



Heat integration of alternative Ca-looping configurations for CO₂ capture



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ABSTRACT

The best option to overcome the energy penalty in Ca-looping is to take advantage of the surplus heat by external integration to produce additional power and increase net efficiency. As calciner represents the main energy consumption, another possibility is to internally use the surplus heat to preheat the solids entering this reactor. The objective of internal integration is to reduce the energy demand per captured tonne of CO₂. It represents a reduction of the coal and oxygen needs and also a total decrease in the CO₂ generation regarding the ordinary configuration. However, the amount of available heat for extra power generation by external integration, essential for the viability of this technology, is also reduced. This is the case of the configurations including a cyclonic preheater or a mixing seal valve. This study assess the energy penalty minimization that may be reached by external integration of these internal energy integration configurations. A methodological 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 the most suitable configuration to improve the viability of this technology.

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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 CO₂ 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 CO₂ 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 CO₂. When the energy required in the calciner is provided by oxyfuel combustion, the oxygen needs and the global amount of generated CO₂ 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 CO₂ 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 CO₂ 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 CO₂ 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.

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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 CO₂ 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 [13–18]. 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.

2. Ca-looping configurations and modelling

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

The three configurations are operated to obtain maximum CO₂ 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.

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).

$$X_{ave} = \sum_{N=1}^{\infty} r_N X_N \quad (1)$$

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].

$$X_N = a_1 f_1^{N+1} + a_2 f_2^{N+1} + b \quad (2)$$

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 CO₂ partial pressures makes necessary to use an advanced calcination model. This model was developed by Martínez et al. [23] and is summarized in Table 3.

The energy required for the sorbent regeneration is obtained from the oxy-fuel combustion of the coal defined in Table 1, to avoid CO₂ 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 CO₂ 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 CO₂ ratios to achieve the same CO₂ 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 CO₂, 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 affecting 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 CO₂ generation. Regarding the mixing seal valve, coal and oxygen savings reach 15% and CO₂ 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 Fig. 2 represent the amount of available heat in each configuration and their corresponding temperature levels. The curves show the pinch point,

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