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Synthesis, Experimental Studies, and Analysis of a New Calcium-Based Carbon Dioxide Absorbent

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Received December 10, 2004. Revised Manuscript Received March 23, 2005

A new kind of Ca-based regenerable CO₂ absorbent, CaO/Ca₁₂Al₁₄O₃₃, was synthesized on the basis of the integration of CaO, as solid reactant, with a composite metal oxide Ca₁₂Al₁₄O₃₃, as a binder, for applying it to repeated calcination/carbonation cycles. The carbonation reaction can be applied in many industrial processes, and it is important for practical calcination/carbonation processes to have absorbents with high performance. The cyclic carbonation reactivity of the new absorbent was investigated by TGA (thermogravimetric analysis). The effects of the ratio of active material to binder in the new absorbent, the mechanics for preparation, and the reaction process of the high-reactivity CaO/Ca₁₂Al₁₄O₃₃ absorbent have been analyzed. The results obtained here indicate that the new absorbent, CaO/Ca₁₂Al₁₄O₃₃, has a significantly improved CO₂ absorption capacity and cyclic reaction stability compared with other Ca-based CO₂ absorbents. These results suggest that this new absorbent is promising in the application of calcination/carbonation reactions.

1. Introduction

The carbonation reaction of CaO with CO₂ is very important in many industrial processes such as CO₂ absorption gasification,¹ hydrogen production from integrated coal gasification,^{2–4} methane steam reforming process,^{5,6} CO₂ separation from flue gas⁷ or syngas,⁸ and chemical heat pump⁹ and energy storage system.¹⁰ The carbonation process is a gas–solid reaction, and Ca-based absorbents are repeatedly used; in other words, after the carbonation reaction, the absorbents must be regenerated in the subsequent cyclic reaction. Therefore, the carbonation/calcination characteristics of absorbents are very important for practical application. The most common Ca-based absorbents suitable for the carbonation/calcination process are either calcium-containing natural materials, such as dolomite, or prepared materials of calcium oxide deposited on a substrate. Some absorbents, such as limestone, undergo a decay of CO₂ absorption capacity and physical breakdown.^{11,12} High-

performance absorbents should have not only a large calcium oxide content but also a high cyclic stability.

Through the experiments on 10-μm CaO powder, Barker¹¹ demonstrated a drop in the absorption capacity from 59 wt % in the first carbonation cycle to 8 wt % at the end of the 25th cycle, and suggested that, because of the formation of a 22-nm-thick product layer, particles smaller than 22 nm in diameter should be able to achieve stoichiometric conversion. Barker¹³ later proved this hypothesis by obtaining repeated 93% conversion of 10-nm CaO particles over 30 cycles with a carbonation time of 24 h under 100% CO₂ at 850 K. Harrison and co-workers developed an absorption-enhanced hydrogen production process from the steam methane reforming reaction by removing CO₂ from the gas mixture through the carbonation of CaO, but the absorption capacity of absorbents decreased rapidly with the increase of cyclic carbonation number.^{14,15} Abanades and Alvarez,¹⁶ on the basis of the changing microporosity within grains and the mesoporosity surrounding them due to sintering, hypothesized the explanation for the drop in capture capacity over multiple carbonation/calcination cycles. In the application of Ca-based chemical heat pump process, CaO showed a drop in CO₂ capture from 53 wt % in the first cycle to 27.5 wt % by the fifth cycle.¹⁷ A lithium zirconate (Li₂ZrO₃)-based sorbent provided 20 wt % capacity over two cycles.¹⁸ In another study, researchers at Toshiba Corp. (New York) observed that the reactivity of lithium orthosilicate (Li₄SiO₄) was better than that

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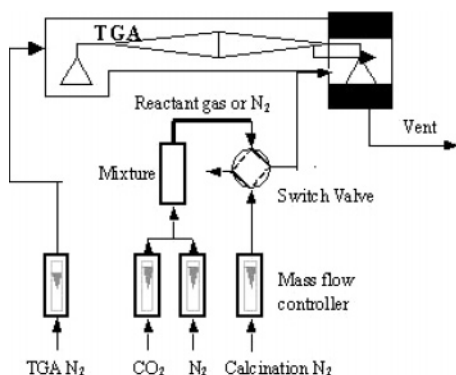


Figure 1. Simplified flow diagram of the experimental setup used for the multiple carbonation and calcination reactions.

