Journal of Cleaner Production xxx (2014) 1–10

Contents lists available at ScienceDirect

Journal of Cleaner Production

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

A laboratory-scale study of the aqueous mineral carbonation of coal fly ash for CO_2 sequestration

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ARTICLE INFO

Article history:
Received 11 November 2013
Received in revised form
28 January 2014
Accepted 1 March 2014
Available online xxx

Keywords:
Accelerated carbonation
Fly ash
CO_2 sequestration
Liquid-to-solid ratio
Reaction temperature

ABSTRACT

Mineral sequestration of waste materials provides a promising method for CO_2 sequestration, due to its potential as a finishing step in industries which produce CO_2 and alkaline solid by-products. However, a number of challenges in mineral carbonation that remain to be resolved, including overcoming the slow kinetics of mineral–fluid reactions, dealing with the large volume of source material required, and reducing the energy needed to hasten the carbonation process. In order to overcome the slow reaction kinetics, experiments on accelerated carbonation are being conducted worldwide. As a result, studies of the operational parameters of the carbonation reaction are progressing. The present study examined the effect of two operational parameters on the mineralization of Australian coal fly ashes for CO_2 sequestration at laboratory scale. In this study, carbonation tests were carried out for three Australian coal fly ash samples (S1, S2, S3) inside a continuously stirred reaction chamber. Different water-to-solid ratios (from 0.1 to 1) and reaction temperatures (20–80 °C) were tested under a moderate initial CO_2 gas pressure of 3 MPa, and the pressure drop due to carbonation with time was recorded until a constant pressure was achieved at the end of each test. The quantity of CO_2 stored in each test was estimated by applying ideal gas law to the test conditions. The formation of carbonates during testing was confirmed by performing micro-structural analysis using scanning electron microscopy. According to the results, a 0.2–0.3 water-to-solid mix ratio recorded the highest sequestration potential for all three fly ashes, and was identified as the optimum for mineralization. The increase of reaction temperature resulted in a faster rate of initial CO_2 transfer into the fly ash material but did not have a significant impact on the overall sequestration. Of the three tested ashes, S3 ash sample showed the highest sequestration potential of 27.05 kg of CO_2 per ton of fly ash under test conditions. The results confirm the possibility of manipulating the water-to-solid mix ratio and the reaction temperature to enhance the carbonation reaction for mineral CO_2 sequestration.

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1. Introduction

The accelerated increase of global temperatures in the past 60 years is attributed largely to anthropogenic greenhouse gas emissions (Tett et al., 1999). The atmospheric concentration of CO_2, the major greenhouse gas, increased by 39% from the pre-industrial era to 2010 (Lal, 2010). There are three main strategies for lowering the atmospheric concentration of CO_2: (i) reducing global energy use; (ii) developing low- or no-carbon fuel; and (iii) sequestering CO_2 from point sources or from the atmosphere using natural and engineering techniques (Schrag, 2007). The need for carbon sequestration is becoming crucial among all available carbon mitigation options due to the large point-source emission of CO_2 during coal-fueled power generation. Coal combustion alone accounts for about 20% of global greenhouse gas emissions (CCES, 2011), and coal produces the most CO_2 per unit energy of all fossil fuels. However, the world's reliance on coal-based power will continue to grow, irrespective of the improvements achieved in efficiency and the growth of nuclear, solar, wind, and biomass energy sources (Schrag, 2007). Therefore, for the long-term viability of the coal-based power industry, the development of carbon capture and storage technologies at coal-fired power plants requires urgent attention (Geman and Gronewold, 2009).

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http://dx.doi.org/10.1016/j.jclepro.2014.03.005
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Please cite this article in press as: Ukwattage, N.L., et al., A laboratory-scale study of the aqueous mineral carbonation of coal fly ash for CO_2 sequestration, Journal of Cleaner Production (2014), http://dx.doi.org/10.1016/j.jclepro.2014.03.005

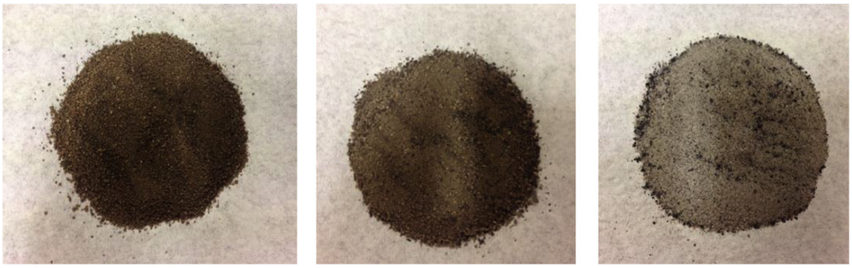
S1 S2 S3

Fig. 1. Dry samples of Latrobe Valley fly ashes.

