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# Solution combustion synthesis of MgO-stabilized CaO sorbents using polyethylene glycol as fuel and dispersant

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The main limit for the calcium looping process is the sharp decrease of the capture capacity of the CO<sub>2</sub> sorbents during multiple cycles. In this research, a solution combustion method was employed to synthesize MgO-stabilized CaO sorbents. Polyethylene glycol (PEG) was used as the fuel and dispersant, with the purpose to enhance the uniformity of the Ca and Mg distributions in the sorbent. The results show that highly reactive MgO-stabilized CaO sorbents can be obtained through a solution combustion method using PEG as the fuel and dispersant. The existence of MgO can effectively restrain the sintering of the sorbent, resulting in a more porous and stable micro-structure of the sorbent. The CO<sub>2</sub> capture capacity of the MgO-stabilized CaO sorbent synthesized under the optimum conditions is 0.40 g(CO<sub>2</sub>)/g(sorbent) after 20 cycles, which is 75.3% higher than CaCO<sub>3</sub>.

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## 1. Introduction

In response to global climate change, the Chinese government has promised the world to reach its carbon peak by 2030 and achieve carbon neutrality by 2060.<sup>1</sup> To this end, it is of utmost importance and a significant challenge to promote CO<sub>2</sub> capture from the major emission sources.<sup>2</sup> The calcium looping process (CLP), which is based on the reversible carbonization reaction of lime, was proposed as a feasible and effective post-combustion CO<sub>2</sub> capture technology.<sup>3</sup> As shown in Fig. 1, this process is achieved in a dual circulating fluidized bed reactor system, including a carbonator and a calciner. Limestone is firstly calcined in the calciner at around 900 °C and decomposed to CaO and CO<sub>2</sub>. The formed CaO is then delivered to the carbonator to capture the CO<sub>2</sub> in flue gas at around 650–700 °C, at which temperature about 90% CO<sub>2</sub> capture efficiency is achieved. A CO<sub>2</sub> free flue gas is obtained at the outlet of the carbonator after gas–solid separation. Then the separated CaCO<sub>3</sub> formed by carbonation in the carbonator is delivered to the calciner to be calcined again. The energy needed in the calciner is provided by oxy-fuel combustion. Therefore, the CO<sub>2</sub> concentration in the flue gas at the outlet of the calciner can be higher than 95%, at which concentration the CO<sub>2</sub> can be directly sent for storage. A key advantage of CLP over other CO<sub>2</sub> capture technologies is the utilization of natural limestone or

dolomite which have low costs, widespread availability and non-toxicity to the environment.<sup>4</sup> Industrial solid wastes such as carbide slag<sup>5</sup> and lime mud<sup>6</sup> were also proposed as CO<sub>2</sub> sorbents. Therefore, CLP was considered as one of the most promising technologies to reach large scale CO<sub>2</sub> emission reduction from coal-fired power plants.<sup>7</sup> The main limit for CLP is the sharp decrease of the capture capacity of the CO<sub>2</sub> sorbents with increasing the number of the calcination/carbonation cycles.<sup>8</sup> This is mainly due to the sintering occurred in the calcination stage under relatively high temperature,<sup>9</sup> while the carbonation process has little effect on the sintering of the sorbents.<sup>10</sup> In order to approach full calcination of the sorbent under high concentration CO<sub>2</sub>, calcination required significantly higher temperatures (900–1000 °C), resulting in changes in pore shape, pore shrinkage and grain growth.<sup>11</sup> More CO<sub>2</sub> sorbent has to be added in the reactor to guarantee high enough CO<sub>2</sub> capture efficiency, which leads to more serious abrasion.

A lot of efforts have been made to enhance the anti-sintering characteristics and increase the CO<sub>2</sub> capture capacity of the natural sorbents. It has been proved that modification by

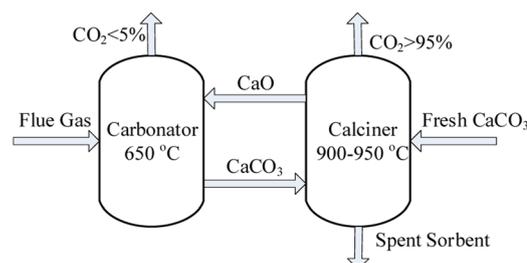


Fig. 1 Flow diagram of calcium looping process.

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organic acid,<sup>12</sup> ball milling,<sup>13</sup> hydration treatment,<sup>14</sup> mixing of a small amount of additives<sup>15</sup> and synthesis of precipitated calcium carbonate<sup>16</sup> can obviously increase the cyclic carbonation conversions during multiple cycles. However, these treatment methods can only optimize the initial microstructure of the calcined sorbent, with little help to the anti-sintering characteristics of the sorbent. Therefore, the treated sorbents still shows relatively fast decay on CO<sub>2</sub> capture capacity. In comparison, inert material supported synthetic sorbents show much better cycle stability. The inert materials such as Mg<sup>17</sup> and Al<sup>18</sup> in the synthetic sorbents can effectively inhibit the growth and fusion of the CaO grain when calcined at high temperature, resulting in stable microstructure during multiple cycles. It is the main mechanism why the synthetic sorbents can achieve higher CO<sub>2</sub> capture capacity. It has been proved that TiO<sub>2</sub>, MgO, La<sub>2</sub>O<sub>3</sub>, ZrO<sub>2</sub>, SiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub> are effective inert materials.<sup>19</sup> Thereinto, MgO has drawn extensive attentions due to its low cost and wide distribution in natural world. The synthesis methods and the MgO precursors have significant effects on the CO<sub>2</sub> capture capacity of the synthetic sorbents. The CaO–MgO sorbent synthesized by mechanical mixing using insoluble MgO precursors shows a little higher cyclic carbonation conversion than natural Ca-based sorbents.<sup>20</sup> When synthesized by sol–gel method using soluble CaO and MgO precursors, the synthetic sorbents show the highest CO<sub>2</sub> capture capacity.<sup>7</sup> However, the synthesis cost is relatively high, limiting its industrialized application. Using Ca(NO<sub>3</sub>)<sub>2</sub> and Mg(NO<sub>3</sub>)<sub>2</sub> as CaO and MgO precursors, Naeem *et al.*<sup>21</sup> yielded highly effective CaO–MgO sorbents *via* a hydrothermal approach. They also found that the presence of a carbonaceous template during synthesis allowed for the formation of multi-shelled microstructures.

