

# Kinetics of carbonation reaction of basic oxygen furnace slags in a rotating packed bed using the surface coverage model: Maximization of carbonation conversion



Shu-Yuan Pan<sup>a</sup>, Pen-Chi Chiang<sup>a</sup>, Yi-Hung Chen<sup>b</sup>, Chung-Sung Tan<sup>c</sup>, E.-E. Chang<sup>d,\*</sup>

<sup>a</sup> Graduate Institute of Environmental Engineering, National Taiwan University, 71 Chou-Shan Rd., Taipei City, Taiwan 10673, Taiwan, ROC

<sup>b</sup> Department of Chemical Engineering and Biotechnology, National Taipei University of Technology, 1, Sec. 3, Zhongxiao E. Rd., Taipei City, Taiwan 10608, Taiwan, ROC

<sup>c</sup> Department of Chemical Engineering, National Tsing Hua University, 101, Sec. 2, Kuang-Fu Rd., Hsinchu, Taiwan 30013, Taiwan, ROC

<sup>d</sup> Department of Biochemistry, Taipei Medical University, 250 Wu-Hsing Street, Taipei City, Taiwan 110, Taiwan, ROC

## HIGHLIGHTS

- Kinetics of BOFS carbonation reaction was well expressed by surface coverage model.
- The model was validated by SEM images which indicate the BOFS was covered by CaCO<sub>3</sub>.
- Carbonation conversion of BOFS with CRW was higher than that in pure-water system.
- Reaction rate in RPB was greater than that in both autoclave and slurry reactors.
- The maximum conversion was determined by kinetic model and response surface method.

## ARTICLE INFO

### Article history:

Received 7 April 2013

Received in revised form 17 July 2013

Accepted 17 July 2013

Available online 8 August 2013

### Keywords:

Carbon capture

Reaction rate constant

Calcium carbonate

Autoclave reactor

Slurry reactor

Response surface methodology

## ABSTRACT

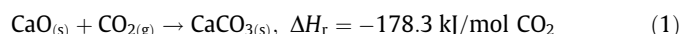
The reaction kinetics of carbon dioxide (CO<sub>2</sub>) capture by the accelerated carbonation of basic oxygen furnace slag (BOFS) in a rotating packed bed (RPB) was evaluated using the surface coverage model. Experimental data were utilized to determine the reaction rate constants and maximum carbonation conversion of BOFS based on the surface coverage model. The results indicate that the carbonation of BOFS in an RPB can be well-expressed by the surface coverage model, with *R*<sup>2</sup> values from 0.98 to 0.99. In addition, the results of reaction kinetics could be validated by observation of SEM and XEDS before and after carbonation, which indicates that the reacted BOFS was surrounded by the CaCO<sub>3</sub> product. On the other hand, the reaction kinetics of steelmaking slag in an RPB was compared with that in various types of reactors, i.e., autoclave and slurry reactors. The overall rate of carbonation in an RPB (i.e., 0.299 min<sup>-1</sup>) was greater than that in both a slurry reactor (i.e., 0.227 min<sup>-1</sup>) and an autoclave reactor (i.e., 0.033 min<sup>-1</sup>). Furthermore, the maximum carbonation conversion of BOFS was initially determined by the results of the surface coverage model and then confirmed statistically by the response surface methodology (RSM). It was thus concluded that accelerated carbonation of BOFS in the RPB is a viable method due to its faster reaction kinetics under relatively milder reaction conditions. Accelerated carbonation of BOFS in the RPB is a promising process for CO<sub>2</sub> capture due to its relatively higher carbonation conversion of BOFS within a shorter reaction time.

© 2013 Elsevier Ltd. All rights reserved.

## 1. Introduction

Fossil fuels will remain the dominant source of energy around the world through at least 2035 and perhaps longer [1]. To keep carbon dioxide (CO<sub>2</sub>) concentration at or below 550 ppm in the atmosphere over the next 100 years, development of appropriate technologies for carbon capture, utilization, and storage (CCUS) is

essential to reduce CO<sub>2</sub> emission into the atmosphere. Accelerated carbonation, which mimics natural weathering processes by promoting the reaction kinetics under a moisture-rich condition or in a liquid solution, is a promising method for removing CO<sub>2</sub> from emission sources since energy consumption and costs may be reduced by the inherent properties of carbonation (an exothermal reaction) [2,3]. CO<sub>2</sub> reacts with metal-oxide bearing materials such as CaO and Ca(OH)<sub>2</sub> to form a thermodynamically stable and insoluble product (CaCO<sub>3</sub>) as shown in Eqs. (1) and (2), respectively:



\* Corresponding author. Tel.: +886 2 2376 9236; fax: +886 2 2736 1661.

E-mail address: [eechang@tmu.edu.tw](mailto:eechang@tmu.edu.tw) (E.-E. Chang).

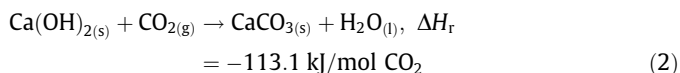
## Nomenclature

### Materials

BHC	blended hydraulic slag cement
FAS	fly-ash slag
UFS	ultra-fine slag
BOFS	basic oxygen furnace slag
CRW	cold-rolling wastewater
EW	effluent water from wastewater treatment plant
DW	de-ionized water

### Analytical methods

PSD	particle size distribution
XRF	X-ray fluorescence
TG	thermo-gravimetric analysis
DTG	differential thermo-gravimetric analysis
DSC	differential scanning calorimetric analysis



