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Solubility of Cu, Ag and Au in Magmatic Sulfur-bearing Fluids as a Function of Oxygen Fugacity

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Magma-derived fluids containing chlorine and sulfur are critical for the transport of ore metals to porphyry ore-forming environments. The effect of redox conditions on ore metal speciation and solubilities in sulfur-bearing fluids required for efficient exploration models have not yet been experimentally constrained. We performed experiments to determine the effect of oxygen fugacity (fO_2) on the solubility of Cu, Ag and Au in high-temperature, low- density, low-salinity fluids and hypersaline brine. The experiments were conducted at $T=900^\circ\text{C}$, $P=2000$ bar varying fO_2 in 7 steps between NNO - 0.5 to NNO + 2.5. The apparent solubility of Cu and Ag at the imposed metal activities increases by a factor of 7 in the $\text{H}_2\text{O-NaCl-KCl-S}$ low-salinity fluid with fO_2 increasing from NNO - 0.5 to NNO + 2.5. The addition of 0.198 m HCl increases the overall solubility of Cu by a factor of 1.2 – 2.4. The apparent solubility of Au decreases by a factor of 9 as fO_2 changes from NNO - 0.5 to NNO + 2.5. The relationship between the logarithms of the apparent Cu and Ag solubilities and fO_2 is linear and the slope of the fitted line corresponds to 1+ oxidation state of these metals indicating that they are dominantly complexed by ligands that are S-free (e.g., chloride). Considering previous studies on silicate melts and our experimental data for volatiles, Cu and Ag likely have constant fluid/melt partition coefficients in the typical fO_2 range of arc magmatism. Our experiments also show that Cu extraction would be less efficient in S-rich systems at intermediate fO_2 . But, the direct exsolution of hypersaline fluids from silicate melts at oxidizing conditions may be a particularly efficient way of Cu sequestration from crystallizing upper crustal magma bodies provided that these brines can be physically mobilized and transferred to the hydrothermal system.