

# Temperature, melt composition and oxygen fugacity effects on sulfur redox state in silicate melts

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Sulfur plays a fundamental role in many different igneous processes (volcanic degassing, the formation of magmatic ore deposits and planetary differentiation for example). The wide range of stable sulfur oxidation states (-2, 0, +4 and +6) controls distribution of the element between the major terrestrial reservoirs, although in silicate melts [1] showed that S occurs almost exclusively as sulfide ( $S^{2-}$ ) and sulfate ( $S^{6+}$  or  $SO_4^{2-}$ ). Here we address the fact that despite this long-standing understanding, experimental measurements of  $S^{2-}$  and  $S^{6+}$  concentration in natural melts have not been comprehensive in terms of composition and temperature.

We investigated experimentally at atmospheric pressure the effects of temperature (1200-1500 °C) and melt composition (14 melt compositions) on both sulfide and sulfate solubility under controlled conditions of oxygen and sulfur fugacity. The S and major element contents of the quenched glasses were determined by electron microprobe with the lowest S contents (i.e., < 60 ppm) determined by SIMS. We used sulfide and sulfate capacities ( $C_{S^{2-}}$  and  $C_{S^{6+}}$ , respectively) to express  $S^{2-}$  and  $S^{6+}$  solubilities as a function of the  $fO_2$  and  $fS_2$ .

The data show that sulfide capacity increases with both FeO concentration in the melt and increasing temperature while sulfate capacity increases with CaO in the melt and with decreasing temperature. Taken together the results enable estimation of the S redox state ( $S^{6+}/S^{2-}$  ratio) as a function of melt composition, temperature and oxygen fugacity. The results are in good agreement with earlier measurements of  $S^{6+}/S^{2-}$  in melts and we calculate a significant increase relative to FMQ (Fayalite-Magnetite-Quartz oxygen buffer) of the oxygen fugacity of the  $S^{2-}$  to  $S^{6+}$  transition as temperature decreases from 1500 to 1200 °C. We used our results to simulate sulfur-degassing paths for various melt compositions under a range of redox conditions (FMQ -2 log  $fO_2$  units to FMQ +2). Degassing an  $H_2O$ -rich fluid should generally lead to an increase in oxygen fugacity both at low  $fO_2$ , through degassing of  $H_2$  and  $H_2S$ , and at high  $fO_2$  through loss of  $H_2O$  and  $SO_2$ .

[1] Fincham and Richardson (1954) Proc. Royal Soc. London. Series A. Math. Phys. Sciences, 223, 40-62.