Alkaline solid wastes such as basic oxygen furnace slags (BOFS) and municipal solid waste incinerator bottom ash (MSWI-BA) are an available source of CaO due to their low cost [2,4–7]. Although the carbonation of steelmaking slag would contribute only 170 Mt/year on reduction of CO<sub>2</sub> emission (the world's annual slag production was estimated to be 220–420 Mt, with a CaO content of 34–52%) [8], it is considered a “win-win” strategy because there are both environmental and economic benefits. For instance, the carbonation process can not only fix CO<sub>2</sub> by steelmaking slag but can also improve the physico-chemical properties of steelmaking slag, which is utilized for construction materials, by eliminating free-CaO and Ca(OH)<sub>2</sub> [7–10]. Therefore, many studies have focused on performance evaluation of different alkaline wastes using various processes to accelerate the carbonation reaction [11,12]. However, the reaction rate of carbonation might be not fast enough to reach the CO<sub>2</sub> emission rate in industry if the reaction occurs in traditional reactors such as autoclave or slurry reactors [5,6].

The carbonation reaction of BOFS slurry can be further accelerated by using a rotating packed bed (RPB) [13,14], which could lead to the formation of thin liquid films via high gravity, thereby increasing the contact surface area and mass transfer between the gas and liquid phases [15–19]. According to the previous study [13], the maximum carbonation conversion of BOFS achieved in an RPB was 93.5% operated under a liquid-to-solid (L/S) ratio of 20:1 mL g<sup>-1</sup>, a pure-CO<sub>2</sub> flow rate of 2.5 L min<sup>-1</sup>, and a rotation speed of 750 rpm at 65 °C for 30 min. In addition, the RPB process was proven effective for ex situ CO<sub>2</sub> capture using hot-stove gas (containing 30% CO<sub>2</sub>) in China Steel Corp. [14]. However, the kinetics of carbonation reaction in an RPB was not determined, and the effects of operating parameters on carbonation have not been critically evaluated in the literature.

The surface coverage model was originally developed by Ho and Shih [20] and then further verified as the most suitable model to describe the reaction kinetics of carbonation for hydrated lime and/or the desulfurization of flue gas using Ca(OH)<sub>2</sub> at low temperatures [21–24]. According to the assumption of the surface coverage model, for instance, the carbonation of Ca(OH)<sub>2</sub> would result in the formation of precipitation products, which would then coat on the active site of the surface of Ca(OH)<sub>2</sub>. As the reaction proceeded, the precipitation products would occupy more of the surface of Ca(OH)<sub>2</sub> and then form a product layer around the Ca(OH)<sub>2</sub>, which might hinder the un-reacted species from further diffusion and reaction [24]. From the viewpoint of the reaction mechanism, those reaction behaviors are similar to the accelerated carbonation for BOFS, on which calcium carbonate was coated, thus leading to surface coverage.

Therefore, the objective of the present study was to introduce the surface coverage model for determining the reaction kinetics of accelerated carbonation for BOFS in an RPB. The morphology

of the fresh and carbonated BOFS was characterized by SEM–XEDS before and after carbonation, which provided evidence of changes to the surface composition and molecular structure, for assessing the suitability of using the surface coverage model. In addition, comparisons of the reaction kinetics for carbonation with different alkaline wastes in various types of reactors (e.g., RPB, slurry reactor, and autoclave reactor) were evaluated by the surface coverage model. Furthermore, the effects of the key operating factors including reaction temperature, slurry flow rate, and rotation speed on the carbonation conversion of BOFS were investigated by both kinetic analysis and response surface methodology (RSM) to obtain the maximum carbonation conversion of BOFS in an RPB.

## 2. Materials and methods

### 2.1. Materials

A high-pressure cylinder of CO<sub>2</sub> gas with a volumetric concentration of 99.8% was provided by Ch'ing-Feng Gas Corp. (Taipei, Taiwan). The BOFS was supplied and ground by CHC Resources Corp. (Kaohsiung, Taiwan). Before each experiment, all BOFS samples were dried at 105 °C for 1 h to remove the adsorbed water, and then placed at room temperature in an airtight container. Since the composition of BOFS varied with batch to batch, it was required to prepare sufficient amount of slag powder with a size of less than 88 μm for composition homogeneity during the entire experiments.

Prior to evaluating the reaction kinetics of carbonation for BOFS in an RPB, the physico-chemical properties of fresh BOFS including true density, BET surface area, particle size distribution (PSD), and chemical composition were analyzed. The true density and BET surface area of the fresh BOFS was measured previously using a pycnometer (Micromeritics Accupyc1340) and low-temperature N<sub>2</sub> adsorption BET apparatus (Micromeritics ASAP2010), respectively, as reported in the literature [13]. The PSD of the fresh BOFS was obtained in accordance with the ISO 13320-1 method by laser diffraction (Malvern, Hydro 2000MU), with a measurement range between 0.02 and 2000 μm. The chemical composition of the fresh BOFS was measured by XRF (PW2430, Phillips), adapted from ASTM method C114 for concrete.

### 2.2. Experimental details

A schematic of the experimental set-up for accelerated carbonation of BOFS slurry in an RPB can be found in the paper describing our previous investigation [13]. In all experiments, BOFS slurry was prepared at a liquid-to-solid (L/S) ratio of 20:1 mL/g, and the carbonation of BOFS slurry was carried out with pure CO<sub>2</sub> gas (99.8 vol.%) in an RPB. The BOFS slurry was stirred constantly at 500 rpm in a repository and then pumped to the inner edge of the RPB for carbonation reaction. The BOFS slurry moved outward from the inner edge of RPB and was mixed counter-currently with

Download English Version:

<https://daneshyari.com/en/article/6691879>

Download Persian Version:

<https://daneshyari.com/article/6691879>

[Daneshyari.com](https://daneshyari.com)