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Comparative study of five Québec ultramafic mining residues for use in direct ambient carbon dioxide mineral sequestration

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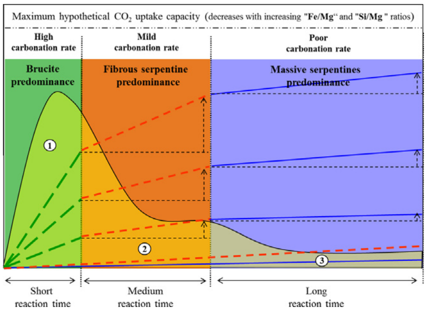
HIGHLIGHTS

- Direct ambient CO_2 uptake by five ultramafic mining residues was monitored.
- Carbonation rate and yield was highly dependent on brucite and chrysotile contents.
- Massive serpentine-enriched mining residues are unsuitable for ambient carbonation.
- $Mg_{brucite}/Mg_{total}$, Mg/Fe, Mg/Si ratios and fiber content govern carbonation rates.

GRAPHICAL ABSTRACT



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ABSTRACT

The mineral carbonation of five different ultramafic mining residues was studied experimentally, namely chrysotile (Black Lake mine [Bl] and Asbestos mine [Asb]), nickel (Ni–Cu Dumont mine project [Du] and Raglan Ni–Cu–EGP mine [Rgl]) and diamond mine residues (Renard mine project [Rnd]). The CO_2 uptake, gas volume and mining residue physical and chemical characteristics were monitored in a fixed-bed diffusion cell to determine their potential for direct ambient carbon dioxide capture and to identify specific parameters influencing their reactivity. The various samples exhibited different behavior when reacted with gaseous CO_2. Brucite- and (fibrous serpentine) chrysotile-containing residues exhibited higher pore-water pH, carbonation rate and yield with brucite content broadly dictating the reaction progress. Massive serpentines such as lizardite/antigorite-containing residues were found to weakly dissolve which significantly reduced their carbonation rate. The Mg_{bru}/Mg_{tot} ratio (brucitic Mg-total Mg atom ratio) and the percentage of fibers were thus the key parameters controlling the direct carbonation of alkaline mining residues. Recourse to alkaline and alkaline-earth metal contents to assess mining residue carbonation capacity may lead to inaccurate estimates as the residue Mg/Si and Mg/Fe ratios were found to greatly control the carbonation capacity through silica gel coating (high Si content) or/and iron (III) hydroxide passivation (high Fe content).

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1. Introduction

Ultramafic mining residues produced during mining operations around the world and accrued, such as in stockpiles, may offer a long-term solid storage buffer of atmospheric CO_2 (IPCC 2007, [1]). Novel techniques are currently under development on the use of such materials on an industrial and global scale to sequester ambient CO_2 while research on the topic is constantly expanding [2]. A number of studies have estimated ultramafic mining residues to contain high enough amounts of magnesium (∼12–19 wt.%) able to chemically bind to ambient CO_2 via above-ground

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natural weathering mechanisms [3]. The resulting carbonates regularly encountered within ultramafic mining residues exposed to weathering are commonly dypingite [$4MgCO_3 \cdot Mg(OH)_2 \cdot 5H_2O$], hydromagnesite [$4MgCO_3 \cdot Mg(OH)_2 \cdot 4H_2O$] and nesquehonite [$MgCO_3 \cdot 3H_2O$] which, as they form, consolidate and cement the residue particles together thereby restricting their air-borne hazard and dissemination by the wind. These hydrated magnesium carbonates are environmentally stable [4] and have been proven as a secure means to store anthropogenic CO_2 emissions [5,6].

The suitability of direct carbonation processes (poor efficiency but cheap) [2] as compared to multi-step mineral processes (high efficiency but costly) [2] for ambient CO_2 sequestration (low CO_2 concentration ~400 ppm, NOAA 2013, [7]) recently led to a resumption of studies on direct carbonation of mining residues dating back to the early 2000s [8]. In the various studies, the abundance of magnesium in mining residues and the detection of magnesium carbonates naturally formed on their mineral surfaces are often underlined as the two key criteria for the ability of mining residues to directly withdraw CO_2 from air or from an industrial exhaust gas. Accordingly, the suitability under specific geochemical conditions of many different ultramafic mining wastes for outdoor carbonation, based on the above two criteria, are broadly exposed in the literature. For instance, the reactivity of residues vis-à-vis CO_2 has been widely investigated, including residues originating from platinum mines (Northam Merensky tailings and BRPM Merensky tailings, South Africa), alumina manufacturing (Australia), the Mount Keith nickel mine (North Eastern Goldfields district, Western Australia), the Clinton Creek asbestos mine (Clinton Creek Yukon, Canada), Vourios Mountain (Western Macedonia, Greece), mined serpentines (Varena, Lithuania), a Nickel mine (Hitura, Finland), the Diavik Diamond Mine (Yukon, Canada), the Dumont Nickel Deposit (Amos, Canada), and the Black Lake Mine (Thetford Mines, Canada). The prospect of using these residues on a large scale through development of geo-engineering technologies to displace substantial amounts of CO_2 from air and reduce its impact on the environment is frequently stressed [4,9–16].

