Speed of sound in liquid Fe-C alloy under high pressures using inelastic X-ray scattering

The structure of the Earth is divided into three layers, the uppermost hydrosphere, the silicate crust and mantle, and a metallic core (Fig. 1). This layered structure was primarily the result of geological processes occurring just after the Earth was formed. Today, structural information in Earth's interior is provided by seismological measurements that accurately give the sound velocities of pressure (P-) and shear (S-) waves and density (Fig. 1). In the past decades, the understanding of the mantle has significantly progressed, thanks to developments in laboratory measurements and theoretical calculations for possible silicate and oxide minerals. However, the composition of the metallic core is still controversial. This is because there are no available rock samples originally from the core, and it remains very challenging to perform laboratory measurements of the physical properties of possible core materials under the relevant extremely high-pressure and high-temperature conditions.

Geochemical and geophysical arguments show that the core consists mainly of iron. However shock compression experiments showed that the sound velocity and density of pure liquid iron are ~3% faster and ~10% lower than those of the liquid core (e.g., [1]), which indicates that some lighter elements are present. The nature of the lighter elements is fundamental to understanding a number of geological questions such as the core dynamo, the core temperature, and indeed the core formation process. Carbon is one of the candidate lighter components in the outer core. In order to constrain the amount of carbon in the outer core, we determined the sound wave velocity of liquid Fe-C alloy up to 70 GPa using inelastic X-ray scattering (IXS) at beamline **BL35XU** [2].

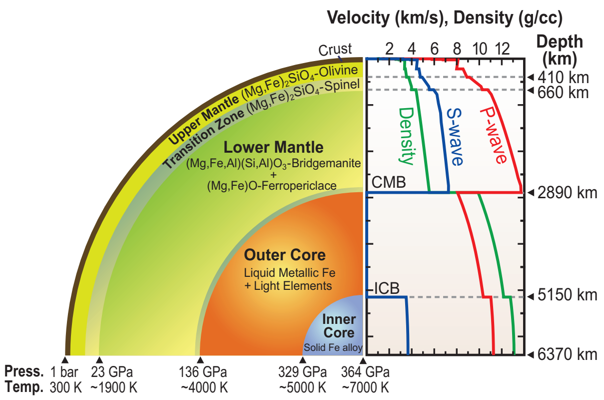

Fig. 1. Schematic of internal structure and seismological parameters of the Earth.

Samples of $Fe_{84}C_{16}$ alloy (4 wt% carbon) were melted at high pressure and temperature in an external-resistance-heated (EH) or a laser-heated (LH) diamond-anvil cell (DAC). The sample was loaded into the DAC with single-crystal sapphire discs that served as thermal and chemical insulators. Pressure was determined from the Raman shift of diamond or using the equation of state for Fe_3C observed before melting. Temperatures were monitored using a type-R thermocouple in the EH-DAC or by a spectroradiometric method in LH-DAC experiments, where uncertainties were less than 20 K or ±10%, respectively. The melting experiments were performed at 7.6–70 GPa and 1500–2800 K. The DACs were placed into vacuum chambers in order to minimize background scattering by air. The molten state of the specimen was confirmed by X-ray diffraction by the absence of diffraction peaks and/or the presence of diffuse signals from the sample. IXS with ~2.8 meV energy resolution, using Si (999) backscattering geometry at 17.79 keV, was used to determine the longitudinal acoustic, P-wave sound velocity. The scattered photons were collected by an array of twelve spherical Si analyzers leading to twelve independent spectra at momentum transfers (Q) between 3.2–6.6 nm^{-1} with a resolution $\Delta Q \sim 0.45$ nm^{-1} (full width). The energy transfer range of ±30 (or –10 to +30) meV was scanned for 1 to 3 h.

The IXS spectra (Fig. 2(a)) included three peaks: the Stokes and anti-Stokes components of the longitudinal acoustic (LA) phonon mode from the Fe-C sample, and a quasi-elastic contribution near zero energy. The excitation energies (E) for the LA phonon mode of the liquid sample obtained in a pressure range of 7.6–70 GPa are plotted as a function of momentum transfer (Q) in Fig. 2(b). The P-wave velocity (V_P) corresponds to the long-wavelength LA velocity at $Q \to 0$ limits. We made a linear fit to the data obtained at low Q below 3.5 nm^{-1} to determine the P-wave velocity, because positive dispersion may appear at higher $Q >> 3$ nm^{-1}. The resulting sound velocity of liquid $Fe_{84}C_{16}$ is plotted in Fig. 3(a).

