

Experimental investigation of sulfidation of silicates, glasses and melts under reducing conditions

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Sulfur is a key volatile element to understand processes from accretion in the solar nebula, to core formation, and planetary volcanism. In most of these environments polyvalent S occurs in its reduced state. S-rich gases are predicted, for example, driving lunar pyroclastic eruptions [1]. Reactions of these gases with silicates only leave indirect evidence, such as in sulfidation textures observed in lunar rocks [2] or enstatite chondrites [3]. To better constrain these processes, we present experimental results on sulfidation reactions between reduced S-C-O gas and silicates.

We ran the experiments in evacuated silica glass tubes with the minerals olivine (Fo_{92}), anorthite, diopside, and three glasses with Mercury compositions; high-Mg, low-Mg and high-Al [4]. The silicates are placed in a graphite cup in the glass tube, above a graphite cup that contains elemental sulfur (Fig. 1). At high temperatures (800-1200 °C) the sulfur forms a gas and reacts with the silicate samples. The graphite cups control the $f\text{O}_2$ at the C-CO buffer [5]. The experiments were run for 24h.

At 800 °C sulfidation is kinetically limited and only occurs on the surface of mineral and glass grains. However, at higher temperatures (1000, 1200 °C) the reaction becomes increasingly pervasive and creates porosity in glasses and minerals (Fig.2). Observed sulfide reaction products include CaS , $(\text{Ca,Mg})\text{S}$, $(\text{Ti,Fe,Ca,Mg})\text{S}$ and $(\text{Fe,Ti})_{1-x}\text{S}$. The Mg/Ca-ratio in the sulfides is a function of the silicate composition and increases with temperature. An additional important reaction product is SiO_2 .

Our experiments show that sulfidation reactions of silicates are an efficient sink for sulfur at reducing conditions. These gas-solid reactions may have played a key role in the enrichment of S at the surface of Mercury. Therefore, the experimental products are used as analog materials and analyzed by mid-infrared spectrometry as a reference for space missions (e.g. MERTIS onboard the ESA/JAXA mission BepiColombo to Mercury [6]).

[1] Renggli et al. (2017) *GCA*, 10.1016/j.gca.2017.03.012. [2] Shearer et al. (2012) *GCA*, 10.1016/j.gca.2011.11.031. [3] Fleet & MacRae *GCA*, 10.1016/0016-7037(87)90333-4. [4] Morlok et al. (2021) *Icarus*, j.icarus.2021.114363. [5] Renggli & Klemme (2021) *JGR Planets*, 2020JE006609. [6] Hiesinger et al. (2020) *Space. Sci. Rev.*, 10.1007/s11214-020-00732-4.

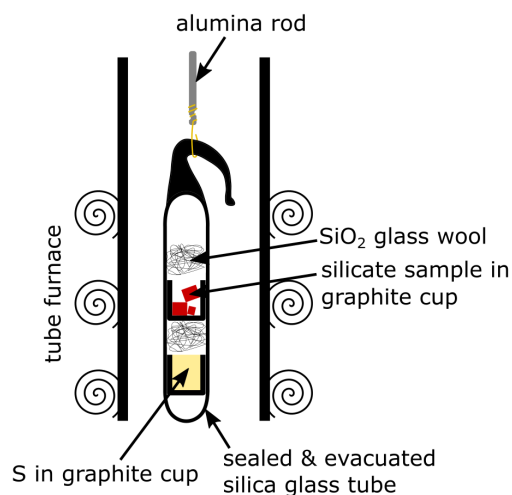


Fig. 1: Experimental set-up. Evacuated silica glass tube suspended in the hot zone of a vertical tube furnace. The graphite cups control the $f\text{O}_2$ at the C-CO buffer and the elemental S forms a S-C-O gas phase at 800, 1000 and 1200 °C.

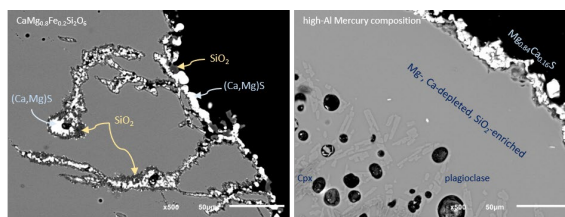


Fig. 2: Back-scattered electron images of cross-sections through diopside (left) and a high-Al Mercury melt [4] (right) reacted at 1200 °C. The diopside reacts to $(\text{Ca,Mg})\text{S}$ and SiO_2 , whereas the Al-rich melt starts crystallizing clinopyroxene and plagioclase in the interior and $\text{Mg}_{0.84}\text{Ca}_{0.16}\text{S}$ forms on the surface. The boundary layer is depleted in Ca and Mg, and enriched in SiO_2 .