RESEARCH NOTES

Sorbent Cost and Performance in CO_2 Capture Systems

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Power plants are prime candidates to apply CO_2 capture for final storage as a mitigation option for climate change. Many CO_2 capture concepts make use of a sorption−desorption cycle to separate CO_2 from flue gas or O_2 from air. These include commercial absorption processes, as well as processes using new sorbent formulations, adsorption, and high-temperature chemical looping cycles for CO_2 and O_2. All of these new processes must confront the large scale of carbon flows typical in a power plant. In this work, a common mass balance for all of these processes is used to define a parameter that highlights the minimum sorbent performance required to keep sorbent makeup costs at an acceptable level. A well-established reference system for which reliable commercial data exist (absorption with monoethanolamine, MEA) is used as a technoeconomic baseline to show that some of the sorbents being proposed in the open literature might need to be tested under laboratory conditions for tens of thousands of sorption−desorption cycles before they can be further considered as viable options for CO_2 capture from power plants.

Introduction

The capture of CO_2 from large stationary sources with storage in geological formations is being widely studied as a mitigation option to achieve deep cuts in CO_2 emissions from fossil fuels.[1] In the longer term, if biomass is used as the primary fuel, power plants implementing CO_2 capture and storage technologies could even become effective net sinks of CO_2 from the atmosphere.[2]

There are a wide range of concepts for CO_2 capture that make use of a sorption−desorption cycle to separate CO_2 from a flue gas (postcombustion capture) or from a fuel gas (precombustion capture). These include commercial processes using amines and physical sorbents, as well as new sorbent formulations for absorption, adsorption, and high-temperature chemical looping cycles for CO_2 and O_2. The general scheme for all of these processes can be represented in Figure 1 in terms of molar flows of CO_2 (F_{CO_2}), sorbent flowing in the capture−regeneration loop (F_R), and sorbent makeup flow (F_0). A similar scheme would be valid for the O_2 chemical looping cycles. A makeup flow of sorbent (F_0) is required to compensate for the natural decay of activity and/or sorbent losses during many sorption−desorption cycles. It must be emphasized that this decay is, to some extent, always unavoidable because of a wide range of chemical and physical interactions of the
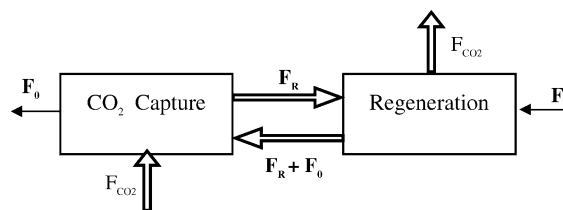
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Figure 1. General scheme of a CO_2 capture system using a sorption−desorption cycle.

sorbent inside the reactors and transport lines and the sorbent losses associated with gases leaving the system.

CO_2 capture systems are in a class by themselves because the mass flows of sorbent circulating in a system such as Figure 1 are very large, at least large enough to match the large molar flow of CO_2 being processed in the power plant. For the sake of comparison, it is useful to compare this to the size and scale of the sorbent flows in a flue gas desulfurization unit. The CO_2 molar flow in the flue gases of a power plant is typically 2 orders of magnitude higher than the SO_2 molar flow, even when processing high-sulfur fuels. Therefore, extremely low values of F_0/F_R are required in order to keep the sorbent makeup cost (and any other costs related to the disposal of spent sorbent) within reasonable limits. This translates into very demanding conditions for the sorbent performance required in the sorption−desorption chemical loop, as will be discussed below. It is beyond the scope of this work to review the wide range of sorbents proposed for capturing CO_2 in power plants and their relative benefits arising from specific process conditions. Our aim is only to establish a common link between the expected cost of a sorbent and its expected performance in the sorption−desorption loop of Figure 1.

Table 1. Outline of Sorbent Characteristics, Estimated Unit Prices, and Performance Criteria ($N_{1/2}$) To Match the Performance of a MEA-Based Absorption System, Used as a Baseline Reference[a]

