

PERCOLATIVE BEHAVIOR OF IMMISCIBLE LIQUIDS AT HIGH PRESSURE AND TEMPERATURE: IMPLICATIONS FOR COMPOSITION OF PLANETARY CORES. Yingwei Fei¹, and Yuki Shibazaki¹,
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Introduction: Internal differentiation of planetary bodies is one of the most important processes that define the internal structure and chemical redistribution. The planetesimals were likely differentiated once the metallic iron alloys reached melting temperature due to heating from sources such as radioactive decay of short-lived isotopes such as ²⁶Al and ⁶⁰Fe, impact, and release of potential energy [1]. It is important to understand how the liquid metal percolated through a silicate matrix during the early differentiation.

Percolation of liquid metal in solid silicate matrix is likely a dominant process in the initial differentiation when the temperature is not high enough to melt the entire planetary body. During the core formation, different amounts of multiple light elements enter the iron dominant core, depended on the initial composition and physical conditions of the core-forming event, such as pressure, temperature, and oxygen fugacity. Therefore, it is critical to understand percolative behavior of metallic liquid with multiple light elements in the solid matrix. The Fe-S-Si, Fe-S-O, and Fe-S-C systems form immiscible liquids at low pressure [e.g., 2-5]. Because the two immiscible liquids have different wetting properties, the immiscible liquids could be physically separated through percolation if one liquid forms smaller dihedral angle than 60° whereas the wetting angle of the second liquid is larger than the critical angle. This potentially is a novel mechanism for compositional separation during core formation for small planetary bodies, which has not been considered before.

Experimental Procedure: We have designed experiments to investigate the percolative behavior of immiscible metallic liquids in San Carlos olivine matrix over the pressure range of 2-6 GPa. The starting material is a mixture of Fe, FeS, FeO, and San Carlos olivine. Experiments at 3, 4, and 6 GPa were conducted in the multi-anvil device with an 18/11 cell assembly [6], whereas the experiments below 3 GPa were performed in the piston-cylinder apparatus. The experiments were maintained at high pressure and temperature for 6 hours to ensure texture equilibrium. The recovered samples were mounted in epoxy resin and polished for compositional mapping and quantitative chemical analysis. A series of experiments were carried out at 1873 K to evaluate the effect of pressure on the percolative behavior.

The efficiency of percolation depends on the dihedral angle, determined by the interfacial energies of the solid-solid and solid-liquid interfaces. We have developed a 3D image technique for quantitative measurements of dihedral angle [7], using a dual beam FIB/SEM (focused ion beam-scanning electron microscopy) instrument at the Carnegie Institution of Washington. The 3D imaging utilizes the combined capabilities of FIB and SEM to produce high-resolution volume rendering at the nano-scale. It provides a new, powerful tool to precisely determine the true dihedral angle. The method is far more superior than the traditional technique based on the measurements of the relative frequency distributions of apparent dihedral angles between the quenched liquid metal and silicate grains on polished 2D cross-sections. It further provides the details of each interface, allowing examination of the wetting ability of liquid in the matrix with multiple crystal phases. Through quantitative calculations, we can obtain volume fraction, surface area ratio, and connectivity.

Results: Melt in the Fe-S-O system forms immiscible liquids. Figure 1 shows a representative SEM image of the quenched texture at 6 GPa and 1873K, showing two immiscible liquids (S-rich in the edge and S-poor in the core) trapped in San Carlos olivine crystals. Similar immiscible liquid texture is also observed at 4 and 3 GPa. Although we introduced 9.4 wt% O into the melt by adding FeO in the starting material, the oxygen content in the metallic melt is limited to less than 3 wt% because of iron loss to the sample capsule.

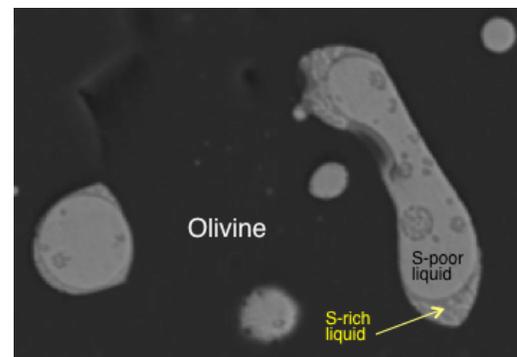


Fig. 1. A representative image of immiscible liquids in the Fe-S-O system at 6 GPa and 1873 K.

The compositions of the observed immiscible liquids are fundamentally different from those reported in previous study [3] that reported immiscible liquid gap between metallic (O-poor) and ionic (O-rich) liquids. The immiscible liquids observed in this study are both metallic liquids with limited dissolved oxygen, characterized by a S-poor and a S-rich liquid. The S-poor/Fe-rich liquid is always located in the core surrounded by the S-rich liquid. Only the S-rich liquid is wetting the olivine grains, as illustrated by the 3D volume rendering (Fig. 2).



Fig. 2. 3D view of Fe-S-O melt in an olivine matrix. The melt contains two immiscible liquids (S-poor and S-rich). The size of the bounding box is $4 \times 4 \times 2 \mu\text{m}$.

The metallic melt pockets trapped at olivine grain corners show significantly larger dihedral angle than the critical angle (60°) at 6 GPa, forming isolated, non-connected melt pockets. The dihedral angle decreases with decreasing pressure. At 3 GPa, we observed interconnected melt network in the system. The melt percolating in the grain boundaries is limited to the S-rich liquid. As visualized, the S-rich liquid controls the percolation in this case.

Discussion: The efficiency of removal of liquid metal alloys from a solid silicate matrix strongly influences the timing of core formation and the composition of the core. The ability of percolating metallic liquid in crystalline silicate depends on the dihedral angle, forming interconnected melt network at small dihedral angles ($<60^\circ$). It has been demonstrated that the dihedral angles could be strongly influenced by the compositions of the metallic melt and the crystalline silicate, particularly the oxygen content in the liquid [8, 9]. There is also significant effect of pressure on the measured dihedral angles [10]. Percolative core formation is only favoured at relatively low pressures, applicable to small planetary bodies. We have also demonstrated that immiscible liquids with different wetting ability could play an important role in percolative core formation. For systems with immiscible liquids which should be common for multi-component systems at relative low pressure, the liquid in direct contact with the silicate grains controls the percolation. If that liquid has a small dihedral angle ($<60^\circ$) while the other liquid has a large dihedral angle, it could separate the two liquids through percolation. Such a mechanism for compositional separation during core formation could have operated for the differentiated planetesimals.

References: [1] Greenwood R. C. et al. (2005) *Nature* 435, 916-918. [2] Sanloup C., and Fei Y. (2004) *PEPI* 147, 57-65. [3] Tsuno K. et al. (2007) *PEPI* 160, 75-85. [4] Corgne A. et al. (2008) *GCA* 72, 2409-2416. [5] Terasaki H. et al. (2011) *EPSL* 304, 559-564. [6] Bertka C. M. and Fei Y. (1997) *JGR* 102, 5251-5264. [7] Fei Y. (2013) *JoVE* 81, e50778. [8] Gaetani G. A. and Grove T. L. (1999) *EPSL* 169, 147-163. [9] Terasaki H. et al. (2005) *EPSL* 232, 379-392. [10] Terasaki H. et al. (2008) *EPSL* 273, 132-137.