

Sulfur radical species form gold deposits on Earth

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Current models of the formation and distribution of gold deposits on Earth are based on the long-standing paradigm that hydrogen sulfide (H₂S) controls gold mobilization and precipitation by fluids across the lithosphere [1] [2]. However, this paradigm may be challenged by recent findings of sulfur radical forms such as the trisulfur (S₃⁻) and disulfur (S₂⁻) ions, which are stable in aqueous solution at elevated temperatures and pressures [3] [4]. To provide a quantitative understanding of the role of these S forms on gold behavior, we combined in situ X-ray absorption spectroscopy and solubility measurements with ab-initio molecular dynamics and thermodynamic simulations on aqueous Au-S-bearing solutions representative of ore-forming hydrothermal fluids.

The results demonstrate the formation of soluble complexes between S₃⁻ and Au⁺ and allow their stability across a wide temperature-pressure range of crustal fluids to be quantified. These species enable extraction, transport, and focused precipitation of gold by S-rich fluids 10-100 times more efficiently than hydrogen sulfide only. As a result, S₃⁻ provides a major control on the source, concentration and distribution of gold and associated metals in economic deposits, such as magmatic-related porphyry-epithermal Cu-Au(-Mo), sedimentary rock-hosted Carlin, and metamorphic orogenic Au deposits. The growth and decay of S₃⁻ (and potentially S₂⁻) during the hydrothermal fluid generation and evolution may thus be a key factor that determines the fate of gold in the lithosphere.

[1] Seward (1973), *GCA* **37**, 379-399. [2] Pokrovski *et al.* (2014), *Geol. Soc. London Spec. Publ.* **402**, 9-70. [3] Pokrovski & Dubrovinsky (2011), *Science* **331**, 1052-1054. [4] Pokrovski & Dubessy (2015), *EPSL* **411**, 298-309.