SMP06-P01　　　　　会場:3 階ポスター会場　　　　　時間:4 月 28 日 18:15-19:30

Hydrogen positions in hydrous ringwoodite determined by pulsed neutron powder diffraction
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The transition zone in the Earth's mantle has been considered potentially large water reservoir. It was experimentally evidenced that its main constituent minerals can uptake significant amount of water as hydroxyl groups in their crystal structures. The ringwoodite $[(\gamma\text{-Mg,Fe})_2SiO_4]$ is one of the high pressure polymorph of olivine, which is the most major phase in the lower part of the transition zone, between 525 to 660 km in depth. It was reported that ringwoodite can incorporate up to 2.6 wt.% of water (Kohlstedt et al., 1996). The hydration of ringwoodite strongly affects its physical and chemical properties such as electrical conductivity, compressibility and seismic velocities. However, crystallographic sites of hydrogen and its incorporation mechanism are still unclear mainly due to insensitiveness of X-ray probe for hydrogen. The previous studies of its structure refinement by X-ray diffraction demonstrated that hydrous ringwoodite has cubic spinel structure with Fd-$3m$ space group (Kudoh et al., 2000; Smyth et al., 2003). Here we applied neutron diffraction for hydrous ringwoodite for the first time to analyze its hydrogen positions.

Deuterated ringwoodite powder samples were synthesized at 1300 ^oC and 18 GPa for 5 minutes using a scaled-up Kawai-type multi anvil apparatus. The run products were evaluated by micro-focused X-ray, Raman spectroscopy and powder X-ray diffractometer to confirm their purity.

Neutron powder diffraction patterns were taken at BL-19 (TAKUMI) at Materials and Life Science Experimental Facility, J-PARC. The two representative deuterated ringwoodite samples were with identical composition mixed and measured together in a sample holder made of TiZr "null" alloy. The obtained diffraction pattern has been analyzed by Rietveld refinement using the "Z-Rietveld" code in order to determine positions and site occupancies of deuterium atoms in the ringwoodite structure.

We can propose two possible models for the deuterium atomic positions, 96g and 192i. These refined models were given with almost identical R factors. They also gave similar site occupancies and temperature factors for the elements except for the deuterium. However, in terms of deuterium temperature factor, the 192i model is more preferable than the 96g model. The refinement results also showed that the (Mg+Fe)/Si ratio is lower than the ideal ratio of dry ringwoodite [(Mg+Fe)/Si = 2.0] while Si in T sites are fully occupied, which demonstrates that deuterium only substitutes Mg and Fe in M sites and Si in T sites is not affected.

References:
[1] Kohlstedt, et al., Contrib. Mineral. Petrol., 123, 345, 1996.
[2] Kudoh, et al., Phys.Chem.Minerals., 27, 474, 2000.
[3] Smyth, et al., Am Mineral., 88, 1402, 2003.

キーワード: ringwoodite, neutron diffraction, Rietveld refinement
Keywords: ringwoodite, neutron diffraction, Rietveld refinement

SMP06-P02　　　　　会場:3 階ポスター会場　　　　　時間:4 月 28 日 18:15-19:30

Stability of Hydrous phase H MgSiO2(OH)2 in the lower mantle
Stability of Hydrous phase H MgSiO2(OH)2 in the lower mantle

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Subducting slabs transport water to Earth's deep interior and its circulation on a global scale is the key to understanding the evolution of the planet. However, it is still a matter of debate how deep water can be transported. Therefore, there are many studies on phase relationships in hydrous minerals or MORB-H2O systems. Most dense hydrous magnesium silicates (DHMS) are stable up to 50 GPa (e.g., Komabayashi et al., 2004). Recently, the synthesis of Mg- and Si- bearing δ-AlOOH, which is a solid solution between 2AlOOH-MgSiO2(OH)2, was reported and it might be transported with Mg-perovskite or Mg-post perovskite up to 135 GPa (Ohira et al., 2012, AGU). Tsuchiya (2013) theoretically reported Phase H, the end member of the system, was stable above 45 GPa and up to 55 GPa. And also it was experimentally synthesized at 50 GPa (Nishi et al., 2014). Although the previous studies claimed that Phase H was broken down above 55 GPa, it may be a host phase of water in the deep Earth interior. Here, we report the stability field of a new candidate phase of water reservoir at the lower mantle conditions by investigating the MgO-SiO2-H2O system up to 75 GPa and 2000 K.

