Heat integration of alternative Ca-looping configurations for CO_2 capture

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Abstract

The best option to overcome the energy penalty caused by the CO_2 separation in Ca-looping cycle is to take advantage of the surplus heat from carbonator, calciner and concentrated CO_2 stream. External integration is the traditional choice to use this energy to produce additional power and increase net efficiency when Ca-looping is coupled with power plants. This integration has been extensively studied when applied to the ordinary configuration of Ca-looping cycle. As calciner represents the main energy consumption within this capture cycle, another possibility to minimize energy penalty is to internally use the surplus heat from the solid and gaseous streams leaving the calciner to preheat the solids entering this reactor. The general objective of internal integration is to reduce the energy demand per captured tonne of CO_2. It represents a reduction of the coal and oxygen needs and also a total decrease in the CO_2 generation with respect to the ordinary configu-

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ration. However, it has been demostrated that the amount of available heat for extra power generation by external integration, which is essential for the viability of this technology, is also reduced. This is the case of the configurations which include a cyclonic preheater or a mixing seal valve. This study aims to assess, through computational simulation, the energy penalty minimization that may be reached by external heat integration when internal energy integration has been previously applied in these configurations. A methodological integration process has been applied to obtain a reduction of the energy penalty with respect to the ordinary configuration. This energy saving combined with the lower size of equipment and reduced capital cost would make the cyclonic preheater configuration the most suitable one to improve the viability of this technology.

Keywords:
Ca-looping, cyclonic preheater, mixing seal valve, heat integration, CO_2 capture

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1. Introduction

One of the main obstacles to the development of CCS technologies for global carbon emissions reduction is the large amount of energy required in the capture processes [1]. Several researches have tackled this issue using different techniques as thermal integration [2], [3] or multi-objective optimization [4]. They have been mainly focused on amine scrubbing technology, although different solutions have been proposed to overcome this problem as the integration with renewable energy [5] or the use of supercritical CO_2 as working fluid [6].

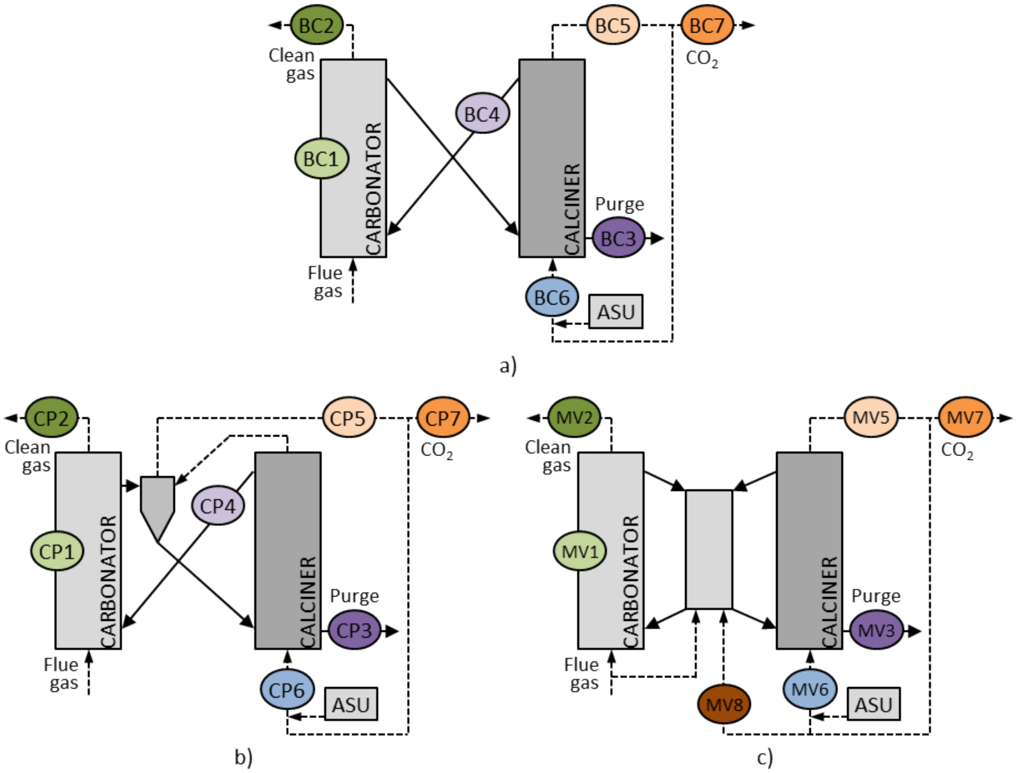
Nevertheless, other CCS options are able to further reduce the energy penalty and costs due to their inherent advantages for thermal integration and the use of less expensive CO_2 sorbents. Among these technologies Ca-looping process is highlighted. The large energy demand in the Ca-looping process is one of the main key issues of this technology [7], [8]. Most significant energy penalties in the Ca-looping cycle arise from the heat requirements in the calciner itself, the oxygen separation process and the compression of captured CO_2. When the energy required in the calciner is provided by oxyfuel combustion, the oxygen needs and the global amount of generated CO_2 are intimately related to the energy consumption in the regeneration reactor. A reduction of the coal consumption in the calciner means a reduction of the ASU requirements. The ASU power consumption and the energy requirement for CO_2 conditioning imply a similar reduction of the overall efficiency in the power plant ranging 3-4 percentage points each one [9]. A decrease of the energy consumption in the calciner implies a lower demand of fuel in this reactor and, thus, a reduction of the additional CO_2 generated in the system that has to be compressed.

