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Comparative study of optimized purge flow in a CO_2 capture system using different sorbents

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Abstract

One of the most promising options for CO_2 capture in large power generation facilities is the system based on the CO_2 sorption loop. This method has gained rapid importance due to promising carbonator CO_2 capture efficiency, the existence of low cost sorbents and the fact that no gas pre-treatment unit is needed before entering the system. The sum of these features results in a competitively low cost CO_2 capture system when using low cost natural sorbents. Different regenerable sorbents are being investigated for large-scale CO_2 capture purposes and high temperature Mg-based, Li-based and Ca-based sorbents are considered as suitable candidates. This study considers the applicability of lithium orthosilicatum, hydrated limestone and raw natural limestone. A basic configuration that makes use of two interconnected circulating fluidized beds (carbonator and calciner) has been studied. Among the key variables that influence the performance of these systems, the carbonation conversion of the sorbent and the heat requirement at calciner are the most relevant. Both variables are mainly influenced by sorbent/CO_2 ratio and make-up flow (purge) of solids. Purge is necessary to mitigate the sorbent deactivation. Large sorbent/CO_2 ratios improve the carbonation conversion but also increase the cost of the system due to a more intensive solid circulation. Large make-up flow also improves the extent of sorption phenomena and hence the CO_2 capture, but increases the heat demand at calciner and the fresh sorbent cost. The aim of this paper is to calculate the optimum make-up flow of fresh sorbent and sorbent/CO_2 ratio for a set of these regenerable sorbents in order to minimize the capture cost of the system integrated into a power plant. Resulting optimal values are compared to assess the energetic performance and CO_2 capture cost of the cycle for each sorbent material.©

Keywords: CO_2 capture; sorbent looping; limestone; coal composition

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1. Introduction

The need to reduce anthropogenic emissions of CO_2 is globally agreed and represents the driving force to reconsider the current technologies used for power generation [1]. However, fossil fuels are predicted to be the main energy sources during the next decades [2]. CO_2 emission reductions can occur as a result of an increase in energy efficiency, substitution of low- or non-carbon fuels, or by the capture and storage of CO_2. CCS has been pointed out by the Intergovernmental Panel on Climate Change (IPCC) as a suitable technology for mitigating climate change in a mid-term [3].

Absorption processes based on chemical solvents, mainly amines, are feasible and commercially available options to carry out CO_2 capture since they offer high capture efficiency and selectivity at acceptable costs. Main

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drawbacks of this technology are the need of flue gas pre-treatment and energy penalties at low temperatures which imply low quality heat flows [3,4]. Among emerging adsorption processes of CO_2 capture, high temperature solid sorbents looping are promising since no pre-treatments are required and plant repowering may be achieved by means of energy integration into the plant [3,5].

This CO_2 separation process relies on the use of sorption/desorption looping technologies based on equilibrium reactions which allow solid sorbent regeneration. Exothermic carbonation reaction (sorption) and endothermic calcination reaction (desorption) occur at different temperatures and they need different chemical environments to take place. Thus, as firstly suggested by Shimizu et al. [6], two interconnected reactors are required to have a continuous process. The use of solids means that, in many cases, fluidized bed systems will represent an optimal method of allowing large amounts of solid material to be transferred from one chemical environment to another [5]. The proposed system makes use of two circulating fluidized beds (CFB) to improve solid circulation, residence time and gas-solid mixing and to control temperatures.

As presented in figure 1, flue gases leaving the reference plant enter the carbonator and after sorption reaction the clean gas flux is released. Carbonated sorbent is regenerated in the calciner producing a high concentrated flux of CO_2. A subsequent compression process is needed in order to make CO_2 transport possible, proximately at 120 bar and 80 ºC [8].

The adsorption capture process implies a strong energy penalty mainly associated with regeneration process, high temperatures and oxygen production for oxyfuel combustion. However, the waste heat fluxes from the cycle may be integrated in an existing power plant improving the efficiency or in a new design steam cycle to increase power plant net power output.