of lithium zirconate.^{19,20} Extended cyclical studies performed on lithium orthosilicate samples attained 26.5 wt % sorption capacity over 50 cycles without any change in the reactivity.²¹

In this study, a new Ca-based absorbent material has been synthesized on the basis of the integration of a new binder, $\text{Ca}_{12}\text{Al}_{14}\text{O}_{33}$. It has high reactivity and stability under the conditions of the multiple carbonation/calcination cycles. Furthermore, we have focused on several other kinds of natural Ca-based absorbents by studying the decay of the CO_2 absorption capacity and the effect of the inert MgO in dolomite on the cyclic stability during carbonation/calcination cycles. Finally, the comparison of the CO_2 absorption capacity and the cyclic stability for these absorbents are given.

2. Experiments

2.1. Experimental System. A Dupont 951 TGA (TA Instrument 1200) was used to study the carbonation reaction of Ca-based CO_2 absorbents. The TGA consists of a quartz tube placed in an oven which can be operated at temperatures up to 1473 K. A computer continuously records reaction temperature and sample weight. The reacting gas contains CO_2 and N_2 , and their proportion can be adjusted by mass flow controllers. The simplified TGA schematic diagram is shown in Figure 1.

A small amount of the absorbent (about <15 mg) was placed in a quartz sample holder and brought to 1173 K under nitrogen flow for calcination. After complete calcination, the temperature was decreased at a rate of 50 °C/min to the carbonation temperature. When approaching the carbonation temperature, the valve was switched to allow the gas mixture of reactant to flow over the calcined absorbent. A manual mated three-way valve was used to switch between the pure nitrogen stream and the reaction gas mixture at certain carbonation and calcination time intervals. The time for carbonation and calcination is 30 and 10 min, respectively. The microstructure of carbonated samples was investigated by scanning electron microscopy (SEM, KYKY2000). The phase composition of the sample was determined by X-ray diffraction (XRD, D8, Advance, BRUKER).

2.2. Experimental Samples. Experimental samples consist of pure CaCO_3 (20 μm), and three dolomites. The compositions of the three dolomites are shown in Table 1.

Table 1. Chemical Composition of Three Dolomites

sample	chemical composition (molar ratio)						particle diameter (μm)
	CaO	MgO	Fe_2O_3	SiO_2	Al_2O_3	other	
dolomite I	1	0.66	0.05	0.03	0.01	0.005	30
dolomite II	1	0.63	0.04	0.04	0.02	0.004	25
dolomite III	1	2.37	0.05	0.03	0.02	0.005	32

2.3. Preparation of High-Reactivity Calcium Oxide. In experiment, the novel samples are from two primary materials, that is, CaO and CaCO_3 . The detailed preparation procedures of the novel absorbent are as follows. Pure CaO (about 20 μm) is calcined in air at 1173 K for 2 h. Aluminum nitrate enneahydrate ($\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$) (28.4 g) and the powdered calcium oxide (7.7 g or 26.2 g) were added into a mixture of 2-propanol (65 mL or 130 mL) and distilled water (760 mL) so that the weight ratios of calcium oxide to newly formed materials ($\text{Ca}_{12}\text{Al}_{14}\text{O}_{33}$) would become 35:65 wt % or 75:25 wt %. This solution was stirred for 1 h at 348 K and was dried at 393 K for another 18 h before it was calcined at 773 K for 3 h in air. By this method, 2-propanol, water, and nitric acid in the solution can be evaporated off at different stages, thereby offering production of fine and porous powder. Spherical particles were made from the paste obtained by adding water to the resultant powder. These particles were dried at 393 K for 2 h and calcined in air at 1173 K for 1.5 h. The mixed powder was ground to a certain size (10 μm). The procedures for particles coming from CaCO_3 are similar to those described in the above method.

It is found from the results of XRD analysis that the new absorbent includes only CaO and $\text{Ca}_{12}\text{Al}_{14}\text{O}_{33}$, and it is assumed that all Al_2O_3 coming from $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ reacts completely with CaO to form $\text{Ca}_{12}\text{Al}_{14}\text{O}_{33}$. Thus, the mass ratio of CaO to $\text{Ca}_{12}\text{Al}_{14}\text{O}_{33}$ in the new absorbents can be calculated according to the original mass of CaO and $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$.

