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Research paper

The production of iron oxide during peridotite serpentinization: Influence of pyroxene

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ABSTRACT

Serpentinization produces molecular hydrogen (H₂) that can support communities of microorganisms in hydrothermal fields; H₂ results from the oxidation of ferrous iron in olivine and pyroxene into ferric iron, and consequently iron oxide (magnetite or hematite) forms. However, the mechanisms that control H₂ and iron oxide formation are poorly constrained. In this study, we performed serpentinization experiments at 311 °C and 3.0 kbar on olivine (with <5% pyroxene), orthopyroxene, and peridotite. The results show that serpentine and iron oxide formed when olivine and orthopyroxene individually reacted with a saline starting solution. Olivine-derived serpentine had a significantly lower FeO content (6.57 ± 1.30 wt.%) than primary olivine (9.86 wt.%), whereas orthopyroxene-derived serpentine had a comparable FeO content (6.26 ± 0.58 wt.%) to that of primary orthopyroxene (6.24 wt.%). In experiments on peridotite, olivine was replaced by serpentine and iron oxide. However, pyroxene transformed solely to serpentine. After 20 days, olivine-derived serpentine had a FeO content of 8.18 ± 1.56 wt.%, which was significantly higher than that of serpentine produced in olivine-only experiments. By contrast, serpentine after orthopyroxene had a slightly higher FeO content (6.53 ± 1.01 wt.%) than primary orthopyroxene. Clinopyroxene-derived serpentine contained a significantly higher FeO content than its parent mineral. After 120 days, the FeO content of olivine-derived serpentine decreased significantly (5.71 ± 0.35 wt.%), whereas the FeO content of orthopyroxene-derived serpentine increased (6.85 ± 0.63 wt.%) over the same period. This suggests that iron oxide preferentially formed after olivine serpentinization. Pyroxene in peridotite gained some Fe from olivine during the serpentinization process, which may have led to a decrease in iron oxide production. The correlation between FeO content and SiO₂ or Al₂O₃ content in olivine- and orthopyroxene-derived serpentine indicates that aluminum and silica greatly control the production of iron oxide. Based on our results and data from natural serpentinites reported by other workers, we propose that aluminum may be more influential at the early stages of peridotite serpentinization when the production of iron oxide is very low, whereas silica may have a greater control on iron oxide production during the late stages instead.

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1. Introduction

Serpentinization is a low-temperature (<500 °C) hydrothermal alteration of ultramafic rocks (typically komatiite and peridotite), where olivine and pyroxene are hydrated to form serpentine, (±) brucite, and (±) magnetite (Eq. (1)):

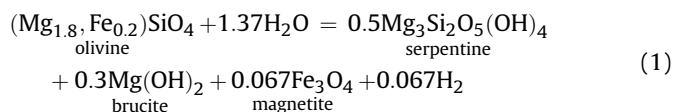
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Serpentinization occurs in many geological settings on Earth, including the ocean floor, mid-ocean ridges and subduction zones (e.g., Charlou et al., 2002; Hyndman and Peacock, 2003; Mével, 2003; Evans et al., 2013). It dramatically modifies the physical, mechanical, and chemical properties of oceanic lithosphere (e.g., Escartín et al., 1997, 2001; Guillot and Hattori, 2013). Serpentinization leads to the uptake of voluminous H₂O (up to ~13 wt.%) and a dramatic decrease in density and strength (e.g., Escartín et al., 1997, 2001; Germanovich et al., 2012). Deformation experiments show that serpentine particularly lizardite is weaker than the other components of oceanic lithosphere, and a small degree of serpentinization (<15%) greatly reduces the strength of peridotite (Escartín et al., 1997, 2001). Moreover, serpentine can incorporate large quantities of fluid-mobile elements, such as B, Sr, and Cs (e.g., Hattori and Guillot, 2003; Scambelluri et al., 2004; Guillot and Hattori, 2013). Thermodynamic and experimental studies show that serpentine can remain stable at depths of greater than 150 km (Ulmer and Trommsdorff, 1995; Schmidt and Poli, 1998), suggesting that serpentinization is significant for the transfer of H₂O and fluid-mobile elements to the mantle.

Fluids derived from serpentine-hosted hydrothermal fields typically contain abundant molecular hydrogen (H₂) and methane (CH₄), which can be utilized by communities of microorganisms to gain metabolic energy (e.g., Schrenk et al., 2004, 2013; Brazelton et al., 2006, 2010; Holm et al., 2015). Molecular hydrogen is produced by the oxidation of Fe²⁺ in olivine and pyroxene to Fe³⁺. Ferric iron is mainly distributed into magnetite and silicate minerals (e.g., serpentine, talc, and tremolite). If all Fe³⁺ is incorporated into silicate minerals, magnetite may be absent during serpentinization (Seyfried et al., 2007; Evans, 2008; Evans et al., 2009).

