



# Production of magnesium-rich solutions by acid leaching of chrysotile: A precursor to field-scale deployment of microbially enabled carbonate mineral precipitation



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## ABSTRACT

Carbonate minerals are one of the primary carbon sinks under investigation for sequestering anthropogenic carbon dioxide (CO<sub>2</sub>). Ultramafic rock has the potential to act as a magnesium source for carbonate precipitation reactions utilizing atmospheric CO<sub>2</sub>. This study characterizes the release of magnesium from chrysotile tailings from the Clinton Creek Asbestos Mine (Yukon, Canada) by means of a 42-day leaching experiment using sulfuric acid. Low acid concentrations (targeting 8.33% and 16.67% dissolution of chrysotile) resulted in stoichiometric, dissolution. Moderate (33.33% and 66.67% dissolution) and high (100% dissolution) acid concentrations resulted in non-stoichiometric dissolution of chrysotile. Here, the concentration ratio of Mg:Si in solution was greater than expected for moderate acid concentrations and lower than expected for high acid concentrations. The moderate acid reaction system demonstrated that as much as 84% of the magnesium found in chrysotile can be released into solution via chemical weathering. However, at high acid concentrations, Mg values were in fact lower than the moderate acid reactivity, presumably resulting from adsorption of soluble magnesium to amorphous silica, a byproduct of extensive magnesium leaching from chrysotile fibers. The production of magnesium-rich solutions in this experiment demonstrates that a highly concentrated starting material for magnesium carbonate precipitation reactions can be produced from chrysotile-bearing tailings, providing ultra-mafic hosted mines with an important resource for developing technological strategies for reducing their net carbon emissions. This process represents a win-win scenario for the management of chrysotile-bearing mine wastes, in which a hazardous material is transformed into a geologically stable mineral sink for 'carbon dioxide storage'.

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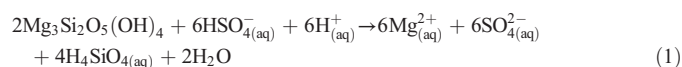
## 1. Introduction

Greenhouse gas emissions produced by fossil fuel combustion are largely responsible for the current increase in atmospheric carbon dioxide (CO<sub>2</sub>) concentration (Lacis et al., 2010; IPCC, 2013). Amongst the sequestration strategies under development, geologic storage of CO<sub>2</sub> in carbonate minerals is of particular interest due to the stability and longevity of these minerals as a carbon sink over geologic time (Wilson et al., 2009a, 2010; Köhler et al., 2010; Renforth and Manning, 2011; Bea et al., 2012; Renforth, 2012; Harrison et al., 2013; Power et al., 2013). Owing to their stability, carbonate minerals will likely be the final sink for a large proportion of the CO<sub>2</sub> produced by human activity over a timeframe of 1 Ma (Seifritz, 1990; Lackner et al., 1995; Kump et al., 2000; Lackner, 2003). Carbonate minerals produced from

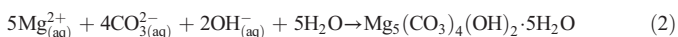
ultramafic rocks, particularly serpentinite, are of interest as a feedstock for carbonation reactions because they weather rapidly under acidic conditions, provide a source of magnesium for carbonation reactions, and are accessible at Earth's surface (Eqs. (1) and (2)) (Stumm, 1992; Park et al., 2003; Park and Fan, 2004; Wilson et al., 2009a). High temperature and pressure industrial processes for carbonate mineral production from magnesium silicate minerals have been examined previously (Lackner, 2003; Zevenhoven et al., 2008; Fagerlund et al., 2009; Koukouzas et al., 2009). Although these strategies are effective for rapid sequestration of CO<sub>2</sub> in carbonate minerals, they are currently not practical for large-scale use due to the financial and energy resources required and the low price on CO<sub>2</sub> (Power et al., 2013). Consequently, other methods of accelerating carbonate mineral precipitation are being examined, including acid leaching of chrysotile under Earth surface conditions:

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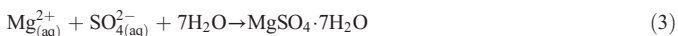
E-mail address: [j.mccutcheon@uq.edu.au](mailto:j.mccutcheon@uq.edu.au) (J. McCutcheon).



