

Stratification in planetary cores by liquid immiscibility in Fe–S–H

The InSight (Interior Exploration using Seismic Investigations, Geodesy and Heat Transport) mission conducted by NASA recently detected seismic waves from the interior of Mars and found that Mars has a large and light liquid core [1]. A large and light core implies that there are substantial amounts of light elements in the core. Sulfur has been a widely accepted candidate light element in the Martian core on the basis of studies on Martian meteorites. However, the low density of the core requires the presence of other light element(s) such as oxygen, carbon, and hydrogen in addition to sulfur. In particular, hydrogen could be another major light element in the core of Mars because of its abundance in planetary building blocks and strong siderophile behavior at high pressures. Since light elements change the chemical and physical properties of the metallic core, their amounts affect core dynamics such as core convection and dynamo action, which generate a planetary magnetic field. While Earth currently has a magnetic field, the Martian magnetic field was lost ~4 billion years ago, which led to the loss of the ocean on Mars. Despite its importance, the property of Fe-S-H liquids has rarely been studied because of experimental difficulties in treating Fe-H alloys. Since hydrogen escapes from an iron lattice at ambient pressure and room temperature, the quantification of the hydrogen content in iron alloys requires X-ray diffraction (XRD) measurements at high pressures.

In the present study [2], we conducted highpressure melting experiments using a diamondanvil cell (DAC) technique and XRD measurements at SPring-8 BL10XU. We loaded a foil of Fe-S alloy and paraffin (C_nH_{2n+2}) as a hydrogen source into a cell assembly. After compression to high pressures, the sample was heated to higher than its melting temperature using a laser-heating system installed at BL10XU (Fig. 1). We collected XRD patterns before, during, and after heating at the center of a heating spot (Fig. 2). The disappearance of XRD peaks of iron alloys during heating was used to confirm melting. The XRD profiles collected after heating indicate volumes of iron alloy crystals that crystallized upon quenching the temperature. Such crystals preserve the composition of the liquid during heating. Because hydrogen expand the lattice volume of iron alloys, we determined the hydrogen content in the liquid from the volumes of hydrogen-bearing Fe and FeS at high pressures. Combining the XRD profiles at high pressures and textural/chemical observations

on sample cross sections at ambient pressure, we determined the composition of Fe–S–H liquid at high pressures and temperatures.

Cross sections of recovered samples showed a homogeneous single liquid or two separate Fe–S and Fe–H liquids. Pressure (*P*)–temperature (*T*) conditions in the experiment indicate that Fe–S–H liquids show immiscibility (separation into two liquids) at relatively low temperatures (e.g., <3000 K at 40 GPa) (Fig. 3(a)). We also conducted silicon- or oxygen-bearing experiments. Silicon and oxygen were preferentially incorporated into S-rich liquids, while carbon from paraffin was included predominantly in H-rich liquids. We observed immiscible S-rich and H-rich liquids to ~120 GPa, while immiscibility gaps in other iron-light element liquids close below 10–30 GPa.

The present-day P-T conditions of the Martian core (~20-40 GPa and ~2000-2500 K) are fully within the ranges required for liquid immiscibility to occur in the



Fig. 1. Microscopy images of a sample chamber (a) before, (b) during, and (c) after heating. Change in the shape of Fe–S alloy between (a) and (c) indicates the melting of the heated portion.

Fe–S–H system (Fig. 3(b)). Also, the compositions of liquids in the present experiments, including hydrogen contents revealed by XRD measurements, are similar to that in one of the Martian core composition models [3]. Therefore, the Martian core may have separated into two liquids as it cooled. The light liquid released by the separation of the liquid core could have driven the core convection and generated the planetary magnetic field. At the same time, separated light and heavy liquids could have formed gravitationally stable layers that halted the core convection. In this way, the Fe–S–H liquid immiscibility may have been responsible for both the onset and termination of the Martian core convection and planetary magnetic field.

On the other hand, P-T conditions of the Earth's outer core overlap the conditions of immiscibility only at its top (Fig. 3(b)). This implies that the Earth's uppermost core is stratified. Indeed, seismology indicates a low-sound-velocity layer at the top of the outer core [4]. While the layer is required to be light to remain at the top of the core, alloying more light elements causes a higher sound velocity. However, since the effect of H on the sound velocity of iron alloy is less than those of other light elements, the low-velocity layer could be formed by the enrichment of H and the depletion of S, Si, and O. Such a compositional anomaly can be formed by immiscibility in Fe–S–H liquid because Si and O are preferentially incorporated into S-rich liquids.



Fig. 2. X-ray diffraction patterns collected before, during, and after heating. Peaks of iron alloys and pressure medium (Al₂O₃) are labeled.



Fig. 3. Pressure-temperature conditions for immiscibility in liquid Fe-S-H. (a) Experimental results showing a homogeneous single liquid (filled) and two immiscible liquids (open) in Fe-S-H (rectangles) and Fe-S-H-C/ Si/O (triangles). Elements other than Fe, S, and H are indicated. The miscible/immiscible boundary (black line) and its uncertainty band (dashed lines) are based on the P-T conditions of the miscible/immiscible border in a sample (diamonds) and those of miscible/immiscible liquids obtained near the boundary. Yellow and blue lines are melting curves in the Fe–Fe₃S and Fe–FeH_x (x > 1) systems, respectively. (b) The blue band indicates the isentropic temperature profile of the Earth's core with the core-mantle boundary temperature $T_{\text{CMB}} = 3600 \text{ K}$. Light-blue lines are the pressure-temperature paths for the segregation of core-forming metals from silicate. Temperature profiles of the initial (pink) and present-day Martian cores (orange and red) are also given.

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References

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