2.4. Experimental Data Analysis Method. To comprehensively analyze the characters of absorbents, we have defined the cyclic stability of absorbent, $C_s(n)$, as the ratio of the ultimate absorbed CO_2 mass amount $m_{n,\text{CO}_2 \text{ abs}}$ (g) for the certain cycle n to the maximum CO_2 mass amount absorbed $m_{\text{max},\text{CO}_2 \text{ abs}}$ (g) in all cycles:

$$C_s(n) = m_{n,\text{CO}_2 \text{ abs}} / m_{\text{max},\text{CO}_2 \text{ abs}} \text{ (g/g)}$$

The cyclic absorption capacity, $C_c(n,t)$, has been defined as the ratio of the absorbed CO_2 mass amount at certain reaction time t in the certain cycle n , $m_{n,\text{CO}_2 \text{ abs}}(t)$ (g), to the initial mass of the absorbent before every cyclic carbonation reaction, m_{initial} (g):

$$C_c(n,t) = m_{n,\text{CO}_2 \text{ abs}}(t) / m_{\text{initial}} \text{ (g-}\text{CO}_2\text{/g-absorbent)}$$

Theoretically, 56 g of unsupported CaO absorbent should react with 44 g of CO_2 corresponding to a maximum CO_2 absorption capacity of 0.786 g- CO_2 /g-CaO at 100% CaO conversion, that is to say, the maximum value of $C_c(n,t)$ is 0.786 g- CO_2 /g-absorbent for all Ca-based absorbents, and the theoretical maximum CO_2 absorption capacity of $\text{CaO}/\text{Ca}_{12}\text{Al}_{14}\text{O}_{33}$ (75/25 wt %) absorbent will be $0.786(0.75) = 0.589$ g- CO_2 /g-absorbent.

The carbonation and calcination temperatures of absorbents are also important parameters for practical application. For example, in the process of steam methane reforming (SMR) reactions with the addition of CO_2 absorbent, the carbonation temperature of absorbent must be in the range of the reforming temperature, otherwise the CO_2 from the SMR reaction cannot be removed from the reacting zone. Because of cyclic use, the absorbent must be regenerated by calcination after carbonation, and the temperature of calcination has important effects on many aspects of reforming process. The disadvantages of an adsorbent with a higher calcination temperature are the

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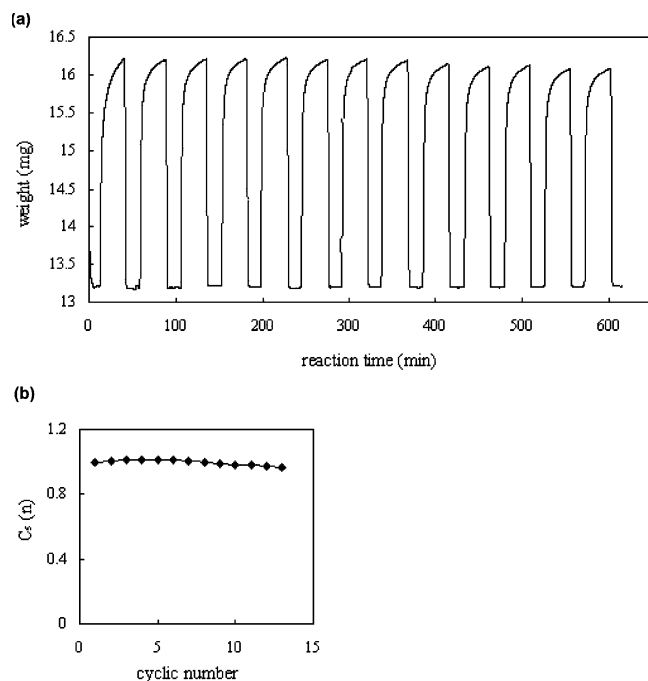


Figure 2. (a) Calcination/carbonation cycles of $\text{CaO}/\text{Ca}_{12}\text{Al}_{14}\text{O}_{33}$ (35/65 wt %) in a TGA with 14% CO_2 stream; (b) cyclic stability of $\text{CaO}/\text{Ca}_{12}\text{Al}_{14}\text{O}_{33}$ (35/65 wt %).

increased energy demand for the regeneration and strict material requirement for the reformer, which result in system efficiency decrease and cost increase. Therefore, the calcination temperature of absorbent cannot be high. In this study, all absorbents were allowed to react 30 min for carbonation (963 K, 14% CO_2) and 10 min for calcination (1123 K, 100% N_2).

