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Towards a consensus on the pressure and composition dependence of sound velocity in the liquid Fe–S system

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A B S T R A C T

Recent advances in techniques for high-pressure and high-temperature experiments enable us to measure the velocity of sound in liquid Fe alloys. However, reported velocities in liquid Fe–S differ among research groups (e.g., by >10% at 5 GPa), even when similar methods are used (i.e., the ultrasonic pulse–echo overlap method combined with a large volume press). To identify the causes of the discrepancies, we reanalyzed previous data and conducted additional sound velocity measurements for liquid Fe–S at 2–7 GPa, and evaluated the potential error sources. We found that the discrepancy cannot be explained by errors in the sound velocity measurements themselves, but by inaccuracies in determining the temperature, pressure, and chemical composition in each experiment. Of particular note are the significant errors introduced when determining pressures from the unit-cell volume of MgO, which is a temperature-sensitive pressure standard, using inaccurate temperatures. To solve the problem, we additionally used h-BN as a pressure standard, which is less sensitive to temperature. The pressure dependence of the sound velocity became smaller than that of the original data because of the revised pressure values. Our best estimate for the seismic velocity of the Moon's liquid outer core is 4.0 ± 0.1 km/s, given a chemical composition $Fe_{83}S_{17}$.

1. Introduction

Quantitative experimental data that can be compared with observations are important for understanding the structure and evolution of the Earth and other planets. Seismological observations are a strong tool for elucidating the internal structure of planetary bodies, although such observations have been made for only the Earth and Moon. Density and sound velocity measurements of potential constituent materials of planets are essential to constrain the composition and structure of planetary interiors (e.g., Li and Liebermann, 2014). High-pressure density and sound velocity data for liquid Fe alloys have recently been reported by several groups, although the data vary among research groups. For example, the sound velocity in liquid Fe–S recently reported by Jing et al. (2014) differs significantly (>10% at 5 GPa; Fig. 1a) from that of Nishida et al. (2013), even though both cases used a similar method. Jing et al. (2014) noted that the reason for the discrepancy is unknown, but questioned the validity of the sound velocity reported by Nishida et al. (2013), because the density value calculated from the linear relationship between sound velocity and pressure found in Nishida et al. (2013) converged to a limit as pressure increased to infinity (red solid curve in Fig. 1b and c). However, this was due to Jing et al. (2014) inadequately extrapolating the linear relationship in the study of Nishida et al. (2013). There is no significant difference in the compression curves from the sound velocities reported in both cases in their considered experimental pressure ranges (Fig. 1b). It is, therefore, inappropriate to judge the validity of the sound velocity from the compression curves.

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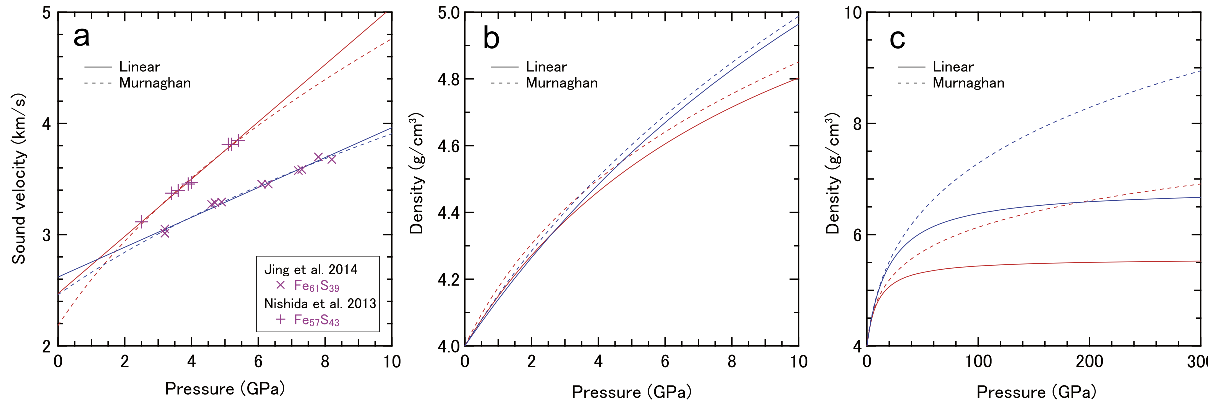

Fig. 1. Comparisons between Nishida et al. (2013) and Jing et al. (2014). (a) Sound velocity up to 10 GPa, (b) calculated density up to 10 GPa, and (c) calculated density extrapolated to 300 GPa. The density was calculated from sound velocity by using a linear relation (solid curve) or the Murnaghan equation of state (dashed curve) assuming $\rho_0 = 4.0$ g/cm^3, as used in Jing et al. (2014).

