



Growth kinetics of forsterite reaction rims at high-pressure



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ABSTRACT

Growth kinetics of forsterite (Fo) reaction rims between periclase (Per) and enstatite (En) were studied experimentally at pressure (P) and temperature (T) conditions of 3.0–11.1 GPa and 1473–1873 K, respectively. Pt markers originally placed at the Per-En interface were always observed at the Per-Fo interface, which indicates that Mg and O are the diffusing species in Fo rim growth (Mg-O coupled diffusion). The presence of some En inclusions in Fo grains and the growth rate of the Fo rim suggests that grain boundary diffusion is dominant rather than lattice diffusion. Considering the very fast grain boundary diffusion of O in olivine, the Mg-O coupled grain boundary diffusion in Fo is deduced to be rate-limited by the diffusivity of Mg. Based on an analysis of data collected under dry conditions, the product of the Mg grain boundary diffusion coefficient (D_{gb}) and the effective grain boundary width (δ) was determined to be $\delta D_{gb} = \delta D_{gb,0} \exp[-(E^* + PV^*)/RT]$ with $\delta D_{gb,0} = 10^{-9.68 \pm 1.51} \text{ m}^3/\text{s}$, $E^* = 379 \pm 44 \text{ kJ/mol}$ and $V^* = -1.9 \pm 1.4 \text{ cm}^3/\text{mol}$. Our results, combined with previously reported data on Mg lattice diffusion in Fo, suggest that for Mg, the significance of grain boundary diffusion increases with depth in the Earth's upper mantle, although lattice diffusion is still dominant for typical mantle grain sizes of 1–10 mm.

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1. Introduction

Diffusion is a key physical property, which controls a number of processes in the Earth's mantle including rock deformation, grain growth, and chemical reaction (e.g. Poirier, 1985; Karato, 2008). Diffusion of elements in mantle minerals occurs not only in the grain interior but also along the grain boundary, and grain boundary diffusion often plays an important role. For example, rheology of fine-grained olivine aggregates is governed by diffusion of silicon along a grain boundary (diffusion creep, Coble creep) (e.g. Hirth and Kohlstedt, 2003). Grain-growth in a forsterite-enstatite two-phase system is rate-limited by Mg-O coupled diffusion along a grain boundary (Tasaka and Hiraga, 2013). Thus, knowledge of diffusion, including grain boundary diffusion, in mantle minerals is of fundamental importance for understanding of the dynamic and chemical processes of the Earth's mantle.

Because olivine is the most abundant mineral in the upper mantle, accurate understanding of diffusion in olivine is highly desirable. Previous studies on diffusion of major elements in olivine revealed that $D_{\text{Si}} < D_{\text{O}} < D_{\text{Mg}}$ (D_{X} denotes diffusion coefficient for an element X) for lattice diffusion (e.g. Chakraborty et al., 1994; Dohmen et al., 2002) and $D_{\text{Si}} \sim D_{\text{Mg}} \ll D_{\text{O}}$ for grain boundary

diffusion (e.g. Watson, 1986; Farver et al., 1994; Farver and Yund, 2000) at relatively low pressures ($< \sim 2$ GPa). On the other hand, although the effect of pressure on diffusion coefficients has been reported by some studies (e.g. Chakraborty et al., 1994; Fei et al., 2012, 2014), understanding of diffusion at high-pressures corresponding to the deep upper mantle is insufficient, especially regarding grain boundary diffusion.

