Sensitivity Analysis of Carbonate Looping Process Using Twin Fluidized Bed Model

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ABSTRACT

CO₂ as the major greenhouse gas plays an important role in environmental problems. Therefore, it is reasonable to find an effective technology for mitigating large CO₂ emissions. Carbonating looping is an efficient post-combustion CO₂ capture technology using limestone sorbent, which is more energy-saving than traditional technologies. In our research, a carbonate looping process model had been developed using ASPEN PLUS software. In detail, the sensitivity analysis of main parameters were adopted. The simulation results indicated that the CO₂ capture rate of the whole process can achieve above 90% and CO₂ concentration in the decarbonated flue gas was less than 4% under the carbonation temperature, the calcination temperature, the F₀/F_{CO₂} and the gas-solid separation were 630°C, 900°C, 0.04, 1.0, separately. This work gave an potential example for the retrofitting current coal-fired power plant combined with carbonate looping process.

Keywords: CO₂ capture; Carbonate looping; Process model; Twin Fluidized bed; Sensitivity analysis.

INTRODUCTION

In recent years, scientists and researchers have put much more concern on the climate change issues, which surely confirm that global warming is mainly caused by greenhouse gas emission (GHG) (Safdarnejad *et al.*, 2016; Cheng *et al.*, 2019; Huang *et al.*, 2019). GHG is mainly emitted from fossil fuel burning which is considered as the main source of global warming. Among all the GHG, CO₂ is the principal gas which related to many large power plants (Symonds *et al.*, 2017; Tritipayanon *et al.*, 2019; Safarian *et al.*, 2020). Therefore, it is necessary to develop effective technologies for reducing large CO₂ emissions (Li *et al.*, 2013; Jiang *et al.*, 2019). CO₂ capture and storage (CCS) is a promising technology to control GHG discharge, which has been widely used in these decades (Zhang *et al.*, 2019; Liu *et al.*, 2020). There are several methods for CO₂ capture from power plants: (i) oxy-fuel combustion (Atsonios *et al.*, 2015); (ii) pre-combustion (Solares *et al.*, 2020); (iii) post-combustion (Alam *et al.*, 2020). The post-combustion method is widely used method for CO₂ capture in power stations at present. The

flue gas passes through CO₂ removal equipment to mitigate CO₂ emission (Liang *et al.*, 2015; Martinez *et al.*, 2018).

There have been many methods in the literature about post-combustion CO₂ captures, such as absorption (Porter *et al.*, 2015), adsorption (Shah and Imae, 2016), cryogenic (Yuan *et al.*, 2014), chemical looping (Leion *et al.*, 2008) and membrane (Zhao *et al.*, 2018; Yang *et al.*, 2019). However, these processes have some bottlenecks including enormous energy losses of 8–14% which increase fuel consumption and environmental concern regarding solvent regeneration (Lasheras *et al.*, 2011; Oh *et al.*, 2016; Liu *et al.*, 2019a, b). In addition, some researchers have proposed that the chemical looping combustion consumes less energy to separate CO₂ from flue gas, but this method still need full-scale experimental development (Abad *et al.*, 2007; Leion *et al.*, 2008). The carbonate looping (CL) is one of the promising methods for post-combustion CO₂ capture, which uses calcium oxide (CaO) as absorption sorbents to reduce CO₂ emission (Diego *et al.*, 2017). The main equipment of this process is two interconnected fluidized beds. The principal of the carbonate looping process is shown in Fig. 1.

In Fig. 1, the flue gas which comes from an upstream power plant firstly enters the carbonator. In the carbonator, most of the CO₂ react with CaO and convert to CaCO₃. The exothermic carbonation reaction happens at 650°C and a pressure of 1 bar which can achieve a better conversion rate of CaO (Strohle *et al.*, 2014). The CaCO₃ then transfers to

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Fig. 1. Carbonate looping process principle.

the second fluidized bed (calciner), where the CO_2 is released from CaCO_3 at about 950°C , forming high purity CO_2 product and ready to compress for storage. The regenerated CaO from calcination reaction is then sent to the carbonator for another cycle. The calcination reaction is endothermic, therefore the extra heat is supplied by the combustion of fuel and pure oxygen. In the whole process, high temperature will cause deactivation of sorbent and the over-time cycle will also lead to solid losses. Therefore, make-up flow should be introduced into the system to keep the mass balance in the system (Jia *et al.*, 2007; Zhang *et al.*, 2016; Alonso *et al.*, 2018). Compared with other post-combustion CO_2 capture technologies, CL has extremely lower energy penalties and the sorbent is widely distributed in nature and easy to obtain (Romano, 2009; Pan *et al.*, 2017). In addition, CL using indirectly heated method will be further improving CO_2 capture efficiency (Hoeftberger *et al.*, 2016; Reitz *et al.*, 2016).

