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## A Novel Swing Adsorption Reactor Cluster (SARC) for cost effective post-combustion CO<sub>2</sub> capture: A thermodynamic assessment

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### Abstract

The SARC consists of a number of standalone reactors where a solid sorbent is carbonated by a flue gas and regenerated by a combination of vacuum and temperature swing. Efficiency is maximized through heat integration between carbonation and regeneration using a heat pump. Initial power plant simulations showed 9.4 %-points energy penalty when integrated into a pulverized coal plant. This is in-line with reported energy penalty for MEA and VPSA technologies, but great potential for further efficiency improvements exists. Future studies will investigate the effect of SARC process parameters and sorbent material selection on the energy penalty.

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## 1. Introduction

Adsorption based CO<sub>2</sub> capture processes using low temperature solid sorbents, hold great potential for reducing CO<sub>2</sub> capture costs [1]. Low temperature adsorption based post-combustion CO<sub>2</sub> capture technology, using alkali-metal and functionalized amine sorbents, has the advantage of easy retrofit to CO<sub>2</sub> production sites, including both industrial and energy sectors. These sorbents have more tolerance to water and can absorb low partial pressure CO<sub>2</sub>, which makes them suitable to capturing CO<sub>2</sub> from power plants [2]. Due to the high sensitivity to the temperature, this category of sorbents was mainly operated in an interconnected fluidized bed loop where the sorbent undergoes a pure temperature swing as it circulates between the two reactors [3, 4]. One main challenge with this configuration is that conventional co-current riser operation cannot achieve the desired degree of CO<sub>2</sub> capture. Plug flow operation in packed beds can solve this problem, but application of temperature swing adsorption (TSA) in a packed bed configuration was also considered inadequate due to the issue of hot spot formation in packed bed reactors, in addition to the long cycle time which can run over several hours [5].

The Swing Adsorption Reactor Cluster (SARC) concept, which is the focus of this paper, aims to reduce the reactor design complexity and bring considerable energy penalty reduction with respect to alternative low temperature post-combustion adsorption systems. It consists of a cluster of standalone reactors where the sorbent in each single reactor is alternately exposed to carbonation and regeneration conditions via periodic switching of the gas feed streams, reactor temperature and operating pressure (Figure 1). This concept allows combination of temperature and vacuum swings, which promises substantial energy penalty reduction in chemisorption based CO<sub>2</sub> capture processes.

To minimize the energy penalty, the temperature swing can be carried out using a heat pump with high coefficient of performance (COP), thanks to the limited temperature difference between evaporation (~55°C) and condensation (~70-75°C), that continuously transfers heat between the reactors in the cluster. In the proposed process, the heat generated by the exothermic carbonation reaction will be supplied to the endothermic regeneration stage by means of the heat pump, ensuring a tight heat integration between the carbonation and the regeneration steps. As a result, the SARC concept does not require waste heat (e.g. integration with the steam cycle of a power plant) to efficiently regenerate the sorbent. This promises simplified retrofits to existing infrastructure and substantial energy penalty reductions when applied to industrial processes with limited availability of waste heat for sorbent regeneration. Application of a vacuum swing will minimize the temperature difference between the carbonation and regeneration steps, thus maximizing the heat pump coefficient of performance, and thereby reducing considerably the energy penalty of CO<sub>2</sub> capture.

The heat pump integration in the process will require tubes insertion in each reactor, in which a working fluid circulates for transferring heat between the reactors in the cluster. Due to its excellent mixing properties, a fluidized bed reactor is best suited for this concept, as it ensures efficient heat transfer between the heat pump working fluid in the tubes and the bed. However, the good mixing makes the sorbents to carbonate equally in the entire bed, thereby causing reduced CO<sub>2</sub> uptake as the CO<sub>2</sub> partial pressure decreases when it rises through the bed. To alleviate this drawback, a multistage dense fluidized bed is proposed for the SARC reactor concept (Figure 1). Other concepts considered multistage fluidized bed systems but with downcomers [6, 7]. In contrast to SARC, these concepts involve solids circulation between the different bed stages (from top to down) while the gas is fed from the bottom; An external mechanism is therefore required to lift the solids to the top of the subsequent reactor. Additionally, the proposed concepts in [6, 7] can only be suitable for a pure temperature swing, because application of a vacuum swing in this type of looping configuration will be practically unfeasible. The multistage fluidized bed reactor proposed in the SARC concept does not involve any solids exchange between the bed stages and no additional devices are needed as no external transport of the sorbents takes place, thereby facilitating the combination of temperature and vacuum swings.

