

Contents lists available at ScienceDirect

Earth and Planetary Science Letters



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Mg lattice diffusion in iron-free olivine and implications to conductivity anomaly in the oceanic asthenosphere

Hongzhan Fei^{a,b,*}, Sanae Koizumi^c, Naoya Sakamoto^d, Minako Hashiguchi^{e,1}, Hisayoshi Yurimoto^{d,e}, Katharina Marquardt^a, Nobuyoshi Miyajima^a, Tomoo Katsura^a

^a Bayerisches Geoinstitut, University of Bayreuth, Bayreuth D95440, Germany

^b Institute for Study of the Earth's Interior, Okayama University, Misasa, Tottori 6820193, Japan

^c Earthquake Research Institute, University of Tokyo, 1-1-1 Yayoi, Tokyo 1130032, Japan

^d Isotope Imaging Laboratory, Creative Research Institution, Hokkaido University, Sapporo 0010021, Japan

^e Department of Natural History Sciences, Hokkaido University, Sapporo 0600810, Japan

ARTICLE INFO

Article history: Received 26 July 2017 Received in revised form 27 November 2017 Accepted 11 December 2017 Available online 27 December 2017 Editor: J. Brodholt

Keywords: Mg lattice diffusion olivine ionic conductivity upper mantle

ABSTRACT

Mg lattice diffusion coefficients in iron-free olivine aggregates were measured as a function of pressure from 1 to 13 GPa, temperature from 1100 to 1300 K, and bulk water content from less than 1 up to 350 wt. ppm using multi-anvil apparatus and secondary ion mass spectrometer in depth profiling mode. The water contents in the samples were analyzed by Fourier transmission infrared spectrometer. The results show that Mg lattice diffusion coefficient increase with increasing temperature and water content, and decrease with pressure. The activation energy, water content exponent, and activation volume are 250 ± 30 kJ/mol, 1.2 ± 0.2 , and 4.3 ± 0.3 cm³/mol, respectively. Since Mg lattice diffusion controls the ionic conduction in olivine based on the Nernst-Einstein relation, the ionic conductivity in the upper mantle appears as a maximum at the top of oceanic asthenosphere due to the negative correction for pressure and positive correction for water content, which well explains the high conductivity anomaly observed at ~70–120 km depth beneath young oceanic plates.

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1. Introduction

Conductivity of ferromagnesian silicate of Earth mantle is controlled by three major mechanisms: small polaron conduction (σ_h) through the hopping of electron holes (h^{\bullet}) between ferrous and ferric iron, proton conduction (σ_p) due to the migration of free protons (H_i^{\bullet}) , and ionic conduction (σ_i) by the migration of metal vacancies (V''_{Me}) [we follow the Kröger–Vink notation (Kröger and Vink, 1956) for defect chemistry in this paper]. Magnetotelluric studies have depicted high-conductivity layers in the order of $10^{-1}-10^{-2}$ S/m at the top of asthenosphere beneath young oceanic plates (Baba et al., 2006, 2010), which is higher than σ_i and σ_h in dry olivine by more than one orders of magnitude (Karato, 1990). Another feature with the high-conductivity layers is the anisotropy, the conductivity in the direction parallel direction to the plate motion is by about 1 order of magnitude higher than that in the normal direction on the horizontal plane (Baba et al., 2006, 2010). This conductivity anomaly was once attributed to the water-enhanced σ_p in hydrous olivine (Dai and Karato, 2014a; Karato, 1990). However, recent experimental results suggested that the magnitude of σ_p is insufficient to account for the considered anomaly (Gardés et al., 2014; Yoshino et al., 2006, 2009). In addition, although carbonate melt could enhance the bulk conductivity of peridotite (Gaillard et al., 2008; Sifre et al., 2014), using the carbonate melt to explain the high-conductivity layers requires ~200–500 wt. ppm of CO₂, which is significantly higher than that in the mid-ocean ridge basalt (Dai and Karato, 2014a; Hirschmann, 2010). These points lead us to consider the role of ionic conduction in the oceanic asthenosphere.

Incorporation of water (hydrogen) into an olivine crystal structure increases defect concentrations in the metal sites (Mackwell and Kohlstedt, 1990; Nakamura and Schmalzried, 1983) and therefore will enhance Mg diffusion (Hier-Majumder et al., 2005). Based on the proportionality of σ_i and Mg diffusivity in olivine indicated by the Nernst–Einstein relation, water is expected to enhance not only σ_p but also σ_i (Hier-Majumder et al., 2005). Additionally, σ_i in olivine dominates at higher temperature than σ_h and σ_p (Yoshino et al., 2009). It may therefore play an essential role in oceanic as-

^{*} Corresponding author at: Bayerisches Geoinstitut, University of Bayreuth, Bayreuth D95440, Germany.

E-mail address: hongzhan.fei@uni-bayreuth.de (H. Fei).

¹ Present address: Research Center for Planetary Trace Organic Compounds, Kyushu University, Motooka, Nishi-ku, Fukuoka, 8190395, Japan.

thenosphere conduction due to its hydrous and high temperature conditions. Although Hier-Majumder et al. (2005) claimed that, even enhanced by water, the σ_i in olivine is still insufficient to explain the high-conductivity layers based on water effect on Mg diffusivity in olivine, the water content (fugacity) in Hier-Majumder et al. (2005) was controlled by confining pressure in the experimental set up. The water fugacity exponent and activation volume for Mg diffusivity are thus correlated parameters and cannot be determined independently although they reported the fitting values of both. Therefore, the enhancing effects of water on Mg diffusivity and σ_I are still under quantified and need to be further investigated.