Among all the synthesis methods, solution combustion method is a kind of facile one-pot synthesis approach to obtain highly effective synthetic sorbents with stable CO<sub>2</sub> capture capacity.<sup>22</sup> Using carbide slag as the CaO precursor, dolomite as the MgO precursor and trace Mn as the dopant, Ma *et al.*<sup>23</sup> synthesized a novel Mn/Mg-copromoted carbide slag *via* a solution combustion method. The byproduct of biodiesel was used as the solvent and fuel, leading to even lower synthesis cost. The yielded sorbents achieved a CO<sub>2</sub> capture capacity of 0.52 g g<sup>-1</sup> after 10 cycles, which was 82% higher than that of carbide slag. The supplement ratio of CaO in calcium looping process is only 3% mole ratio of CO<sub>2</sub> in the flue gas. Although there is a certain amount CO<sub>2</sub> released during the solution combustion process, compared with the CO<sub>2</sub> amount that can be captured by the calcium based sorbent, the CO<sub>2</sub> amount that released during the synthesis process can be ignored. Also, the CO<sub>2</sub> released during the synthesis process can be delivered into the carbonator of the CLP process to be captured, avoiding the CO<sub>2</sub> emission by synthesis process. By capturing the undesirable CO<sub>2</sub> released from the synthesis process through CLP process, the solution combustion synthesis methods can be eco-friendly when synthesizing the CaO–MgO sorbents. Polyethylene glycol (PEG) is a kind of organic polymer that is widely used as dispersant in chemical industry. For example, as binder for dispersed phases, PEG was often used in propellant formulations since it well maintains viscoelastic behaviour

undergone stretch.<sup>24</sup> In this research, polyethylene glycol was employed as the fuel to synthesize the highly reactive CaO–MgO sorbents by solution combustion method, with the purpose to enhance the dispersion of Mg and Ca in the synthetic sorbents.

## 2. Sorbents and experiments

### 2.1. Sorbents preparation

All the reagents used in this research were analytically pure from Titan Technology Co., Ltd (Shanghai, China). Ca(NO<sub>3</sub>)<sub>2</sub> and Mg(NO<sub>3</sub>)<sub>2</sub> were employed as CaO and MgO precursors. Polyethylene glycol was employed as fuel and dispersant. The molecular weight of the polyethylene glycol used here was 200–1500. The synthesis process of the CaO–MgO sorbent by solution combustion method was shown in Fig. 2. A certain amount of deionized water and polyethylene glycol were firstly added into a beaker, and then the solution was heated to 80 °C. After that, Ca(NO<sub>3</sub>)<sub>2</sub> and Mg(NO<sub>3</sub>)<sub>2</sub> were put into the beaker. The solution was stirred in a magnetic stirrer until all materials were dissolved. Then, the solution was poured into the porcelain boat and sent to the muffle furnace for combustion synthesis. The CaO–MgO sorbents were obtained after calcination at 700–850 °C for 30–120 min. The detailed experimental settings were shown in Table 1. The synthetic CaO–MgO sorbent was sent for XRD analysis to check the compounds. As shown in Fig. 3, the main compounds of the CaO–MgO sorbents were CaO and MgO. No other compound was found through the XRD results. As the contrast sample, the CaO from calcination of CaCO<sub>3</sub> was defined as CaO–C.

### 2.2. Cyclic calcination/carbonation tests

As shown in Fig. 4, the cyclic calcination/carbonation tests of the sorbent were accomplished in a dual fixed bed reactor (DFR), which was detailed elsewhere.<sup>25</sup> The N<sub>2</sub> and CO<sub>2</sub> feed were controlled by mass flow controllers and introduced into the reactor. The sample was firstly calcined for 10 min at 850 °C in pure N<sub>2</sub> and then was carbonated for 10 min at 700 °C in 15% CO<sub>2</sub> (balanced by N<sub>2</sub>). Li *et al.*<sup>26</sup> has proved that the optimum carbonation time for calcium based sorbent is 650–700 °C. Criado *et al.*<sup>27</sup> attributed the lower conversion under lower

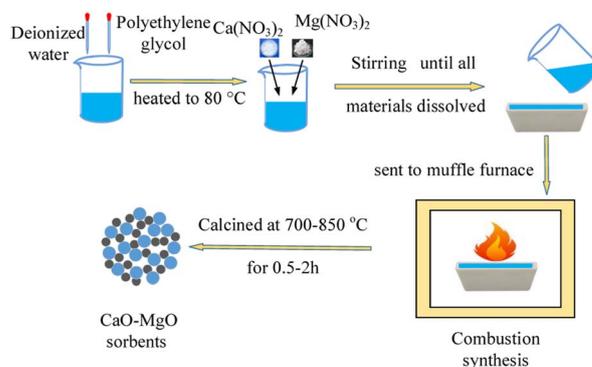


Fig. 2 Synthesis process of the CaO–MgO sorbent by solution combustion method.