sorbent	C_s ($/kg)	M_s (kg/mol)	b ($/mol of CO_2)	F_R/F_{CO_2} ($/mol of CO_2)	C_s^* ($/mol of CO_2)	F_0/F_R ($/mol of CO_2)	$N_{1/2}$ ($/mol of CO_2)	ref
MEA[3]	1.250	0.061	2	3.57	0.544	0.000 152	4561	3
$CaCO_3$ (crushed stone)[5]	0.005	0.1	1	3[b]	0.0015	0.0552	13	10−14
K_2O (potash)[5]	0.155	0.0942	1	3[b]	0.044	0.001 89	367	7
Na_2CO_3 (soda ash)[5]	0.105	0.106	1	3[b]	0.033	0.002 48	280	8, 25
Li_2CO_3 (lithium)[5]	4.47	0.0739	1	3[b]	0.99	0.000 0835	8299	18, 19
active carbons	0.5	0.5[c]	1[d]	1[d]	0.25	0.000 75	925	6−8
zeolites	1.0	0.2[c]	1[d]	1[d]	0.20	0.000 375	1849	6−8
hydrotalcites	2.0	2.0[c]	1[d]	1[d]	4.0	0.000 020 7	33505	9
Fe_2O_3 (iron ore)[5]	0.026	0.160	2[e]	3	0.025	0.003 32	209	20
Co (cobalt)[5]	33.43	0.059	2[e]	2	7.9	0.000 010 5	66001	21
Cu (copper)[5]	1.812	0.1271	2[e]	2	0.46	0.000 18	3858	21
Ni (nickel)[5]	8.638	0.0587	2[e]	2	2.0	0.000 040 8	16986	21−24
Mn (manganese)[5]	0.592	0.055	6[e]	2	0.39	0.000 21	3270	21

[a] In all data from ref 5, prices are for the year 2000. The sorbent is considered to be the raw material in the form at which the price is given. [b] Assuming an average carbonation conversion of $^1/_3$. [c] Assuming 2 mol of CO_2 adsorbed/kg of active carbon, 5 mol of CO_2/kg of zeolite (see ref 5), and 0.5 mol of CO_2/kg of hydrotalcite (Figure 4 of ref 9). [d] The definition of M_s for these sorbents already gives the mass of sorbent required to absorb 1 mol of CO_2. [e] This is per mole of O_2 transported, assuming $2Fe_2O_3 = 4FeO + O_2$, $2Me + O_2 = 2MeO$ (for Me = Co, Cu, Ni), and $6MnO + O_2 = 2Mn_3O_4$.

Discussion

Table 1 shows several characteristics for the monoethanolamine (MEA) reference system and a number of alternative solid sorbents being proposed in the open literature. These have been divided into three generic groups: compounds containing Li, Na, K, or Ca that can react with CO_2 to form a carbonate; two generic substrates for adsorption-based systems (that may or may not contain some of the metals listed above as active components); and a group of metals with oxides suitable for use in O_2 chemical looping cycles (that may or may not require support on a solid substrate). A series of references (which is not intended to be comprehensive), illustrative of studies where these sorbents are proposed for CO_2 capture application, are also included in Table 1.

The cost of CO_2 capture is the sum of several terms encompassing the large capital costs associated with the capture plant, the new operating and maintenance costs, the cost of additional fuel resources needed to compensate for the efficiency penalty introduced by the capture plant, and the cost of CO_2 compression. Any new capture system following the scheme of Figure 1 will have to compete economically with existing based systems that already can deliver total CO_2 capture costs below $40/tonne of CO_2 separated.[3] Because the cost of the makeup flow of sorbent in Figure 1 is only one component of the total capture cost, to keep total capture costs at reasonable values, the sorbent makeup costs cannot be excessive.

A commercially proven CO_2 capture system employing absorption with MEAs can be used as a benchmark. Rao and Rubin[3] have shown that in order to minimize the overall cost of CO_2 capture the makeup cost of MEA must be kept reasonably low. Thus, strict operational requirements are necessary for the desulfurization unit in a power plant in order to keep concentrations of SO_2 in the flue gases below 10 ppmv so as to minimize the sorbent makeup that otherwise deactivates rapidly because of reactions with SO_2 and other pollutants in the flue gases. If this is the case for MEA, a well-known chemical that has been used in industry for many decades, a comparably low sorbent makeup cost per unit of CO_2 separated will be essential for any other sorption−desorption system for CO_2 capture that follows the

scheme of Figure 1. We can define the cost of sorbent per kilogram of CO_2 removed (COS) using the notation of Figure 1, as follows:

$$COS = \left(\frac{F_0}{F_R}\right)\left(\frac{F_R}{F_{CO_2}}\right)\frac{bM_s}{M_{CO_2}}C_s = \left(\frac{F_0}{F_R}\right)\frac{C_s^*}{M_{CO_2}} \quad (1)$$