Although a fast and complete calcination reaction can be assumed, the reverse carbonation reaction is more challenging. Therefore, sorbent/CO_2 molar ratio represents a key factor in carrying out capture with success. High sorbent/CO_2 ratio directly increases carbonation reaction efficiency but it also implies high energy penalties since a larger sorbent internal circulation enhances calcination heat requirements [7]. Another relevant factor is the sorbent used in the cycle whose chemical and physical properties will influence on the performance of the system.

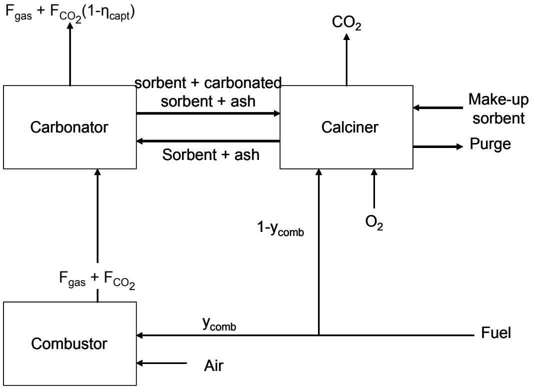

Figure 1. Carbonation-Calcination Looping[7]

In this study, several high-temperature sorbents have been assessed focusing on both energetic efficiency and economics. The addition of a postcombustion capture system in a reference power plant and its subsequent integration in a supercritical cycle have been simulated. Regenerable sorbents feasibility has been evaluated by comparing the minimum avoided CO_2 costs. The efficiency of CO_2 capture systems after energetic integration with the supercritical plant (optimized for limestone as sorbent) has also been assessed for each sorbent.

2. CO_2 sorbents behaviour

Many metal oxides exhibit the carbonation and calcination reactions under specific operational conditions. A wide range of regenerable solid sorbents have been investigated in the literature. Apart from CaO, gas-solid carbonation of other sorbents has not been studied in depth. They may be classified as natural or synthetic. Natural sorbents, like $CaCO_3$ [10-1], $NaHCO_3$ [16,14] or $MgCO_3$ [10,15] present faster reaction kinetics and usually are cheaper and widely available. Otherwise, synthetic ones, like Li_2ZrO_3 [16,17] or Li_4SiO_4 [10,18] with slow carbonation kinetics have extremely high production costs that must be offset with a much longer durability to compete with natural sorbent current prices. Depending on the calcination temperatures of the solid adsorbent, they are also classified as high temperature, i.e.$CaCO_3$, 930º C [7,20], or low temperature sorbents, i.e. $NaHCO_3$, 120-170º C [13,14]. High temperature sorbents will be more suitable for integration of the capture cycle with the power plant. There exist several criteria to determine the potentiality of a sorbent to be used in post-combustion capture processes integrated into a power plant:

i) High reaction rates in the temperature range of flue gases from the reference plant or, in case of boiler-carbonator configuration, proper range of temperature for fuel combustion. Therefore, low temperature sorbents present a less favourable position for integration into a new power plant because recoverable heat from the capture process has a lower quality. On the other hand, sorbents that decompose beyond 1000 °C are not adequate for integration because their calcination would impose severe energy penalties and restrictions with constructive materials.
ii) Low temperature spring between carbonation and calcination reaction minimizes the energy penalties to heat up the solid mass circulating between both reactors.
iii) Low specific cost of the sorbent are desirable.
iv) High specific adsorption ability of the sorbent diminishes the required inventory within the loop, simplifying operation and reducing energy consumption.
v) Long term stability of the sorbent has a strong influence on final cost of avoided CO_2. Minimization of required purge in the system due to slow degradation and high residual activities of the sorbents may counterbalance specific costs. The cyclic sorption behaviour of these sorbents is described by expression (Eq. 1) where Xr is the residual activity of the solid sorbent after a long number of cycles [7].

$$X_N = \frac{1}{\frac{1}{(1-X_r)} + kN} + X_r \qquad \text{(Eq. 1)}$$

Three different sorbents have been chosen to simulate the carbonation looping and the integration of this capture cycle with a new supercritical power plant: limestone, hydrated limestone and lithium orthosilicate. Limestone is the most spread solid sorbent for looping cycles of CO_2 capture [7,8,12,20] and is considered the baseline case to compare the results in this study.

