Resolution of inconsistencies in experiments of iron-silicate sulfur partitioning based on ab initio calculations

Mr. Kei Ito

PhD Student (D1)
Geodynamics Research Center

2023.11.10 (Fri.) 16:30 ~

Venue: Meeting Room #486

Science Research Bldg. 1, 4th floor.
Ehime Univ.

Keywords: 1. Sulfur
2. Metal-silicate Partititoning
3. ab initio Calculation

Depletion of S in the mantle relative to its cosmic abundance (Lorand et al., 2013) makes S one of the most promising elements that could explain the density deficit in the core (Birch, 1952). However, there are significant discrepancies in past experimental results on the partitioning of S between liquid iron and molten silicate. A study using a diamond anvil cell (DAC) (Suer et al., 2017) reported a partitioning coefficient ($\log D_S$) of ~10 for S at high pressure (40-90 GPa). But, this result differs significantly from the extrapolated values (100~10000) of the results using a multi-anvil (MA) or piston cylinder (PC) (Rose-Weston et al., 2009 etc.). For this reason, studies using DAC indicate that S cannot be a major light element in the core. To resolve this discrepancy, we calculated the sulfur partitioning using theoretical methods.

In this study, we investigate the S partitioning behavior between liquid iron and molten silicate over a wide range of pressure and temperature conditions (3000-5000 K, 0-135 GPa) using ab initio free energy simulations based on the thermodynamic integration method (Taniuchi & Tsuchiya, 2018).

The $\log D_S$ has a positive pressure dependence, and it becomes smaller at higher pressures than at lower pressures. The $\log D_S$ has a negative dependence on the oxygen content in liquid iron. S has a large pressure dependence from 0-20 GPa, similar to the MA&PC results; from 20-150 GPa, the apparent pressure dependence becomes nearly zero or negative as the amount of oxygen in liquid iron increases, closer to the DAC results with increasing pressure. This suggests that the major discrepancy between the experiments may be due to the fact that the experiments were not performed over a wide pressure range and the effect of oxygen dissolution in the liquid iron at high pressure.

Contact: Steeve Gréaux, PhD. (e-mail: greaux@ehime-u.ac.jp)