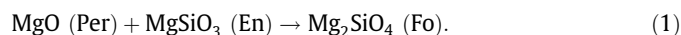
In order to constrain diffusion in mantle minerals, growth kinetics of reaction rims have been studied (e.g. Fisler and Mackwell, 1994; Gardés et al., 2011; Gardés and Heinrich, 2011; Nishi et al., 2013). In these studies, attached pairs of two minerals (reactants) were annealed at high-temperatures to yield diffusion-controlled growth of a layer of reaction product(s) between the reactants. By studying rim growth kinetics, multiple constraints for diffusion coefficients of major elements are obtainable simultaneously: position of an inert marker, which is originally placed at the interface of two reactants, provides information regarding diffusing components during the rim growth; and growth rate of the rims gives a diffusion coefficient of the rate-limiting species. The growth kinetics of orthopyroxene reaction rims between quartz and olivine at relatively low pressures (≤ 2 GPa) have been studied by many researchers (e.g. Fisler et al., 1997; Yund, 1997; Milke et al., 2001, 2007, 2009, 2013; Gardés et al., 2011, 2012; Gardés and Heinrich, 2011). In contrast to orthopyroxene, there have been no studies that have explored growth kinetics of olivine single

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layer reaction rims except for Yund (1997) who reported some data at $P = 0.7$ GPa and $T = 1273$ K. Although there are some studies on growth of olivine-orthopyroxene double layers at pressures ≤ 1.5 GPa (Götze et al., 2010; Gardés et al., 2011; Gardés and Heinrich, 2011), interpretation of double layer rim growth is complicated because the thickness of each layer is affected by diffusion in both layers.

In this study, we experimentally examined growth kinetics of forsterite (Fo), an Mg-endmember of olivine, reaction rims at a wide range of pressure, 3–11 GPa, at temperatures of 1473–1873 K. The Fo reaction rims were formed between periclase (Per) and enstatite (En) due to the following reaction:



Based on analysis of recovered samples, diffusion coefficients of rate-limiting species and their P - T dependencies were determined. Element diffusion and other related processes in the Earth's deep upper mantle are discussed.

2. Experimental procedures

2.1. Sample assembly

The annealing experiments were carried out using single crystal Per and a pre-sintered aggregate of orthoenstatite. The single crystal Per was provided by Furuuchi Chemical Corporation, Tokyo. Prior to sintering of the En aggregate, MgSiO_3 powder was synthesized from a mixture of MgO and SiO_2 reagents using 1-atm furnace at a temperature of 1773 K with a heating duration of 20 h. The En aggregate was sintered from the MgSiO_3 powder at 3 GPa and ~ 1400 K for 1 h using a multi-anvil apparatus, ORANGE-1000. The sintered En aggregate was confirmed to be orthoenstatite by X-ray diffraction. Both Per single crystals and En aggregates were shaped into discs with approximate dimensions of $\phi 1.2 \times 0.4$ mm, and one of their flat surfaces was polished with 1 μm diamond paste. The Per single crystal was oriented so that the polished surface was parallel to the (100) plane. A small amount of Pt paste was placed onto the polished surface of Per single crystals to determine the diffusing species during the reaction rim growth (Pt marker). Prior to sampling, Per single crystals and En aggregates were annealed at 900 °C for 1000 min and 1200 °C for 60 min, respectively, at 0.1 MPa in air to remove volatiles.

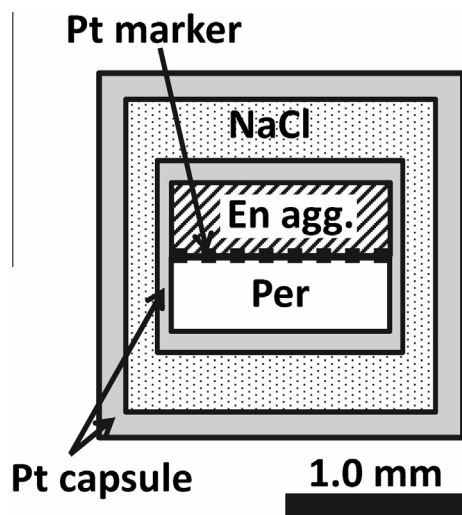


Fig. 1. Capsule design for the annealing experiments. En agg.: enstatite aggregate, Per: single crystal periclase.