Table 1 Details of the experimental settings

No.	Molar ratio of Ca/Mg	Weight of Ca(NO <sub>3</sub> ) <sub>2</sub> ·4H <sub>2</sub> O (g)	Weight of Mg(NO <sub>3</sub> ) <sub>2</sub> ·6H <sub>2</sub> O (g)	Weight of PEG (g)	Weight of distilled water (g)	Molecular weight of PEG (g mol <sup>-1</sup> )	Synthesis temperature (°C)	Synthesis time (min)
1	7 : 3	41.33	19.23	100	100	600	750	60
2	8 : 2	47.23	12.82	100	100	600	750	60
3	9 : 1	53.13	6.41	100	100	600	750	60
4	7 : 3	41.33	19.23	100	100	200	750	60
5	7 : 3	41.33	19.23	100	100	1500	750	60
6	7 : 3	41.33	19.23	100	100	600	700	60
7	7 : 3	41.33	19.23	100	100	600	850	60
8	7 : 3	41.33	19.23	100	100	600	750	30
9	7 : 3	41.33	19.23	100	100	600	750	120

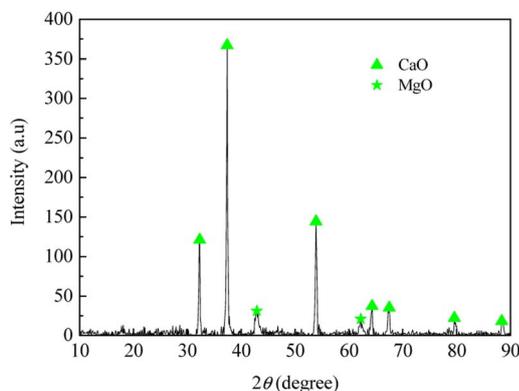


Fig. 3 XRD analysis of the synthetic CaO–MgO sorbent.

carbonation temperature to product layer thickness decreases in accordance with an Arrhenius dependence. Therefore, the carbonation temperature chosen in this manuscript is 700 °C.

Carbonation conversion ( $X_N$ ) and CO<sub>2</sub> capture capacity ( $C_N$ ) were employed to reflect the CO<sub>2</sub> capture performance of the sorbents. Carbonation conversion denotes conversion of CaO in the sorbent to CaCO<sub>3</sub>, mol mol<sup>-1</sup>. CO<sub>2</sub> capture capacity indicates CO<sub>2</sub> adsorption amount per unit mass of sample, g(CO<sub>2</sub>)/g(sorbent). The sample mass after calcination and carbonation were measured by an electronic balance, and the  $X_N$  and  $C_N$  were calculated according to the mass change during the carbonation and calcination stage, as shown in equal (1) and (2).

$$X_N = \frac{m_N - m_{\text{cal}}}{m_0 a} \cdot \frac{M_{\text{CaO}}}{M_{\text{CO}_2}} \quad (1)$$

$$C_N = \frac{m_N - m_{\text{cal}}}{m_{\text{cal}}} \quad (2)$$

where  $m_0$  is the initial mass of the sample, mg;  $m_N$  is the mass of the carbonated sample after  $N$  cycles, mg;  $m_{\text{cal}}$  is the mass of the completely calcined sample (the mass of the sample after each calcination is the same), mg;  $M_{\text{CaO}}$  and  $M_{\text{CO}_2}$  are molar masses of the CaO and CO<sub>2</sub> respectively, g mol<sup>-1</sup>;  $a$  is the content of CaO in the initial sample, %.

### 2.3. Microstructure analysis

The samples of CaO–C and CaO–MgO sorbent were collected and sent for SEM and EDS analysis (Hitachi, Regulus8100), to determine the micro morphology and the distribution of element. The pore volume and pore area distributions of the CaO–C and CaO–MgO sorbents after different cycles were examined by a nitrogen adsorption analyzer (Micromeritics, ASAP 2020-M). The pore volume and pore size distribution of the sample were computed by BJH model, the BET surface of the sample was calculated by BET model.

## 3. Results and discussions

### 3.1. Effect of the MgO content on CO<sub>2</sub> capture capacity

With the Ca/Mg molar ratio ranging from 9 : 1 to 7 : 3, the MgO-stabilized CaO sorbents were synthesized by solution combustion method. The solution combustion synthesis temperature and time were 750 °C and 60 min and the molecular weight of the polyethylene glycol is 600 in this case. The DFR test results were shown in Fig. 5. It can be seen that the MgO-stabilized CaO sorbents shows much higher and more stable CO<sub>2</sub> capture capacity compared with that of the CaO–C. In the 20th cycle, the carbonation conversions of the MgO-stabilized CaO sorbents were 0.57, 0.53 and 0.42 when the Ca/Mg molar ratio were 7 : 3, 8 : 2 and 9 : 1, while the value for CaO–C was only 0.29. Take the MgO-stabilized CaO sorbent with Ca/Mg molar ratio is 7 : 3 as example, the SEM and EDS analysis were conducted to determine the element distribution on the surface of the sorbent, as shown in Fig. 6. The SEM analysis result shown in Fig. 6(a) was employed to calculate the EDS spectrum and the result was

