocity to avoid fluidization of the falling solid sorbent.	(O) Varying the feed rate to the reactor could enable flexible load operation. This will however be affected by the mechanism used for solids circulation.
Rotary bed	(−) Similar drawbacks as fixed bed, although smartly engineered structed beds can reduce these challenges. Cooling the rotating system in TSA is complex.	(O) Direct heating with steam or CO2 is the best option, but this would involve a drying step. VSA requires a sophisticated sealing system.	(−) Acceptable CO_2 recovery and purity require implementing purge zones between the adsorption and regeneration feeding substantial amounts of steam. Complex sealing in VSA mode.	(+) A relatively simple steady state operation strategy if the rotation speed and the zone of the different steps of the capture cycle are designed properly	(O) If smartly engineered structures are used to remove the heat and mass transfer limitation, a reasonable footprint could be achieved	(O) Flexible load operation is possible by varying the feed rate to the reactor and adjusting the rotational speed.
Circulating fluidized bed	(−) The high mixing results in fast CO_2 breakthrough and low working adsorption capacity, requiring large sorbent recirculation rates.	(+) Both direct and indirect heating are possible. Indirect heating is ideal when low-grade heat is available (e.g., from a power plant).	(−) The high mixing requires very long risers in the adsorption and involves larger circulation rate to achieve acceptable CO_2 recovery.	(+) A simple steady state operation strategy due to the use of only two reactors with the sorbent circulating between them.	(+) Small footprint due to the high flue gas velocities permitted by fluidized bed risers.	(−) The range of flue gas feed rates over which stable sorbent circulation will be maintained will be limited.
Counter-current multistage circulating fluidized bed	(+) Counter-current behavior maximizes the working adsorption capacity. The advantage of good heat transfer of fluidized bed is maintained for effective TSA operation.	(+) Similar to conventional circulating fluidized bed.	(+) High CO_2 recovery and purity could be achieved with this configuration if gas leakage between reactors is effectively minimized.	(O) Similar to conventional circulating fluidized bed.	(O) Counter-current operation will increase the adsorber footprint relative to conventional circulating fluidized beds to maintain down-flowing sorbent.	(−) Similar to conventional circulating fluidized bed.
Multistage dense fluidized bed (switching)	(O) Similar to counter-current multistage circulating fluidized bed, but the transient reaction front will move through the stages over time, requiring a taller reactor.	(+) High heat transfer rates from fluidization and practical vacuum swing from standalone reactors allow for easy VTSA operation, synergistically combining heat and vacuum pumps.	(+) Good CO_2 recovery and purity can be achieved by implementing an evacuation step directly after the adsorption step.	(−) A cluster of transient reactors is needed for continuous flue gas processing and heat pump working fluid must be continuously redirected between different reactors.	(O) Moderately large footprint to avoid elutriation of particles, but sufficient gas velocities can be achieved as particles are allowed to fluidize.	(O) The fluidization velocity could be varied over an order of magnitude while maintaining the dense regime (bubbling and turbulent regime).

[a]Symbols: + (advantage), O (neutral), and − (drawback) are used to describe the overall performance of each reactor configuration against the different factors.

with the different families of sorbents. Therefore, identifying the best combinations of reactor configuration, regeneration mode, and sorbent families along with taking into consideration the CO_2 source (CO_2 partial pressure in the flue gas) would bring breakthrough insights to the adsorption-based CO_2 capture community. This would set up clear roadmaps and directions for development and scaleup of the most promising combinations for the different industrial sectors. Such an approach was partially followed for screening sorbents based on key performance indicators such as CO_2 recovery, purity, and avoidance costs, but it was limited to a fixed bed configuration using a PVSA regeneration mode.[120] Future studies should also investigate the technical and economic aspects of flexible load operation and partial capture of the different combinations of reactor configuration and regeneration modes. As observed in Table 2, most of the technoeconomic assessment studies were completed for capturing CO_2 from coal power plants, so there is a clear need for extending them to other CO_2 sources covering power generation (natural gas and waste-to-energy plants), cement, metal production, and biofuel.

As for the technical aspect, thorough experimental testing and validation at the lab and prepilot scale should be dedicated to promising reactor configurations, such as the multistage fluidized bed and the structured reactors. For this latter, the focus should be on investigating the feasibility of integrating the designed regeneration modes and confirmation of achieving sufficiently high separation performance and sufficiently short cycle time. Measuring the extent of heat recovery and addition in the appropriate configurations and determining its limitations is also of high importance for heat integration purposes. Several recent studies have already touched on this for specific reactor configurations, but the studied designs and achieved performance remain far below expectations.[68,99,121,122]

8. SUMMARY AND CONCLUSIONS

This paper provides a detailed overview on the different reactor configurations proposed for adsorption-based CO_2 capture. Various gas—solid contacting systems and sorbent regeneration modes are identified and discussed in terms of strengths and limitations. In addition, the suitability of the different combinations of contacting systems and regeneration modes are discussed.