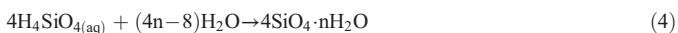
and precipitation of magnesium carbonate minerals:



At high concentrations of sulfuric acid, and under evaporative conditions, alternative precipitation reactions may result in formation of Mg-sulfate minerals:



and/or precipitation of amorphous silica:



Selective extraction of  $\text{Mg}_{(\text{aq})}^{2+}$  from chrysotile during incongruent dissolution may yield both formation of amorphous silica and remnant chrysotile, which can be represented by combining reactions (1) and (4).

Ultramafic mine tailings provide an excellent substrate for carbon mineralization because the ore extraction process significantly increases the surface area of the tailing minerals (White et al., 1996; Molson et al., 2005; Wilson et al., 2009a). Increased exposure of reactive surfaces accelerates the rates of silicate weathering and carbon mineralization relative to those observed for unprocessed ultramafic bedrock. As a result, various methods of accelerating carbonation reactions in mine tailings have been explored. Passive carbon mineralization has been observed at active and historic tailings storage sites, with the potential for greater rates of carbonate mineralization if the geochemical conditions needed for mineral formation are encouraged (Wilson, 2009; Wilson et al., 2009a,b, 2010, 2011, 2014; Pronost et al., 2011). Passive carbonate mineral formation has been documented in milled chrysotile at Clinton Creek Asbestos Mine [Yukon, Canada; (Wilson et al., 2009a)], Cassiar [British Columbia, Canada; (Wilson et al., 2009a)], Black Lake Mine [Québec, Canada; (Pronost et al., 2011)], Woodsreef Mine [New South Wales, Australia (Oskierski et al., 2013)]; processed kimberlite at the Diavik Diamond Mine (Northwest Territories, Canada; Wilson et al., 2009b, 2011); and tailings from the serpentinite-hosted Ni deposit at Mount Keith Nickel Mine [Western Australia; (Wilson et al., 2010)].

The carbonate minerals formed by alteration of tailings at these sites consist primarily of hydrated magnesium carbonate minerals. In some cases, this mineralization is found in association with microbial communities (Power et al., 2011a). This association occurs because photosynthetic microbes induce saturation of carbonate minerals by increasing solution pH via generation of hydroxyl anions ( $\text{OH}^{-}$ ). The hydroxyl ions react with bicarbonate to form water and carbonate ions ( $\text{CO}_3^{2-}$ ), which can subsequently react with divalent cations, such as magnesium and calcium, to form carbonate minerals (Pentecost and Bauld, 1988; Thompson and Ferris, 1990). Microbial cells also act as nucleation sites for carbonate minerals by attracting magnesium ions to their negatively charged surfaces (Beveridge, 1988; Schultze-Lam et al., 1996). Magnesium ions in solution are surrounded by strong hydration shells, composed of six water molecules in octahedral coordination around the central cation (Kluge and Weston, 2005; Hänchen et al., 2008). Carbonate mineralization reactions are promoted by the ability of microbial cells to disrupt these hydration shells while also generating localized high concentrations of magnesium to react with carbonate ions.

Previous studies have demonstrated the ability of some phototrophic microbes to increase the rate of magnesium carbonate mineral precipitation in leachates from ultramafic materials (Power et al., 2009, 2010, 2011a,b; McCutcheon et al., 2014). Based on this accelerated reaction, there is potential for such microbial processes to be utilized for carbon sequestration as long as the microbes are provided with a suitable geochemical environment. Magnesium-rich waters generated by leaching ultramafic mine tailings could act as the magnesium source for microbial, and other, carbonate mineral precipitation reactions. It has been proposed that some mining companies could utilize

biotechnology as a method to reduce net carbon emissions by leaching and carbonating the large volumes of reactive tailings that they produce (McCutcheon et al., 2014; Power et al., 2014). Natural weathering rates limit the availability of magnesium for microbially-mediated carbon mineralization reactions; however, these can be enhanced by the use of stronger acids. Therefore, this study provides geochemical and mineralogical characterization of chrysotile collected from the Clinton Creek Asbestos Mine, and investigates the ability of this material to act as a magnesium source for carbonate mineralization reactions by way of a leaching experiment.

