

Effects of pressure and temperature on the silicon diffusivity of pyrope-rich garnet



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ABSTRACT

We have determined the pressure and temperature dependencies of Si volume diffusion rates in a single crystal of $\text{Pyp}_{75}\text{Alm}_{15}\text{Gr}_{10}$ garnet at 6–25 GPa and 1400–1800 °C by the ^{29}Si tracer diffusion method. High-pressure experiments were conducted using the Kawai-type multi-anvil high-pressure apparatus. Diffusion profiles were obtained using secondary ion mass spectrometry in the depth-profiling mode. The Si diffusion coefficient in garnet (D_{gt}) is expressed by the Arrhenius equation: $D_{\text{gt}} = D_0 \exp(-(E^* + PV^*)/RT)$, with $\log_{10}D_0 = -7.1 \pm 1.7 \text{ m}^2/\text{s}$, $E^* = 345 \pm 56 \text{ kJ/mol}$, and $V^* = 4.7 \pm 0.5 \text{ cm}^3/\text{mol}$. Si diffusion seems to be the slowest in the major constituent elements in a pyrope-rich garnet and control rates of plastic deformation. The comparison between Si diffusion rates in garnet and wadsleyite/ringwoodite suggests that garnet has similar or slightly lower strength (at most ~2 times) compared with wadsleyite, and has similar or slightly higher strength (at most ~3 times) compared with ringwoodite at temperatures ranging from 900 to 1300 °C. Thus, strength contrast between subducted oceanic crust and underlying peridotite layer may be small at the mantle transition zone conditions. This result suggests that the separation of subducted oceanic crust from an underlying peridotite layer may not occur.

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1. Introduction

Silicate garnet is believed to be one of the major constituent minerals in the Earth's mantle. In the upper mantle, the volume fraction of garnet is about 20% (e.g., Ringwood, 1991). At the conditions present at the mantle transition zone, the pyroxene component progressively dissolves into garnet (e.g., Akaogi and Akimoto, 1977; Irifune et al., 1986), resulting in a volume fraction of garnet (majorite) of about 40% (e.g., Ringwood, 1991). In subducted oceanic crust, garnet is the most abundant mineral at mantle transition zone conditions, making up about 80% of the volume (e.g., Irifune and Ringwood, 1987). Thus, investigation of the physical properties of garnet is crucial for understanding the dynamics of the mantle and subducting slabs.

Garnet has been considered to be rheologically stronger than olivine or spinel, based on results that show the dislocation density

in garnet is less than that in coexisting olivine or spinel in deformed samples (e.g., Ando et al., 1993; Ingrin and Madon, 1995). Deformation experiments that use uniaxial compression and microhardness tests (Karato et al., 1995), Griggs-type apparatus (Katayama and Karato, 2008), and deformation-DIA apparatus (Li et al., 2006) have also demonstrated that garnet is stronger than olivine or spinel. The strength contrast between garnet and olivine/spinel has been considered as a cause for the separation of subducted oceanic crust from the underlying peridotite layer (van Keken et al., 1996; Karato, 1997). Although deformation experiments of garnet have been widely conducted, quantitative high-pressure and high-temperature deformation experiments have been conducted under conditions no deeper than those of the middle upper mantle (6.8 GPa and 1300 °C) (Li et al., 2006). Therefore, a discussion of the rheological properties of garnet at higher pressure conditions requires a large extrapolation from low pressure data.

