The solubility and speciation of thorium in chloride-bearing aqueous solutions at elevated temperatures

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The presence of Th and its intrinsic radioactivity is a critical issue in ore deposits, particularly with the extraction of the rare earth elements (REE). Two of the primary REE ore minerals, monazite and xenotime, commonly have high Th contents, and this has led to significant increases in REE recovery costs and the production of undesirable radioactive waste [1]. As modern exploration is based on genetic models, an understanding of the factors controlling the transport and deposition of both Th and the REE is required. While data on the REE are reasonably available [2], progress in determining geochemical parameters for thorium under hydrothermal conditions has been minimal; with experimental studies of thorium solubility and speciation largely restricted to ambient temperature. In this study, we determined the solubility of ThO_2 at elevated temperatures (175-250 °C) with variable chloride concentration (0.25-2.5 m [mol/Kg] NaCl), and variable pH (1.6-3.4).

The experiments in our study employed the autoclave solubility method, developed to determine the solubility of metals in ligand-bearing aqueous solutions [3]. Under the conditions of our experiments, the amount of Th dissolved in the aqueous solution was determined to be independent of the NaCl concentration, and rather conrolled by the hydrolysis and formation of the complexes, $Th(OH)_2^{2+}$ and $Th(OH)_4^0$. Thus, unlike the REE, for which chloride complexation is important in hydrothermal mobilization, Th complexation with chloride is negligeable. We propose the concentrations of Th observed in our experiments likely represent a minimum solubility level that can be developed in natural hydrothermal systems in equilibrium with Th-bearing minerals.

[1] Findeiß and Schäffer (2017) *J. Sustain. Metall.* **3**, 179–189 [2] Migdisov et al. (2016) *Chem. Geol.* **439**, 13–42. [3] Migdisov et al. (2009) *Geochim. Cosmochim. Acta* **73**, 7087-7109.