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Modeling the Calcium Looping Process with an Emphasis on the Bed Hydrodynamics and Sorbent Characteristics

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ABSTRACT: The calcium looping process is considered a promising technology to CO_2 capture emissions from combustion plants in recent decades. To model this process, the bed hydrodynamics as well as the sorbent characteristics will affect the calcium looping efficiency. In this study, CaO/Al_2O_3 sorbent is first synthesized by sol-gel method and then its performance is compared with pure CaO sorbent through 20 carbonation/calcination cycles. In addition, a general model based on bed hydrodynamics as well as sorbent properties for this process is presented and then the influence of parameters such as superficial gas velocity, carbonator height and sorbent inventory on process efficiency is investigated. Thermogravimetric experiments reveal that CaO/Al_2O_3 sorbent preserves 73% of its activity at the end of 20 cycles, whereas it is obtained as 21 for pure CaO sorbent. The results obtained from modeling show that the adsorption efficiency is decreased from 78.69 to 22.68% for pure CaO, whereas, it is decreased from 86.5 to 74.1% for modified CaO/Al_2O_3 sorbent. Finally, by studying the affective parameters it is obtained that the solid inventory has a significant impact on the process efficiency while the gas velocity and the height of the carbonator are far less effective.

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1. Introduction

Carbon Capture and Storage (CCS) technology is recognized as a promising technology to reduce CO_2 emissions from fossil fuel power plants and is classified into three categories including post-combustion, pre-combustion and oxy-fuel. The Ca-Looping (CaL) process which is an efficient post-combustion CO_2 capture technology using limestone-based sorbents is based on a reversible chemical reaction (so-called carbonation–calcination reaction of $CaO/CaCO_3$) as described in Eq. (1) [1]:

$$CaO_{(s)} + CO_{2(g)} \leftrightarrow CaCO_{3(s)};\ \Delta H_r = -178\ \text{KJ mol}^{-1} \quad (1)$$

The CaL process firstly proposed by Shimizu et al. [2], is a system consisting of two interconnected fluidized bed reactors acting as carbonator and calciner. In the carbonator reactor, particles of CaO adsorb CO_2 and the partially carbonated solids are driven into the calciner, where the endothermic calcination reaction occurs at high temperatures, typically above 900°C.

Since the CaL process is still a technology at an early stage of development, modeling works are of great importance for studying the effect of the diverse parameters governing the CaL process. This work is aimed to present a model of the carbonator reactor taking into account the fluidized bed hydrodynamics as well as the sorbent characteristics. Furthermore, the effects of important parameters such as gas velocity, height of the carbonator and solid inventory on the CO2 capture efficiency achieved by CaO and synthesized CaO/Al_2O_3 sorbents as a function of the cycle number are assessed.

2. Methodology

To prepare CaO/Al_2O_3 sorbent, sol-gel auto-combustion synthesis method is used [3]. The CO_2 adsorption efficiency of the carbonator is calculated by a 1D CFB model for fast fluidization as presented by Kunii and Levenspiel [4]. In this model, the reactor is divided into two regions: the dense region and the lean region.

To obtain the values for Hl and εse, Eqs. (2) and (3) are combined and subsequently solved using the Newton–Raphson method.

$$\varepsilon_{se} = \varepsilon_s^* + (\varepsilon_{sd} - \varepsilon_s^*)e^{-aH_l} \quad (2)$$

$$\frac{W_t}{A_t \rho_s} = \frac{\varepsilon_{sd} - \varepsilon_{se}}{a} + H_t \varepsilon_{sd} - H_l(\varepsilon_{sd} - \varepsilon_s^*) \quad (3)$$

In the recent years, Oritiz et al. [5] proposed a modified equation for describing the sorbent CO_2 carrying capacity with the number of cycles as the Eq. (4):

$$\frac{X_N}{X_1} = \frac{X_r}{X_1} + \left(\frac{1}{\kappa(N-1) + (1 - \frac{X_r}{X_1})^{-1}}\right) \quad (4)$$

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The CO_2 concentration at the top of the dense region, CCO2,d, is derived from the Eqs. (5) to (8):

$$X_{\max,ave} = \sum_{N=1}^{N=\infty} r_N X_N \tag{5}$$

$$K_{ri} = k_s \frac{X_{\max,ave} S_0 \rho_{CaO}}{M_{CaO}} (1-X)^{2/3} \tag{6}$$

$$K_{ff} = \gamma_{core} K_r + \frac{1}{(1/K_{be}) + (1/\gamma_{wall} K_r)} \tag{7}$$

$$\ln \frac{C_{CO_2,in}}{C_{CO_2,d}} = \frac{K_{ff}\, \delta H_d}{u_0} \tag{8}$$

The CO_2 concentration at the carbonator exit, CCO_2,ex, is calculated according to Eqs. (9) and (10):

$$\ln \frac{C_{CO_2,d}}{C_{CO_2,ex}} = \frac{(1-\varepsilon_f)_{bed} K_r}{u_0 a} \left[(1-e^{-aH_l}) - \frac{1-\eta_{bed}}{1+(a/a')} (1-e^{-(a+a')H_l}) \right] \tag{9}$$

$$\eta_{bed} = \left(\gamma_{core} + \frac{1}{(K_r/K_{be}) + (1/\gamma_{wall})} \right) \frac{\delta}{1-\varepsilon_f} \tag{10}$$

