Adsorption technology for direct recovery of compressed, pure CO₂ from a flue gas without pre-compression or pre-drying

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Abstract The effects of the sorption and the regeneration temperatures on the performance of a novel rapid thermal swing chemisorption (RTSC) process (Lee and Sircar in AIChE J. 54:2293–2302, 2008) for removal and recovery of CO₂ from an industrial flue gas without pre-compression, pre-drying, or pre-cooling of the gas were mathematically simulated. The process directly produced a nearly pure, compressed CO₂ by-product stream which will facilitate its subsequent sequestration. Na₂O promoted alumina was used as the CO₂ selective chemisorbent, and the preferred temperatures were found to be, respectively, 150 and 450 °C for the sorption and regeneration steps of the process. The specific cyclic CO₂ production capacity of the process and the pressure of the by-product CO₂ gas were substantially increased over those previously achieved by using the sorption and regeneration temperature of, respectively, 200 and 500 °C (Lee and Sircar in AIChE J. 54:2293-2302, 2008). The net compressed CO₂ recovery from the flue gas (\sim 92%) did not change. However, substantially different amounts of high and low pressure steam purges were necessary for comparable degree of desorption of CO₂. A first pass estimation of the capital and the operating costs of the RTSC process was carried out for a relatively moderate size application (flue gas clean up and CO_2 recovery from a ~ 80 MW coal fired power plant). Both costs were substantially lower than those for a conventional absorption process using MEA as the CO₂ solvent (Desideri and Paolucci in Energy Convers. Manag. 40:1899-1915, 1999).

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1 Introduction

Emission of carbon dioxide, the primary green house gas, to the atmosphere via industrial flue gases has become a major universal concern due to the potential global warming problems. Consequently, development of efficient and cost-effective means for post-combustion removal and recovery of CO₂ from a flue gas for possible sequestration is a major topic of current research and development in the field of chemical engineering. Three different generic separation technologies have been extensively evaluated for this application. They are (a) physical or chemical absorption of CO₂ by a liquid solvent, (b) physical adsorption of CO₂ by a micro-porous solid adsorbent, and (c) selective permeation of CO₂ through a porous or a non-porous membrane. The key features of these technologies have been highlighted and a list of major references on the topic is provided in a recent publication by Lee and Sircar (2008). Many of these technologies are fairly well developed and continue to be improved through process modifications and development of new materials used in the processes. Several key articles describing the state of the art are listed in the reference section (Barchas and Davis 1992; Chakravarti et al. 2001; Chue et al. 1995; Daniels et al. 1998; Desideri and Paolucci 1999; Ishibashi et al. 1995; IEA GHG 2010; Karasaki et al. 1995; Leci and Goldthorpe 1992; Makita et al. 1993; Matsumoto et al. 1992; Mimura et al. 1995; Riemer 1996; Suzuki 1996).

The crucial technical issues associated with the problem of CO₂ removal and recovery from an industrial flue gas are



that (a) the CO_2 is present in dilute or moderate concentrations (\sim 5–30 mole %), (b) the volumetric flow rate of the gas to be treated is very high, (c) the flue gas is generally available at a near ambient pressure, (d) the gas is saturated with water, and (e) the gas is generally produced at an elevated temperature of 130–180 °C (Lee and Sircar 2008).

Many of the above-mentioned separation technologies may require pre-drying, pre-compression, and pre-cooling of the flue gas prior to effective CO₂ removal and recovery. The cost associated with these pretreatments of the flue gas may render the over-all separation idea to be economically non-viable. Furthermore, most of these separation methods only produce a concentrated stream of CO₂ at a near ambient pressure along with the CO₂ free (or lean) effluent gas. Thus, sequestration of the recovered CO₂ enriched gas by many of these technologies will require a significant compression power and capital cost. A typical sequestration pressure may be 140 bar.

The recently proposed rapid thermal swing chemisorption process (RTSC) concept for removal and recovery of CO_2 from a flue gas circumvents some of the short comings of the previous methods. It does not require precompression, pre-drying or pre-cooling of the flue gas and simultaneously and directly produces a CO_2 free effluent gas at the flue gas pressure, and an essentially pure and compressed CO_2 by-product gas (Lee and Sircar 2008). This can potentially reduce the cost and energy of subsequent sequestration of the CO_2 by-product.