-A significant amount of the energy consumed in the calciner is used to heat up the solids recirculated at a lower temperature from the carbonator. The remainder corresponds to endothermic reaction of sorbent regeneration, which is an unavoidable energy intake if CO_2 capture efficiency and make-up flow are to be kept constant. The temperature difference between entering solids from the carbonator and those in the calciner may be as high as 300 ° C.

Solids preheating prior to calciner inlet diminishes the temperature difference thus reducing the calciner energy requirements. Martínez et al. [10] proposed various configurations of the Ca-looping process that internally integrate a fraction of the available heat for this purpose. Among them, the inclusion in the ordinary configuration of a cyclonic preheater and a mixing seal valve appear as the most promising ones, reducing coal, oxygen specific consumption and CO_2 generation [11], [12]. However, as a result of these efforts it is also observed a decrease of the waste heat available for external energy recovery.

-Ca-looping shows an important potential for external heat integration since high-quality waste heat flows may be used to drive a steam cycle, reducing the energy penalty imposed to the power plant [13], [14], [15], [16], [17], [18]. Energy penalties as low as 5.17 percentage points may be achieved when applying a methodological procedure to define the external integration to the Ca-looping basic configuration which consists of two CFB reactors interconnected by independent loop seal valves which allow the exchange of solids from one to other fluidized bed and will be further described in the next section [9].

In this work, the external heat integration methodology developed by Lara et al. [9] to properly define external heat integration was applied to two alternative configurations which include (a) a cyclonic preheater and (b) a mixing seal valve. These systems were modelled and simulated to carry out an energy assessment of the whole system. The objective was to determine to which extent the reduction of the available heat in these novel configurations affect the energy penalty of the complete system when compared to the ordinary configuration.



a)

b)

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Figure 1: Configurations schemes

2. Ca-looping configurations and modelling

Three different Ca-looping systems were modelled: an ordinary configuration (BC), figure 1a, with no internal heat integration for comparison purposes, the cyclonic preheater configuration (CP), figure 1b, and the mixing seal valve configuration (MV), figure 1c.

All of them are assumed to be fed with the flue gas from a 500 MW_e coal power plant with a 40% energy efficiency. This plant burns the coal defined in table 1 with a 20% of oxygen excess. The flue gas is fed to the Ca-looping system at 180 °C.

Table 1: Coal composition and heating value

C	H	N	S	ash	H_2O	LHV
$72.04\%_{db}$	$4.08\%_{db}$	$1.67\%_{db}$	$0.65\%_{db}$	$7.36\%_{db}$	8.1%	25,372 kJ/kg

The three configurations are operated to obtain maximum CO_2 capture only limited by carbonation equilibrium at operating conditions which imply a 93.01% efficiency capture. Carbonation model was developed by Alonso et al. [19] and Charitos et al. [20] and it is summarized in table 2.

Table 2: Carbonation model

Carbonation efficiency	$\eta_{CR} = k_{CR}\,\varphi\, f_{a,CR}\,\tau_{CR}\,X_{ave}\,(v_{\bar{C}O_2} - v_{eq})$
Fraction of active sorbent	$f_{a,CR} = 1 - exp(\frac{-t^*_{CR}}{n_{Ca}/\dot{n}_{Ca}})$
Fast stage reaction time	$t^*_{CR} = \dfrac{X_{ave} - X_{in}}{k_{CR}\,\varphi\,X_{ave}\,(v_{\bar{C}O_2} - v_{eq})}$
Carbonator space time	$\tau_{CR} = \dfrac{n_{Ca}}{\dot{n}_{CO_2}}$

