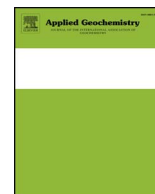




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Carbon sequestration via enhanced weathering of peridotites and basalts in seawater

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ABSTRACT

Enhanced weathering of mafic and ultramafic rocks has been suggested as a carbon sequestration strategy for the mitigation of climate change. This study was designed to assess the potential drawdown of CO₂ directly from the atmosphere by the enhanced weathering of peridotites and basalts in seawater. Pulverized, and ball milled dunite, harzburgite and olivine basalt were reacted in artificial seawater in batch reactor systems open to the atmosphere for two months. The results demonstrate that the ball-milled dunite and harzburgite changed dramatically the chemical composition of the seawater within a few hours, inducing CO₂ drawdown directly from the atmosphere and ultimately the precipitation of aragonite. In contrast, pulverized but unmilled rocks, and the ball-milled basalt, did not yield any significant changes in seawater composition during the two-month experiments. As much as 10 wt percent aragonite was precipitated during the experiment containing the finest-grained dunite. These results demonstrate that ball milling can substantially enhance the weathering rate of peridotites in marine environments, promoting the permanent storage of CO₂ as environmentally benign carbonate minerals through enhanced weathering. The precipitation of Mg-silicate clay minerals, however, could reduce the efficiency of this carbon sequestration approach over longer timescales.

1. Introduction

Human activities over the past century, particularly fossil fuel consumption, have caused a dramatic increase of CO₂ concentration in the atmosphere (e.g. IPCC, 2007; Oelkers and Cole, 2008). This anthropogenic impact on the global carbon cycle is considered to be the main reason for the observed climate change over the past decades (IPCC, 2005; Solomon et al., 2009). Global climate change has been linked to various other phenomena, including hurricanes, droughts, floods, glacier retreat, and rising sea levels (Emanuel, 2005; Rignot, 1998; Schiermeier, 2011; Trenberth et al., 2014). As such, large efforts have been made to develop effective carbon capture and storage (CCS) methods that remove CO₂ from the atmosphere (e.g. Gerdemann et al., 2007; Gislason and Oelkers, 2014; Lackner et al., 1995; Matter and

Kelemen, 2009; Matter et al., 2016; Michael et al., 2010; Oelkers et al., 2008; Power et al., 2013, 2016; Wilson et al., 2014).

Chemical weathering is a slow process that controls atmospheric CO₂ concentrations over geological time scales. A number of scientists have proposed accelerating chemical weathering to counter global climate change (Griffioen, 2017; Hartmann et al., 2013; Köhler et al., 2010, 2013; Lackner, 2003; Montserrat et al., 2017; Schuiling and De Boer, 2011; Schuiling and Krijgsman, 2006; Seifritz, 1990; Taylor et al., 2016). The goal of enhanced weathering is to hasten silicate mineral weathering rates to accelerate the removal of CO₂ from the atmosphere as dissolved inorganic carbon and/or as carbonate minerals. Because of their relatively rapid dissolution rates, enhanced weathering has focused on mafic and ultramafic rocks (Hartmann et al., 2013; Hauck et al., 2016; Moosdorf et al., 2014; Renforth, 2012; Taylor et al., 2016).

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Experimentally measured dissolution rates of Wolff-Boenisch et al. (2011) suggest that ground mafic and ultramafic rocks could lead to the efficient carbon dioxide mineralization in seawater. One method to accelerate weathering rates is to increase rock surface area by crushing, grinding, and/or milling (Renforth, 2012). The grinding process can increase specific dissolution rates through “mechano-chemical activation” (Balaz et al., 2008; Gerdemann et al., 2007). Past studies have also demonstrated that milling can dramatically increase the reactivity of Mg-silicates by reducing particle size to $> 1 \mu\text{m}$ (e.g. Haug et al., 2010; Rigopoulos et al., 2015, 2016a, 2016b; Turianicová et al., 2013). The effect of mechanical activation on the carbon sequestration efficiency of ultramafic rocks/mine waste materials has been reported by Li and Hitch (2016a, b; 2017a, b). Schuiling and De Boer (2011) suggested that even relatively large olivine grains might completely dissolve within 1–2 years in high-energy shallow marine environments. In contrast, Hangx and Spiers (2009) estimated that olivine particles of $10 \mu\text{m}$ need approximately 23 years to completely dissolve, although Renforth (2012) suggested this estimate was 30% too high. Moreover, Köhler et al. (2013) suggested that only olivine particles with a grain size on the order of $1 \mu\text{m}$ would sink slowly enough to enable their nearly complete dissolution.

The goal of this study is to assess the potential for the enhanced weathering of peridotites and basalts in seawater to facilitate the drawdown of atmospheric CO_2 . Towards this goal, we reacted two ultramafic rocks and one mafic rock in artificial seawater in open system reactors. The rock samples were subjected to different degrees of ball milling to produce powders of distinct sizes and surface area. The purpose of this communication is to report the results of this experimental study and to use these results to illuminate the potential role of enhanced ultramafic and mafic rock weathering in seawater as a viable CCS technique.

