

Sulfur solubility in sediment melts as a function of oxygen fugacity at 3 GPa, 1050°C

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If sulfur (S) recycling during subduction is accomplished through the fluid mediums formed during slab devolatilization and fluxed-melting, the amount of S being recycled depends on the solubility of S in the fluids, which, in turn, is a function of pressure, temperature, fluid composition and oxygen fugacity (fO_2). Here we report an experimental investigation of the fO_2 effect on S solubility in sediment melts at 3 GPa, 1050°C. The experimental pelite starting material (EPSM) contains ~7 wt% H₂O and ~1.9 wt% S, which produced a simple assemblage of melt+pyrrhotite/anhydrite+Garnet+coesite±kyanite±rutile. Oxygen fugacity was varied by employing solid buffers in a “double capsule” configuration. Sulfur content at sulfide saturation (SCSS) in melt was found to increase with decreasing fO_2 from ~200 ppm at FMQ+0.7 (FMQ stands for the Fayalite-Magnetite-Quartz buffer) to ~1900 ppm at FMQ-6. Sulfur content at sulfate saturation (SCAS) is ~2600 ppm at FMQ+2.7. Raman spectra of experimental melts have shown that S exists as H₂S/HS at reduced conditions and as SO₂ at oxidized conditions. The solubility minimum, i.e., the beginning of the transition from S²⁻ to S⁶⁺ is estimated to be at FMQ+0.7, while the transition finishes at about FMQ+2.7. Therefore, at relatively oxidized conditions ($fO_2 > FMQ+1$), sediment melts can contain hundreds, even thousands ppm S, potentially act as the transfer medium for S, especially in its oxidized form. Our SCSS data do not fit the popular Liu et al. (2007) [1] model, but were reproduced nearly perfectly using the empirical model proposed by Clemente et al. (2004) [2] for hydrous rhyolitic melts. Moreover, we observed a linear correlation between logSCSS and logXFeO (the mole fraction of FeO in melt) with a slope close to -1. Based on the solubility mechanism of H₂S in hydrous silicate melt: $H_2S_{gas} + OH_{melt} = H_2O_{gas} + HS_{melt}$ (Burnham, 1979 [3]), we defined a hydro-sulfide capacity C_{HS} with the relation: $\ln C_{HS} = \ln[HS] + \ln(f_{HSO}/f_{HS})$. In combination with the equilibrium between melt and FeS, we obtained an expression for SCSS, which explains the negative dependence of SCSS on FeO content in melt.

[1] Liu et al. (2007) *Geochim. Cosmochim. Acta* **71**, 1783–1799. [2] Clemente et al. (2004) *J. Petrol.* **45**, 2171–2196. [3] Burnham (1979) *The evolution of the igneous rocks.*