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Pressure, water, and, diffusion controls on sulfur speciation and solubility in silicate melts

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Sulfur is a key magmatic volatile controlling the composition of volcanic gases, the timing of sulfide saturation, and the release of chalcophile metals. In silicate melts sulfur occurs primarily as sulfide (S^{2-}) and sulfate (S^{6+}), with speciation known to be governed by temperature, composition, and oxygen fugacity. While sulfur speciation at 1 bar is well characterised, the combined effects of pressure, dissolved H_2O, and diffusion kinetics during magma ascent remain debated.

We present new experimental constraints on sulfur solubility, speciation, and diffusion in silicate melts at 1200-1500 °C and pressures from 1 bar to 2.5 GPa. Sulfide capacities were determined using experiments in which sulfur fugacity was buffered with Ag–Ag_2S assemblages and oxygen fugacity was fixed at the C–CO–CO_2 equilibrium, while sulfate capacities were obtained from anhydrite-saturated melts. Both sulfide and sulfate capacities decrease systematically with increasing pressure, reflecting positive partial molar volume differences between sulfur species and calcium oxide in the melt. When combined with 1-bar datasets, results show that increasing pressure stabilises sulfate relative to sulfide, shifting the S^{2-}–S^{6+} crossover to lower oxygen fugacity relative to FMQ by ~0.4 log units per GPa at 1400 °C, due to S-Fe electron exchanges through degassing during magma ascent.

Initial results for hydrous experiments, with melt H_2O contents from ~1 to 8 wt%, demonstrate that increasing dissolved water systematically reduces total sulfur content at sulfide saturation, under fixed redox conditions, opposite to trends inferred from previous unbuffered studies. This suggests that hydrous melts may reach sulfur-saturation at greater depths and begin degassing earlier than predicted by existing models.

Finally, diffusion experiments in andesite to basaltic melts show that sulfate diffuses significantly more slowly than sulfide, and more slowly than H_2O and CO_2. This kinetic contrast may limit sulfur transfer into bubbles during decompression, potentially leading to transient disequilibrium between sulfur redox in the melt and the composition of the exsolved volatile phase.

Together, these results show that pressure, water, and diffusion jointly regulate sulfur degassing pathways, and that volcanic gas compositions and melt inclusions may not faithfully record equilibrium sulfur speciation during magma ascent.