

## 一种基于乙二醇-丁二醇体系的新颖的二氧化碳捕集利用方法

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**摘要:** 在温和条件下, 发展了一种以乙二醇-乙二胺体系新颖、高效地固定  $\text{CO}_2$  的方法。在此方法中,  $\text{CO}_2$  被快速激活并转化为一种固态的  $\text{CO}_2$  储集材料( $\text{CO}_2\text{SM}$ ), 通过 XPS、XRD、FTIR 和  $^{13}\text{C}$  NMR 等技术表征证实为烷基碳酸胺。基于 TGA 结果,  $\text{CO}_2\text{SM}$  的水溶液可以与  $\text{Ca}(\text{OH})_2$  和  $\text{Ba}(\text{OH})_2$  反应制备  $\text{CaCO}_3$  和  $\text{BaCO}_3$  微粒, 还可用于循环吸收和解吸  $\text{CO}_2$  的过程。此外, 乙二醇-乙二胺水溶液在 20  $^\circ\text{C}$  下吸收  $\text{CO}_2$  并在 98.6  $^\circ\text{C}$  下解吸  $\text{CO}_2$ , 没有明显的溶液损失。因此, 乙二醇-乙二胺体系提供了一种绿色、高效、低成本的二氧化碳捕集利用方法。

**关键词:**  $\text{CO}_2$  捕获、储集和利用;  $\text{CO}_2$  储集材料;  $\text{CaCO}_3$  和  $\text{BaCO}_3$  微粒; 吸收和解吸循环

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## A $\text{CO}_2$ Capture and Utilization Approach Using the System 1,4-Butanediol and 1,2-Ethanediamine

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**Abstract:** A novel and high effective way to fix  $\text{CO}_2$  agilely through the system 1,4-butanediol (BDO)+1,2-ethylenediamine (EDA) under mild condition was developed. In this process, the bubbling  $\text{CO}_2$  could be activated efficiently and directly converted into a novel solid  $\text{CO}_2$ -storage material ( $\text{CO}_2\text{SM}$ ), which was extensively confirmed as an alkylcarbonate salt using XPS, XRD, FTIR and  $^{13}\text{C}$  NMR analyses. As a material, the aqueous  $\text{CO}_2\text{SM}$  solution could be used to prepare controllable morphologies  $\text{CaCO}_3$  and  $\text{BaCO}_3$  micro-particles by reacting with  $\text{Ca}(\text{OH})_2$  and  $\text{Ba}(\text{OH})_2$  according to thermogravimetry analysis (TGA) result. Additionally, the aqueous BDO+EDA system could be recycled multiple times without any important loss of  $\text{CO}_2$  capturing and releasing capability at absorption temperature of 20  $^\circ\text{C}$  and desorption temperature of 98.6  $^\circ\text{C}$ . As a result, the combination of BDO+EDA seems to provide a green  $\text{CO}_2$  capture and utilization (CCU) approach featuring high efficiency and low cost.

**Keywords:**  $\text{CO}_2$  capture, storage and utilization;  $\text{CO}_2$ -storage material;  $\text{CaCO}_3$  and  $\text{BaCO}_3$  micro-particles; absorption-desorption cycle

### 0 Introduction

Carbon dioxide ( $\text{CO}_2$ ) is a major anthropogenic greenhouse gas (GHG) in the atmosphere<sup>[1]</sup>. Due to

international efforts to decrease greenhouse gases, the reduction of  $\text{CO}_2$  has been extensively studied using electrochemical and photochemical reactions<sup>[2-3]</sup>; however, the results obtained are still not satisfactory.