The feasibility of the CL process has been evaluated by many researchers through large-scale test rigs and simulations. In 2009, Symonds *et al.* (2009) testified that humid environment can increase carbonation conversion. In 2010, Alonso *et al.* (2010) designed and built a $30 \text{ MW}_{\text{th}}$ test facility in Spain, and conducted a series of experiments about CO_2 capture performance. The results showed that CO_2 capture efficiency can be achieved above 70% as the reactors had enough solid inventory. A $10 \text{ kW}_{\text{th}}$ dual fluidized bed (DFB) was erected in the University of Stuttgart (Germany) by Charitos *et al.* (2010). Some parameters, such as carbonate temperature, calcium looping ratio, and carbonation reaction time, etc., were investigated through this experiment. The experiments demonstrated that the increased carbonate temperature and calcium looping ratio can significantly boost CO_2 capture efficiency, meanwhile, the CO_2 capture efficiency of the whole system can reach above 90%. On the other hand, Lasheras *et al.* (2011) developed a process model using a 1D fluidized bed model through ASPEN PLUS and Strohle *et al.* (2014) established a 1 MW_{th} test rig to validate the simulation. These two works showed that make-up flow

had a great influence on the CO_2 absorption rate and the experiment had a great agreement with simulation. In 2019, Hilz *et al.* (2019) established a $20 \text{ MW}_{\text{th}}$ CL demonstration power plant, which showed good prospects for both economical and technical applications.

Present work focused on CO_2 capture using carbonate looping based on earlier reported work by Shimizu *et al.* (1999). The flue gas came from $350 \text{ MW}_{\text{th}}$ net electrical power, which had a high temperature and flow rate. The whole process was introduced and set up into ASPEN PLUS software for numerical simulation of CO_2 capture performance. The carbonation and calcination temperature, make-up flow, gas-solid separation efficiency, and SO_2 concentration in the flue gas were analyzed and optimal parameters were obtained. The work will provide important data for the application of carbonate looping.

METHODOLOGY

Model Description

In the present research, a carbonate looping process model was established using ASPEN PLUS software and the process flow diagram was demonstrated as shown in Fig. 2. A coal-fired power plant that has $350 \text{ MW}_{\text{th}}$ net electrical power and 36% net efficiency was used as an upstream plant in this study. The sulfur dioxide gas was removed when the flue gas passed through the carbonate looping process. The main parameters of flue gas were shown in Table 1.

The flue gas initially entered the carbonator and the two chemical reactions occurred in carbonator:

The CO_2 absorption reaction (R1) happened in the first fluidized bed (CARBONATE) and the SO_2 absorption reaction rate (R2) was set as 100%. The solid stream and the

Fig. 2. A process model of carbonate looping in ASPEN PLUS.

Table 1. The properties of the flue gas.

Main parameters	Value
Mass flow (kg s^{-1})	446
Temperature ($^{\circ}\text{C}$)	120
Pressure (kPa)	101.325
Component of flue gas	Mass fraction (wt%)
CO_2	19.4
N_2O	15.53
SO_2	0.1
O_2	2.478
N_2	62.64

decarbonated flue gas were separated by the cyclone (SEP1), in which gas-solid efficiency was 100%. After separation, the decarbonated flue gas was cooled down to around 100°C (COOLER-1) and discharged directly into the atmosphere. On the other hand, the solids were partly removed from the system (PURGE) due to the deactivation of sorbent and the rest solids were transferred into the calciner (CALCINER).

In the calciner, the reversed reaction happened at around 900°C and CO_2 released from CaCO_3 . Because the reversed reaction was endothermic, the whole system needed external heat, which we adopted coal combustion to supply the heat. The coal was first pulverized into powder (DECOMP) and pushed into the calciner to combust with pure oxygen. Meanwhile, the make-up flow (MAKE-UP) consisted of 100% CaCO_3 continually entered the calcinator for the

supplement of emitted sorbent, which made sure the mass balance of the whole process.