It is experimentally difficult to directly determine σ_i in waterrich olivine at asthenospheric temperatures because of water loss from the sample and also the high conductivity paths produced by resulting fluid phases during σ_i measurement (Yoshino et al., 2006). To quantitatively evaluate the water effect on σ_i , here we measured the diffusivity of Mg instead of directly measuring the conductivity because Mg diffusion experiments can be performed at much lower temperatures (e.g. Hier-Majumder et al., 2005), and controlling the water content is much easier (e.g. Fei et al., 2013). than σ_i measurement experiments. Since atomic diffusion occurs both within lattice and along grain-boundaries, we obtained the effective diffusivities (lattice and grain-boundary, D_{Mg}^{lat} and D_{Mg}^{gb} , respectively) in an iron-free olivine (forsterite) as functions of pressure, temperature, and water content. This paper will focus on the experimental results of D_{Mg}^{lat} and its implications to the conductiv-ity in the upper mantle. The experimental procedures used in this study have been described elsewhere (Fei et al., 2016). We provide a brief description below.

2. Experimental and analytical methods

2.1. Starting material

Fine-grained pure iron-free olivine aggregates were synthesized from a mixture of SiO₂ and Mg(OH)₂ at ~1630 K using a vacuum furnace at The University of Tokyo (Koizumi et al., 2010). The major impurities were Al₂O₃, FeO, MnO, TiO₂, Na₂O, K₂O, and P₂O₅, with concentrations of <0.1 wt.% each examined by X-ray fluorescence (Fei et al., 2016). The average grain sizes were 0.6 µm. The initial bulk water contents ($C_{\rm bulk}^{\rm bulk}$) were <1 wt. ppm. Several samples were further heated at 1700 K, after which the average grain sizes were 2 µm (Fei et al., 2016).

2.2. Water equilibrium and diffusion annealing

Each sample was loaded into a Pt capsule together with graphite powder to prevent the sample from mechanical damage, and enstatite powder to buffer the SiO₂ activity. An additional talc + brucite mixture (4:1 weight ratio) was placed at the bottom of the capsule as a water source (Fei et al., 2013, 2016). The talc + brucite produced orthopyroxene after dehydration, which buffered the SiO₂ activity, as well. The capsules were welded by arc-welding in liquid nitrogen, loaded into the cell assembly (Fig. 1), and annealed at 8 GPa, 1100–1300 K in a multianvil apparatus for water equilibrium. The annealing duration was 4–11 h, which was sufficiently long based on the hydrogen and Mg vacancy diffusion rates in olivine (Demouchy, 2010; Demouchy and Mackwell, 2003). Dry condition experiments were performed at 1–13 GPa and 1300 K in the same method but without the talc + brucite.

The recovered samples were polished using diamond powder and an alkaline colloidal silica solution, deposited with 600– 1000 nm of 25 Mg enriched Mg₂SiO₄ thin films and 100 nm of ZrO₂ films using the pulsed laser deposition system at Ruhr-University



Fig. 1. High pressure cell assembly for water-equilibrium and diffusion experiments. The thin film for diffusion couple (green color) is only 600–1000 nm and therefore not scaled in the figure. The sample and talc + brucite water source are separated by graphite + enstatite powder. Water released from talc + brucite diffused slowly through the graphite+enstatite powder and most of the water reacted with graphite. As a result, undersaturated water conditions were generated. The water content in the sample was controlled by the ratio (talc + brucite)/(graphite + enstatite). (For interpretation of the references to color in this figure, the reader is referred to the web version of this article.)

Bochum, and then annealed again for diffusion at 1–13 GPa, 1100–1300 K, and 0.5–30 h using the same experimental set up and chemical conditions as those in the water-equilibrium experiments (Fei et al., 2013, 2016).

For precise determination of C_{H_2O} dependence of Mg diffusivity, it is essential to perform experiments under constant pressure and temperature, but various C_{H_2O} conditions, namely, water should be undersaturated in the samples. In our experiments, no free water was observed both after water equilibrium and after diffusion annealing when the capsules were opened. That is reasonable because most of water released from talc and brucite should react with graphite, leading to undersaturated conditions (Fei et al., 2013, 2016). The undersaturated condition is also confirmed by the much lower water content in our samples than the solubility in iron-free olivine (Bali et al., 2008).

Since water is undersaturated in the capsules, it is unable to know the exact oxygen fugacity in the experiments although it is for sure within the graphite stability field. Nevertheless, the Mg diffusivity in iron-free olivine is independent with oxygen fugacity (f_{O_2}) because the Mg defect concentration is f_{O_2} -independent (Smyth and Stocker, 1975), as confirmed by our additional experiments (Appendix A).

2.3. FT-IR analysis

The water contents before and after diffusion annealing were analyzed by Fourier transform infrared (FT-IR) microscopy. The water content in the lattice and bulk water in the aggregates $(C_{H_2O}^{bulk(lat)})$ and $C_{H_2O}^{lat}$, respectively) were obtained by integration of the sharp peaks and the total absorptions including the broad bands, respectively (Keppler and Rauch, 2000), from 3000 to 4000 cm⁻¹ (Fig. 2) using the Withers et al. (2012) calibration:

$$C_{\rm H_2O} = \int \frac{M \times A(\nu)}{k \times \rho \times \tau} d\nu \tag{1}$$

where $C_{\text{H}_2\text{O}}$ is the $C_{\text{H}_2\text{O}}^{\text{lat}}$ or $C_{\text{H}_2\text{O}}^{\text{bulk}}$ in wt. ppm, $A(\nu)$ is the absorption coefficient at wavenumber ν , k is the molar absorption coefficient, 45200 L/(mol cm²), M is the molecular weight of H₂O,

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