Earth and Planetary Science Letters 468 (2017) 11–26

Contents lists available at ScienceDirect

Earth and Planetary Science Letters

www.elsevier.com/locate/epsl

Subarctic physicochemical weathering of serpentinized peridotite

O.I. Ulven [a,*], A. Beinlich [a,b], J. Hövelmann [a,c], H. Austrheim [a], B. Jamtveit [a]

[a] *Physics of Geological Processes (PGP), University of Oslo, 0316 Oslo, Norway*
[b] *The Institute for Geoscience Research (TIGeR), Department of Applied Geology, Curtin University, 6845 Perth, Australia*
[c] *German Research Centre for Geosciences (GFZ), Interface Geochemistry, 14473 Potsdam, Germany*

ARTICLE INFO

Article history:
Received 18 November 2016
Received in revised form 17 March 2017
Accepted 24 March 2017
Available online xxxx
Editor: M. Bickle

Keywords:
peridotite
sub-arctic weathering
brucite dissolution
carbonation
fracturing

ABSTRACT

Frost weathering is effective in arctic and subarctic climate zones where chemical reactions are limited by the reduced availability of liquid water and the prevailing low temperature. However, small scale mineral dissolution reactions are nevertheless important for the generation of porosity by allowing infiltration of surface water with subsequent fracturing due to growth of ice and carbonate minerals. Here we combine textural and mineralogical observations in natural samples of partly serpentinized ultramafic rocks with a discrete element model describing the fracture mechanics of a solid when subject to pressure from the growth of ice and carbonate minerals in surface-near fractures. The mechanical model is coupled with a reaction–diffusion model that describes an initial stage of brucite dissolution as observed during weathering of serpentinized harzburgites and dunites from the Feragen Ultramafic Body (FUB), SE-Norway. Olivine and serpentine are effectively inert at relevant conditions and time scales, whereas brucite dissolution produces well-defined cm to dm thick weathering rinds with elevated porosity that allows influx of water. Brucite dissolution also increases the water saturation state with respect to hydrous Mg carbonate minerals, which are commonly found as infill in fractures in the fresh rock. This suggests that fracture propagation is at least partly driven by carbonate precipitation. Dissolution of secondary carbonate minerals during favorable climatic conditions provides open space available for ice crystallization that drives fracturing during winter. Our model reproduces the observed cm-scale meandering fractures that propagate into the fresh part of the rock, as well as dm-scale fractures that initiate the breakup of larger domains. Rock disintegration increases the reactive surface area and hence the rate of chemical weathering, enhances transport of dissolved and particulate matter in the weathering fluid, and facilitates CO_2 uptake by carbonate precipitation. Our observations have implications for element cycling and CO_2 sequestration in natural gravel and mine tailings.

1. Introduction

Chemical and physical processes within the critical zone control the extent and rate of mass transport in surface-near environments (Martin and Meybeck, 1979; Milliman and Syvitski, 1992; White and Blum, 1995; Gaillardet et al., 1999). Weathering has significant effects on the atmospheric CO_2 concentration due to the dissolution of Ca- and Mg-silicate minerals and subsequent carbonate precipitation (e.g. Berner and Kothavala, 2001). High concentrations of Ca- and Mg-silicate minerals can be found in mafic (e.g., basalt, gabbro) and ultramafic (e.g., dunite, peridotite) rocks which weather at the Earth's surface during interaction with surface waters. Numerous experimental (Hövelmann et al., 2011, 2012a, 2012b; van Noort et al., 2013; Andreani et al., 2009; Giammar et al., 2005; King et al., 2010; Daval et al., 2011; Harrison et al., 2013, 2015, 2016; Wilson et al., 2010) and natural analogue studies (Beinlich and Austrheim, 2012; Beinlich et al., 2010, 2012; Kelemen and Matter, 2008; Oskierski et al., 2013a, 2013b; Boschi et al., 2009; Hansen et al., 2005; Kelemen et al., 2011; Wilson et al., 2009, 2011; Power et al., 2009) have investigated the potential of ultramafic rocks as a feedstock for carbonate formation to mitigate anthropogenic CO_2 emissions. Other aspects of societal importance associated with ultramafic rock weathering include the release of asbestos and fluid mobile toxic Ni- and Cr-bearing compounds into agricultural land and drinking water (Bales et al., 1984; Dublet et al., 2012; Garnier et al., 2013; Holmes et al., 2012; Oze et al., 2007). Due to the interest in soil toxicity and the economic significance of Ni deposits (Golightly and Arancibia, 1979; Wells et al., 2009), most studies of chemical weathering of ultramafic rocks focus on laterite formation in tropical and temperate climate zones that often results in depletion in Ca, Mg and Si and enrichment in Fe and potentially toxic metals such

---

[*] Corresponding author.
E-mail address: o.i.ulven@fys.uio.no (O.I. Ulven).

http://dx.doi.org/10.1016/j.epsl.2017.03.030

as Co, Cr and Ni (Oze et al., 2007; Schwertmann and Latham, 1986).

