Temperature and melt composition effects on sulfate solubility in silicate melts at atmospheric pressure

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Knowledge of the solubility of sulfur (S) in silicate melts is fundamental to an understanding of a number of different igneous processes (e.g., volcanic degassing, the formation of magmatic ore deposits and planetary differentiation). Several oxidation states of sulfur are present in natural environments and these exert control over the distribution of sulfur between Earth’s different geochemical reservoirs (mantle, core, crust, atmosphere, oceans). Although the solubility of sulfide (S\(^2\)) in silicate melts is well constrained under the reducing conditions (below the fayalite-magnetite-quartz FMQ oxygen buffer) where it is stable, only a few data are available for sulfate (S\(^6\) or SO\(_4^{2-}\)) solubility under the more oxidizing conditions relevant to many volcanic systems.

In this study, we investigated the influence of temperature and melt composition on sulfate solubility in silicate melts using equilibration experiments at atmospheric pressure under fixed oxidizing \(f_{O_2}\) (i.e., \(\log f_{O_2} = -1.13\)) and relatively high sulfur fugacity. The temperature was varied from 1200 to 1500°C and 14 different silicate melt compositions were studied (e.g., andesite, basalt, basanite, phonolite). The S contents of the quenched run products (i.e., silicate glasses) were determined by electron microprobe with \textit{in-situ} SIMS being used for the lowest S concentrations.

The data obtained here highlight a fundamental control of both the temperature and melt composition on the sulfate solubility. At constant \(f_{O_2}\), as the temperature increases, the sulfate solubility decreases significantly. For example, with a fixed An/SO\(_2\) ratio of 0.6, the sulfate solubility decreased by a factor \(~4\) as temperature was increased from 1300 to 1400°C (e.g., for a basalt composition, the S content decreased from 448±20 ppm to 118±17 ppm). Concerning the melt composition effect, a strong positive correlation is observed between S solubility and CaO content (e.g., from 229±16 ppm to 1935±25 ppm at 1300°C for a basalt with 9.7% CaO and a CMAS like composition with 21.2% of CaO respectively). The new sulfate solubility results are used to provide new equations to determine the sulfate capacity of silicate melts at atmospheric pressure in order to constrain S behavior in oxidizing magmatic processes (e.g., Earth near-surface environments).