Carbonator is assumed to operate at 650 °C. The model for the average capture capacity, X_{ave}, is given by a different expression for each configuration. As a general case, it may be calculated by means of equation 1.

$$X_{ave} = \sum_{N=1}^{\infty} r_N\, X_N \tag{1}$$

where X_N defines the degradation of the sorbent as it accomplishes complete carbonation/calcination cycles. A curve to model this deactivation of the sorbent, equation 2, was proposed by Zhen-shan et al. [21], and the parameters ($a_1 = 0.1045$, $a_2 = 0.7786$, $b = 0.07709$, $f_1 = 0.9822$ and $f_2 = 0.7905$)

were later fitted by Rodríguez et al. [22].

$$X_N = a_1\, f_1^{N+1} + a_2\, f_2^{N+1} + b \tag{2}$$

r_N, in equation 1, is the age distribution of particles population, which means that r_i is the fraction of particles whose capture capacity is X_i. The sorbent degradation rate depends on the configuration since partial carbonation and calcination reactions may take place out of the principal reactors, in the heat exchangers. The definition of the models used to evaluate the age distribution of the particles, r_N, and the average capture capacity, X_{ave}, in each configuration are out of the scope of this paper and they may be found elsewhere [11] [12].

Even when the calciner is assumed to operate at 950 °C, which represents a sufficiently high temperature to achieve instantaneous and complete calcination, the high CO_2 partial pressures makes necesary to use an advanced calcination model. This model was developed by Martínez et al. [23] and is sumarized in table 3.

Table 3: Calcination model

Calcination efficiency	$\eta_{CL} = \dfrac{f_{a,CL}}{ln(1/(1-f_{a,CL}))}$
Fraction of active sorbent	$f_{a,CL} = 1 - exp(\frac{-t^*_{CL}}{n_{Ca}/\dot{n}_{Ca}})$
Full calcination time	$t^*_{CL} = \dfrac{-3\,X_{in}}{k_{CL}(C_{eq} - C_{CO_2})}$

The energy required for the sorbent regeneration is obtained from the oxy-fuel combustion of the coal defined in table 1, to avoid CO_2 dilution in the calciner. A fraction of the gas generated in this reactor is recirculated to reduce the inlet oxygen concentration to 60 %v. and increment the flow of fluidization agent

The cyclonic preheater configuration makes use of the gaseous stream leaving the calciner to preheat the solids entering this reactor. This device provides an excellent heat transfer between gas and solids, due to the high swirl and turbulent motion of the flow inside, and it implies low investment costs. Martínez et al. [11] determined the two-stage preheater as the most adequate one for Ca-looping application. Particles may leave the carbonator only partially carbonated and therefore, carbonation may take place in the cyclonic preheater since, in this device, the sorbent is put into contact with a highly concentrated CO_2 stream. As well, as particles temperature increases in the cyclones, also calcination may take place. The extent of carbonation and/or calcination reactions in the cyclones, and their effect on temperature and composition of the gaseous and solid streams leaving the cyclonic preheater, are included in the model.

The mixing seal valve configuration makes use of the sensible heat of calcined particles to heat up the solids from the carbonator. In this system, particles from both reactors are collected in a single seal valve that also feeds both reactors. Solids at different temperatures can directly exchange heat since they are put into contact and mixed in this device. Therefore, heat is transferred through conduction, convection and radiation inside this seal valve. Then, the new mixture of solids is directed to both reactors through two different recirculation pipes. The mixing of carbonated and regenerated sorbent particles reduces the fraction of active calcium oxide entering the carbonator. Complete mixing of solid particles, which is the most unfavourable case, is assumed in the model. Thus, this configuration requires high purge fractions or higher CaO to CO_2 ratios to achieve the same CO_2 capture efficiency. Martínez et al. [12] determined that the most suitable way to operate this system is to use two gaseous streams, flue gas and concentrated CO_2, to aerate the mixing seal valve and to distribute the solids leaving this device by directing 15% of them to the carbonator and the remaining amount to the calciner. As in the cyclonic preheater configuration, carbonation or calcination may take place in the mixing seal valve, thus afecting the temperatures and composition of the solid and gaseous streams leaving this device. This fact is taken into account in the model.

Table 4 summarizes the main figures. Cyclonic preheater configuration shows 13% of coal and oxygen savings compared to the base case which means a 6.5% reduction of the CO_2 generation. Regarding the mixing seal valve, coal and oxygen savings reach 15% and CO_2 generation diminishes 7.4% whereas the flow of solids between reactors undergoes a significant increase associated with the lower active fraction of calcium oxide in the carbonator due to the mixing of carbonated and regenerated particles.

