



# Is the mantle chemically stratified? Insights from sound velocity modeling and isotope evolution of an early magma ocean



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## ABSTRACT

The upper mantle is widely accepted to be pyrolitic, but the bulk composition of the lower mantle remains highly disputed. Recent modeling of the lower mantle shear wave velocity profile has suggested that the lower mantle is enriched in bridgmanite, therefore implying a higher Si/Mg than that of the upper mantle. We improve upon such modeling by taking into account Ca-perovskite and considering the distribution of Fe between bridgmanite and ferropericlase, more appropriate for Al-bearing systems. Using available experimental data, we derive a means to constrain Fe–Mg partitioning for bridgmanite and ferropericlase, constrain suitable values for the lower mantle, and apply these to lower mantle shear wave velocity calculations. Calculations that consider the effects of minor chemical components such as Ca and Al suggest that the lower mantle shear wave velocities can resolve PREM for a pyrolitic composition to within 1%. We also model chemical fractionations of the <sup>147</sup>Sm–<sup>143</sup>Nd and <sup>176</sup>Lu–<sup>176</sup>Hf systems induced by a crystallizing magma ocean that would produce a putative Si-enriched lower mantle. The comparison of the calculated <sup>143</sup>Nd/<sup>144</sup>Nd and <sup>176</sup>Hf/<sup>177</sup>Hf signatures with those of the terrestrial array shows that a Si-enriched lower mantle, if ever formed, no longer exists. Both mineralogical calculations and geochemical magma ocean modeling support the idea that the Earth's lower mantle is likely pyrolitic and that the mantle as a whole need not be chemically stratified.

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## 1. Introduction

The solid Earth is composed of three major chemical layers – the crust, the mantle, and the core. These features were first identified by analyzing seismic waves generated by large-scale earthquakes, penetrating into the interior of the Earth (e.g., Williamson and Adams, 1923). Later, when the physical properties of putative inner Earth materials were measured in high pressure and temperature experiments, inner Earth seismic wave velocities and density profiles were linked to the chemical compositions of these layers (e.g., Birch, 1952).

The silicate mantle is the Earth's volumetrically largest division. Its chemical composition and structure are a constant subject of inquiry and debate as they tie into many issues pertaining to the origin of the Earth and its evolution (e.g., Javoy et al., 2010). One of the most prominent seismic features of the mantle is the 660-km discontinuity, characterized by a sharp jump in seismic velocity. This discontinuity, which serves as the boundary between the upper and lower mantles, has traditionally been attributed

to the temperature and pressure-induced breakup of ringwoodite, (Mg,Fe)<sub>2</sub>SiO<sub>4</sub>, into bridgmanite, (Mg,Fe)SiO<sub>3</sub> (formerly referred to as “Mg-silicate-perovskite”), and ferropericlase, (Mg,Fe)O (Ito and Takahashi, 1989).

There is yet no consensus on whether this discontinuity is also a chemical boundary (e.g., Irifune et al., 2010; Murakami et al., 2012), which would imply different compositions for the lower and upper mantles. The upper mantle composition is well established from samples of mid-ocean ridge volcanism and mantle peridotites, and is widely presumed to be pyrolitic (McDonough and Sun, 1995). Pyrolite, a theoretical composition calculated as three parts peridotite and one part normal mid-ocean ridge basalt (Ringwood, 1962), is depleted in Si compared to chondrites. Chondrites are often considered to be the building blocks of the Earth. If the bulk Earth has a CI-chondritic Si/Mg ratio, then the compensation for the apparent depletion of Si in the upper mantle is explained either by sequestering of some Si into the core (e.g., Allegre et al., 1995; Georg et al., 2007), or having a higher Si/Mg ratio in the lower mantle (e.g., Taylor and McLennan, 1985), or both.

