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Sulfur in cooling magmas: new experiments at crustal temperatures (1050–1250 °C) and implications for arc oxidation and metal mobilisation

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Sulfur, though a minor element in magmatic systems, strongly influences magmatic redox state, volatile evolution, and metal mobility. Its speciation between sulfide and sulfate dictates the behaviour of important metals-notably Cu, Ag, Au-linking melt evolution to the formation of ore deposits. In volcanic gases, H_2S and SO_2 evolution can indicate renewed unrest and constrain magma storage and redox conditions. Interpretations of this range of behaviours exhibited by sulfur in magmatic and volcanic processes depend on quantitative models of sulfur solubility and speciation in the melts which transport it.

Existing constraints on sulfide (S^{2-}) and sulfate (S^{6+}) in melts, commonly expressed as sulfide and sulfate capacities (C_{S2-} and C_{S6+}), come from 1 atmosphere experiments at the high temperatures of 1200-1500 °C. We present new experimental measurements for 14 silicate melt compositions at 1050-1250 °C, a temperature interval that has been little studied but is directly relevant to storage and differentiation in the crust. Using these data, we parameterise capacities as functions of temperature and melt composition, demonstrating excellent agreement with previous 1 atm experiments in the overlapping temperature range. Our results show that extrapolating earlier datasets to <1200 °C reproduces sulfide solubility but substantially overestimates sulfate solubility at low temperature. The implication is that the sulfide to sulfate cross-over shifts more strongly than previously believed to higher oxygen fugacity as temperature declines.

The stronger observed temperature dependence of the S^{2-} to S^{6+} transition implies significant Fe-S redox exchange during cooling, ascent, and differentiation of hydrous arc magmas, raising fO_2 by 2-3 log units. We propose that this temperature-driven auto-oxidation contributes to the higher redox state recorded in arcs (to ~ΔFMQ +2) relative to MORB (near ΔFMQ), because arcs typically differentiate to ~900°C whereas MORB remain >1150°C. We also find that even highly oxidised magmas (>ΔFMQ +2) remain sulfide-saturated under crustal conditions. Given that such stable sulfides would scavenge Cu and other key-metals we suggest that the occurrence of arc-associated porphyry deposits is governed less by the capacity of oxidised magmas to remain Cu-rich, and more by the efficiency of Cl-rich fluids at remobilising metals from stable crustal sulfides.