



Impact of temperature and oxygen availability on the dynamics of ambient CO₂ mineral sequestration by nickel mining residues



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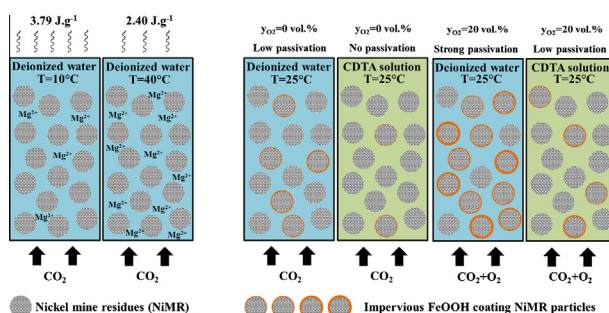
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HIGHLIGHTS

- Ambient temperature variations strongly impacted the rate of NiMR CO₂ uptake.
- Heat generated by carbonation under various temperatures was estimated.
- Presence of oxygen negatively affected both rate and yield of ambient carbonation.
- Use of dilute CDTA chelator reduces undesired precipitation of FeOOH.

GRAPHICAL ABSTRACT



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ABSTRACT

A detailed study of the sensitivity of mineral carbonation to temperature variations and air oxygen content in a saturated nickel mining residue (NiMR) under humid environments was carried out. The dynamics of CO₂ uptake in NiMR beds subjected to carbonation was monitored as a function of temperature and oxygen content in the range 10–40 °C and 0–20 vol.%, respectively. An increase in the rate of carbonation was noticed along with elevated carbonation temperatures. A temperature rise from 10 to 40 °C was accompanied by a ten-fold increase in initial reaction rate and was corroborated by a corresponding stimulated release of magnesium in pore water as evidenced by residue leaching experiments. Furthermore, significant heat was generated during reaction progress which led to a rise in temperature up to 4.9 °C at a 10 °C carbonation temperature. The rate of carbonation was found to be reduced by the increase of iron (III) content in the pore water fostered by the presence of dissolved oxygen. Formation of iron hydroxides induced pore water acidification which stimulated carbonation. This was counterbalanced by the ensuing precipitation of iron (III) hydroxide as a thin coating which slowed the progress of carbonation. Using weakly concentrated CDTA-chelating ligands helped curb iron (III) precipitation which ensured higher carbonation rates under an oxic environment.

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1. Introduction

The vast amounts of solid mining wastes which have accumulated from mining operations across the globe are generating great interest regarding their potential for atmospheric (outdoor) CO₂ capture [1–8]. The outdoor carbonation of mining residues is

expected to gradually capture CO₂ from air therefore reducing its impact upon the environment [9–11], yet the use of mining residues to capture CO₂ faces economic constraints due to their low reaction rates [9]. To improve the reactivity of mining residues as well as to exploit geological formations from which they originate, several techniques have been developed over the past decade [6,8,12–21]. The various methods used to speed up carbonation encompass mechanical, thermal and chemical preprocessing techniques as encountered in most carbonation process schemes

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[22,23]. However, those pre-reaction treatments add cost and energy penalties, and limit applicability to atmospheric CO₂ capture in terms of energy and chemical reagent consumption as compared to the concentration of CO₂ to be captured [3,8,24–30].

Several recent studies are exploring atmospheric CO₂ capture using mining residue sinks to enhance their reactivity under ambient environmental conditions [22,23,31,32]. With the aim to estimate the potential of mining residues to sequester CO₂, the effects of mineralogical and elemental composition of mining residues as well as the impact of liquid saturation and gas moisture on their spontaneous mineral carbonation were investigated under environmental conditions and much insight has been gained on how ambient carbonation can be improved [9,11,33]. In these past studies, the impact of seasonal rainfall on reaction behavior was thoroughly addressed by our group in terms of repercussions of pore water saturation and simulated periodic watering on mining residue carbonation. Moreover, field observations of residue heaps in winter have also been conducted to detect if any interaction occurs between mining waste heaps and the atmosphere via snow coverage. Such observations revealed the formation of diffuse snow-free warm air vents with temperatures up to 20 °C when ambient air temperatures were as cold as –13 °C [34]. The impact of gas diffusion/convection on carbonation as the main supply routes of CO₂ in mining tailings [35–38] has also been discussed [33]. Researchers have also investigated the presence of various natural acid-generating substances such as metal sulfides and elemental sulfur to enhance the ambient dissolution process either under biotic or abiotic conditions [7,39–41]. These various studies have shed light on the potential of mining residues to naturally capture atmospheric CO₂ in a direct way and at the same time give rise to a better understanding of their behavior when subjected to dry or wet meteorological conditions. Still, the influence of all ambient parameters governing natural carbonation has not yet been fully explored.