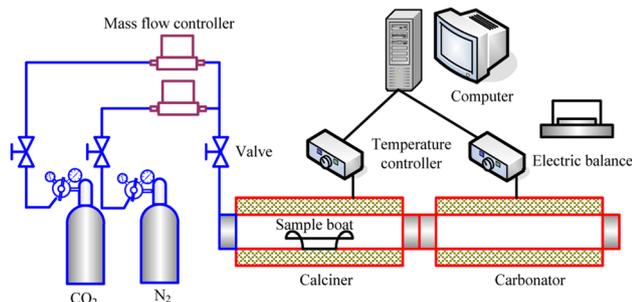


Fig. 4 Schematic diagram of dual fixed-bed reactor (DFR).



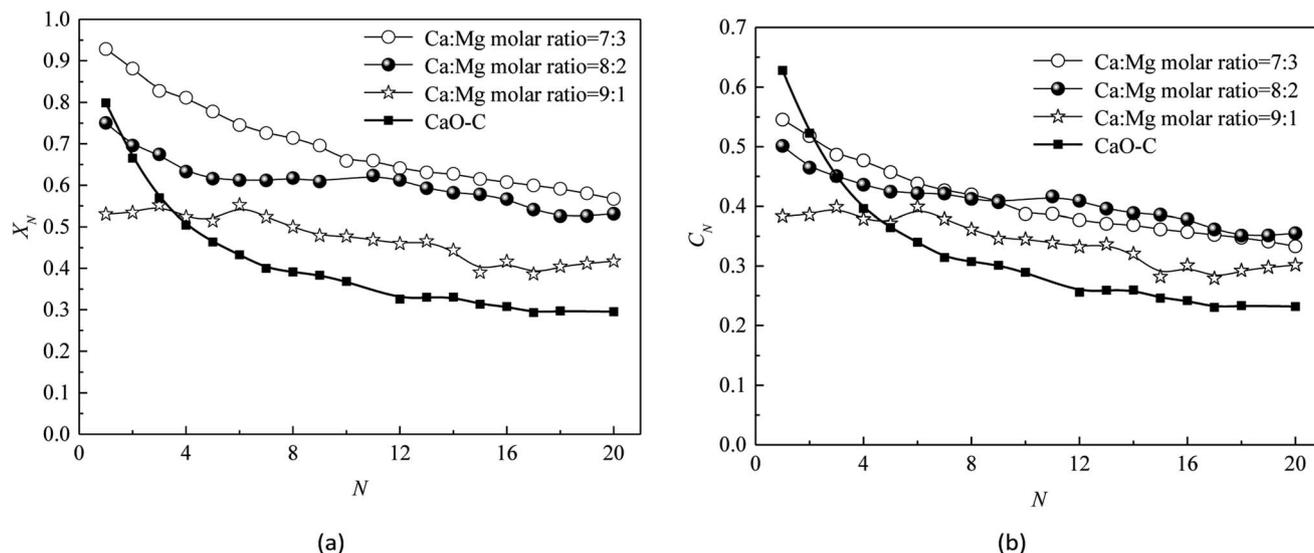


Fig. 5 Effect of the MgO content on the (a) carbonation conversion and (b)  $\text{CO}_2$  capture capacity of the MgO-stabilized CaO sorbent.

shown in Fig. 6(d). According to the EDS spectrum, the Ca/Mg molar ratio on the surface of the sorbent was calculated as 6.95 : 2.97, which was agree with the synthesis process. It proved that the EDS result was accurate. The Ca and Mg distributions on the sorbent surface were shown in Fig. 6(b) and (c). It revealed a homogeneously evenly distribution of Ca and Mg on the surface of the sorbent. As shown in Fig. 7, the calcined MgO-stabilized CaO sorbents showed more porous microstructure compared with the CaO-C during the 1<sup>st</sup> cycle. The existence of MgO can effectively restrain the sintering of the sorbent. As shown in Fig. 8, the calcined MgO-stabilized CaO sorbents can

maintain effective enough pore structure for  $\text{CO}_2$  diffusion in the sorbent, while CaO-C occurred obvious sintering after 20 cycles. Thus, the MgO-stabilized CaO sorbents showed higher and more stable  $\text{CO}_2$  capture capacity compared with CaO-C.