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Nowadays, the ways to reduce  $\text{CO}_2$  mainly have CCS ( $\text{CO}_2$  capture and storage/sequestration) and CCU ( $\text{CO}_2$  capture and utilization)<sup>[4-6]</sup>. Although CCS has helped to reduce continuing damage to the environment by  $\text{CO}_2$ , it has significant disadvantages that it resulted in waste of C1 resources. In comparison, CCU offers some distinct advantages because it not only consumes  $\text{CO}_2$ , but uses  $\text{CO}_2$  to afford an environmentally friendly C1 feedstock and to produce value-added chemicals. To maximally use  $\text{CO}_2$ ,  $\text{CO}_2$  had to be chemically activated because carbon atom in  $\text{CO}_2$  is very stable<sup>[8]</sup>. Recently, it was reported that  $\text{CO}_2$  could be converted to carbamate or carbamic ester<sup>[9]</sup> with amines, including monoethanolamine<sup>[10-11]</sup>, diethanolamine<sup>[12]</sup>, triethanolamine<sup>[13]</sup>, N-methyldiethanolamine<sup>[14]</sup>, and diglycolamine<sup>[15]</sup>. However, these processes have the major drawbacks, such as the corrosive nature and volatility of the amines, their occasional decomposition and the high energy cost of their regeneration. Recently, Jeesop et al.<sup>[16]</sup> found that the diazabicyclo [5.4.0]-undec-7-ene (DBU), which contains amidogen, could be combined with alcohols to fix  $\text{CO}_2$ . Nevertheless, these systems are still expensive and are not able to attract significant industrial attention.

Our previous studies<sup>[17-20]</sup> showed that ethylene glycol (EG) and its derivatives compounds presented native hydrogen bonding sites and the potential absorption properties for acid gases. Meanwhile, this work showed that EDA could react readily with  $\text{CO}_2$  with an absorption ability of about 0.46 mol  $\text{CO}_2$  per mol EDA, which was in good agreement with Parks work<sup>[21]</sup>. However, the volatility of EDA significantly weakened its  $\text{CO}_2$  absorption ability. Recently, Sengwa<sup>[22]</sup> had reported the hydrogen bonding

interaction and hydrogen bonded structures in amine-alcohol mixed solvents might decrease the loss of amines. Moreover, our previous work<sup>[23]</sup> also indicated that BDO had multiple hydrogen bonding sites and could form hydrogen bond with EDA to reduce amine loss. Therefore, as a fixing agent of EDA, BDO, which has favorable properties such as low vapor pressure, low toxicity, and low melting temperature, was added into EDA to form a mixed system. On the one hand, this could reduce the loss of EDA and improve the absorption performance of  $\text{CO}_2$  when  $\text{CO}_2$  was exposed into the binary system. On the other hand, this could form the novel solid  $\text{CO}_2$ -storage material ( $\text{CO}_2\text{SM}$ ) when  $\text{CO}_2$  was brief introduced into the mixed solution. At the same time,  $\text{CO}_2\text{SM}$  was took advantage of  $\text{CO}_2\text{SM}$  to prepare  $\text{CaCO}_3$  and  $\text{BaCO}_3$  micro-particles. In addition, the absorption and desorption capability of the binary mixed solution was also investigated. As a result, the combination of EDA + BDO seemed to provide a green CCU approach featuring high efficiency and low cost.

## 1 Experimental

### 1.1 Materials

The analytical grade EDA and BDO were purchased from Tianjin Reagent Company. They are used after drying over 0.4 nm molecular sieves and decompression filtration before measurements, and they were degassed by ultrasound just before the experiment. Doubly distilled water with a conductivity lower than  $0.1 \text{ ms} \cdot \text{cm}^{-1}$  ( $25^\circ\text{C}$ ) was used. Compressed  $\text{CO}_2$  (99.999%, V/V) was purchased from the Standard Things Center (China). All the materials were used as received.

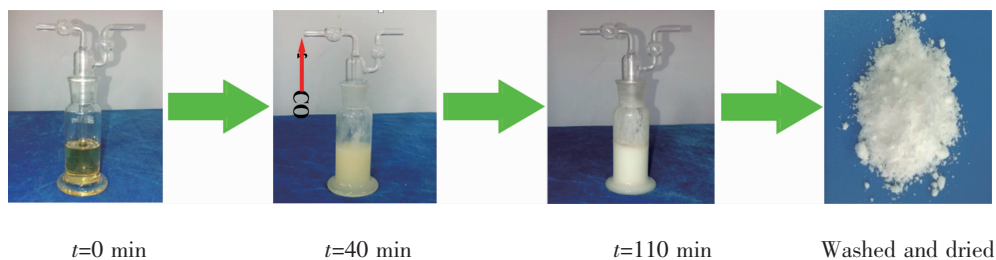


Fig.1 Reaction processes of the system EDA+BDO with  $\text{CO}_2$  at various time

## 1.2 Agile CO<sub>2</sub> Fixation into the CO<sub>2</sub>SM

CO<sub>2</sub> was bubbled into the system BDO+EDA under mild condition. In this process, an interesting phenomenon was found that the system BDO+EDA became turbid at  $t=20$  min and changed into solid powder at  $t=110$  min. And then, the solid powder was washed three times with ethanol and dried under vacuum at 60 °C for 3 h, which was stored at room temperature and named as the CO<sub>2</sub>SM (Fig.1).

