The mineralogical factor in the assessment of repository conditions - Inferences from natural analogues

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Retardation processes that may affect radionuclide transport in geological media include adsorption, ion-exchange, matrix diffusion, precipitation and mineralization, and they are therefore beneficial for repository safety. Both irreversible sorption and mineral precipitation are important to long-term predictions of repository safety because they can immobilise radionuclides efficiently. However, they are difficult to demonstrate.

Studies of element transport and retardation in natural systems such as U orebodies can provide convincing evidence for such mechanisms. Investigations on the Cigar Lake deposit (Canada) and the natural nuclear fission reactors of Gabon were carried out to evaluate the mobility of performance assessment-relevant elements in response to the alteration and to identify the mechanisms which could retard the migration of radionuclides. Both deposits display hydrothermal alteration of U-bearing sandstones and the formation of a clay halo around the ore. This clay zone provides effective redox buffering and played a significant role in the long-term preservation of the U ore. Natural uraninite analogous to spent fuel UO2 retained most of the radionuclides produced by the nuclear reactions with the exception of noble gases, alkaline and alkaline earths. High amounts of Ca, Sr and REE were coprecipitated by fluoroapatite and Al-phosphates in the clay halo. Clays (illite, chlorite) and Ti-oxide were found to have sorbed significant amounts of U. Sorption onto mineral surfaces was followed by the formation of coffinite (USiO4 nH2O) that displays variable chemical composition and multiple substitutions. Alteration of uraninite under reducing conditions lead to the precipitation of newly formed Si-P-S- and Y-REE-Th-Zr-U-bearing minerals in the form of polyphased and chemically complex coffinite.

The formation of secondary U-silicates and phosphates in clayey environments is an important factor because these minerals 1) are ubiquitous and they form at low Eh and at various temperatures (50-300 degrees), and 2) represent effective traps for several critical radionuclides including U, Th, Pu, Np, Am and Cm, therefore retarding their mobility in the near-field of a repository.