



Capture of carbon dioxide from flue/fuel gas using dolomite under microwave irradiation

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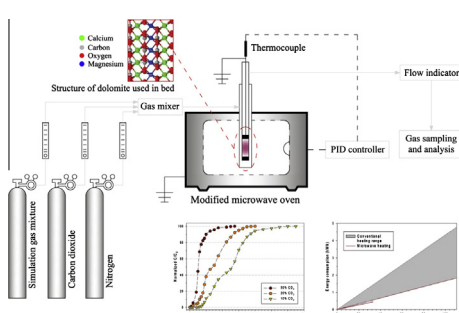
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HIGHLIGHTS

- We develop microwave heating system to capture CO₂ by a packed bed of dolomite.
- Dolomite shows consistent CaO conversion of 90% within 15 cycles in TGA tests.
- We capture CO₂ from producer gas using a packed bed of dolomite heating under microwave.
- We find that carbonation kinetics is governed by both chemical reaction and diffusion.

GRAPHICAL ABSTRACT



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ABSTRACT

Naturally occurring dolomite is a low cost CO₂ sorbent with superior performance to well known limestone. In this work, a microwave heating system was developed to carry out the CO₂ capture experiment using a packed bed of calcined dolomite. Preliminary tests were performed in thermogravimetric analyzer (TGA) from which the calcination–carbonation temperatures of 750–700 °C were found as the suitable condition to obtain a conversion of 90% that could be sustained during 15 cycles of CO₂ capture and release. Dynamic breakthrough curves of CO₂ capture were developed by introducing different concentrations of CO₂ (10–50 vol% in N₂) into a packed bed of calcined dolomite heating under microwave irradiation. The appropriateness of the developed system was further examined through its implementation for CO₂ capture from a gas stream simulating air gasification producer gas. The regenerability of the bed was assessed in three cycles of calcination–carbonation (750–700 °C) in microwave where the sorbent revealed a high and relatively sustainable performance for CO₂ capture. Kinetic studies based on shrinking core model deduced that the carbonation reaction mechanism in the packed bed was governed by a combination of chemical reaction and CO₂ diffusion.

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1. Introduction

Clean energy technologies are all about supplying power from sustainable resources while meeting the challenge of reducing carbon footprint. These technologies have experienced impressive progresses in recent years. Yet, surging demand for fossil fuels has

overshadowed the deployment of clean energy technologies. Considering that carbon-based fossil fuels contribute to 75% of anthropogenic CO₂ emission [1], the long-standing relationship between industrial growth and carbon dioxide emissions would exist.

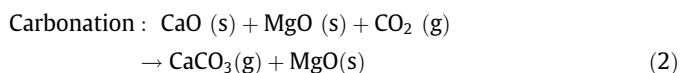
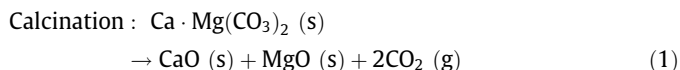
Carbon capture from combustion or gasification plants to produce a pure stream of CO₂ for sequestration in the earth, commonly known as CCS, has been considered as one of the key technologies to mitigate the anthropogenic CO₂ emissions. However, the current state-of-the-art technology imposes considerable energy penalty to the industry [2]. Implementation of inexpensive

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and enduring sorbent material which can withstand high temperatures (up to 900 °C) dictated by flue gas or product gas of gasification process would definitely have considerable impact on the economics of CO₂ capture. Calcined limestone is plentifully available in low cost and is well known for its ability to scavenge CO₂ from exhaust gas, virtually coming from any industry, to very low concentrations [3]. However, the disadvantage associated with limestone is its diminishing CO₂ capture capacity upon several cycles of capture and release [3,4]. This decay in real time performance is exacerbated in each cycle and the calcium oxide carbonation conversion drops to below 20% within 20 cycles [5]. Such decay in performance was found to emanate from sintering of CaO at temperatures above 530 °C (Tammann temperature of CaCO₃) which blocks the pores and impedes the access of CO₂ to the sorbent interior [6].

Silaban et al. [7] found that naturally occurring carbonates of calcium and magnesium, known as dolomite (Ca·Mg (CO₃)₂), in calcined form presented a superior CO₂ capture performance compared to calcined CaCO₃ with improved multi-cycle durability based on the following reactions:



This superior performance of calcined dolomite is believed to be attributed to MgO which does not react with CO₂ at high carbonation temperatures; only CaO converts to carbonate [8,9]. The un-reacted MgO creates excess pore volume and provides free passage for CO₂ diffusion; this keeps virtually the entire active sites of the sorbent accessible and results in a favorable cycling performance [10,11].

