Microtexture Characterization on Fe-rich Alloy Melt in Mantle Minerals: Microtomography on Samples Recovered from High-pressure and High-temperature Experiments

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Introduction

The density deficit of the Earth's core as compared to that of pure iron suggests that certain light elements are present in the core [1, 2]. So far S, H, C, Si, and O have been considered as candidate elements, with no consensus on which one is the most predominant because of the lack of knowledge about this portion of the Earth [3]. The experimental simulation of possible core formation processes in the presence of controlled amounts of light elements (S, H, C, and Si) and mantle minerals (Mg_2SiO_4, $MgSiO_3$, and MgO), summarized in Tables 1 and 2, has been performed with synchrotron x-ray radiography and diffraction techniques under high pressure and high temperature at the GSECARS facility at the APS. In the present study, we use x-ray microtomography to explore the microstructure produced during melting of the Fe-rich components in the simulation experiments, by examining a series of recovered samples quenched from various pressure and temperature conditions.

Methods and Materials

Two series of high-pressure experiments were carried out during the Fe-rich melt segregation experiments. In one, natural enstatite was used to represent the mantle mineral, and the Fe segregation process was examined with different kinds of light elements (S, H, C, and Si) (Table 1). In the other, various mantle minerals (Mg_2SiO_4, $MgSiO_3$, and MgO) were used, and further observations were made that focused on S as the light element (Table 2).

X-ray microtomography [4] enabled the spatial and size distribution of iron droplets in the recovered samples to be observed nondestructively (Fig. 1). The data were collected on the sector 13 bending magnet beamline at the APS. The energy of the monochromatic x-ray beam was selected to be 45 keV, which is optimistic, to obtain a good contrast between the Fe-rich melt and surrounding mantle minerals. The transmitted x-rays were imaged with a single-crystal YAG scintillator (0.2-mm thick), a microscope objective, and a 1242×1152-pixel fast charge-coupled device (CCD) detector. The field of view was slightly less than approximately 3×3 mm, which resulted in the spatial resolution being about 4.0 μm (after 2×2 binning). More than 720 x-ray projections were collected by rotating the sample through 180°, then the tomographic reconstructions were done by using filtered backprojection for all of the angles in a given row. Reconstructed volume data were visualized as 2-D slices or 3-D movies.

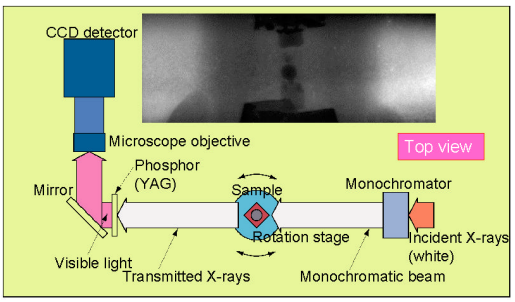

FIG. 1. Schematics showing the setup of computed microtomography. Transmitted x-rays are converted by the YAG single crystal to visible light, which is then reflected by the mirror through a microscope objective into the CCD detector.

Table 1. Starting materials.[a]

Light element	Reagents	Mixing ratio (by weight)
H	Fe, $Mg(OH)_2$, SiO_2	Fe Hx:En = 1:1, 2:1, 1:2, 1:4
S	Fe, FeS, $MgSiO_3$ (enstatite)	Fe 23% S:En = 1:1, 2:1, 1:2
Si	Fe 17% Si, $MgSiO_3$ (enstatite)	Fe 17% S:En = 1:1, 2:1
C	Fe, C (graphite), $MgSiO_3$ (enstatite)	Fe 7% C:En = 1:1, Fe 15% C:En = 1:2

[a] Fe:En = 1:1 by weight and ≈2:5 by volume.

Table 2. Starting materials and experimental conditions

Run No.	HP module	Mantle mineral	Fe:FeS_2:mantle mineral[a]	Pressure (GPa)	Temp. (°C)
D0319	DIA	$MgSiO_3$	5:1:6	4	977
D0320	DIA	Mg_2SiO_4	5:1:6	4	1027
D0321	DIA	MgO	5:1:6[b]	4	777
T0317	T-cup	$MgSiO_3$	5:1:6	16	827
T0279	T-cup	Mg_2SiO_4	5:1:6	16	1127
T0257	T-cup	MgO	5:1:6	16	827

[a] Mixing ratio is represented by weight.
[b] Pure Fe includes 33% of large grain powder.

