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Structural and chemical changes in mine waste mechanically-activated in various milling environments

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ABSTRACT

This paper evaluates the mechanical activation of mine waste (e.g. partially serpentinized olivine) using different milling machines, with a special focus on changes in microstructure and chemical transformation. The mechanical activation experiments were carried out using lab-scale high-energy planetary and vibratory mills, as well as a pilot-scale stirred mill, and laser diffraction, nitrogen adsorption, X-ray diffraction, and infrared spectroscopy were employed to identify mechanically-induced changes in the mine waste. Direct aqueous carbonation was used to identify the best type of mechanical activation for carbon storage in the mine waste. The experimental results demonstrate that agglomeration of the particles takes place during extended milling in dry conditions, and that there is an effective mechanical activation limit for crystallite size reduction during dry grinding; the higher the milling intensity, the smaller the forsterite's crystallite size will be at the mechanical activation limit. Additionally, stirred milling in wet conditions produces the largest specific surface area, and vibratory milling in dry conditions generates the most disordered materials. The serpentine content was slightly dehydrated during dry milling and was not activated at all during wet milling. The stirred mill proved to be the most efficient form of mechanical activation vis-a-vis the direct aqueous carbonation process, followed by the planetary mill and the vibratory mill, respectively.

1. Introduction

The enhanced ex-situ carbonation of mine waste rock or tailings from the processing of ultramafic ores, such as nickel mine waste, is currently recognized as a promising way of mitigating anthropogenic CO_2 [1,2]. The ex-situ carbonation of ultramafic rocks can be achieved using either a direct or indirect method in a gas-solid or aqueous phase [3]. In a direct carbonation process, both the dissolution of magnesium silicate and CO_2 and the precipitation of magnesium carbonate occur in a single reaction step; the indirect carbonation process differs from the direct process in that it introduces an additional step wherein the reactive compound is extracted from the matrix before carbonation. A pre-treatment method is often applied prior to both carbonation process in order to enhance the rate of reaction, with the appropriate method being determined depending on the primary mineral in the feedstock: thermal and chemical activation is most effective when serpentine is the majority mineral [4], while mechanical activation is preferable when olivine is the majority mineral in the feedstock [5–7].

Mechanical activation, which uses mechanical energy such as intensive grinding to enhance a reaction [8], is one of the most effective pre-treatment methods for sequestering CO_2 via mineral carbonation [9]. From a thermodynamic perspective, mechanical activation stores excess energy in the minerals in the form of long-lived defects, such as the formation of new surface area, new crystallite phases, amorphous materials, and lattice distortion [10]. This excess energy lowers the activation energy of the downstream reaction [11].

Research into enhancing mineral carbonation through the use of mechanical activation is constantly leading to new advances in this technique [12–14]. Numerous investigations have focused on how various grinding conditions, such as grinding modes [15,16], grinding solutions [12,17], grinding atmosphere [18,19], and grinding instruments [15,20], affect on the structural changes of mechanically-activated ultramafic rocks. Sandivik et al. has compared the mechanical activation effects of olivine under various grinding modes, include wet, moisture (10 wt% H_2O), dry modes. They found that grinding under wet conditions typically produces finer particle sizes and larger surface areas than dry grinding at the same energy input. While prolonged dry grinding tends to create aggregates and agglomerates with large particle sizes and a high degree of change to the crystal structure. However, intensive grinding under moisture mode generates surface areas as large as wet grinding and disorders the crystal structure as high as dry grinding [16]. Haug has studied the structure changes of olivine in different mills using milling intensity as the important parameter for comparison.

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For example stirred mill and nutation mill are best compared at medium intensity for industrial applications [15]. However, most previous studies have focused on the changes in specific surface area and the degree of amorphization [15–17,20–22], there has been little investigation into the changes in lattice micro-structure (e.g. crystallite size and micro-strain) [23] and chemical transformation [5] induced by mechanical activation. The structure amorphization of minerals that do not undergo chemical transformation during mechanical activation (i.e. pure olivine) can simply be quantified using the peak area of the X-ray diffraction pattern. For ultramafic mine waste, which is partially serpentinized and weathered, the milling system would be very complicated; thus, a study on the long-lived defects of new phase formation and lattice distortion could provide a more robust understand of how mechanical energy activates the mine waste.

This study investigates the mechanical activation of mine waste under various milling conditions by using different characterization methods to examine the long-lived defects (surface, micro-structure, and chemical changes) that occur as a result of milling. Ideally, the results of this analysis will indicate which conditions are optimal for mechanically activating mine waste for use in mineral carbonation.

