



The impact of calcium sulfate and inert solids accumulation in post-combustion calcium looping systems

M. Elena Diego*, Borja Arias, Mónica Alonso, J. Carlos Abanades

Spanish Research Council, INCAR-CSIC, C/Francisco Pintado Fe, 26, 33011 Oviedo, Spain

HIGHLIGHTS

- ▶ The effects of sulfur and the accumulation of inerts are quantitatively discussed.
- ▶ Mass and energy balances were solved together with an updated carbonator model.
- ▶ It was found that the main impact on CaL performance is due to the sulfur inlet.
- ▶ A minimum purge is required even when an effective reactivating step is carried out.

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ABSTRACT

Postcombustion CO₂ capture by calcium looping (CaL) is being rapidly developed for coal combustion applications. This work discusses the impact of the accumulation of CaSO₄ and other inert solids on CO₂ capture efficiency and the overall CaL process performance. Several process configurations are considered, and the mass and energy balances and an updated carbonator reactor model are solved for each configuration. The minimum fresh sorbent requirements for sustaining a certain level of CO₂ capture efficiency are quantified as well as the effects of an increase in the make-up flow. It was found that the main effect on the CaL process is produced by the sulfur present in the coal fed to the calciner and in the flue gas entering the carbonator. For a typical set of operating conditions it was calculated that the deactivating effect caused by an increase of 0.5% in the sulfur content with respect to a reference coal (low ash content) fed to the calciner is similar to the effect caused by the accumulation of inerts when using a coal with 15% more ash.

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1. Introduction

CO₂ capture and storage (CCS) has emerged as a suitable option for reducing CO₂ emissions from large stationary sources such as coal power plants [1]. Nowadays there are mature CO₂ capture technologies that could be commercially deployed if there are reasonable incentives due to carbon prices [1]. In order to reduce the costs associated with CO₂ capture, a number of new technologies are also emerging. One of the most promising processes is post-combustion Ca-looping (CaL), which has experienced a rapid scale up in the last few years. It has already been tested in small facilities (10s kW_{th}) operating in full continuous mode [2,3] and has currently reached the experimental testing phase in a 1.7 MW_{th} pilot plant in La Pereda – Spain [4,5], a 200 kW_{th} facility at IFK – Stuttgart [6,7] and a 1 MW_{th} pilot plant at ETS in Darmstadt [8,9].

* Corresponding author. Tel.: +34 985119090; fax: +34 985 297662.

E-mail address: marlen@incar.csic.es (M.E. Diego).

Postcombustion CaL was first proposed by Shimizu et al. [10], and is based on the use of lime as a sorbent to capture CO₂ by means of carbonation/calcination cycles. The most suitable configuration for the application of CaL on a large scale involves the use of two interconnected circulating fluidized bed (CFB) reactors (carbonator and calciner as shown in Fig. 1). In this process, the flue gas generated in the power plant is directed to a carbonator, where CO₂ reacts at temperatures between 600 and 700 °C with a stream of CaO particles. As a result, CaCO₃ is formed and a CO₂ depleted gas leaves the carbonator. The partially carbonated solid stream enters the calciner together with a continuous make-up flow of limestone fed to this reactor to compensate for the decay of the CO₂ capture capacity of the sorbent with the number of carbonation/calcination cycles. In the calciner coal burns under oxy-fired conditions [10] to attain the temperatures required to convert both the CaCO₃ from the carbonator and the fresh sorbent back to CaO (around 900 °C). Although the heat demand in this reactor (coal and O₂) is high [10,11], the overall energy penalty of the CaL process is low [10,12–19], since energy can be recovered from

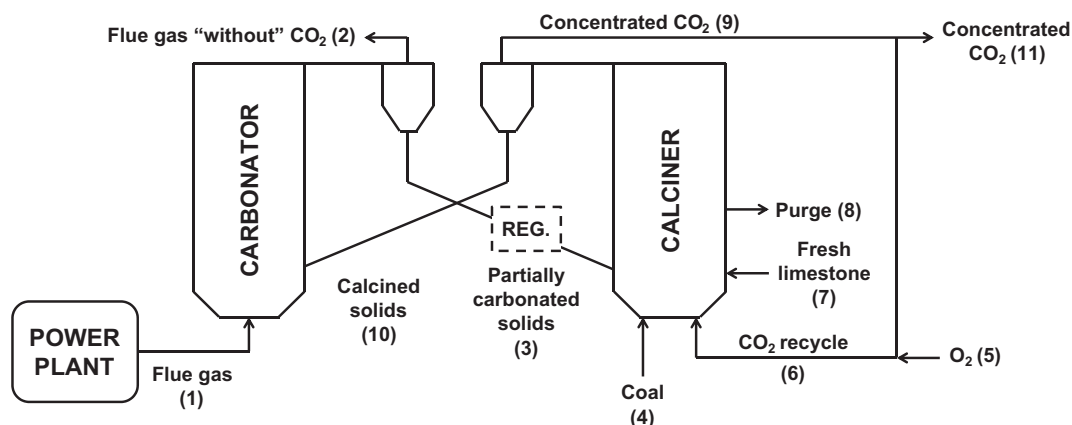


Fig. 1. General scheme of the CaL process for the three studied configurations. Dotted line indicates a possible location for the sorbent regenerator (REG.) in Configuration 3.

high-quality heat sources (the solids streams between reactors, the carbonator and the high temperature gases abandoning the reactors).