We can find from Fig. 5(a) that the carbonation conversion of the MgO-stabilized CaO sorbents increased with increasing the MgO content in the sorbents. In Fig. 7 and 8, the MgO-stabilized CaO sorbents with higher MgO content also demonstrated more porous microstructure. However, it doesn't mean that the more the MgO content, the better the  $\text{CO}_2$  capture capacity. As the inert support materials in the sorbent, increasing the MgO

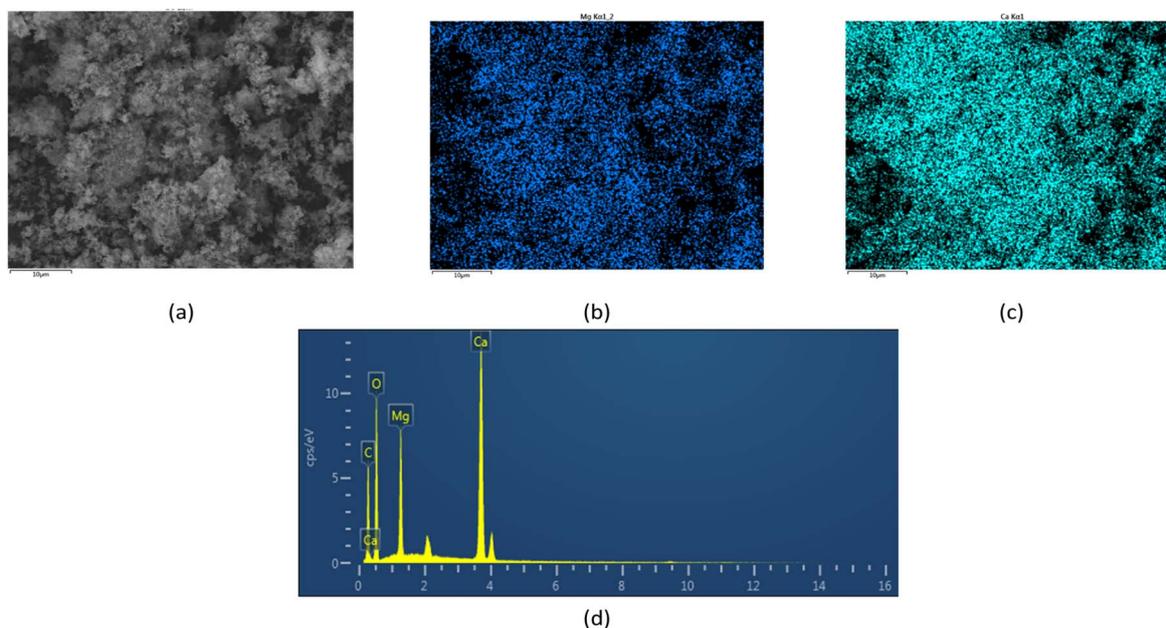


Fig. 6 SEM and EDS analysis of the calcined MgO-stabilized CaO sorbent with the Ca/Mg molar ratio is 7 : 3 (a) SEM imagine; (b) Mg distribution; (c) Ca distribution; (d) EDS spectra.



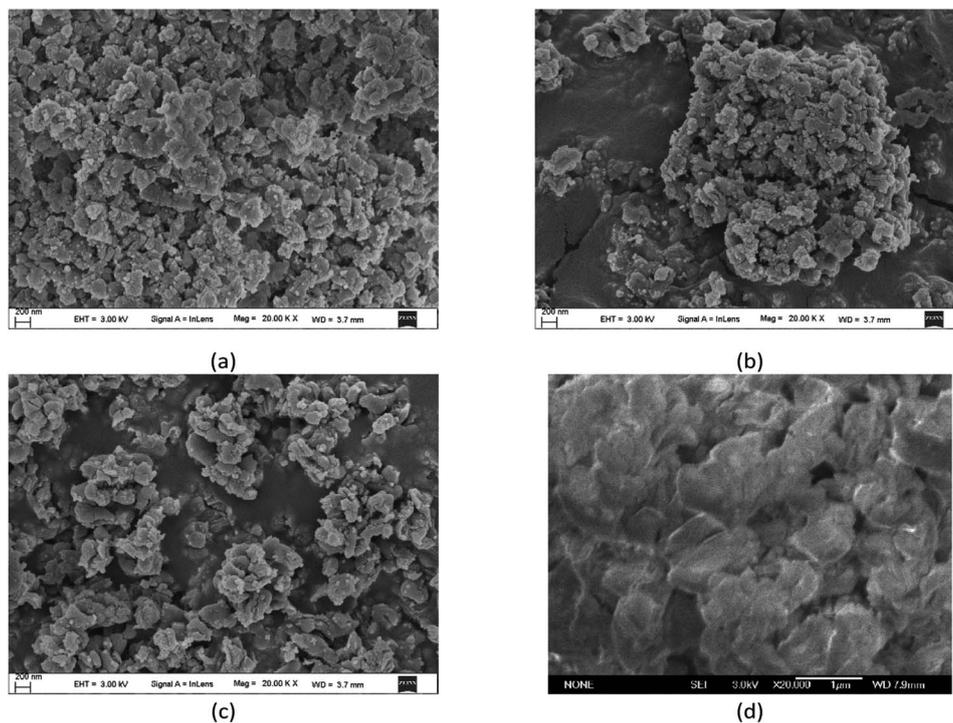


Fig. 7 SEM analysis of the calcined (a–c) MgO-stabilized CaO sorbent and (d) CaO–C in the 1st cycle: (a) Ca/Mg molar ratio is 7 : 3; (b) Ca/Mg molar ratio is 8 : 2; (c) Ca/Mg molar ratio is 9 : 1.

content means the active constituent content, *i.e.* CaO, will be less, which will decrease the CO<sub>2</sub> capture capacity of the sorbent. As shown in Fig. 5(b), also the carbonation conversions

were lower, the MgO-stabilized CaO sorbent with Ca/Mg molar ratio is 8 : 2 represents higher CO<sub>2</sub> capture capacity from the 10<sup>th</sup> cycle. Therefore, the optimum Ca/Mg molar ratio is 8 : 2.