The fundamental behavior of the conventional fixed and fluidized bed during adsorption is inadequate for extracting the full potential of the technology. Fixed beds exhibit high pressure drop and formation of heat waves in addition to mass and heat transfer limitation issues. The good mixing in fluidized beds results in early CO_2 breakthrough that, in turn, results in poor CO_2 recovery or low sorbent utilization.

To overcome the challenges associated with these two reactor configurations, recent research trends focused on structured packings for the fixed bed and counter-current multistage for fluidized bed. Smartly designed structures can minimize the pressure drop, while maintaining a high gas—solids contacting area, thus maximizing the flue gas throughput rate and minimizing the reactor size and footprint. As for a fluidized bed, the multistage configuration can minimize the negative effect of the mixing on the CO_2 recovery and working capacity, while maintaining the benefits of the good heat transfer characteristics for heat integration purposes. Both these configurations pose considerable scale-up challenges due

to their increased complexity, but they appear to hold the greatest promise for the future of adsorption-based CO_2 capture.

Alternative configurations include rotating beds, with similar properties to structured fixed beds, and moving beds, with similar properties to multistage fluidized beds. Relative to fixed beds, rotating beds offer steady state process operation, but impose challenges with sealing when a vacuum needs to be drawn. Moving beds can further increase sorbent working capacity relative to multistage fluidized beds, but impose challenges regarding heat transfer rate, mass transfer resistance on the larger particles, and pressure drop. Another noteworthy alternative is the swing adsorption reactor cluster that synergistically combines vacuum swing and temperature swing using a heat pump.

A range of energy duties has been reported for different adsorption-based CO_2 capture technologies, although the variance in flue gas composition, CO_2 recovery, and purity, and process modeling assumptions makes it difficult to compare between different studies. However, several studies find that the sorbent-based processes outperform MEA benchmarks. In addition, VSA and heat pump concepts that consume only electrical energy show great promise for industrial processes without large amounts of low-grade heat that can be used for TSA. In general, there is a clear need for studies using standardized assumptions for comparing the different reactor configurations and benchmarking them using key performance indicators, particularly the cost of CO_2 avoidance that combines both energy efficiency and capital cost into a single metric. It is likely that different sorbent—reactor combinations work best for different flue gas streams and desired CO_2 avoidance rates. Such a standardized benchmarking study therefore needs to be completed for several key future applications of postcombustion CO_2 capture.

Other aspects such as the complexity of the operation strategy of the selected combinations, ease of retrofitting, and the ability to operate under a flexible load should also be considered in this comparison. Following such a standardized study, research efforts can be focused on development and scale-up of the most promising sorbent—reactor combinations to accelerate the commercial deployment of adsorption-based postcombustion CO_2 capture.

■ ASSOCIATED CONTENT

ⓢ Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.iecr.0c04547.

> Advantages and limitations of the different reactor configurations and of the different regeneration modes for adsorption-based CO_2 capture (PDF)

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Notes

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ABBREVIATION

SARC	swing adsorption reactor cluster
VPSA	vacuum pressure swing adsorption
VSA	vacuum swing adsorption
VTSA	vacuum combine temperature swing adsorption
TSA	temperature swing adsorption
PSA	pressure swing adsorption
ESA	electric thermal swing
CCUS	carbon dioxide capture, utilization and storage
COP	coefficient of performance
P&ID	process and instrumentation diagram
MFC	mass flow controller
PEI	polyethyleneimine
LPM	liters/minutes
GJ	gigajoules
MJ	megajoules
t	tonne
MOF	metal organic framework
KX35	potassium carbonate-based solid sorbent
Sorb	sorbent (adsorbent)
HP	heat pump
SA-VSA	steam-aided vacuum swing adsorption
C	gas phase CO_2 concentration at the section of reactor
C_0	gas phase CO_2 concentration at the feed section of the reactor
θ	vacant site available for the adsorption at the section of reactor
θ_0	equilibrium vacant site available for the adsorption at the feed CO_2 partial pressure
L_1 and L_2	adsorption capacity (mol/kg)
NaUSY	type of zeolite (adsorbent)
MWth	megawatt thermal
CHP	combined heat and power
MW	megawatt
k_{LDF}	LDF mass transfer coefficient for pore diffusion control (s^{-1})
h_i	heat transfer coefficient (W/(m^2 k))
k_f	thermal conductivity of sorbent (W/(m K))
D_e	molecular diffusivity (m^2/s)

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