5–5 Elucidation of the Site-Occupancy of Hydrogen Atoms in Iron
— In situ Neutron Diffraction under High Pressure and High Temperature —

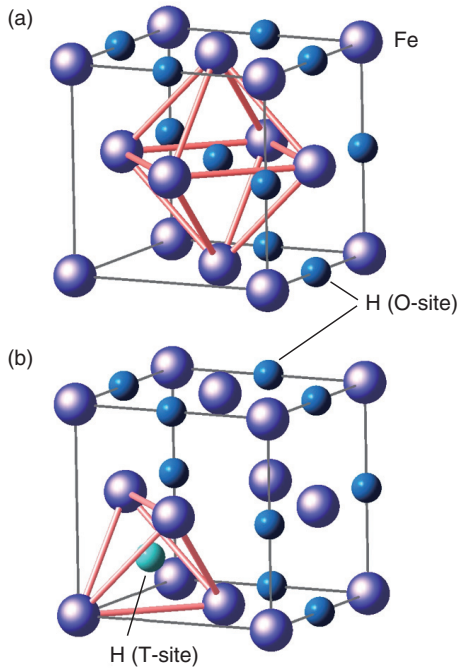

Fig.5-15 Structural models of FeD_x with an fcc metal lattice
The H atoms are accommodated in (a) the only O-site and (b) both the O- and T-sites. Frames colored in red show typical T- and O-sites.

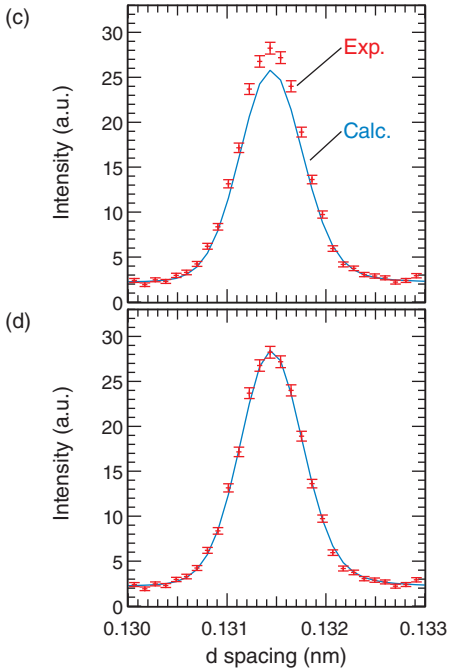

Fig.5-16 *In situ* neutron diffraction profile at 988 K, 6.3 GPa
A typical diffraction peak is shown. Red crosses are the experimental results; the cyan curve is the fitting result using the fcc structural model with the D atoms occupying (c) only the O-site and (d) both the O- and T-sites.

The hydrogen (H) occupation state in a metal lattice provides important information concerning some properties of metal hydrides, such as their stability. It is well-known that slightly dissolved H atoms induce changes in the mechanical properties of a metal. In addition, the crystal structure and physical properties are changed by high concentrations of H atoms in a metal lattice. The high H-concentration state in an iron (Fe) metal lattice is realized only at a high H_2 pressure of several GPa. Hence, it is difficult to experimentally determine the H-occupation state. In this study, we have investigated the H-occupation state of a face-centered cubic (fcc) iron deuteride (FeD_x) using *in situ* neutron diffraction measurement.

The high-pressure neutron diffractometer (PLANET) constructed at the Materials and Life Science Experimental Facility in J-PARC enables us to perform *in situ* neutron diffraction experiments at a pressure of several GPa and temperature of several hundred K. Based on the high-pressure reaction cell for the *in situ* synchrotron radiation X-ray diffraction experiment, we developed such a device for the *in situ* neutron diffraction experiment. The neutron diffraction profile of the deuteride is generally suitable for neutron structural analysis rather than that of the hydride; hence, we performed the *in situ* neutron diffraction experiment on the formation process of FeD_x instead of FeH_x.

At high pressure and high temperature, the Fe metal exhibited fcc structure, which has two interstitial sites available for accommodating H atoms: octahedral (O) and tetrahedral (T) sites (Fig.5-15). We succeeded in observing the formation of fcc-FeD_x. It is believed that D atoms occupy only the O-sites in the fcc-FeD_x; however, the refinement of the neutron-diffraction profile measured at 988 K and 6.3 GPa showed a misfit for some reflection peaks (Fig.5-16(c)). Hence, another structural model was applied for the analysis; the D atoms were accommodated in both the O- and T-sites. Using this model, the profile was well-reproduced and the refined occupancies of the D atoms in the O- and T-sites were 0.532(9) and 0.056(5), respectively (Fig.5-16(d)). We discovered that the D atoms slightly occupied the T-sites of the fcc-Fe. This result provides important information for understanding certain properties of Fe infused with dissolved H atoms.

This study was partly supported by Japan Society for the Promotion of Science (JSPS) KAKENHI Grant-in-Aid for Scientific Research (A) (No.24241032).

Reference

Machida, A. et al., Site Occupancy of Interstitial Deuterium Atoms in Face-Centered Cubic Iron, Nature Communications, vol.5, 2014, p.5063-1-5063-6.