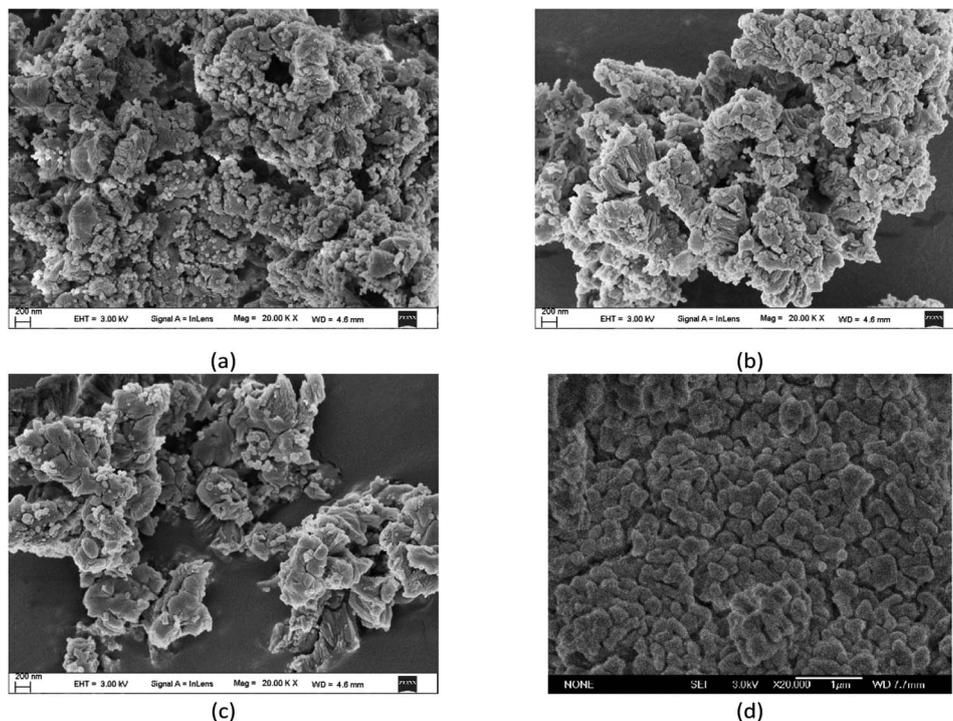


Fig. 8 SEM analysis of the calcined (a–c) MgO-stabilized CaO sorbent and (d) CaO–C after 20 cycles: (a) Ca/Mg molar ratio is 7 : 3; (b) Ca/Mg molar ratio is 8 : 2; (c) Ca/Mg molar ratio is 9 : 1.



### 3.2. Effect of molecular weight of the polyethylene glycol on CO<sub>2</sub> capture capacity

The molecular weight of the polyethylene glycol (PEG) differs from 200 to tens of thousands, according to its extent of polymerization. In view of the fact that the polyethylene glycol exists in solid state under 50 °C when the molecular weight is higher than 2000, the molecular weights of the polyethylene glycol used in this research were chosen as 200 (PEG 200), 600 (PEG 600) and 1500 (PEG 1500). The effect of molecular weight of the polyethylene glycol on CO<sub>2</sub> capture capacity of the MgO-stabilized CaO sorbent was shown in Fig. 9. It is obvious that the MgO-stabilized CaO showed the best carbonation conversion and CO<sub>2</sub> capture capacity when synthesized using PEG 600 as fuel and dispersant. Firstly, increasing the molecular weight can enhance the CO<sub>2</sub> capture capacity of the MgO-stabilized CaO sorbent. When the molar weights of PEG is higher than 600, increasing the molecular weight of PEG reduces the CO<sub>2</sub> capture capacity of the MgO-stabilized CaO sorbent on the contrary. As is shown in Fig. 10, the molecular weight of PEG can dramatically affect the Ca and Mg uniformity coefficient when synthesized by solution combustion method in the furnace. It is obvious that the Ca and Mg distributed much more uniformly on the surface of the MgO-stabilized CaO when

synthesized by PEG 600. As shown in Fig. 10(a) and (c), when synthesized by PEG 200 and PEG 1500, the Mg presents aggregated distribution in local. Some place such as signed by red cycles in Fig. 10(a) and (c), there was less Mg distributed there. The CaO distributed in these local parts will be evenly sintered during multiple calcination/carbonation cycles. Thus, lower CO<sub>2</sub> capture capacity is presented. To sum up, PEG 600 is the optimum fuel and dispersant for MgO-stabilized CaO sorbent.

### 3.3. Effect of synthesis temperature on CO<sub>2</sub> capture capacity

The synthesis temperature was chosen as 700 °C, 750 °C and 850 °C in this research. The effect of synthesis temperature on CO<sub>2</sub> capture capacity of the MgO-stabilized CaO sorbent was shown in Fig. 11. The MgO-stabilized CaO sorbent showed the highest CO<sub>2</sub> capture capacity when the synthesis temperature was 750 °C. In the 1<sup>st</sup> cycle, when synthesized at 750 °C, the carbonation conversion of the MgO-stabilized CaO sorbents was 0.75, which was equal to the sorbent synthesized at 700 °C and 28.9% higher than that synthesized at 850 °C. After 20 cycles, when synthesized at 750 °C, the carbonation conversion of the MgO-stabilized CaO sorbents was 0.53, which was 18.7% and 12.2% higher than that synthesized at 700 °C and 850 °C. It has been proved that synthesizing under high temperature is

