Goldschmidt 2022 Abstract
https://doi.org/10.46427/gold2022.11439

Uptake of uranium by crystallization of phosphate minerals.

RINAT GABITOV[1], ANGEL JIMENEZ[1], ARTAS A. MIGDISOV[2], JUEJING LIU[3], XIAOFENG GUO[3], KAT ROSE[1], ALBERTO PEREZ-HUERTA[4], VARUN PAUL[1], PADMANAVA DASH[1], CHANAKA NAVARATHNA[1], TODD MLSNA[1], FLORIE CAPORUSCIO[2], HONGWU XU[2] AND ROBERT ROBACK[2]

[1]Mississippi State University
[2]Los Alamos National Laboratory
[3]Washington State University
[4]The University of Alabama

Presenting Author: rinat.gabitov@gmail.com

Aiming at the development of a new engineered backfill material, we have investigated the uptake of uranium by apatite at a range of temperatures similar to those expected at waste repository sites, where thermal peak of the waste package (heated by radioactive decay) is ~300°C (Greenburg and Wen 2013). Experiments were conducted in plastic containers or autoclaves at saturated water pressure and 40-350°C, where metastable phosphate (brushite) was transformed to apatite/monetite in NaCl solutions. Uranium was introduced as aliquots or as a solid oxide at oxidized and reduced conditions. The oxidation state of dissolved uranium (U^{4+} or U^{6+}) was controlled by addition of solid redox buffers into autoclaves at 250-350°C.

X-ray diffraction (XRD) and backscattered electron diffraction (EBSD) of crystalline products allowed evaluation of crystal structures. Uranium content in crystals and its concentration in the solutions were evaluated with inductively coupled plasma mass spectrometry (ICP-MS) and laser ablation ICP-MS, which allowed the calculation of uranium partition coefficient ($D=U_{calcite}/U_{fluid}$) between crystal and fluid phases.

Overall, our results showed:

1) Crystal structure of the final phosphate phase (monetite or hydroxyapatite) depends on pH of the solution;

2) In experiments with uranium aliquot, apatite crystallization extracted more than 95% of dissolved uranyl at pH=3-4;

3) Uranium uptake by phosphates was not as redox sensitive as was shown for calcite, where U^{4+} was more compatible with calcite by a few orders of magnitude compared to U^{6+} (Gabitov et al. 2021).

Acknowledgements: DOE NEUP.

References

Greenburg H.R. and Wen J. (2013) LLNL-TR639869-DRAFT, 38.

Gabitov R.I., Migdissov A., Nguyen A., Van Hartesveldt N., Perez-Huerta A., Sadekov A., Sauer K.B., Baker J., Paul V., Caporuscio F., Xu H., and Roback R.C. (2021) Chem. Geol. 564, 120054.