This paper investigates the SARC concept for integration in a coal power plant through reactor and power plant simulations. The reactor, heat pump and power plant models will be described first then followed by a result section presenting the SARC reactor behavior and estimated energy penalty imposed in a base case.

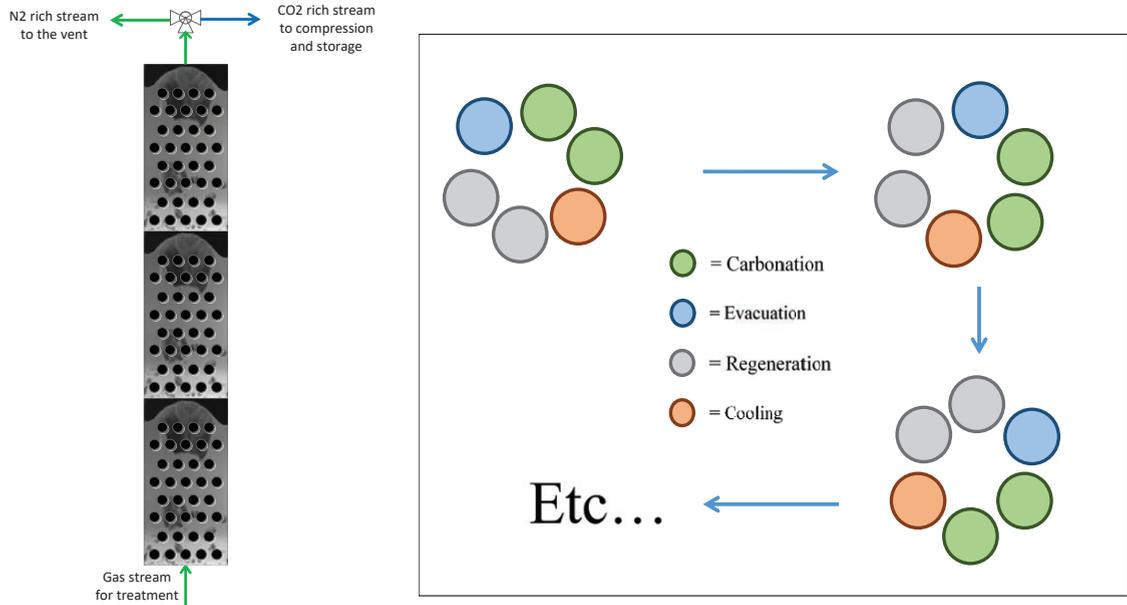


Figure 1: A conceptual design of the SARC concept: left, a typical multistage fluidized bed SARC reactor with tube insertion for heat addition/ recovery using the heat pump; right, SARC reactor cluster for steady operation [8].

## Nomenclature

APH	Air preheater
COP	Heat pump coefficient of performance
CSTR	Continuous Stirred Tank Reactor
FF	Fabric filter
FGD	Flue gas desulfurizer
HP	High pressure
LHV	Lower heating value
LP	Low pressure
MP	Medium pressure
PFR	Plug flow reactor
SARC	Swing Adsorption Reactor Cluster
SCR	Selective catalytic reduction
SPECCA	Specific primary energy consumption for CO <sub>2</sub> avoided
TSA	Temperature Swing Adsorption
USC	Ultra supercritical
VSA	Vacuum Swing Adsorption
VPSA	Vacuum/pressure Swing Adsorption

## 2. Reactor simulations

A simple reactor model was developed for simulating the SARC concept where the reactor was considered as a series of Continuous Stirred Tank Reactors (CSTR) containing solid material and gas. The feed gas was considered to be an ideal mixture of ideal gases. Thermal and chemical equilibrium between gas and solid was assumed.