The rheological properties of minerals can also be constrained from diffusion studies, because the slowest diffusing species in minerals control the rates of plastic deformation involving the climb-controlled dislocation creep and diffusion creep (e.g., Kirby and Raleigh, 1973). It has been demonstrated that Si is the slowest diffusing species in various silicate minerals such as quartz (Béjina

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and Jaoul, 1996; Cherniak, 2003), plagioclase (Cherniak, 2003), diopside (Béjina and Jaoul, 1996), olivine (Dohmen et al., 2002), wadsleyite (Shimojuku et al., 2009), and ringwoodite (Shimojuku et al., 2009). The slow diffusion rates of Si may be attributed to the strong covalent bonding of Si–O in silicate minerals. Thus, it is highly likely that Si diffusion also controls the rates of plastic deformation in garnet. However, to date, garnet diffusion data are limited to divalent cation diffusion (e.g., Chakraborty and Ganguly, 1992; Elphick et al., 1985; Chakraborty and Rubie, 1996; Ganguly et al., 1998; Freer and Edwards, 1999), Si–Al interdiffusion (Nishi et al., 2013; van Mierlo et al., 2013), and rare earth element diffusion (Van Orman et al., 2002; Tirone et al., 2005).

In this study, we examined the pressure and temperature dependencies of Si diffusion rates in a single crystal of pyrope-rich garnet under the conditions of the upper mantle to the uppermost lower mantle (6–25 GPa, and 1400–1800 °C). Diffusion coefficients were determined by fitting the measured diffusion profiles using a numerical simulation that takes the convolution effect into account. Based on the results, we discuss: (1) the plausible deformation mechanism of garnet in the mantle and subducted oceanic crust, and (2) the strength contrast between garnet and wadsleyite/ringwoodite and possibility of the separation of subducted oceanic crust from the underlying peridotite layer.

2. Experimental procedures

Starting material was a single crystal of natural garnet from the Czech Republic whose composition was measured to be $\text{Py}_{75(1)}\text{Alm}_{15(0)}\text{Gr}_{10(0)}$ from electron probe microanalysis, where values in parentheses represent one standard deviation. The detailed chemical composition is given in Nishi et al. (2013). The garnet was cut into several pieces with dimensions of about $500 \times 500 \times 300 \mu\text{m}$. Samples with no visible cracks were used in the diffusion experiments. The surfaces of the samples were polished with a $0.25\text{-}\mu\text{m}$ -diameter diamond compound, and subsequently polished by colloidal silica to remove the damaged surface layer. The crystallographic orientation of the garnet was not determined, because diffusivity is isotropic in cubic crystal.

High-vacuum thermal evaporation was used to coat a ^{29}Si -enriched SiO_2 ($^{28}\text{Si}:^{29}\text{Si}:^{30}\text{Si} = 0.39:99.35:0.26 \text{ at.}\%$) thin film as a diffusing source. A tungsten foil with a thickness of $25 \mu\text{m}$ was used as the heating source. The thickness of the deposited ^{29}Si -enriched SiO_2 thin film was determined to be about 70 nm by measuring the coated sample without annealing with a secondary ion mass spectrometer (SIMS).

High-pressure and high-temperature experiments were carried out using the Kawai-type multi-anvil high-pressure apparatus installed at the Institute for Study of the Earth's Interior at Okayama University. The pressure conditions were 6, 16, and 25 GPa, and the temperature conditions ranged from 1400 to 1800 °C. The OEL/TEL (octahedral edge length/truncation edge length, in mm) of the experiments at 6, 16, and 25 GPa were 18/11, 10/4, and 8/3, respectively. The pressure medium was Cr-doped MgO. The furnace was graphite for the 6 GPa experiments, and LaCrO_3 for the 16 and 25 GPa experiments. Temperature was monitored with $\text{W}_{97}\text{Re}_3\text{-W}_{75}\text{Re}_{25}$ thermocouples whose junction was set at the center of the furnace. No corrections were made for the pressure effect on emf.

