Sound velocity measurements in dhcp-FeH by inelastic X-ray scattering method: Implications for the hydrogen concentration in the Earth's core

The distributions of density and sound velocities of the Earth's interior are well investigated through geophysical observations. According to proposed models (e.g., Preliminary Reference Earth Model (PREM) [1]), the inner core, which is solid, is about 2–5% less dense than pure iron. Therefore, it is accepted that the core consists of iron and light elements. Hydrogen is one of the most plausible light elements in the core.

A large number of high-pressure experiments on iron alloys, including iron hydride (FeH), have been carried out in order to discuss the core composition. However, most of the previous works have constrained the abundances of light elements in the core by matching *only* density *or* sound velocity of the iron alloy to the PREM data. The important point is that iron alloy, the density ρ *and* sound velocity V of which are the same as ρ and V of the Earth's core *at* the pressure and temperature conditions of the core, can be a candidate for the constituent of the core. Therefore, we determined the density evolution of the sound velocity of FeH by inelastic X-ray scattering (IXS) and X-ray diffraction (XRD) analyses, and thereby constrained the abundance of hydrogen in the Earth's core by matching *both* density *and* velocity to the PREM data [2].

The IXS and XRD experiments were conducted up to 70 GPa at room temperature. High-pressure conditions were generated using a symmetric diamond anvil cell. The iron sample was compacted from a powder into a foil and hydrogen initially pressurized to 0.18 GPa was loaded to the sample chamber. Double-hexagonal-close-packed (dhcp) FeH was synthesized at around 4 GPa.

The IXS experiments were performed at **BL35XU**. We used the Si (9 9 9) configuration, which provides an incident photon energy of 17.794 keV with an energy resolution of 2.8 meV full width at half-maximum. The beam size was focused to 15 μm × 17 μm using a Kirkpatrick-Baez (KB) mirror pair. The scattered X-rays were analyzed with 12 crystals arranged in a 2-dimensional array (3×4 array). The XRD experiments were carried out at **BL10XU**. The X-ray beam was collimated to 15 μm in diameter and a typical wavelength used was 0.41348(7) Å. An XRD pattern of the sample was collected after each IXS measurement in order to obtain the density data of the sample at each pressure.

A typical IXS spectrum collected at 70 GPa and room temperature is shown in Fig. 1. The

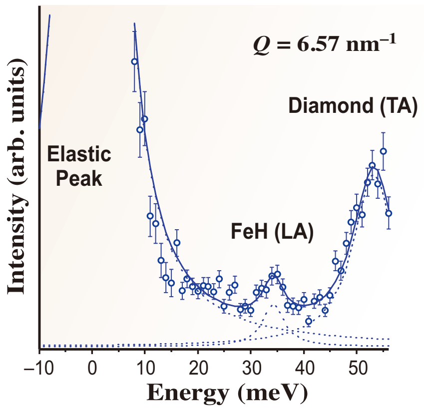

Fig. 1. Typical IXS spectrum of dhcp-FeH at 70 GPa and room temperature (Q = 6.57 nm^{-1}). The peak at zero energy is attributed to elastic scattering. Solid line is fitted result for elastic, longitudinal acoustic (LA) phonons of dhcp-FeH, and transverse acoustic (TA) phonons of diamond, contributions and dotted lines are the corresponding individual components.

compressional sound velocity (V_P) was determined by fitting the phonon dispersion with a sine function:

$$E\,[\mathrm{meV}] = 4.19\times10^{-4}\, V_P\,[\mathrm{m/s}] \times Q_{max}\,[\mathrm{nm}^{-1}]\,\sin\left(\frac{\pi}{2}\,\frac{Q\,[\mathrm{nm}^{-1}]}{Q_{max}\,[\mathrm{nm}^{-1}]}\right) \quad (1)$$

where E and Q are the energy and momentum of the phonon, respectively, and Q_{max} is approximately the first Brillouin zone edge. Figure 2 shows the dispersion curves up to 70 GPa. The determined velocity is an aggregate sound velocity averaged over crystal orientations because our sample was polycrystalline.

The measured V_P for dhcp-FeH is presented as a function of density in Fig. 3, and compared with PREM in the inner core [1]. The V_P for dhcp-FeH above 8.0 g/cm^3 (about 30 GPa) follows Birch's law (a linear relation between velocity and density). In contrast, the velocity is not linear when density is below 8.0 g/cm^3. Since it has been reported that dhcp-FeH loses its magnetism between 22 and 42 GPa (a ferromagnetic to nonmagnetic transition), the difference below and above 8.0 g/cm^3 is due to the loss of magnetism. This indicates that magnetism significantly affects the sound velocity.

We discussed the hydrogen concentration in the Earth's inner core using a linear mixing model under the assumption that the average density ρ and sound velocity V of a two-component ideal solid are given by

$$\rho = t\rho_{\mathrm{FeH}} + (1-t)\rho_{\mathrm{Fe}} \quad (2)$$

and

$$V = \frac{V_{\text{FeH}}\,V_{\text{Fe}}}{(1-t)\,V_{\text{FeH}} + t V_{\text{Fe}}} \qquad (3)$$

where t is the volume fraction of FeH. We assigned $\rho = \rho_{\text{PREM}}$ and $V = V_{\text{PREM}}$, where ρ_{PREM} and V_{PREM} are the density and compressional sound velocity of the PREM inner core [1], respectively. The density of iron ρ_{Fe} was estimated from the equation of state [3], and the sound velocity of iron V_{Fe} was calculated from the shock wave data [4]. In addition, the shear sound velocity V_S was derived by combining V_P with ρ and the bulk modulus K derived from the equations of state of dhcp-FeH [5] or hcp-Fe [4] according to the relation:

