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Energy integration of high and low temperature solid sorbents for CO_2 capture

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Abstract

It is crucial to reduce the energy penalties related to CO_2 capture processes if CCS is to be implemented at industrial scale. In this context, gas-solid sorption has become a relevant technology. The absence of large amounts of water when using dry solid sorbents and their high heat capacity reduce the energy requirements in the gas-solid sorption CO_2 capture process. Depending on the sorbent composition, the gas-solid sorption process carries out at high or low temperatures. High temperature sorbents allow the utilization of waste energy while energy requirements in low temperature processes will be less demanding. This study is focused on the assessment and comparison of the final energy penalty of low-temperature (amine impregnated alumina-based solid particles) and high-temperature solid sorbents capture process (calcium oxide).

Keywords: Gas-solid sorption; solid sorbent; energy integration; waste energy; energy penalty; carbon capture

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1. Introduction

Amine chemical absorption is the most developed CO_2 capture technology due to its extensive use in the chemical and oil industries for the removal of CO_2 from gaseous streams. The significantly high energy requirements associated with the regeneration step implies an important energy penalty. As well, large sorbent make-up flow is needed to counterbalance amines degradation. The high costs derived from these drawbacks limit the implementation of this technology for CO_2 capture from power plants. It is crucial to reduce the energy penalties related to CO_2 capture processes if CCS is to be implemented at industrial scale. In this sense, gas-solid sorption is one of the most promising technologies. Main causes of energy penalty are: the sorbent regeneration energy (endothermic process), the energy losses due to evaporated water when aqueous solvent are used, and the CO_2 compression process. The absence of large amounts of water when using dry solid sorbents and the fact that its heat capacity is comparatively lower than that of aqueous amine solvents reduces the regeneration energy requirements. Depending on their composition, solid sorbents may react with CO_2 at low and high temperature. High temperature sorbents allow for the reduction of energy penalty by making use of waste energy. Low temperature sorbents present minor energy demand in the regeneration step. This study is focused on amine impregnated alumina-based solid particles as low temperature sorbent and its comparison with results from calcium oxide cycles as high temperature sorbent.

Nomenclature

ASU	Air separation Unit
CCS	Carbon capture and storage
GCC	Grand Composite Curve
HRSG	Heat recovery steam generator
HT	High-temperature
LT	Low-temperature
NGCC	Natural gas combined cycle
RR	Regeneration reactor
SR	Sorption reactor

1.1. High temperature sorbents

The carbon capture process which makes use of calcium oxide as solid sorbent is known as Ca-looping process. The regeneration step of the Ca-looping process leads to a strong energy consumption that might compromise the competitiveness of this technology. However, waste heat flows in the CO_2 capture cycle are available for integration which may contribute to the reduction of the global energy consumption of the process. Furthermore, the use of high temperature sorbents results in high quality heat flows that facilitate their subsequent utilization.

The heat integration in the Ca-looping process may follow two strategies: internal and external heat integration. The former uses a fraction of the available heat to reduce the energy requirement of the process itself. In the later approach, the heat flows are integrated in an external power cycle to generate electricity thus, increasing the power production of the plant and reducing the energy penalty associated to the CO_2 capture process.

Most recent proposals have tackled the internal heat integration in two different ways. On the one hand, the energy is recovered in the loop itself using a basic Ca-looping layout. On the other hand, the traditional configuration is modified in order to avoid the oxy-fuel combustion. Regarding the former works, Martínez *et al.* [1] analyzed the options to recover energy from the gaseous or solid streams leaving the calciner to heat up the carbonated sorbent entering this reactor. The idea was to reduce the energy required to increase the temperature of the particles that leave the carbonator at 650 ºC and are directed to the calciner that operates at temperatures as high as 950 ºC. Different heat exchange devices where analyzed including an additional fluidized bed, working as a heat recovery system, and a mixing seal valve in which solid streams from both reactors are mixed facilitating the heat transfer between them. Martínez *et al.* [2] later proposed a further modification of the process consisting in the inclusion of a cyclonic preheater in which the gas leaving the calciner transfers heat to the carbonated particles before entering this reactor.

