Synchrotron Radiography and Microtomography on Fe-Rich Melt Segregation from Silicates at High Pressure and High Temperature

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Introduction

Formation of the core is perhaps the single most important event in the Earth's history since the birth of this planet. Various models of core formation fall between the following two end-member cases in terms of scale of the iron-silicate separation process. The "microscale" models involve predominantly small iron droplets that either percolate through a solid silicate matrix, or migrate through silicate liquid.[1] The "macroscale" models, on the other hand, all assume pre-existence of large liquid-iron blobs (iron "ponds") lying on top of solid silicates. Diapirs from iron ponds migrate downward by Rayleigh-Taylor instability[1,2] or through flotation of silicate grains from beneath a layer of molten iron to merge with the mantle above.[3] As pointed out by Stevenson,[1] the main problem with the macroscale models still lies in the microscale issue of whether liquid iron can percolate through a solid matrix.

The Earth's core consists of a liquid outer core and a solid inner core.[4] Both are believed to be made predominantly of iron (Fe) from geophysical and cosmochemical evidence.[5,6] However, there is a mismatch between the density of the core and that of pure iron at corresponding pressure (*P*) and temperature (*T*). The liquid outer core is about 10% less dense than the pure iron, while the solid inner core may be as dense as pure iron. This mismatch is ascribed to the presence of light elements.[5] A recent result of high *P* and high *T* experiment supports the notion[7] that light element(s) must be present in the inner core, as well as outer core, based on newly estimated thermal expansivity of pure iron.[8]

The nature of light elements in the cores is strongly linked to the core formation process. This research focuses on the effects of light elements on the dynamics and mechanisms of core formation and tries to address the "microscale" issue experimentally. To examine the mechanism(s) by which light elements were incorporated in the core during earlier core-formation process, we combined diffraction techniques with imaging capability.

Methods and Materials

High *P* and high *T* *in situ* x-ray experiments were performed using the 250 ton press installed at the GSECARS 13-BM-D beamline at the APS, with a cubic-anvil "DIA" high-pressure apparatus. *In situ* x-ray diffraction measurements were carried out based on the energy dispersive method with an energy range of 20-100 keV. Figure 1 shows a schematic of the x-ray diffraction and imaging setup. For x-ray diffraction, the incident x-ray beam is collimated by the front slits (100 x 300 μm), and diffracted x-rays are detected by a Ge solid-state detector (SSD) at a fixed diffraction angle of 6º. For the x-ray imaging, an aluminum attenuator (10 mm thick) replaces the front slits. This helps us to enlarge the beam size (2 x 2 mm) and to control intensity to optimize the image contrast. Transmitted x-rays are converted by the YAG single crystal to visible light, which is then reflected by the mirror through a microscope objective into the CCD camera. Diffraction and imaging modes can be interchanged by driving the incident slits in (for diffraction) and out (for imaging) of the x-ray beam path.

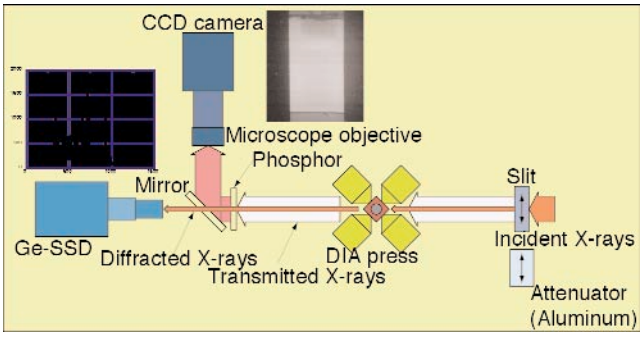

FIG. 1. X-ray diffraction and imaging setup (top view). Diffracted x-rays go above the YAG single crystal and mirror assembly and are detected by a Ge SSD. The YAG phosphor converts transmitted x-rays to visible light, which enables us to "see" inside the cell using the CCD camera. The mirror assembly for the imaging is small and far enough from the press so that it does not conflict with the diffraction assembly.