Chemical equilibria binding CO<sub>2</sub> in Active Material + moisture in solid were considered as shown by Equations (1) and (2), where Am is the active material. Those equations can be written in form a stoichiometric matrix as described by Equation (3) which can also be divided into a gas and solids part (Equation (4)). The solids part is converted to mass basis by multiplication with molar weights. Chemical equilibria for CO<sub>2</sub> and water were modelled based on experimental data for a functionalized amine sorbent [9]. A Toth isotherm was used to describe the CO<sub>2</sub> adsorption capacity  $q_{CO_2}$  [molCO<sub>2</sub>/kg<sub>sorbent</sub>] as a function of temperature and CO<sub>2</sub> partial pressure  $P_{CO_2}$  [bar] (Equation (5)). Experimental data was also provided for H<sub>2</sub>O adsorption capacity  $q_{H_2O}$  at different relative humidities  $\phi$  [9]. The data could be well described by a second order polynomial, resulting in the model using the equilibrium vapour pressure of water  $P^*_{H_2O}$  described by Equation (6).

Table 1: Reactor model and isotherms for CO<sub>2</sub> and H<sub>2</sub>O adsorptions on a functionalized amine sorbent.

<b>GAS SOLIDS ADSORPTION REACTIONS</b>	
Am(s) + CO <sub>2</sub> (g) ↔ AmCO <sub>2</sub> (s) (ΔH= 75 kJ.mol <sup>-1</sup> )	(1)
H <sub>2</sub> O(g) ↔ H <sub>2</sub> O(s) (ΔH= 43 kJ.mol <sup>-1</sup> )	(2)
$\begin{pmatrix} CO_2 \\ N_2 \\ H_2O(g) \\ O_2 \\ Am \\ AmCO_2 \\ Inert \\ H_2O(s) \end{pmatrix} \rightleftharpoons 0$	(3)
$\begin{pmatrix} -1 & 0 & 0 & 0 & -1 & 1 & 0 & 0 \\ 0 & 0 & -1 & 0 & 0 & 0 & 0 & 1 \end{pmatrix}$	(4)
Stoichiometric matrix, S	$S = (S_{gas}, S_{solid})$
Chemical species, κ	
<b>ISOTHERM FOR CO<sub>2</sub> ADSORPTION</b>	<b>ISOTHERM FOR H<sub>2</sub>O ADSORPTION</b>
$b = 408.84 \exp\left(29.542 \left(\frac{353}{T} - 1\right)\right)$	(5)
$t = 0.3 + 0.14 \left(1 - \frac{353}{T}\right)$	(6)
$q_{CO_2} = \frac{3.4bP_{CO_2}}{\left(1 + \left(bP_{CO_2}\right)^t\right)^{\frac{1}{t}}}$	$P^*_{H_2O} = 770529 e^{\left(\frac{-5049.9}{T}\right)}$
	$\phi = 100 \frac{P_{H_2O}}{P^*_{H_2O}}$
	$q_{H_2O} = 9.01124 E-4 \phi^2 + 2.79362 E-2 \phi + 1.1965$
<b>MASS ANDN HEAT BALANCE</b>	
$\frac{dN}{dt} = F^{in} y^{in} - F_y + S^T_{gas} R$	(7)
$\frac{dM_{solid}}{dt} = S^T_{solid} R$	(8)
$(M^T_{solid} C_{p,solid} + N^T C_{v,gas}) \frac{dT}{dt} = F^{in} h^{in} - Fh - R^T \Delta H_{rx} + Q$	(9)
$Q = AU \times \Delta T$	(10)
$pV_{gas} = \text{sum}(N)R_0T$	(11)

The mass balance is described in Equations (7) and (8) and are solved for gas and solid in each CSTR. N [kmol] is a vector containing the gas holdup of each gas species; F<sup>in</sup> and F [kmol/s] are the gas flow rates into and out of the CSTR; y=N/sum(N) [-] is a vector of gas mole fractions; M [kg] is a vector of the mass holdup for each solid species; and R is a vector of chemical reaction rates. The reaction rates are determined so as to satisfy the chemical equilibria (similar approach was used in simulating a multistage fluidized bed system running a pure TSA CO<sub>2</sub> capture [7]). This

is considered a reasonable assumption given the long gas residence times and good gas-solids contact in the large SARC reactors operating in the bubbling/turbulent regime.