Results and Discussion

The microstructure of the molten iron solid or partially molten silicate assemblies varies depending on the kind of light element involved (Fig. 2). In the systems containing sulfur and hydrogen, the iron melt forms spherical droplets that migrate rapidly downward through the silicate matrix (solid or partial melt). They form large spheres at the bottom of the sample chamber, indicating that the viscosities of the melts are low and the surface tension (dihedral angle) is high. In the systems containing silicon and carbon, no obvious melt droplets are observed even at the highest temperature (1500°C) in the experimental timescale (about 1 h), whereas x-ray diffraction indicates that iron-alloy has been melted. These results suggest that different mechanisms may be operating in systems involving different light elements to produce the difference in the texture.

Figure 3 compares microtomographs before and after the run in the systems Fe-FeS_2-MgO and -Mg_2SiO_4. In the system containing MgO (Fig. 3 top), the sample involved 33% (by weight) of large grains of pure Fe, which was expected to disappear and to show more uniform texture by the reaction between Fe, FeS, and MgO. Although no Fe and FeS peaks were observed at the highest temperature of 777°C in the *in situ* high-pressure experiment, suggesting that both Fe and FeS were melted, no obvious difference in texture was observed, and the edge of each Fe grain was still sharp. The system containing Mg_2SiO_4 (Fig. 3 bottom) did not show any difference, either. From the x-ray diffraction, either MgO or Mg_2SiO_4 peaks were observed at the highest temperatures, meaning the solid phase of these materials. Thus, the correlation between the reaction and sphere-forming has not been verified. Some more experiments under higher temperature (over 1500°C) are required.

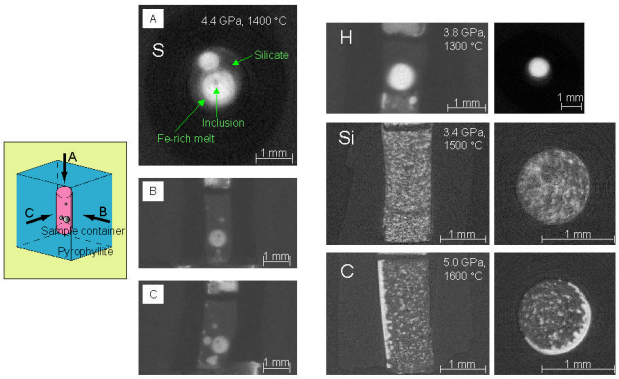

FIG. 2. Representative x-ray microtomographs on recovered samples. Bright areas represent higher absorption (containing heavier elements). The picture at the far left shows the schematics of the recovered sample of the system, including sulfur. In the system containing sulfur, large spheres can be seen. Silicate inclusions, one of which is indicated, can be seen in the molten metal. In the system containing carbon, Fe may react with the surrounding sample container because most of the Fe was seen to be accumulated along the capsule. In the system including silicon and carbon, we were not able to see any spheres, although the temperature is higher than in the experiments of sulfur- and hydrogen-contained systems.

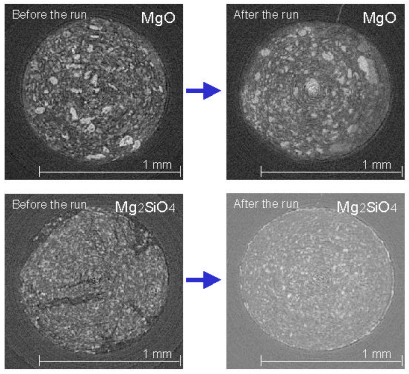

FIG. 3. Comparison of x-ray microtomographs before (left) and after (right) the runs (top: D0321, bottom: D0320). Although in situ x-ray diffraction showed no Fe or FeS peaks at the highest temperature, no obvious sphere was observed in the texture.

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References

[1] A. P. Jephcoat and P. Olsen, Nature **325**, 332-335, (1987).

[2] T. Uchida, Y. Wang, M. L. Rivers, and S. R. Sutton, J. Geophys. Res. **106**, 21799-21810 (2001).

[3] J.-P. Poirier, Phys. Earth Planet. Inter. **85**, 319-337 (1994).

[4] M. L. Rivers, S. R. Sutton, and P. Eng, in *Developments in X-ray Tomography II*, edited by U. Bonse (International Society for Optical Engineering, Bellingham, WA, 1999), pp. 78-86.