The RTSC process employs a reversible CO_2 chemisorbent like Na_2O promoted alumina which (a) selectively chemisorbs CO_2 from a flue gas in presence of water, (b) offers a decent CO_2 working capacity for a thermal swing sorption concept for CO_2 removal and recovery by operating between temperatures of $150-550\,^{\circ}C$ without precompression of the flue gas, (c) exhibits fast kinetics for CO_2 sorption and desorption, (d) permits thermal desorption of CO_2 by purging the sorbent with super-heated steam, and (e) is thermally stable at a temperature of $450\,^{\circ}C$ as indicated by limited studies (Lee et al. 2007b).

Figure 1 shows a conceptual diagram of the five-step cyclic RTSC process concept, which can be practiced in a multi-column configuration similar to that of a commercial poly-bed pressure swing adsorption (PSA) process concept (Fuderer and Rudelstorfer 1976), where more than one parallel columns simultaneously undergoes a certain step of the RTSC process at a given time (Lee and Sircar 2008).

The RTSC process steps include (a) sorption of CO_2 from a flue gas at near ambient pressure and at a temperature of 150–200 °C to produce an effluent (vent) gas containing $\leq \sim 1$ mole % CO_2 , (b) co-current CO_2 rinse under similar conditions to purge out the void impurities from the sorber and recycling the effluent as feed, (c) heating the sorber, which is saturated with pure CO_2 at the conditions of

step (b), in order to desorb the CO_2 in a batch-wise manner and produce a void gas comprised of essentially pure but compressed CO_2 , (d) counter current steam purge at high pressure to desorb the CO_2 at high pressure followed by condensation of steam in a constant pressure heat exchanger (CPHE) to produce the compressed, pure CO_2 by product gas, and (e) a multi-tasking step consisting of simultaneous column pressure reduction and high temperature steam purging for further desorption of CO_2 , and then simultaneously cooling the sorber under steam purge to desorb some more of the CO_2 and preparing the sorber to start a new cycle.

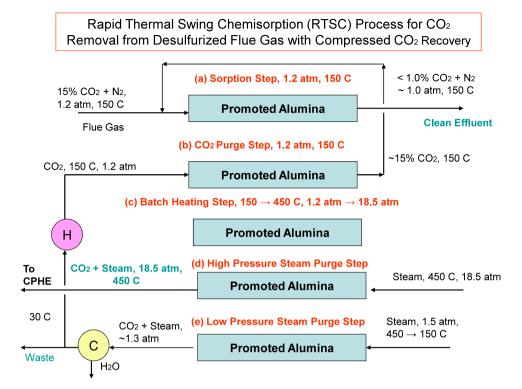
The process uses a number of parallel shell and tube type sorption columns where the tubes are packed with the chemisorbent and the shell side is used to heat or cool the tubes by cross- flow of superheated steam (Lee and Sircar 2008; Lee et al. 2008b). Several steam inlets and outlets along the length of the shell side of the sorber may be used to reduce any time lag during the heating and cooling steps. This arrangement permits the use of a rapid thermal swing adsorption (RTSA) process concept employing a cycle time of only several minutes for each individual step of the process (Sircar 2003).

The performance of the RTSC process concept was simulated earlier using the experimentally measured CO₂ chemisorption and desorption characteristics on Na₂O promoted alumina and a mathematical model to describe the mass and energy balances inside a sorber undergoing the steps of the process (Lee and Sircar 2008; Lee et al. 2007a). The step (a) of the process was operated at a temperature of 200 °C and a pressure of 1.1 bar, while the highest temperature in steps (c) and (d) of the process was 500 °C. The results showed that the RTSC concept was capable of removing CO_2 from a flue gas containing $\sim 15\%$ CO_2 and deliver a 99+% CO₂ by-product gas at a pressure of \sim 13.0 bar with a recovery of $\sim 93\%$ from the feed flue gas, and its performance was far superior to that of a pressure swing chemisorption (PSC) process designed for removal and recovery of CO2 from a wet, hot inert gas at super-ambient pressure (Sircar and Golden 2001).

The purpose of the present work is to demonstrate that (a) the performance of the RTSC concept can be significantly improved by carrying out the chemisorption step at a temperature of $150\,^{\circ}$ C and using a final regeneration temperature of $450\,^{\circ}$ C, and (b) the capital and the operating costs of the RTSC concept under these conditions of operation can be substantially lower than those of a conventional liquid- absorption process for CO_2 removal and recovery using MEA as the solvent (Desideri and Paolucci 1999).



