



Experimental Constraints on the Redox and Compositional Control of S Solubility in Hydrous Felsic to Mafic Magmas

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Knowledge on the solubility of sulphur in natural silicate melts provides important constraints on geochemical processes occurring in magmatic and hydrothermal systems. The solubility of S in magmas is mainly controlled by the transformations of S species into sulphide or sulphate form due to changes in the redox conditions of the system. The saturation of a silicate melt in respect of sulphide- or/and sulphate-bearing phase is responsible for the maximum concentrations of dissolved sulphur. Hence, the composition of the silicate melt and activities of melt components, especially Fe and Ca, are also critical parameters controlling S solubility. Although the experimental database on S solubility is relatively large, it is mainly focused on hydrous silicic or dry basaltic melts and little is known about the behaviour of S in hydrous intermediate and mafic systems in a wide range of redox conditions.

Here we present new experimental data on the solubility of S in rhyolitic to basaltic magmas equilibrated with aqueous S-bearing fluids of constant bulk composition (5 wt.% H₂O and 1 wt.% S in the system) at 1050°C, 200 MPa and redox conditions corresponding to that of log $fO_2 \sim$ QFM-2 to QFM+3 (where QFM is quartz-fayalite-magnetite oxygen buffer). Additional data have been obtained for alkali-rich phonolitic and tephriphonolitic compositions as well as for Fe-rich basaltic melt. For typical

magmatic melts in the compositional range from rhyolite to basalt, the solubility of S systematically increases with increasing fO_2 and decreasing SiO_2 content of the melt. At $\log fO_2 < \sim QFM$, S solubility is controlled by the stability of FeS phase while at $\log fO_2 > \sim QFM+2$, $CaSO_4$ is a stable phase. Both S-bearing phases coexist at intermediate fO_2 values. The largest difference in solubilities of sulphide and sulphate S is observed for basaltic melts, where the concentration of sulphide S is 0.07 wt.%, being approximately ten times lower than the concentration of sulphate S of about 0.6 wt.%. Other melt compositions show lower concentration differences between dissolved S species, having, however, similar positive dependence on fO_2 . For all investigated melt compositions, the transition between concentrations of sulphide and sulphate S in the fO_2 range from QFM to QFM+2 is continuous which is consistent with continuous change in S speciation in the melt as a function of the redox state (Wilke et al., EGU 2008). At given T, P and redox conditions, the solubility of S is controlled by the melt composition with the best correlation with SiO_2 concentration in rhyolitic to basaltic melts. Alkali-rich and Fe-rich compositions show significant deviations from the S vs. SiO_2 trends. However, at $\log fO_2 > QFM+1$, S solubility for all studied compositions correlates well with the concentration of Ca in the melts but at lower fO_2 it seems that neither Ca nor Fe have main control on the amount of dissolved S. Also no significant correlation is observed for alkalis. The obtained experimental data can be used to predict the concentrations of dissolved S and to estimate the prevailing redox conditions in natural magmas saturated with S-bearing phase(s) and to calibrate the existing thermodynamic models.

Wilke et al. (2008) Sulfur and iron oxidation state in quenched melts as a function of oxygen fugacity. EGU 2008, Session GMPV12.