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Outline of doctoral disertation

“Sound velocity and density of liquid Fe-Ni-S alloy at high pressure”

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This doctoral thesis is organized into 5 chapters for the density and sound velocity of liquid Fe-Ni-S alloys, 2 appendixes for high-pressure stability and compressibility of two aluminum-rich phases, hexagonal new aluminum-rich phase and calcium-ferrite type phase, in subducted mid-oceanic ridge basalt (appendix 1, 2), and one appendix for thermal conductivity of MgO at high pressure.

The liquid Earth’s outer core is mainly constructed of iron (Fe)-nickel (Ni) alloy. The Earth’s core was found to be less dense than pure iron comparing the core density with density of solid pure iron estimated from its equation of state, which is called core density deficit problem. From cosmochemical abundances and geochemical model, hydrogen (H), carbon (C), nitrogen (N), oxygen (O), silicon (Si), and sulfur (S) are thought to be possible candidates to induce the core density deficit. The density contrast between the outer core and the pure iron has been estimated to be 5-10%, depending on the assuming outer core geotherm. Both the nature and the amount of core light elements have been left as one of the biggest enigma of the Earth science for more than 60 years. In this thesis, we focused on sulfur as a light element in the core. Sulfur has high solubility to liquid iron even at low pressure and missing in the mantle compared to the other volatile elements. This implies that sulfur has been trapped into the liquid iron during metal-silicate partitioning in magma ocean.

Through this thesis, we have investigated density and sound velocity of liquid $(Fe,Ni)_3S$ under high pressure and high temperature conditions to constrain the chemical composition of the Earth and the other planetary cores.

Chapter 1: "General introduction: The light components in the Earth's outer core and liquid planetary cores"

We introduce the history of research for the chemical composition of the Earth's core, density and sound velocity of pure iron as a reference to discuss the density deficit problem. In addition, we review previous works which investigated the phase relations, physical properties, and chemical properties of the iron-light element(s) alloys.

Chapter 2: "Basis and data analysis of density measurements from X-ray diffraction and sound velocity measurements from inelastic X-ray scattering spectroscopy"

We first mention principles of density determination from diffused scattering in X-ray diffraction patterns and sound velocity measurements via inelastic X-ray scattering spectroscopy. We also introduce experimental development to achieve sound velocity measurement on liquid iron alloy at high pressure in diamond anvil cell (DAC) high pressure apparatus.

Chapter 3: "Density measurements of liquid Fe-Ni-S alloy at high pressure and temperature"

We report density of liquid $Fe_{46.5}Ni_{28.5}S_{25}$ measured in a pressure range of 25-70 GPa at 3200 K by X-ray diffraction method using laser-heated DAC. Obtained density data were fit to the third-order Vinet equation of state, and determined bulk modulus and its pressure derivative at 1 bar to be K_0=17.05(626) GPa and K_0'=6.96(150) with fixed ρ_0 =4680 kg/m^3. Our data indicate that alloying 25 atm% sulfur reduced density about 14% from liquid pure iron.

Chapter 4: "Sound velocity of liquid Fe-Ni-S at high pressure and temperature via inelastic X-ray scattering spectroscopy"

We describe the project on sound velocity measurements of liquid $(Fe,Ni)_3S$ via high-resolution inelastic X-ray scattering spectroscopy in DAC up to 46 GPa. We determined density dependence of sound velocity of liquid $(Fe,Ni)_3S$ using density data obtained in Chapter 3. In this study, significant temperature effect on sound velocity was not observed in the experimental temperature range from 1500 K to 2500 K. Such a small temperature dependence is consistent with the conclusion in previous theoretical studies. Our study reveals that alloying sulfur increases the sound velocity of liquid iron-nickel alloys.

Chapter 5: “Implication for the composition of liquid planetary core”

The density and sound velocity of liquid (Fe,Ni)3S were estimated under the Earth’s outer core pressure-temperature conditions, combining with present density and sound velocity data, and thermal expansion coefficient proposed in previous study. It was found that the sound velocity of liquid (Fe,Ni)3S is slightly faster than P wave speed of PREM, and density of liquid (Fe,Ni)3S is almost consistent with the density of the Earth’s outer core. Only Fe-Ni-S system can not explain the density and sound velocity of the outer core perfectly. Sulfur strongly partition in liquid iron in equilibrium of silicate melt and iron alloy, amd has been found in iron meteorite as FeS troilite, iron-sulfur alloy. Sulfur is depleted silicate Earth form CI chondrite, which is accepted as a building block of the Earth. In addition, liquid Fe-S alloy has a similar bulk modulus to that of the outer core. Therefore, though there should be the other light elements to explain both the density and sound velocity in the outer core, we conclude that sulfur is a major light element in the Earth’s core from the consideration of geochemistry and physical properties of liquid iron-the other light element(s) proposed in previou works. The combination technique of inelastic X-ray scattering spectroscopy and diamond-anvil cell that we developed will provide sound velocity data of liquid iron alloys with various chemical compositions. Mars has long been believed to have sulfur enriched core relative to the Earth. We also consider the Martian interior model using our density data of liquid $(Fe,Ni)_3S$ combined with estimated mineral assemblages in the Martian mantle. Our modeling indicates magnesium silicate perovskite layer would be a few tens kilometer thick. Such thin magnesium silicate perovskite layer would have little influence to the thermal convection in the Martian mantle. We also calculate the heat flow at the core-mantle boundary to be 0.4 TW using thermal conductivity of MgO (Appendix 3), magnesium silicate perovskite, and garnet.