Fig. 1 Schematic cartoon of the five-step RTSC process concept



2 Chemisorption of CO₂ on Na₂O promoted alumina

The key properties of Na₂O promoted alumina for sorption and desorption of CO₂ used in the model simulation were experimentally measured in our laboratory (Lee et al. 2007b, 2008a). Figure 2 shows the CO₂ chemisorption isotherms at different temperatures. They were estimated using a new analytical model which accounted for simultaneous Langmuirian chemisorption on the surface of the sorbent and an additional complexation reaction between the gaseous and the chemisorbed CO₂ molecules. The validity of the model was extensively tested using experimental isotherm data at different temperatures. The model parameters at different temperatures are reported elsewhere (Lee et al. 2007b). The low heats of CO₂ chemisorption (64.9 kJ/mole) and complexation reaction (37.5 kJ/mole) on the promoted alumina promoted easy desorption of CO₂ from the material by an inert gas purge.

The kinetics of chemisorption of CO_2 on the promoted alumina could be described by the Linear Driving Force (LDF) model (Lee et al. 2008a). Figure 3 shows that the LDF mass transfer coefficient ($k_{\rm LDF}$, minute⁻¹) for CO_2 chemisorption on the promoted alumina exponentially increases with increasing temperatures (Lee et al. 2008a). The effective activation energy for the CO_2 mass transfer on the sorbent was only 6 kJ/mole.

It has also been shown that the LDF model described the desorption of CO_2 from the promoted alumina by reduction

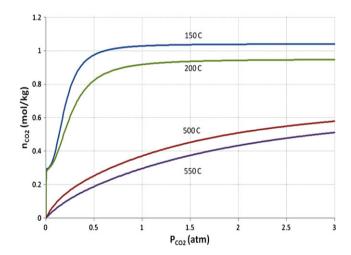


Fig. 2 Chemisorption isotherms of CO₂ on promoted alumina at different temperatures

of gas phase CO_2 partial pressure (decrease in total pressure or purge with an inert gas) (Lee et al. 2008a). The mass transfer coefficient for CO_2 desorption was identical to that for CO_2 sorption at the same temperature, and the coefficients were independent of CO_2 mole fraction (Lee et al. 2007b, 2008a).

The chemisorbent was developed by Air Products, Inc. A sample of the material, which was produced in commercial scale, was donated to Lehigh University. Long term stability of the material under the conditions of operation of the RTSC concept needs to be evaluated.



3 New simulations of RTSC process concept

The performance of each step of the RTSC process concept (Fig. 1) was simulated using the above-described thermodynamic and kinetic parameters for chemisorption of CO₂ on the promoted alumina and a mathematical model to simulate column dynamics of CO₂ sorption and desorption (Lee et al. 2007a). A shell and tube heat exchanger type sorber was used. The inside diameter of the tubes was 2.54 cm (=D), and they were 213.4 cm long $(=L_c)$. They were packed with spherical beads of Na₂O promoted alumina of ~ 0.35 cm diameter (= d_p). The sorbent was heated or cooled by cross-flow of super-heated steam at ambient pressure through the shell side having multiple steam entry and exit points. The over-all heat transfer coefficient from the shell to the tube side was found from the literature to be \sim 67 W/m² K⁻¹. The individual cycle times of steps (a)–(e) were, respectively, 3.0, 1.0, 3.0, 1.0 and 3.0 minutes. The model assumed ideal gas behavior, absence of axial dispersion, negligible pressure drop inside the sorber, and instantaneous thermal equilibrium between the gas phase and the sorbent. The flue gas was at 1.1 bar pressure and it contained

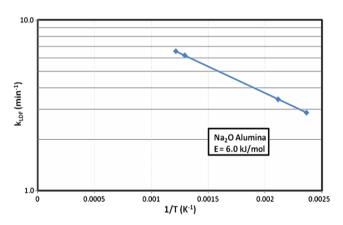


Fig. 3 LDF mass transfer coefficients for CO₂ chemisorption on promoted alumina as function of temperature

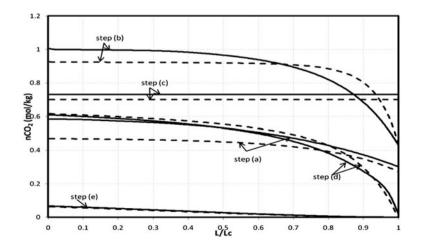
Fig. 4 CO₂ loading profiles in sorber at the end of the steps of RTSC process. *Solid lines* (150–450 °C), *dashed lines* (200–500 °C)

15% CO₂ in mixture with an inert gas (N₂). Figures 4–8 show some of the key process simulation results employing sorption and regeneration temperatures of, respectively, 150 and 450 °C.