The Grand Composite Curves shown in figure 2 represent the amount of available heat in each configuration and their corresponding temperature levels. The curves show the pinch point, where there is no heat available, at 950 °C, which classifies these configurations as threshold problems, with only cooling requirements. The reduction of available heat for running the supercritical power plant from the base case to the configurations including internal integration can be identified in figure 2. The most significant loss of available heat is associated with those gaseous and solid streams at 950 °C

Table 4: Comparison of energy savings and main operating variables in the Ca-looping system

Configuration	Solids flow [kg/s]	Specific coal consumption [kg_{coal}/kg_{CO_2}]	Specific O_2 consumption [kg_{O_2}/kg_{CO_2}]	Reduction of CO_2 generation
Base case	2,221	0.45	1.03	-
Cyclonic preheater	2,147	0.39	0.89	6.5%
Mixing seal valve	4,802	0.38	0.88	7.4%

whose energy content is invested in preheating the solid stream at 650 °C. However, temperature levels still seem to be suitable to generate supercritical steam at around 600-620 °C. It should be noticed that the heat from the carbonator, at a constant temperature (650 °C), may be used for this aim. In the case of the mixing seal valve, the surplus heat from the carbonator partially compensates for the loss of available heat at the highest temperatures. Cyclonic preheater configuration presents 14% reduction of available energy while this reduction amounts 17% in the mixing seal valve case.

3. HEN design

The available heat from the Ca-looping is recovered to drive a steam cycle. The heat integration was carried out applying a systematic procedure defined in [9] through which an optimized HEN for each configuration is established. This methodology was designed to cover the heating needs of every cold stream just by exhausting the available energy from the hot streams.