After the reaction in the calcinator, the stream passed through the separator (SEP) first to discharge the CaSO_4 , the rest stream then flowed to the second cyclone (SEP2), where the CO_2 of the whole system was separated from the solid and part of the CO_2 ($\text{CO}_2\text{-2}$) was recycled to the calcinator, the another was compressed for storage. Meanwhile, the CaO sorbent was sent into the carbonator for another cycle.

Simulation Assumptions

The simulation of the carbonate looping process was based on ASPEN PLUS and the main equipment parameters were shown in Table 2. To simplify the calculation, the following assumptions were adopted: (1) The reaction time did not affect the whole process; (2) There was no pressure loss in the whole process; (3) The combustion of coal was complete; (4) The by-product (ash and CaSO_4) was completely extracted from the process.

Table 2. Main equipment parameters.

Main equipment	Items	Value
Carbonator	Temperature ($^{\circ}\text{C}$)	650
Calcinator	Temperature ($^{\circ}\text{C}$)	900
	Make-up flow (kmol h^{-1})	355
	Coal flow (t h^{-1})	120
Cyclone	Separation efficiency (%)	100

Process Evaluation

To evaluate the performance of the whole carbonate looping process, two main indexes were introduced into this work: CO₂ capture in the carbonator, and CO₂ capture in the whole process. The calculation of these two indexes was summarized as follows:

$$X_{carb} = 1 - \frac{CO_{2, flueout}}{CO_{2, fluegas}} \quad (1)$$

The CO₂ capture rate in the carbonator, X_{carb} , was defined as the relationship between CO₂ concentration in the decarbonate flue gas, $CO_{2, flue out}$, and the CO₂ concentration in the flue gas, $CO_{2, flue gas}$.

$$X_{CO_2} = \frac{CO_{2, captured}}{CO_{2, fluegas} + CO_{2, makeup} + CO_{2, coal}} \quad (2)$$

The CO₂ adsorption efficiency in the whole process, X_{CO_2} , was explained as the relationship between captured CO₂, and produced CO₂. The produced CO₂ mainly contained two parts: the make-up flow, $CO_{2, make up}$, and the combustion of the coal, $CO_{2, coal}$.

RESULTS AND DISCUSSION

Effect of Carbonation Temperature

Carbonation temperature had an effect on the CO₂ capture rate which had been presented in Fig. 3. The basic simulation conditions were shown in Tables 1 and 2.

In Fig. 3, with increasing carbonation temperature from 600 to 650°C, the CO₂ capture rate in carbonator increased steadily first and then dropped sharply to 82%. For CO₂ concentrations in the decarbonated flue gas, when carbonation

temperature increased, it decreased firstly and then increased rapidly. This trend showed that high carbonation temperature can boost the conversion rate of the absorption reaction (R1), which was a benefit for CO₂ capture. When the carbonation temperature was 630°C, the CO₂ capture rate in the carbonator reached the highest value which was 90% and CO₂ concentration in the decarbonated flue gas was 2.5% which was the lowest point. However, the higher carbonation temperature was not good for the CO₂ capture. When the temperature was above 630°C, the over-high temperature had a bad effect on both the CO₂ capture rate and CO₂ concentrations in the flue gas. That was because CaO absorption was an exothermic reaction and higher temperature was detrimental to the adsorption reaction.

The effect of carbonation temperature on CO₂ capture of the whole process and CaCO₃ circulating flow rate were shown in Fig. 4 and obtained parameters were represented in Tables 1 and 2. It can be seen from Fig. 4 that the CO₂ capture rate of the whole process and CaCO₃ circulating flow rate had the same trend when the carbonation temperature was increased from 600 to 650°C. The increasing carbonation temperature had two contrary effects. On the one hand, when the temperature increased from 600 to 630°C, more CO₂ was captured and more limestones were generated and circulated in the whole process, therefore, both CO₂ capture rate and CaCO₃ circulating flow grew. On the other hand, over-high temperature hindered the absorption reaction rate. That was why both two lines dropped sharply when the temperature increased from 630 to 650°C. From the obtained results, it was concluded that the carbonation temperature significantly affected the performance of the process. Meanwhile, when the carbonation temperature was 630°C, the CO₂ capture rate and CaCO₃ circulating flow rate had the highest value. So, we chose 630°C as the optimal carbonation temperature for further considerations.

