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

MICHAEL SCHINDLER1,*, AARON J. LUSSIERS1, JACOB BELLROSE1, SERGEI ROUVIMOV2, PETER C. BURNS2,3, AND T. KURT KYSER4

1Department of Earth Sciences, Laurentian University, Sudbury, Ontario P3E 2C6, Canada
2Department of Chemistry and Biochemistry, University of Notre Dame, Notre Dame, Indiana 46556, U.S.A.
3Department of Civil and Environmental Engineering and Earth Sciences, 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 C-rich phases chromeladonite (C-rich mica), ideally KMgCr2(Si4O10)(OH)2, eskolaite, ideally Cr2O3, bracewellite, Cr3+OOH, and an amorphous C-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 C-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 Fe3+ 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 Fe2+ (e.g., hematite and other Fe3+-phases).

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

INTRODUCTION

There is increasing interest in the occurrence of colloidal and crystalline nanoscale materials in the environment, as nanoparticles can transport metal(loid)s over significant distances and effectively sequester them in contaminated sites as well as ore deposits (Vilks et al. 1993; Kretzschmar and Schäfer 2005; Hochella et al. 2008; Weber et al. 2009). Colloids and crystalline nanoparticles are, in this regard, environmentally relevant for remediation strategies of U-contaminated sites and the design of nuclear waste repositories as they can transport and sequester radionuclides (Traexler and Ewing 2002; Bargar et al. 2008; Dreissig et al. 2011; Schmid et al. 2012, 2013; Bots et al. 2014; Suzuki et al. 2015). Uranium(IV) is typically thought to be immobile under reducing conditions, and that sequestration and containment of U and other radionuclides occurs under reducing conditions (Long and Ewing 2004). Uranium (IV)-bearing nanoparticles such as uraninite, coffinite, and U-bearing silicates have been, however, identified in numerous studies (e.g., Bargar et al. 2008; Dreissig et al. 2011, Wang et al. 2013; Fuchs et al. 2015; Riegler et al. 2015; Suzuki et al. 2015), suggesting that the mobility of U(IV) may be significantly enhanced by the presence of colloidal material under reducing conditions.

Uranium ore deposits have been examined as natural analogs of nuclear waste repositories as their alteration halos provides valuable information on the mobilization and retardation of U and other radionuclides over millions of years (Pearcy et al. 1994; Fayek et al. 2003; Schindler et al. 2010). Recent nano- to microscale studies of finely disseminated uraninite in sandstone-hosted roll-front uranium deposits in northwest China (Min et al. 2005), organic matter at the Carbon Leader Reef, Witwatersrand...