We note that the group $C_s^* = M_s C_s b(F_R/F_{CO_2})$ is a suitable variable to compare cost of different sorbents because this is the cost of the amount of sorbent required to react (or absorb) 1 mol of CO_2 (or O_2). Furthermore, because the ratio F_R/F_{CO_2} is related to the expected average conversion of the sorbent, and this can be estimated from laboratory data, C_s^* can be readily estimated when the price of the sorbent, C_s ($/kg of sorbent), is known. For a typical MEA-based system,[3] $b = 2$, $M_{MEA} = 0.061$, $C_s = $1.25/kg of MEA, and the makeup flow is 1.5 kg of MEA/tonne of CO_2. The latter corresponds to values of $F_0/F_R = 0.000\ 152$ and $F_R/F_{CO_2} = 3.57$ (based on a lean solvent loading of 0.22 mol of CO_2/mol of solvent and a maximum rich loading of 0.5 mol of CO_2/mol of solvent, which gives $F_R/F_{CO_2} = 1/[0.5 − 0.22] = 3.57$). This translates into $C_s^* = 0.544$ $/mol of CO_2 and a reported sorbent makeup cost for a large-scale MEA-based system of around $COS_{MEA} = $0.0019/kg of CO_2, or approximately $2/tonne of CO_2 captured.

The rationale behind the present study is that the sorbent performance in any new CO_2 capture system should be of sufficient quality to ensure that sorbent makeup costs are comparable to those being achieved today for a commercially proven technology, i.e., absorption with MEA. This is assumed to be a necessary (but not sufficient) condition because we consider it unlikely that other benefits arising from a new, unproven technology—such as a major reduction in process energy requirements—could compensate for much higher values of this component of total capture cost if the overall cost of CO_2 capture is to be low. Nevertheless, one could accommodate the potential for such improvements by considering costs on the basis of CO_2 avoided rather than CO_2 captured. This measure accounts for the effects of process energy penalties on the total CO_2 emissions associated with producing a unit of product (e.g., a kilowatt·hour of electricity). For regenerative

sorbents, energy requirements are a major contributor to overall capture costs.[3] Thus, we define

$$(COS)_a = COS(R_{a/c}) \tag{2}$$

where $R_{a/c}$ is the ratio of \$/tonne avoided to \$/tonne captured. It can be shown that this ratio is a simple function of the process energy penalty and CO_2 capture efficiency. For current MEA systems applied to power plants, this ratio is roughly 1.4.[3] Thus, as a quantitative guideline for evaluating different sorbents, we suggest that an acceptable level of sorbent losses for any new capture system using regenerable sorbents should be such that

$$(COS)_a \leq (COS_{MEA})_a \tag{3}$$

or if process energy requirements are not significantly different,

$$COS \leq COS_{MEA} \tag{4}$$

Because some precommercial capture systems[4] can achieve total removal costs of less than \$20/tonne of CO_2 captured, or about \$25/tonne of CO_2 avoided, the example above for MEA suggests that sorbent makeup costs for emerging processes cannot exceed more than a few dollars per tonne if the process is to be economically competitive. There are no reliable estimates of sorbent makeup costs for new, noncommercial systems because there is no information on the key flow ratios in eq 1, and the unit cost of sorbent is also uncertain. In particular, the flow ratio F_0/F_R can only be estimated when sufficient information is available on the performance of the sorbent at realistic process conditions and at a relevant number of sorption−desorption cycles. The unit sorbent cost is also an important variable in eq 1. Reasonable minimum values can be assumed for some of the sorbents listed in Table 1, considering the price of similar solids (for example, industrial active carbon-based adsorbents or synthetic catalyst based on zeolites) or the market price of the raw material (minerals and metals). A common source has been used to estimate the prices of the raw materials.[5]

Employing the minimum unit price for the sorbents of Table 1, we can now use the MEA system as a reference case to ascertain how much the sorbent makeup flows must decrease (or increase) to keep COS equal to COS_{MEA}. The results, with some reasonable assumptions adopted for the degree of utilization of the sorbent (F_R/F_{CO_2}), are also listed in Table 1 in the form of the sorbent recycle ratio F_0/F_R. While this ratio alone gives a direct measure of the quality of sorbent required in the system, we consider it useful to also display another variable typically used when investigating properties of a sorbent for a multicycle operation: the number of sorption−regeneration cycles, N, experienced by a particle of sorbent.