2. Materials and methods

2.1. Sample selection, preparation and characterization

The rocks used in this study were collected from the Troodos ophiolite complex; this complex was formed in a supra-subduction zone environment around 92–90 Ma ago (Mukasa and Ludden, 1987; Robertson, 2002; Robinson and Malpas, 1990). In the present study, two peridotites were collected from the Troodos mantle section: one dunite and one harzburgite, both of which are partially serpentinized. Additionally, an olivine basalt was collected from the “Upper” Pillow Lava unit of the Troodos ophiolite. The mineralogical and textural characteristics of these rock samples were determined by petrographic analysis of representative thin sections using a polarizing microscope (Figs. S1–S3 in the electronic supplement). Whole-rock chemical analyses were also performed using a combination of lithium metaborate/tetraborate fusion inductively coupled plasma (ICP), inductively coupled plasma mass spectroscopy (ICP-MS) and instrumental neutron activation analysis (INAA) techniques (Tables S1 and S2 in the electronic supplement). Fine-grained samples were initially prepared using a stainless steel pulverizer and then sieved to obtain the $104\text{--}150 \mu\text{m}$ size fraction. This fraction was cleaned ultrasonically ten times in ethanol to remove fine particles; and then dried overnight at 50°C . The specific surface area of this fraction for each rock sample was measured by the BET method (Table 1). A portion of this size fraction was used directly in the experiments, while the remainder was ball-milled to further reduce its grain size as described in section 2.2. The non-ball-milled size fraction is henceforth referred to as “unmilled”. Additional enhanced weathering experiments were performed using selected ball-milled samples.

2.2. Ball milling

Ball milling (BM) was performed using a Fritsch Pulverisette 6

Table 1

Ball milling conditions and specific surface area values of the unmilled and milled rock materials used in the experiments (data for the milled samples of basalt and dunite acquired from Rigopoulos et al. (2015, 2016a), respectively).

Sample code	Ball milling conditions		BET ($\text{m}^2 \text{g}^{-1}$) Specific surface area
	Milling time (h)	Type of milling	
SM15 (Unmilled Dunite)	–	–	0.4
BM38 (Milled Dunite)	4	Wet (50 wt% Ethanol)	35.7
BM46 (Milled Dunite)	20	Wet (50 wt% Ethanol)	64.6
SM17 (Unmilled Harzburgite)	–	–	0.5
BM72 (Milled Harzburgite)	16	Wet (50 wt% Ethanol)	53.1
SM1 (Unmilled Basalt)	–	–	2.0
BM7 (Milled Basalt)	4	Wet (50 wt% Ethanol)	58.9

planetary mono mill. The optimum milling conditions for basaltic and ultramafic rocks were applied, based on the results of previous studies (Rigopoulos et al., 2015, 2016a). The peridotites and basalt were wet-milled in an 80 mL tungsten carbide bowl using ethanol as process control agent (PCA). The selection of this PCA is based on recent experimental results (Rigopoulos et al., 2015, 2016a), which demonstrated that the use of ethanol during ball milling promotes the formation of smaller, more uniform and rounded particles compared to H_2O . In our experiments, the ball-to-powder mass ratio was 20:1 w/w, the fluid-to-powder mass ratio was 1:2 and the rotation speed was 300 rpm. The process was performed using 30 tungsten carbide balls with a 10 mm diameter. Tungsten carbide bowl and balls were used to avoid possible contamination due to the hardness of ultramafic and mafic rocks. Ball milling was automatically interrupted every 5 min for 5 min to avoid sample heating. This periodic interruption avoids phase transformations and reduces the evaporation of the PCA. After milling was complete, the recovered rock powders were dried overnight at room temperature. The enhanced weathering experiments described below were performed using the ball-milled samples with the highest BET specific surface area (see Fig. 1). For the dunite, an additional ball-milled sample was used to clarify the role of ball milling duration on chemical weathering rates. The correlations between the specific surface area and the ball milling duration for each rock type are shown in

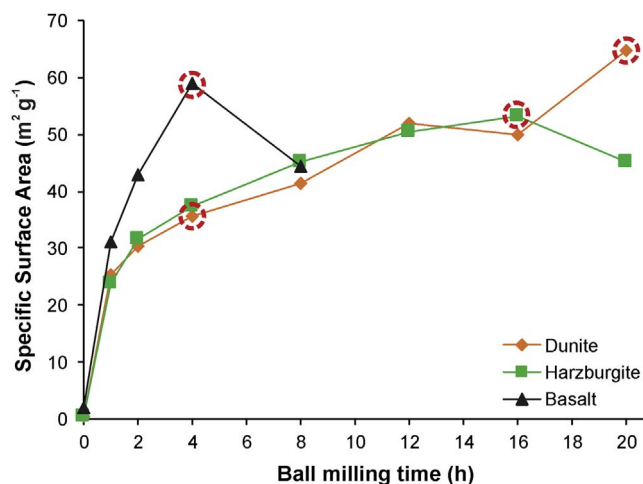


Fig. 1. BET ($\text{m}^2 \text{g}^{-1}$) specific surface area versus ball milling time for the studied rock materials (red circles show the milled samples used during the experiments). The BET values for the milled basalt and dunite samples were acquired from Rigopoulos et al. (2015, 2016a), respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

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