



## Dynamics of carbon dioxide uptake in chrysotile mining residues – *Effect of mineralogy and liquid saturation*

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### ABSTRACT

Carbonation of chrysotile mining residues (CMR) was studied to expose the role of residue size and mineralogy, gas composition, liquid saturation and watering schemes in saturation-controlled porous beds. CO<sub>2</sub> uptake dynamics and evolution of carbonating residue were monitored *in situ* in terms of gaseous CO<sub>2</sub> absorbed and relative humidity, bed liquid saturation, electrical conductivity, pore-water pH, and pressure drop. CO<sub>2</sub> uptake was contributed both by facile carbonation of chrysotile fines and “domestic” brucite, and slow-paced carbonation of coarser magnesium silicate particles. Chrysotile carbonation was a function of fiber length while inhibited lizardite carbonation was indirectly observed. A CO<sub>2</sub>-lean carbonation regime was identified where CO<sub>2</sub> uptake increased *linearly* with the CO<sub>2</sub> fraction. This enabled extrapolating at very low CO<sub>2</sub> gas contents to assess CMR carbonation under natural atmospheric conditions. Reduction of saturation and backmixing in the liquid proved effective for the proliferation of Mg-supersaturated zones to enhance carbonation. Maintaining low liquid saturation *via* periodic liquid additions translated into improved carbonation because of inhibition of silanol-polymerization passivation. Partial pore saturation proved effective in stimulating carbonation, both in flow-through and in diffusive modes, especially at lower CO<sub>2</sub> fractions thus foreseeing implementation of useful optimization strategies to enhance ambient carbonation of CMR heaps.

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## 1. Introduction

Research and development initiatives are underway at a global scale with an objective of deploying reliable CO<sub>2</sub> capture and storage (CCS) solutions to help curb anthropogenic carbon dioxide emissions (O'Connor et al., 2001a; Park et al., 2003; Carey et al., 2004; Jia et al., 2004; Giammar et al., 2005; Dufaud et al., 2009; Prigiobbe et al., 2009; Garcia et al., 2010; Larachi et al., 2010). One effective way to curtail emissions involves trapping highly-concentrated CO<sub>2</sub> streams in suitable geological formations such as ultrabasic rocks (Goff and Lackner, 1998), sedimentary formations and saline aquifers (Bachu and Adams, 2003; Kaszuba et al., 2003), and depleted oil or gas reservoirs (Holloway, 1997; Wildenborg and Lokhorst, 2005) where physicochemical carbonation processes are controlled by the high pressure and high-temperature conditions prevailing underground (Felmy et al., 2012). At the opposite end of the spectrum, above-ground ultramafic mining tailings, exposed to meteoric weathering, have been recently recognized to carbonate and crust under ambient conditions in the presence of meteoric

water and atmospheric CO<sub>2</sub> (Beaudoin et al., 2008; Wilson et al., 2009; Bea et al., 2012). Consequently, harnessing such residues to sequester significant quantities of CO<sub>2</sub> directly from the atmosphere could open up new attractive avenues to add to the current CCS portfolio.

Mine tailings often consist of vast stacks containing up to 20 wt.% Mg mainly in the form of magnesium silicates, which are able to bind with CO<sub>2</sub> molecules thus removing CO<sub>2</sub> from the atmosphere by chemical storage in an array of solid carbonates (Seifritz, 1990; Lackner et al., 1995; Oelkers et al., 2008) *via* mineral sequestration (Lackner et al., 1995; O'Connor et al., 2000; Andreani et al., 2009; Teir et al., 2009). Mine wastes serving as an above-ground CCS solution may offer obvious economic advantages. The approach is anticipated to require only simple and low-cost technology, unlike its underground CCS option, especially for exploiting areas where the already-mined residues lend themselves to carbonation. One example of such mine tailings is the serpentinized waste residues of Black Lake (Thetford Mines, Québec) which extend over an area of 1.7 km<sup>2</sup> and have a height of 130 m, providing a maximum storage capacity of *ca.* 700 megatons of carbon dioxide (Pronost et al., 2012). To give perspective to this figure, such a quantity would be tantamount to emitting 660 kilotons CO<sub>2</sub> per year burning petroleum carbonaceous residues in thirty 500 MW power plants over a 35-year horizon.

