実験報告書様式（一般利用課題・成果公開利用）

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CROSS TOKAI **Experimental Report** J-PARC	承認日 Date of Approval 2017/7/27 承認者 Approver Takanori Hattori 提出日 Date of Report 2017/7/26
課題番号 Project No. 2017A0093 実験課題名 Title of experiment: Site occupancy and volume of interstitial hydrogen atoms in iron hydride polymorphs 実験責任者名 Name of principal investigator Eiji Ohtani 所属 Affiliation Tohoku University	装置責任者 Name of Instrument scientist Takanori Hattori 装置名 Name of Instrument/（BL No.） Planet/BL11 実施日 Date of Experiment April 8th -20th, 2017

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 the same as that used for the previous proposal of 2016B, which was the 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 a uniaxial hand press to the pressure of 100-200 MPa, and an iron 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:

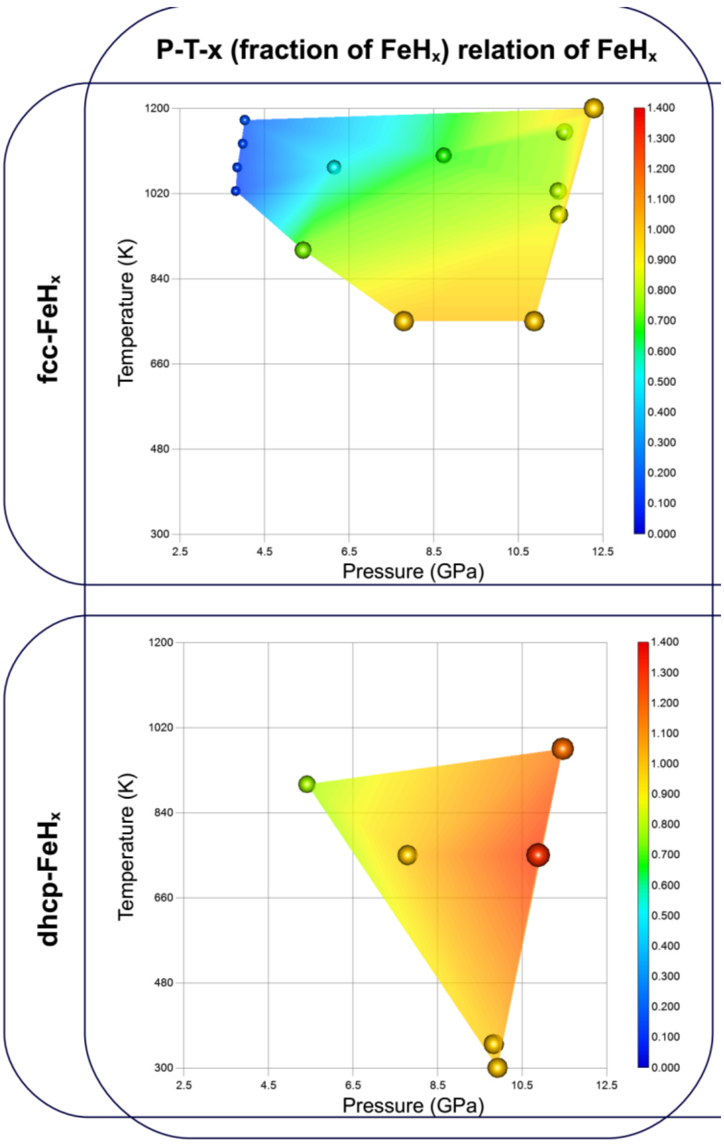
A cubic high-pressure apparatus (Atsuhime) installed at BL11 was used for high pressure generation. An Fe disc specimen (3mm in diameter and 2.5mm in height) is placed at the center of a hydrogen-sealing capsule made of NaCl (5.5mm in diameter and 8mm in height) with internal hydrogen sources of NH_3BH_3 pellets above and below. The NaCl capsule was inserted in a cylindrical graphite heater and embedded in a pressure-transmitting medium made of ZrO_2 (17-mm-edge cube). Two high pressure high temperature diffraction experiments was conducted at around 9.5-12 GPa in the stability field of dhcp- and fcc-iron and 300-1200 K using anvils with a 7 mm truncated edge length, and around 3.6-4 GPa and 850-1200 K using anvils with a 10 mm truncated edge length in the stability field of fcc-Fe. 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 NH_3BH_3. The composition and site occupancy of H atom in the fcc-and dhcp-lattice of FeH_x were determined for the

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

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 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. 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 9-12 GPa using the pressure cell for the cubic anvils with 7 mm truncated edge length. Run 2 was made at around 3.6-4 GPa using the anvils with 10 mm truncated edge length.



Diffraction profiles obtained at 10.9 GPa and 750 K revealed that the four typical peak intensities of dhcp-FeHx can be fitted by FeHx with x values greater than unity (x>1) model more reasonably. This process can be explained as follows: After complete filling (x=1) of the interstitial sites, hydrogen go into the lattice defect space. Hydrogen atoms kicked out iron and appears to fill the vacant iron sites, and thus hydrogen atoms becomes larger than unity. This mechanism is similar to hydrogen in TiHx compound. Figure 1 shows the change of hydrogen content x in FeHx (fcc and dhcp-FeHx), The amount of hydrogen, x, increases with increasing pressure and decreasing temperature. The amount of hydrogen, x, exceeds unity in dhcp-Fe at pressures greater than 10 GPa.