To complement the current knowledge gaps on the reactivity of mining residues and to provide a better understanding of the reaction under ambient environmental conditions, the emphasis of the present study is on the effect of seasonal temperature variations and oxygen content of air on the dynamics of CO₂ capture by nickel mining residues. In some northern countries such as Canada, winter and summer periods experience significant temperature contrasts [42] often from –18 to +23 °C (Québec) and occasionally from –30 to +30 °C, with weather records in Canada of 45 °C (5 July 1937, Saskatchewan) and –65 °C (3 February 1947, Yukon) according to the National Oceanic and Atmospheric Administration (NOAA, 2012, [43]). These extreme shifts in temperature could significantly impact the reactivity of mining residues and solubility of gaseous CO₂ as well as affect precipitation of magnesium carbonates [44,45]. Furthermore, CO₂ diffusion in mining residue piles is inherently accompanied by a simultaneous oxygen supply which often generates oxygen gradients in the range of 20.9–2 vol.% between the surface and a depth of 30 m of the tailings piles [46]. Oxygen concentration profiles develop via coupled physical and chemical processes such as molecular diffusion in air pockets, transport through the water-filled voids and oxidation of the sulphidic minerals making up the waste rock or the mill tailings [47–49]. Mining residues often contain minerals susceptible to oxidize in the presence of oxygen thereby leading to the formation of (by-) products which can affect the medium composition and hinder the carbonation process, thus affecting the dynamic of carbonation depth profiles. This may have long-term effects on the rate and yield of CO₂ uptake, especially in areas covered with layers of snow that may strongly impede air and CO₂ infiltration.

The above literature survey has highlighted many factors that may potentially influence the carbonation of mining residues that are stock-piled aboveground. It further shows that some major

factors, which may play a significant role in outdoor carbonation of mining residues, might have been overlooked in previous laboratory studies dedicated to assess their CO₂ storage capacity; these include temperature and oxygen partial pressure. Therefore, our study will focus on a laboratory study of nickel mining residues subject to controlled levels of temperature and oxygen level to assess their influence on:

- (1) the carbonation kinetics of a magnesium-rich mining residue as a function of temperature;
- (2) the dissolution kinetics of magnesium from the residue as a function of temperature as Mg dissolution is intimately linked to carbonate formation;
- (3) the interference of dissolved oxygen on oxidizable mineral species with mineral carbonation kinetics.

To the best of the authors' knowledge, this is the first time all three aspects have been studied comprehensively in an integrative approach for a given mining residue and for such an industrial application.

2. Experimental section

2.1. Sample characterization

Nickel mining residues (NiMR) collected from the output of the defibering process of a Ni–Cu mine pilot plant (Dumont Nickel project, Royal Nickel Corporation, 25 km northwest of Amos, Québec, Canada) were selected for this study. The NiMR consisted of particles of various sizes and shapes with $D_{20} = 106 \mu\text{m}$, $D_{50} = 212 \mu\text{m}$ and $D_{80} = 850 \mu\text{m}$. The NiMR native crystalline phases were identified by means of powder X-ray diffraction (PXRD) using a Siemens D5000 X-ray powder diffractometer (Cu K α radiation) at 1°/min rate (0.02° step size) over a 5–65° scattering angle range. Chrysotile and lizardite ($\text{Mg}_3[\text{Si}_2\text{-}_x\text{O}_5]\cdot(\text{OH})_{4-4x}$) are the major minerals coexisting with minor components such as brucite [$\text{Mg}(\text{OH})_2$], magnetite [$\text{Fe}^{2+}\text{Fe}^{3+}\text{O}_4$] and wustite [FeO] (Fig. 1) [9,11,33,50].

Pre-existing crystalline carbonates were not detected in the residues. However, amorphous carbonates were detected as revealed by CO₂ infrared analysis with estimated indigenous amorphous carbonates amounting to 0.4 wt.%. Such erstwhile carbonates may either have existed before mining as a result of the local geochemical conditions or upon exposure of the ore to the atmosphere after being mined, processed and disposed of in the piles. Their prior quantification is necessary to correctly interpret the carbonation conversion data. The native brucite in the NiMR was estimated through a thermal decomposition methodology

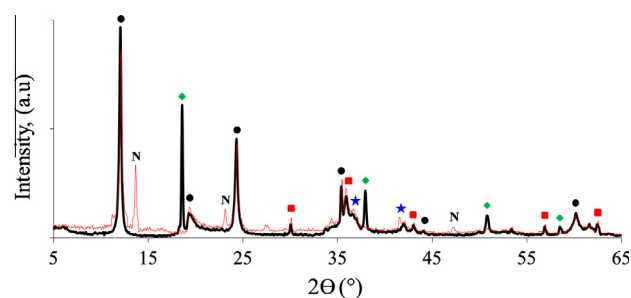


Fig. 1. PXRD spectra in the 5–65°2 θ region (Cu K α) of native NiMR – symbols refer to peaks corresponding to chrysotile/lizardite (filled black circle), brucite (filled green diamond), Magnetite (filled red square), wustite (filled blue star) and nesquehonite (N). Black thick curve = native NiMR, red thin curve = NiMR after 3-day carbonation. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

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