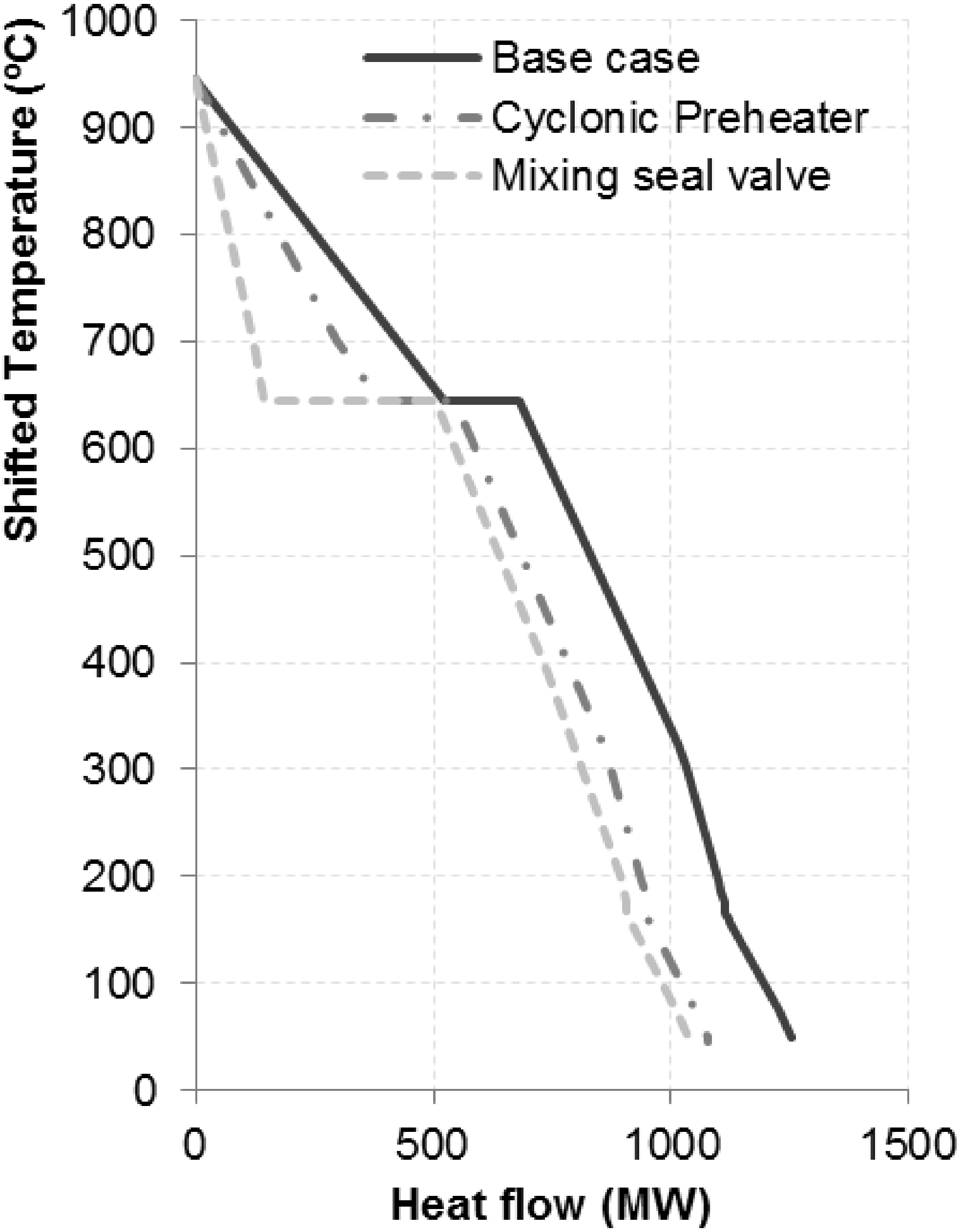

Figure 2: Grand Composite Curves

The procedure proposes an specific order to determine the matches between streams according to the pinch method, which is a widely applied technique used to define the hot and cold streams and obtain their minimum energy requirements. This integration technique allows rapid design of an energy and cost-efficient heat exchanger network for this kind of problems, despite the high amount of streams involved.

The whole integrated systems comprises the Ca-looping, a supercritical steam cycle and the compression train. Figure 3 shows the steam cycle and the compression train. The steam cycle contains a heat recovery steam generator that produces steam at 618.5 °C and 29 MPa. The high pressure turbine is followed by a reheating process. Two bleeds from the intermediate pressure turbine are used to drive the CO_2 compressors and the pump. The compression train consists of 4 compression steps alternated with cooling processes. CO_2 leaves the system at 80 °C and 12.1 MPa.

Tables 5, 6 and 7 show the heat streams in each configuration organized by temperatures. The main difference between the cyclonic preheater and the base case appears in the CO_2 stream to compression (BC5 and CP5). The inlet temperature and the amount of heat are reduced in the cyclonic preheater configuration since this concentrated stream of CO_2 is previously used to heat up the solids from the carbonator. With regard to the mixing seal valve configuration two main differences may be observed. There is no heat stream from the solids recirculation to carbonator equivalent to BC4. As well, the amount of surplus heat from this reactor is significantly higher in the mixing seal valve case (MV1) than in the base case (BC1) since solids enter the carbonator at a higher temperature.

