（ ※本報告書は英語で記述してください。ただし、産業利用課題として採択されている方は日本語で記述していただいても結構です。）

CROSS TOKAI **Experimental Report** J-PARC	承認日 Date of Approval 2017/03/06 承認者 Approver Takanori Hattori 提出日 Date of Report 2017/03/06
課題番号 Project No. 2016B0017 実験課題名 Title of experiment Hydrogen volume in iron hydride at high pressure and temperature 実験責任者名 Name of principal investigator Eiji Ohtani 所属 Affiliation Graduate school of Science, Tohoku University	装置責任者 Takanori Hattori 装置名 Name of Instrument/（BL No.） ATSUHIME 6-axis press at Planet beamline (BL11) 実施日 Date of Experiment January 14-19

試料、実験方法、利用の結果得られた主なデータ、考察、結論等を、記述して下さい。（適宜、図表添付のこと）
Please report your samples, experimental method and results, discussion and conclusions. Please add figures and tables for better explanation.

1. 試料 Name of sample(s) and chemical formula, or compositions including physical form.

The starting material was powdered sample of iron (99.9% purity, Wako pure chemical industries, Ltd) with a grain size about 300 um. The powdered Fe was compressed by uniaxial hand press to the pressure of 100-200 MPa, and a Fe disc with 3 mm diameter and 2.5 mm length was prepared for the starting material.

2. 実験方法及び結果（実験がうまくいかなかった場合、その理由を記述してください。）

Experimental method and results. If you failed to conduct experiment as planned, please describe reasons.

Experimental method:

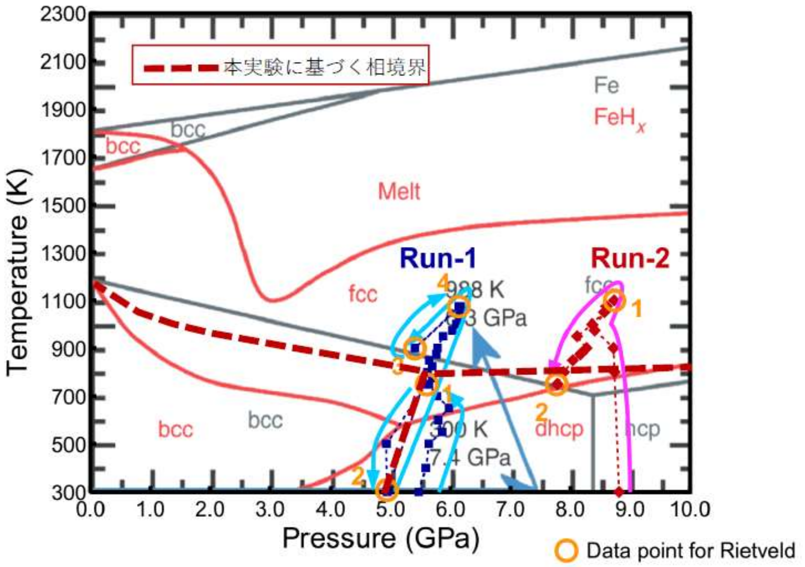
A cubic high-pressure apparatus (Atsuhime) installed at BL11 was used for high pressure generation. An Fe disc specimen (3mm in diameter and 2.5mm in height) is placed at the center of a hydrogen-sealing capsule made of NaCl (5.5mm in diameter and 8mm in height) with internal hydrogen sources of NH3BH3 pellets above and below. The NaCl capsule was inserted in a cylindrical graphite heater and embedded in a pressure-transmitting medium made of ZrO2 (17-mm-edge cube). High pressure high temperature diffraction experiments was conducted at around 5-9 GPa in the stability field of fcc-iron and 300-1200 K. Neutron diffraction data was corrected at high pressure and temperature. Neutron-diffraction profiles were accumulated during the heating process. The temporal evolution of the diffraction profile was monitored at several fixed temperatures above the hydrogenation temperature of NH3BH3. The composition and site occupancy of H atom in the fcc-and dhcp-lattice of FeHx were determined for the equilibrium state at a fixed pressure. The temperature was kept constant and the temporal evolution of the diffraction profile was monitored to confirm that the solid solution of

2. 実験方法及び結果（つづき） Experimental method and results (continued)

FeHx reached equilibrium with the surrounding H_2 fluid. The structure of FeHx and the content of hydrogen were determined by Rietveld refinement.

Results and discussion

We have successfully determined the hydrogen atomic volume in iron hydride lattice at various pressure and temperature for the first time. The phase transition and hydrogenation of iron were observed by dhcp-FeHx and fcc-FeHx phase, which are high pressure and high temperature phases of FeHx. We have conducted two separate runs. Run 1 was made in the pressure range around 6 GPa using the pressure cell for the cubic anvils with 10 mm truncated edge length. Run 2 was made at around 8 GPa using the anvils with 7 mm truncated edge length. The pressure-temperature paths of our experiments are shown in Figure 1. The phase boundaries of FeHx compounds determined here are significantly different from those reported previously as shown in this figure; stability field of dhcp-FeHx is wider as shown in this figure. We have successfully conducted Rietveld analyses for six runs, and the atomic fraction of hydrogen, x, was determined for fcc and dhcp-FeHx. We found that x for fcc-FeHx varies from 0.45 to 0.75 in the pressure and temperature range studied, whereas x for dhcp-FeHx is close to unity. We determined the hydrogen volume in iron hydride lattice based on our Rietveld analyses and previous equation of state of iron at high pressure and temperature. The atomic volume of hydrogen is greater than those estimated previously (e.g., Fukai et al., 2003) as was determined for the deuterium volume in FeDx (Machida et al., 2014). Our data provide important clues for evaluation of the compression behavior of hydrogen atom in metallic iron lattice and for more reliable estimation of the hydrogen content in the Earth's core by comparing with the density of the Earth's core derived from seismological studies.



We have studied the SEM-EBSD analysis of the recovered iron samples which was transformed to bcc-Fe after release of hydrogen during decompression and recovery. The recovered bcc-Fe shows characteristic micro–textures with large lattice strain and oriented micro-cracks which might be formed during release of hydrogen from the solid iron hydride.

References:

Fukai et al., Jour. Alloy Compound, 348, 105-109 (2003).
Machida et al., Nature communications, 5:5063 doi: 10.1038/ncoms6063 (2014)