

氏名	PUREVJAV Narangoo
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学位論文の題目	Hydration mechanism in Earth's mantle transition zone investigated by neutron diffraction (中性子回折による地球マントル遷移層鉱物中の水素の解析)
論文審査委員	准教授 奥地 拓生 教授 神崎 正美 教授 河村 雄行 准教授 山下 茂 准教授 Jung-Fu Lin (University of Texas at Austin)

学位論文内容の要旨

The Earth's mantle transition zone at depths from 410 to 660 km is considered to be the largest reservoir of water transported by subduction of oceanic plates. This is because its major constituent minerals, wadsleyite and ringwoodite, are capable of storing up to 3 wt.% water in their crystal structures. This is their unique property compared with the other mantle constituent minerals. Water shows significant effects on the physical properties of wadsleyite and ringwoodite such as increasing their electrical conductivity and reducing their elasticity. In order to understand the effect of water on these physical properties, hydrogen positions and hydration mechanisms need to be understood in the relevant minerals structures. Neutron diffraction is the most direct way to probe the hydrogen in condensed matter. Thus here we conducted neutron diffraction for hydrous ringwoodite and hydrous wadsleyite. Several types of the qualified samples suitable for neutron diffraction measurements were separately synthesized using a scaled-up Kawai type cell. The synthesis procedures and the sample characterization procedures are shown in detail in this thesis. We successfully measured our D₂O-bearing ringwoodite sample by time-of-flight powder neutron diffraction at J-PARC spallation neutron source facility. By this study, for the first time we demonstrated the nature of hydrogen positions and cation exchanging mechanisms in hydrous ringwoodite structure. We found that the deuterium atoms exchanged with magnesium, iron and silicon, which were occupying both of octahedral and tetrahedral sites. The result of this study was reconfirmed by single-crystal neutron diffraction that was conducted at Oak Ridge National Laboratory (ORNL), where a single crystal of H₂O-bearing ringwoodite with 600 μm in size was measured. We successfully refined the hydrogen position and occupancy in the crystal structure of hydrogenated ringwoodite. We also applied single-crystal neutron diffraction for H₂O-bearing wadsleyite. The measurement was also carried out at ORNL, where H₂O-bearing wadsleyite crystal with 800 μm in size was measured. By the study, we demonstrated that hydrogen in wadsleyite exchanged only with magnesium at M3 octahedral site, while it did not exchange with the other magnesium and silicon cations. These single-crystal diffraction works were its first applications for deep mantle hydrous minerals. From these studies, we conclude that hydration mechanisms between hydrous wadsleyite and hydrous ringwoodite are qualitatively different each other. Thus their observed differences in physical properties are primarily induced by their distinct hydration mechanisms in their crystal structures.

論文審査結果の要旨

This study is the first systematic application of time-of-flight neutron diffraction for analyzing hydrated structures of the two major mantle transition minerals, wadsleyite and ringwoodite. The author successfully analyzed the crystal structures of deuterated and hydrogenated ringwoodite. She also successfully analyzed the crystal structure of hydrogenated wadsleyite. These were all made for the first time. To conduct these works, the recently-established couple of the strongest spallation neutron sources in Japan and USA were combined with a newly-developed synthesis scheme of high-quality hydrous mineral crystals, where large and homogeneous sample crystals were grown at high pressure and high temperature deep mantle conditions.

Water plays a crucial role for the evolution of the Earth. It exists as the oceans on the Earth's surface, as well as hydroxyls within hydrous minerals in its deep interior. The surface water is transported into the deep interior of the Earth by plate tectonics. Mantle transition zone at depths from 410 to 660 km is considered to be the largest reservoir of the transported water, which consists mainly of wadsleyite and ringwoodite mineral phases. Wadsleyite and ringwoodite can accept ~3 wt% of water within their crystal structures, while the absorbed water significantly affects the physical properties of these minerals, such as increasing their electrical conductivities, reducing their elastic constants, and modifying their phase boundaries. In order to understand microscopic processes induced by the addition of water for affecting these properties, hydrogen sites and hydration mechanisms are needed to be analyzed in the relevant crystal structures.

The results outlined above provide novel insights for the role of water in the two representative minerals in the Earth's mantle transition zone. In particular, it was newly established that wadsleyite and ringwoodite showed a clear contrast in their cation species to be exchanging with hydrogen. The representative results for wadsleyite and ringwoodite were separately published in two peer-reviewed international scientific journals. Based on these achievements, we, the dissertation committee, recommend that Purevjav Narangoo be awarded the degree of Doctor of Philosophy in Science.