Appendix 1: “Stabilities of NAL and Ca-ferrite type phases on the join $NaAlSiO_4$-$MgAl_2O_4$ at high pressure”

Imada, S., Hirose, K., & Ohishi, Y. (2011). Stabilities of NAL and Ca-ferrite-type phases on the join NaAlSiO4-MgAl2O4 at high pressure. Physics and Chemistry of Minerals, 38(7), 557-560.

Stabilities of hexagonal new aluminous (NAL) phase and Ca-ferrite-type (CF) phase were investigated on the join $NaAlSiO_4$-$MgAl_2O_4$ in a pressure range from 23 to 58 GPa at approximately constant temperature of 1850 K, on the basis of in-situ synchrotron X-ray diffraction measurements in a laser-heated diamond-anvil cell. The results show that NAL is formed as a single phase up to 34 GPa, NAL + CF between 34 and 43 GPa, and only CF at higher pressures in 40%$NaAlSiO_4$-60%$MgAl_2O_4$ bulk composition. On the other hand, both NAL and CF coexist below 38 and 36 GPa and only CF was obtained at higher pressures in 60%$NaAlSiO_4$-40%$MgAl_2O_4$ and 20%$NaAlSiO_4$-80%$MgAl_2O_4$ composition, respectively. These results indicate that NAL appears only up to 46 GPa at 1850 K and CF forms continuous solid solution at higher pressures on the join $NaAlSiO_4$-$MgAl_2O_4$. NAL has limited stability in subducted mid-oceanic ridge basalt crust in the Earth's lower mantle and undergoes a phase transition to CF in deeper levels.

Appemdix 2: "Compression of Na0.4Mg0.6Al1.6Si0.4O4 NAL and Ca-ferrite type phases"

Imada, S., Hirose, K., Komabayashi, T., Suzuki, T., & Ohishi, Y. (2012). Compression of Na0.4Mg0.6Al1.6Si0.4O4 NAL and Ca-ferrite-type phases.Physics and Chemistry of Minerals, 39(7), 525-530.

Compression behaviors of two Al-rich phases in the lower mantle, hexagonal new aluminum-rich (NAL) phase and its high-pressure polymorph Ca-ferrite-type (CF) phase, were examined for identical $Na_{0.4}Mg_{0.6}Al_{1.6}Si_{0.4}O_4$ (40%$NaAlSiO_4$–60%$MgAl_2O_4$) composition. The volumes of the NAL and CF phases were obtained at room temperature up to 31 and 134 GPa, respectively, by a combination of laser-annealed diamond-anvil cell techniques and synchrotron X-ray diffraction measurements. Fitting of such pressure-volume data to the third-order Birch-Murnaghan equation of state yields bulk modulus K_0 = 199(6) GPa at 1 bar and its pressure derivative K_0' = 5.0(6) for the NAL phase and K_0 = 169(5) GPa and K_0' = 6.3(3) for the CF phase. These results indicate that the bulk modulus increases from 397 to 407 GPa across the phase transition from the NAL to CF phase at 43 GPa, where the NAL phase completely transforms into the CF phase on $Na_{0.4}Mg_{0.6}Al_{1.6}Si_{0.4}O_4$. Density also increases by 2.1 % at the phase transition.

Appemdix 3: "Measurements of lattice thermal conductivity of MgO to core-mantle boundary pressures"

Imada, S., Ohta, K., Yagi, T., Hirose, K., Yoshida, H., & Nagahara, H. (2014). Measurements of lattice thermal conductivity of MgO to core-mantle boundary pressures. Geophysical Research Letters, 41(13), 4542-4547.

The pressure response of lattice thermal conduction in MgO periclase has been a matter of interest for many decades to estimate the thermal conductivity profile of the lower mantle. Using the pulsed light heating thermoreflectance technique, we measured the lattice thermal diffusivity of MgO at pressures up to 137 GPa at 300 K to determine its lattice thermal conductivity under deep lower mantle conditions. Considering the temperature effect estimated by previous high-temperature measurements, we calculated the lattice part of the thermal conductivity of MgO to be 17.9 ± 1.1 W/m/K at 135 GPa and 3600 K. Additionally, we observed that the lattice conductivity of MgO has little dependence on its grain size under core-mantle boundary conditions.