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Mats JONSSON
Department of Chemistry, KTH Royal Institute of Technology, Sweden

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« Radiation induced corrosion of uranium based nuclear fuel materials »

UO_2 is the most commonly used commercial nuclear fuel material. For this reason, the chemistry of UO_2 has been studied extensively for many decades. In this presentation, mechanistic aspects of radiation induced dissolution of UO_2 under conditions relevant for a geological repository for used nuclear fuel will be discussed. Particular focus will be put on the reactivity of H_2O_2 towards UO_2 since H_2O_2 has been shown to be the most important radiolytic oxidant under the expected repository conditions. Several studies have shown that H_2O_2 can undergo catalytic decomposition on oxide surfaces forming surface-bound hydroxyl radicals as an intermediate. On UO_2, this intermediate can react in two different ways: (1) oxidation of the surface and (2) reaction with H_2O_2. In other words, the two competing reactions have a common intermediate. In a system containing HCO_3^-/CO_3^{2-}, U(VI) will be readily dissolved once it is formed. This enables the formation of uranyl-peroxo-carbonato complexes in solution which, at higher uranyl concentrations, can dominate the peroxide speciation. These negatively charged complexes display a completely different reactivity towards UO_2 as well as other oxide surfaces and thereby change system quite dramatically. The impact of these complexes will be discussed in detail as well as some of the features of catalytic H_2O_2 decomposition on oxide surfaces.
In addition to the numerous studies performed on UO_2, similar studies of alternative fuel materials have been conducted in recent years. Here we will focus on UN, UC and U_3Si_2 and discuss the mechanism and kinetics of radiation induced dissolution of these materials.