Investigation of multi light elements in iron-silicate-water system using high pressure and temperature neutron experiments: Implications for the Earth's evolution

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Introduction:
The Earth’s core is considered to consist of Fe alloy and some light elements (O, S, Si, H, C). Hydrogen (H) is the most abundant element in the universe and one of the promising candidates existing in the Earth’s core. However, its amount dissolved in the core and its process are still unknown because H cannot be detected by X-ray diffraction (XRD) and it easily escapes from iron by the release of pressure. Recently, H content in *fcc*-Fe at high pressure and high temperature (high-PT) has been determined using in-situ neutron diffraction measurements at J-PARC [1], suggesting that H had preferentially dissolved into iron before any other light elements have dissolved in the very early stage of Earth’s evolution. It is important to further study the partitioning of the other light elements between iron hydride (not pure iron) and silicates. To clarify the effects of multi light elements on the core-mantle segregation in the Earth’s formation, we have focused on sulfur (S) and investigated its effect on hydrogenation of iron.

Experimental:
Iron fine powder was pelletized with/without S powder and placed at the center of the graphite sample capsule. Powder mixture of quartz (SiO_2), brucite $Mg(OD)_2$ (or MgO for comparison of the water existence) surrounds the Fe pellet. Sample composition of Fe:$Mg(OD)_2$:SiO_2=2:1:1 of molar ratio with 5wt.% of S simulates an ideal condition of the primitive Earth.
In-situ high-PT neutron experiments were carried out using a six-axis multi-anvil press “Atsuhime” installed at MLF, PLANET (BL11), J-PARC. An originally improved multi-anvil 6-6 type assembly was applied. Three samples were pressurized up to 6–7 GPa and then heated up to 700 ˚C step by step. The reaction processes (dehydration of $Mg(OD)_2$, phase transformations of Fe (*bcc*-*fcc*) and FeS, formation of silicates) were carefully checked in real time. The diffraction patterns of *fcc*-Fe were measured at 550 ˚C and 700 ˚C for long time and analyzed using the Rietveld refinement method to determine structural and atomic parameters of *fcc*-Fe and sub-products. The recovered samples were examined by XRD and SEM-EDS to more precisely identify the products, their compositions, and the elements partition.

Results and Discussion:
The cell volume changes in *fcc*-Fe and sub-phase of FeS-V were investigated for every hour during the long-time measurements at 700 ˚C. For the sample including water, the volume of *fcc*-Fe was significantly increased with time, while the sample without water showed no change regardless of temperatures. The cell volume of FeS-V did not increase, suggesting that the formation of FeS was promoted in the existence of water, but its hydrogenation was negligible. Fe gradually incorporated D in the existence of water and FeD_x formed (x~0.10 at a maximum). From the results of SEM, the inhibition of deuterization might be due to the formation of FeS layer surrounding Fe pellet, whereas such a thick layer was not observed when using the Fe+S pellet. The sample without water did not include FeS layer nor Fe-bearing silicates.
It is suggested that both S and Fe were not mobile and could not react with each other without the existence of water. H and S can be preferentially incorporated into solid Fe at lower-T (~700 ˚C) before

melting. The other light elements (C, O, Si) could have dissolved into molten iron hydride and/or FeS in the later process of Earth’s core-mantle differentiation at much higher-PT condition.

Reference:
[1] R. Iizuka-Oku, T. Yagi, H. Gotou, T. Okuchi, T. Hattori, A. Sano-Furukawa, Nature Commun. 8, 14096 (2017).

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