However, the mechanisms that control H₂ production and the formation of magnetite are poorly understood (Bach et al., 2006; Beard et al., 2009; Frost et al., 2013). Analyses of serpentinized dunite and harzburgite suggest that magnetite is absent at the early stage of serpentinization when Mg-rich serpentine and Fe-rich brucite (around 27 wt.% FeO) form, whereas it is produced at the late stage of serpentinization through the breakdown of Fe-rich brucite (Bach et al., 2006). By contrast, Frost et al. (2013) argued that magnetite resulted from the extraction of iron from brucite and serpentine. However, the factors that control the distribution of iron between brucite and serpentine are poorly constrained. Based on the formation temperatures of serpentinites obtained according to oxygen isotopic compositions of serpentine minerals, Klein et al. (2014) proposed that magnetite is absent during serpentinization at temperatures below 200 °C. Consistently, experimental studies of peridotite serpentinization have shown that magnetite is not formed at 200 °C and 500 bar (Seyfried et al., 2007). By contrast, magnetite is produced at 200 °C after olivine serpentinization (McCullom et al., 2016), indicating that the process of magnetite formation during olivine serpentinization differs greatly from that during peridotite alteration. Compared to olivine, peridotite serpentinization produces one to two orders of magnitude more silica (e.g., Allen and Seyfried, 2003). Silica greatly controls the production of iron oxide during serpentinization, as it changes the composition and stability of Fe-bearing minerals (e.g., Bach et al., 2006; Frost and Beard, 2007; Klein et al., 2009). Frost and Beard (2007) reported that magnetite should theoretically be unstable at high silica activity. By contrast, analyses of natural serpentinites show that silica promotes magnetite production (Miyoshi et al., 2014). The controversy

motivates us to investigate the influence of pyroxene and silica on magnetite production during serpentinization.

In this study, we performed serpentinization experiments at 311 °C and 3.0 kbar on natural ground orthopyroxene, olivine (with <5% pyroxene) and peridotite. Serpentinized harzburgite from the Lichi Mélange, Taiwan, was examined in order to compare with experimental observations in this study. This work aimed to (1) study the production of iron oxide during peridotite serpentinization, (2) illustrate the sequence of serpentinization reactions of olivine and orthopyroxene, and (3) study the influence of pyroxene on the formation of iron oxide during peridotite serpentinization.

2. Methods

2.1. Preparation of starting materials

The experimental strategy used in this study is to react olivine, orthopyroxene and spinel-bearing peridotite with a saline solution (0.5 mol/L NaCl). The peridotite was sampled at Panshishan (Jiangsu Province, China) where it occurs as xenoliths in alkaline basalts (Chen et al., 1994; Sun et al., 1998; Xu et al., 2008; Yang, 2008). It is composed of 60–65 vol.% olivine, 20–25 vol.% orthopyroxene, 15 vol.% clinopyroxene, and 1–2 vol.% spinel. The peridotite is fresh, as evidenced by a very small loss on ignition (typically less than 0.5%) upon heating at 1200 °C (Yang, 2008). Olivine and orthopyroxene were picked from crushed peridotite grains (<60 mesh) using a binocular microscope, and their fractions include less than 5% of other mineral phases. Peridotite, olivine and orthopyroxene were then ground in an agate mortar and sieved into a starting grain size of 100–177 μm. Small grains formed during crushing were removed with an ultrasonic bath. The starting fluid was prepared with fresh pure water and reagent-grade sodium chloride.

2.2. Preparation of gold capsules and experimental procedures

The solid reactants and starting solution were loaded into gold capsules (4.0 mm outer diameter, 0.2 mm wall thickness, and 30 mm long). The water/rock ratios, *i.e.*, mass ratios between the starting solution and the solid reactants, are ~1.0. Gold is commonly used in serpentinization experiments (e.g., Berndt et al., 1996; Malvoisin et al., 2012) because it is chemically inert and does not form any Fe-Au alloy at low temperatures (e.g., ≤500 °C). The capsules were double-sided welded with a tungsten inert gas high-frequency pulse welder (PUK3). Leaks were checked before and after all experiments by putting capsules in a drying furnace at 100 °C for at least 2 h. Only capsules with mass differences less than 0.5% from the initial mass before heating were used in experiments.

All experiments were performed in cold-seal hydrothermal vessels at the Guangzhou Institute of Geochemistry, Chinese Academy of Sciences (Table 1). The capsules were loaded into the end of hydrothermal vessels, followed with a filler rod (~6 cm in

Table 1
Experimental conditions.

Sample No.	Starting material	Water/rock ratios ^a	Time (days)	Products
Fe37	Olivine	1.1	26	Srp, Mgt, Ol
HR42	Orthopyroxene	0.82	18	Srp, Opx
HR25	Peridotite	1.4	19	Srp, Mgt, Ol, Opx, Cpx
HR61	Peridotite	0.82	120	Srp, Mgt, Ol, Opx, Cpx
HR77	Peridotite	1.2	28	Srp, Mgt, Ol, Opx, Cpx

^a Mass ratios between the starting solution and the solid reactant loaded into gold capsules.

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