Such a chemical stratification in the mantle, if it exists, would have most likely been caused by the gravitational settling of dense bridgmanite crystals during the solidification of a magma ocean

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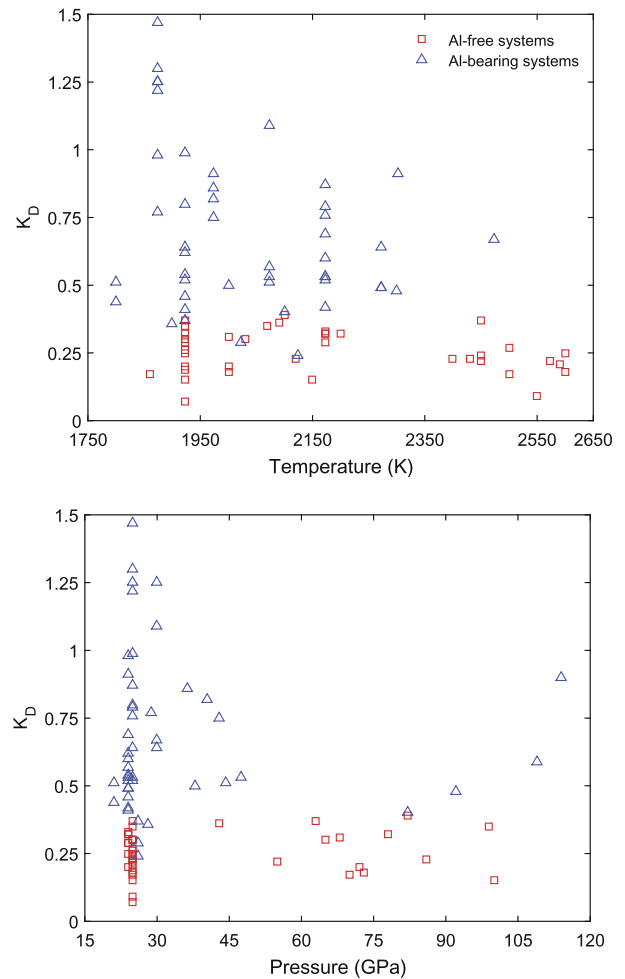
not long after the Earth formation (Agee and Walker, 1988). The preservation of such a chemically layered mantle throughout billions of years would imply a lack of material transport between the upper and lower mantles throughout the history of the Earth. Although seismic tomography has revealed slabs penetrating the 660-km discontinuity (Van der Hilst et al., 1997), the extent of material exchange between the upper and lower mantles remains unclear.

The composition of the lower mantle has been evaluated by modeling sound velocity profiles using experimentally derived physical properties of mantle minerals under proper  $P$ - $T$  conditions (Lee et al., 2004; Stixrude et al., 1992). However, in most experiments, the chemical composition of the lower mantle is approximated by the Fe–Mg–Si–O system, neglecting the less abundant major elements such as Al and Ca. The effect of Al on the shear properties of bridgmanite, having only been explored in the past decade (Jackson, 2004; Murakami et al., 2012), has profound implications for interpreting the lower mantle composition. Using experimental data obtained under lower mantle  $P$ - $T$  conditions, recent modeled seismic shear wave velocities of a pyrolitic lower mantle (Murakami et al., 2012) were found to have large discrepancies from the shear wave velocities of the 1D seismic profile, PREM (Dziewonski and Anderson, 1981). Specifically, the modeled shear wave velocity profile for a pyrolitic lower mantle with Al-bridgmanite was much lower than PREM ( $\sim 3\%$  offset). The difference was resolved by increasing the bridgmanite/ferropericase ratio in the lower mantle compared to the pyrolitic composition, leading Murakami et al. (2012) to infer a Si-enriched lower mantle.

