

Speciation and solubility of gold in CO₂-HCl-H₂O fluids: MD simulations and solubility experiments

WEIHUA LIU¹, YUAN MEI^{1,2}, JOËL BRUGGER²,
ARTASHE MIGDISOV³ AND ANTHONY WILLIAMS-JONES³

¹CSIRO Mineral Resources Flagship, Melbourne,
weihua.liu@csiro.au

²Monash University, Melbourne, Australia

³McGill University, Montreal, Canada

Increasing evidence of the abundance of water-poor, CO₂-rich fluids in orogenic gold deposits indicates that 'exotic' volatile-rich fluids may have played a role in their formation (e.g., Ashanti Belt, Ghana [1]; Red Lake, Canada [2]; Sunrise Dam, Australia [3]). In water-rich, CO₂-bearing fluids, CO₂ has been suggested to be a pH buffering agent, optimising gold solubility as gold-hydrosulphide complexes [4]. However, the role of CO₂ for gold mobility in water-poor, CO₂-rich supercritical fluids is yet to be explored.

We have conducted preliminary gold solubility experiments and *ab initio* molecular dynamics simulations to investigate this problem. The solubility experiments have been conducted in a titanium autoclave at 340 °C with 0.01 m HCl, and the amount of water and CO₂ were loaded to the cell to ensure the fluid density is in the vapour phase with CO₂ mole fraction (CO₂/(H₂O+CO₂)) ranging from 0.1 to 0.84. The results showed that gold solubility has a negative correlation to the CO₂ content in the fluids, i.e., decreasing with decreasing water fugacity.

Ab initio MD simulations of the AuCl⁰ complex (linear [H₂O-Au-Cl]⁰) in the CO₂-H₂O system were conducted at 340 °C with CO₂ mole fraction from 0.1-0.98 at densities between 0.78-0.15 g/cm³. The MD simulations indicate that the number of hydration water and H-bonds near the AuCl⁰ complex decreases systematically with increasing CO₂ mole fraction. These results are consistent with the experiments, suggesting that H₂O as a polarized molecule plays a more active role than the un-polarized CO₂ molecule in the fluids, and hydrated chloride species are the main form for transporting gold in the CO₂-H₂O-HCl system.

[1] Schmdit Mum et al., 1997, *Min. Dep.* **32**:107-118 [2] Chi et al., 2006, *Min. Dep.* **40**:726-741 [3] Baker et al., 2010, *Econ. Geol.* **105**:873-894 [4] Philips and Evans, 2004, *Nature*, **429**:860-863