From an economic point of view, naturally occurring carbonates such as limestone and dolomite may offer the cheapest option for CCS technology. As mentioned, a major hurdle in the implementation of CCS technology is the severe energy penalties imposed to power plants. Current industrial CO₂ capture technologies reduce the net electricity output by 13–37% [2]. Whereas, a feasible CO₂ capture technology must be able to achieve 90% CO₂ capture with no more than 10% energy penalty [12]. This signifies the importance of implementation of cheap sorbents and energy efficient technologies for CO₂ capture and sorbent regeneration with lowest possible energy penalties imposed to the plant.

Microwave technology has been explored as one of the energy efficient systems in heating applications. Microwave systems are known with key features such as high energy efficiency and short processing time. Considering the fact that in microwave heating no time is wasted to heat the surrounding area of the material and microwave irradiation is directly delivered to the material through molecular interaction with the electromagnetic field, a higher heating rate is achieved [13]. With this high heating rate, it takes few minutes to reach a high temperature, whereas this time might be prolonged to hours in conventional heating systems [14]. Microwave heating systems lead to considerable savings in both energy consumption and process time.

Microwave systems have been implemented for several heating applications such as pyrolysis [14,15], decomposition of tar and

volatiles [16,17] and development of new materials [18]. However, use of microwave heating system for CO₂ capture, in spite of its high energy efficiency, merited very little attention so far. To the best of authors' knowledge, there is no report in the open literature regarding CO₂ capture using microwave heating. With the emerging emphasis on utilization of cost-efficient methods in CCS technology, the aim of this study was to employ dolomite as a low cost sorbent for CO₂ capture under microwave irradiation. For this purpose, preliminary CO₂ capture experiments were performed in thermogravimetric analyzer (TGA) to monitor the sorption capacity of the calcined dolomite and its cyclic carbonation-calcination durability. Further CO₂ capture experiments were carried out in a packed bed reactor under microwave irradiation with varying CO₂ concentration. In addition, to show the applicability of the developed system, CO₂ was captured from a gas stream which simulated the product gas of air gasification and the regenerability of the dolomite sorbent was examined in some carbonation-calcination cycles under microwave irradiation. Kinetics of the carbonation reaction under microwave heating was also studied to determine the rate controlling step of the reaction.

2. Materials and methods

2.1. Sorbent material

Naturally occurring dolomite (Ca·Mg (CO₃)₂) was obtained from a dolomite mine located at Alborz Mountains in Iran. It was crushed and sieved to a grain size of 2–3 mm. The ground dolomite was then calcined at 900 °C for 3 h to obtain the oxide form. The calcined dolomite was ground to a particle size of around 75–100 μm and stored in desiccator for CO₂ capture experiments. The chemical composition of the calcined dolomite was determined by X-Ray Fluorescence (XRF) analysis (Rigaku RIX3000, Japan). For this purpose, a sample of raw dolomite was ignited at 950 °C for 5 h. The LOI was calculated as the weight percent loss on ignition. The ignited sample was used to prepare the fused disk for XRF analysis. The results of XRF analysis are summarized in Table 1.

2.2. CO₂ capture in TGA

The preliminary CO₂ capture experiments were carried out in a Thermogravimetric analyzer (TGA, SDTQ-600) to find the optimum operating temperatures at which the highest CO₂ capture could be achieved. A small amount of the sorbent (10–12 mg) was loaded in a ceramic pan and heated at a rate of 30 °C/min under a N₂ flow rate of 50 ml/min to the pre-set calcination temperature (750 or 850 °C). The sorbent was calcined for 15 min, then the temperature was reduced to the desired carbonation temperature (750, 700 or 650 °C). At the onset of carbonation, N₂ was switched to CO₂ at the same flow rate to initiate the CO₂ capture test. The variation in the sample weight and temperature was recorded continuously as a function of time. Based on the recorded results, molar CaO conversion was calculated as [19]:

$$x_{\text{CaO}}\% = \frac{W_{\text{CaO}}\%}{0.786 \times \alpha_{\text{CaO}}} \quad (1)$$

where $W_{\text{CaO}}\%$ represents the weight percentage change of the sorbent during the course of CO₂ capture, 0.786 (g CO₂/g CaO) is the

Table 1
Chemical composition of natural dolomite.

Composition	CaO	MgO	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	Na ₂ O	SO ₃	K ₂ O	TiO ₂	P ₂ O ₅	MnO	SrO	LOI
wt.%	34.69	15.06	2.34	1.07	0.61	0.44	0.16	0.12	0.04	0.02	0.02	0.02	45.4

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