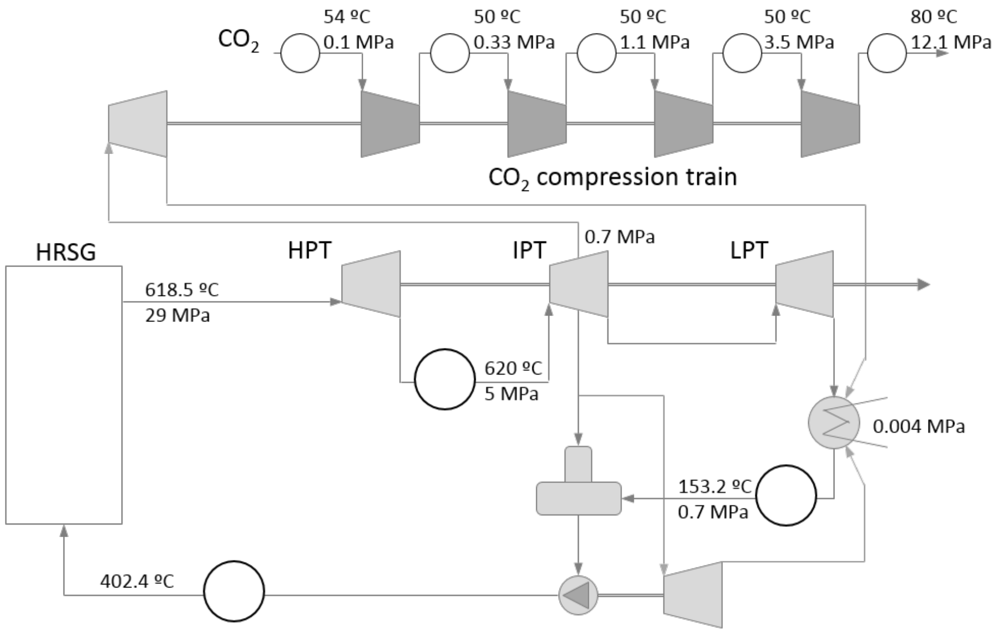

Figure 3: Steam cycle and compression train diagram

Figure 4 shows the steam cycle and the compression train heat integration in every configuration. Streams number 3 and 6 are not integrated in the external steam cycle but they internally exchange heat between them in all configurations. The base case integrated system, figure 4a, consists of 13 heat exchangers which allows to accomplish CO_2 compression and to drive a 364 MW steam cycle. It should be noticed that streams BC2, BC4, BC5 and BC7 have to be divided in 2 stretches at different temperatures since, according to the methodology, it is the most adequate way to take advantage of this streams. The same number of heat exchangers are required in the cyclonic preheater configuration, figure 4b. In this case, the most relevant changes with respect to the reference case are the net power, 307 MW, and the fact that the CO_2 stream to compression is relocated from the HRSG to

Table 5: Reference case streams

	Flow description	T_{in} [°C]	T_{fin} [°C]	$\dot{Q}$[MW]
BC3	Purge flow	950.0	200.0	26.4
BC4	Recirculation to CR	950.0	650.0	365.0
BC5	CO_2 to compression	950.0	329.7	307.8
BC1	Surplus heat CR	650.0	650.0	157.1
BC2	Gas flow leaving CR	650.0	190.0	230.6
BC7	CO_2 to compression 2^{nd} step	329.7	190	44.3
BC6	$CO_2 + O_2$ to CL	180.0	296.4	-26.4

the preheating process. The number of heat exchangers is reduced in 2 in the mixing seal valve configuration, figure 4c. The main changes in this case are the net power, 292 MW, and the fact that the reheating process recovers heat from the carbonator, MV1, instead of making use of the solid stream recirculated to carbonator, since this stream is not available in this case as it is in the other configurations, BC4 and CP4.

4. Energy analysis

Heat integration methodology leads to the recovery of almost all the available heat in every configuration. There are only minor cooling requirements that amount 307 kW in the base case, 572 kW in the cyclonic preheater and 7 kW in the mixing seal valve.

As shown in figure 2, the temperature distribution of the available heat differs in each configuration. The base case has a greater fraction of high quality heat. Then, a question arises: will it be possible to drive a steam