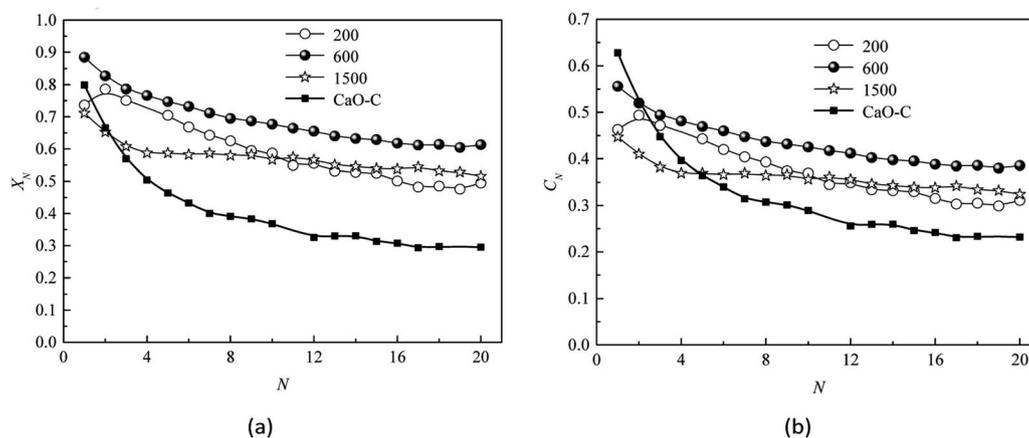


Fig. 9 Effect of molecular weight of the polyethylene glycol on (a) carbonation conversion and (b) CO<sub>2</sub> capture capacity of the MgO-stabilized CaO sorbent.

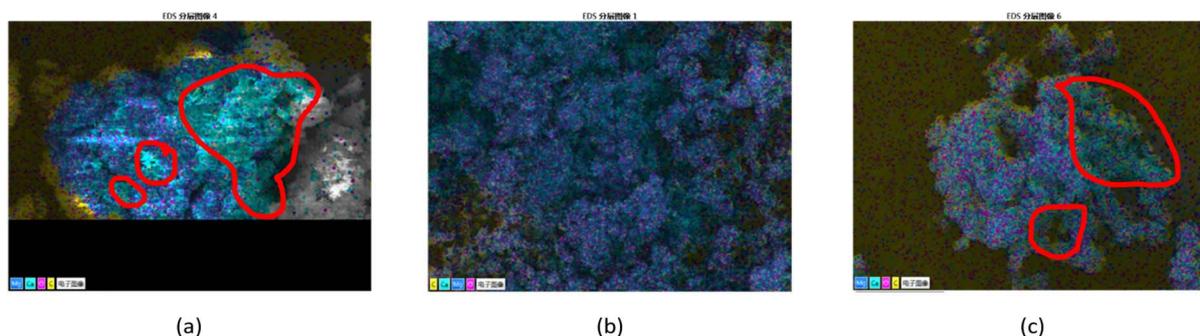


Fig. 10 SEM and EDS analysis of the calcined MgO-stabilized CaO sorbent synthesized using PEG with different molecular weight: (a) PEG 200; (b) PEG 600; (c) PEG 1500.



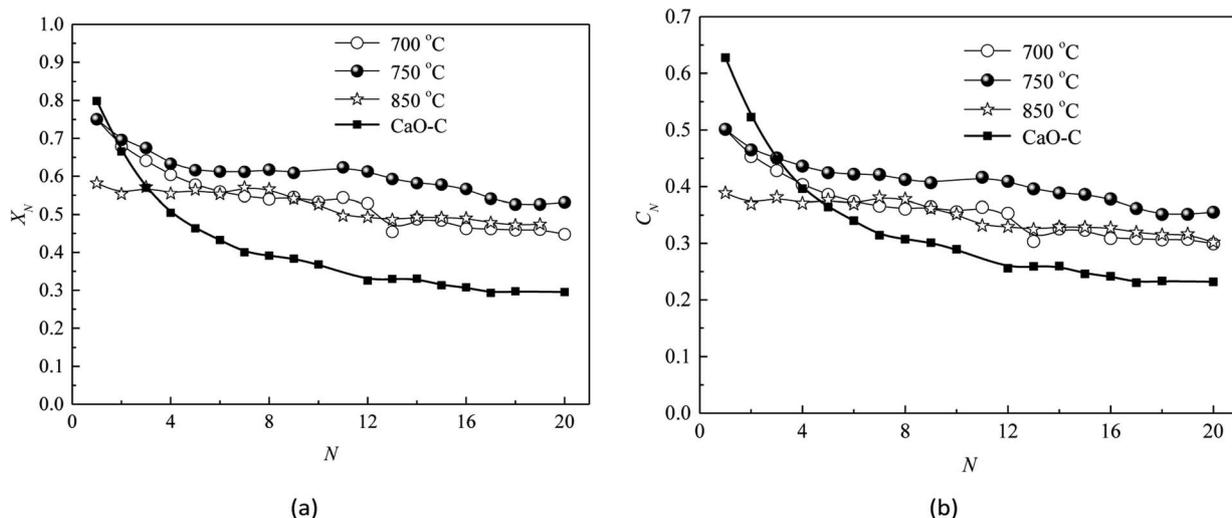


Fig. 11 Effect of synthesis temperature on (a) carbonation conversion and (b) CO<sub>2</sub> capture capacity of MgO-stabilized CaO sorbent.