3. Results and Discussion

3.1. Cyclic Carbonation/Calcination Characteristics of the High-Reactivity Calcium Oxide. Some researchers reported^{11,16} that, for the carbonation reaction, after a rapid chemically controlled initial period, a much slower product layer stage diffusion follows. The transition between the fast and slow reaction stages was attributed to the formation of a CaCO_3 product layer, which limits the carbonation conversions. Abanades et al.¹⁶ developed a simple correlation to explain the fast drop in the absorption capacity of absorbent tested under a wide range of conditions used by other authors. To find an appropriate high-temperature CO_2 absorbent, we prepared two kinds of calcium-based CO_2 absorbents, using $\text{Ca}_{12}\text{Al}_{14}\text{O}_{33}$ as the binder. Its cyclic reaction characteristics are observed and recorded as shown in Figure 2. It can be seen from Figure 2a that the cyclic reaction capacity of $\text{CaO}/\text{Ca}_{12}\text{Al}_{14}\text{O}_{33}$ (35/65 wt %) with CO_2 is very stable and changed little with the increase of cyclic numbers. The reaction kinetics of $\text{CaO}/\text{Ca}_{12}\text{Al}_{14}\text{O}_{33}$ (35/65 wt %) with CO_2 is depicted in Figure 2a, being similar to the patterns of other calcium-based absorbents. The surface chemical reaction with fast reaction rate dominates the beginning stage and is followed by a rather slow product layer diffusion stage. Particularly, the reaction path of no significant changes along with the cyclic numbers suggests that the sample is a promising CO_2 absorbent. It can be seen from Figure 2b that the cyclic stability of $\text{CaO}/\text{Ca}_{12}\text{Al}_{14}\text{O}_{33}$ (35/65 wt %) is superior and almost remains at the same level even in the 13th cyclic reaction process.

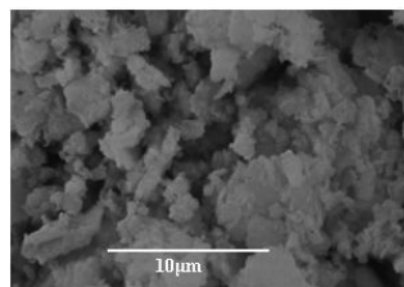


Figure 3. SEM image of $\text{CaO}/\text{Ca}_{12}\text{Al}_{14}\text{O}_{33}$ (75/25 wt %) after the first carbonation (carbonation at 963 K with 14% CO_2 , calcination at 1123 K with 100% N_2).

After the reaction of CaO with CO_2 , the reaction product CaCO_3 must undergo a calcination process in order to regenerate CaO to be used repeatedly. In the process of calcination, some pores are produced inside the CaO particle. At the same time, CaO sintering may occur during high-temperature calcination. The decline of CO_2 absorption capacity and cyclic stability with the increasing carbonation/calcination cycles can be attributed to changes in the particle structure during calcination. When small CaO particles are heated to certain temperatures high enough yet below the melting point, CaO coalesces and sinters, and the surface of nascent CaO decreases with the increase of residence time. Surface and porosity are very important for the reaction of CaO with CO_2 , but sintering reduces surface area and porosity sharply, which in turn affects the reaction rates and conversion of CaO with CO_2 .^{22,23} Abanades et al.¹⁶ attributed the decay of CaO conversion to a certain loss of small pores and an increase of large pores. They carried out observations with SEM, which revealed that the pore distribution of the calcines was continuously changing with cyclic numbers; more and bigger macro-pores were being formed under the condition that micropores decreased. The reason for high cyclic stability of $\text{CaO}/\text{Ca}_{12}\text{Al}_{14}\text{O}_{33}$ may be that, via introducing binder and the CaO hydration procedures during its preparation stage, the particles become ultra-fined. In preparation calcination stage, the $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ and CaO form $\text{Ca}_{12}\text{Al}_{14}\text{O}_{33}$, a more stable material which is distributed uniformly among CaO crystallites and does not take part in reaction. Hence, the sintering of CaO particles in the calcination stage is efficiently prevented and results in the high CaO cyclic stability. By the image of SEM (Figure 3 and Figure 4), it can be found that, with the increase of cyclic number, the conglomeration of CaO particles does not occur. The phenomena of particle growing and pore space decreasing have not been found when comparing the microgranular appearance of particles after the 11th reaction (Figure 4) with the microgranular appearance of particles after the 1st reaction (Figure 3). Abanades et al.¹⁶ found that their CaO sintered severely and the particles become much more compact with the increase of cyclic number, but such phenomena have not been observed in $\text{CaO}/\text{Ca}_{12}\text{Al}_{14}\text{O}_{33}$ (75/25 wt %) samples.