Figure 4 shows the CO₂ loading profiles [moles of CO₂/kg of sorbent as function of normalized distance (L/L_c) from the feed gas end] inside the sorber at the end of steps (a)-(e) of the process for operation using sorptionregeneration temperature pairs of 150-450 °C (solid lines) and 200–500 °C (dashed lines). A very small amount of CO₂ was allowed to break through the sorber during step (a) of the process so that the average CO₂ mole fraction of the effluent gas from that step was $\sim 1\%$. The step (b) introduces extraneous amount of CO₂ in the sorber (chemisorbed and void) in order to saturate the sorber with nearly pure CO₂. Steps (d) and (e) desorbed about 98.2% of the CO₂ present in the sorber at the end of steps (b) and (c). It may also be seen from the figure that the residual CO_2 in the sorber at the end of step (e) was very small, and nearly 25% of the sorber volume towards the product end was essentially CO2 free at the end of that step. The integrated area under the profiles for steps (a) and (e) shown by Fig. 4 provides the net cyclic CO₂ capacity of the sorbent. The residual amount of CO₂ is practically the same when a regeneration temperature of 450 °C is used instead of 500 °C.

Figure 5 shows the heating and cooling profiles of the sorber tubes during steps (c) and (e) of the process. It shows that a period of \sim 3 minutes was sufficient to heat the sorbent from 150 to 450 °C and to cool it from 450 to 150 °C. The shell side steam temperatures used during steps (c) and (e) of the process were, respectively, 475 and 100 °C. The sorber was purged with low pressure steam at 450 °C during the initial part (\sim 1 minute) of step (e) before the cooling process was initiated.

Figure 6 explains the pressure rise inside the tubes during batch-wise heating (step c) of the process. It plots the total specific amount of pure CO_2 in the sorber tubes (chemisorbed + void) as functions of gas phase pressure at





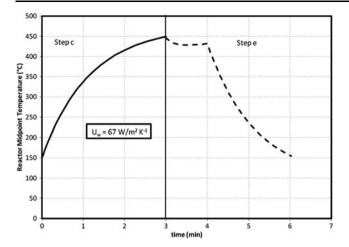


Fig. 5 Transient heating and cooling of the sorber

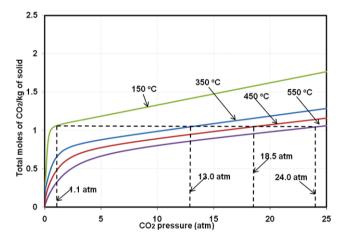


Fig. 6 Total equilibrium amount of CO₂ in sorber

the sorption (150 °C) and final regeneration (450 °C) temperatures of the RTSC process. The sorbent was assumed to be in equilibrium with the gas phase during this step. The CO_2 is desorbed during the batch heating process and it accumulates in the gas phase raising its pressure. The process is controlled by CO_2 mass conservation and the chemisorption equilibrium isotherm at the sorbent temperature. Figure 6 shows that the CO_2 gas pressure in the tubes increased from ~ 1.1 bar to ~ 18.5 bar due to batch-wise heating of the sorbent from 150 to 450 °C.

Figure 7 shows the transient mole fraction of CO_2 in the sorber effluent gas during steps (d) and (e) of the process as functions of the specific amount of steam purge per unit amount of net CO_2 produced by the process (ton/ton). The effluent gas is essentially a binary mixture of CO_2 and steam. The effluent gas during step (d) was initially pure CO_2 for a period of time. Then the CO_2 mole fraction progressively decreased as more purge steam was passed. The temperature of the effluent gas was $450\,^{\circ}C$ and its total pressure (super-ambient), which remained unchanged during the

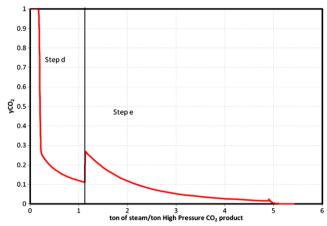


Fig. 7 Effluent gas CO₂ mole fractions during steps (d) and (e) of the RTSC process

step, was equal to the compressed CO_2 pressure inside the sorber at the end of step (c). The steam from this effluent can be condensed out in a constant pressure heat exchanger (CPHE) to recover the CO_2 by-product gas at \sim 18.5 bar pressure. In other words, the RTSC process serves as a de facto CO_2 compressor for the by-product CO_2 and facilitates its sequestration after further compression. A detailed mathematical model describing steam condensation in the shell side and water vaporization in the tube side of a shell and tube heat exchanger was developed for simulation of the performance of a constant pressure heat exchanger (CPHE) to be used in conjunction with the RTSC process for recovery of compressed CO_2 from step (d) of the process (Jeong and Caram 2010). The model confirmed the viability of design and operability of a CPHE concept.