The energy balance is described in Equation (10) where  $C_{p,solid}$  [J/kg.K] is a vector of solid heat capacities;  $C_{v,gas}$  [J/kmol.K] is a vector of gas heat capacities;  $T$  [K] is the reactor temperature;  $h^{in}$  and  $h$  [J/kmol] are the enthalpies of the inlet and outlet gas;  $\Delta H_{rx}$  [J/kmol] is a vector of the enthalpies of reaction; and  $Q$  [W] is the heat addition or removal by the heat pump which is evaluated as shown by Equation (11).  $A$  [m<sup>2</sup>] is the total heat transfer surface area,  $U$  [W/m<sup>2</sup>.K] is the heat transfer coefficient and  $\Delta T$  [K] is the temperature difference between the reactor and the heat transfer fluid in the heat pump tube bundles. Finally, the ideal equation of state was used for the gas (Equation (12)), where  $p$  [Pa] is the pressure;  $V_{gas}$  [m<sup>3</sup>] is the gas volume; and  $R_0$  [J/kmol.K] is the universal gas constant.

Boundary conditions will be specified for individual cases in the results and discussion section of this paper.

The heat and mass balance for each of the CSTR's in series was solved using ode15s, which is a differential-algebraic equation solver in Matlab.

### 3. Process integration

A process integration study of the proposed SARC process integrated in a large-scale pulverized coal (PC) ultra-supercritical (USC) power plant (Figure 3) has been performed. The power plant generates superheated and reheated steam at 270/60 bar and 600/620°C, by burning 1657 MW<sub>LHV</sub> of low sulphur South African bituminous coal. The mass and energy balances of the power plant are calculated with the in-house code "GS" [10]. The main assumptions used for the calculations of the power plant are defined according to EBTF guidelines [11]. For more details on the power plant model, the reader is directed to [12].

The heat pump is based on a conventional vapor compression cycle using ammonia as working fluid. The excess heat available from de-superheating the ammonia exiting the compressor is used to preheat a stream of water from the steam cycle condenser, which is returned to the water preheating line (#21). Process integration includes low temperature heat recovery from the intercoolers of the CO<sub>2</sub> compressors and of the gas evacuation vacuum pump. This low temperature heat is used to heat up the condensate from the steam cycle condenser (#22) and allows to completely avoid the first two feed water heaters, improving the overall efficiency of the plant. The assumptions adopted for the process integration of the SARC unit are resumed in **Error! Not a valid bookmark self-reference.**

Table 2: Assumptions for the process integration of the SARC process in the power plant.

Heat pump compressors isentropic efficiency, %	85
Heat pump compressors electric-mechanical efficiency, %	94
Vacuum pumps isentropic efficiency, %	85
Vacuum pumps electrical-mechanical efficiency, %	95
Vacuum pumps intercooling temperature, °C	35
Regenerator recycle fan isentropic efficiency, %	80
Regenerator recycle fan electrical-mechanical efficiency, %	94
Pressure loss in gas distribution nozzles, % of solid inventory weight	10