3.2. Effects of Different Binder Contents on Cyclic Reaction Characteristics. $\text{CaO}/\text{Ca}_{12}\text{Al}_{14}\text{O}_{33}$ (35/65 wt %) has been proved to have high cyclic

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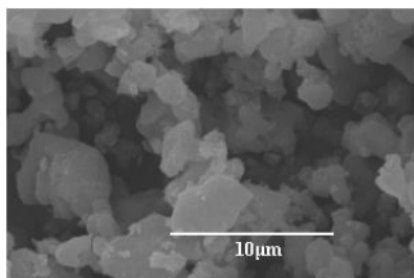


Figure 4. SEM image of CaO/Ca₁₂Al₁₄O₃₃ (75/25 wt %) after the eleven carbonation (carbonation at 963 K with 14% CO₂, calcination at 1123 K with 100% N₂).

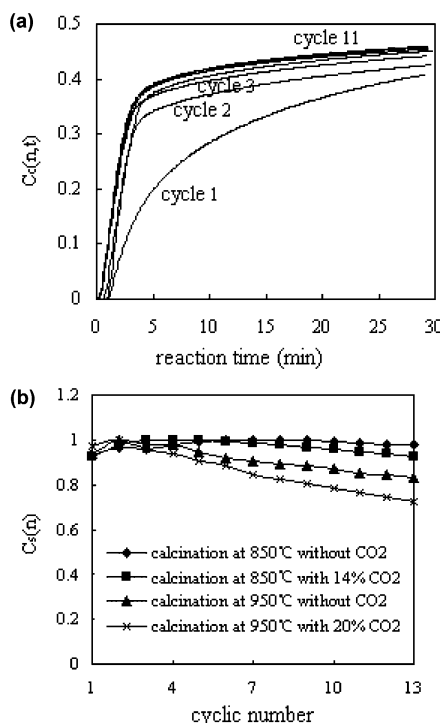


Figure 5. (a) Cyclic absorption capacity of CaO/Ca₁₂Al₁₄O₃₃ (75/25 wt %) (carbonation at 963 K with 14% CO₂, calcination at 1123 K with 100% N₂); (b) cyclic stability of CaO/Ca₁₂Al₁₄O₃₃ (75/25 wt %) at different calcination conditions.

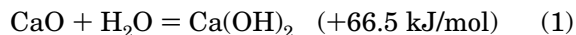
stability; however, its absorption capacity is limited because of its low CaO content. To study the effect of binder content on cyclic reaction characteristics, we have increased the content of CaO to 75 wt %. The experimental results are shown in Figure 5. The shape of the carbonation curve (Figure 5a) shows that the reaction of CaO/Ca₁₂Al₁₄O₃₃ (75/25 wt %) with CO₂ can also be divided into two different stages. The first stage is a rapid, chemically controlled initial reaction period that is followed by a much slower product diffusion stage. Because of the smaller porosity inside particles, the overall reaction rate is controlled by intraparticle diffusion and the cyclic stability and cyclic absorption capacity increase with cycle numbers for the first three cycles. From the fourth cycle onward, the cyclic stability is stabilized. In comparison with the CaO/Ca₁₂Al₁₄O₃₃ (35/65 wt %), with the decrease of binder content, the time of chemical reaction kinetic control stage for CaO/Ca₁₂Al₁₄O₃₃ (75/25 wt %) increases a little, but it also can be found from Figure 5a that the CO₂ absorption capacity of CaO/Ca₁₂Al₁₄O₃₃ (75/25 wt %) is greatly enhanced. More importantly, it can be seen from Figure 5b that the cyclic stability of absorbent does not decline

sharply with the increase of binder content for calcination at 850 °C without CO₂.