The mass balance of Figure 1 can be written to yield the fraction, r_N, of sorbent (molecules, particles, or any other mass unit) that has gone through the sorption−desorption cycle *exactly* N times:

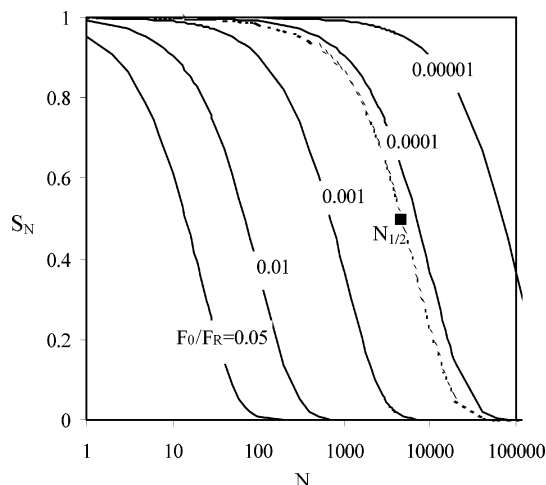$$r_N = \frac{F_0 F_R^{N-1}}{(F_0 + F_R)^N} \tag{5}$$

Figure 2. Sorbent mass balance in a sorption−desorption system for CO_2 capture applications, representing the fraction of sorbent units that have cycled the system more than a given number of times, N, for different sorbent makeup ratios. The dotted line is the MEA baseline reference case $F_0/F_R = 0.000\ 152$.

Therefore, the fraction of sorbent, S_N, that has been cycling in the sorption−desorption system *at least* N times is

$$S_N = 1 - \sum_{N=1}^{N} r_N = \left(\frac{1}{\dfrac{F_0}{F_R} + 1} \right)^N \tag{6}$$

Figure 2 represents eq 6 for a range of typical ratios F_0/F_R expected in a CO_2 capture system in power plants. As mentioned above, for the reference case of a MEA-based system, the ratio F_0/F_R is as low as 0.000 152, as long as the SO_2 concentration in the flue gas is kept below 10 ppmv.

To better handle the mass balance represented by eq 6, we can define a characteristic number of cycles, $N_{1/2}$, that indicates that 50% of the sorbent units (molecules or particles) have completed the sorption−desorption cycle more than $N_{1/2}$ times.

$$N_{1/2} = \frac{\ln 2}{\ln\left(\dfrac{F_0}{F_R} + 1\right)} \tag{7}$$

For the case of the typical MEA-based system described earlier, $N_{1/2}$ is 4561. The meaning of this parameter for a molecule like MEA is that, on average, it cycles approximately 4500 times before it is lost from the system or encounters a poison that irreversibly deactivates it. If there are no poisons and no material losses, the molecule can, in principle, absorb and desorb CO_2 indefinitely. However, for systems using gas−solid reactions (all of the remaining sorbents in Table 1) and continuous transport of solid between reactors, the implications of high values of $N_{1/2}$ are very different. Solids reactants are known to attrite, escape from solid capture devices (cyclones, filters, etc.), change properties during cyclic reactions, be covered by surface deposits, or suffer chemical poisoning of the active part of the sorbent. All of these decay processes are bound to increase and to accumulate in the particles as the number of cycles increases. As a consequence, it seems reasonable to expect that the testing of new sorbents to
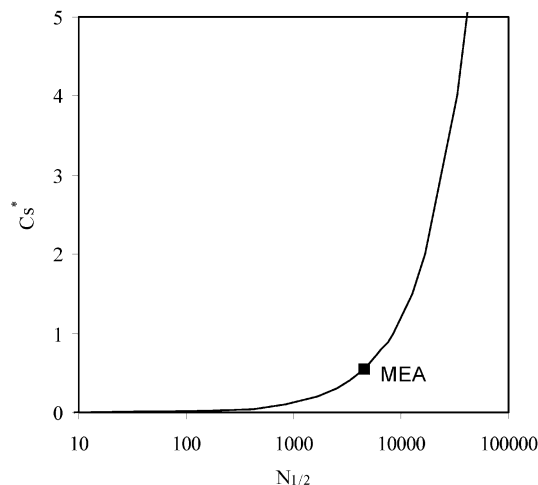
Figure 3. Required performance index of a sorbent ($N_{1/2}$) against the manufacturing cost of a unit of sorbent needed to react with 1 mol of CO_2 or O_2 (C_s^*). This is in order to keep sorbent makeup costs below the MEA reference case (black square). Different cost thresholds will generate different curves.