Results showed that a two-step cyclonic preheater leaded to a 13.3% reduction of the mass ratio of coal consumed to CO_2 captured as compared to the ordinary configuration (0.39 kg_{coal}/kg_{CO2} vs 0.45 kg_{coal}/kg_{CO2}) and an 11% reduction of the oxygen needs. The inclusion of a mixing seal valve in the Ca-looping system was thoroughly analyzed by Martínez *et al.* [3]. In this configuration, the two seal valves which direct the particles from one reactor to the other are substituted by a single mixing seal valve that collects the solids from both reactors and distributes them to carbonator and calciner again. A 15.5% reduction of the consumed coal to captured CO_2 and a 14.5% decrease of oxygen consumption were achieved in this case.

Among the Ca-looping configurations that aims to avoid the oxy-fuel combustion step, the process called "HotPSA" operates with pressure swing absorption [4,5]. The objective is to reduce the CO_2 partial pressure in the calciner by means of steam injection or depressurization which diminishes the regeneration temperature requirements. Thus, the system may reuse part of the reaction heat from carbonation for subsequent calcination in the same reactor. Ball *et al.* [6–8] developed the Endex configuration for the direct heat transfer between combustor and calciner or between calciner and carbonator. The endothermic regeneration is assumed to occur at a lower temperature than the carbonation, which can be achieved by reducing the calciner pressure as in the "HotPSA" process. The difference lies in the system configuration which permits continuous operation mode in each reactor. In this case, carbonator is housed inside the calciner, to permit heat conduction through the reactors wall, and the entire unit is insulated. Kim *et al.* [9] proposed a system combining multiple stages of capture cycles with different sorbents operating at diverse temperature levels. Each stage consists of an absorber and a regenerator. Thus, the exothermic absorption of a high temperature stage is used to provide energy to the endothermic regeneration of a lower temperature stage.

External integration is essential to reduce the energy penalty of the CO_2 capture process even when a fraction of the available heat is internally integrated. Romeo *et al.* [10] proposed the energy integration in a new design supercritical steam cycle to repower the existing power plant, obtaining a 7.89 energy penalty. The inclusion of the CO_2 compression train in heat integration leads to lower energy penalties. Integration proposed by Hawthorne et al. [11] reduced the energy penalty down to 6.4 percentage points, while Lisbona *et al.* [12,13] achieved 5.78 energy penalty, and Vorrias *et al.* [14] 4.96. However, the values of energy penalty obtained by Martínez *et al.* were somehow larger (8.3-10.3) [15].

Some works have also considered the possibility of integration in the reference existing power plant whose flue gas flow is fed into the capture plant. Romeo *et al.* [16] analyzed this integration through an exergetic point of view. Yang *et al.* [17] studied various possibilities of integration including the retrofitting of the existing power plant and the development of a new design power plant concluding that the best efficiency option is to use the available heat in a HRSG to drive new turbines.

As shown, many studies are focused on external heat integration to analyze and evaluate the Ca-looping process. However, the optimization of this integration through a well stablished methodology becomes essential. To this aim, Lara *et al.* [18] combined the pinch methodology with economic and exergetic analysis to optimize the design of the heat exchangers network that constitutes the heat integration system. This work served as a basis for the development of an algorithm with the objective of providing a systematic procedure for heat integration to find an energy and cost-efficient solution for the heat exchanger network in this type of systems [19]. This procedure was tested in two different cases in which the available heat was used to drive a supercritical steam cycle obtaining an energy penalty as low as 5.17 percentage points.

1.2. Low temperature sorbents

Amine impregnated alumina-based solid sorbent consists of a porous solid support in which surface the amine is allocated. These chemisorbents present great potential to reduce the energy penalty and to increase the capture capacity and CO_2 selectivity compared to amine processes. In this cyclic process, sorption occurs at 40-45 ºC and 1.3 bar and sorbent regeneration takes place at 80-85 ºC and 0.1 bar. Energy integration analysis shows a significant potential of this sorbent to achieve energy penalties similar or even lower to those obtained with the calcium oxide.

The use of amine-impregnated as solid sorbent is a promising option for CO_2 separation from flue gases with a sorption/desorption cycle. It works at low temperatures in sorption, 75-90ºC, and desorption processes, 100-140ºC. Other advantages include the reduction of energy demand for regeneration [20] due to less water evaporation [21] and lower specific heat capacity of the sorbent [22]. For this reason, the use of these materials is a suitable option to reduce energy penalty in CO_2 capture systems [23,24] and be energetically compared with high temperature solid sorbent.

