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Doctoral Thesis

**Chemical evolution and stratification
of the primordial mantle and core**

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Abstract

The primordial Earth is supposed to be fully molten after moon-forming giant impact. Crystallization and chemical evolution of the mantle and the core is important to understand chemical structure of the primordial mantle and core, and to elucidate the origin of geochemical signatures formed in the early Earth. Based on melting experiments over core-mantle boundary pressure conditions using laser-heated diamond anvil cell techniques, (1) melting (solidus) temperature of pyrolite lower mantle, (2) partitioning of iron and density contrast between silicate solid and melt in $(\text{Mg}_{0.89}\text{Fe}_{0.11})_2\text{SiO}_4$ bulk composition, (3) fractional crystallization of a basal magma ocean, and (4) partitioning of lithophile elements between molten iron and silicate melt were investigated. With these experimental data I construct a forward model on the co-evolution of the mantle and the core. (*127 words*)

Thesis summary

The thesis entitled “Chemical evolution and stratification of the primordial mantle and core” is composed by six chapters.

In chapter 1, the results and problems of the previous studies, aim of this thesis on the chemical aspect of Earth’s formation and evolution are summarized. The primordial Earth is supposed to be fully molten immediate aftermath the moon-forming giant impact. Crystallization and chemical differentiation of the mantle and the core are important to understand chemical structure of the primordial mantle and core, and to elucidate the origin of geochemical signatures formed in the early Earth. However, experimental studies have been limited to relatively low pressure (P) and temperature (T) conditions by multi-anvil press experiments. In this study, I developed and performed various melting experiments using laser-heated diamond anvil cell techniques over core-mantle boundary pressure conditions.

In chapter 2, the results of melting experiments performed on samples with bulk composition $(\text{Mg}_{0.89}\text{Fe}_{0.11})_2\text{SiO}_4$ at pressures from 20 to 159 GPa are reported. A precipitous change in Fe-Mg distribution coefficient between solid silicate and melt at pressures greater than ~ 76 GPa was found. These results imply that $(\text{Mg,Fe})\text{SiO}_3$ liquid becomes more dense than coexisting solid at ~ 1800 km depth in the lower mantle. $(\text{Mg,Fe})\text{SiO}_3$ perovskite is on the liquidus at deep mantle condition and predict that fractional crystallization of dense magma would have toward an iron-rich and

silicon-poor composition, consistent with seismic inferences of structures in the core-mantle boundary region.

In chapter 3, serial crystallization experiments on pyrolite were performed at ~75 GPa and ~135 GPa in order to understand crystallization processes and chemical consequences of a magma ocean on cooling of the fully molten mantle immediate aftermath the giant impact. The cotectic composition of Mg-rich perovskite and ferropericlasite in pyrolitic system was determined to (Mg+Fe)/Si molar ratio of 2.71 at ~75 GPa. The density crossover between silicate melt and solid should occur at least after 35 % crystallization of silicate magma. Possible chemical stratification in the primordial mantle was modeled based on the results obtained in these experiments.

In chapter 4, the partitioning of lithophile elements (K, O, Si, Mg, Al, and Ca) between liquid metal and silicate melt was investigated up to 138 GPa and 5450 K in S-free/S-bearing Fe + K-doped pyrolite system, in order to constrain the amounts of radioactive ⁴⁰K and other light elements in the core. The obtained iron-potassium exchange coefficients show strong temperature dependence but negligible effects of pressure and sulfur content, not supporting the transition-metal-like behavior of potassium at high pressure. As a consequence, the present experiments suggest only ~10 ppm potassium in the core, which yields present-day heat production of ~0.1 TW, even when we assume the entire core-mantle chemical equilibrium at 136 GPa and 5300 K (liquidus of pyrolitic mantle). On the other hand, the core dissolves substantial amounts

of silicon and oxygen as a result of reaction with a basal magma ocean at 4500-5000 K, which account for the 10% core density deficit. In addition, quenched liquid iron obtained in relatively high-temperature experiments included certain amounts of Mg, Al, and Ca, suggesting that these elements may have been once incorporated into the core at the time of giant impact. A more realistic model for the Earth's core must consider both materials that equilibrated at modest P - T and those added at very high P - T , and the effect of latter components are focused in this chapter.

In chapter 5, I showed that the solidus temperature of a primitive (pyrolitic) mantle is as low as 3570 ± 200 kelvin at the core-mantle boundary (CMB), based on the observation of small amounts of partial melt by three-dimensional x-ray micro-tomography imaging. It gives the upper bound of the CMB temperature (T_{CMB}) since the mantle side of the CMB is not globally molten. Such remarkably low T_{CMB} implies that post-perovskite is present not only in cold regions but in wide areas of the lowermost mantle. The low T_{CMB} also requires that the melting temperature of outer core alloy is depressed largely by the impurity effect, suggesting that hydrogen is an important alloying element.

In chapter 6, the chemical model on the coevolution of the primordial mantle and the core was constructed based on the results on mantle crystallization (chapter 2 and 3), and on chemical reaction between liquid core and molten mantle (chapter 4). The additional constraint of the large amount of hydrogen in the core (chapter 5) strongly restricts the style of the formation and chemical coevolution of the mantle and the core

in the primordial Earth. Furthermore, the kind of building blocks of the Earth was discussed from the aspect of view of major element compositions of the modeled bulk Earth.