After obtaining the CCO_2,d and CCO_2,ex by Eqs. (8) and (9) respectively, the CO_2 capture efficiency is calculated by:

$$E_{CO_2} = 1 - \frac{C_{CO_2,ex}}{C_{CO_2,in}} \tag{11}$$

3. Discussion and Results

The CO_2 capture efficiency achieved by CaO and CaO/Al_2O_3 sorbents during 100 carbonation/calcination cycles is calculated. Capture efficiency achieved by CaO sorbent is decreased from 78.69% to 22.68% at the end of the cycle, indicating a very severe deactivation of CaO under CaL conditions, while it is decreased from 86.5% to 74.1% in the case of modified CaO/Al_2O_3 sorbents.

In order to evaluate different parameters affecting the CO_2 capture efficiency achieved by CaO and CaO/Al_2O_3 sorbents, solid inventory, gas velocity and height of the carbonator are investigated. The effect of each parameter is discussed as below:

An increase of the incoming solids flow results in a larger fraction of CaO in the bed and also increases the height of the dense phase, which is definitely the main factor affecting the CO_2 adsorption efficiency of the carbonator. As seen in Fig. 1, increasing the solid inventory of the CaO sorbent has a greater effect on CO_2 capture efficiency, but this effect is less in the case of the modified CaO/Al_2O_3 sorbent.

The gas velocity affects the particle distribution in the fast fluidized bed so that an increase of the superficial velocity of the gas results in a decrease in the height of the dense phase. As seen in Fig. 2, a smaller overall CO_2 capture efficiency is achieved at higher superficial gas velocities.

Keeping the solids inventory constant, the height of the dense region, Hd, decreases with increasing the height of the carbonator, according to the combination of Eqs. (2) and (3). As seen in Fig. 3, the overall CO_2 capture efficiency slightly

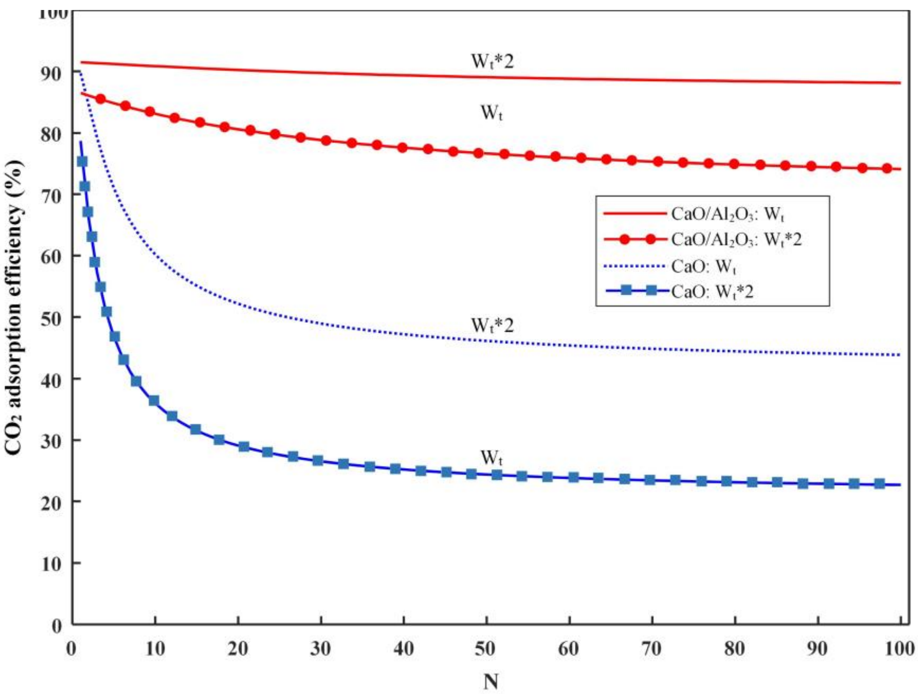

Fig. 1. Effect of the solids inventory on CO_2 adsorption efficiency achieved by CaO and CaO/Al_2O_3 sorbents