Fig. 1 shows a schematic illustration of the capsule assembly used in this study. The Per and En were attached and surrounded by 20 μm thick Pt foil and then placed in NaCl powder. The NaCl was used to provide quasi-hydrostatic conditions for the sample. The outermost Pt tube (OD: 2.0 and ID: 1.8 mm, where OD and ID are outer and inner diameters, respectively) was welded to avoid possible uptake of water from surrounding ceramic parts during annealing at high pressures and high temperatures (e.g., Nishihara et al., 2006).

2.2. High pressure and high temperature experiments

The reaction rim growth experiments were carried out using the Kawai-type multi-anvil apparatus, ORANGE-1000, installed at the Geodynamics Research Center, Ehime University, Japan. The annealing experiments were conducted at pressures (P) of 3.0–11.1 GPa and temperatures (T) of 1473–1873 K for durations of 0–780 min. The experiments at $P \leq 8.0$ GPa and $P > 8.0$ GPa were carried out using 18 M/11 and 14M/8 cell assemblies, respectively, where 18M/11(14M/8) denotes (Mg,Co)O octahedral pressure medium with an 18(14) mm edge length and tungsten carbide anvils (Tungaloy, F grade, 26 mm cube) with 11(8) mm truncation edge length.

In the 18M/11 assembly, a cylindrical graphite heater (OD: 3.5 mm, ID: 3.0 mm, length: 8.7 mm), a LaCrO_3 thermal insulator, and Mo/Cu electrodes were employed. The same 18M/11 assembly was used in Matsukage and Kawasaki (2014) except that only one capsule was placed at the center of the assembly. Pressure calibration for the 18M/11 assembly was performed using phase transition of Bi at room temperature ($P = 2.55$ and 7.7 GPa) and quartz-coesite transition in SiO_2 at $T = 1473$ K (3.2 GPa, Akaogi et al., 1995) (Matsukage et al., 2013). In the 14M/8 assembly, a cylindrical LaCrO_3 heater (OD: 5.5 mm, ID: 3.5 mm, length: 8.0 mm) and a Mo/Ta electrode were employed. Pressure calibration for the 14M/8 assembly was performed using phase transition of Bi and ZnTe (6.0, 9.6 and 12.0 GPa, Kusaba et al., 1993) at room temperature, and coesite-stishovite transition in SiO_2 at $T = 1573$ K (9.5 GPa, Zhang et al., 1996), and olivine-wadsleyite transition in Mg_2SiO_4 at $T = 1573$ –1873 K (14.0–15.1 GPa, Morishima et al., 1994). Temperature in both assemblies was measured using W3Re-W25Re thermocouple without pressure correction for emf. Samples were pressurized to the target value slowly, then heated to the target temperature with a rate of ~ 50 K/min; the temperature was kept constant (for 0–780 min) and then quenched by shutting off electric power.

2.3. Microstructural and chemical analysis

Recovered samples were cut perpendicular to the sample interface using a wire-saw. Half of the cut samples were analyzed using a scanning electron microprobe equipped with a field emission gun (FE-SEM) for microstructural observation. The remaining samples were analyzed by Fourier-transform infrared (FT-IR) spectroscopy for quantification of water content.

The cut samples for the FE-SEM analysis were mounted on epoxy resin, and the surfaces were polished with 1 μm diamond paste and 50 nm colloidal silica. The polished surfaces were etched using 35% nitric acid, typically for 3 min, to make the grain boundary visible. Microstructural images were obtained using FE-SEM (JEOL JSM-7000F) and an energy-dispersive spectrometer (Oxford INCA). Operating conditions included accelerating voltages of 10 or 15 kV and probe currents of 1 or 1.5 nA. Crystallographic orientation of Fo grains in the reaction rim was analyzed based on the electron back-scattered diffraction (EBSD) technique for selected samples (Nishihara et al., 2014; Ohuchi et al., 2015). EBSD patterns were generated via interaction of a vertical incident electron beam

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