The CO_2 mole fraction of the effluent gas during step (e) progressively decreased as more purge steam was passed. The mole fraction initially rises above that at the end of step (d) because of facilitated desorption of CO_2 due to lowering of the sorber pressure from that of step (d) to near ambient. It should be noted that the effluent CO_2 mole fraction became very low near the end of the step (e).

Figure 8 summarizes the CO_2 desorption pattern by the process. It gives the fraction of total CO_2 present in the sorber at the end of step (c) that is desorbed by steam purge (ton of steam/ton of net CO_2 product by the over-all process) during steps (d) and (e). It shows that $\sim 55\%$ of CO_2 is desorbed during the high pressure steam purge step which can be recovered as high pressure CO_2 by-product from the process. This translates to $\sim 93\%$ of flue gas CO_2 recovered as high pressure CO_2 by product (see Table 1).

The over-all simulated process performances of the RTSC concept for removal and recovery of compressed CO_2 from a flue gas containing 15% CO_2 at a pressure of 1.1 atm are summarized in Table 1. Process performance for two sets of temperatures for CO_2 chemisorption (150 and 200 °C)



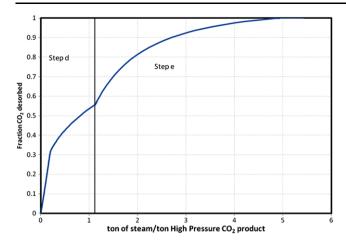


Fig. 8 Over-all CO₂ desorption pattern from the RTSC process

and sorbent regeneration (450 and 500 °C) are reported. It may be seen from the table that (a) the productivity of 99+% CO₂ by-product gas (moles/kg of sorbent) from a flue gas was \sim 33% higher and (b) the product CO₂ gas pressure was substantially larger (\sim 18.5 atm compared to \sim 13.0 atm) when the RTSC process was operated between the sorption and regeneration temperatures of 150 and 450 °C (case 1) instead of 200 and 500 °C (case 2) which was reported earlier (Lee and Sircar 2008).

This translates to $\sim\!22\%$ reduction in the power needed to further compress the CO_2 for sequestration at 140 atm, as well as a reduction of $\sim\!17\%$ in the capital cost of the compressor. The specific quantities of high and low pressure steam purges, however, increased for case 1. Approximately 98.2% of total CO_2 present in the sorber at the end of step (c) was removed during steps (d) and (e) in both cases.

4 First pass cost comparison

We put together a first past cost estimate for the RTSC concept operating between sorption and regeneration temperatures of 150 and 450 °C for removal and recovery of CO₂ from a flue gas using the simulated process performance data reported in Table 1. The purpose was to obtain a ball park economic comparison between the RTSC concept and a conventional CO₂ removal system by absorption using MEA. An extensive design and economic scenario was available for the latter case in the literature (Desideri and Paolucci 1999) for removal and recovery of CO₂ from a flue gas generated by a coal fired power plant of ~ 80 MW capacity. The flue gas was compressed to ~ 1.34 atm (~ 5 psig) using blowers to overcome the pressure drop in the MEA process train consisting of cooling towers, heat exchangers, absorption towers etc. The MEA was regenerated by steam heating.



 Table 1 Simulated performance of RTSC process

	TSC process	
	Case 1	Case 2
Feed gas conditions		
Temperature (°C)	150	200
CO ₂ mole fraction	0.15	0.15
Pressure (atm)	1.1	1.1
quantity of CO ₂ per cycle (mol/kg)	0.503	0.458
Effluent vent gas conditions		
Average CO ₂ concentration	~1.3%	~1.0%
Recovered CO ₂ product gas		
CO ₂ mole fraction	0.99+	0.99+
Temperature (°C)	450	500
Pressure (atm)	18.5	13.0
Recovery from Feed (%)	92.6	93.4
quantity of CO ₂ per cycle (mol/kg)	0.534	0.402
kg HP steam/kg CO ₂	1.1	0.44
kg LP steam/kg CO ₂	4.5	3.1
CO ₂ product compression		
Final CO ₂ Pressure (atm)	140	140
# of Compression Stages	3	4
Compression Power (MW)	2.3	2.8
Capital Cost for CO ₂	2.25	2.64
compressors, \$/ton CO2 captured		