## 2. Material and methods

### 2.1. Description of chrysotile sampling site and sample collection

Chrysotile asbestos was mined at the Clinton Creek Asbestos Mine (77 km NW of Dawson City, Yukon Territory, Canada; Fig. 1) by Cassiar Asbestos Corporation Limited from four open pits, producing 60 million tons of tailings between 1967 and 1978 (Wilson et al., 2010; Yukon, 2010). The bedrock is composed of serpentinite and peridotite (harzbergite, lherzolite, dunite, and pyroxenite) (Htoon, 1979). Most of the peridotite has been serpentinized to chrysotile and antigorite, with the fibrous chrysotile present in veins cutting through the non-fibrous serpentinite wall rock (Fig. 1). Passive carbon mineralization in the tailings has resulted in the formation of nesquehonite ( $\text{MgCO}_3 \cdot 3\text{H}_2\text{O}$ ) as evaporative crusts, and dypingite [ $\text{Mg}_5(\text{CO}_3)_4(\text{OH})_2 \cdot 5\text{H}_2\text{O}$ ] and hydromagnesite [ $\text{Mg}_5(\text{CO}_3)_4(\text{OH})_2 \cdot 4\text{H}_2\text{O}$ ] as cement between silt- to cobble-sized serpentinite grains within the tailings (Wilson et al., 2009a). For the purposes of this study, an aggregate 1 kg grab sample of chrysotile from 'intact' veins, and a hand sample of solid ore were collected within the Porcupine Pit. Of particular relevance to this study, weathering of this vein material likely plays a role in formation of microbialites: Thrombolites composed of acicular aragonite crystals, and containing preserved diatoms, algae, and cyanobacteria, have formed in the Porcupine Pit pond since closure of the mine in 1978 (Power et al., 2011a).

### 2.2. Characterization of chrysotile samples

A 50 g sub-sample of chrysotile fibers from the Porcupine Pit at Clinton Creek was milled with a TM Engineering Ltd. ring mill prior to analysis for major oxide composition and loss on ignition (LOI) via X-ray fluorescence (XRF) using a PANalytical PW-2400 Wavelength Dispersive X-ray Fluorescence Spectrometer. Powder X-ray diffraction (XRD) data were used to determine mineral composition of powdered chrysotile and leached vein material from experiments using a Bruker D8 Focus Bragg-Brentano diffractometer at the University of British Columbia. A step size of  $0.03^\circ$  over a range of  $3\text{--}80^\circ 2\theta$  was used with a dwell time of 0.7 s/step. This instrument is equipped with a LYNXEYE 1D position sensitive detector and a long, fine-focus Co X-ray tube that was operated at 35 kV and 40 mA. Mineral phases were identified with reference to the ICDD PDF-2 Database using the program DIFFRAC.EVA V.3 (available from Bruker AXS).

Polished thin-sections of serpentinite ore were characterized using a Nikon Eclipse LV 100 POL petrographic microscope. Samples were also examined with scanning electron microscopy and energy dispersive X-ray spectroscopy (SEM-EDS) using a Zeiss Leo 1540 XB microscope equipped with an Oxford Instruments INCA x-sight energy dispersive spectrometer for elemental analysis. Sections were coated with 10 nm of osmium using a Filgen OPC80T osmium plasma coater prior to examination using SEM-EDS at 10 kV and a working distance of 12 mm.

### 2.3. Leaching experiment

The ability of the tailings to act as a source of soluble magnesium for carbonate precipitation was quantified by leaching the tailings with

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