

# Grain-growth kinetics in wadsleyite: Effects of chemical environment

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## Abstract

Grain-growth kinetics in wadsleyite was investigated using a multianvil high-pressure apparatus. Fine-grained wadsleyite aggregates were synthesized by isostatic hot-pressing and were subsequently annealed under high pressure and temperature in a controlled chemical environment. Wadsleyite samples show normal grain-growth characterized by a log-normal grain-size distribution following the relation,  $L^n - L_0^n = kt$  where  $n$  is a constant,  $L$  the grain-size at time  $t$ ,  $L_0$  the grain-size at time  $t=0$  and  $k$  is a rate constant that depends on temperature  $T$  and chemical environments ( $f_{O_2}$ : oxygen fugacity in Pa,  $C_{OH}$ : water content in H/10<sup>6</sup>Si) as:

$$k = A'_D f_{O_2}^{r_D} \exp\left(\frac{-H_{D'}^*}{RT}\right) + A'_W f_{O_2}^{r_W} C_{OH}^q \exp\left(\frac{-H_{W'}^*}{RT}\right)$$

with  $A'_D = 10^{-4.9 \pm 6.1(-8.0 \pm 7.4)} (\text{m}^n \text{s}^{-1} \text{Pa}^{-r_D})$ ,  $r_D = 0.12 \pm 0.11(0.20 \pm 0.14)$ ,  $H_{D'}^* = 410 \pm 230(500 \pm 270) \text{ kJ/mol}$ ,  $A'_W = 10^{-18.2 \pm 1.4(-24.0 \pm 1.7)} (\text{m}^n \text{s}^{-1} \text{Pa}^{-r_W})$ ,  $r_W = 0.14 \pm 0.05(0.22 \pm 0.06)$ ,  $q = 1.7 \pm 0.3(2.2 \pm 0.3)$  and  $H_{W'}^* = 120 \pm 60(160 \pm 70) \text{ kJ/mol}$  with assumed value of  $n = 2(3)$  (values in parentheses denote parameters for  $n = 3$ ). Both water and oxygen fugacities significantly enhance grain-growth kinetics. The large value of the parameter describing the water fugacity dependence,  $q \sim 1.5$ – $2.5$ , cannot be explained solely by a simple model in which grain-growth is controlled by diffusion of atoms (defects) across the grain-boundaries. The interaction of grain-boundaries with charged defects or the density of hydrated ledges may be important factors that control the grain-growth kinetics of wadsleyite. When compared at similar thermo-chemical conditions, grain-growth of wadsleyite is found to be more sluggish than grain-growth of olivine. The present results show that a small wadsleyite grain-size ( $<1 \text{ mm}$ ) in subducting slabs can be maintained for a significant geological time ( $\sim 1 \text{ My}$ ) under “dry” ( $<200 \text{ H/10}^6\text{Si}$ ) conditions when the temperature is lower than  $1500 \text{ K}$ , whereas when a large amount of water ( $>100,000 \text{ H/10}^6\text{Si}$ ) is present, a small grain-size ( $<1 \text{ mm}$ ) can only be maintained for a significant time at low temperatures ( $<600 \text{ K}$ ).

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## 1. Introduction

Grain-size has an important effect on rheological properties (e.g., Karato et al., 1986). The grain-size of Earth's materials can change by a factor of  $\sim 10$ – $100$  under various environments, which may result in a vari-

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ation in viscosity of as much as several orders of magnitude. Consequently, the variation of grain-size has an important effect on mantle dynamics (e.g., Riedel and Karato, 1997; Karato et al., 2001; Solomatov, 2001; Hall and Parmentier, 2003; Solomatov et al., 2002).

Grain-growth is one of the important processes by which the grain-size of a rock may be controlled. The kinetics of grain-growth has been investigated for quartz and calcite (Tullis and Yund, 1982), olivine (Karato, 1989a; Nichols and Mackwell, 1991), anorthite (Dresen et al., 1996)  $\text{CaTiO}_3$ -perovskite + FeO wüstite (Wang et al., 1999) and silicate perovskite + magnesiowüstite (Yamazaki et al., 1996). However, no comparable studies exist for transition zone minerals, although grain-growth plays a critical role in controlling the rheology of subducting slabs (e.g., Karato et al., 2001).

