Journal of Hazardous Materials 195 (2011) 107–114

Contents lists available at ScienceDirect

Journal of Hazardous Materials

journal homepage: www.elsevier.com/locate/jhazmat

CO_2 sequestration by carbonation of steelmaking slags in an autoclave reactor

E.-E. Chang[a], Shu-Yuan Pan[b], Yi-Hung Chen[c], Hsiao-Wen Chu[b], Chu-Fang Wang[d], Pen-Chi Chiang[b,*]

[a] Department of Biochemistry, Taipei Medical University, Taipei, Taiwan
[b] Graduate Institute of Environmental Engineering, National Taiwan University, No. 71 Chou-shan Rd., Taiwan
[c] Department of Chemical Engineering and Biotechnology, National Taipei University of Technology, Taiwan
[d] Biomedical Engineering and Environmental Sciences, National Tsing Hua University, Taiwan

ARTICLE INFO

Article history:
Received 13 May 2011
Received in revised form 3 August 2011
Accepted 4 August 2011
Available online 10 August 2011

Keywords:
Accelerated carbonation
Alkaline solid waste
Calcite
Surface coverage model
Life cycle assessment

ABSTRACT

Carbon dioxide (CO_2) sequestration experiments using the accelerated carbonation of three types of steelmaking slags, i.e., ultra-fine (UF) slag, fly-ash (FA) slag, and blended hydraulic slag cement (BHC), were performed in an autoclave reactor. The effects of reaction time, liquid-to-solid ratio (L/S), temperature, CO_2 pressure, and initial pH on CO_2 sequestration were evaluated. Two different CO_2 pressures were chosen: the normal condition (700 psig) and the supercritical condition (1300 psig). The carbonation conversion was determined quantitatively by using thermo-gravimetric analysis (TGA). The major factors that affected the conversion were reaction time (5 min to 12 h) and temperature (40–160 °C). The BHC was found to have the highest carbonation conversion of approximately 68%, corresponding to a capacity of 0.283 kg CO_2/kg BHC, in 12 h at 700 psig and 160 °C. In addition, the carbonation products were confirmed to be mainly in $CaCO_3$, which was determined by using scanning electron microscopy (SEM) and X-ray powder diffraction (XRD) to analyze samples before and after carbonation. Furthermore, reaction kinetics were expressed with a surface coverage model, and the carbon footprint of the developed technology in this investigation was calculated by a life cycle assessment (LCA).

1. Introduction

Carbon sequestration is a promising option for reducing carbon dioxide (CO_2) emissions and alleviating global warming. Both CO_2 captured from emission sources and subsequent transport of the captured CO_2 to isolated reservoirs are essential for carbon sequestration. Carbon capture is affected by environmental factors, capacity, and cost. Mineral sequestration is a method of carbon capture that accelerates the natural weathering of silicate minerals, allowing them to react with CO_2 to form stable products, carbonate minerals, and silica for further usage or disposal [1]. In addition, carbonation is an exothermal reaction; thus, energy consumption and costs may be limited by its inherent properties [1,2]. In all cases, the sequestration chemicals must provide base ions such as monovalent sodium and potassium, or divalent calcium and magnesium ions to neutralize the carbonic acid. Other carbonate-forming elements such as iron carbonates are not practical due to their unique and precious features [3].

In addition to controlling the reaction conditions, choosing suitable mineral feedstocks and properly designing the reactor are crucial to achieving high CO_2 sequestration efficiencies. One possible feedstock for CO_2 sequestration by accelerated carbonation is industrial solid waste, including steelmaking slags, combustion residues, and fly ash, which generally are alkaline and rich in calcium. The use of industrial waste is advantageous because of its low cost and widespread availability in industrial areas [4]. Interest in using industrial alkaline solid wastes as sources of calcium or magnesium oxide for CO_2 sequestration has arisen because these materials are readily available, cheap, and usually produced near large-emission sources of CO_2 [5]. In this study, carbonation reactions were performed primarily via the reaction of CO_2 with raw CaO-based materials, and calcium carbonate ($CaCO_3$) was observed to be the predominant carbonation product [6]. The use of this material simultaneously can reduce the amount of waste and neutralize a hazardous material.