The cross section of the capsule assembly is schematically shown in Fig. 1. The garnet coated with the ^{29}Si -enriched SiO_2 thin film was embedded by a fine-grained powder of CsCl (for experiments at 6 GPa) or NaCl (for experiments at 16 and 25 GPa) in a semi-sintered MgO capsule. The coated surface was surrounded with Au foil to minimize reaction with CsCl or NaCl. Ni foil and NiO powder were placed at the end of the capsule to control the

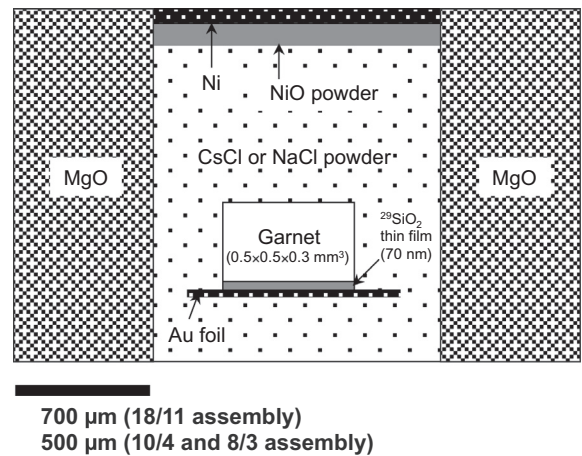


Fig. 1. Schematic illustration of the sample configuration. The garnet coated with ^{29}Si -enriched SiO_2 thin film (70 nm) was embedded by a fine-grained powder of CsCl (for experiments at 6 GPa) or NaCl (for experiments at 16 and 25 GPa). The diffusion plane was contacted with the Au foil. Ni foil and NiO powder were placed at the end of the MgO capsule to control the oxygen fugacity.

oxygen fugacity with an NNO buffer. In each experiment, the sample was first pressurized to the desired pressure (6–25 GPa) at $27 \text{ }^\circ\text{C}$ and then heated to the desired temperature (1400–1800 °C). After diffusion annealing, the sample was quenched by shutting off the power supply to the heater.

The diffusion profiles were measured by a depth-profiling mode using the SIMS (Cameca ims-6f) installed at Tohoku University. The surface was coated with Au film with a thickness of about 80 nm in order to eliminate any electronic charge on the surface. The primary ion beam was mass filtered O^- accelerated to -12 kV with a beam current of $17\text{--}40 \text{ nA}$ and defocused to have a spot size of $25 \mu\text{m}$ in diameter. Secondary ions of ^{24}Mg , ^{25}Mg , ^{26}Mg , ^{28}Si , ^{29}Si , and ^{30}Si were accelerated with 4.5 kV and measured at a mass resolving power of ~ 4000 using an electron multiplier. The counting time for all the secondary ions was 0.5 s . Secondary ion intensities were measured from the central area ($10 \times 10 \mu\text{m}$) of the sputtered crater (about $80 \times 80 \mu\text{m}$) in order to eliminate the crater edge effect.

After SIMS analysis, the depth of the crater was measured using an atomic force microscope (AFM; digital Instruments Nanoscope IIIa). The sputtering rate was estimated to be $0.34 \pm 0.08 \text{ nm}$ based on the relationship between the crater depth and the sputtering time. By assuming a constant sputtering rate, the temporal data were converted to depth data.

Depth resolution of the depth-profiling mode in SIMS can be degraded by atomic mixing and surface roughness (e.g., Hofmann, 1998). Therefore, measured depth profiles can always contain distortion. The experimentally measured concentration profile involving the distortion, $C_{\text{conv}}(y)$, is expressed as a convolution integral (e.g., Ganguly et al., 1988; Dohmen et al., 2002):

$$C_{\text{conv}}(y) = \int_{-\infty}^{+\infty} C(y)F(y' - y)dy \quad (1)$$

where y is depth, $C(y)$ is true concentration profile without distortion arising from atomic mixing and surface roughness, $F(y)$ is depth resolution function. We determined the $F(y)$ by accounting for the distortion arising from the atomic mixing and surface roughness. The effect of the atomic mixing can be derived from the depth profile of the coated sample without annealing, because the surface roughness of the coated sample without annealing is negligibly small due to the mirror polishing. It has been suggested that the

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