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Neptunium (V) Solubility in the Presence and Absence of WIPP Relevant Ligands

UGRAS KAPLAN[1], ADRIANNE ELICE NAVARRETTE[1], CEREN ASLANI[2], JANDI LEE POTTER[2] AND JONATHAN PAUL ICENHOWER[1]

[1]Los Alamos National Laboratory
[2]Los Alamos National Lab.

Presenting Author: ukaplan@lanl.gov

Neptunium (V) Solubility in the Presence and Absence of WIPP Relevant Ligands

Kaplan U, Navarrette A. E., Icenhower, J. P.

Actinide Chemistry and Repository Science Program (ACRSP) / Los Alamos National Laboratory (LANL)-Carlsbad Operations / NM-USA

Email: ukaplan@lanl.gov

The aqueous chemistry of Np(V) in 5 M NaCl and synthetic WIPP brines in high ionic strength (5.3-7.4 M) as a function of pC_{H+} in presence and absence of borate, WIPP relevant ligands (EDTA, oxalate, citrate, acetate), and carbonate at = 23 ± 2 °C was thoroughly studied by long-term batch solubility experiments (approximately 330 days) from an undersaturation approach. Applying a comprehensive set of experimental and spectroscopic techniques, including UV-VIS-NIR and Np L_{III}-edge X-ray Absorption Spectroscopy (XAS), the solubility controlling Np(V) solid phases and the predominant aqueous Np(V) species were identified in all studied samples. Also, the impact of borate on the aqueous speciation and, especially, on the solubility of Np(V) in 5 M NaCl was confirmed. The interactions of organic ligands with Np(V) differ from each other in 5 M NaCl and in synthetic WIPP solution. The interactions of organics in the WIPP brine vary depending on kinetics, and it has been observed that this effect disappears over time, whereas their interactions in 5 M NaCl are more stable. The effect of carbonate on Np(V) solubility is mostly seen at pC_{H+} 9 and 11. Model calculation showed that the total soluble Np species at pC_{H+} 9.5 was 4.02 x 10^{-7} M under full repository conditions; a similar Np concentration of 5.82 x 10^{-7}M was found in the present work. Also, solid phase characterization by EXAFS indicates that the redox controlling solid phase consists of $KNpO_2CO_{3(s)}$ or $K_3NpO_2(CO_3)_2$ and this result confirms the WIPP model prediction. The data obtained from this work quantifies the effects of WIPP-relevant concentrations of organics/borate/carbonate on the solubility of Np(V) to confirm the predictions of the WIPP actinide model and inform decisions and recommendations made in the upcoming recertification of the WIPP.