

EFFECT OF CARBON AND SULFUR ON IRON MELTING AT HIGH PRESSURE: IMPLICATIONS FOR COMPOSITION AND EVOLUTION OF THE TERRESTRIAL PLANET CORES. Y. Fei¹, L. Deng^{1,2}, A. Corgne³, ¹Geophysical Laboratory, Carnegie Institution of Washington, 5251 Broad Branch Rd., N.W., Washington DC 20015, USA (e-mail: fei@gl.ciw.edu), ²Institute of Physics, Southwest Jiaotong University, Chengdu 610031, P.R. China, ³GEMOC, Macquarie University, North Ryde, NSW 2109, Australia

Introduction: It is certain that planetary differentiation involves extensive melting of planetary materials through the decay of the short-lived radionuclides ²⁶Al and ⁶⁰Fe, and impact heating. Evidence from cosmochemical observations, refined seismic data, high-pressure experimentation, and theories of geomagnetism leads to the idea of iron being the dominant component of the cores of the Earth and other planetary bodies [1]. Detailed comparison between the experimentally measured density of pure iron and the observed density of the Earth's core from seismic data revealed the density deficit in the core (both liquid outer core and solid inner core). The density deficit may be explained by incorporation of certain amounts of light elements (lighter than iron) such as sulfur (S), carbon (C), silicon (Si), and oxygen (O).

During the core formation, it is likely that different amounts of light elements enters the iron dominant core depended on the initial composition and physical conditions of the core-forming event, such as pressure, temperature, oxygen fugacity. The composition and evolution of the core during cooling are strictly controlled by the melting relations and element partition in this multi-component system. Therefore, it is critical to understand melting behavior of Fe alloying with light elements at high pressure and temperature.

Among all the potential light elements that may present in the Earth's core, carbon and sulfur are the prominent candidates. Fe-S melt was the first melt to percolate through the surrounding silicates during differentiation because of the low eutectic melting temperature of the Fe-FeS system. On the other hand, carbon is extremely abundant in the solar system and has strong chemical affinity for Fe. Adding a small amount of carbon into Fe-S will cause a vast liquid immiscibility gap, which may have great effect on the melting relationship of the Fe-C-S ternary system. We have previously determined the eutectic temperature as a function of pressure in the Fe-FeS [2-5] and Fe-C [6] systems. The eutectic temperature in the Fe-C system increases linearly with increasing pressure, from 1420 K at 1 bar to 1975K at 25 GPa, whereas the eutectic temperature in the Fe-FeS system initially decreases with increasing pressure, and then increases at pressures above 14 GPa after new high-pressure iron-sulfur compounds (Fe₃S₂, Fe₂S, and Fe₃S) forms. In this study, we focus on determining melting relations

in the Fe-C-S system at high pressure and temperature, with implication for evolution of the planetary core.

Experimental procedure: Melting experiments in the systems Fe-C and Fe-C-S were performed up to 25 GPa and 1973 K, using a piston-cylinder and multi-anvil apparatus at the Geophysical Laboratory. Starting materials with different carbon and sulfur contents were prepared by mixing fine powder of pure iron (Fe), iron sulfide (FeS), and graphite (C). Compositions of the starting material in the Fe-C system were described in [6]. Two compositions in the Fe-C-S system were prepared, 90Fe-5C-5S wt% and 90Fe-2C-8S wt%. MgO capsules were used to contain the samples. For the multi-anvil experiments, well-calibrated high-pressure assemblies (18/11, 10/5, 8/3) were used to achieve high pressures up to 25 GPa [7]. After reached the experiment pressures, samples were heated to the targeted temperatures, measured with W5%Re-W26%Re thermocouple. Samples were quenched to ambient condition and prepared for composition and texture analysis. Melting relations were determined with a JEOL JXA-8900 electron microprobe and SEM, based on quench textures and chemical composition analyses of the quenched phases. Powder X-ray diffraction technique was also used to identify phases and determine unit cell parameters.

Results: Based on the knowledge of eutectic melting in both Fe-C and Fe-S binary systems, we conducted experiments in the Fe-C-S ternary system to evaluate the effect of carbon and sulfur on melting relations. At low pressure (P = 3.5 GPa), we observed two immiscible C-rich and S-rich Fe-C-S liquids at 1873 K. As temperature decreases, Fe₃C crystallizes, coexisting with a Fe-C-S melt. At a pressure of 5 GPa, we observed only one Fe-C-S liquid, indicating miscibility gap closure at a pressure between 3.5 GPa and 5 GPa. The observation is consistent with result of a recent study [8] in a chondrite model composition in which miscibility gap closure was observed at a pressure between 4.8 and 6.2 GPa. The miscibility gap closure at 4.9 GPa has important implications for planetary differentiation and core composition stratification for small planetary body.

We conducted experiments using starting materials containing 90 wt% metallic iron and 10 wt% C and S with different C/S ratios. All experiments at pressures of 5, 10, and 20 GPa produced a single homogenous Fe-C-S liquid above the liquidus temperature. With

decreasing temperature, Fe-C compounds first solidify, coexisting with a S-rich Fe-C-S melt. The solid Fe-C compounds contain no sulfur, whereas the melt is S-rich and its composition varies depending on the starting composition and temperature. The melt also contains detectable amounts of oxygen. In addition to the change of melting temperature as a function of pressure, we also observed a change of melting relations between 5 and 10 GPa. At 5 GPa, S-rich Fe-C-S melt coexists with Fe_3C , whereas the melt coexists with Fe_7C_3 at 10 and 20 GPa for the same starting composition, indicating Fe_3C melts incongruently at pressures above 5 GPa.

Discussion and implications: Our experimental results in the Fe-C-S system should provide necessary information for understanding core evolution in differentiated planetary bodies with the cores containing both S and C. The observed S-rich and C-rich immiscible liquids at low pressures (< 5 GPa) would lead to a stratified core for a small planetary body, with a denser C-rich liquid inner core and S-rich liquid outer core. As the core cools down, the C-rich liquid would crystallize Fe_3C first and then coexisting Fe_3C and metallic Fe. Significantly lower temperature is required to solidify the S-rich melt.

For a planetary core that is sufficiently large so that the core pressures are greater than 5 GPa, no composition stratification in the core is expected. Based on crystallization sequence, iron carbide or metallic iron would crystallize depending on the initial Fe/(C+S) ratio. With 90%Fe in the starting materials, iron carbide is the liquidus phase. In order to crystallize metallic iron with dissolved carbon, it is necessary to increase the initial Fe/(C+S) ratio. If the Earth's core contains both carbon and sulfur, the solid inner core is nearly S-free, but it could contain a significant amount of carbon. On the other hand, the liquid outer core would be S-rich and C-poor.

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