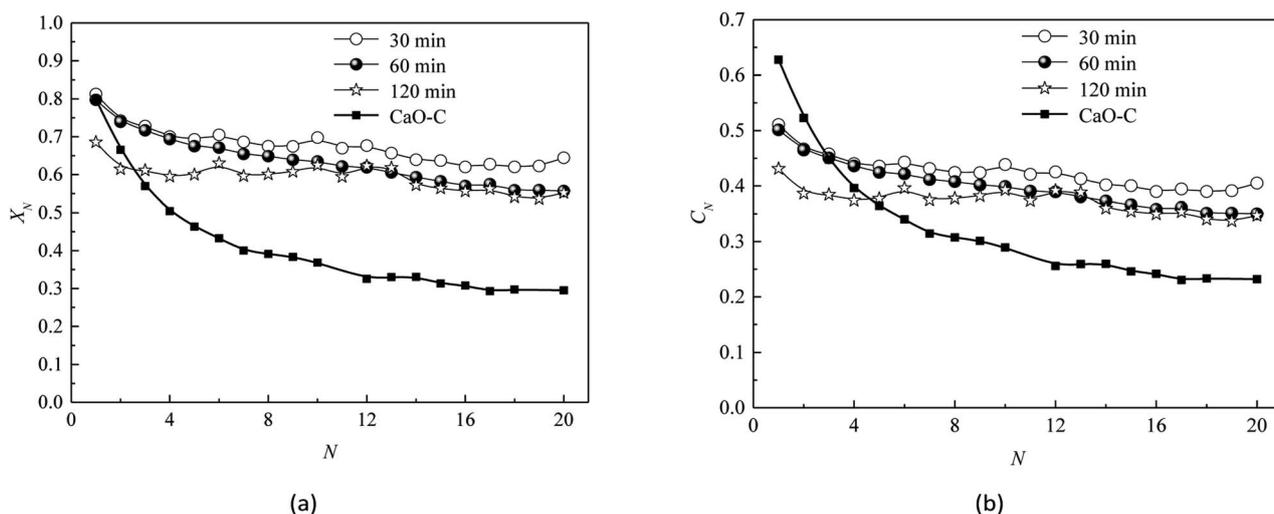


Fig. 12 Effect of the Synthesis time on (a) carbonation conversion and (b) CO<sub>2</sub> capture capacity of MgO-stabilized CaO sorbent.

helpful for the sorbent to form more stable microstructure during multiple calcination/carbonation cycles. So, initially increasing the synthesis temperature can enhance the microstructure of the sorbent. However, if the synthesis temperature is higher than the optimum temperature, severe pre-sintering will occur during the synthetic process, decreasing the CO<sub>2</sub> capture capacity of the sorbent. To sum up, the optimum synthesis temperature is 750 °C.

### 3.4. Effect of synthesis time on CO<sub>2</sub> capture capacity

The effect of synthesis time on CO<sub>2</sub> capture capacity of the MgO-stabilized CaO sorbent was shown in Fig. 12. If the synthesis time is too short, the combustible material in the solution can not burn out during the synthesis process. It need about 30 min for the PEG in the solution to finish its combustion. Go on lengthening the synthesis time will lead more severe pre-sintering of the sorbent, which is adverse for the CO<sub>2</sub> capture.

The experimental test results in the DFR also proved this conclusion. The CO<sub>2</sub> capture capacity of the MgO-stabilized CaO sorbent slightly decreased when the synthesis time lengthened from 30 min to 120 min. After 20 cycles, the CO<sub>2</sub> capture capacity of the MgO-stabilized CaO sorbent reduced by 14.4% when the synthesis time lengthened from 30 min to 120 min. Therefore, 30 min is the optimum synthesis time when synthesizing the MgO-stabilized CaO sorbent.

## 4. Conclusions

In this research, using polyethylene glycol as the fuel and dispersant, solution combustion method was employed to synthesize MgO-stabilized CaO sorbents by soluble MgO and CaO precursors. The main conclusions are presented as follows:

(1) A kind of highly reactive MgO-stabilized CaO sorbents was obtained through solution combustion method using PEG



as fuel and dispersant. The Ca and Mg distributed homogeneously on the surface of the MgO-stabilized CaO sorbents. The existence of MgO can effectively restrain the sintering of the sorbent, resulting in a more porous and stable microstructure of the sorbent. The microstructure is benefit to the CO<sub>2</sub> diffusion and the carbonation reaction. That's why the MgO-stabilized CaO sorbents can achieve so high CO<sub>2</sub> capture capacity.

(2) The synthetic conditions including Ca/Mg molar ratio, molecular weight of PEG, synthesis time and synthesis temperature have apparent effects on the CO<sub>2</sub> capture capacity of the MgO-stabilized CaO sorbents. The optimum synthetic conditions obtained this research: the optimum Ca/Mg molar ratio is 8 : 2; the optimum molecular weight of PEG is 600; the optimum synthesis temperature and time are 750 °C and 30 min respectively. The CO<sub>2</sub> capture capacity of the MgO-stabilized CaO sorbent synthesized under the optimum conditions is 0.40 g(CO<sub>2</sub>)/g(sorbent) after 20 cycles, which is 75.3% higher than CaO-C.

## Conflicts of interest

There are no conflicts to declare.

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