In this study, we examine the potential error sources affecting high-pressure high-temperature sound velocity measurement in liquid Fe–S. Then, the data of Nishida et al. (2013) are reanalyzed, and additional experiments are performed. We consider the potential causes of the discrepancy between the data of Nishida et al. (2013) and Jing et al. (2014), and propose an improved method. We report on the sound velocity in the liquid Fe–FeS system at pressures up to 7 GPa and consider the implications for the Moon's outer core.

2. Experimental methods

High-pressure experiments were conducted at the BL04B1 beamline at the SPring-8 synchrotron facility, Japan and the AR-NE7A beamline at the KEK PF-AR synchrotron facility, Japan. Pressure was generated using a Kawai-type high-pressure apparatus. We used three different types of cell assembly (Types A to C) designed for a truncated-edge length (TEL) of 8 mm (Fig. 2). The Type A cell assembly was the same as that used by Nishida et al. (2013). The Type B cell assembly was a modified version of the Type A cell assembly. The BN sleeve was changed to a flat-bottomed cylindrical container to improve the parallelism between the front and back surfaces of the sample and reduce the risk of leakage of the molten sample. The different thermocouple configuration was adopted to reduce the sample deformation and to facilitate the construction of the cell. In the Type C cell assembly, we removed the thermocouple from the Type B cell assembly (see Section 3.2 for more details) and placed an inner sleeve into the sample container to minimize leakage of the molten sample. The experiments at SPring-8 were carried out using Type A and B cell assemblies. All the experiments at KEK PF-AR were conducted using the Type C cell assembly.

The starting materials were pellets consisting of a mixture of Fe and FeS powders. We measured sound velocity using the ultrasonic pulse-echo overlap method, following Nishida et al. (2013). X-ray radiography was employed to measure the sample length. A three-cycle sine wave burst with a frequency of 37 or 42 MHz was used as an input electrical signal to measure the travel time. Sample melting was judged from X-ray diffraction patterns of the sample during experiments, and was confirmed afterwards from textural observations of the run products using optical and electron microscopes. We determined the pressure and temperature simultaneously without a thermocouple from the unit-cell volumes of h-BN and MgO by employing their equations of state (EOSs).

We increased the temperature along a single heating path for each experiment, considering the high reactivity and mobility of liquids. In the case of earlier experiments using Type A and B cell assembly, we gave priority to investigation of temperature dependence. In the case of additional experiments using the Type C cell assembly, we measured the sound velocity at 100–200 K above the melting temperature of the Fe–S sample, and then promptly quenched the sample with the highest priority to preventing chemical contamination. In addition, we performed a multiple heating cycles at different ram loads in one run, as done by Jing

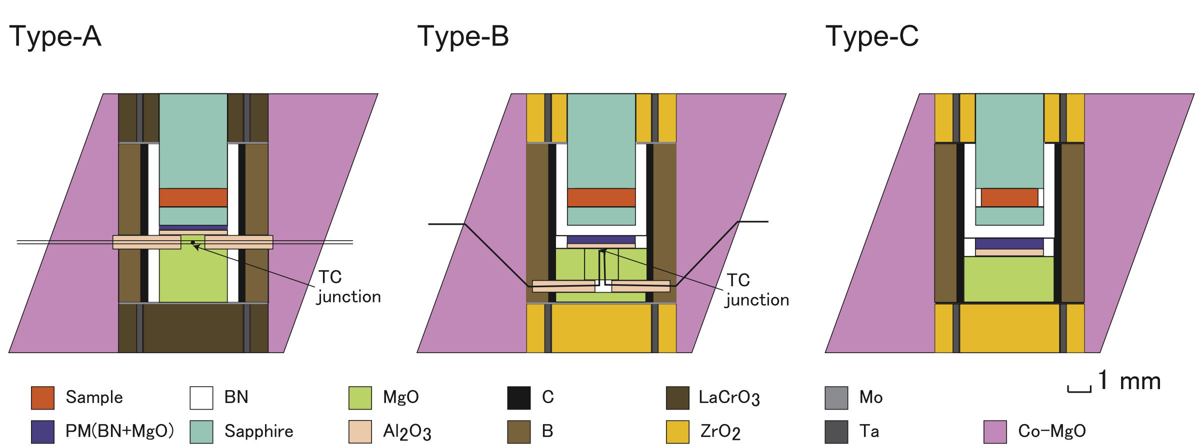

Fig. 2. Schematic illustrations (cross-sections) of the cell assemblies.