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In engineering terms, mining residues undergoing carbonation under ambient atmospheric conditions are merely gigantic reactors that entail less cost than processes purposely designed for atmospheric CO<sub>2</sub> capture which, on account of the low CO<sub>2</sub> concentrations to be intercepted, are capital and energy intensive (Ranjan and Herzog, 2011; Simon et al., 2011). Moreover, engineered reactors often require expensive chemical reagents, pre/post-treatments, pumping devices operating across the entire process and reagents regeneration at the loop end (Lackner et al., 1999; Baciocchi et al., 2006; Zeman, 2007; Storaloff et al., 2008; Mahmoudkani et al., 2009; Nikulshina et al., 2009). In this respect, contemplating the requirements from pre/post-combustion mineral carbonation projects is far more challenging as these rely on high temperature/pressure and chemical reagents to stimulate the naturally slow reaction (Huijgens and Comans, 2003–2005; Bachu, 2008; Gaus, 2010) for achieving higher performances in a short time (O'Connor et al., 2001b; Huijgen and Comans, 2003; Park and Fan, 2004; McKelvy et al., 2004; Alexander et al., 2007).

The above survey indicates that research on carbonation of mine tailings under ambient atmospheric conditions has hitherto been scarce and still leaves numerous unanswered issues. A better understanding of the main parameters affecting natural carbonation of mine residue can provide insights into improvements that can be brought into the reaction, filling the existing knowledge gaps and thus enabling incorporation of tailings piles among the major strategies for atmospheric CO<sub>2</sub> sequestration. Therefore, with a goal to enhance ambient carbonation activity, CO<sub>2</sub> sequestration in mining residues foresees implementation of new approaches leading to long-term and inexpensive means with minimal impact on the environment. Due to their regional abundance, chrysotile mining residues were investigated experimentally in this work. The few existing studies focused on the role of water in the tailings as an essential factor for CO<sub>2</sub> and magnesium dissolution as well as on the dependence of the rate of CO<sub>2</sub> capture on CO<sub>2</sub> partial pressure during ambient mineral carbonation (Pronost et al., 2011). Diffusion of CO<sub>2</sub> in accelerated carbonation was also identified to influence carbonation reactions (Bertos et al., 2004; Bea et al., 2012). The impact on carbonation efficiency due to varying liquid saturation in the residue pores and of its linkage to CO<sub>2</sub> gas composition is not fully understood. Also, the convective and diffusive transport modes of CO<sub>2</sub> through the residue layers impart, beside gas composition, depth-wise heterogeneity of the carbonation profiles that has not been addressed. It is here proposed to scrutinize carbonation reactions under varying liquid saturation and watering schemes, and gas composition in both convective and diffusive modes. Mine residue heterogeneity is a subject which has not received sufficient coverage in the literature when it comes to assessing its potential for CO<sub>2</sub> sequestration. Hence, the role of residue granulometry and mineralogical heterogeneity on CO<sub>2</sub> uptake will be specifically addressed. This work provides a further step on improving our understanding of carbonation mechanisms within mine tailings under ambient atmospheric conditions.

## 2. Experimental

### 2.1. Sample characterization

Chrysotile mining residues (CMR) were collected from the Black Lake Mine tailings (Thetford Mines, Québec). The CMR native crystalline phases were identified by means of powder X-ray diffraction (PXRD) registered on a Siemens D5000 X-ray powder diffractometer (Cu K $\alpha$  radiation) at 1°/min (0.02° step size) over the 5–40° scattering angle range. In agreement with literature reports on similar residues (Voormeij and Simandl, 2004; Power et al., 2011; Pronost et al., 2011), chrysotile and lizardite were the