Fig. 3. CO₂ capture rate in carbonator and CO₂ concentrations in the decarbonated flue gas along with the changes of carbonation temperature.

Fig. 4. CO₂ capture rate of the whole process and CaCO₃ circulating flow rate as the function of carbonation temperature.

Effect of Calcination Temperature

When the calcination temperature changed from 850 to 1250°C, the carbonation temperature and coal mass flow were set as 630°C and 120 t h⁻¹, respectively. The variation of CO₂ capture rate in the carbonator and the whole process, CO₂ concentrations in the decarbonated flue gas, and CaCO₃ circulating flow along with the calcination temperature were calculated and analyzed.

Fig. 5 showed that the CO₂ capture rate in the carbonator decreased firstly and then increased with the calcination temperature rose, on the contrary, CO₂ concentrations in the decarbonated flue gas rose firstly and then dropped down. The reason for the trend was that the rising temperature caused the deactivation of sorbent which called sintering effect. Sintering can weaken CO₂ capture capacity and reduce cycle times of the sorbent. When the calcination temperature was 1175°C, the CO₂ capture rate in the carbonator was only 80% and CO₂ flue gas reached the highest point of 4.5%. That was unacceptable for the system. However, as the calcination temperature varied from 1150 to 1250°C, more CaSO₄ decomposed into CaO and make-up introduced into the whole process. Thus, the more CO₂ was capture in the carbonator, which increased the CO₂ capture rate in the carbonator and decreased the CO₂ concentration in the flue gas.

The effect of calcination temperature on the CO₂ capture rate in the whole process and CaCO₃ circulating flow was shown in Fig. 6. As the calcination temperature increased, the CO₂ capture rate in the whole process and CaCO₃ circulating flow showed the same pattern (See Fig. 5). Sintering can change and deactivate the micro-granular of the sorbent, which caused a rapid decline of the CO₂ capture rate. On the other hand, when the temperature was higher than 1175°C, the CaSO₄ decomposition and make-up flow lines moved upward by indicating a sharp increment of the CO₂ capture rate.

The relationship between the energy consumption and the calcination temperature was shown in Fig. 7. The energy

consumption increased steadily with the calcination temperature increased. This behavior was observed because a higher calcination temperature needed more coal combustion to provide extra heat, and it increased the system load and caused much more energy consumption.

From the above-obtained results (Figs. 5, 6 and 7), it can be concluded that when the calcination was 900°C, the CO₂ capture rate in carbonator and the whole process, CaCO₃ circulating flow and energy consumption were found to be 89%, 91%, and 74.9 MW, respectively, which were the best in the process. Therefore, 900°C was chosen as the optimal calcination temperature.

Effect of Make-up Flow

Make-up flow (F_0) was related to CO₂ concentration in the flue gas (F_{CO_2}), therefore F_0/F_{CO_2} had been chosen to be a variable in further study. The carbonator and calcination temperature, coal consumption was set at 630°C, 900°C, and 120 t h⁻¹, respectively. The effect of F_0/F_{CO_2} on CO₂ capture in the carbonator and CO₂ concentration in the decarbonated flue gas were calculated and analyzed.

It can be seen from Fig. 8 that, with the increase of make-up flow, CO₂ capture in the carbonator had a significant increase and CO₂ concentrations in the decarbonated flue gas decreased, which indicated that more make-up flow was good for the system. On the other side, a certain level of make-up flow increase was not adequate for CO₂ capture conditions. When the F_0/F_{CO_2} was above 0.04, the trend of CO₂ capture rate in the carbonator and CO₂ concentrations in the decarbonated flue gas was stabilized gradually due to the oversaturation of CaO in the system. The trend showed that the appropriate amount of CaCO₃ supplement was necessary, and it can update the deactivated sorbent.