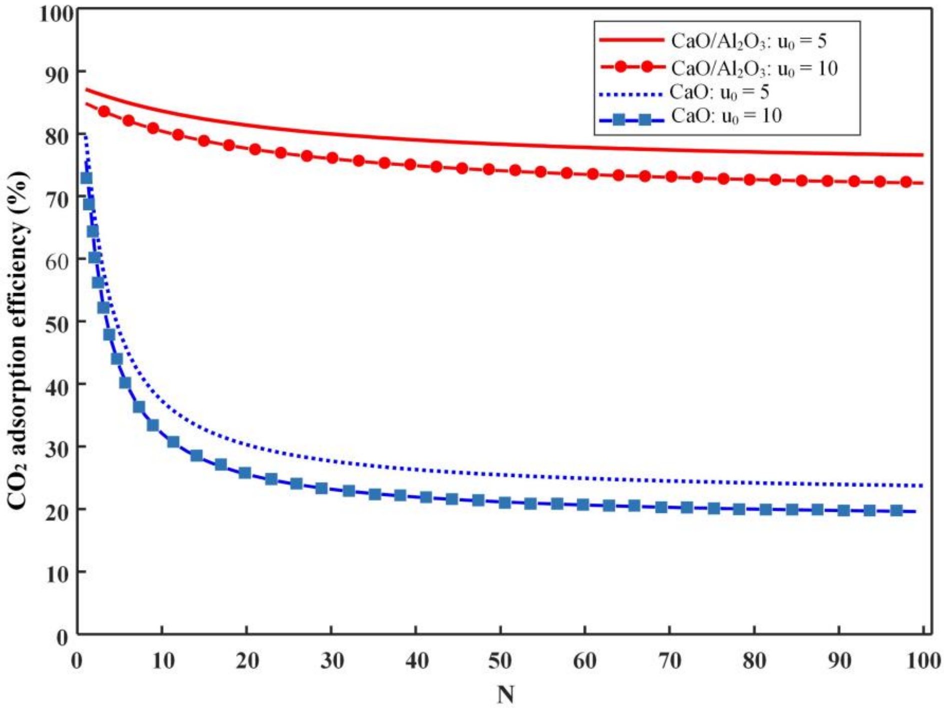

Fig. 2. Effect of the superficial gas velocity on CO_2 adsorption efficiency achieved by CaO and CaO/Al_2O_3 sorbents

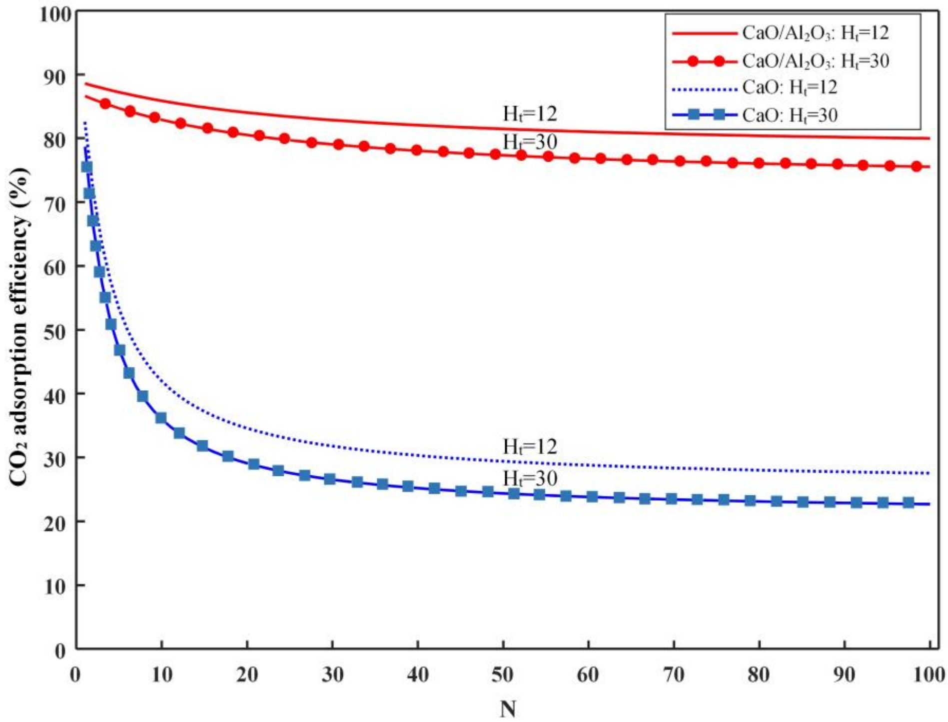

Fig. 3. Effect of total height of the carbonator on CO_2 adsorption efficiency achieved by CaO and CaO/Al_2O_3 sorbents

decreases because the CO_2 adsorption occurs mainly in the dense region. It seems that the importance of the effect of height of the carbonator and gas velocity on both types of sorbents is the same.

4. Conclusions

In this work a model was presented for calculating CO_2 capture efficiency of the carbonator, taking into account the fluidized bed hydrodynamics in the fast fluidity regime as well as the sorbent characteristics. As a main conclusion from the simulation results, the capture efficiency was significantly improved by applying CaO/Al_2O_3 as the CO_2 sorbent. During 100 multi-cycles, the capture efficiency by CaO/Al_2O_3 sorbent decreased from 86.5% to 74.1% and it decreased from 78.69% to 22.68% using CaO. Furthermore, the effects of three parameters including solid inventory, gas velocity and height of the carbonator on the CO_2 capture efficiency were investigated. It was obtained that the solid inventory has a significant impact on the process efficiency while the other two are far less effective.

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