The solid CO<sub>2</sub>SM was characterized by X-ray photoelectron spectroscopy (XPS), X-ray diffraction (XRD), Fourier transform infrared reflectance (FTIR) and <sup>13</sup>C NMR spectroscopic techniques. XPS data were obtained with a KRATOS Axis ultra X-ray photoelectron spectrometer with a monochromatized AlK $\alpha$  X-ray ( $h\nu=1\,486.6$  eV) operated at 150 W. XRD patterns were collected on a powder X-ray diffractometer (Siemens D/max-RB) with Cu K $\alpha$  ( $\lambda=0.154\,06$  nm) radiation and scanning rate of  $0.05^\circ\cdot\text{s}^{-1}$  at working voltage of 40 kV and working current of 40 mA. FTIR spectra of CO<sub>2</sub>SM were taken as 1% dispersion in KBr powder using a Nexus 670 FTIR spectrometer with a resolution of  $1\text{ cm}^{-1}$  in the range from  $4\,000\sim 400\text{ cm}^{-1}$ , and a base line correction were made for the spectra that were recorded in air at room temperature. <sup>13</sup>C NMR spectra was recorded on a Bruker ARX-400 nuclear magnetic resonance spectrometer equipped with a 4 mm standard bore CP/MAS probe head. In addition, for monitoring the reaction real-time, the liquid <sup>13</sup>C NMR of mixing system was measured as follows: (I) pure EDA and EDA in DMSO- $d_6$ ; (II) the system BDO+EDA (molar ratio 1:1); (III) CO<sub>2</sub> bubbled into the system BDO+EDA system 15 min, 30 min, and 45 min, respectively; and (IV) the liquid <sup>13</sup>C NMR spectra of the system CO<sub>2</sub>+BDO+EDA were not measured after 45 min, and only the solid CO<sub>2</sub>SM was measured by the solid <sup>13</sup>C NMR spectra.

At the same time, thermogravimetry analysis (TGA, Q50 V20.6 Build 31) was employed to measure the weight change of CO<sub>2</sub>SM. In the thermal stability experiments, the sample was heated up to 400 °C with a rate of  $10\text{ }^\circ\text{C}\cdot\text{min}^{-1}$  in the nitrogen atmosphere.

## 1.3 Absorption and desorption processes

Absorption-desorption cycles of the aqueous BDO+EDA system with CO<sub>2</sub> were carried out at an absorption temperature of 20 °C and a desorption temperature of 98.6 °C at ambient pressure. The absorption process was conducted for 15 min and desorption process was run for 90 min. All reactions were carried out in a 100 mL gas-washing bottle, and all measurements of mass were performed on an electronic balance with an accuracy of 0.1 mg (Sartorius BS224S). The desorption experiments were carried out after the absorption. The CO<sub>2</sub>-saturated absorption system was heated in a pre-heated oil bath with condensation and stirring. And then a follow-up absorption cycle was marched after the system cooled.

## 2 Results and discussion

### 2.1 Study on CO<sub>2</sub> absorption process

Firstly, the loading capacity of CO<sub>2</sub>, which was expressed as molar ratio of CO<sub>2</sub> (absorbed)/EDA (starting), in pure BDO and pure EDA were studied at  $400\text{ mL}\cdot\text{min}^{-1}$  of CO<sub>2</sub>, respectively. The result showed that the pure BDO only had little-to-no CO<sub>2</sub> binding capacity (about 0.003 15 mol of CO<sub>2</sub> per mole BDO) through physical absorption and the pure EDA had strong CO<sub>2</sub> capability of approximately 0.467 mol CO<sub>2</sub> per mol EDA. However, in the system BDO+EDA, it showed an unimaginable capability to capture CO<sub>2</sub> (0.709 mol per mol), indicating BDO participated in the reaction