A mixture of quartz and brucite (molar ratio 1 : 1) powders were used as starting materials. The high pressure and high temperature experiments were performed by using a double-sided Laser heated diamond anvil cell. A pellet with thickness of about 15 μ m was made by a cold compression technique. In situ XRD experiments in the MgO-SiO2-H2O system were performed at BL10XU, SPring-8. In this study we confirmed that hydrous phase H does exist in the MgO-SiO2-H2O system and its stability fields expands at least up to 75 GPa and above 2000 K in contrary with previous reports (Tsuchiya, 2013; Nishi et al., 2014).

If Phase H exists under high pressure conditions corresponding to the pressure of CMB, it may transport water to CMB and thus the core may contains hydrogen as a light element.

キーワード: hydrous phase, lower mantle, subduction
Keywords: hydrous phase, hydrous phase, subduction

Partitioning of carbon between metallic- and silicate-liquids in carbonaceous chondrite compositions at high pressure

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Major volatile elements in the terrestrial planets are oxygen, sulfur, carbon, hydrogen, and nitrogen. They are also candidates for light components in the earth's core which were incorporated into the core at terrestrial magmaocean stage. Partitioning behavior of carbon has not been determined well though it is one of the strong candidates for light elements in the earth's core. We investigated partitioning of carbon with sulfur and oxygen between metallic- and silicate liquids at 6 GPa and 2073 K in carbonaceous chondrite composition (Allende meteorite; CV3). Effect of nitrogen and water as accessory components were also examined. High pressure experiments were conducted with multi-anvil high pressure apparatus. Graphite was used as capsule material. Composition of coexisting metallic- and silicate liquids were measured by electron microprobe with wavelength dispersion type spectrometer except for carbon in silicate liquid. Carbon concentration of bulk recovered sample was measured by elemental analyzer. Then, carbon concentration in silicate liquid was obtained by subtraction of carbon amounts in metallic phase which obtained by electron microprobe and SEM image analyses. Present result suggests that in oxidized carbonaceous chondrite composition, partitioning coefficient of carbon [$D^{Metallicliquid/Silicateliquid} = C^{Metallicliquid}/C^{Silicateliquid}$; C is concentration of carbon in wt.%] is close to 1, and it may increase with increasing the Fe^{metal}/Fe^{oxide} ratio in the carbonaceous chondrite composition.

SMP06-P04　　　　　　　会場:3 階ポスター会場　　　　　　時間:4 月 28 日 18:15-19:30

Microanalysis of H2O and CO2 in silicate melt using laser Raman spectroscopy

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<Introduction>

Water and carbon dioxide are the major volatile components in the crust and mantle. Development of microanalytical techniques of these volatiles has made it possible to investigate mechanisms of igneous and volcanic processes. FTIR has been used as a fundamental tool for this purpose, but its spatial resolution is too large ($\sim$30 μm) to analyse small melt inclusions and micro-scale volatile distribution within a high-pressure experimental sample. In this study, we developed a new technique for volatile analysis in silicate glasses with $\sim$1 μm spatial resolution using laser Raman spectroscopy.