Fig. 9 reflects the effect of make-up flow on the CO₂ capture rate in the whole process and CaCO₃ circulating flow rate, which indicated the same increase pattern when the make-up flow increased. When F_0/F_{CO_2} was lower than 0.04,

Fig. 5. CO₂ capture rate in the carbonator and CO₂ concentrations in the decarbonated flue gas along with the changes of calcination temperature

Fig. 6. CO₂ capture rate in the whole process and CaCO₃ circulating flow rate along with the changes of calcination temperature.

the CO₂ capture rate of the whole process increased rapidly. This was observed because of the initial CaCO₃ produced by carbonation was fewer than the initial circulation flow rate and when the CaCO₃ stream and the make-up flow mixed, the circulating flow rate surged.

In combination with Figs. 8 and 9, when the optimal F_0/F_{CO_2} was 0.04, the CO₂ capture rate in the carbonator and whole process, CO₂ concentration in the decarbonated flue gas and CaCO₃ circulating flow rate reached the best value, which were 86.4%, 91.6%, 3.1%, and 642.9 t h⁻¹, respectively.

Effect of Gas-solid Separation Efficiency

When the gas-solid separation efficiency varied from 0.95

to 1.0, the carbonation temperature, calcination temperature, and F_0/F_{CO_2} were set at 630°C, 900°C, and 0.04, respectively. The change of CO₂ capture in the carbonator and CO₂ concentrations in the decarbonated flue gas were calculated and analyzed.

From Figs. 10 and 11, it can be seen that the CO₂ concentrations in the decarbonated flue gas and CaCO₃ circulating flow rate were linear with gas-solid separation efficiency, which indicated that gas-solid separation efficiency had a great impact on the system stability. The CO₂ concentrations in the decarbonated flue gas linearly decreased with the gas-solid separation efficiency increased, however, CO₂ capture rate in the carbonator and the whole process and

Fig. 7. Energy consumption of the calcination as a function of calcination temperature.

Fig. 8. CO₂ capture rate in the carbonator and CO₂ concentrations in the decarbonated flue gas along with the changes of make-up flow.

CaCO₃ circulating flow rate showed the opposite trend. When the gas-solid separation efficiency was in the range of 0.95 to 1, the CO₂ capture rate in the carbonator increased from 87% to 89%, the CO₂ concentrations in the decarbonated flue gas decreased from 3.1% to 2.59%, and the CaCO₃ circulating flow also increased accordingly. Therefore, the gas-solid separation efficiency directly affected the system performance. It is observed that the higher gas-solid separation efficiency can achieve more favorable CO₂ adsorption rate. Herein, the design of the cyclone should try to ensure a high gas-solid separation efficiency. When the gas-solid separation efficiency was too low, the loss of effective CaO sorbents increased, and the concentration of the sorbent gradually

decreased after circulation, which caused CO₂ cannot be captured. In the present work, we chose 1.0 as gas-solid separation efficiency.

Effect of SO₂ Concentration in the Flue Gas

When the SO₂ concentrations in the flue gas varied from 0.1% to 1.0%, the carbonation temperature, calcination temperature, F_0/F_{CO_2} , and gas-solid separation efficiency was set at 630°C, 900°C, 0.04 and 1.0, separately. The change of CO₂ capture rate in the carbonator and CO₂ concentrations in the decarbonated flue gas were calculated and analyzed.

It can be seen from the Fig. 12 that the SO₂ concentrations in the flue gas had a great effect on the CO₂ capture rate in

Fig. 9. CO_2 capture rate in the whole process and $CaCO_3$ circulating flow rate along with the changes of make-up flow.

Fig. 10. CO_2 capture rate in the carbonator and CO_2 concentrations in the decarbonated flue gas along with the changes of gas-solid separation efficiency.

the carbonator. When the SO_2 concentrations in the flue gas increased from 0.1% to 1%, the CO_2 capture rate in the carbonator decreased from 88.3% to 63.7%, which dropped by 21.6%. This trend also showed in the carbonator and CaO not only reacted with CO_2 , but with SO_2 , and meanwhile, the density of $CaSO_4$ was larger than $CaCO_3$, which was not good for CO_2 diffusion process and greatly reduced the CO_2 capture rate in the carbonator. Therefore, the CO_2 concentrations in the decarbonated flue gas increased.

Fig. 13 showed CO_2 capture rate in the whole process and $CaCO_3$ circulating flow rate decreased with SO_2 concentrations in the flue gas increased. That was because the large increase of SO_2 in the flue gas, the more CaO was converted into $CaSO_4$. Meanwhile, $CaSO_4$ was not completely decomposed at $900^\circ C$

in the calcinator. Therefore, more $CaSO_4$ was discharged from the system, which significantly reduced the CO_2 capture in the whole process and $CaCO_3$ circulating flow.

