

Anhydrous ringwoodites in the mantle transition zone: Their bulk modulus, solid solution behavior, compositional variation, and sound velocity feature

Xi Liu ^{a,b,*}, Zhihua Xiong ^{a,b}, Linlin Chang ^c, Qiang He ^{a,b}, Fei Wang ^{a,b},
Sean R. Shieh ^d, Chunming Wu ^c, Baosheng Li ^e, Lifei Zhang ^{a,b}

^a Key Laboratory of Orogenic Belts and Crustal Evolution, MOE, Peking University, Beijing 100871, China

^b School of Earth and Space Sciences, Peking University, Beijing 100871, China

^c College of Earth Science, University of Chinese Academy of Sciences, Beijing 100049, China

^d Department of Earth Sciences, University of Western Ontario, London, Ontario, N6A 5B7, Canada

^e Mineral Physics Institute, State University of New York, Stony Brook, NY 11794, USA

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Abstract

The isothermal bulk moduli of anhydrous Mg_2SiO_4 -ringwoodite (Rw) and Fe_2SiO_4 -Rw, and other 4–2 oxide spinels at ambient P - T condition have been evaluated, and empirically fitted to a model as $K_{\text{T0}} = 270.8(300) + 0.343(59) \cdot V_0 + 23.04(269) \cdot EN_{\text{-total}}$, where K_{T0} is the isothermal bulk modulus in GPa, V_0 the unit-cell volume in \AA^3 and $EN_{\text{-total}}$ the electronegativity total of all cations in the chemical formula. This model well reproduces all data used in its calibration, and may be used to predict the K_{T0} of other 4–2 oxide spinels. Combined with the generally linear volume–composition relationship of the Rw solid solutions along the join Mg_2SiO_4 – Fe_2SiO_4 , this model leads to a much smaller composition effect on the K_{T0} : $K_{\text{T0}} = 185.0(1) + 7.0(1) \cdot X_{\text{Fe}}$, where X_{Fe} is the atomic ratio $\text{Fe}/(\text{Fe} + \text{Mg})$. Furthermore, a bulk composition-independent compositional variation with P has been disclosed for the Rw at the P - T conditions of the lower part of the mantle transition zone (MTZ): $X_{\text{Fe}} = 0.222(41) - 0.0053(19) \cdot P$, with P in GPa. The nearly ideal mixing behavior, much smaller composition effect on the bulk modulus, and significant compositional variation of the Rw in the lower part of the MTZ substantially increase the gradients of the V_s - P and V_p - P profiles to generally match those constrained by the seismic reference models PREM and AK135. If there is any global low- T anomaly at the depth of 660 km, its required magnitude is most likely not larger than 200 K.

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Keywords: 4-2 spinel; Bulk modulus; Compositional variation; Mantle transition zone; Ringwoodite; Solid solution behavior; Sound velocity

1. Introduction

Due to the extremely limited availability of pristine rock samples from the deep interior of the Earth, the mineralogical model for the upper mantle is usually built by comparing the elastic data of geochemically plausible minerals to the observed seismic velocity data (Weidner and Ito, 1987; Li and

Liebermann, 2007; Irifune et al., 2008). Ringwoodite (Rw), with a conventionally-accepted composition of approximately $(\text{Mg}_{0.89}\text{Fe}_{0.11})_2\text{SiO}_4$, is commonly regarded as the most abundant mineral in the lower part of the mantle transition zone (MTZ) by the geological scientific community (Irifune and Ringwood, 1987; Ita and Stixrude, 1992). Since a complete series of Rw solid solutions exists between Mg_2SiO_4 and Fe_2SiO_4 , compositionally characterized by the parameter $X_{\text{Fe}} = \text{Fe}/(\text{Fe} + \text{Mg})$ (in molar units), the correlation between the elastic properties, used in the sound velocity calculation, and composition of the Rw might be important indicator to the compositional and mineralogical model of the MTZ. Many

* Corresponding author. School of Earth and Space Sciences, Peking University, Beijing 100871, China. Tel.: +86 10 62753585; fax: +86 10 62762996.

E-mail address: xi.liu@pku.edu.cn (X. Liu).

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efforts, both experimental and theoretical, have thus been made to constrain the elastic features of the ringwoodites with different compositions, and significant advances have been achieved in the last half century or so (e.g., Mao et al., 1969; Sato, 1977; Weidner et al., 1984; Zerr et al., 1993; Kiefer et al., 1997; Sinogeikin et al., 1998; Nishihara et al., 2004; Higo et al., 2006; Li et al., 2006; Matsui et al., 2006; Liu et al., 2008a; Nunez Valdez et al., 2012).

Isothermal bulk modulus at ambient P - T condition (K_{T0} ; GPa) is one of the most important elastic properties for understanding the density and sound velocity features of Earth materials at the P - T conditions of the Earth's interior. Although the K_{T0} value of the Mg_2SiO_4 -Rw has been reasonably well determined (e.g., Weidner et al., 1984; Hazen, 1993; Li, 2003), that of the Fe_2SiO_4 -Rw is still in discrepancy, ranging from 187.3(17) to 212(10) GPa (Mao et al., 1969; Nestola et al., 2010). As a result, the dependence of the K_{T0} on the X_{Fe} of the Rw, varying from ~15 to 36 GPa/ X_{Fe} (Weidner et al., 1984; Sinogeikin et al., 1998), is currently still a topic to debate. Another potential compositional factor which may strongly affect the bulk modulus of the Rw is its water content (e.g., Inoue et al., 1998; Ye et al., 2012). Although Rw can host up to ~2.7 wt% water (Kohlstedt et al., 1996; Bolfan-Casanova et al., 2000), it may be nearly anhydrous in the MTZ since recent studies using seismicity (Green et al., 2010), electromagnetic induction (Kelbert et al., 2009), electronic conductivity measurement (Yoshino et al., 2008), and sound velocity analysis (Mao et al., 2012) suggested that the water content in the MTZ probably does not exceed 0.1 wt%. On the other hand, the only terrestrial occurrence of Rw observed as inclusion in a diamond from Brazil indicates ~1 wt% water in the MTZ (Pearson et al., 2014), although it remains unclear whether this phenomenon has global implication or not.