The approach of Murakami et al. (2012) neglects the contribution of a Ca-bearing phase and the effect of Al on Fe–Mg partitioning between bridgmanite and ferropericase, which in turn, affects the sound wave speeds of these minerals. Here, we model the shear wave velocities of the lower mantle, taking these effects into account. The pyrolitic mantle contains  $\sim 3$ – $4$  wt% CaO (e.g., McDonough and Sun, 1995), which in the lower mantle resides in Ca-perovskite,  $\text{CaSiO}_3$ , occurring alongside two other minerals, bridgmanite and ferropericase. Although the shear properties of Ca-perovskite have not been measured experimentally, *ab initio* calculations (e.g., Karki and Crain, 1998) of cubic Ca-perovskite have demonstrated high shear velocities which can potentially resolve the discrepancy between a pyrolitic lower mantle and observed seismic wave velocities.

Calculations of lower mantle sound wave velocities usually consider bridgmanite and ferropericase with Fe–Mg partitioning characteristic of Al-free systems (e.g., Zhang et al., 2013). Al, however, residing exclusively in bridgmanite, affects Fe–Mg partitioning to a substantial degree. Although Al is reported to increase the amount of Fe in bridgmanite due to the coupled substitution of ( $\text{Fe}^{3+}$  and  $\text{Al}^{3+}$ ) for ( $\text{Mg}^{2+}$  and  $\text{Si}^{4+}$ ) (Wood and Rubie, 1996), the extreme variability in the data (Fig. 1) has made the subject controversial. In particular, relations among pressure, temperature and Fe–Mg partitioning behavior in coexisting Al-bearing bridgmanite and ferropericase have so far remained unclear. Another possible complication is the spin transition occurring in ferropericase, a high pressure effect involving a change in an electron configuration from high spin to low, which subsequently affects both the bulk and shear properties of ferropericase. As Fe–Mg partitioning between Al-bridgmanite and ferropericase in the lower mantle can affect shear wave velocity calculations, there is a necessity to critically assess and analyze the available experimental data. Through such an analysis, we present a statistically significant correlation that is capable of predicting  $K_D$  values, use this relation to derive  $K_D$  suitable for the lower mantle, and apply these to shear wave velocity calculations.

In addition, our study also explores the fractionation of lithophile trace elements in a putative Si-enriched lower mantle that



**Fig. 1.** Variations of  $K_D$  values as defined in Eq. (5), with temperature and pressure. Blue, upward-pointing triangles and red squares denote data for Al-bearing (Frost and Langenhorst, 2002; Irifune et al., 2010; McCammon et al., 2004; Murakami, 2005; Nishiyama and Yagi, 2003; Sinmyo and Hirose, 2013; Wood and Rubie, 1996; Wood, 2000) and Al-free systems (Auzende et al., 2008; Frost and Langenhorst, 2002; Nakajima et al., 2012; Sakai et al., 2009), respectively. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

would have formed in a differentiating magma ocean, by taking advantage of primordial fractionation effects in the  $^{176}\text{Lu}$ – $^{176}\text{Hf}$  and  $^{147}\text{Sm}$ – $^{143}\text{Nd}$  radiogenic isotope systems between a silicate melt and high-pressure lower mantle phases.

Using approaches from mineral physics and geochemistry, we evaluate the likelihood of a chemically stratified mantle, with the lower mantle being enriched in Si compared to the upper mantle.

## 2. Methods

### 2.1. Shear wave velocity modeling

Modeling a seismic velocity profile of a planetary layer such as the lower mantle involves a series of calculations for different depths of the layer. Each step calculates the compound shear wave velocities for a mineral aggregate representing a bulk chemical composition at a given pressure and temperature. For the calculations of the aggregate shear wave velocity of a pyrolitic lower mantle, the mineral proportions of lower mantle minerals ( $\sim 77.5$  vol% bridgmanite [ $\text{Al}-(\text{Mg}, \text{Fe})\text{SiO}_3$ ]– $15$  vol% ferropericase [ $(\text{Mg}, \text{Fe})\text{O}$ ], and  $\sim 7.5$  vol% Ca-perovskite [ $\text{CaSiO}_3$ ]) (e.g., Irifune, 1994) are used. Since the amount of Fe in ferropericase or bridgmanite affects the wt% proportions of these minerals, we consider minor variations in the volume and weight proportions of lower

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