Previous studies pointed out an increase of 3.7 net efficiency points for a PC plant and 1.5 net efficiency points for NGCC plant when applying amine solid sorbents instead of aqueous amines have been previously reported [20]. Other works limited this the increment to 1.7 efficiency points from a penalty of 8 efficiency points for MEA to 6.3 of polyethyleneimine (PEI)-based solid sorbent [24]. As in those processes which make use of high temperature solid sorbents, the optimum design to circulate the sorbent between sorption and desorption stages includes fluidized bed reactors [24].

In this case, low-grade heat may be recuperated from the sorption process and CO_2 compression stage. Although temperature level is low, it is still worthy to develop a procedure for heat integration to find the optimum efficiency for this kind of solid sorbents.

1.3. Objective

The objective of this study is the assessment of the energy integration of low temperature solid sorbents for CO_2 capture to determine the common characteristics and the main differences between high and low temperature technologies and their effect on CCS energy penalty. For this aim, the energy integration of the low temperature capture systems is carried out and optimized, and the results are compared to those obtained for the calcium looping process.

In both cases, CO_2 is captured by means of a regenerable solid sorbent. The capture and regeneration process is carried out in two reactors: a sorption one where CO_2 capture takes place, and a regeneration one, where the CO_2 is released. The regenerated sorbent is then redirected to the first reactor to start a new cycle. A global system has been defined for each case, consisting in a reference power plant; the corresponding capture system and a compression train. For the high-temperature case, a secondary supercritical steam cycle has also been defined. The main difference between them is the process temperature, which conditions the energy integration targets.

In the calcium looping, the regeneration energy is provided by oxycombustion and available heat may be used to produce as much additional power as possible. In the amine-impregnated alumina sorbent capture system, the regeneration heat is provided by the reference power plant turbine bleeds; so the integration is focused on using as less steam as possible. While in high-temperature capture the objective is to take advantage of as much heat as possible for the secondary steam plant, thus, reducing the energy penalty by using the heat to provide additional power; in low-temperature capture the objective is to integrate the capture cycle in the reference plant in the most efficient way in order to reduce the heat requirements of the capture cycle, thus limiting the amount of turbine bleeds.

A diminution of the energy needs associated with the CO_2 capture systems is essential to achieve the goals necessary to make these processes feasible at commercial scale. Adequate heat integration is critical to minimize the energy consumption in the capture system and, thus, the energy penalty related to the CCS.

2. High temperature solid sorbents. Ca-looping cycle.

A 500MW power plant fed with high-rank coal supplies the Ca-looping CO_2 capture system with flue gas. The flue gas from the reference power plant at 180 °C enters the carbonator where CO_2 capture takes place at 650 °C. The clean gas is emitted through the stack. The solids are directed to the calciner, where the sorbent is regenerated, and a concentrated stream of CO_2 leaves the reactor to the compression train. The energy required to heat up the solids to 950 °C (temperature at which calcination takes place), is provided by oxy-combustion, so an air separation unit (ASU) is also part of the system. A CaO/CO_2 molar ratio of 5 in the carbonator and a make-up flow of 2.5% over the total solid stream leaving the calciner are considered, leading to a capture efficiency in the carbonator around 70%. In this case, both reactors are interconnected circulating fluidized beds. A compression train is the last system in the process, conditioning the CO_2 for transport, up to 120 bar and 80 °C.

The main energy streams of the Ca-looping process to integrate in a new power cycle, Table 1, Figure 1, are the heat released in the carbonation reaction (S1); the gas flow leaving the carbonator to the stack (S2); the CO_2 generated in the calciner (S5, S7, S11), the intercoolers at the compression train (S12-S15) and the solids streams (S3, S4). The recirculated stream of CO_2 and the oxygen entering the calciner (S6) must be heated up.