Steam reactivation of lime is a promising method to improve spent sorbent performance [21-23]. Hydrated limestone presents a 600% increase in the residual carbonation activity [23] as a consequence of the reduced sintering phenomena affecting the sorbent morphology. Reactions taken into consideration after calcination are i) the exothermic hydration of lime and ii) carbonation of calcium hydroxide. An extra energy penalty corresponding to lime hydration has been considered and calculated as the coal consumption required to generate the steam which hydrates the whole lime inventory in the system. Make-up flow fed to the calciner is natural limestone. Once the solid material is calcined, lime is hydrated in a different reactor where generated steam is introduced.

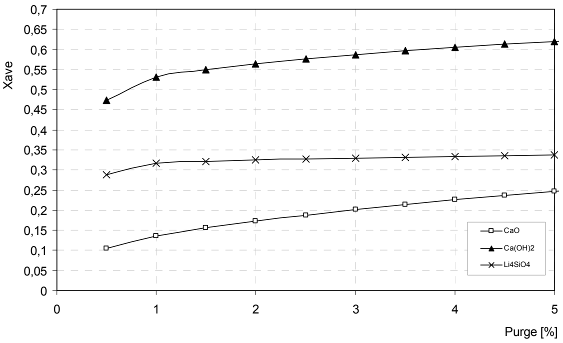

Figure 2. *Xave for different sorbents*

Lithium orthosilicate, synthetic sorbent, also presents a residual activity four times higher than natural limestone. Fresh sorbent introduced in the regeneration reactor is assumed to be a 1:1 molar mixture of Li_2CO_3 and Li_2SiO_3. Specific cost of the fed mixture was assumed to be 150 €/kg.

Parameters of the model were modified for each sorbent, their values are presented in table 1. Decay of activity with time for the simulated sorbents has been modelled following Eq. 1 and parameters adjusted, X_r and k, considering data from open literature [7,10,23,]. Heat of carbonation reaction for each considered sorbent is different and will determine the consumption of fuel in the regeneration stage of the sorbent having a strong influence on the energetic balance of the integrated system [7,10,23,24]. Also the stoichiometric of the carbonation reaction may vary from one sorbent to another, modifying the definition of the percentage of solid sorbent population with N cycles in the loop, r_N. However, the selected sorbents have 1:1 stoichiometric. Figure 2 represents the evolution of this parameter when varying purge percentage in the system for each sorbent under study. Physical properties of the involved species have been taken from [26-28].

Table 1. Simulated CO_2 sorbents properties

Sorbent	Carbonation reaction	ΔH^o_{car}	T_{carb}	T_{regen}	Xr	k	Cs	Ref.
$CaCO_3$	$CaO + CO_2 \leftrightarrow CaCO_3$	168 kJ/mol	650º C	930º C	0.075	0.520	6 €/tonne	7
$Ca(OH)_2$	$CaO + H_2O \leftrightarrow Ca(OH)_2$	73 kJ/mol	780º C	960º C	0.480	0.505	6 €/tonne	21-23,24
	$Ca(OH)_2 + CO_2 \leftrightarrow CaCO_3$	105 kJ/mol						
Li_4SiO_4	$Li_4SiO_4 + CO_2 \leftrightarrow Li_2SiO_3 + Li_2CO_3$	142 kJ/mol	500º C	710º C	0.309	4.590	150 €/kg	10, 25-28

3. Technical and economical model of the integrated cycle

In order to evaluate the technical and economical consequences of using these sorbents for CO_2 capture in power plants, it is necessary to integrate the sorbents chemical behaviour, mass and heat flows into a steam cycle that reduce penalties associated with capture processes.

