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Mobilization and agglomeration of uraninite nanoparticles: A nano-mineralogical study of samples from the Matoush Uranium ore deposit

MICHAEL SCHINDLER^{1,*}, AARON J. LUSSIER², JACOB BELLROSE¹, SERGEI ROUVIMOV²,
PETER C. BURNS^{2,3}, AND T. KURT KYSER⁴

¹Department of Earth Sciences, Laurentian University, Sudbury, Ontario P3E 2C6, Canada

²Department of Civil and Environmental Engineering and Earth Sciences, University of Notre Dame, Notre Dame, Indiana 46556, U.S.A.

³Department of Chemistry and Biochemistry, University of Notre Dame, Notre Dame, Indiana 46556, U.S.A.

⁴Department of Geological Sciences and Geological Engineering, Queen's University, Kingston, Ontario K7L 3N6, Canada

ABSTRACT

The occurrence of uraninite nanoparticles in the alteration zones of uranium ore deposits suggests potential mobilization of U(IV) under reducing conditions, which is important for understanding the mobility of uranium in contaminated sites and potential repositories for nuclear waste. This study investigates the occurrence of uraninite nanoparticles in the outer alteration zone of the Matoush uranium ore deposit, Quebec, Canada. Selected samples with finely disseminated uraninite from the outer alteration zone of the deposit are examined by X-ray fluorescence spectroscopy, scanning electron microscopy, and high-resolution transmission electron microscopy on specimens prepared using the focused ion beam milling technique. Uraninite nanoparticles occur as single particles, in clusters, and in larger aggregates in close association with the Cr-rich phases chromceladonite (Cr-rich mica), ideally $\text{KMgCr}^{3+}(\text{Si}_4\text{O}_{10})(\text{OH})_2$, eskolaite, ideally Cr_2^+O_3 , bracewellite, Cr^{3+}OOH , and an amorphous Cr-rich oxide matrix as well as with fluorapatite and galena. Nanoparticles on the surface and in the outer rim of single uraninite crystals indicate the growth of larger uraninite crystals via crystallization through particle attachment and Oswald ripening. The flow texture of the uraninite nanoparticles in the amorphous Cr-rich oxide matrix, their aggregation on the surface of nanocrystals of bracewellite, the absence of products of a redox reaction involving U(VI) and Fe(II), and the occurrence of amorphous Fe-depleted alteration layers between uraninite and eskolaite, and uraninite and Cr-rich mica indicate that the uraninite nanoparticles have been mobilized under reducing conditions (leaching of Fe^{2+} from the alteration layer) at low T (amorphous character of the alteration layer) after the main mineralization event from the center of the mineralization to the outer parts of the Matoush dike complex. These results indicate that fluids can mobilize U(IV) under reducing conditions in the form of uraninite nanoparticles albeit over limited distances. The potential mobilization of these nanoparticles may also explain the occurrence of proximal mineralized zones in U-ore deposits that lack common products resulting from the reduction of U(VI) by Fe^{2+} (e.g., hematite and other Fe^{3+} -phases).

Keywords: Uraninite nanoparticles, uranium mobilization, uranium ore deposit, natural analog, spent nuclear fuel repository, alteration