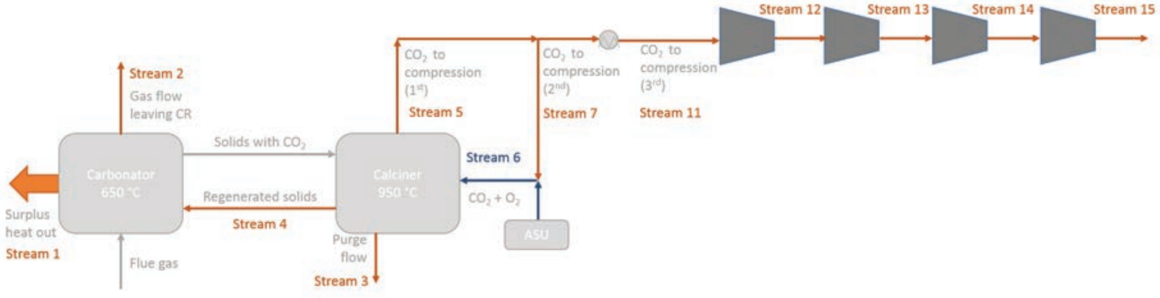


Figure 1. High temperature capture process scheme

Table 1. Energy streams Ca-looping cycle. HT capture process.

		Stream	Tin[°C]	Tout[°C]	Q [MW_{th}]	Type
Capture cycle	1	Surplus heat carbonator	650.00	650.00	157.10	HOT
	2	Gas flow leaving carbonator	650.00	190.00	230.60	HOT
	3	Purge flow	950.00	200.00	26.40	HOT
	4	Solids from RR to SR	950.00	650.00	365.00	HOT
	5	Captured CO_2 to compression	950.00	329.70	307.80	HOT
	6	CO_2+O_2 to calciner	180.00	296.40	26.40	COLD
	7	CO_2 to compression (2nd step)	329.70	190.00	44.30	HOT
	11	CO_2 to compression (3rd step)	190.00	55.22	37.38	HOT
Compression train	12	1st intercooler	171.50	50.00	30.02	HOT
	13	2nd intercooler	163.20	50.00	27.86	HOT
	14	3rd intercooler	164.60	50.00	28.22	HOT
	15	4th intercooler	173.60	80.00	23.48	HOT

3. Low temperature solid sorbents. Amine-impregnated alumina solid sorbent.

For the low-temperature capture cycle, it is assumed that the flue gas has been through FGD processes and enters the system, without SO_x, at 180 °C. A condensation to avoid steam in the flue gases has been implemented prior entering the capture plant. After the condensation, the flue gas flow needs to increase their temperature and pressure before entering the sorption reactor. The temperature increase has been taken into account in the integration process and a fan has been added just before the sorption reactor to increase the pressure of the gas to the reactor conditions. In this case, the sorption reactor, which operates at 42.50 °C and 1.3 bar, has been considered to be a circulating fluidized bed. A 78% of sorption capacity has been assumed; and the global capture efficiency is around 80%. Due to the regeneration times for this sorbent, the regeneration reactor has been assumed as a fixed bed with complete regeneration.

The flue gas enters the sorption reactor where CO_2 capture takes place at 42.50 °C. The clean gas is emitted through the stack. The solids are directed to the regeneration reactor, and a concentrated stream of CO_2 leaves the reactor to the compression train. The energy required to heat up the solids to 85.00 °C (temperature at which regeneration takes place), is provided by steam of the reference power plant turbine bleeds.