The objective function for optimization is the avoided CO_2 cost, as defined in [3] (€/tCO_2). The independent variables are the purge of sorbent material and the molar ratio sorbent/CO_2.This purge is needed to compensate conversion decay of sorbent and other solid losses in the system. The purge in the system has been defined as a percentage over the total mass flow leaving the calciner when no purge is considered. Constraint equations include the sorbent characteristics: heat of reaction, carbonation and regeneration temperatures, residual activity of the sorbent, sorbent degradation velocity, and sorbent costs; and also include mass and energy balances of the proposed integration of waste heat flows from capture and compression processes into a new supercritical steam cycle. The optimization process of the energetic integration has been only developed for natural limestone as sorbent in the capture cycle. The resulting configuration of heat exchangers has been used to simulate the system considering different sorbents in the capture cycle. Obtained results in those cases do not represent the lowest possible cost of tonne CO_2. Further work should be done to correctly address the real heat flows from the capture cycle for different sorbents into the supercritical cycle.

The reference power plant generates 500 MWe and the net efficiency of the unit is 40.32% LHV. It has been supposed for calculations a high-rank coal, table 2, with negligible sulphur content. Flue gases flow of the reference power plant, 546.80 kg/s, 21.72%w CO_2 is fed to the carbonation loop. A detailed description of integration could be found elsewhere [8,29]. Summarising, waste heats from carbonator and calciner are used to design the high-pressure equipments of a supercritical cycle. Heat recovery steam generators take advantage of the flue gases from carbonator at 650 ºC and from calciner at 930 ºC. A solid-gas heat exchanger is used to reduce the purge temperature from the corresponding temperature operation of the purged reactor down to 180º C. When CaO is hydrated, an extra energetic penalty, calculated as the extra coal required to generate the steam for hydration, has been included in the model. Intercooling CO_2 compression heat is used in low-pressure heat exchangers in the condensate section of the steam cycle. No steam turbine bleeds are necessary for water preheating due to heat integration with CO_2 compression train and flue gases from calciner and carbonator used as high-pressure heaters.

An air separation unit (ASU) is needed to provide oxygen for oxyfuel combustion. The required oxygen flow is ranged from 36 to 107 kg/s. A typical value of 220 kWh/tonne O_2 has been taken for ASU power consumption [3].

Reference plant and capture plant capital costs have been taken from literature [30]. Amortization costs, fixed costs, O&M costs, fuel costs and sorbent costs of the capture plant have been considered. The extra auxiliary consumption in the capture system is mainly generated by the ASU whose electrical consumption ranges broadly depending on the case under study. The main assumptions for these economic calculations are an interest rate of 8.78%, 25 year project life (fixed charge factor of 10%); fixed cost has been taken as 1% and yearly maintenance costs around 2% of the overall capital cost of the total capital requirement; coal cost of 1.43 €/GJ; capital investment of 1100 €/kWgross for the reference power plant and a conservative value of 2070 €/kWgross (2383 €/kWnet) according [31] for the capture system comprising an oxy-CFB calciner, a CFB carbonator and the steam cycle. The total investment is 550 M€ for the reference case with a cost of electricity (COE) of 31.90 €/MWh and emission of 905.10 kgCO_2/MWh. These figures change for the different carbonation loop sizes obtained depending on type of sorbent, sorbent/CO_2 molar ratio and purge percentage.

4. Comparative results

Thermal and economical baseline results are those corresponding to the CO_2 capture cycle using natural limestone as sorbent, figure 3. The optimal integration of the capture process heat flows with the new supercritical cycle leads to a minimum value of 13.80 €/CO_2 tonne for low purge percentages, 2.50%, and high molar ratio fresh sorbent flow over CO_2 flow, 5. The results for limestone slightly differ from the operating conditions presented in [8] because a less optimistic expression has been used to describe sorbent activity, X_N. The lower residual activity and faster deactivation, the higher purge required in the system.

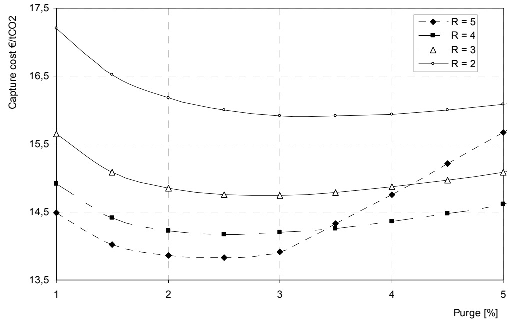

Figure 3. CO_2 avoided cost for different R and purge. Limestone.