2. Materials and experiments

The experimental proceeded according to the following steps: mechanical activation, autoclave carbonation, and physical and chemical characterization (Fig. 1). Firstly, the material is sampled for characterization before being mechanically activated in a high-energy mill for different periods of time. Once the designated time interval had expired, the material was then re-sampled for characterization. The physical changes in materials were characterized by testing the particle size distribution (PSD) and BET surface area, and its structural and chemical information was obtained by using X-ray diffraction (XRD) and Fourier Transform Infrared (FTIR) spectroscopy. The initial and mechanically-activated samples were then carbonated using a direct aqueous process, and, once this step had been completed, the carbonation conversion of the materials after mechanical activation was obtained via quantitative analysis of the carbonated products using their XRD patterns.

2.1. Material

The mine waste material was provided by Hard Creek Nickel's (HCN) Turnagain project in Northern British Columbia, Canada. The received material was ground using a laboratory bond ball mill and screened with a 140 mesh Tyler sieve (160 μm). The undersized materials were used as the initial material for mechanical activation. The initial material had an 80% passing size of 73 μm, a specific surface area of 3.1 m^2/g, and a density of 3.1 g/m^3. X-ray powder diffraction (XRPD) analysis revealed that the starting material was composed of approximately 64.7% Forsterite (Mg_2SiO_4) and 29.6% Lizardite ($Mg_3Si_2O_5(OH)_4$), with trace amounts of magnetite, quartz, and brucite also being present. Table 1 shows the oxide values of samples analyzed using X-ray fluorescence (XRF), the loss on ignition (LOI), and the total C (TOT/C) tested using the Leco method at Acme Analytical Laboratories Ltd., Canada.

2.2. Mechanical activation

In order the test the behavior of mine waste materials under various milling conditions, three types of high-energy mill were used for the mechanical activation:

(1) Planetary mill (PM): The selected planetary mill was a Pulverisette 7 (Fitsch GmbH, Germany) containing seven, 15 mm zirconium oxide balls in a 45 ml zirconium oxide mixing bowl. The mill was loaded with 10 g of feed material, and the ball-to-solid weight ratio was 7:1. The rotational speed of the mill was kept constant at 500 rpm under dry atmospheric conditions and at ambient temperature.
(2) Vibratory mill (VM): The selected vibratory mill was an SPEX 8000 (SPEX Industries, Inc., Edison, NJ) that was loaded with a hardened 25 ml steel vial containing 10 g of feed material and two 6 mm hardened steel balls. The vial was vibrated at an amplitude of 50 mm and a frequency of 20 Hz under dry atmospheric conditions and ambient temperature.
(3) Stirred mill (SM): The stirred mill chosen for the experiments was a horizontal stirred ball mill LME-4 (Netzsch, Germany) which had a 2.48 L effective milling chamber that was loaded with 2 L of 2 mm Keramax MT1 ceramic beads. Pulp with 30 wt% solids content was prepared by mixing 5 kg of sample with 11.7 kg water before being fed into the charged chamber. The milling products were collected, sampled, and then re-fed into the mill, and this cycle was repeated up to nine times until the slurry temperature reached 40 °C. The sampled slurry was dried in an oven at 100 °C for 12 h and subsequently deagglomerated. During the tests, the pulp flow rate was controlled at 2.3 l/min, and the mill agitator speed was maintained at 1500 rpm.

Table 2 lists the samples collected from each mill and the milling variables. For the characterization and carbonation tests, the samples were collected, sealed in plastic tubes, and stored in a freezer at 0 °C. The milling energy consumption recorded in Table 2 indicates the net energy consumed of each sample, which was calculated as the difference between the power draw of milling with an empty vial and milling with a vial filled with sample and grinding media. The calculation of milling energy consumption for the stirred mill is the same as used in [6], and the calculations for the planetary mill and the vibratory mill are the same as in [5]. Mill intensity is equal to the specific energy input divided by time.

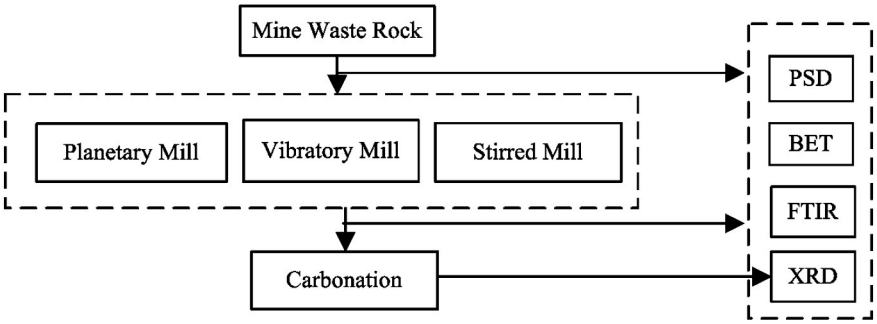

Fig. 1. Procedures of the experiments.