The RTSC process had eleven parallel shell and tube $(2.54 \text{ cm diameter} \times 214 \text{ cm long})$ heat exchanger-type sorbers and the associated gas manifolds, and switch valves. The process cycle times were the same as those of simulation. The over-all dimension of each sorber was ~ 7.0 ft tall and ~ 20.9 ft in diameter. The shell and tube sorbers were designed to withstand the product CO₂ pressure using standard chemical engineering protocol. The CO₂ chemisorbent cost was assumed to be \$3.08/kg. The centrifugal CO₂ product compressors were designed using a maximum compression ratio of 2.0 per stage and an adiabatic efficiency of 0.75. The flue gas was compressed to 1.17 atm (2.5 psig) to overcome the pressure drop in the RTSC train. Blowers (efficiency = 0.5) were used for this purpose. The cost of electricity was assumed to be \$0.10/kWh. The cost of high and low pressure steam purge used in the processes was obtained from published literature (Smith and Varbanov 2005). The costs of the components of the RTSC system, the compressors, the heat exchangers, and the other accessories were estimated using a standard reference book for process design (Seider et al. 2003). An installation multiplier of 5 was used for all units. Table 2 shows the results where the costs are reported as \$/ton of compressed CO2 product from the RTSC process.

Table 2 Comparison of first pass economics of MEA and RTSC processes

Item	MEA (CO ₂ Product at 1.7 bar) Recovery = 90%	NTSC (150 °C sorption, 450 Desorption) (CO ₂ Product at 18.5 bar) Recovery = 92.6%
Separation Unit ^c	5.15	7.38
•	2.64	1.12
Feed Compressor or Blower	(blowers 5 psia feed, Cooling Tower, and HX)	(2.5 psia feed)
CO ₂ Product Compressor	5.37	2.25
Chemisorbent (\$1.40/lb)		0.18
MEA (\$ /lb)	1.06	_
Heat Exchangers (inter-stage cooling)	0.64	0.11
Total Capital Cost	14.86	11.32
Relative Cost	1.00	0.76
Power for Feed Gas Blower/Compressor ^d	8.52	Internally Supplied
Power for Product CO ₂ Compressor ^d	10.17	Internally Supplied
MEA Make up	1.5	_
Regeneration Steam Cost ^e	1.4	4.65
Total Operating Cost	21.59	4.65
Relative Cost	1.00	0.22
Additional Power Generated, MW	_	16.6
Power to Pre-Compress Feed (MW)	_	2.2
Power to Compress CO ₂ to 140 atm (MW)	_	2.3
Net Power Produced (MW)	_	12.1
Additional Heat for Thermal Swing \$/ton	_	45.23
Revenue Generated From Sale of Additional Power ($\frac{1}{N}$ ton CO ₂ , assumed price = $\frac{30.15}{N}$ Wh)	-	33.35
Net cost for additional Heating (\$/ton)	_	~11.00
Savings for enhanced CO ₂ recovery over plant life relative to MEA by RTSC case, assuming CO ₂ price of \$30/ton (\$/ton)	-	0.85

A first pass heat integration of the RTSC process was carried out for optimization. A certain amount of steam was generated from recovery of the sensible heats of the sorber vessels during step (e) of the RTSC process which was then used to generate electricity. A part of that electric energy was used to drive the blowers and compressors and the balance was credited. Table 2 reflects these credits. It may be seen from Table 2, that the RTSC concept can potentially reduce the capital and the operating costs of CO_2 removal and recovery from a flue gas, as well as its subsequent compression costs to \sim 140 bar for sequestration, compared to those by a conventional MEA absorption process. In particular, the reduction in the operating cost offered by the RTSC concept can be substantial.

It should also be mentioned here that the absorption and steam regeneration steps of a conventional MEA process for

removal of CO_2 are typically operated at pressures of ~ 1.34 and 1.7 atm and at temperatures of ~ 65 and 90 °C, respectively (Desideri and Paolucci 1999). The vapor pressures of MEA at 65 and 90 °C are, respectively, 6.9 and 29.4 mm of Hg. Consequently, the effluent gas steam from the MEA process during the regeneration step contains a substantial amount of MEA, even after partial condensation by cooling, which may require additional clean up (OSHA limit is ~ 3 ppm) before it is vented. This will incur additional cost which is not considered here. This problem is not applicable for the RTSC process.