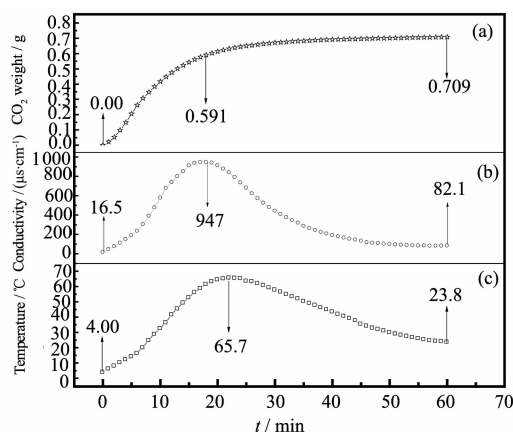


Fig.2 Variation of weight (a), electrical conductivity (b), and temperature (c) changes for the system EDA+BDO at CO<sub>2</sub> flow rate of  $400\text{ mL}\cdot\text{min}^{-1}$

of CO<sub>2</sub> with EDA. In the process, the varieties in weight, electrical conductivity and temperature of the system BDO+EDA were recorded with the bubbling CO<sub>2</sub> (Fig.2).

As shown in Fig.2(a), the loading capability of CO<sub>2</sub> rapidly increased before 20 min, and then slowly increased with the lengthening absorption time. Moreover, the solution became turbidity after 20 min. Before turbidity, the system CO<sub>2</sub>+BDO+EDA promoted mass transfer and intensified intermolecular collision. After turbidity, solid granules prevented mass transfer, slowed down intermolecular collision and decreased saturation time. Therefore, the concentration of solid granules increased with the increasing reaction time, and the solution finally changed into solid. Compared with the previously-reported loading performance of CO<sub>2</sub> in various aqueous alkanolamines<sup>[24]</sup>, including MEA (0.462 8 mol CO<sub>2</sub> per mol amine), DEA (0.236 0 mol CO<sub>2</sub> per mol amine) and TEA (0.194 4 mol CO<sub>2</sub> per mol amine), the novel system BDO+EDA with much higher CO<sub>2</sub> loading capacity because BDO, as a reactant, participated in the reaction between CO<sub>2</sub> and EDA.

From Fig.2 (b), the conductivity of the system BDO+EDA kept on increasing from 16.49  $\mu\text{S}\cdot\text{cm}^{-1}$  to 947  $\mu\text{S}\cdot\text{cm}^{-1}$  rapidly within the first 19 min, which

indicated that the molecules violently motioned among BDO, EDA, and CO<sub>2</sub> to form ionic compound. With the increase of reaction time, the conductivity values decreased from 947  $\mu\text{S}\cdot\text{cm}^{-1}$  to 82.1  $\mu\text{S}\cdot\text{cm}^{-1}$  within another 41 min, which was mainly due to the gradually increased viscosity of this system, the generation of white solid granules, and the decreased ion movement. Simultaneously, the conductivity change also demonstrated the dramatic change in charge between the low ionic strength solution and the high ionic strength solution caused the increasing viscosity of the system BDO+EDA. This also proved that the system firstly translated into ionic compound when CO<sub>2</sub> was exposed in solution, and then precipitated in the form of solid precipitation.

As shown in Fig.2 (c), the change trend of temperature was in good agreement with that of conductivity. Obviously, CO<sub>2</sub> capture process was exothermal, in which the temperature quickly increased from 4.0 to 65.7  $^{\circ}\text{C}$  within 22 min, and then slowly decreased by 23.8  $^{\circ}\text{C}$  with the increasing absorption time. According to the temperature monitoring, the dramatical change of temperature suggested that the BDO+EDA system capture CO<sub>2</sub> through chemical reaction rather than physical adsorption only and the change trend of temperature

