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The role of Iron on the dissolution of Sulfur in hydrous silicate melts

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It is well established that the sulfur solubility in silicate melts is a function of fO_2 and melt compositions [1]. At high fO_2, S^{6+} is dominant and more soluble in natural silicate melts than S^{2-}, which is dominant at low fO_2. Primitive melt compositions such as basalt can contain an order of magnitude more sulfur than more evolved melt compositions such as rhyolite. The compositional control on the sulfur solubility is yet not fully understood but a positive correlation with FeO has often been observed. Thus, determining the S^{2-}- and S^{6+}-speciesincorporated in the silicate glass structure is crucial to unravel the sulfur dissolution mechanism in silicate melts. Together with solubility measurements such information is essential to quantify the sulfide and sulfate capacity of silicate melts and to understand the sulfur flux in subduction zones.

Here we present Raman and XANES measurements on experimentally equilibrated sulfur-bearing, hydrous silicate glasses, including soda-lime (SLG), $K_2Si_4O_9$ (KSG), albite and trondhjemite (TROND) compositions. A series of SLG and KSG glasses, doped with small quantities of Fe, were also studied in order to determine the effect of Fe/S on the S solubility. The experiments were performed in internally heated and cold seal pressure vessels at 200 MPa, 1000 and 850 °C and fO_2 ranging from $\log fO_2$ = QFM-2.4 to QFM+4.

The systematic correlation of features in Raman and XANES spectra allows the identification of at least four different S-species in the glasses depending on fO_2 and Fe/S of the system. In Fe-free melts S is dissolved as SH^-, H_2S and/or SO_4^{2-} depending on the prevailing fO_2. S^{2-} is more soluble than S^{6+}.The total sulfur solubility depends on the degree of polymerisation of the melt and increases with increasing NBO/T. This correlation is much more pronounced for SH^- and H_2S than for SO_4^{2-}. Adding Fe results in the formation of Fe-S-complexes at the expense of SH^- and H_2S, which are still observed up to Fe/S $\sim$ 2.6. The S^{6+}/S^{2-} equilibrium in Fe-free systems is at $\sim$1.5 log units lower fO_2 than observed for Fe-rich basaltic and andesitic systems [2,3].

This may have consequences for the S-cycle in subduction zones as Fe-poor slab-derived liquids are able to carry S^{6+} which may act as an oxidising agent in the mantle wedge by successively oxidising Fe^{2+} to Fe^{3+}.

References:

[1] Carroll & Rutherford (1985) *Am Mineral* 73, 845-849.

[2] Jugo et al. (2010) *GCA* 74, 5926-5938.

[3] Botcharnikov et al. (2011) *Nature Geo 4*, 112-115.