major minerals coexisting with minor components such as magnetite, brucite, albite, chlorite, talc and phlogopite (Assima et al., 2012). Free brucite in CMR was estimated to amount to 2.94 wt.%. Elemental analysis of the residue was carried out by digesting CMR samples and analyzing their leachate using Perkin-Elmer inductively coupled plasma – optical emission spectrometry (ICP-OES 43000DV). Si, Mg and Fe were the major elements while Al, Ni, Ti, Ca, Cr, Na, and K assayed elements were minor (Assima et al., 2012). Electron microprobe analysis on a CAMECA SX-100 revealed an intra-framework substitution of Mg and Fe in serpentine with formula (Fe<sup>II</sup><sub>0.025</sub>Mg<sub>0.975</sub>)<sub>3</sub>Si<sub>2</sub>O<sub>5</sub>(OH)<sub>4</sub>. This stoichiometry was close to that established via Mössbauer spectroscopy for pure chrysotile fibers (Sarvaramini and Larachi, 2011; Larachi et al., 2012). For the Brunauer–Emmett–Teller (BET) specific surface area determination, CMR samples were dried and then characterized through N<sub>2</sub> sorption and desorption at 77 K after degassing at 150 °C overnight, and residual pressures down to 10<sup>−5</sup> Torr, using a Micromeritics TRISTAR 3000 BET analyzer. CMR BET surface was 6.4 m<sup>2</sup>/g.

### 2.2. Residue heterogeneity

Multi-kg samples of CMR were sieved to isolate sufficient quantities for the fraction passing the 2 mm sieve. The fraction thus obtained was composed of rock particles and fibers with passing sizes  $D_{5\%} = 75$ ,  $D_{10\%} = 150$ ,  $D_{20\%} = 212$ ,  $D_{30\%} = 425$ ,  $D_{50\%} = 850$  and  $D_{60\%} = 1800$   $\mu\text{m}$ . This fraction was deemed sufficiently heterogeneous to reflect the various microscale phenomena taking place at the heap site while matching the size of laboratory-scale carbonation testing setups. Hence, it was further fractionated into narrower sieved fractions  $\leq 75$ ; [75–150]; [150–212]; [212–300]; [300–425]; [425–600]; [600–850]; [850–1800] and [1800–2000]  $\mu\text{m}$  (Supporting information Fig. S1) to quantify the extent of carbonation for each size class and to highlight any size-dependent anomaly in the carbonation pattern. Sample fractions, 30 g each, were dried to eliminate weakly bound water and then placed into quartz cells. The residue layers were thenceforth fully saturated with deionized water before upward sparging of a moist CO<sub>2</sub> stream at a volumetric flow rate of  $100 \pm 3$  mL/min/cell. Carbonation was monitored for 4 days in a temperature-controlled hood ( $22 \pm 0.5$  °C) at 0.1 MPa using deliberately CO<sub>2</sub>-rich feeds to emulate accelerated ambient carbonation.

The extent of CO<sub>2</sub> fixation was quantified after the residue fractions were dried in an oven at 110 °C for 48 h and then calcined under N<sub>2</sub> stream at 810 °C in an induction furnace. The evolving gases were desiccated over a 4–20 mesh powder of anhydrous CaCl<sub>2</sub> (Fisher Scientific) and then analyzed using a CO<sub>2</sub> infrared analyzer (Advance optima continuous gas analyzer A02000 Series, Uras 14). Assuming a 1:1 Mg:CO<sub>2</sub> stoichiometry, indigenous carbonates accounted for 0.37, 0.31, 0.22, 0.25, 0.29, 0.28, 0.27, 0.20 and 0.15 wt.% of the residue fractions, respectively, from finer to coarser fraction, and overall ca. 0.35 wt.% of the 2-mm-passing CMR.

### 2.3. Carbonation kinetics

A dynamic technique was implemented to study residue carbonation at atmospheric pressure while the main parameters governing the carbonation reactions were monitored *in situ*. The parameters thus monitored during the reaction were the volume of CO<sub>2</sub> absorbed per unit mass of CMR sample, the temperature and relative humidity, *HR*, of the recirculating gas, the CMR bed temperature, liquid saturation,  $\beta$ , electrical conductivity,  $\sigma$ , and pore-water pH, and the pressure drop across the CMR bed,  $\Delta P$ . Only the 2-mm-passing CMR fraction was investigated.

At the heart of the carbonation setup, shown in Fig. 1, is a 30-cm-long and 5-cm-ID cylindrical column packed up to  $17 \text{ cm} \pm 0.2 \text{ cm}$  with an unconsolidated residue layer (1). This reactive layer was

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