In addition to ore magnesium content and climate differences (temperature, humidity, and precipitation) that impact reaction [17,18], the carbonation rate of ultramafic mining wastes has also been shown to be strongly affected by both their constituent minerals [19] and the microbial activity on the residue heaps [16,20]. Power and co-workers have identified the role of microorganisms in accelerating the weathering of alkaline mining residues at least by 100 to possibly 1000 times, therefore enhancing their dissolution and the precipitation of carbonates [6,20–22]. The recent monitoring of carbon dioxide uptake in ultramafic mining residues demonstrated the improved efficiency of CO_2 mineral sequestration with increased amounts of native brucite in the residues [19]. Carbonation of lizardite/antigorite was observed to be slow while chrysotile carbonation was likely to be a function of fiber length; shorter fibers appeared to be less easily carbonated than longer ones. Iron content, in the form of magnetite or incorporated in serpentines lattices, also proved to be detrimental to carbonation due to Fe^{III} hydroxide precipitation [17,18].

In contrast to microbial processes which are less demanding to implement on an existing mining residue heap, the mineralogy of the mining residue is inextricably linked to the feedstock exploited. Accordingly, the combined elemental and mineralogical compositions is a mandatory indicator that must be carefully used when assessing the hypothetical CO_2 specific sequestration capacity and the overall rate of direct CO_2 uptake. In order to investigate the suitability of alkaline mining waste for ambient carbonation and to provide reliable figures on their capacity to sequester CO_2, the present study proposes to scrutinise the reactivity of five different and well characterized ultramafic mining residues from the Québec mining industry subjected to an accelerated carbonation in a controlled environment. The kinetics and extent of CO_2 uptake are discussed on the basis of the residues' physicochemical properties while key criteria are proposed to assist in categorizing ultramafic mining residues based on their suitability to bind to CO_2.

2. Materials and methods

2.1. Origin of ultramafic mining residues

Ultramafic mining residues used in this work were freshly collected from mine tailing heaps of five different mining projects in Québec (Canada). They were received from the Asbestos [Asb] and Black Lake [Bl] chrysotile mines (Asbestos and Thetford Mines, Québec), the Dumont [Du] Ni–Cu mine project (Royal Nickel, Amos, Québec), the Raglan [Rgl] Ni–Cu–EGP mine (Xstrata Nickel, Nunavik, Québec), and from the Renard [Rnd] Diamond project (SOQUEM/Stornoway Diamond, in north-central Québec).

2.2. Sample characterization methodology

Particle size analysis was performed using an electric sieve tower of various mesh sieves with horizontal electric vibratory motion and vertical shaking. Brunauer–Emmett–Teller (BET) specific surface area was measured using a Micrometrics TRISTAR 3000 BET analyzer through N_2 sorption and desorption at 77 K after degassing at 150 °C overnight and residual pressures down to 10^{-5} Torr. Residues sample bulk densities were measured using a Micrometric AccuPyc 1330 pycnometer. The crystalline phases of the samples were identified from X-ray diffraction (XRD) spectra registered on a Siemens D5000 X-ray powder diffractometer (Cu Kα radiation) at 1°/min rate (0.02° step size) over the 5–65° scattering angle range. Elemental composition of the residue samples was obtained by first digesting samples using combined microwave and HCl, H_3PO_4, and HF solutions, from which surplus HF was eliminated using boric acid (H_3BO_3) [17]. Dissolved samples were thereafter analyzed with a Perkin–Elmer inductively coupled plasma-optical emission spectrometer (ICP-OES 4300 DV) to assay Si, Mg, Fe, Ca, K, Cr, Al, Na, Ni, and Ti. The distribution of iron in different coordination and oxidation states in the materials was assessed through room-temperature ^{57}Fe Mössbauer spectroscopy using a 50 mCi^{57} CoRh source on a conventional Mössbauer spectrometer operated in a sine mode and by electron microprobe analysis performed on a CAMECA SX-100. The indigenous carbonates contained in the samples were estimated by calcination under a N_2 stream in an induction furnace at 810 °C while the evolving gases were desiccated (anhydrous calcium chloride 4–20 mesh powder, Fisher Scientific) and analyzed using a CO_2 infrared analyzer (Advance optima continuous gas analyzer A02000 Series, Uras 14). The sample brucite content was determined through thermal decomposition and a thermogravimetry technique [23].

2.3. Sample carbonation methodology

Carbonation experiments were performed in a vertical reactor configured into two main compartments as presented in Fig. 1. The two compartments, hereafter referred to as upper and lower, are connected to each other by a gate. The upper compartment of 1542.4 mL was used as a CO_2 container while the lower compartment of 506.7 mL housed a thin layer (ca. 1.2 mm) of the ultramafic mining residue sample to be carbonated. The residue layer was held horizontally packed at the top of the lower compartment by a double mesh structure bonded to the reactor inner wall that served as sample holder and gas distributor. Both compartments are equipped with a CO_2 sensor (GMT221 CO_2 transmitter display 0–10%, Vaisala), an integrated hygrometer–thermocouple

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