Transporting uranium in acidic brines under reducing conditions

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Although uranium has two common oxidation states, U^{4+} and U^{6+}, it is only as U^{6+} that it is believed to be mobile in hydrothermal solutions. Models developed to explain the mobility of uranium in nuclear waste repositories, nuclear reactors, and unconformity-type ore deposits, one of the world's main sources of uranium, rely heavily upon this belief. This may be justified for ambient temperature for which there is a considerable body of supporting data. It is less justified for high temperature (>100 °C), however, because of the paucity of data. In this study, we determined the solubility of uranium oxides in aqueous solutions at high temperature (200-350 °C), and variable chloride activity ($\sim 10^{-2}$-$10^{-0.5}$ aCl^-), and oxygen fugacity (MoO_2-MoO_3 and Ni-NiO buffers for oxidizing and reducing conditions, respectively).

Our experiments were performed using the autoclave solubility method, which has been developed to determine the solubility of metals in ligand-bearing aqueous solutions [1]. The accuracy of the data was verified by comparing results obtained for both UO_2 and U_3O_8.

The results of the above experiments clearly demonstrate that reducing conditions do not necessarily impede uranium dissolution. Increasing chloride activity resulted in a corresponding increase in the amount of uranium dissolved in the aqueous solution. Under reducing conditions, the species UCl_4^0 was favored whereas at oxidizing conditions, $UO_2Cl_2^0$ dominated. Surprisingly, at high chloride activity, the solubility of uranium under reducing conditions exceeded that at oxidizing conditions for the same uranium oxide. These results underscore the need for a re-evaluation of the conditions and nature of mobility of uranium in settings of interest to industry, government and academia. Regardless of the scenario, reducing conditions can no longer be considered a guarantee of uranium immobility.

[1] Migdisov *et al.* (2009) *Geochimica et Cosmochimica Acta* **73**, 7087-7109.