Many advanced experimental methods such as direct high- P compression, ultrasonic measurement and Brillouin spectroscopy have been used to determine the isothermal bulk modulus of the Rw, and extensive and invaluable knowledge has been obtained. Direct high- P compression is commonly carried out by using a diamond-anvil cell (DAC) with a hydrostatic (usually $P < 10$ GPa) or quasihydrostatic ($P > 10$ GPa) pressure state since most liquid pressure media solidify at pressures lower than ~10 GPa (Klotz et al., 2009; Liu et al., 2011a). Due to the large isothermal bulk modulus of the Rw (~200 GPa), a hydrostatic pressure of about 10 GPa can press the Rw to about 95% of its ambient volume only, leading to relatively low accuracy in the determined isothermal bulk modulus. Ultrasonic measurement is intrinsically precise when single crystal or polycrystalline sample with homogeneous grain size, random grain orientation and full density (or zero porosity) is used (Sato, 1977; Liu et al., 2011b). Further, ultrasonic measurements can now be conducted at pressures higher than 3 GPa (Li et al., 1996, 1998; Li, 2003), at which a full elimination of any residual micropores and microcracks in the studied samples has been demonstrated (Liebermann, 1975; Liebermann et al., 1977; Rigden and Jackson, 1991). Ideally, Brillouin spectroscopy measurements can fully constrain the elastic tensors of a phase (Weidner et al., 1984; Wang et al., 2003a). Other experimental

methods have been used as well; for a recent review, see Li and Liebermann (2014). On the other hand, it is well known that there are some specific difficulties with the synthetic Rw samples. Ringwoodites synthesized at different P - T conditions with different experimental durations might contain different amounts and types of defects (Smyth et al., 2003), and attain different magnitudes of order-disorder phenomenon (Hazen et al., 1993; Kudoh et al., 2000; O'Neill et al., 2003). They might have different quantities of contaminant water in their structures (Higo et al., 2006), since hydrogen can easily penetrate through most experimental capsule materials (Liu et al., 2006) and subsequently enter the Rw structure. If the dependence of the isothermal bulk modulus of the Rw on the X_{Fe} variable is relatively small, which is probably the case (Mao et al., 1969; Finger et al., 1986), all these experimental complexities then would add together to prevent a very accurate determination of this dependence and lead to controversial results (Sinogeikin et al., 1998; Higo et al., 2006).

One way around this problem is to build an empirical model which relates the bulk moduli to some other physical properties of some materials with the same crystal structure but different compositions (Anderson and Anderson, 1970; Duffy and Anderson, 1989). By extending the ranges of all the relevant properties, those uncertainties in the employed experimental techniques and variations of the physical and chemical states of the investigated samples become unimportant, and the correlation of the physical properties can be accurately established. Rw is crystallographically a cubic 4–2 oxide spinel ($Fd\bar{3}m$ with $Z = 8$) (Yagi et al., 1974; Sasaki et al., 1982), and shares the same crystal structure with a large number of phases such as Ni_2SiO_4 -Sp (spinel), Co_2SiO_4 -Sp, Mg_2GeO_4 -Sp, Fe_2GeO_4 -Sp, Co_2GeO_4 -Sp, Mg_2TiO_4 -Sp, Fe_2TiO_4 -Sp, Co_2TiO_4 -Sp and Zn_2TiO_4 -Sp. Because of the similarity in the crystal structures, these spinels have been widely used as analogues to explore the high- P physical properties of Mg_2SiO_4 -Rw (Mao et al., 1970; Liu et al., 1974; Liebermann, 1975; Liebermann et al., 1977; Sato, 1977; Finger et al., 1979; Weidner and Hamaya, 1983; Bass et al., 1984; Rigden et al., 1988; Rigden and Jackson, 1991).

This study aims at evaluating the isothermal bulk moduli of the Mg_2SiO_4 -Rw and Fe_2SiO_4 -Rw, and those cubic 4–2 oxide spinels obtained by different experimental techniques in recent years, and building an empirical model to describe the relationship between the isothermal bulk moduli and other physical properties such as the unit-cell volume and the electronegativity of the cations. It has been demonstrated that such an empirical model can reproduce well all the data used in its calibration. In addition, this model has been examined with some extra experimental data which have not been used in its construction, and satisfactory agreement has been achieved as well. To apply this model to the Rw solid solutions in the system Mg_2SiO_4 - Fe_2SiO_4 , a well-established volume–composition relationship is a prerequisite. We have summarized all the Rw volume–composition data reported so far, and found a nearly linear solid solution behavior for the Rw solid solutions. Eventually a much smaller composition effect on the bulk modulus of the Rw has been revealed.

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