However, with an increase of CO₂ concentration and temperature in calcination processes, the cyclic stability of CaO/Ca₁₂Al₁₄O₃₃ (75/25 wt %) begin to decline. The sintering rate of CaO at a given temperature can be accelerated by the presence of CO₂ in the sintering atmosphere.²³ Values for cyclic stability of CaO/Ca₁₂Al₁₄O₃₃ (75/25 wt %) at different calcination conditions are compared in Figure 5b. The cyclic stability of calcination at 850 °C with 14% CO₂ declines a little compared with that of calcination at 850 °C without CO₂. However, when calcination proceeded at 950 °C, the cyclic stability of CaO/Ca₁₂Al₁₄O₃₃ (75/25 wt %) in the 13th cycle carbonation declines, respectively, to 0.85 and 0.74 without and with CO₂ in calcination processes. The temperature of absorbent calcination, 950 °C, is sufficient to achieve complete calcination of absorbent under 1 atm of CO₂, and the absorption capacity of CaO/Ca₁₂Al₁₄O₃₃ (75/25 wt %) at this calcination temperature is about $0.45 \times 0.74 = 0.333$ g-CO₂/g-absorbent, and still higher than that of limestone (0.2 g-CO₂/g-absorbent) and dolomite I (0.25 g-CO₂/g-absorbent).

The calcination temperature and the CO₂ concentration in the calcination atmosphere have an important effect on the cyclic stability of CaO/Ca₁₂Al₁₄O₃₃ (75/25 wt %). When calcium-based absorbents are used to capture CO₂ from gases in order to decrease CO₂ emission into atmosphere, a higher calcination temperature, such as 950 °C, is necessary to produce pure CO₂ suitable for sequestration in greenhouse gas mitigation. When calcination temperature and CO₂ concentration are higher than 950 °C and 20%, the cyclic stability of CaO/Ca₁₂Al₁₄O₃₃ (75/25 wt %) would begin to decline faster than those at lower calcination temperature, but CaO/Ca₁₂Al₁₄O₃₃ (75/25 wt %) is still promising in many other industrial processes such as the methane steam reforming process with CO₂ absorbent addition, chemical heat pump, energy storage system, etc., which need relatively lower calcination temperatures.

3.3. Mechanics of Preparation and Reaction Process of High-Reactivity Ca-Based CO₂ Absorbent. The hydration and calcination of CaO and aluminum nitrate consist of many physical processes and chemical reactions. When CaO is added into water, the reaction of hydration changes CaO to Ca(OH)₂ according to eq (1):



Because of the volume increase from CaO (16.9 cm³/mol) to Ca(OH)₂ (33 cm³/mol) and the expansion caused by the large quantity of heat released (66.5 kJ/mol) in hydration, ultrafine Ca(OH)₂ particle (compared in Figure 6 and Figure 7) can be generated from the cracking and refining of CaO particles suspended in water.

The solution of aluminum nitrate and 2-propanol is mixed with Ca(OH)₂ ultrafine particles with stirring at constant temperature. The addition of 2-propanol makes the particle size distribution of the hydration product Ca(OH)₂ more uniform. The water in the sample can be removed via drying. When the sample is calcined at 773 K, Ca(OH)₂ is heated to decompose to CaO according to eq (2):

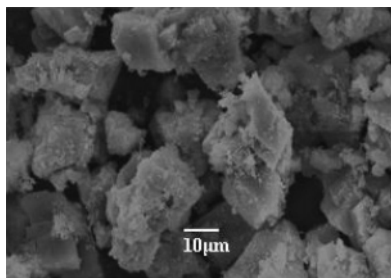


Figure 6. SEM image of original CaO particle.

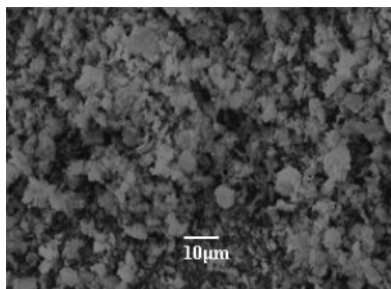
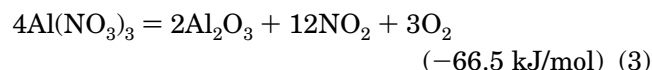


Figure 7. SEM image of CaO/Ca₁₂Al₁₄O₃₃ (75/25 wt %).

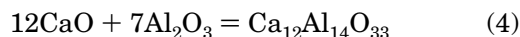


At the same time, aluminum nitrate begins to decompose and generate Al₂O₃ according to the reaction shown in eq (3):



The penetration of aluminum nitrate into particles enables more porosity during the process of decomposition of aluminum nitrate.

