

Sulphur species and gold transport agents in magmatic fluids unveiled

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Magmatic fluids exsolving from arc-related calc-alkaline magmas play an important role in the transport of volatile species and metals toward the Earth's surface and the formation of magmatic-hydrothermal ore deposits, including giant porphyry Cu-Mo-Au deposits and epithermal Au-Ag-Cu deposits. It is now well-established that chlorine- and sulphur-bearing complexes transport much of the metal content in high-temperature magmatic fluids. Which exact sulphur species transport metals in these fluids, however, remains highly debated.

To study the speciation of sulphur at a range of temperature and redox conditions characteristic of shallow magmatic degassing and to identify which sulphur species may be responsible for the transport of gold, we applied a three-step procedure. In step one, we trapped pre-equilibrated gold-saturated sulphur-bearing aqueous fluids (initial composition: $\text{H}_2\text{O} + 1 \text{ mol NaCl/kg H}_2\text{O} + 5 \text{ mol\% H}_2\text{SO}_4$) as synthetic fluid inclusions (SFI) in quartz at 200 MPa and 750-1000°C using a prototype externally heated molybdenum-hafnium carbide vessel apparatus. In step two, we reheated the SFI to their entrapment temperature in a microscope heating stage and analysed them by *in situ* Raman spectroscopy with carefully chosen laser wavelengths to prevent the signal enhancement of sulphur radical species by Raman resonance. Finally, we employed LA-ICP-MS to determine the concentration of gold in the SFI, and thus constrain gold solubility in the experimental fluids.

Our experimental design allowed for the quantification of different sulphur-bearing aqueous species, including sulphide, SO_2 and sulphur radical species, at previously inaccessible temperature and oxygen fugacity conditions [1]. We show that sulphur radicals are stable at a wide range of temperature and redox conditions; however, their concentration remains below the detection limit in their respective non-resonant Raman spectra. The measured gold concentrations are generally very high and show a remarkable ~ 1 log unit increase with decreasing oxygen fugacity from $129 \pm 52 \text{ }\mu\text{g/g}$ at NNO+2.6 to $1,420 \pm 360 \text{ }\mu\text{g/g}$ at NNO-0.9. Our observations suggest that HS^- rather than sulphur radical species are the primary carriers of gold in magmatic fluids.

[1] Farsang & Zajacz (2025), *Nature Geoscience* 18, 98-104.