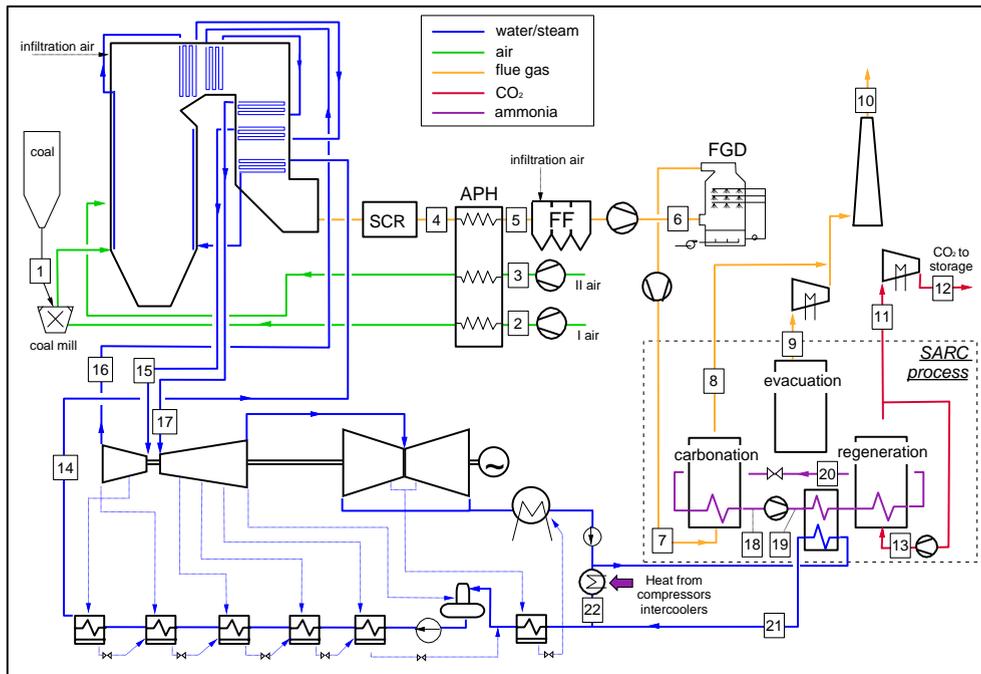


Figure 2: PC-USC power plant integrated with the SARC process and ammonia heat pump

## 4. Results

### 4.1. Reactor sizing

The dynamic SARC reactor cycle contains four steps: carbonation, evacuation, regeneration and cooling. Heat must be supplied during regeneration and extracted during carbonation and cooling. Heating and cooling rates in the different process steps are calculated assuming an overall heat transfer coefficient,  $U$ , of 300 W/m.K, a heat transfer tube diameter of 30 mm, a tube spacing of 45 mm and a staggered geometry. The assumed heat transfer coefficient is of the same order as expected for bed to immersed tubes heat transfer in bubbling fluidized beds [13]. In fact, it is expected that heat transfer is limited by bed to tube thermal resistance, being the heat transfer coefficient of the heat pump working fluid in condensation and evaporation at least one order of magnitude higher than the assumed overall value. The total reactor volume must be large enough to accommodate all the tubes required to extract the heat released when absorbing CO<sub>2</sub> from the incoming flue gas stream given a certain design  $\Delta T$ , assumed to be 5 °C in this case.

The SARC simulated in this work processes a flue gas stream with 13.4% of CO<sub>2</sub>, entering with a flowrate of 610 Nm<sup>3</sup>/s. It is assumed that the carbonation step of each reactor in the cluster receives one tenth of this flue gas stream, i.e. 61 Nm<sup>3</sup>/s. This gas stream is fed at an assumed inlet superficial velocity of 1 m/s and an average carbonation temperature of 50 °C. These flow conditions will require a circular reactor with a diameter of 9.6 m.

When assuming 90% CO<sub>2</sub> capture and 100% steam adsorption [9] for heat exchanger sizing purposes (while in the simulations for the mass and energy balance calculation, CO<sub>2</sub> and H<sub>2</sub>O adsorbed were properly modelled based on the adsorption isotherms presented above), the resulting overall reaction enthalpy (including steam) is 114 kJ/mol of CO<sub>2</sub> adsorbed. The incoming stream requires a heat extraction rate of 37.4 MW. The tube arrangement outlined earlier can extract 4.54 MW/m of reactor height, implying that the bundle of horizontal tubes must be 8.25 m in height. Assuming an additional reactor height of 80% the tube bundle height, a total reactor height of 14.85 m can be calculated. Given the sorbent density of 880 kg/m<sup>3</sup> and a static bed void fraction of 0.4, the total mass of sorbent needed to fill the bed up to the height of the tube bundle is 205 ton (or 2835 kg/m<sup>2</sup> of reactor cross section). This will lead to a pressure drop of 30.59 kPa, including 10% of assumed additional pressure drops due to gas distribution nozzles.

#### 4.2. Typical SARC behavior

This section will present an example of the behaviour of a typical SARC cycle. The main steps of the SARC cycle are the carbonation and regeneration steps. The carbonation takes place at low temperature and atmospheric pressure while a combination of vacuum and temperature swings is applied simultaneously in the regeneration step. A primary vacuum pump is used to carry the vacuum in the regeneration step (it is referred to in the rest of the manuscript as the regeneration pump). An evacuation step is applied after the carbonation step in order to evacuate the N<sub>2</sub>-rich gas accumulating in the reactor from the carbonation step to avoid that it mixes and reduces the purity of the CO<sub>2</sub>-rich gas stream released by the sorbent in the regeneration step. A secondary vacuum pump is used for this purpose where the evacuated gas is vented to the atmosphere (it is referred to in the rest of the manuscript as the evacuation pump). A cooling step is applied after the regeneration step combined with a re-pressurization of the reactor, to bring the pressure and temperature in the SARC reactor back to the set conditions for starting a new carbonation step. A small flowrate of flue gas is fed to this step at 4.6 times the minimum fluidization velocity (i.e. 0.0228m/s) to maintain fluidization while minimizing CO<sub>2</sub> losses (it is assumed here that the mean sorbent diameter of 150 μm and the gas stream consists of only N<sub>2</sub>). The heat addition and removal to the different steps are carried out using the heat pump.