When the sample is calcined at higher temperature, CaO and Al₂O₃ inside the particle react to generate a new kind of material, Ca₁₂Al₁₄O₃₃, according to eq (4):



XRD analysis (Figure 8) of the calcined sample shows that only two materials exist: CaO and Ca₁₂Al₁₄O₃₃. During the cyclic carbonation/calcination of CaO/Ca₁₂Al₁₄O₃₃, the Ca₁₂Al₁₄O₃₃ binder does not take part in the reaction and may be considered as inert. Due to the uniform distribution of Ca₁₂Al₁₄O₃₃ among CaO micro-

grains, the sintering of CaO micrograins is retarded by binder Ca₁₂Al₁₄O₃₃ during calcination. The absorbent shows a stable and high CO₂ absorption capacity.

The possible mechanism of hydration for this high-reactivity absorbent is given by Figure 9. The main processes include the following: chemical reaction (CaO hydration) → thermal expansion → chemical reaction calcination → material structure reestablishment (formation of binder among CaO particles) → formation of high surface area. It can be seen from XRD analysis that the binder in the absorbent after high-temperature calcination is Ca₁₂Al₁₄O₃₃. In other words, the preparation process of absorbent depends on not only the chemical reactions but also the physical effects. The ultrafine Ca(OH)₂ with high surface area can be formed in this process. After being calcined at high temperature, the binder formed among CaO ultrafine particles retards the sintering of CaO particles to give the absorbent high reactivity.

3.4. Comparison of Several Ca-Based CO₂ Absorbents. To test the cyclic carbonation/calcination characteristics of CaO/Ca₁₂Al₁₄O₃₃ in detail, the cyclic absorption capacity and cyclic stability of seven absorbents are investigated. The comparison of cyclic absorption capacity is based on ultimate carbonation CO₂ absorption capacity, which is defined as the absorption capacity achieved after 30 min in every carbonation process.

From the comparisons of the CO₂ absorption capacity and cyclic stability among seven absorbents, the stabilities of three dolomites are better than that of pure CaCO₃. The reason for the cyclic reaction stability of dolomite being better than that of pure CaCO₃ may be that the inactive MgO in dolomite does not take part in the carbonation reaction and thus partially hinders the sintering inside the CaO particle during calcination. However, it has been noted that the cyclic absorption capacity of dolomite III becomes lower when more inactive materials are being included. There are many disadvantages for absorbents with a high portion of inactive materials. On one hand, it needs larger quantities of absorbents to capture a certain amount of CO₂ as the activity is low; on the other hand, more energy will be required for the calcination process as the inactive part must also be heated to the decomposition temperature of CaCO₃. Although dolomite III also shows a stable cyclic stability, it has a poor cyclic absorption

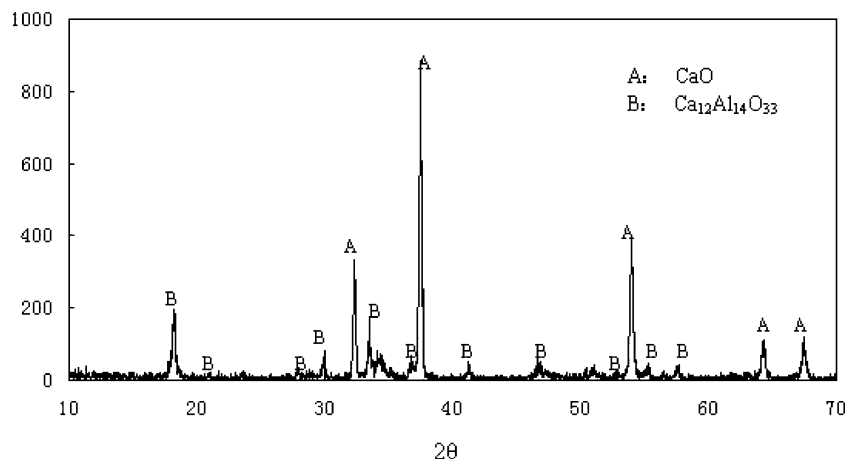


Figure 8. XRD image of the new absorbent CaO/Ca₁₂Al₁₄O₃₃ (75/25 wt %).