<Experimental and analytical procedure>

Standard glasses were synthesized by using a piston-cylinder apparatus in M. Nakamura's laboratory. Basaltic glass power was loaded into a platinum-sleeved nickel capsule together with oxalic acid and run at 1473 K and 0.5-1.2 GPa. After quench, the H_2O and CO_2 (dissolved as CO_3^{2-}) contents of the glasses were measured by using FTIR. The same glasses were then analysed with a Thermofisher DXR laser-Raman spectrometer. Wave length, power and diameter of the laser beam were 532 nm, 10 mw and 0.7 μm, respectively. We normalized the Raman spectra by the intensity of a peak at 500 cm^{-1} (T-O-T bond) and subtracted the spectrum of the volatile-free glass. The intensities of peaks at 3550 cm^{-1} (H_2O) and 1080 cm^{-1} (CO_3^{2-}) in the resulting spectra were compared with the H_2O and CO_2 contents determined by FTIR spectroscopy.

<Results>

The H_2O and CO_2 contents were determined to be 0.7-2.1 wt% and 0.05-0.82 wt%, respectively. Raman spectroscopy showed that the intensities of peaks at 3550 and 1080 cm^{-1} increased with increasing H_2O and CO_2 contents, respectively. We fitted a linear equation to the data and obtained H_2O (wt%) = (3.58$\pm$0.14)$\times$I$_{3550}$ and CO_2(wt%) = (4.61$\pm$0.21)$\times$I$_{1080}$.

<Application>

We applied this technique to volatile analysis of an experimentally-produced bubble-bearing basaltic glass. In the experiment, basaltic melt was first equilibrated with H_2O-CO_2 mixture fluid at 1473 K and 1 GPa, and then decompressed to 0.5 GPa and kept for 10 minutes. After quench, the H_2O and CO_2 contents around bubbles were measured along the radial direction at 2-μm intervals for a total of 50 μm. The CO_2 contents decreased on moving towards the bubble, indicating that CO_2 was diffusing into the bubble. Fitting the diffusion equation to this profile, we estimated the diffusivity of CO_2 to be 1.2×10^{-12} m^2/s. This value is consistent with that reported by Zhang et al. (2007). In contrast to CO_2, H_2O showed a flat profile, suggesting that H_2O was already equilibrated with the fluid in the bubble. This is because the diffusivity of H_2O is one order of magnitude greater than that of CO_2. Such a diffusive fractionation was observed also in rhyolitic melt (Yoshimura and Nakamura, 2010).

キーワード: CO2, H2O, glass, Raman
Keywords: CO2, H2O, glass, Raman

地球内部の炭素含有鉱物相の循環過程
Dynamic and cyclic process of carbon-bearing phases of the terrestrial interior

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本研究は次のようにまとめられる。

１）地球内部の炭素含有鉱物相は、物質状態の動的変化（気体、液体、固体）を活動的な地球惑星上で解明するために議論する。

２）本研究で用いた試料は、ダイヤモンド（コンゴ、アフリカ）、石灰岩（秋吉等、日本）、カーボナタイト（レンガイ-タンザニア-アフリカ、ヨーロッパ、北米）とシンガイト（シュンガ、ロシア）そして炭酸塩粒子を有するリビアガラス（アフリカ）などで、それらを FE 分析型 SEM などで炭素含有物質のマイクロ状ナノ結晶粒等を観察している。

３）本件の研究データでは、マイクロ炭素含有粒子が容易に三状態の物質相変化し、地表から内部における高圧衝撃波現象（地震、火山と衝突）で固化したガラス相等の存在を示す。

４）局地的な炭素を含む流体相を含む堆積が、不規則表面および動的な地球の内部に分布するのは、原始地球の地球外衝突の動的プロセスにより物質状態を変化し、さらに進化した地球の海底衝突で固化した混合物が地球内部の動的な混合形成したことによります。

５）創成期の衝突過程で形成された不均質な地表面と内部は、マクロ生命活動を安定的な動的な炭素を含む三物質状態（空気、液体や固体）を生成し、その後継続的地表や内部の変化過程で複雑で局地的な分布を示している。

キーワード: 炭素, 地球内部, 循環過程, 衝撃波現象, 不規則分布, 局部形成
Keywords: carbon, interior, cyclic process, shock wave event, irregular distribution, local deposit