$$V_S^{\,2} = \frac{3}{4}\left(V_P^{\,2} - \frac{K}{\rho}\right) \qquad (4)$$

From the set of equations (2) and (3), we obtain x = 0.13(3) in FeH_x (0.23(6) wt% H), and ρ_{FeH} = 9.8(7) g/cm^3 at the inner core boundary (ICB) and 10.5(7) g/cm^3 at the center of the Earth (COE). According to the equation of state of dhcp-FeH [e.g., 5], the values of ρ_{FeH} = 9.8(7) and 10.5(7) g/cm^3 are reasonable for the densities of dhcp-FeH at ICB (328.9 GPa and 5000 K) and COE (363.8 GPa and 6000 K) conditions within the range of uncertainty, respectively.

On the other hand, the estimated V_S for $FeH_{0.13(3)}$ (lower gray diamonds in Fig. 3) is much higher than the PREM V_S data. Recent *ab initio* calculations of hcp-Fe showed that V_S significantly decreases with temperature owing to anharmonic effects, i.e., V_S is reduced by about 29% from 0 K to 5000 K at the ICB density. Considering the anharmonic temperature effect, V_S for $FeH_{0.13(3)}$ at 5000 K and ICB pressure (white diamond in Fig. 3) is in good agreement with the PREM data at ICB condition.

Since the density *and* both V_P and V_S of $FeH_{0.13(3)}$ (0.23(6) wt% H) *at* the ICB pressure and temperature are consistent with the PREM data at ICB, $FeH_{0.13(3)}$ determined in this work is suitable as the Earth's inner core composition, assuming that the light element in the core is only hydrogen.

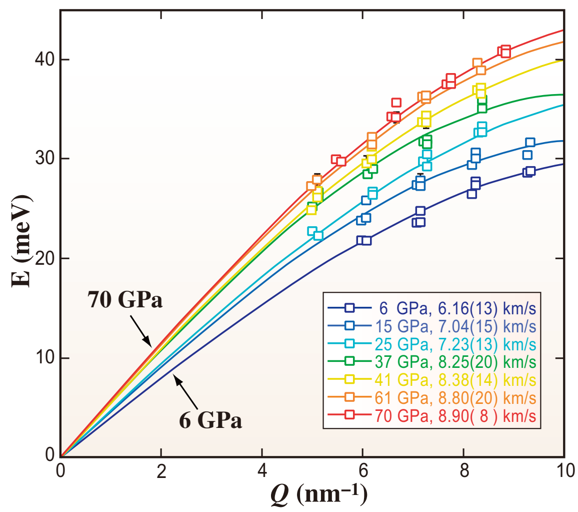

Fig. 2. Dispersion curves of LA phonons of dhcp-FeH at room temperature and pressures of 6, 15, 25, 37, 41, 61, and 70 GPa from bottom to top, together with each V_P value. Solid lines are the dispersion curves obtained by fitting the experimental data with equation (1) in the text. Vertical error bars (about 1% uncertainty) fall within the size of symbols if they are not apparent.

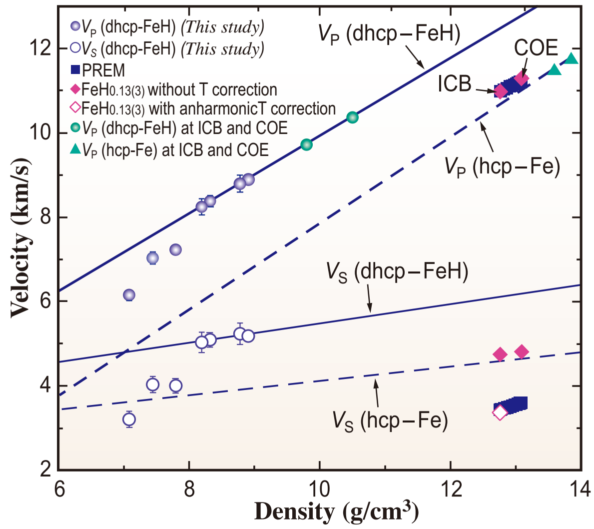

Fig. 3. V_P and V_S as a function of density, compared with the seismic observations. Solid and open circles are V_P and V_S for dhcp-FeH in this study, respectively. The solid squares are the PREM data in the inner core [1]. The solid lines indicate Birch's laws for nonmagnetic dhcp-FeH in this study and the dashed lines indicate those for hcp-Fe calculated from the shock data [4]. The solid triangles are V_P for hcp-Fe at ICB and COE conditions [3,4]. The green solid circles and pink solid diamonds denote our estimated densities and sound velocities for dhcp-FeH and $FeH_{0.13(3)}$ at ICB and COE pressures, respectively. The open diamond denotes our estimated V_S for $FeH_{0.13(3)}$ at 5000 K and the ICB pressure. Uncertainties in our estimations (sound velocities of $FeH_{0.13(3)}$) are about 2% for V_P and about 4% for V_S, which are not plotted for clarity.

Yuki Shibazaki[a,*], Eiji Ohtani[b] and Hiroshi Fukui[c,d]

[a] Geophysical Lab., Carnegie Institution of Washington, USA
[b] Department of Earth and Planetary Material Sciences, Tohoku University
[c] Graduate School of Material Science, University of Hyogo
[d] SPring-8/RIKEN

*Email: yshibazaki@ciw.edu

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