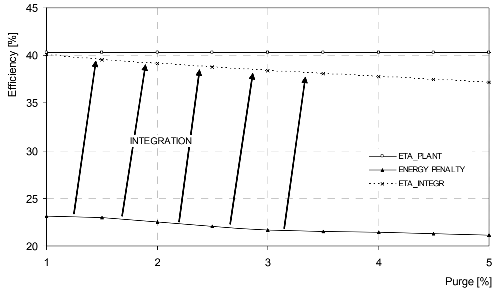

Figure 4. Efficiencies and energy penalties in the system. Limestone

In figure 4, the efficiency of the original power plant, the efficiency of the capture process without further utilization of waste heat flows and the efficiency of the fully integrated system and the original power plant are represented. These three efficiencies were calculated for purge values from 1 to 5% and molar ratio of 5 which corresponds to the minimum cost of avoided CO_2 tonne, figure 3. Energetic penalties between 16-18 points are observed when considering the coal consumption required for the capture process and lead to unacceptable values of cycle efficiencies. This strong detriment of energetic efficiency is relieved after integration with a supercritical cycle and the losses of efficiency referred to original power plant are confined to 0.10-3.13 points.

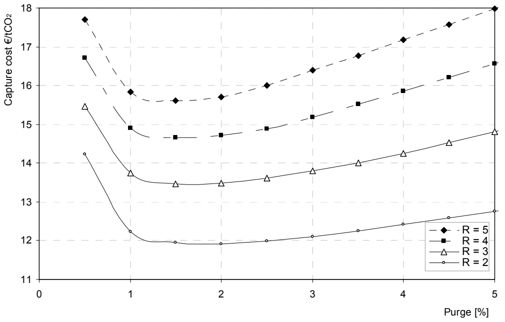

Figure 5. CO2 avoided cost for different R and purge. Hydrated limestone.

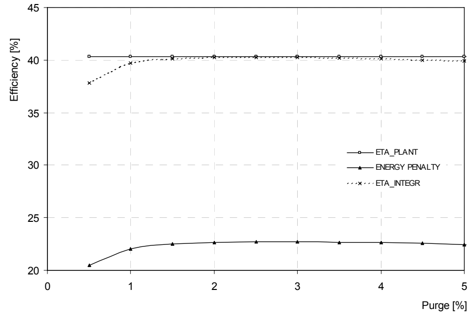

Figure 6. Efficiencies and energy penalties in the system. Hydrated limestone.

Considering hydrated limestone as CO_2 sorbent, the minimum cost of avoided CO_2 tonne, 11,92 €/CO_2 tonne, was found for the lowest molar ratio of fresh sorbent, 2, and purge values of 2%. There exist an inversion of cost curve trends when comparing raw limestone and hydrated limestone for different molar ratios, figure 5. In the case of raw limestone, the advantages of operating at low ratios (energetic standpoint) were counterbalanced with poor carbonation efficiencies meaning that slightly lower energy consumptions are required for significantly smaller amounts of avoided CO_2. However, hydrated limestone presents a better sorbent performance, X_N, and its theoretical maximum carbonation efficiencies at low ratios (about 100%) are comparable to those obtained for raw limestone at high ratios (68-100%). Due to this strong improvement in carbonation efficiency, solid particle flux circulating along the loop becomes the only key parameter to evaluate the influence of operation conditions on CO_2 cost. Since heat exchangers configuration has not been optimized for each sorbent but only for limestone, not all the waste heat from the capture process may be integrated in the supercritical cycle. Thus, obtained efficiencies of the integrated system may be improved by designing a new configuration of heat exchangers capable of using up all the heat released in the carbonation looping. Figure 6 shows an energy penalty around 17 points. After integration, the achieved efficiency is comparable to the original one for any purged value higher than 1%.