It is, therefore, our conclusion that the proposed process for CO_2 removal and recovery by the RTSC concept is economically attractive considering that the penalty for emitting CO_2 to the atmosphere may be as high as \$50–100/ton.



10-20 MW

7.8%

Market amenability

It should be mentioned here that the cost comparison between RTSC and conventional MEA processes shown in Table 2 is for removing and recovering CO_2 from a ~ 80 MW coal fired power plant which is a relatively small-sized unit. The technical, economic, and practical feasibility of scalingup this technology to much larger power plants may not be foreseeable at this time. The adsorber sizes for larger plants will also be relatively big, and may require multi-train operation. But that will be true of any other currently available separation technology for removal and recovery of CO₂ from a flue gas. For example, an MEA process designed for removing CO₂ from the flue gas of a 320 MW power plant consisted of four parallel trains, each comprised of an absorber column (tray type) having a diameter of ~ 27.7 feet and a height of ~ 154 feet, and a stripping column (tray type) which was ~ 17 feet in diameter and ~ 131 feet tall (Desideri and Paolucci 1999).

About $\sim 35\%$ of all coal fired power plants and $\sim 70\%$ of all natural gas fired power plants in the U.S.A. are less than ~ 100 MW in size. Thus, there may a sizable number of applications of RTSC technology in the size range evaluated in this work. The size distribution of U.S power plants is given by the pie charts in Fig. 9 (http://eia.doe.gov, 2008).

5 Summary

Recently a rapid thermal swing chemisorption (RTSC) process concept was proposed for removal and recovery of CO₂ from a flue gas without pre-compression, predrying and pre-cooling, using a CO₂ selective, hydrophobic chemisorbent such as Na₂O promoted alumina (Lee and Sircar 2008). The process directly produced a compressed, pure CO₂ by-product gas for subsequent sequestration. New model simulations of the steps of the cyclic RTSC process show that the operation of the process at sorption and regeneration temperatures of 150 and 450 °C (case 1), respectively, instead of previously reported temperatures of 200 and 500 °C (case 2), substantially increase (a) the cyclic CO₂ productivity (moles of CO₂/kg of sorbent/cycle) of the process, and (b) the pressure of the by-product CO₂. Thus, the capital cost of the RTSC system and the energy and capital costs for subsequent compression of the product CO2 for sequestration at higher pressure are substantially reduced. However, larger quantities of high and low pressure steam are used for purging the sorber for case (1).

The relevant CO_2 chemisorption characteristics of the promoted alumina used in the simulation are reviewed, and key column dynamic simulations of CO_2 sorption and desorption steps for the process are analyzed. A first pass economic evaluation of the RTSC process was carried out and

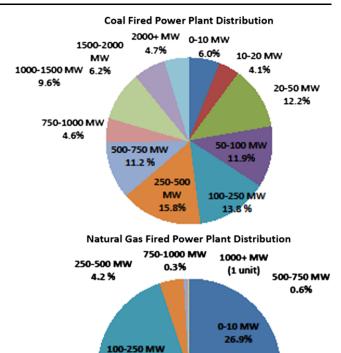


Fig. 9 Power plant size distribution in the USA

24.6%

50-100 MW

18.9%

compared with the published cost data for a conventional absorption process using MEA for removal and recovery of CO₂ from a flue gas (Desideri and Paolucci 1999). The RTSC process can potentially reduce the capital and operating costs for this application by a significant amount.

20-50 MW

References

Barchas, R., Davis, R.: The Kerr-McGee/ABB Lummus Crest technology for the recovery of CO₂ from stack gases. Energy Convers. Manag. **33**, 333–340 (1992)

Chakravarti, S., Gupta, A., Hunek, B.: Advanced technology for the capture of carbon dioxide from flue gases. In: First National Conference on Carbon Sequestration, Washington, DC, 15–17 May

Chue, K.T., Kim, J.N., Yoo, Y.J., Cho, S.H., Yang, R.T.: Comparison of activated carbon and zeolite 13X for CO₂ recovery from flue gas by pressure swing adsorption. Ind. Eng. Chem. Res. 34, 591–598 (1995)