As a consequence of the nature of the CaL process, these systems have a continuous input of inert solids, mainly due to the coal fed into the circulating fluidized bed calciner but also because of the SO_2 in the flue gas entering the circulating fluidized bed carbonator. The SO_2 tends to react with the CaO present in both reactors of the system and forms CaSO_4 . In order to prevent the accumulation of inerts in the system, solids should be purged from the calciner, which will contain mainly CaO, CaSO_4 and ashes. The flow rate of the solids purge is defined from a mass balance of the inerts fed to the process and the fresh limestone added to the calciner. The ratio between these two variables also determines the composition of the total inventory of solids in the system, which is known to affect the performance of the calcium looping process in terms of CO_2 capture efficiency and heat requirements in the calciner [11,20,21]. Some previous works give an overall view of the CaL process by formulating the mass and energy balances of the whole system, and they analyze the performance of CaL under certain operating conditions, such as different make-up flows of limestone or different solids circulating rates between reactors [11,21], even in the presence of sulfur [12,20]. However, these studies do not analyze the influence of ashes and the formation of CaSO_4 on CO_2 capture efficiency from a carbonator reactor point of view. This is a critical relationship to be quantified in the system. For a certain set of operating conditions, the solids inventory in the circulating fluidized bed carbonator will be fixed, and an increase in the concentration of inert solids in the system will translate into a low inventory of active Ca inside the reactor, thereby reducing the CO_2 capture efficiency. The aim of this work therefore is to quantitatively discuss these effects by analyzing several scenarios in relation to different power plant and CO_2 capture configurations. For this purpose, mass and energy balances were solved together with an updated carbonator reactor model, allowing us to calculate the CO_2 capture efficiency for each scenario. This simulation exercise provided useful information to determine the minimum make-up flow of limestone required to sustain a certain level of CO_2 capture efficiency as a function of the quality of the coal fed to the calciner and the SO_2 content in the flue gas entering the carbonator reactor from the power plant.

2. Methodology for process simulation

The process configurations analyzed in this work follow the general scheme depicted in Fig. 1, in which the flue gas coming from the power plant is fed into the carbonator of the CaL facility.

Table 1
Outline of the process configurations used in this work.

Process configuration	Power plant	Sorbent reactivation
1	PC	No
2	CFBC	No
3	CFBC	Yes

Table 1 summarizes the different configurations of the process, depending on the type of power plant emitting flue gases, the availability of SO_2 capture from the flue gas (with a flue gas desulfurization (FGD) unit in the pulverized coal (PC) power plant or an in situ SO_2 capture in the circulating fluidized bed combustion (CFBC) power plant) or the presence of a reactivation process.

Configuration 1 consists of a PC power plant that uses low sulfur fuel with no flue gas desulfurization unit. Some previous works study the effect of SO_2 on the sorbent activity in postcombustion CaL [22–26]. They have shown that SO_2 accelerates the decrease in CO_2 carrying capacity during cycling. Therefore, some authors [23] have highlighted the need for desulfurization of the flue gas entering the carbonator. However, the possibility of using the CaL process as a CO_2 – SO_2 co-capture system translates into capital cost savings that may compensate for the additional limestone make-up requirements.

Configuration 2 is a CFBC power plant fitted with a CaL facility. High-efficient SO_2 capture (typically 90%) is assumed at the interior of the combustion chamber of the CFBC. For the purpose of this work, a similar PC + FGD + CaL configuration would give almost identical results to those obtained for this CFBC case. The only difference in favor of a CFBC power plant is that the purged material from the calciner can be used inside the CFBC as a sorbent to capture SO_2 from the flue gases.

Configuration 3 shows a similar scheme to that of Configuration 2, different only in that it incorporates a regenerator in the process. This reactivation step could be one of hydration [26–34], recarbonation [35] or any other means to increase the average activity of the circulating material or a fraction of such a solid stream. In order for the simulation to embrace any sorbent reactivation strategy no specific procedure to regenerate the solids is specified. The impact of the reactivation step is only considered through the increase in the average carrying capacity of the circulating material.

Mass and energy balances were solved for each configuration using an updated version of the carbonator model proposed by Alonso et al. [36]. This model assumes that the carbonator behaves as a continuous stirred tank reactor (CSTR) for the solids, so that the conversion of the particles is based on their residence time

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