determine their critical properties for reactor design (reactivity, selectivity, CO_2 capture capacity, mechanical strength, etc.) should be carried out under conditions that can be extrapolated, at least, to the region of $N_{1/2}$ cycles. Figure 3 shows in a logarithmic scale how the requirements in terms of the quality of the sorbent ($N_{1/2}$) increase with the unit cost of the sorbent C_s^*. Application of this criterion to sorbents of unit cost comparable to that of MEA is already a major technical challenge ($N_{1/2}$ is already very high for MEA). We can illustrate this point by discussing the three main groups of solid sorbents compiled in Table 1. Synthetic adsorbents and catalyst based on a substrate like active carbon or zeolites are common in many industrial separations. When their target is to remove a minor contaminant in a large gas flow, or to produce a high value-added product, the cost of the sorbent referring to 1 tonne of the impurity removed, or to the product produced, is allowed to be high. However, this cannot be the case in CO_2 capture systems, where the flow of the impurity is very large and the price of the product has to be very low. If CO_2 sorbents are to be developed using these substrates,[6–9] their performance needs to be demonstrated after hundreds to thousands of sorption–desorption cycles. Many well-established, large-scale separation processes in industry today, often used as examples to introduce a new CO_2 separation concept, could not exist if the "price" of the separated product had to be as low as 1 tonne of CO_2 separated from a power plant.

Natural limestones have also been investigated as a regenerable sorbent for CO_2 separation.[10–14] However, the sorbent cannot maintain a high capture capacity beyond 20 cycles, and large quantities of limestone makeup are required for CO_2 control.[15] This can only be acceptable because of the extremely low price of crushed limestone and because the exhausted calcines might have some downstream value as a feedstock for the cement industry. Equally, we have, for the sake of simplicity, ignored the loss of sorbent due to the formation of $CaSO_4$, whose quantitative removal would represent an advantage for such systems because excess sorbent would no longer be required to remove SO_2. It is obvious that improved performance with synthetic or reactivated sorbents would be highly beneficial for the

lime carbonation–calcination system.[16,17] However, these new sorbents will have to be tested for reactivity and capture capacity at a much higher number of cycles, given the previous analysis.

Similar comments apply to other sorbents based on lithium carbonates[18,19] or to sorbents for O_2 chemical looping concepts.[20–24] In the latter case, the cheapest O_2 carrier in Table 1 (Fe_2O_3) has significant limits in terms of performance,[20] so that NiO-based carriers are increasingly considered as a much more promising route for these systems.[21,22,24] However, because Ni is an intrinsically expensive material, the resulting value of $N_{1/2}$ is around 17 000, and this means that the key information needed to assess the performance of these materials in these applications must be obtained after many oxidation–reduction cycles.

In general, we propose that new sorbents for CO_2 capture applications should always undergo testing to the extent suggested by eq 7 in order to adequately assess the cost of sorbent makeup in any large-scale system used for CO_2 capture. While this condition is necessary for reliable economic evaluations, we note that systems using "cheap" sorbents[10–14,25] also may face a wide range of other problems that could prevent or limit their near-term application.

Conclusions

There is scope for technology breakthroughs to reduce the cost of CO_2 capture with respect to well-known, precommercial systems, and a range of interesting sorption–desorption systems are being proposed in the open literature to achieve this objective. However, to develop and characterize these sorbents under realistic process conditions, one must bear in mind the need to keep the cost of sorbent makeup flow (which will always be required to maintain the necessary activity in the sorption–desorption loop) at a reasonable level if overall goals of low capture costs are to be achieved. To properly evaluate proposed new process designs, sorbent properties should be obtained under conditions resembling the state of the sorbent after a representative number of sorption–desorption cycles that may vary from tens to tens of thousands of cycles, depending on the unit cost of the sorbent.

Acknowledgment

Notation

b = moles of sorbent required to stoichiometrically react with 1 mol of CO_2 (or O_2)

COS = cost of sorbent makeup per tonne of CO_2 removed ($/kg of CO_2)

$(COS)_a$ = cost of sorbent makeup per tonne of CO_2 avoided ($/kg of CO_2)

C_s = unit cost of sorbent ($/kg)

C_s^* = cost of the amount of sorbent necessary to absorb or react with 1 mol of CO_2 (or O_2)

($/mol)

F_0 = makeup flow of sorbent in Figure 1 (mol/s)

F_{CO_2} = flow of CO_2 removed by the capture system of Figure 1 (mol/s)

F_R = flow of sorbent required to react with the flow of CO_2 (mol/s)

M_{CO_2} = 0.044 kg/mol

M_s = molecular weight of the sorbent (kg/mol)

N = number of times that a unit of sorbent has cycled the sorption−regeneration loop of Figure 1

$N_{1/2}$ = characteristic number of cycles that indicates that 50% of the sorbent has completed the sorption−desorption cycle more than $N_{1/2}$ times

r_N = fraction of sorbent that has completed the sorption−desorption cycle N times

$R_{a/c}$ = ratio of \$/tonne avoided to \$/tonne captured

S_N = fraction of sorbent that has cycled the sorption−desorption system more than N times

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