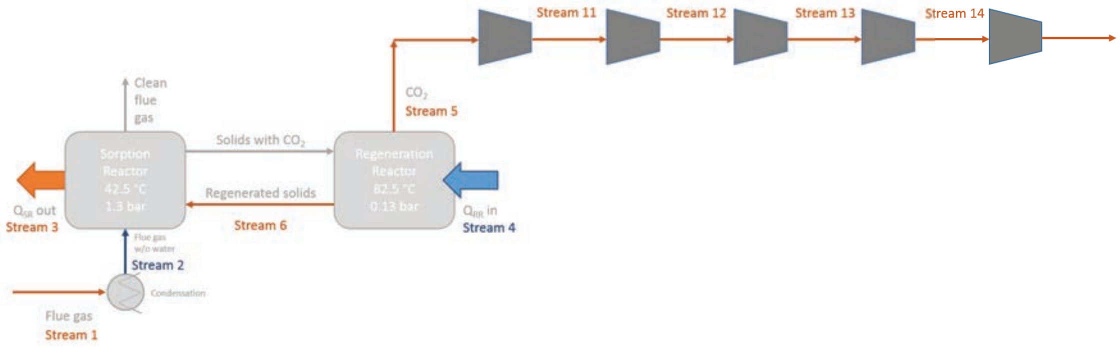


Figure 2. Low temperature capture process scheme

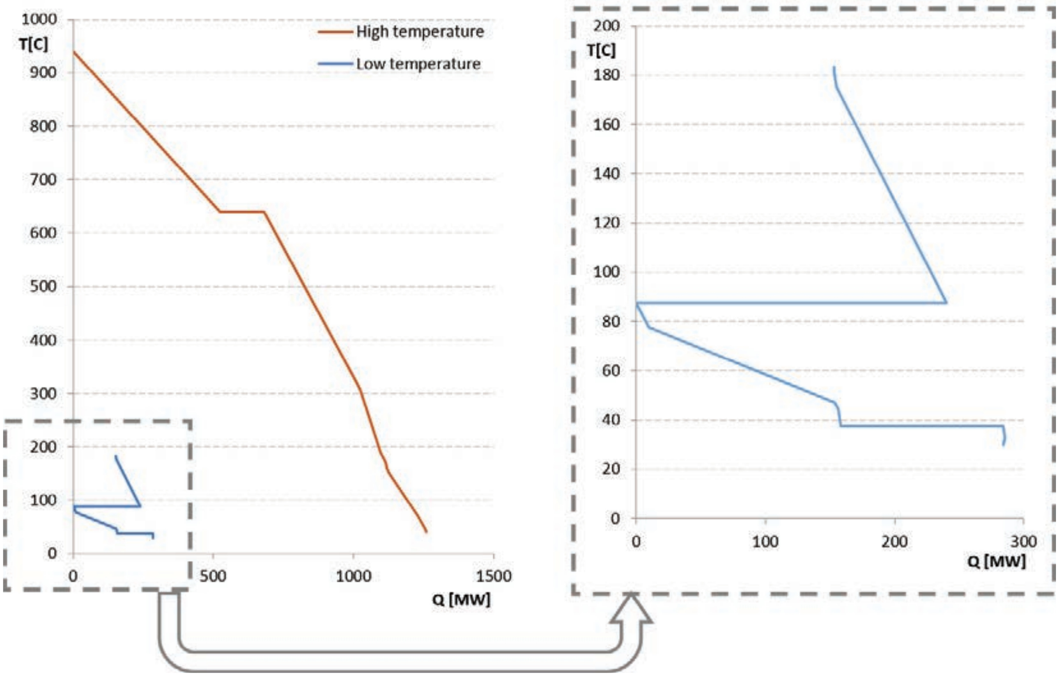
The energy streams of the capture plant are listed in Table 2, Figure 2. These streams are divided between heat sources (type hot) and heat sinks (type cold). As presented, the more demanding energy stream is the heat that must be provided to the regeneration reactor (RR), which works at 82.5°C and 0.13 bar. The intercoolers of the compression train provide an amount of energy in a temperature range adequate for the integration with the power plant and the CO_2 capture cycle.

Table 2. Energy streams amine-impregnated sorbents cycle. LT capture process.

		Stream	Tin[°C]	Tout[°C]	Q [MW_{th}]	Type
Capture cycle	1	Flue gas before condensation	180.00	37.70	100.65	HOT
	2	Flue gas after condensation	25.00	39.41	6.26	COLD
	3	Sorption heat	42.60	42.50	126.00	HOT
	4	Regeneration heat	82.50	82.60	240.60	COLD
	5	Captured CO_2	82.50	50.00	2.11	HOT
	6	Solids from RR to SR	82.50	52.00	112.03	HOT
Compression train	11	1st intercooler	186..30	50.00	9.32	HOT
	12	2nd intercooler	186.40	50.00	9.33	HOT
	13	3rd intercooler	186.70	50.00	9.35	HOT
	14	4th intercooler	188.20	50.00	9.46	HOT

4. Process integration.

Once the streams are defined, the pinch analysis is applied to both systems. The Grand Composite Curve of both cases, according to data provided in Table 1 and Table 2 is shown in Figure 3. Figure 3 a) shows the streams corresponding to the capture cycle and compression train for both, HT and LT cases, while Figure 3 b) enlarges the GCC of the low temperature case. The Grand Composite Curves represent the amount of available heat in each configuration and their corresponding temperature levels. The curves show the pinch point correspondent to each case. For the low temperature case, the pinch point is located at 87.5 °C; while in the high temperature one; the point is located at 940 °C. This last one is a threshold problem, which only has cooling requirements.



a) HT and LT cases b) LT case

Figure 3. Grand Composite Curves. HT and LT cases including compresion train.

