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Process modelling of the calcium looping process and validation against 1 MW_{th} pilot testing

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Abstract

The calcium carbonate looping (CaL) process is a promising post-combustion technology for CO₂ capture from fossil-fired power plants and carbon intense industries like steel and cement manufacturing. A CaL system consists basically of two interconnected circulating fluidized bed (CFB) reactors using natural limestone as a sorbent. Inside the carbonator, CO₂ contained in the flue gas is absorbed by the exothermic carbonation reaction, during endothermic sorbent regeneration in the calciner, CO₂ is released consequently. The CaL technology has proven its feasibility in semi-industrial scale by pilot testing in various test rigs all over the world. For the further development of the process, models are required for the prediction of process characteristics in terms of heat and mass flows. Most critical to the calculation of an overall process performance is the CO₂ absorption efficiency in the carbonator and, moreover, the decreasing CO₂ carrying capacity of the sorbent during operation. The enrichment of impurities like ash or calcium sulfate in the circulating sorbent as well as the reduction in particle size due to attrition needs also to be taken into account when assessing the performance of a CaL system. The main focus of this paper is the development of an advanced steady state process model and its validation against 1 MW_{th} long term pilot testing. The validation of the process model showed good agreement in predicting the CO₂ absorption efficiency of the carbonator as well as the sorbent regeneration in calciner. Furthermore, the composition, and the effect of attrition to particle size distribution of the circulating sorbent are reproduced with good accordance. The process model is further applied for a sensitivity study to show the influence of crucial parameters on the particle size reduction as well as on the calciner efficiency.

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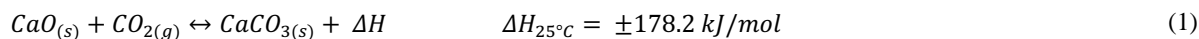
Keywords: CCS; calcium carbonate looping; process modelling; attrition; carbonation; calcination

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1. Introduction

In recent years, most of the world's energy is supplied by means of fossil fuels. According to a report of the International Panel on Climate Change (IPCC), the worldwide CO₂ emissions in 2013 were about 32.2 GtCO₂ from which 46 % can be directly attributed to the combustion of coal [1]. The application of carbon capture and storage (CCS) technologies account for the potential of efficient reduction of global CO₂-emission from large point sources. In addition to the end-of-pipe-storage of the CO₂ being captured, its utilization by means of carbon capture and utilization (CCU) processes has gained increasing interest during the past years [2, 3]. First generation CO₂-capture processes such as IGCC, solvent scrubbing and oxy-fuel combustion have the disadvantage of net efficiency losses up to 8 - 14 % points and high CO₂-avoidance costs [4]. A promising carbon capture technology first proposed by Shimizu et al. [5] is the CaL process using natural limestone as sorbent. The feasibility of the CaL process has been confirmed through successful operation of various pilot plants at different scale, varying from 200 kW_{th} in Stuttgart [6] up to 1 MW_{th} in Darmstadt [7, 8]. A 1.7 MW_{th} test facility is in operation at La Pareda, Spain [9], showing also promising results. In addition to the standard CaL process with directly heated calciner, an indirectly heated CaL process is currently under investigation. In order to show the feasibility of such process configuration, a 300 kW_{th} pilot plant with has been erected and successfully tested at TU Darmstadt [10, 11].

The CaL process takes advantages of the reversible reaction of calcium oxide and carbon dioxide, stated by Equation (1).



The process consists of two interconnected circulating fluidized beds reactors, operating at atmospheric pressure. In the carbonator, the CO₂ contained in the flue gas is absorbed by calcium oxide (CaO) forming calcium carbonate (CaCO₃). The exothermic carbonation reaction occurs between 600 and 750 °C, it has become evident, that the ideal temperature regarding kinetics and chemical equilibrium for the carbonation is around 650 °C. Due to the exothermic nature of the carbonation reaction, a constant reactor cooling is necessary to keep the temperature in the desired range for carbonation. The CO₂-lean flue gas is led to the atmosphere, while the solids mixture is transferred to the calciner where it is regenerated in an endothermic reaction (1). The required heat in the calciner is supplied by means of oxy-fuel combustion of coal, aiming at a high CO₂-concentration at the calciner outlet. In order to control the combustion temperature inside the reactor, a certain split stream from the calciner off gas is recirculated for oxygen dilution. To ensure an almost complete regeneration of the sorbent under high CO₂-partial pressure, calciner operation temperatures above the chemical equilibrium are required. However, by increasing the calciner temperature, sorbent deactivation is facilitated due to the sintering of the outer particle pores.

Since the CaL process in an emerging technology, the up-scaling and efficient design to an industrial size is faced with several challenges. Thereby process modelling and simulation tools are a matter of importance to understand the potential for optimization and effective process control strategies. The first carbonator models by Shimizu et al. [5] and Abanades et al. [12] were based on the bubbling bed model proposed by Kunii and Levenspiel [13]. More sophisticated models by Ströhle et al. [8, 14] and Romano [15] consider the hydrodynamics in the riser of the carbonator based on the work by Kunii and Levenspiel [16]. In contrast to the increasing knowledge on the carbonator modelling, merely little attention has been paid on a detailed analysis of the sorbent regeneration in the calciner. A satisfying performance of the calciner is obviously essential to guarantee the performance of the process.

Since fluidized bed reactors inhere a good mixing and large surface area, particle attrition is an important variable, which affects the reactor performance due to the particle elutriation and change in the particle size distribution (PSD) [17]. Attrition is the result of mechanical, thermal and chemical stresses that arise in and between the particles during operation. It is defined as the degradation of bed materials and leads to a change in size and number of particles [18]. A better understanding of attrition effects is needed to obtain an improved operation of fluidized bed reactors (FBR). The attrition of limestone in fluidized bed reactors has been researched in numerous works [19, 20]. The particle attrition is influenced by factors like particle porosity, particle size of the fresh limestone, material hardness, particle velocity and exposure time. If fresh material is exposed to attrition, the attrition rate peaks in the beginning and decays to a stationary attrition rate level. This has been attributed to the rounding off of the particles. The fresh particles, which are mainly irregular and angular, are smoothed and rounded

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