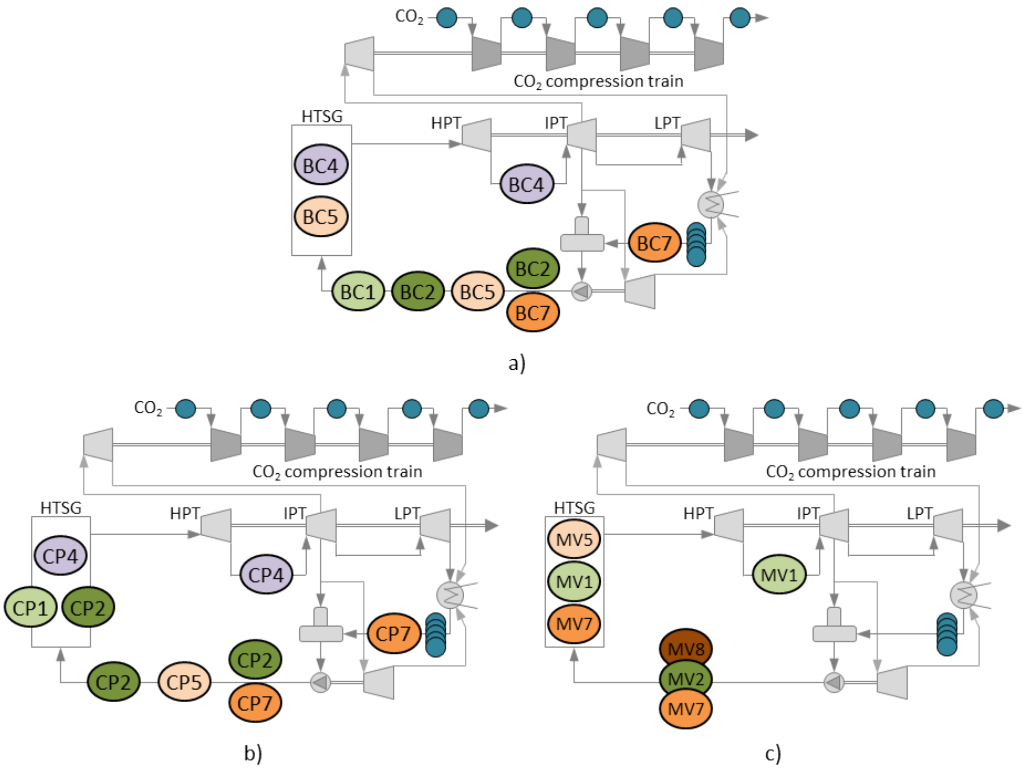

Figure 4: Steam cycle and compression train heat integration

Table 6: Cyclonic preheater streams

	Flow description	T_{in} [°C]	T_{fin} [°C]	$\dot{Q}$[MW]
CP3	Purge flow	950.0	200.0	25.4
CP4	Recirculation to CR	950.0	650.0	352.7
CP5	CO_2 to compression	705.0	330.0	163.6
CP1	Surplus heat CR	650.0	650.0	157.1
CP2	Gas flow leaving CR	650.0	190.0	230.6
CP7	CO_2 to compression 2^{nd} step	330.0	190	41.0
CP6	$CO_2 + O_2$ to CL	180.0	309.3	-25.4

cycle with the same energy efficiency? The answer is not straightforward since a fraction of the mechanical power production is directly used to drive the compression train. The steam cycle energy efficiency is calculated with the gross power production, $\dot{W}_{gross}$, that is the sumation of the mechanical power used to generate electricity and that consumed in the CO_2 compressors, table 8. Similar energy efficiencies of the steam cycle, about 45%, are thus obtained in every configuration. The difference lies in the size of the steam cycle, in terms of $\dot{W}_{gross}$, that is reduced a 14% in the case of the cyclonic preheater and a 17% in the case of the mixing seal valve.

The set containing the Ca-looping and the steam cycle was assessed, table 9. As in the steam cycle analysis, the gross power comprises the power used to generate electricity and that used in the CO_2 compression train. The efficiency also keeps constant in every configuration, about 37%. It is the size of the ASU which is reduced a 13% for the cyclonic preheater case and a 15% for the mixing seal valve case. The size of the Ca-looping is expected

Table 7: Mixing seal valve streams

	Flow description	T_{in} [°C]	T_{fin} [°C]	$\dot{Q}$[MW]
MV3	Purge flow	950.0	200.0	25.2
MV5	CO_2 to compression	950.0	696.4	115.9
MV7	CO_2 to compression 2^{nd} step	696.4	190	156.7
MV8	CO_2 to MV	696.4	180	37.8
MV1	Surplus heat CR	650.0	650.0	361.9
MV2	Gas flow leaving CR	650.0	190.0	230.6
MV6	$CO_2 + O_2$ to CL	180.0	388.1	-25.2

Table 8: Energy parameters of the steam cycle

Configuration	$\dot{Q}_i$[MW]	$\dot{W}_{gross}$[MW]	η_{SC}
Base case	1256	562	44.7%
Cyclonic preheater	1079	485	45.0%
Mixing seal valve	1037	465	44.9%

to diminish a 13% in the cyclonic preheater due to the coal consumption reduction. The size comparison in the case of the mixing seal valve is far more complicated since the solids circulation flow is significantly increased while the coal consumption is reduced. The reduction of the equipment sizes is important since it entails a decrease of the capital cost.

To assess the energy penalty related to the carbon capture (CC) process, CO_2 compression train has also been included in the integrated system, table 10. Analyzing the CC system which comprises the calcium looping cycle, the steam cycle and the CO_2 compression train, a slight reduction of the energy

Table 9: Energy parameters of the set containing calcium looping and steam cycle

Configuration	$\dot{Q}_{coal}$[MW]	$\dot{W}_{ASU}$[MW]	$\dot{W}_{CaL+SC}$[MW]	η_{CaL+SC}
Base case	1268	91	471	37.1%
Cyclonic preheater	1098	79	406	37.0%
Mixing seal valve	1053	76	390	37.0%

efficiency of 0.7 percentage points in the case of the cyclonic preheater and 1 percentage point in the mixing seal valve may be observed. It is due to the fact that the amount of CO_2 directed to the compression train is reduced in a lower extent than the steam cycle power. The CO_2 capture efficiency and, thus, the CO_2 from the power plant flue gas are equal in each configuration. Only the fraction of CO_2 coming from the coal combustion in the calciner is reduced in the new configurations.

Table 10: Energy parameters of the global system

Configuration	$\dot{W}_{CT}$[MW]	$\dot{W}_{CC}$[MW]	η_{CC}	η_{global}	EP
Base case	107	364	28.7%	34.3%	5.7%
Cyclonic preheater	100	307	28.0%	34.4%	5.6%
Mixing seal valve	98	292	27.7%	34.4%	5.6%

The global system consists of the initial 500 MW_e power plant and the CC process. As mentioned before, the initial power plant has a 40% energy efficiency. Table 10 shows the energy efficiency of the global system and the energy penalty associated with the CO_2 capture process. The configurations in which a fraction of the available heat is internally integrated to reduce the energy consumption in the calciner, that are the cyclonic preheater and the mixing seal valve configurations, present a slight reduction of the energy penalty compared to the ordinary base case.

