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Modeling of the oxy-combustion calciner in the post-combustion calcium looping process

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HIGHLIGHTS

- ▶ Two model frames were created for the calcium looping process.
- ▶ 1-D model describing the carbonator, calciner and solid return system.
- ▶ 3-D model which describes the calciner reactor in more detail.
- ▶ A 1.7 MW pilot calciner reactor was examined with both models and validation of models was done.
- ▶ The process fuel consumption was then optimized by adjusting recirculation of solids.

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ABSTRACT

The calcium looping process is a fast-developing post-combustion CO_2 capture technology in which combustion flue gases are treated in two interconnected fluidized beds. CO_2 is absorbed from the flue gases with calcium oxide in the carbonator operating at 650 °C. The resulting $CaCO_3$ product is regenerated into CaO and CO_2 in the calciner producing a pure stream of CO_2. In order to produce a suitable gas stream for CO_2 compression, oxy-combustion of a fuel, such as coal, is required to keep the temperature of the calciner within the optimal operation range of 880–920 °C. Studies have shown that the calcium looping process CO_2 capture efficiencies are between 70% and 97%. The calciner reactor is a critical component in the calcium looping process. The operation of the calciner determines the purity of gases entering the CO_2 compression. The optimal design of the calciner will lower the expenses of the calcium looping process significantly. Achieving full calcination at the lowest possible temperature reduces the cost of oxygen and fuel consumption. In this work, a 1.7 MW pilot plant calciner was studied with two modeling approaches: 3-D calciner model and 1-D process model. The 3-D model solves fundamental balance equations for a fluidized bed reactor operating under steady-state condition by applying the control volume method. In addition to the balance equations, semi-empirical models are used to describe chemical reactions, solid entrainment and heat transfer to reduce computation effort. The input values of the 3-D-model were adjusted based on the 1-D-model results, in order to model the behavior of the carbonator reactor realistically. Both models indicated that the calcination is very fast in oxy-fuel conditions when the appropriate temperature conditions are met. The 3-D model was used to study the sulfur capture mechanisms in the oxy-fired calciner. As expected, very high sulfur capture efficiency was achieved. After confirming that the 1-D model with simplified descriptions for the sorbent reactions produces similar results to the more detailed 3-D model, the 1-D model was used to simulate calcium looping process with different recirculation ratios to find an optimal area where the fuel consumption is low and the capture efficiency is sufficiently high. It was confirmed that a large fraction of the solids can be recirculated to both reactors to achieve savings in fuel and oxygen consumption before the capture efficiency is affected in the pilot unit. With low recirculation ratios the temperature difference between the reactors becomes too low for the cyclic carbonation and calcination. As a general observation, the small particle size creates high solid fluxes in the calcium looping process that should be taken into account in the design of the system.

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1. Introduction

Carbon dioxide emissions from stationary sources like power plants burning fossil fuels have been acknowledged to be a contributor to the greenhouse gas phenomenon causing global warming. Carbon dioxide capture and storage (CCS) has emerged as an option to provide a partial solution to this problem [1]. Oxy-fuel combustion and post-combustion solvent techniques have been tested and demonstrated in various pilot scale units [2]. However, the need for more efficient and environmental friendly CCS techniques is a pressing issue [3,4]. The concept of the post-combustion calcium looping has already been proved at the laboratory scale and now testing on larger scale is being implemented [5,6]. In post-combustion calcium looping the CO_2 from flue gases is separated with two-interconnected fluidized beds. In the carbonator calcium oxide reacts with carbon dioxide from the flue gases at 650 °C forming calcium carbonate. After carbonation calcium carbonate is transferred to the calciner where it calcines around 900 °C to regenerate calcium oxide and a CO_2 flow suitable for transport and storage. Several models have been created and validated for the calcium looping carbonator however little interest has been paid to the calciner [7–10]. In this work, two modeling approaches are used to study the calciner of a calcium looping process, namely, the 1-D dynamic process model [11] and the 3-D steady-state calciner model. In the 1-D model the full calcium looping process including two interconnected fluidized bed reactors is simulated (Fig. 1) albeit with some simplifying assumptions, such as: no sulfur capture, no make-up flow, no ash accumulation and a uniform particle size. The use of the 1-D approximation and other simplifications for the selected case are justified by carrying out more detailed 3-D simulations for the calciner of the same case. After the validation based on the 3-D model, the 1-D process model is used to study the control of solid circulation between the reactors, and the optimal operation conditions based on capture efficiency and thermal power have been found for the selected cases. The 1.7 MW pilot unit built in Spain is used as the reference case for the simulations [6].