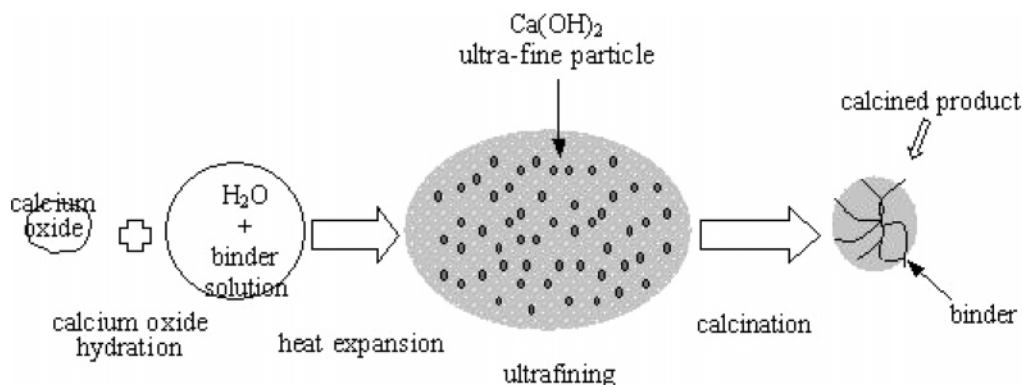


Figure 9. Schematic diagram of hydration preparation of absorbent.

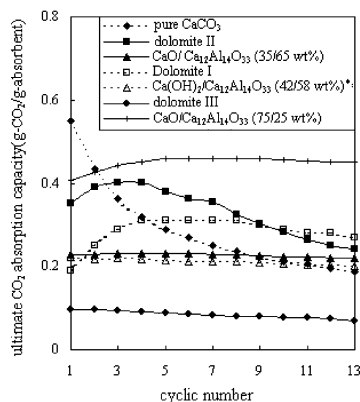


Figure 10. Comparison of ultimate CO₂ absorption capacity of several Ca-based absorbents (carbonation time of 30 min at 963 K with 14% CO₂ and calcination time of 10 min at 1123 K with 100% N₂ for seven absorbents above; ultimate CO₂ absorption capacity is defined as the capacity achieved after 30 min in every carbonation process).

capacity due, to a large extent, to the extraordinarily large MgO content (see Table 1); therefore, it is unsuitable to be a CO₂ cyclic absorbent. The cyclic absorption capacity of dolomite I is lower than that of dolomite II for the first eight cycles, and when the cycle number continue to increase, dolomite I shows higher cyclic absorption capacity than dolomite II as a result of its better cyclic stability. Considering the cyclic stability and cyclic absorption capacity, dolomite I is the preferable CO₂ absorbent among three types of dolomites and pure CaCO₃.

It can be seen from Figure 10 that the absorbent CaO/Ca₁₂Al₁₄O₃₃ (75/25 wt %) has the largest CO₂ absorption capacity and the highest cyclic stability among all absorbents; therefore, CaO/Ca₁₂Al₁₄O₃₃ (75/25 wt %) is

most promising for applications including steam methane reforming. When the mass ratio of CaO to Ca₁₂Al₁₄O₃₃ is 3 to 1 and CO₂ absorption capacity is 0.45 g-CO₂/g-absorbent, it can be calculated that the conversion of CaO is 0.75, indicating its high CaO conversion along with the high cyclic stability. However, more comprehensive tests are needed before CaO/Ca₁₂Al₁₄O₃₃ absorbent can be applied to industrial processes.

4. Conclusions

A new Ca-based CO₂ absorbent, CaO/Ca₁₂Al₁₄O₃₃, has been developed by the integration of CaO with the metal oxide Ca₁₂Al₁₄O₃₃. In comparison with other absorbents, this novel material is able to give high conversion and high cyclic CO₂ absorption capacity for the carbonation reaction. In particular, this material has excellent regeneration capacity in cyclic use, which is one of the key factors for many carbonation applications. The promising results suggest that the new Ca-based absorbent, CaO/Ca₁₂Al₁₄O₃₃, may provide an outstanding performance for the absorption of CO₂ in the carbonation/calcination process. In addition, experimental investigations have been performed by testing pure CaCO₃ and three dolomites, and results show that chemical compositions have important effects on cyclic stability. With an inactive part in dolomite, the cyclic stability of absorbents becomes higher and the cyclic carbonation/calcination characteristics of dolomite I and dolomite II are better than that of pure CaCO₃.

Acknowledgment. This work was supported by the National High Technology Research & Development Program of China (No. 2003AA501330).

EF0496799