Carbon capture at coal combustion sites can be achieved in many ways. The research literature proposes membrane separation technologies, sorbent technologies involving pressure or temperature swing processes, and the use of solvents as capture technologies for CO_2 from the flue gas stream (Reddy et al., 2010). Storage of captured industrial carbon is mainly done through mineral carbonation (Mazzotti et al., 2005). Mineral carbonation is the transformation of CO_2 into calcium, magnesium and other forms of stable carbonates (Eloneva et al., 2012). Coal combustion fly ash contains alkaline oxides such as CaO and MgO which can be carbonated in the presence of CO_2. This process ideally represents the chemical weathering of alkaline earth minerals in the presence of atmospheric CO_2 dissolved in rain water (Lackner et al., 1995). Similarly, alkaline solid waste materials from industries, such as coal combustion fly ash (Reddy et al., 1986), municipal solid waste incineration (MSWI) bottom ash (Rendek et al., 2006), steel slag (Huijgen et al., 2005; Said et al., 2013) and oil shale solid residues (Essington, 1991), have the potential to be used as materials for CO_2 storage. The physical and chemical properties of coal fly ash depend basically on the mineralogy of the parental coal and type of coal burning process (Jo et al., 2012). However, most of the coal fly ashes produced in the world are alkaline and contain considerable amounts of CaO (Soong et al., 2006).

The carbonation reaction of coal fly ash under ambient conditions proceeds at very slow rates (on geological time scales) and its economic feasibility for widespread application is not fully known (Huntzinger et al., 2009). The enhancement of the natural mineralization of fly ash through aqueous carbonation was first studied by Reddy et al. (1986). Thereafter, a number of laboratory-scale research studies has been reported on coal fly ash carbonation (Back et al., 2008; Montes-Hernandez et al., 2009; Uliasz-Bocheńczyk et al., 2009). A pilot scale study was conducted by Reddy et al. (2010), who observed that the moisture content of flue gas reacting with fly ash particles was the most important factor in mineralization (Reddy et al., 2010). Several other authors have used brine solution with coal fly ash to increase CO_2 retention in fly ash (Soong et al., 2006; Nyambura et al., 2011). Brine acts as an additional Ca and Mg source to enhance the reaction efficiency of carbonation (Soong et al., 2006). Soong et al. (2006) proposed the use of mineralogical sequestration inside a controlled reactor as a viable approach to reduce CO_2 emissions.

According to Costa et al. (2007), the rate and the efficacy of the aqueous mineral carbonation of combustion residues depends on four main process-related parameters: (i) sample water content (ii) particle size (iii) temperature and (iv) partial pressure of CO_2. The effects of these parameters need to be examined extensively for the design of carbon capture and storage facilities at industrial sites (Reddy et al., 2010). Laboratory-scale studies of parameter optimization and carbonation process assessment are therefore necessary, before putting the concept of on-site mineral sequestration into practice. Hence, the main objective of the present study was to investigate the effect of two reaction parameters (fly ash–water content and temperature) on the mineral CO_2 sequestration of three types of Australian brown coal fly ashes.

Section 2 below introduces the fly ash materials used in the study, the experimental apparatus and the testing procedure. The following sections describe the results and discuss them.

2. Fly ash material characterization

The fly ash samples for the tests were collected from three power plants located in the Latrobe Valley, Victoria, Australia. Victorian brown coal is the parental material for all of these ashes. The ash was collected from the waiting ponds located at the power plant sites and the bulk material was sieved with a 1.18 mm sieve to obtain uniform-sized particles for testing. The samples were then oven dried at 105 °C to a constant weight, to remove all the moisture in the samples. Fig. 1 shows the state of the dry fly ashes before adding water for testing.

For the characterization of fly ash, morphological analysis was done using a scanning electron microscope Nova Nano 450 which is equipped with a field emission gun. According to the images, the Latrobe Valley ash particles are generally irregular and range from sub-angular through to rounded, sometimes spherical. The mineralogy of the fly ashes was determined by X-ray diffraction spectroscopy using a PANalytical Empyrean diffractometer coupled with Cu-Kα radiation at 45 kV and 40 mA. Elemental analysis was conducted using the X-ray fluorescence method and the results are shown in Table 1 below.

The Brunauer, Emmett and Teller (BET) single point method with N_2 was used to measure the surface area of the particles. The absolute and bulk densities were determined using a Micromeritics Accupyc 1330 helium pycnometer and Micromeritics Autopore 9520 mercury porosimeter, respectively.

3. The experimental system and the carbonation test procedure

Carbonation tests were carried out inside continuously stirred autoclave reactors connected to a plumbing system. Fig. 2 shows a schematic diagram of the experimental apparatus.

Table 1
Chemical composition of Latrobe Valley ashes.

	S1	S2	S3
SiO_2	36	54	13
Al_2O_3	2	9.5	2.1
Fe_2O_3	16	3	23
K_2O	0.4	1.2	0.4
MgO	10.7	5.5	13
Na_2O	4.2	2.1	9
CaO	12.5	18.5	24.8
SO_3	12	2.5	12.8

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