Measurement 97 (2017) 15–22

Contents lists available at ScienceDirect

Measurement

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

Steel-making slag for mineral sequestration of carbon dioxide by accelerated carbonation

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A R T I C L E I N F O

Article history:
Received 20 March 2015
Received in revised form 21 October 2016
Accepted 31 October 2016
Available online 1 November 2016

Keywords:
CO_2 sequestration
Mineral carbonation
Steel making slag
Operational parameters

A B S T R A C T

Mineral sequestration of carbon dioxide (CO_2) in alkaline waste materials is an innovative carbon capture and storage (sequestration) technology that can potentially be introduced as a finishing step for industries those generate alkaline solid by-products and emit flue gas CO_2. For example, steel making industry can capture its gaseous CO_2 emissions with the solid by-product steel slag, which is a rich source of alkaline oxides such as CaO and MgO. These oxides can be converted into stable carbonates at the presence of CO_2 through a series of reactions called mineral carbonation. However, mineral carbonation is not widely practised as a carbon capture and storage technique due to some of the barriers including slow kinetics of carbonation and the cost involved. Therefore, a growing number of research is currently focused on investigating ways and means to overcome the slow reaction kinetics while decreasing the associated costs in order to achieve industrial-level and economical sequestration. As a result, optimization of the operational parameters of carbonation reaction of alkaline wastes such as coal fly ash, municipal solid waste incinerator ash and steel-making slag are progressing rapidly. The present study examines the effect of three operational parameters (CO_2 pressure, temperature and water-to-solid ratio) on the rate and efficiency of the mineral carbonation reaction of steel-making slag at laboratory-scale. Carbonation tests for commercially available steel making slag samples were carried out at different initial CO_2 pressures from 1 MPa to 6 MPa, reaction temperatures from 20 °C to 80 °C, and water to slag mixing ratios from 0.25:1 to 3:1. Each test was allowed to run for 48 h in a continuously stirred tank reactor under CO_2 rich environment. The drop of initial CO_2 pressure with time was recorded until a constant pressure was achieved at the equilibrium. The results were analysed to estimate the quantity of CO_2 stored as carbonates within steel slag in each case. According to the results, the increased initial CO_2 pressure inside the chamber could shorten the time required to complete the carbonation reaction, but the effect on the overall stored CO_2 quantity was negligible. In contrast, with the increase of water-to-solid ratio, the overall carbonation continued to increase significantly. Of the tested reaction temperatures 50 °C was the most favourable for the carbonation to proceed at its optimum. Application of ideal gas law under optimum test conditions revealed a sequestration potential of 29.47 kg of CO_2 per tonne of steel slag. X-ray diffraction analysis and scanning electron microscopic imaging confirmed the formation of carbonate crystals in the slag samples during the carbonation reactions. The study confirmed the possibility of manipulating the reaction parameters to enhance the carbonation reaction for mineral CO_2 sequestration in alkaline solid by products.

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1. Introduction

The main cause of global warming is the increased concentrations of greenhouse gases (GHGs); CO_2, CH_4, N_2O and CO, in the atmosphere of which CO_2 contributes to two-thirds of the enhanced warming effect [1]. The possibility of great loss of environment and social disruption will increase when the concentration of CO_2 in the atmosphere reaches 550 ppm [2]. Therefore, the identification of feasible strategies to stabilize atmospheric CO_2 levels requires urgent attention. As a result, technologies for

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[1] Editor, Geomechanics and Geophysics for GeoEnergy and GeoResources: 4G Journal; Assistant Editor, Journal of Unconventional Oil and Gas Resources.

http://dx.doi.org/10.1016/j.measurement.2016.10.057
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CO_2 capture and storage (sequestration) are being studied extensively all over the world. Of the different sequestration methodologies, the most common and widely practiced process is the geological storage of CO_2 in reservoirs such as saline aquifers, depleted oil or gas wells and deep un-mineable coal seams [3]. In this process, captured CO_2 is injected into the reservoir in the form of super-critical fluid [4,5]. However, for large and point-source CO_2 emission sites such as fuel, heat, and power production facilities, onsite capture and mineralization is the most applicable technique [6]. Capture of the gaseous emissions at such industrial sites can be achieved in different ways including solvent capture, membrane separation technologies and sorbent technologies involving pressure or temperature swing processes [7]. After the capture, the carbon in the form of gaseous CO_2 is converted to stable and solid forms of carbonates through mineral sequestration.

The fundamental concept behind the mineral sequestration is the principle of natural rock weathering [8]. During weathering, Ca and Mg silicate mineral-bearing rock materials react naturally with the ambient CO_2 to transform into solid carbonates which are then stored in the ground permanently [9]. Similarly, primary minerals such as wollastonite ($CaSiO_3$), serpentine ($Mg_3Si_2O_5(OH)_{4)}$ and olivine (Mg_2SiO_4) can be carbonated to achieve a more efficient CO_2 storage process as they contain oxides of Ca and Mg at concentrations of 30–50% by weight [8]. Moreover, the abundance of these materials on earth offers the potential for their use as feed-stocks for sequestration [10,11].