A simulation of SARC reactor behaviour over one process cycle was completed using a multistage reactor composed of four beds. The carbonation and regeneration step times were fixed at 300 s. The heat pump temperature difference was set to 18 K and the vacuum pressure to 0.1 bar. Data of transient evolutions of reactor temperature, pressure and outlet CO<sub>2</sub> mole fraction were collected and plotted in Figure 3. The heat pump maintained the temperature in the carbonation step almost constant at 333 K despite the high uptake of CO<sub>2</sub> and steam in this step, and associated large heat generation. The temperature has however slightly decreased in the evacuation step due to the slight release of CO<sub>2</sub> resulting from the initiation of sorbent regeneration at the end of the evacuation step as the pressure is reduced. The regeneration step appears to occur in two phases; the first phase is driven by the vacuum swing (under the reaction assumptions employed in the reactor model, this CO<sub>2</sub> release would be instantaneous, but to account for various limiting factors such as vacuum pump capacity, regeneration kinetics and the need to prevent particle elutriation, the maximum outlet flow rate was limited to 1 kmol/s). Despite the endothermic regeneration reaction, the reactor temperature continuously increases because the difference between the heat pump working fluid temperature (condensation temperature) and reactor temperature is large at the start of the regeneration. In the second phase of the regeneration, the reactor pressure reaches the constant minimum of 0.1 bar and further CO<sub>2</sub> release must be driven by continued temperature swing. The rate of regeneration gradually reduces as the reactor temperature approaches the condensation temperature of the heat pump working fluid and the resulting heat transfer rate slows down.

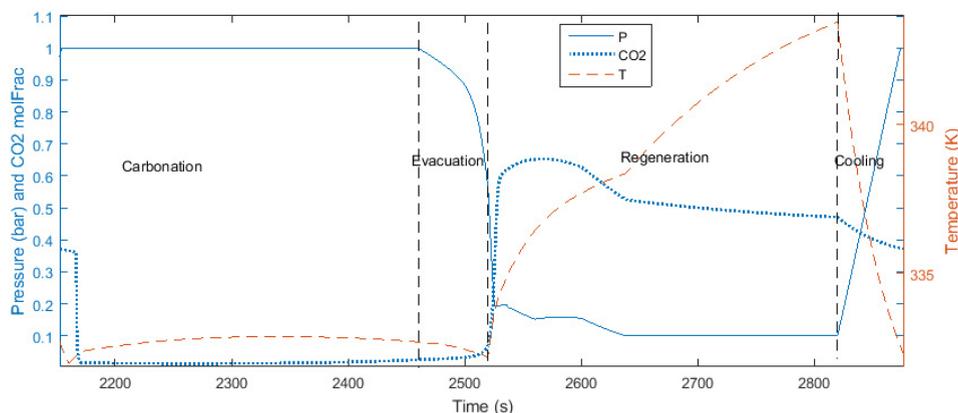


Figure 3: Transient reactor behaviour in a typical SARC cycle.

#### 4.3. Power plant simulation of base case

The energy balance and the main performance indicators of the complete power plant integrated with the SARC

process with the reference operating parameters of the base case simulated in the previous section are shown in Table 2 together with the energy balance of the reference power plant without CO<sub>2</sub> capture [12]. The unavoidable CO<sub>2</sub> compression represents the largest penalty, with 3.3 %-points of efficiency loss. The two SARC vacuum pumps impose the second largest energy penalty with 2.9 %-points. Thirdly, the NH<sub>3</sub> compressor of the heat pump cycle imposes an energy penalty of 2.2 %-points. The low heat pump power consumption is remarkable given the relatively high heat duty of 3.27 MJ/kg of CO<sub>2</sub> required to regenerate and heat up the sorbent. This illustrates the high heat pump efficiency facilitated by the small temperature difference between the carbonation and regeneration steps. Finally, the fan required to overcome the pressure drop when blowing the flue gas through the carbonation step of the SARC cycle also imposes a significant energy penalty of 1.6 %-points. On the whole, an efficiency penalty of 9.64 percentage points has been calculated with respect to the reference case without CO<sub>2</sub> capture. As a result, SPECCA of 3.26 MJ<sub>LHV</sub>/kgCO<sub>2</sub> has been calculated. Such efficiency penalty is in line with the values reported in the literature on post-combustion capture by MEA and VPSA [14-16]. Dedicated process parameter optimization and material selection studies, have the potential to improve significantly the SARC process efficiency. For example, significant energy saving can come from increasing the number of bed stages in the SARC reactor (this improves the CO<sub>2</sub> capture efficiency for the same energy input due to shifting the reactor behaviour from a CSTR towards a plug flow reactor (PFR) as more bed stages are added) and finding proper combinations between the heat and vacuum pumps energy consumptions. Additionally, almost no isotherm data is available for sorbents developed for temperature swing applications, thus not allowing for material optimization studies.