Daniels, E.J., Body, B.J., Brockmei, N.F., Wolsky, A.M.: CO₂ capture from fossil fueled power plants. In: Meyers, R.A. (ed.) Encyclopedia of Environmental Analysis and Remediation, pp. 890–913. Wiley, New York (1998)

Desideri, U., Paolucci, A.: Performance modeling of a carbon dioxide removal system for power plants. Energy Convers. Manag. 40, 1899–1915 (1999)



- Fuderer, A., Rudelstorfer, E.: Selective adsorption of gases. U.S. Patent 3.986.849 (1976)
- Ishibashi, M., Ota, H., Akutsu, M., Kitamura, H., Ando, Y., Matsumoto, H., Kageyama, Y.: Removal of CO₂ from flue gases. Japan Patent 9,070,521 (1995)
- IEA GHG: Carbon dioxide capture from power stations. IEA Greenhouse Gas R&D Programme, Cheltenham, UK (2010). www.ieagreen.org.uk
- Jeong, K.K., Caram, H.S.: Modeling of heat recovery in novel thermal swing sorption enhanced reaction process for hydrogen and carbon dioxide production. AIChE J. (2010, in press)
- Karasaki, M., Iijima, M., Shigeaki, M.: Removal of CO₂ from flue gases. Japan Patent 7,313,840 (1995)
- Leci, C.L., Goldthorpe, S.H.: Assessment of CO₂ removal from power station flue gas. Energy Convers. Manag. **33**, 477–485 (1992)
- Lee, K.B., Sircar, S.: Removal and recovery of compressed CO₂ from flue gas by a novel thermal swing chemisorption process. AIChE J. **54**, 2293–2302 (2008)
- Lee, K.B., Beaver, M.G., Caram, H.S., Sircar, S.: Novel thermal swing sorption enhanced reaction process concept for hydrogen production by low temperature steam methane reforming. Ind. Eng. Chem. Res. **46**, 5003–5014 (2007a)
- Lee, K.B., Beaver, M.G., Caram, H.S., Sircar, S.: Chemisorption of carbon dioxide on sodium oxide promoted alumina. AIChE J. **53**, 2824–2831 (2007b)
- Lee, K.B., Beaver, M.G., Caram, H.S., Sircar, S.: Reversible chemisorbents for carbon dioxide and their potential applications. Ind. Eng. Chem. Res. 47, 8048–8062 (2008a)
- Lee, K.B., Beaver, M.G., Caram, H.S., Sircar, S.: Production of fuelcell grade hydrogen by thermal swing sorption enhanced reaction concept. Int. J. Hydrog. Energy **33**, 781–790 (2008b)

- Makita, T., Hirayama, K., Higuchi, K., Nawata, H., Izumi, J., Oshima, K.: Basic test results on CO₂ removal from boiler flue gas by PSA method. Enerugi Shigen **14** (1993)
- Matsumoto, H., Kitamura, H., Kamata, T., Nishikawa, N., Ishibashi, M.: Fundamental study of CO₂ removal from thermal power plant flue gas by hollow-fiber gas-liquid contactor. Kagaku Kogaku Ronbunshu **18**, 804–812 (1992)
- Mimura, T., Shimojo, S., Suda, T., Iijima, M., Mitsuoka, S.: Research and development on energy saving technology for flue gas carbon dioxide recovery and steam system in power plant. Energy Convers. Manag. **36**, 397–400 (1995)
- Riemer, P.: Greenhouse gas mitigation technologies, an overview of the CO₂ capture, storage and future activities of the IEA greenhouse gas R&D programme. Energy Convers. Manag. **37**, 665–670 (1996)
- Seider, W.D., Seader, J.D., Lewin, D.R.: Product & Process Design Principles: Synthesis, Analysis and Evaluation, pp. 505–557. Wiley, New York (2003)
- Sircar, S.: Rapid thermal swing adsorption. U.S. Patent 20030037672 (2003)
- Sircar, S., Golden, C.M.A.: PSA process for removal of bulk CO₂ from a wet high temperature gas. U.S. Patent 6,322,612 (2001)
- Smith, R., Varbanov, P.: What's the price of steam? Chem. Eng. Prog. 29–33 (2005)
- Suzuki, M.: Application of adsorption technology for environmental control. In: Levan, M.D. (ed.) Proceedings of 5th International Conference on Fundamentals of Adsorption. Pacific Grove, CA, pp. 1–14. Kluwer, Boston (1996)