New features of the 1-D model have been described in Section 2. The main differences between the 1-D and 3-D models are described in Section 3. In Section 4.1 the results of the 1-D and 3-D simulations are compared and in Section 4.2, the hydrodynamics and operation of the calcium looping process can be studied using developed 1-D model. The results show clearly that the efficiency of the process is greatly influenced by the process hydrodynamics. In order to achieve the optimal operation conditions for the complicated dual fluidized bed system, effective solid control systems are needed. This is one of the first attempts to model the whole calcium looping process using a 1-D method, including all key reactions dealing with carbon capture and oxy-fuel combustion. Additionally, the calciner reactor has not been studied with such a detailed model as the 3-D model used in this work. After the experimental validation the developed models will be useful tools in the scale-up studies of the calcium looping process.

2. One-dimensional calciner model

2.1. Model frame

The one-dimensional model frame for the circulating fluidized bed reactor is similar to that presented in previous work [11]. Each reactor is discretized using the control volume method into vertical 1-D control volumes. Spatial derivatives are discretized using first order approximations with central difference or upwind scheme for convective fluxes. Time dependent balance equations for mass and energy are written for each element. A set of time dependent equations is solved using the fixed-step explicit ordinary differential equation solver in Simulink/Matlab system.The major difference between the carbonator and the calciner models is the description of chemical reactions, for mainly combustion. The calciner acts as an oxy-fuel combustor with the purpose of converting $CaCO_3$ to CaO. Following sections describe the additions to the existing model frame.

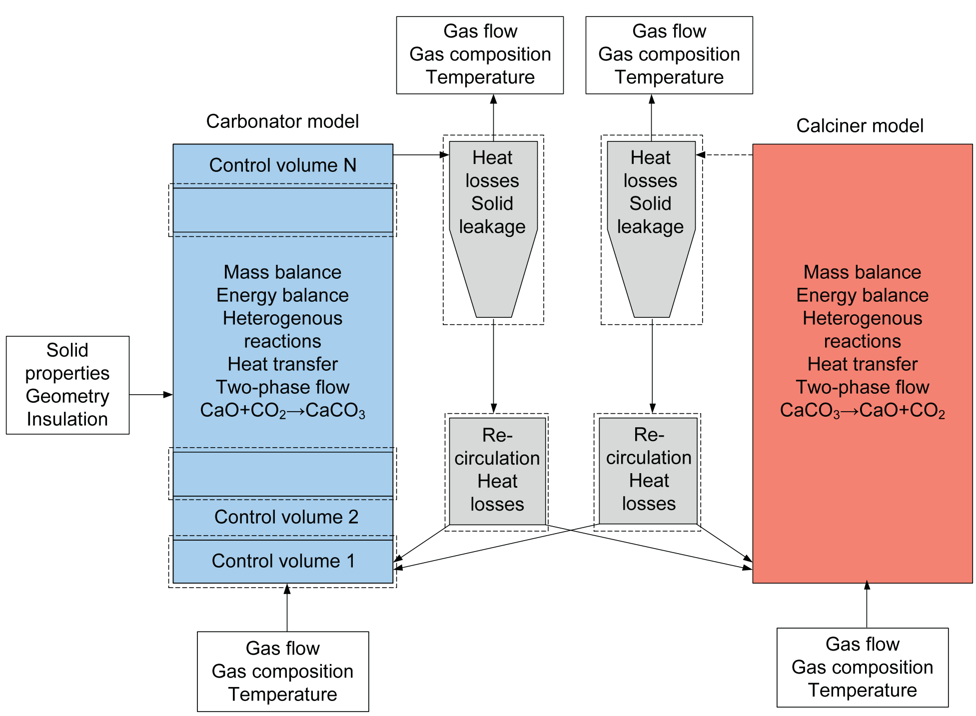

Fig. 1. 1-D dynamic model frame for the calcium looping process, including the carbonator and calciner, return leg and cyclones.

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