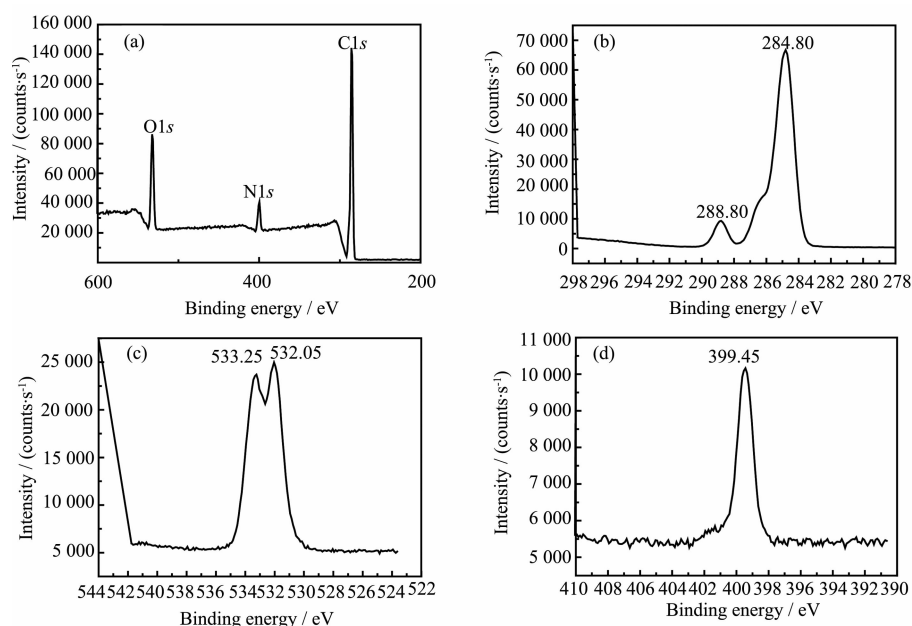


Fig.3 XPS spectra of CO<sub>2</sub>SM: (a) the total element, (b) oxygen element, (c) carbon and (d) nitrogen element

could disclose the extent of capturing process.

## 2.2 Agile CO<sub>2</sub> fixation into the solid CO<sub>2</sub>SM

In this work, a series of experiments were designed to absorb CO<sub>2</sub> by pure BDO, pure EDA, and BDO+EDA system, respectively. In surprise, no solid products were formed in pure BDO or pure EDA. However, the white solid precipitate was formed after CO<sub>2</sub> was bubbled into the system BDO+EDA (molar ratio 1:1) under mild condition for approximately 110 min. And then, the white solid precipitate, named as the CO<sub>2</sub>SM, was thoroughly washed three times with ethanol, dried under vacuum at 60 °C for 3 h. And the properties of CO<sub>2</sub>SM were characterized with XPS, XRD, FTIR and <sup>13</sup>C NMR techniques.

XPS spectra of CO<sub>2</sub>SM were measured and shown in Fig.3. The peak at 288.80 eV was due to the carbon atoms perturbed by three oxygen bonds<sup>[25]</sup>, and the peak at 284.80 eV was due to single carbon oxygen bond (Fig.3(a) and (b))<sup>[26]</sup>. At the same time, two peaks at 532.05 and 533.25 eV were due to O1s (Fig.3(c)) in carbonate-like structures<sup>[27-28]</sup>. In addition, the peak at

399.45 eV, which was due to N1s (Fig.3 (d)), and the peak at 284.80 eV (Fig.3(b)) showed NH<sub>3</sub><sup>+</sup> existed<sup>[29]</sup>. Based on above result, it was found that the CO<sub>2</sub>SM contained the groups of NH<sub>3</sub><sup>+</sup> and CO<sub>3</sub><sup>2-</sup>.

The CO<sub>2</sub>SM had a structure with 17.50°, 21.48°, 22.40°, 22.60°, 29.24°, and 35.42° (Fig.4(a)), which was similar to ethlenediamine carbamate (-NH-COO-) characteristic diffraction peaks (JCPDS 34-1993). The FTIR peaks at 3 307 and 2 189 cm<sup>-1</sup>, which could be assigned to stretching vibration mode of N-H and characteristic absorption peak of -NH<sub>3</sub><sup>+</sup> of CO<sub>2</sub>SM, suggested that the CO<sub>2</sub>SM was a primary amine salt<sup>[30-31]</sup>. Two intense absorption peaks at 1 574 and 1 483 cm<sup>-1</sup> were due to asymmetric and symmetric stretching mode of -CO<sub>2</sub><sup>-</sup><sup>[32]</sup>, respectively. In addition, the characteristic absorption peak at 1 373 cm<sup>-1</sup> denoted carbonate (CO<sub>3</sub><sup>2-</sup>) rather than bicarbonate because the typical peaks of bicarbonate appeared at 1 360 and 835 cm<sup>-1</sup> (Fig.4(b)). Thus, the obtained solid products was likely composed of alkylcarbonate salt<sup>[33-35]</sup>.

