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# Investigation of the potential of coal combustion fly ash for mineral sequestration of CO<sub>2</sub> by accelerated carbonation



N.L. Ukwattage a, P.G. Ranjith a,\*, S.H. Wang b

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#### ABSTRACT

Mineral carbonation of alkaline waste materials is being studied extensively for its potential as a way of reducing the increased level of  $CO_2$  in the atmosphere. Carbonation converts  $CO_2$  into minerals which are stable over geological time scales. This process occurs naturally but slowly, and needs to be accelerated to offset the present rate of emissions from power plants and other emission sources. The present study attempts to identify the potential of coal fly ash as a source for carbon storage (sequestration) through ex-situ accelerated mineral carbonation. In the study, two operational parameters that could affect the reaction process were tested to investigate their effect on mineralization. Coal fly ash was mixed with water to different water-to-solid ratios and samples were carbonated in a pressure vessel at different initial  $CO_2$  pressures. Temperature was kept constant at 40 °C. According to the results, one ton of Hazelwood fly ash could sequester 7.66 kg of  $CO_2$ . The pressure of  $CO_2$  inside the vessel has an effect on the rate of  $CO_2$  uptake and the water-to-solid ratio affects the weight gain after the carbonation of fly ash. The results confirm the possibility of the manipulation of process parameters in enhancing the carbonation reaction

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#### 1. Introduction

The global atmospheric concentration of carbon dioxide has increased from a pre-industrial value of about 280 ppm—379 ppm in 2005 [1]. The CO<sub>2</sub> level in the atmosphere needs to be maintained at levels not exceeding 550 ppm and this requires global emission reductions of 50—60% by 2050 relative to the current trend [2]. Potential ways to reduce CO<sub>2</sub> emissions include substituting biofuel, hydrogen, or electric power for hydrocarbons in the transportation and electric power generation sectors, substituting natural gas for coal in electric power generation and capturing and sequestering carbon dioxide produced by combustion [3]. Carbon capture and storage (sequestration) technology is being researched widely all over the globe, and the scientific community is finding ways to optimize its cost-effectiveness and the rate of CO<sub>2</sub> removal from the atmosphere.

E-mail address: ranjith.pg@monash.edu (P.G. Ranjith).

URL: http://eng.monash.edu.au/civil/about/people/profile/ranjithp, http://users.
monash.edu.au/~ranjithp, http://www.monash.edu.au/research/profiles/profile.
html?sid=5502&pid=3763

Mineral sequestration of CO<sub>2</sub> using alkaline materials is another novel CO<sub>2</sub> sequestration concept which provides a beneficial use for certain rapidly-accumulating waste materials [4]. Solid wastes produced during coal combustion (fly ash and bottom ash), municipal solid waste incineration (fly ash, bottom ash and air pollution control residues) and by-products such as steel slag are some of the residues which are being evaluated for their potential for CO<sub>2</sub> sequestration [4–7]. In mineral sequestration, atmospheric CO<sub>2</sub> is taken up by alkaline oxides and hydroxides present in the candidate materials to form thermodynamically stable forms of carbonates [4,8]. Basically, mineral carbonation is an acid base reaction where an acid formed by the dissolution of CO<sub>2</sub> in water (H<sub>2</sub>CO<sub>3</sub>) is neutralised by a solid base (alkaline mineral). It is one of the steps in natural rock weathering which comprises a complex series of chemical and mineralogical transformations [9].

Under ambient conditions Ca- and Mg-bearing silicate minerals are naturally carbonated to form Ca and Mg carbonates [10]. In this process, carbonic acid (H<sub>2</sub>CO<sub>3</sub>) resulting from the interaction of atmospheric CO<sub>2</sub> with rain water converts the alkaline earth materials into carbonate minerals [5]. The sequence of reactions involved in the carbonation mechanism can be written as follows:

$$H_2O + CO_2 \rightarrow H_2CO_3 \tag{1}$$

<sup>&</sup>lt;sup>a</sup> Deep Earth Energy Lab, Bld. 60, Monash University, Victoria 3800, Australia

<sup>&</sup>lt;sup>b</sup> College of Resource and Civil Engineering, Northeastern University, Shenyang, 110819, PR China

<sup>\*</sup> Corresponding author. iEnergy Initiative — Innovative Deep Earth Energy Research Centre, Bld. 60, Clayton Campus, Monash University, VIC 3800, Australia. Tel.: +61 3 99054982; fax: +61 3 99054944.

$$H_2CO_3 \rightarrow H^+ + HCO_3^- \rightarrow 2H^+ + CO_3^{-2}$$
 (2)

$$CaO + H_2O \rightarrow Ca(OH)_2 \tag{3}$$

$$Ca(OH)_2 \rightarrow Ca^{2+} + OH^-$$
 (4)

$$Ca^{2+} + CO_3^{2-} \rightarrow CaCO_3 \tag{5}$$

Coal combustion fly ash contains oxides such as CaO and MgO that can be converted to carbonates in the presence of  $CO_2$  through the above sequence of reactions. Coal fly ash is an abundant solid waste, production of which is expected to increase as a result of the world's increasing reliance on coal-fired power generation over the next few decades (www.iea-coal.org). Although there are many potential uses of fly ash including cement production, road subbase construction, mine reclamation and as aggregate substitute material, it is presently identified as a resource that is significantly under-utilized [11]. In 2000, only 20% of the total global coal fly ash production (600 million tonnes) was used beneficially, mostly in the cement industry [12]. Each year a large mass of fly ash is collected in storage ponds, which emphasises the need for the investigation of large-scale applications for this waste product.