The objectives of this study were to investigate the carbonation of several steelmaking slags, including ultra-fine (UF) slag, fly-ash (FA) slag, and blended hydraulic slag cement (BHC), in an autoclave reactor. The effects of the operational conditions, including the type of steelmaking slag, reaction time, liquid-to-solid ratio (L/S), temperature, CO_2 pressure, and initial pH, on the performance of the carbonation process were evaluated. In addition, reaction kinetics of the carbonation process were tested using a surface coverage model.

---

* Corresponding author. Tel.: +886 2 23622510, fax: +886 2 23661642.
E-mail address: pcchiang@ntu.edu.tw (P.-C. Chiang).

0304-3894/$ – see front matter © 2011 Elsevier B.V. All rights reserved.
doi:10.1016/j.jhazmat.2011.08.006

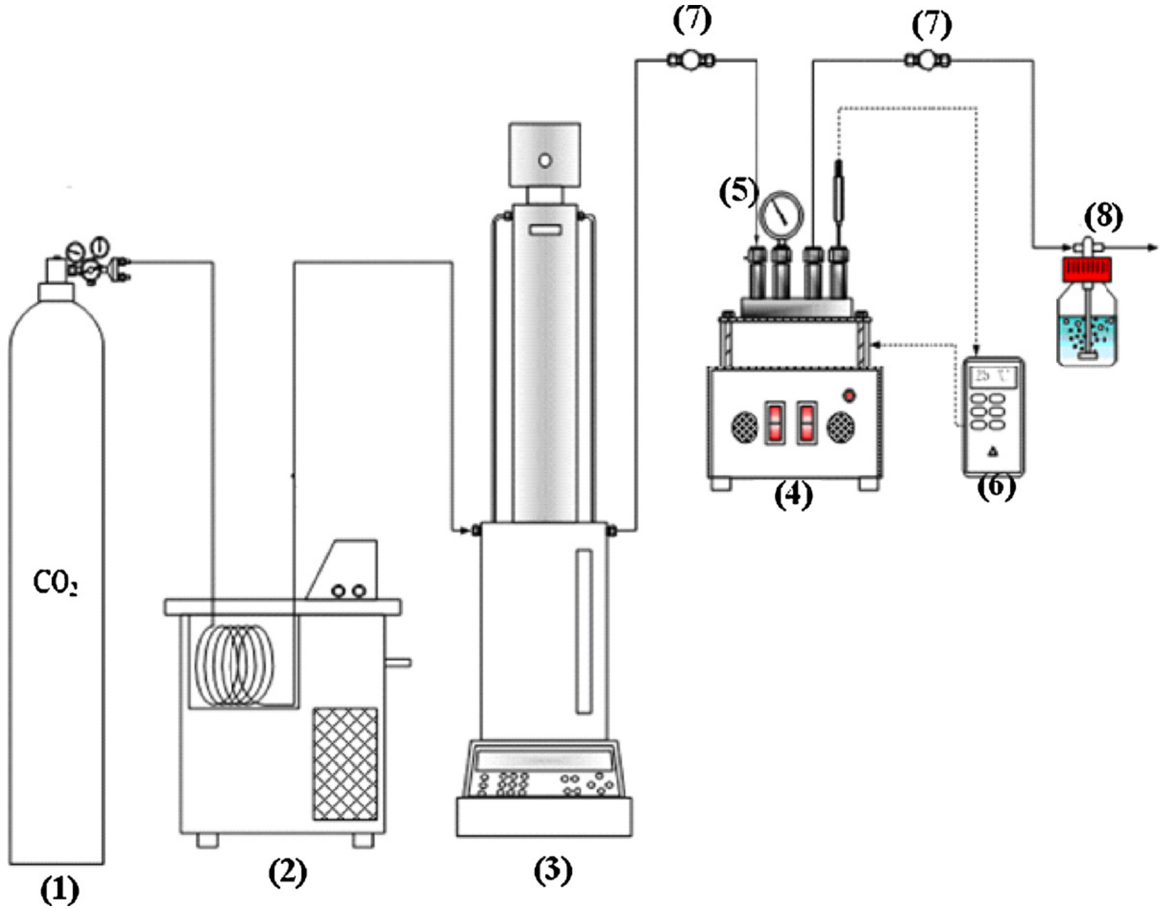

Fig. 1. Schematic diagram of the experimental set-up for the carbonation of steelmaking slag in an autoclave reactor. 1. CO_2 gas cylinder; 2. Circulating bath; 3. Syringe pump; 4. Magnetic stirrer and heater; 5. Reactor (autoclave); 6. Thermo couple; 7. Needle valve; 8. Vent to hood.

2. Materials and methods

2.1. Experiments

The aqueous carbonation of UF slag, FA slag, and BHC were conducted in an autoclave reactor that contained distilled water at a designated temperature of 40–160 °C. The UF slag, FA slag, and BHC with a diameter of approximately 1 cm were provided by the CHC Resources Corporation (Kaohsiung, Taiwan). All slags were ground and sieved to less than 44 μm for all experiments. The BHC contains an intimate and uniform blend of Portland cement and fine granulated blast furnace (BF) slag. The BHC used in this investigation