The use of this available heat differs between both cases. In the high-temperature capture process, the available heat is recovered by a heat exchanger network to drive a secondary supercritical steam cycle. This is possible due to the high quantity and quality of the heat. In the low temperature case, the integration is oriented to minimize the requirements of the steam from the reference power plant turbine bleeds, thus decreasing the energy penalty of the capture cycle.

For the high-temperature case, the supercritical steam cycle is characterized by the streams shown in Table 3. On the contrary, the low temperature case requires the use of the turbine bleeds involved. These streams are shown in Table 4.

Table 3. Supercritical steam cycle streams. HT capture process.

	Stream	Tin[°C]	Tout[°C]	Q [MW_{th}]	Type
20	HRSG	402.40	618.50	400.80	COLD
21	Reheating steam	328.10	620.00	224.41	COLD
22	Preheating steam	168.80	402.40	464.42	COLD
23	Condensate	30.00	153.20	161.90	COLD

Table 4. Reference power plant streams. LT capture process.

	Stream	Tin[°C]	Tout[°C]	Q [MW_{th}]	Type
7	Condensate	31.10	151.40	136.54	COLD
8	IP6 turbine bleed	263.70	145.10	57.29	HOT
9	IP7 turbine bleed	146.10	106.90	28.33	HOT
10	LP8 turbine bleed	81.85	81.80	50.93	HOT

For the low-temperature case, two main targets were expected from heat integration; to provide the energy required for sorbent regeneration while maintaining the amount and objective temperature of Stream 7 (*Condensate*). The approach adopted in this study forces Streams 4 (*Regeneration heat*) and 7 to reach their objective temperatures. They represent the two main heat sinks in the low-temperature system. The heat integration focuses on the management of turbine bleeds to supply both, the low pressure water required to heat up the condensate, Stream 7, and the regeneration energy, Stream 4.

The objectives of the heat integration differ between the high and the low temperature sorbent systems. In the case of high temperature sorbents, the target of the integration is to take advantage of all the available heat streams. In the case of low-temperature sorbents, the aim of the integration is to cover the main heat sinks in the system to reach their objectives.

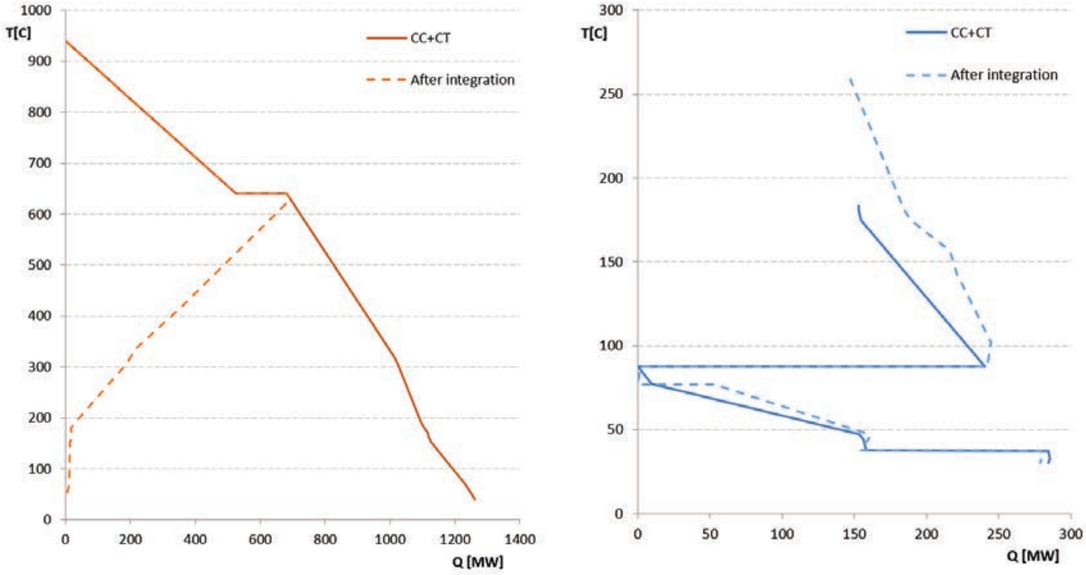
The integration steps are the following:

