



Study on mineral carbonation of heat activated lizardite at pilot and laboratory scale



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ABSTRACT

This investigation examines the experimental data obtained from a 30 L pilot batch reactor, used for single stage aqueous carbonation and for the dissolution of heat activated lizardite and compares this data to the laboratory scale data obtained under similar reaction conditions. Two mineral feeds, heat activated lizardite produced in large quantities in a gas fired industrial kiln (“P.H.A”) and heat activated lizardite sample synthesised in a laboratory kiln (“L.H.A”) were characterised and their reactivity was compared.

Characterisation of the two heat activated samples showed that the P.H.A material contained relatively high concentrations of crystalline phases and a lower proportion of amorphous magnesium silicate, as well as a higher average particle size compared to L.H.A. These properties resulted in a reduction in magnesite yield and extent of magnesium extraction when P.H.A samples were used for carbonation and dissolution experiments, compared to the results obtained with L.H.A. as feed.

The results of aqueous carbonation experiments in the pilot and the laboratory scale reactors using P.H.A exhibited less than 5% variation in repeat experiments. Results indicate that the magnesite yield obtained in the pilot batch reactor was $35 \pm 2\%$ higher than that produced in the laboratory scale reactor. The higher yield was attributed to improved mixing in the pilot batch reactor compared to the laboratory scale reactor and consequent removal of the precipitated phases on the surface of heat activated particles in the pilot reactor.

The yield of magnesite in mineral carbonation experiments using demineralised water and tap water was almost identical. Undertaking reaction in 1 M NaCl solution did not have a significant effect, but adding 0.64 M sodium bicarbonate had a notable effect on magnesite yield. Low pressure, low temperature dissolution experiments using L.H.A sample displayed similar rates of magnesium extraction in the pilot and the laboratory scale reactors.

1. Introduction

Global warming, a result of the accumulation of greenhouse gases in the atmosphere, is widely considered to be one of the most serious of all environmental problems. Motivated by the desire to reduce the impact of global warming, the emissions of CO₂ into the atmosphere must be mitigated by utilising various available options including CO₂ capture, utilization and storage (CCUS) [1].

Studies on ex-situ mineral carbonation, a technology being developed to capture, utilize and store carbon dioxide in a safe and permanent form, was initially conceived by Seifritz in 1990 [2] and developed further by Lackner et al. [3]. However, research activities in this field moderated, following the conclusions published by the IPCC in 2005

[1] which asserted that mineral carbonation (especially heat activation of serpentine) was an energetically and economically unfavourable process compared to conventional underground storage. Since 2008 and due to recognition of erroneous assumptions made in the economic and energy consumption analysis of serpentine heat activation [4,5], work in this area has been reinvigorated and mineral carbonation is once again being developed as a technology which could be exploited for large-scale CO₂ storage and utilization [6].

The process mimics naturally occurring rock weathering and involves reacting mineral silicates with carbon dioxide to produce stable carbonates [3]. Process strategies for mineral carbonation can be divided in to two main groups. Direct carbonation (DC) is the simplest application of the technique, where magnesium silicate minerals (i.e.

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lizardite, antigorite, olivine, etc.) are carbonated in a single step process. Various temperatures, pressures and reagents have been used in single stage carbonation to increase carbonation yield. O'Connor et al. [7] and Chizmeshya et al. [8], performed single stage carbonation experiments under different temperatures and partial pressures of CO₂, with and without NaCl and NaHCO₃. They found that the presence of these reagents, typically 1.0 M NaCl and 0.64 M NaHCO₃, enhanced the reaction rate and carbonation extent of olivine and serpentine compared to experiments at the same temperature and P_{CO2} conducted in the absence of these reagents.

Indirect carbonation (IC) consists of two or more separate stages of reaction involving the extraction of magnesium (Mg²⁺) from the feedstock before reacting the leached cations with CO₂ to form carbonate precipitates in subsequent step(s) [9–11].

In spite of advances in state-of-the-art process simulation, predicting the performance behaviour of complex systems relevant to process scale-up is difficult. As a result, the construction and operation of a research pilot plant and the analysis of data generated therein is a necessary first step in the commercialisation of ex-situ mineral carbonation. Only a few researchers have published studies on pilot scale of mineral carbonation [12–14].

The mineral carbonation research pilot plant constructed by Mineral Carbonation international (MCI) at The University of Newcastle, Australia aims to transform captured CO₂ emissions into carbonates and silicates for potential use in construction materials such as cements, bricks and plasterboard. Our previous work [15] showed that the by-products of mineral carbonation processes (especially two stage process) were enriched with amorphous silica which exhibited some pozzolanic activity. Sanna et al. [16] showed that the residual silica by-product of the mineral carbonation process could replace up to 10 wt% of cement and resulted in higher compressive strength compare to control cubes. Imbabi et al. [17] published a patent on the applications of nesquehonite for construction purposes. They claimed that it might have a number of different useful applications such as mortar or render, board products, concrete, construction blocks, sound insulator, thermal insulator or fire retardant. The products synthesised in the pilot plant were also used in rheological studies of the carbonated slurry, as well as in assessing the feasibility of developing cement replacement materials for use in mortars and as fillers in concrete from the materials generated during mineral carbonation.