The footprint of the SARC reactors can be calculated by noting that 10 reactors, each 9.6 m in diameter, will always be needed in the carbonation step (Section 4.1). In the base case scenario, the carbonation and regeneration steps are 300 s in length, while the cooling and evacuation steps are 60 s long. This implies that 24 reactors are required in the SARC cluster with a total footprint of 1588 m<sup>2</sup>. In comparison, the footprints of MEA absorption columns and VPSA technology were previously calculated as 674 and 13285 m<sup>2</sup> respectively for a plant with almost the same primary energy input [16].

Table 3: Energy balance and performance indicators of the reference SARC power plants and the benchmark plant without CO<sub>2</sub> capture.

	Reference case w/o capture	SARC – Ammonia heat pump
Power balance, MW <sub>e</sub>		
Steam turbine	795.5	804.1
Steam cycle pumps	-26.57	-26.57
Auxiliaries for heat rejection	-6.18	-7.89
SARC fans	-	-27.92
Heat pump compressor	-	-36.50
Regeneration vacuum pump	-	-39.93
Evacuation vacuum pump	-	-8.23
CO <sub>2</sub> compression	-	-53.96
Other auxiliaries	-24.21	-24.21
Net power, MW <sub>e</sub>	738.6	578.8
Heat input, MJ <sub>LHV</sub>	1657.1	1657.1
Net electric efficiency, % <sub>LHV</sub>	44.57	34.93
Efficiency penalty, points % <sub>LHV</sub>	-	9.64
Specific CO <sub>2</sub> emissions, kg/MWh	782.1	99.6
CO <sub>2</sub> avoided, %	-	87.3
SPECCA, MJ <sub>LHV</sub> /kgCO <sub>2</sub>	-	3.26

## 5. Summary and conclusions

This paper investigated a novel swing adsorption reactor cluster (SARC) for post-combustion CO<sub>2</sub> capture through combined reactor modelling and power plant simulations. Each reactor in the cluster consists of a multi-stages fluidized bed operating under bubbling/turbulent regime conditions, which offers multiple advantages, such as simpler reactor design, high CO<sub>2</sub> capture efficiency and ability to combine temperature and vacuum swings. This last one can bring significant energy penalty reduction if the temperature swing is completed using a heat pump transferring heat from the exothermic carbonation to the endothermic regeneration. Vacuum swing reduces the required temperature difference between carbonation and regeneration, which allows the heat pump to transfer heat between the SARC steps in a highly efficient manner.

Integration of the SARC concept with a conventional pulverized coal ultra-supercritical power plant showed an energy penalty of 9.64 %-points for the base case with ammonia as the heat pump working fluid, which is in-line with reported energy penalty for MEA and VPSA technologies. The footprint of the base case was about 2.4 times larger than state-of-the-art MEA absorption columns, but it is much smaller than a vacuum/pressure swing adsorption system due to the higher throughput and avoidance of a second process stage to purify the CO<sub>2</sub>-rich stream from the main vacuum swing stage. Additional reactor footprint can be traded for increased process efficiency.

Dedicated process parameter optimization and material selection studies have the potential to significantly improve the SARC process efficiency. Finally, the SARC system requires almost no complex integration with the steam cycle, making it attractive for retrofit applications. Also, since no steam is required for sorbent regeneration, the SARC concept promises great energy penalty reductions in industrial applications with low availability of waste heat. The simple standalone nature of SARC reactors will also simplify further process scale-up and demonstration efforts.

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