5. Conclusions

The use of a fraction of the available heat from the Ca-looping to increase the temperature of the solids entering the calciner reduces the energy consumption in this reactor at the expense of available heat for external integration. In this study, the external heat integration of cyclonic preheater and mixing seal valve configurations has been carried out by means of a systematic procedure.

The reduction of the calciner energy needs implies a size diminution of various subsystems including the ASU, the steam cycle and the compression train. ASU power demand is reduced around 13-16%, steam cycle (gross), steam cycle between 14 and 17% and CO_2 compression train between 6.5 and 8.5%. The Ca-looping cycle is also smaller in the case of the cyclonic preheater configuration. Regarding the mixing seal valve configuration, the reactors' size of the Ca-looping cycle might be greater since significantly higher solid flows are required. The diminution of the CC components size entails lower capital costs, increasing the feasibility of the technology.

An energy efficiency assessment of the global system was carried out. The cyclonic preheater and the mixing seal valve configurations present a slight decrease of the energy penalty, around 0.1 efficiency points, associated with the carbon capture process compared to an ordinary Ca-looping configuration. This fact, combined with the size reduction of the equipment and, consequently, the expected investment savings makes the cyclonic preheater configuration the most adequate one to improve the viability of the technology. Further research is required in the case of the mixing seal valve configuration since, even when the energy efficiency is similar, the Ca-looping cycle costs might be higher.

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Nomenclature

$v_{\bar{CO_2}}$	Average volume fraction of CO_2
$\dot{Q}$	Heat available in a stream [MW]
$\dot{n}_{Ca}$	Inlet molar flow of CaO and $CaCO_3$ [kmol/s]
$\dot{n}_{CO_2}$	Inlet molar flow of CO_2 [kmol/s]
$\dot{Q}_{coal}$	Coal consumed in the calciner [MW]
$\dot{Q}_i$	Heat transferred in the HEN [MW]
$\dot{W}_{ASU}$	Power consumed in the ASU [MW]

$\dot{W}_{CaL+SC}$	Power generated in the set comprissing the Ca-looping and the steam cycle [MW]
$\dot{W}_{CC}$	Net power production of the set comprising the Ca-looping, the steam cycle and the compression train [MW]
$\dot{W}_{CT}$	Power consumed in the CO_2 compression train [MW]
$\dot{W}_{gross}$	Gross power generated in the steam cycle [MW]
$f_{a,CR}$	Fraction of active sorbent reacting in the carbonation fast reaction regime
k_{CR}	Surface carbonation rate constant $[s^{-1}]$
n_{Ca}	Molar sorbent inventory in the carbonator [kmol]
r_N	Age distribution of particles, fraction of particles that has accomplished N carbonation/calcination cycles
t^*_{CR}	Time for maximum fast kinetic-stage carbonation, X_{ave} [s]
T_{fin}	Stream final temperature [°C]
T_{in}	Stream initial temperature [°C]
v_{eq}	Volume fraction of CO_2 in equilibium conditions
X_N	Capture capacity of a fraction of sorbent that has accomplished N carbonation/calcination cycles
X_{ave}	Average maximum capture capacity of the sorbent

X_{in}	Inlet molar fraction of $CaCO_3$ with respect to CaO and $CaCO_3$
η_{CaL+SC}	Energy efficiency of the set comprissing the Ca-looping and the steam cycle
η_{CC}	Energy efficiency of the set comprising the Ca-looping, the steam cycle and the compression train
η_{CR}	Carbonation efficiency
η_{global}	Energy efficiency of the whole system comprising the initial 500 MW_e power plant, the Ca-looping, the steam cycle and the compression train
η_{SC}	Steam cycle energy efficiency
τ_{CR}	Carbonator space time. Molar inventory of calcium compounds (CaO and $CaCO_3$)) per molar flow of CO_2 [s]
φ	Gas-solid contacting effectivity factor
EP	Energy penalty
ASU	Air separation unit
BC	Base case
CC	Carbon capture
CCS	Carbon Capture and Storage
CL	Calciner

CP	Cyclonic preheater
CR	Carbonator
HEN	Heat energy network
HPT	High pressure turbine
HRSG	Heat recovery steam generator
IPT	Intermediate pressure turbine
LPT	Low pressure turbine
MV	Mixing seal valve

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