- To quantify the heat streams involved in each case, classifying them into heat sources and heat sinks.
- To calculate the amount of heat that can be internally exchanged according to pinch analysis methodology.
- To calculate the hot and cold energy requirements that must be externally provided.
- To define the corresponding heat exchanger network, by applying a systematic procedure [19] that achieves the objectives.
- To define the amount of energy which has to be exchanged.
    - To define the amount of energy from the turbine bleeds to satisfy the hot energy requirements of the heat sinks (for low-temperature case)
    - To define the amount of cold energy requirements from the Steam cycle streams (heat sinks), to be fulfilled by the available heat sources (for high-temperature case).
- To implement the heat exchanger network in the model to calculate the new parameters of the plant.
- To define the new streams and analyze the optimization possibilities.
- To restart the process from the beginning according to the new results until the optimization is finished.

In both cases, the systematic procedure defined in [19] has been applied to define the best suited heat exchanger network. While the methodology was designed to fulfil the heating needs of every cold stream by exhausting the available energy from the hot ones; this approach was not adequate for the low-temperature case, in which the objective was different. So, the methodology was adapted to be suitable to solve the low-temperature sorbent integration problem. With this modifications, the procedure has been extended to both possible situations in pinch analysis, above and below pinch point, which must be addressed separately.

The procedure proposes a specific order to match hot and cold streams according to pinch methodology, to allow rapid design of an energy and cost-efficient heat exchanger network.

Figure 2 shows the GCC of the final heat exchanger networks obtained for a) High-temperature and b) Low-temperature case. The high-temperature HEN consists of 15 heat exchangers and drives a 364 MW supercritical steam cycle, leaving only 307 kW of cooling requirements. The low-temperature HEN consists of 10 heat exchangers. In this way, two turbine bleeds can be avoided in comparison to the non-integrated low-temperature system.



a) HT case

b) LT case

Figure 4. Initial and final Grand Composite Curves.

The energy penalty of the capture process has been calculated once both integrations are performed. While energy penalty of the high-temperature case is around 5.7 percentage points, the energy penalty of the low-temperature one is around 6 percentage points. Thus leads to the suitability of using each one of the configurations based on the modifications that the power plant requires, Table 5.

Table 5. Main figures obtained with integration analysis

	Power produced in the reference plant [MWe]	Additional power obtained through integration [MWe]	Power penalty avoided through integration [MWe]	Energy penalty of capture process [%]
HT	500	364	--	5.7
LT	500	--	32	6.0

5. Conclusions.

This study assesses and compares the energy integration potential of two capture systems based on gas-solid sorption. These two systems make use of two different sorbents: (i) amine impregnated alumina-based solid particles, which reacts with CO_2 at low-temperatures around 45ºC and (ii) calcium oxide which reacts with CO_2 at temperatures around 650ºC. The regeneration processes takes place at higher temperatures: (i) around 85ºC for amine impregnated particles and (ii) above 900ºC for calcium carbonate regeneration. This difference in temperature levels will lead to different energy integration problems which will require specific approaches.

The heat recovered in the HT case through the implementation of 15 heat exchangers drives an external supercritical steam cycle of 364 MW. Cooling requirements are reduced to only 307 kW.

The heat exchange network built after energy integration in the LT case includes 10 heat exchangers which allow to remove two of the required turbine bleeds in the initial case. The energy content associated to these two bleeds is near 32 MW.

The energy penalty imposed in the power plant by the capture processes after heat integration is 5.7 percentage points when high-temperature sorbent is used and around 6 percentage points when a low-temperature sorbent is applied. These values are comparable pointing out the importance and the potential of both type of sorbents. However, high temperature capture process leads to a slightly better global energy efficiency performance.

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