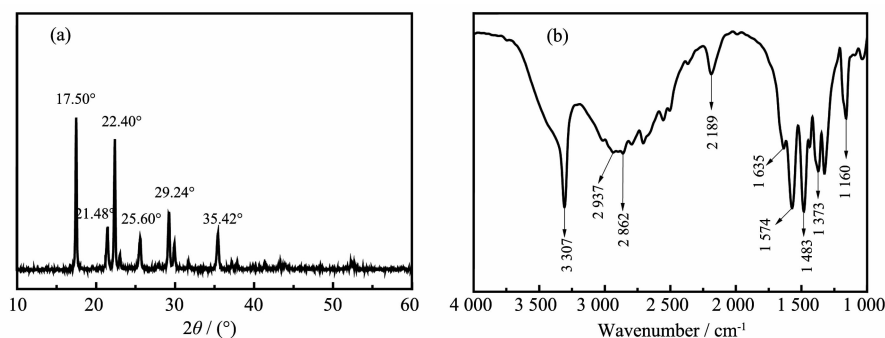
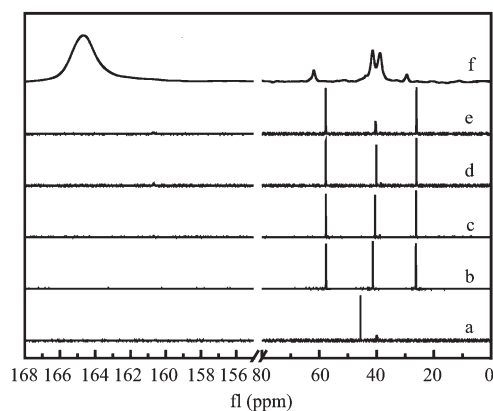


Fig.4 XRD pattern (a) and FTIR (b) of CO<sub>2</sub>SM

The <sup>13</sup>C NMR spectra of EDA in DMSO-d<sub>6</sub> were firstly tested and the result showed that a lathy singlet appeared at 45.594 ppm<sup>[36]</sup>, which was attached to carbon of EDA, and heptet of carbon in DMSO-d<sub>6</sub> appeared at 39.952 (Fig.5a). And the system BDO+EDA (molar ratio 1:1) presented three singlets at 60.923, 44.574 and 29.580 in the <sup>13</sup>C NMR spectrum (Fig.5b), in which the chemical shift (45.594) of carbon in EDA shifted towards high-field (44.574) because of the formation of hydrogen bond between BDO and EDA<sup>[23]</sup>, and other new peaks at 60.923 and 29.580 were assigned to carbon in BDO. At the same

time, a new peak appeared and stabilized at 163.965 after CO<sub>2</sub> was introduced into the system BDO+EDA for 15, 30 and 45 min, respectively; and the characterization peak of solid CO<sub>2</sub>SM was observed at 164.664 (Fig.5c~f). And this indicated not only the CO<sub>2</sub>SM was immediately formed and steadily existed after CO<sub>2</sub> was introduced into the system BDO+EDA, but the CO<sub>2</sub>SM was alkylcarbonate salt rather than bicarbonate because bicarbonate was often found at about 160 in <sup>13</sup>C NMR<sup>[37-43]</sup>. Therefore, the <sup>13</sup>C NMR analysis further illustrated that the system BDO+EDA could agilely activate CO<sub>2</sub> to form the alkylcarbonate

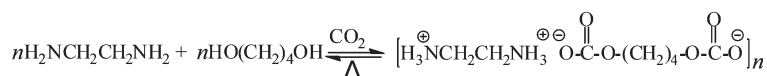




(a) DMSO EDA, (b) EDA+BDO system, (c) EDA+BDO system absorbed CO<sub>2</sub> 15 min, (d) EDA+BDO system absorbed CO<sub>2</sub> 30 min, (e) EDA+BDO system absorbed CO<sub>2</sub> 45 min, (f) solid CO<sub>2</sub>SM