Mechanically homogenized silicate and iron compounds, containing controlled amounts of light elements (with controlled volumetric ratios of Fe/silicate from 10 to 50%) were compressed to high *P* and then heated until melting occurrs, with a cell assembly shown in Fig. 2. Starting compositions are summarized in Table 1. Droplets of Fe-rich melt, with higher x-ray absorption coefficients than the surrounding silicate, were imaged continuously throughout the entire iron-silicate separation process by synchrotron x-ray radiography, which provides real-time, two-dimensional images of melt droplets and their growth as they move through the silicate matrix or melt at high *P* and *T*. Spatial and size distribution of the iron droplets is then examined at ambient condition by computed microtomography on a series of samples quenched from various stages of the melt-silicate separation process.

Table 1. Starting materials. A mixing ratio of Fe:En=1:1 by weight corresponds to 2:5 by volume.

Light element	Reagents	Mixing ratio (by weight)
H	Fe, $Mg(OH)_2$, SiO_2	FeHx:En=1:1, 2:1, 1:2, 1:4
S	Fe, FeS, $MgSiO_3$ (enstatite)	Fe23%S:En=1:1, 2:1, 1:2
Si	Fe17%Si, $MgSiO_3$ (enstatite)	Fe17%Si:En=1:1, 2:1
C	Fe, C(graphite), MgSiO3 (enstatite)	Fe7%C:En=1:1 Fe15%C:En=1:2

Results and Discussion

The separation dynamics varied depending on the kinds of light elements involved (Fig. 3). In the systems including sulfur

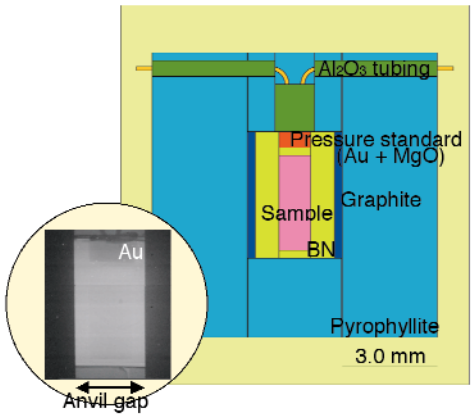

FIG. 2. Cross section of cell assembly. A pyrophyllite cube with 9 mm edge length is employed as the pressure medium. The sample is surrounded by BN and heated by the outer graphite tube. The radiograph at ambient condition (inset) shows homogeneous intensity before melting occurs. The contrast between the homogeneously mixed sample and BN capsule is poor. Darker area in the sample chamber is an Au+MgO mixture, which is used as pressure marker.

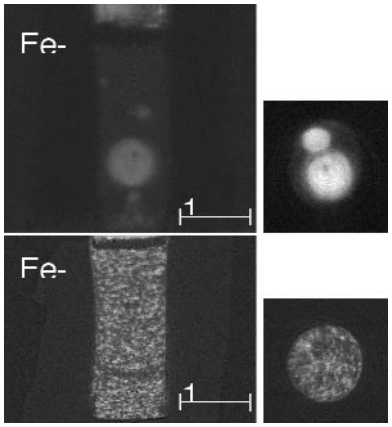

FIG. 3. X-ray microtomographs on the recovered samples (top: Fe-S system quenched from 4.4 GPa and 1400ºC; bottom: Fe-Si system quenched from 3.4 GPa and 1500ºC).

and hydrogen, iron-melt forms spherical droplets, which migrate rapidly through the silicate matrix or melt, forming large spheres at the bottom. We reduced the Fe content to see if the same features were observed with a small amount of Fe present. The sample with 10% volumetric Fe content still showed small droplets at the bottom of the capsule. These observations indicate that the viscosities of the melts are low and the dihedral angles between the melts and the silicates are quite large. In the systems including silicon and carbon, on the other hand, no melt droplets are observed even at the highest temperature of 1500ºC in the experimental time scale (about 1 h), although x-ray diffraction indicated that the iron alloy had been melted. Microtomography on these samples with the resolution of 5 μm showed small channels inside the sample capsule, indicating smaller dihedral angles. These results indicate that the precise segregation mechanism is quite sensitive to the identity of the light element.

Acknowledgments

We thank N. Lazarz, F. Sopron, M. Jagger, G. Shen, M. Newville, P. Eng, J. Pluth, P. Murray, C. Pullins, L. Gubenko, and P. Dell for their valuable contributions. Work was performed at GSECARS, APS at Argonne National Laboratory. GSECARS is supported by the National Science Foundation-Earth Sciences, Department of Energy-Geosciences, the W. M. Keck Foundation, and the United States Department of Agriculture. Use of the Advanced Photon Source was supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, under Contract No. W-31-109-ENG-38.

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