Data from literature [16,19] indicate that new improved sorbents should maintain an acceptable performance after hundreds to thousands sorption/desorption cycles in order to be able to compete with commercially available CO_2 capture technologies. To find the optimum cost of avoided CO_2 tonne when lithium orthosilicate is used as sorbent, a different range of purge percentages has been simulated (0.10-0.90%) for molar ratios between 2-5. The rationale behind the selection of these conditions is to compensate the high costs of synthetic sorbents with a low consumption. Purge flows much below 0.10% would be required for lithium orthosilicate to be competitive. Nevertheless, this cannot be carried out in practice since so low purge flows lead to a dramatic increase in recirculated ash flux. When drawing avoided CO_2 cost curves versus purge values, figure 6, a threshold purge value of 0.20% is brought to light. Below this purge percentage, the energetic integration model generates inconsistent results. Assessing heat fluxes in the integrated system, it was observed that internal solid circulation goes up as a consequence of coal ash accumulation in the loop when the capture process is operated at low purge values.

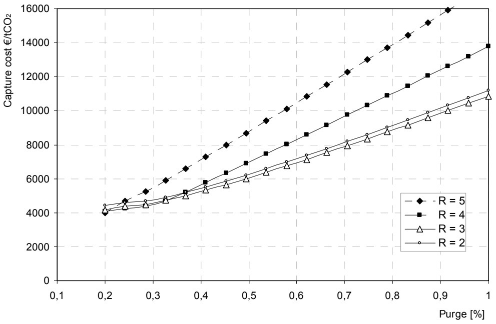

Figure 7. CO2 avoided cost for different R and purge. Lithium orthosilicate.

Two different coals whose ash content is presented in table 3, have been simulated with lithium orthosilicate as sorbent to assess the influence of coal composition on the behaviour of the system with extremely low purge flows. Figure 8 represents ashes internal circulation for different operating conditions. It shows that purge flows required to reach acceptable avoided CO_2 costs mean unworkable particle fluxes due to an increase in ash flux. Hence, low purge and make-up flows are not limited by sorbent behaviour when operating the system with coal but by coal ashes content and its accumulation in the loop.

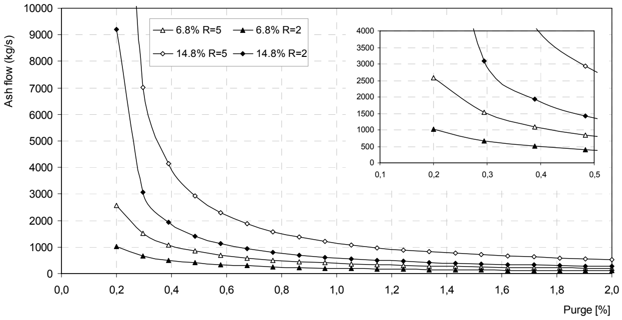

Figure 8. Ash flow for different coal compositions, varying R and purge. Lithium orthosilicate

Table 2. Simulated coal compositions.

C	Z	H_2O	O_2	H_2	N_2
66.2	13.65	8.1	6.76	3.75	1.54
76	6.8	8.5	3.8	3.9	1

5. Conclusions

Hydration enhances limestone activity allowing higher efficiencies for low sorbent/CO_2 ratios. The energetic penalty associated to hydration is compensated by the lower degradation of the sorbent. Therefore, avoided CO_2 cost may be reduced by means of limestone hydration. As the model corresponds to the optimal configuration for raw limestone, proper optimization of the energetic integration using $Ca(OH)_2$ may reduce capture cost below 11.92 €/tCO_2 which is by far a competitive result.

While a better performance of the system was expected under minimum purge conditions with lithium orthosilicate as sorbent, a minimum threshold limit for purge has been set depending on the ash content of the coal fed to the power plant and the calciner. The required purge percentages calculated to offset the extremely high prices of synthetic sorbents are technically forbidden. The mass fraction of the inert material in the solid internal circulation flow grows exponentially at low purge values and the amount of solids moving in the system cannot be handled. It may be concluded that the better sorbent performance, the better quality coal (less ash/inert content) must be used in the capture cycle. High activity sorbents will be preferred to operate in systems fed with inert-free fuels, i.e. natural gas, instead of applications operating with coal.

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