

Sulfur speciation at 2.6 GPa and 700-900°C in hydrous fluids: a key to understand sulfur transfer from subducted slabs to the sub-arc mantle

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Sulfur potentially plays a significant role in the oxidation of the sub arc mantle and the redox evolution of arc magmas, thus it is important to understand its mobility in high-pressure fluids and melts. Currently available data is mainly limited to silicate melts due to the challenging nature of experimentation with sulfur-bearing fluids at these extreme conditions.

We conducted high-pressure experiments with sulfur-bearing aqueous fluids at controlled redox conditions in a piston cylinder apparatus to shed light on the fO_2 range of sulfide-sulfate transition at $P=2.6$ GPa and $T=700-900^\circ\text{C}$. Since fluids are non-quenchable, we sampled the pre-equilibrated fluid at the experimental run conditions by the entrapment of synthetic fluid inclusions in quartz, which was fractured *in situ* during the experiment by passing the coesite to quartz transition [1]. As the average oxidation state of sulfur in the fluid inclusions cannot be significantly modified during rapid quenching, the speciation of sulfur in the fluid inclusions was determined at ambient conditions by using Raman spectroscopy, and the average oxidation state of sulfur was calculated based on these data.

The run products from the Ni-NiO buffered experiments contained H_2S as the only significant sulfur species in the fluid, whereas synthetic fluid inclusions from the Re-ReO₂ buffered experiments conducted over the entire temperature range contained sulfur in the form of SO_4^- , H_2S and native sulfur daughter mineral. The presence of a large fraction of the total sulfur budget in the form of native sulfur implies significant comproportionation of reduced and oxidized sulfur species upon quenching. Mass balance calculations show that sulfur was present dominantly in reduced form at the experimental P - T conditions, contrary to the results of studies conducted at lower pressures, which found the predominance of oxidized sulfur species at fO_2 buffered by Re-ReO₂ [2]. These results are well aligned with the sulfide-sulfate transition in silicate melts shifting to higher fO_2 with increasing pressure [3].

[1] Tsay et al. (2016) *American Mineralogist* 101, 10, 2199–2209. [2] Binder and Keppler (2011) *Earth and Planetary Science Letters* 301, 1–2, 190–198. [3] Matjuschkin et al. (2016) *Contributions to Mineralogy and Petrology*, 171, 7, 66.