We constructed an equation of state (EoS) for liquid $Fe_{84}C_{16}$ to extrapolate the present V_P data and to calculate its density ρ in the core pressure range. The adiabatic Murnaghan EoS can be described as a function of pressure P:

$$\rho = \rho_0 \left(1 + \frac{K'_S}{K_{S0}} P\right)^{\frac{1}{K'_S}} \qquad (1)$$

where ρ_0 is the density, K_{S0} is the adiabatic bulk modulus and K'_S is its pressure derivative at zero pressure. With the relationship $V_P = (K_S/\rho)^{0.5}$, the above EoS can become

$$V_P = \sqrt{\frac{K_{S0}}{\rho_0}}\left(1 + \frac{K'_S}{K_{S0}}P\right)^{\frac{1}{2}-\frac{1}{2K'_S}} \qquad (2)$$

The zero pressure density was obtained from literature data [3]. We fitted Eq. (2) to the present P-V_P data set, and then extrapolated to core pressures (Fig. 3). In this fitting, the temperature effect on V_P was ignored as in previous studies (e.g., [1]). Using the fitted parameters of K_{S0} and K'_S, the density profiles (Fig. 3(b)) for liquid $Fe_{84}C_{16}$ were also calculated along two adiabatic temperatures (Fig. 3(c)). The temperature profiles were calculated with a Grüneisen parameter for liquid pure Fe [1] to give possible core temperatures.

The P-wave velocity of liquid $Fe_{84}C_{16}$ is much faster than that of pure Fe at core pressures (Fig. 3(a)). Therefore, under the assumption of a linear relation between V_P and the carbon content of liquid Fe, only 5.2–4.0 atom% carbon in liquid Fe is required to match the seismological velocity [4] of the liquid outer core. However, much more carbon of 15.4–12.0 atom% is required to account for the core density (Fig. 3(b)). Therefore, carbon alone cannot explain the seismological observations and its maximum content will be less than 5.2 atom%, which corresponds to 1.2 wt%.

This result is also consistent with other geochemical considerations. The $^{13}C/^{12}C$ isotopic ratio of the present silicate mantle suggests that ~1 wt% carbon could have been incorporated into the core [5]. In addition, the mantle abundances of molybdenum and tungsten could be explained simply by the partitioning between the silicate magma ocean and the core-forming metal during the core formation process if the core-forming metal contained ~0.6 wt% carbon [5]. The present constraint on the carbon content of the liquid outer core is in agreement with those geochemical arguments.

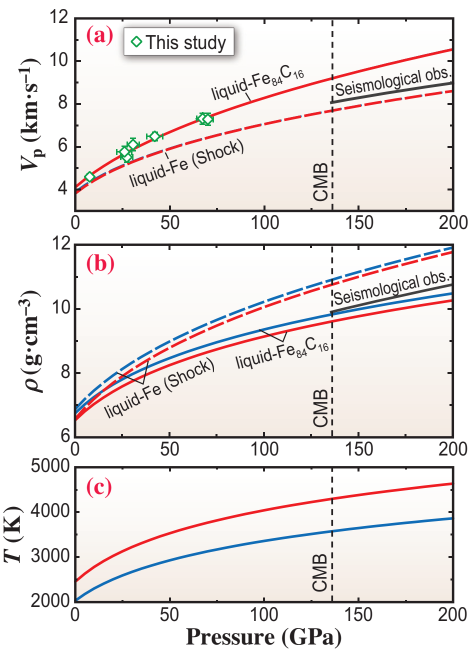

Fig. 3. Velocity and density of liquid $Fe_{84}C_{16}$. The P-wave velocity **(a)** and density **(b)** profiles of liquid $Fe_{84}C_{16}$ are calculated along two adiabatic temperature curves **(c)** of 3600 K (blue) and 4300 K (red) at the CMB, and are compared with that of liquid pure Fe (broken lines) [1] and seismological observations [4] for the liquid outer core. The open symbols are the present experimental data.

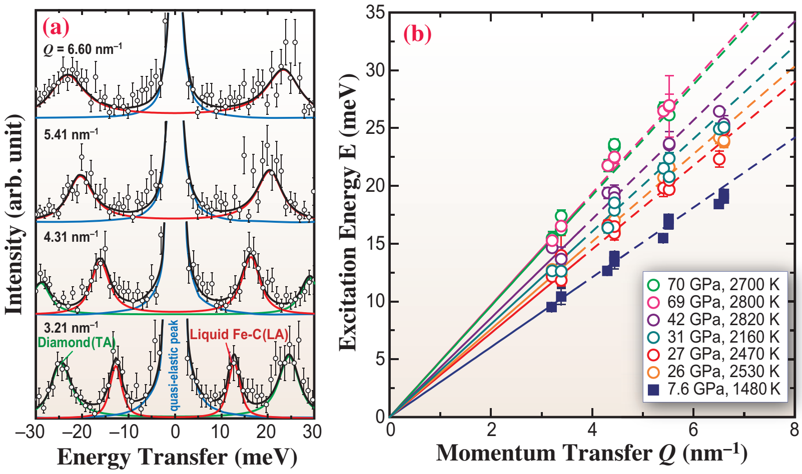

Fig. 2. Typical inelastic X-ray scattering spectra and phonon dispersion of liquid $Fe_{84}C_{16}$. **(a)** IXS spectra were collected at 26 GPa and 2530 K. Three components can be seen; quasi-elastic peak, longitudinal acoustic (LA) phonon mode of liquid sample, and transverse acoustic (TA) phonon mode of diamond from a diamond anvil cell. **(b)** Longitudinal acoustic phonon dispersions were obtained up to 70 GPa. Open and solid symbols exhibit laser-heated and external-resistance-heated DAC experiments, respectively.

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