Combining Figs. 12 and 13, when the SO_2 was introduced into the system, the CaO sorbent reacted with SO_2 firstly and then with CO_2 , which formed the stable $CaSO_4$ solid and reduced circulating flow rate in the whole system. Therefore, the SO_2 concentration in the flue gas had a huge influence on CO_2 capture performance and to ensure the CO_2 capture rate, the SO_2 was mostly removed from the flue gas before came into the process and SO_2 concentration in the flue gas was set as 0.1 in the work.

Under the condition that the carbonation temperature was $630^\circ C$, the calcination temperature was $900^\circ C$, the F_0/F_{CO_2}

Fig. 11. CO₂ capture rate in the whole process and CaCO₃ circulating flow along with the changes of gas-solid separation efficiency.

Fig. 12. CO₂ capture rate in the carbonator and CO₂ concentrations in the decarbonated flue gas along with the changes of SO₂ concentrations in the flue gas.

was 0.04, the gas-solid separation was 1.0 and the SO₂ concentration in the flue gas was 0.1, the simulation was carried out, see Table 3. The results could satisfy for the CO₂ capture rate of over 90% and CO₂ concentration in the decarbonated flue gas was less than 4.0%. By contrast, Lasheras *et al.* (2011) obtained CO₂ capture rate of 80% in a 1052 MW_{el} coal-fired power plant. The carbonate looping used in this study can achieve above 90% CO₂ capture rate. Thus, the results of this work was reasonable.

CONCLUSIONS

In order to remove and capture CO₂ from flue gas, the

carbonate looping process was more suitable for operating conditions with large flow flue gas. In the work, the proposed carbonate looping process was established with ASPEN PLUS to obtain optimal parameters of the carbonate looping process. The process adopts a reversible reaction $\text{CaO} + \text{CO}_2 \leftrightarrow \text{CaCO}_3$ into account and formed a circulation that would obtain higher CO₂ product gas and continually recycle the sorbent. Also, due to particle attrition and sintering of CaO, the make-up flow should be introduced into the system. Meanwhile, the sensitivity analysis of several parameters was conducted and the optimal parameters were selected and listed in Table 3. The main results of this work were as follows:

Fig. 13. CO₂ capture rate in the whole process and CaCO₃ circulating flow rate along with the changes of SO₂ concentrations in the flue gas.

Table 3. Summary of carbonate looping process.

Results parameters	Numerical value
Carbonation temperature (°C)	630
Calcination temperature (°C)	900
F_0/F_{CO_2}	0.04
Gas-solid separation efficiency	1.0
SO ₂ concentration in the flue gas	0.1

1. The proper temperature increase of carbonate and calcination would be beneficial to the reaction, which can increase the CO₂ capture rate. However, the over-high temperature can cause deactivation of sorbents and decrease of the solid circulating flow;
2. The increase of make-up flow increased the solid circulating flow and the activated CaO sorbent which significantly promoted CO₂ capture rate. when the F_0/F_{CO_2} was 0.04, the CO₂ capture rate in the whole system was 91.6% and reached the highest value;
3. The gas-solid efficiency and SO₂ concentrations in the flue gas both had huge influence on the CO₂ capture rate. When the gas-solid efficiency decreased from 1.0 to 0.95, the CO₂ capture rate dropped by 2%. Meanwhile, as the SO₂ concentrations in the flue gas increased from 0.1% to 1.0%, the CO₂ capture rate decreased by 21.6%;
4. When the carbonation temperature was 630°C, the calcination temperature was 900°C, the F_0/F_{CO_2} was 0.04, the gas-solid separation was 1.0 and the SO₂ concentration in the flue gas was 0.1, the target of the CO₂ capture of > 90% and CO₂ concentrations in the decarbonated flue gas < 4.0% could be achieved.

The simulation results were applicable to the current situation of most stations which provide a good example for the utilization of CO₂ capture in power plants. However, there were also some important parameters, such as pressure drop, solid fraction, superficial velocity of carbonator, and

so on, which we did not discuss in the work. These parameters also play an indispensable role in system performance, which will be a challenge for us in the future.

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