The carbonation reaction for such silicate minerals can be written as in Eq. (1):

$$(Ca,Mg)SiO_3 + CO_2 \rightarrow (Ca,Mg)CO_3 + SiO_2 \tag{1}$$

The overall chemistry of the carbonation is exothermic and therefore optimization and process integration should allow its operation at zero or negative net energy input [6,12]. In addition, when compared to geological sequestration, mineral carbonation is unquestionably a permanent form of CO_2 fixation where the end products; carbonate minerals, are stable over millions of years [6,13]. Moreover, for countries where no suitable geological formations are known to exist, this is a feasible sequestration option to practise [14].

The use of alkalinity in industrial solid waste materials, such as coal fly ash, municipal solid waste incinerator (MSWI) ash and steel slag to chemically bind CO_2 is also a well-recognized mineral sequestration option due to its potential application as a finishing step in industrial plants [15,16]. The low cost and widespread availability of such waste materials are added benefits [15]. Compared to primary minerals, industrial solid wastes contain less available metal oxides for carbonation and thus show comparatively limited capacity for carbon sequestration [12]. However, these residues tend to be more reactive during carbonation due to their chemical instability, whereas in minerals the surface needs to be activated either by physical or chemical pre-treatment methods in order to increase the effective carbonation yield [15,17,18]. Furthermore, the use of solid wastes facilitates cost-effective sequestration and provides environmental benefits by helping to manage both CO_2 and the wastes [15].

Carbon sequestration using steel-making slag has the possibility to create a niche market for steel production facilities similar to coal combustion sites and municipal solid waste incineration plants [19]. Slag is a by-product from either the conversion of iron to steel in a basic oxygen furnace, or the melting of scrap to make steel in an electric arc furnace [20]. Depending on the process used, the carbon dioxide intensity of steel making can vary from 0.45 to 1.97 tCO_2/t crude steel [21]. From a tonne of steel, around 0.13–0.2 tonne of slag is produced and it is therefore a plentiful resource [22]. According to the US Geological Survey, the global production of steel making slag is around 200 Mt per year in recent years [23]. In addition, the increasing demand for steel will enable the availability of by-product steel slag to be even higher in the future. Slag is usually used as a supplementary cementitious material, which is a replacement for cement and other similar materials. Steel slag contains around 40% of CaO and therefore is considered to be an ideal material for mineral sequestration of CO_2 [9,15,22,24–26]. Unfortunately, the natural weathering of all of the feedstock materials discussed above, including steel making slag, proceeds at extremely slow rates (over geological time scales) which does not permit the achievement of economically viable carbon capture and storage [12,15,27]. Therefore, effective acceleration of mineral carbonation has become the main aim of research in the discipline of mineral sequestration.

Huijgen and Comans [15] were the first to conduct a systematic study of the reaction mechanisms of CO_2 sequestration by carbonation of an alkaline solid residue: steel-making slag. According to their findings, the diffusion of calcium towards the surface appeared to be the rate-determining step of the carbonation reaction. Therefore, Eloneva attempted to extract calcium selectively from slag prior to carbonation, which enables the production of a pure, and possibly marketable, calcium carbonate [28]. Several other researchers have worked on the effect of pre-leaching of Ca from the slag matrix on the efficiency of mineral sequestration [29,30]. Most research on steel slag carbonation to date has focussed on the acceleration of sequestration by extracting or activating the reactive Ca/Mg component from the silicate minerals present in slag. However, as identified in the very early stages of mineralization research, speeding up the carbonation chemistry kinetics also presents another alternative which can contribute to enhanced sequestration reaction. Therefore, it has been identified as one of the major challenges for large-scale mineralization of CO_2 [6].

The research literature presents various acceleration approaches to overcome the kinetics barrier of the carbonation reaction. These methodologies have led to the development of aqueous processes [17] to the current multi-stage process proposals [6]. However, the development of mineral sequestration remains at the laboratory demonstration scale and over twenty years of research and development work has not yet resulted in a mature technology that can be applied on a large scale in an economically viable way. Research on further enhancement of the mineral carbonation reaction may contribute to the development of cost-effective CO_2 sequestration.

Accelerated carbonation inside a closed reactor in a CO_2 rich environment is an interesting technology for the industrial mineralization of slag and other alkaline solid residues [15]. Soong et al. proposed this method as being viable [31]. According to Costa and Baciocchi [18,27], the rate and efficacy of such carbonation process of combustion residues depends on four main process-related parameters: (i) sample water content (or amount of water mixed), (ii) particle size, (iii) temperature and (iv) partial pressure of CO_2. This emphasises the importance of laboratory-scale parameter optimization studies before putting the concept of on-site mineral sequestration into practice. Therefore, the main aim of the present research is to investigate the effect of three process parameters (pressure, temperature and water-to-solid ratio) on the rate and efficacy of the mineral sequestration of steel-making slag at laboratory-scale.

2. Materials and research methods

This section discusses the important properties of slag used in the present study and the experimental procedure.

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