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Platinum speciation and transport in sulfur-rich hydrothermal fluids

C. LASKAR[1*], G.S. POKROVSKI[1], M. KOKH[1], J.L. HAZEMANN[2], E.F. BAZARKINA[2], E. DESMAELE[3] AND R. VUILLEUMIER[3]

[1]Géosciences Environnement Toulouse (GET), UPS, CNRS, IRD, OMP, 31400, Toulouse, France
(*correspondence : clement.laskar@get.omp.eu)
[2]Institut Néel, UGA, CNRS, 38042, Grenoble, France
[3]Ecole Normale Supérieure (ENS), UPMC, CNRS, Sorbonne Universités, 75005, Paris, France

Understanding PGE deposit formation requires knowledge of the chemical state and mobility of platinum group elements (PGE) in magmatic-hydrothermal fluids. Yet, existing data on aqueous chloride, sulfate, and hydroxide complexes of PGE predict far too small metal contents (<ppt to ppb) in fluids from most geological settings, thus calling upon an important role of sulfide complexes [1-3], and, potentially, trisulfur ion ligands in PGE transport [4]. In an attempt to quantify the effect of sulfur on PGE mobility, here we combined solubility measurements, in-situ X-ray absorption spectroscopy (XAS), thermodynamic (TD) and molecular dynamics (MD) modeling, based on recent advances of our knowledge of sulfur speciation in crustal fluids [5]. Our new data at 300°C and 500 bar suggest formation of two main complexes transporting Pt in the fluid at concentrations of 10s ppm over a wide pH range (4-8): $Pt(HS)_4^{2-}$ in hydrogen sulfide H_2S/HS^- solutions, and $Pt(HS)_2(S_3)_2^{2-}$ in sulfide-sulfate $H_2S/SO_4^{2-}/S_3^{\bullet -}$ solutions. Notably, the obtained concentrations of $Pt(HS)_4^{2-}$ in H_2S/HS^- solutions, using an improved protocol for accurate fluid sampling from a flexible-cell reactor, are 1000 times higher than those reported in previous quench-based studies [6]. TD and MD simulations, based on combined solubility and in-situ XAS data confirm the large stability of the $Pt(HS)_2(S_3)_2^{2-}$ complex, analogous to that proposed for gold [4], with Pt(II) as the major oxidation state. Thus, our new data highlight an important role of previously ignored sulfur complexes that significantly enhance PGE transport by ore-forming fluids in the Earth's crust.

[1] Bazarkina *et al.* (2014) *GCA* **146**, 107–131. [2] Kokh *et al.* (2017) *GCA* **197**, 433–466. [3] Tagirov *et al.* (2019) *GCA* **254**, 86-101. [4] Pokrovski *et al.* (2015) *PNAS* **112**, 13484–13489. [5] Pokrovski and Dubessy (2015) *EPSL* **411**, 298-309. [6] Gammons and Bloom (1993) *GCA* **57**, 2451–2467.