Fig.5 <sup>13</sup>C NMR spectra of EDA+BDO system absorbed CO<sub>2</sub> and formed solid CO<sub>2</sub>SM

salt. Therefore, a possible formation mechanism (Scheme 1) of the CO<sub>2</sub>SM was proposed based on above analytic results. Similarly, Jessop and his co-



Scheme 1 Formation of CO<sub>2</sub>SM from CO<sub>2</sub>, EDA and BDO

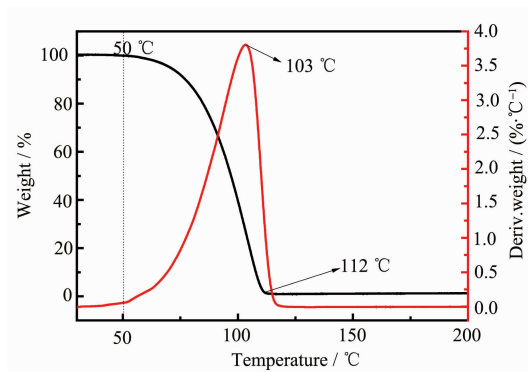


Fig.6 TGA curve for CO<sub>2</sub>SM under nitrogen at a temperature scan rate of 10 °C·min<sup>-1</sup>

Based on TGA-DSC result, CO<sub>2</sub>SM could release CO<sub>2</sub> at 112 °C. Therefore, a potential application of CO<sub>2</sub>SM for preparing CaCO<sub>3</sub> and BaCO<sub>3</sub> was developed. And thus, 0.05 g CO<sub>2</sub>SM was added into Ca(OH)<sub>2</sub> saturated limpid solution, which hydrothermal reaction 2 h at 110 °C in bake oven, and the dumbbell-like CaCO<sub>3</sub> micro-particles were obtained (Fig.7a). The XRD pattern and FTIR spectrum of the

workers [16,44] also reported an innovative class of CO<sub>2</sub>BOL with an alcohol and an amidine or guanidine superbase, which led to the formation of amidine or guanidine alkylcarbonate salts after CO<sub>2</sub> capturing.

TGA-DSC of the CO<sub>2</sub>SM was firstly investigated and shown in Fig.6. The TGA-DSC results showed that the CO<sub>2</sub>SM began to decompose at approximately 50 °C, losing up to 8% of its mass from 50 to 75 °C. Then, the decomposition of CO<sub>2</sub>SM was accelerated at about 103 °C with massive release of CO<sub>2</sub> and the evaporation of EDA. As it could be seen, this decomposed step in the temperature range of 75 to 112 °C decreased 90.5%. And at about 112 °C, a large exothermic peak was due to thermal volatilization of EDA, and the release of CO<sub>2</sub>, which indicated the CO<sub>2</sub>SM completely decomposed. In addition, TGA result also indicated that the CO<sub>2</sub>SM was stable before 50 °C and released CO<sub>2</sub> after heating.

dumbbell-like CaCO<sub>3</sub> suggested that the polymorph of dumbbell-like CaCO<sub>3</sub> was most stable calcite (Fig.S1). Spindle-like BaCO<sub>3</sub> micro-particles could be obtained after the reaction, which 5 g CO<sub>2</sub>SM was added into 0.01 mol·L<sup>-1</sup> Ba(OH)<sub>2</sub> solution, then hydrothermal reaction at 110 °C oven for 2 h (Fig.7b).

Meanwhile, the XRD pattern and FTIR spectrum of the Spindle-like BaCO<sub>3</sub> indicated that BaCO<sub>3</sub> samples could be indexed as typical orthorhombic phase (Fig.S2). In this process, CO<sub>2</sub> released from CO<sub>2</sub>SM by heating reacted with Ca(OH)<sub>2</sub> and Ba(OH)<sub>2</sub> solution to generate amorphous CaCO<sub>3</sub> and BaCO<sub>3</sub>, respectively. The released EDA and rested BDO, which acted as a dispersant, make amorphous CaCO<sub>3</sub>, BaCO<sub>3</sub> to rearrange to form a regular, orderly dumbbell-like CaCO<sub>3</sub> and spindle-like BaCO<sub>3</sub> micro-particles. As for the specific formation process of CaCO<sub>3</sub>, BaCO<sub>3</sub> will be detailedly reported in the future work.