Assessment of the repeatability of the pilot plant operation and establishing the consistency (or otherwise) between pilot and laboratory data is essential. The aim of this work was to study the reactivity of heat activated lizardite produced in large quantities and subsequently carbonated in a pilot plant. Experimental data on single stage aqueous carbonation and dissolution of heat activated lizardite in a pilot batch reactor was gathered and these results compared to those obtained in the laboratory scale experiments under the same or similar reaction conditions.

2. Materials, methods and samples characterisation

2.1. Raw and heat activated lizardite

Raw lizardite was collected from the Great Serpentine Belt in New South Wales, Australia. To prepare laboratory heat activated lizardite (L.H.A), crushed lizardite samples were initially wet milled in a 2 L stainless steel ball mill (MSK-SFM-14, MTI Corporation U.S.A) then wet sieved to pass through a standard 75 µm sieve. The sieved particles were then dried at 110 °C for 24 h, heat activated in different batches and thoroughly remixed. In each heat activation run, 200 g of raw lizardite (< 75 µm) was heat activated in an electrically heated stainless steel rotary kiln (RSR 80/500/11, Nabertherm, Germany) at 630 °C for 4 h.

To produce heat activated lizardite in large quantities for use in the pilot plant (P.H.A), powdered lizardite was fed to a rotary kiln. Heat

activation was conducted in an indirect, natural-gas fired industrial rotary kiln with 3.6 m heated zone length and 0.4 m tube diameter. The operation conditions of the industrial rotary kiln were selected to achieve a similar temperature and residence time to heat activation in the laboratory kiln.

2.2. Single stage aqueous mineral carbonation in the laboratory scale reactor

Experiments were performed in a high pressure, 600 mL stainless steel batch reactor (series 4540, Parr Company, U.S.A) under conditions developed and described by ARC researchers [7]. 56 g heat activated lizardite was mixed with 300 mL distilled water and 16.3 g sodium bicarbonate solution (0.64 M) which buffered the pH of the solution to facilitate both extraction and precipitation processes. The slurry, with a 15 wt% solid to liquid ratio, was charged into the reactor vessel which was limited to operate at a maximum pressure and temperature of 200 bar and 250 °C respectively. An O ring (model FKM 2–7, Parr Company, U.S.A) was used to seal the reactor for operating temperatures to 225 °C, and a rupture disc was used to vent the reactor at 200 bar safely, if required. A stirrer with magnetic coupling, with two sets of four stirrer blades pitched at 45° each was used to agitate the slurry. While stirring was in progress, the reactor was purged with CO₂ to remove residual air. The reactor was then heated to 150 °C. An electrical jacket heater, operating at a maximum temperature of 300 °C, was used to heat the slurry in the reactor. Reactor temperature was controlled using a PID controller (Parr) with feedback from a thermocouple located in the centre of the slurry mixture. Water running through a coil located in the reactor was used to cool the reactor. The reactor was pressurized to 150 bar CO₂ through a dip tube, via a CO₂ pressure booster connected to a CO₂ cylinder. Aliquots of the reacting slurry were collected using a 5 mL sampler, using a dip tube. Each sample was removed from the reactor due to the pressure difference between the reactor and sampler. To analyse the supernatant solution obtained from the sampler, an aliquot of 1 mL of the supernatant solution was taken, passed through a 200 nm syringe filter and then diluted with 2% nitric acid (1:1 vol%) to avoid precipitation. The remaining solid was rinsed three times with distilled water to remove sodium bicarbonate, dried at 110 °C for 24 h then analysed. At the end of each experiment, the reactor was cooled down to 30 °C, de-pressurised through a vent line and the slurry removed from the reactor. Due to the risk of corrosion, a consequence of the effect of chloride (even at trace levels) in high pressure and temperature conditions, the reaction mixture was free of any chloride (below 1 ppm). Consequently, experiments with additives such as sodium chloride, sea water or even tap water were not performed in this laboratory-scale batch reactor.

2.3. Single stage aqueous mineral carbonation in the MCI pilot batch reactor

Experiments in the research pilot plant were performed in a 30 L batch reactor under conditions developed and described by ARC researchers [7]. The reactor (a custom designed vessel, Estanit GmbH, Germany) was designed to operate at pressures up to 250 bar and temperatures up to 200 °C. The reaction vessel, piping and further product wetted surfaces in the high pressure, high temperature environment are made of Hastelloy, an alloy which is resistant to corrosion by chloride, thus enabling experiments with chloride in solution to be undertaken. The slurry of 20 L water, 3 kg of heat activated lizardite (P.H.A) with/without additives i.e. 0.64 M NaHCO₃ and 1 M NaCl was charged into the reactor through a feed funnel located on the top of the reactor while the mixing impeller was operating. The reactor is equipped with a hollow shaft impeller having six 45° pitched blades and four internal baffle plates to enhance mass transfer and mixing performance. The reactor was heated to 150 °C using an electrical heater jacket and held at this temperature during experiments. Tap water was used to cool the reactor and magnetic stirrer coupling. After reaching

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