is classified as CEM III/C (∼90% BF slag content) according to EN standards [7]. A schematic diagram demonstrating the carbonation of the steelmaking slag in an autoclave reactor is shown in Fig. 1. CO_2 was injected continuously into the reactor at a designated pressure and a constant flow rate.

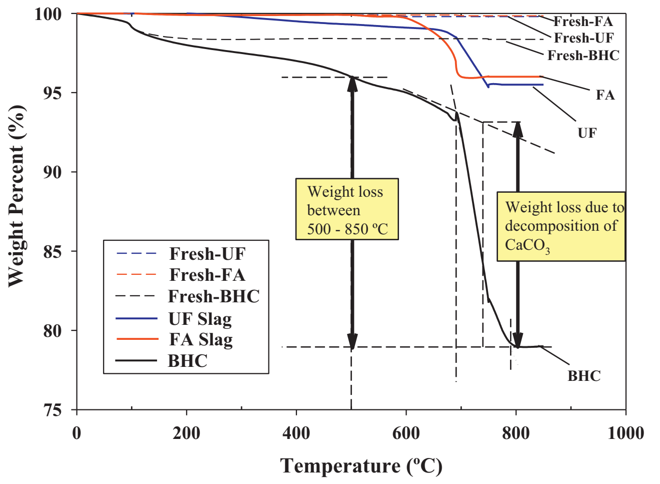

Fig. 2. TGA curves of fresh and carbonate of UF slag, FA slag, and BHC (Carbonation conditions: $P_{CO_2} = 650$ psig; $T = 60\,^{\circ}C$; $t = 1$ h; particle size < 44 μm; L/S = 10 mL g^{-1}).

The operational factors, including the reaction time (t), liquid-to-solid ratio (L/S), reaction temperature (T), CO_2 pressure (P), and initial pH, systematically were varied with the various feedstocks to minimize energy and chemical consumption. After the reaction, the samples of reacted slurry immediately were filtered through a PTFE membrane filter (Millipore, 45-μm pore size and 47 mm diameter), and then heated in an oven (105 °C) for use. The conversion of the carbonation products was determined quantitatively by thermo-gravimetric analysis (TGA) and qualitatively by X-ray diffraction (XRD) and scanning-electron microscopy (SEM).

2.2. Composition analysis

Prior to examining the capacity for CO_2 capture, the chemical compositions of steelmaking slags were characterized by inductively coupled plasma atomic emission spectroscopy (ICP-AES), after total digestion using aqua regia to dissolve the solid materials in the sample, by the chemistry analysis laboratory in the China Hi-ment Corporation. However, SiO_2 was dissolved further by using hydrofluoric acid with increasing temperatures and pressures in a