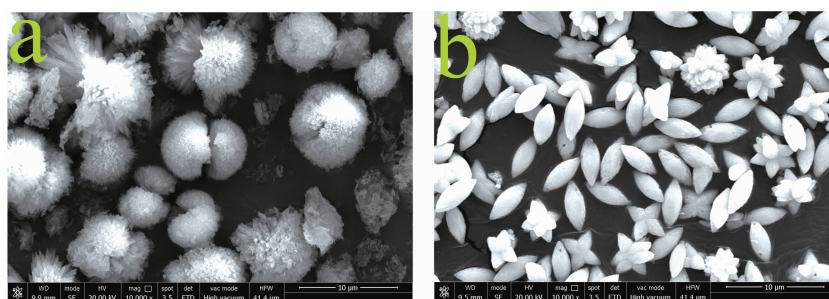


Fig.7 (a) SEM image of dumbbell-like  $\text{CaCO}_3$ ; and (b) SEM image of spindle-like  $\text{BaCO}_3$

### 2.3 Absorption-desorption cycles

Firstly, the experiments of  $\text{CO}_2$  absorption with the aqueous BDO +EDA system were conducted at  $20.0^\circ\text{C}$  under atmospheric pressure and the changes in sample weight were recorded by using an electronic balance. Secondly, the desorption of  $\text{CO}_2$  from the loaded solution, prepared as described above, were carried out at  $98.6^\circ\text{C}$  with stirring until  $\text{CO}_2$  stopped being released from the solution, which lasted for 90 min. Finally, the continuous cycles of simultaneous  $\text{CO}_2$  absorption-desorption were carried out and the results were shown in Fig.8.

As shown in Fig.8, the aqueous BDO +EDA solution could be recycled at least for five continuous absorption-desorption cycles without any important loss of  $\text{CO}_2$  capturing and releasing capability. Whats more, the regeneration temperature was lower than the traditional MEA system in the range from  $100$  to  $140^\circ\text{C}$ <sup>[39]</sup>. It might be due to much lower thermostability of the formed alkylcarbonate salts. Moreover, the maximum  $\text{CO}_2$  loading capacity could reach  $1.540$  mol  $\text{CO}_2$  per mol EDA in the absorption-desorption cycles.

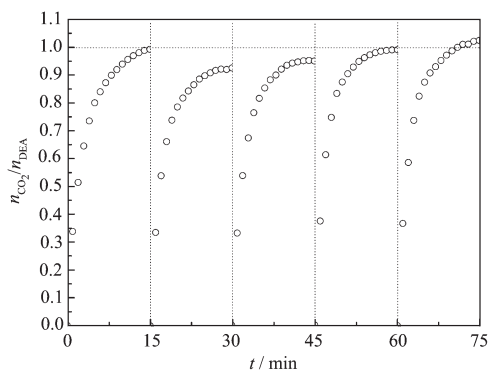


Fig.8 Reversible  $\text{CO}_2$  capture and release by 6% (w/w) aqueous EDA/BDO system

Overall, all the above results indicate that the aqueous BDO +EDA (molar ratio 1:1) system provide a high loading capacity, stable absorption-desorption cycles and low energy requirement processes for the application in industry. In short, all results showed that such a process not only restrained the volatilization of EDA, but also fulfilled the capture, low energy loss and utilization of  $\text{CO}_2$ .

### 3 Conclusions

In conclusion, an innovative CCU approach featuring cost-effectiveness, high capturing capacity and acceptable regeneration energy for industrial processes was developed. Particularly, it was found that  $\text{CO}_2$  gas can be bubbled into the EDA-BDO system under mild condition to generate the novel solid  $\text{CO}_2\text{SM}$ , which was extensively confirmed as an alkylcarbonate salt. Especially, the aqueous EDA + BDO system is superior  $\text{CO}_2$  storage system, exhibiting remarkable  $\text{CO}_2$  capturing and releasing capability after multiple cycles. In addition, the aqueous  $\text{CO}_2\text{SM}$  solution could be employed to prepare  $\text{CaCO}_3$  and  $\text{BaCO}_3$  micro-particles.

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