Over the last few decades fly ash has attracted attention as a potential source in the mineral sequestration process, which involves the capture and storage of atmospheric C in alkaline materials [5,13,14]. The utilization of fly ash to capture and fix  $CO_2$  has an added advantage, since it assists in the management of this potentially hazardous waste. Since the carbonation process helps to alter the chemical stability of the fly ash, the leachability of heavy metals such as Pb and Zn tends to reduce after carbonation [15,16]. This allows the safe disposal of carbonated fly ash into landfills or dumpsites.

However, this mineral carbonation reaction proceeds at an extremely slow rate under natural ambient conditions, to the extent that it limits the achievement of any economic benefits of CO<sub>2</sub> sequestration [17]. Therefore, researchers have been experimenting on possible pathways for accelerating the mineralization reaction. Ex-situ carbonation of alkaline solid wastes inside a reactor with enhanced reaction conditions has been found to be a possible solution [18]. Reddy and colleagues first studied aqueous mineral carbonation by bubbling CO<sub>2</sub> through alkaline solid waste slurry [19]. Subsequently, a number of researchers have focussed on the accelerated carbonation of various alkaline solid wastes [5,20-22]. In these studies some researchers tried to identify the parameters which govern the rate and efficiency of the carbonation reaction [7,13,14]. Accordingly, the reaction has been found to be affected by the inherent properties of the candidate material and the conditions supplied inside the reactor [23].

In accelerated carbonation the mixture is typically carbonated in a gaseous CO<sub>2</sub>-rich environment [7]. The partial pressure of CO<sub>2</sub> inside the reactor is one of the parameters that influence the carbonation process [14]. In addition, researchers have been experimenting to optimize some other process conditions such as the reaction temperature, moisture content of the mixture (ratio of mixing solid with water), contact time with the gas and the provision of proper mixing of the gas and the material in order to achieve an efficient mineral carbonation process [7]. The present paper focuses on the optimisation of two process parameters to achieve the maximum carbonation rate and efficiency of coal combustion fly ash from a large power plant in Australia. The objectives of the study are to evaluate the potential for CO<sub>2</sub> sequestration of coal fly ash and to study the effect of CO2 pressure and solid-to-liquid ratio on the rate and the efficiency of CO2 sequestration.

#### 2. Materials and methods

This section discusses the important properties of the fly ash used, the procedure of accelerated carbonation and the methods used in the analysis of results.

#### 2.1. Fly ash

Fly ash samples for the present study were collected from the Hazelwood power plant, Latrobe Valley, Victoria. This ash is generated by the combustion of Victorian brown coal and collected by electro-static precipitation. After capture from the flue gas stream, the ash is dumped in collection ponds and weathered there until it is transported for use. The powder used in the study came from these collection ponds and was dark brown in colour. The ash samples initially contained large clods of ash and needed to be broken into individual particles for testing.

According to sieve analysis of particle size distribution, the average particle diameter is around 0.9 mm. The true density of the particles is 1.875 gcm<sup>-3</sup>. Energy-dispersive X-ray spectroscopy analysis of the chemical composition shows that Hazelwood fly ash is rich in Ca (39.8% w/w) and Fe (34.3% w/w). In addition, it contains 8.7% Si, 7.3% Mg, 7.2% S and 2.7% Al. The high Ca content in the ash matrix contributes to the alkaline pH of 10.3 before carbonation.

#### 2.2. Accelerated carbonation for CO<sub>2</sub> sequestration

The aqueous mineral carbonation reaction was carried out inside a pressure chamber of 2 L capacity. The chamber is connected to a  $\mathrm{CO}_2$  gas inlet line which provides it with the gas from a cylinder which is pressurized by the pump to the required pressure. The gas outlet line allows the extraction of all the gas after the experiment. After filling the chamber with the mixture to be carbonated, it can be isolated from the gas lines using the valves fixed close to the chamber on both lines. The chamber is also equipped with a pressure transducer which is connected to the data acquisition system and records the chamber inside pressure at set time intervals. The temperature inside the chamber is regulated by a heating element and is recorded by a thermocouple. Fig. 1 shows the components of the experimental set-up used in the study.

In order to investigate the effect of two operational parameters, the pressure of the  $CO_2$  and the solid-to-liquid ratio, on the mineralization of fly ash, a series of experiments was carried out,

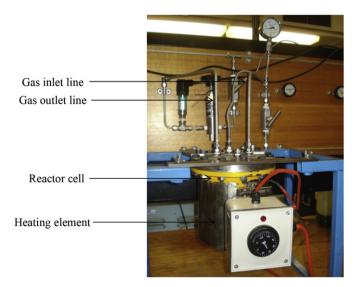


Fig. 1. Experimental set-up used for carbonation.

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