Very recently, Yamazaki et al. (in press) investigated grain-growth kinetics in ringwoodite, but they did not study the influence of chemical environment, particularly that of water content. However, many physical processes in silicate minerals, including grain-growth kinetics, depend on chemical environment factors such as water fugacity, oxygen fugacity etc. (e.g., olivine: Karato, 1989b). The influence of water on grain-growth in wadsleyite (and ringwoodite) is particularly important because of the large solubility of water in these minerals (Smyth, 1987; Inoue, 1994; Kawamoto et al., 1996; Kohlstedt et al., 1996). The water content of wadsleyite in the Earth may range from nearly zero (e.g.,  $<100 \text{ H}/10^6\text{Si}$ ) to saturated ( $500,000 \text{ H}/10^6\text{Si}$ ). Therefore, it is critical to investigate the influence of water on grain-growth kinetics in this mineral.

In this paper, we present the results of the first systematic study of grain-growth kinetics in wadsleyite under controlled water (hydrogen) content and oxygen fugacity. The grain-growth kinetics is shown to strongly depend on water content, oxygen fugacity and temperature. Based on the present results, the grain-size evolution of wadsleyite in subducting slabs is discussed.

## 2. Experimental procedures

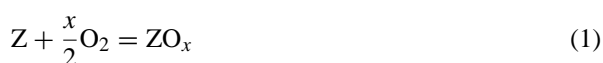
### 2.1. Generation of high-pressure and high-temperature and control of chemical environment

Experiments were performed using a KIWI 1000-ton Kawai-type multi-anvil apparatus. Pressure was generated by eight  $26 \text{ mm}^3$  tungsten carbide anvils (Toshiba F-grade) with 7 or 8 mm truncated edge length. A 5%  $\text{Cr}_2\text{O}_3$ -doped semi-sintered MgO octahedron with a 14 mm edge length was used as a pressure medium.

To reduce the temperature gradient, a stepped  $\text{LaCrO}_3$  heater was used and inserted in a  $\text{ZrO}_2$  thermal insulation sleeve. The sample capsule was electrically insulated from the furnace by an enclosing it in an MgO sleeve, and was placed in the central part of the furnace. Temperature was measured and controlled using a W5%Re–W26%Re thermocouple in a 4-bore alumina sleeve that was inserted axially into the furnace through an MgO sleeve. The hot-junction was in contact with the top part of the capsule. The temperature gradient in a capsule was estimated to be less than 80 K by means of the results of Walter et al. (1995). A Mo disk and a ring were used as electrodes. Each part of assembly, including the sample, was vacuum-dried at  $\sim 400 \text{ K}$  for at least 10 h before experiment.

The pressure was calibrated against the applied ram load using the room temperature phase transitions in Bi (2.55 and 7.7 GPa), and ZnTe (6, 9.6 and 12 GPa; Kusaba et al., 1993). The pressure calibrations at  $T = 1340$ – $1573 \text{ K}$  were performed using the coesite–stishovite transition in  $\text{SiO}_2$  (Zhang et al., 1996) and the olivine–wadsleyite transition in  $\text{Mg}_2\text{SiO}_4$  (Katsura and Ito, 1989; Morishima et al., 1994). Based on the reproducibility of these results, we consider the uncertainties in pressure in these experiments to be  $\pm 0.5 \text{ GPa}$ .

In this type of experiment, special attention must be paid to the control (or characterization) of the chemical environment (Rubie et al., 1993). We controlled oxygen fugacity using three different metal-oxide buffers (Mo– $\text{MoO}_2$ , Ni–NiO and Re– $\text{ReO}_2$ ). The control of oxygen fugacity by solid-state buffers in a multi-anvil apparatus has been well established (e.g., Rubie et al., 1993). The oxygen fugacities corresponding to thermo-chemical equilibrium for each buffer system can be calculated as follows. Consider a chemical reaction:



where Z is Mo, Ni or Re,  $x$  is a constant that depends on the valence state of Z. When the reaction (1) is in equilibrium and both metal and oxide coexist, oxygen fugacity  $f_{\text{O}_2}$  is expressed by the equilibrium constant for the reaction (1) as a function of pressure  $P$  and temperature  $T$ :

$$f_{\text{O}_2} = B^{2/x} \exp \left\{ \frac{2P(V_{\text{ZO}_x} - V_{\text{Z}})}{xRT} \right\} \quad (2)$$

where  $V_{\text{ZO}_x}$  and  $V_{\text{Z}}$  are the molar volumes of the oxide and metal, respectively, and  $B$  is an equilibrium constant at ambient pressure that depends on temperature.

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