Investigating the effect of pressure on the sulfide-sulfate transition in silicate melts: Implications for the behaviour of chalcophile elements and redox state of magma sources

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Sulfur is a minor but ubiquitous element in magmas, and is behind major volatile species in magmatic fluids and volcanic gases. It has been shown that sulfur dissolves in silicate melt either as reduced species (sulfide, $S^{2-}$) at oxygen fugacity ($f_O^2$) values typically below that of the FMQ buffer, or as oxidized species (sulfate, $S^{6+}$) at $f_O^2$ generally above ~FMQ+2. In between these two domains lies the sulfide-sulfate transition in the course of which the solubility of sulfur at sulfide saturation (SCSS) increases drastically until it reaches the solubility of sulfur at anhydrite saturation (SCAS). Over this limited $f_O^2$ range, sulfur dissolves in the silicate melt as both $S^{2-}$ and $S^{6+}$. It is widely considered in the literature that the $f_O^2$ range at which the sulfide-sulfate transition occurs is fixed, independently of the intensive parameters (pressure, temperature, melt composition).

Yet, recent experimental studies (Jégo et al., 2016, GCA; Matjuschkin et al., 2016, CMP) suggest that this transition would be shifted towards higher (i.e., more oxidizing) $f_O^2$ with increasing pressure. If such pressure effect proves to be real, it could have important consequences as for the behavior of chalcophile elements during mantle melting, and magma rise and fractionation, as well as for the actual redox state of magma sources.

We tested this hypothesis by performing melt sulfur solubility experiments over a wide pressure range, imposing oversaturation in both sulfide and sulfate so as to target the transition, and measuring the corresponding $f_O^2$ with solid sensors. Melt sulfur content and speciation were determined by EMP and Raman analyses.