In contrast, little is known about the processes acting during ultramafic rock weathering in the arctic and subarctic climate zones besides recent investigations of the natural carbon uptake potential of ultramafic mine wastes in Canada (e.g. Pronost et al., 2011; Wilson et al., 2009, 2011). The limited availability of water and slow mineral dissolution kinetics at low temperature suggest that mechanical weathering may be relatively more important for bedrock disintegration compared to tropical climate zones. Mechanical weathering results from freezing of water in open fractures and precipitation of carbonate minerals, which has been suggested to drive reaction-induced rock fracturing (e.g. Kelemen and Matter, 2008; Rudge et al., 2010; Beinlich and Austrheim, 2012). Frost weathering may result from the 9% volume increase when liquid water transforms into solid ice and from ice segregation, which involves migration of water to freezing centers due to cryosuction (Walder and Hallet, 1985; Rempel et al., 2004; Vlahou and Worster, 2015). The volume expansion of freezing water effectively drives fracturing only on short time scales during rapid freezing in diurnal freeze-thaw cycles within a few centimeters from the surface of nearly water saturated rocks (Matsuoka and Murton, 2008; Girard et al., 2013). In contrast, ice segregation acts on time scales of a winter season, and involves migration of water from rock volumes above the freezing point or water present in small pores where freezing is suppressed due to curvature effects, or redistribution of ice initially in thermodynamically less favored settings (e.g. Rempel et al., 2016). This typically produces fractures on larger scales, potentially reaching several meters into the bedrock (Matsuoka and Murton, 2008). Refreezing of melt water in fractures during thawing may further accelerate the process (Matsuoka, 2008).

A rock's susceptibility to frost weathering depends on the accessibility for liquid water, i.e. porosity and permeability, and on its mechanical strength. Lautridou and Ozouf (1982) argue that rocks with less than 6% porosity are insensitive to ice fracturing, while Matsuoka and Murton (2008) state that low porosity rocks disintegrate along pre-existing joints only. However, the preconditioning of low porosity rocks to frost weathering by localized dissolution reactions resulting in high porosity zones at the rock's surface and near pre-existing tectonic fractures has received less attention.

Here we present new insight into chemical and physical processes taking place during subarctic weathering of the Feragen Ultramafic Body (FUB) in SE-Norway. The development of different fracture phenomena eventually resulting in gravel formation from ultramafic bedrock can be described by a model that couples chemical weakening with subsequent breaking of the fresh rock driven by carbonate precipitation and ice growth in fractures and pore space. The rate of rock weakening is controlled by mineral reaction kinetics during chemical weathering. The amount of gravel produced thus depends on the rock's composition. Rock disintegration controls the sediment transport and composition of the local runoff and contributes to natural CO_2 uptake via alkalinity production and carbonate formation.

2. Geological setting and climate

The FUB is situated in east central Norway (62°32.9′N 11°48.4′E) at an altitude between 700 and 900 m a.s.l. between the town of Røros and the border with Sweden (Fig. 1). The area is among the coldest regions in Europe and located in the subarctic climate zone with monthly mean temperatures ranging between -25 and $+8\,^\circ$C. Autumn and spring temperatures fluctuate around $0\,^\circ$C, giving rise to fast freeze-thaw cycles, and stable temperatures well below zero are common throughout the winter months (see http://eklima.met.no). As with most parts of Scandinavia, the FUB was completely glaciated during the Weichselian which constrains the maximum age for the observed surface weathering, gravel, and saprolite formation to <10 ka.

The FUB is one of several ultramafic bodies that are aligned over a distance of >200 km across the Norwegian–Swedish border and has been regarded as an alpine peridotite in gneissic Cambro–Silurian sequences (Moore and Hultin, 1980). The area is barren with prominent exposure of ultramafic rocks that disintegrate into a thin (<1 m) layer of unconsolidated gravel containing little soil and locally saprolite (Fig. 2a). The FUB comprises partly serpentinized dunite and peridotite together with harzburgite and chromitite layers. The degree of serpentinization varies between almost complete in the outer parts of the FUB and partial in the central part. The northern contact with the surrounding country rocks is covered by a Devonian serpentine conglomerate indicating that the FUB was exposed to surface weathering already in the Paleozoic. Moore and Hultin (1980) report on small occurrences of Mg-carbonate minerals (magnesite, nesquehonite) along fractures and cleavage surfaces in serpentine and chlorite.

3. Analytical methods

3.1. Raman spectroscopy

A confocal Raman spectrometer (Horiba Jobin Yvon XploRA) operating with the 638 nm line of a He–Ne laser was used for spatially resolved identification of mineral phases within the serpentine and carbonate veins of weathered and fresh parts of the samples. Single spectra were taken with a 500 μm hole, 100 μm slit and 1200 grooves per millimeter grating using an acquisition time of 4 × 45 s. Corrections for system drift were done using the 520.7 nm Raman band of a silicon standard taken at the beginning and the end of each measuring session. Raman maps across the weathering interface were obtained using a step size of 1 × 1 μm and an acquisition time of 2 × 60 s. At each point of the map, a Raman spectrum was taken in the 100–800 cm^{-1} region. The Labspec software was used for peak fitting and for producing intensity maps of selected Raman bands.

3.2. Backscattered electron (BSE) imaging

Back-scattered (BSE) images of thin sections and element distribution maps were collected with a Cameca SX100 electron microprobe and a JEOL JSM 640 LV scanning electron microscope at the Department of Geosciences, University of Oslo. Element distribution maps were acquired with a step size of 1 μm using an acceleration voltage of 15 kV, a beam current of 20 nA and counting time of 50 ms per pixel.

3.3. Whole rock geochemical analyses

Whole rock geochemical analyses including CO_2 and FeO were performed by Actlabs Laboratories Ltd., using the lithium metaborate/tetraborate fusion ICP Whole Rock and the trace element ICP/MS packages (www.actlabs.com). FeO was determined through titration, using a cold acid digestion of ammonium metavanadate, and hydrofluoric acid in an open system. Ferrous ammonium sulphate was added after digestion, and potassium dichromate was used as titrating agent. The whole rock concentration of CO_2 was determined by thermally decomposing 0.2 g of sample material in a resistance furnace in a pure nitrogen environment at $1000\,^\circ$C, using an ELTRA CW-800, directly releasing CO_2. H_2O is removed in a moisture trap prior to the detection of carbon dioxide in the infrared (IR) cell.