How ionizing radiation affects the chemical stability of spent nuclear fuel under deep geological repository conditions

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In Sweden, Finland and many other countries, the highly radiotoxic spent nuclear fuel generated when operating nuclear reactors will be placed in deep geological repositories to prevent contact with the biosphere for extremely long time periods. The deep geological repositories have several natural and engineered barriers to prevent groundwater from reaching the fuel surface and to prevent radionuclide migration from the fuel surface. The decision to build and take a repository into use rests on a number of safety assessments where numerous foreseeable processes must be accounted for. A key-process in this is the dissolution of the spent nuclear fuel and the subsequent release of radionuclides into the groundwater.

Spent nuclear fuel from most commercial nuclear reactors is mainly UO_2 with heavier actinides and fission products in low concentrations. UO_2 is fairly stable under the reducing conditions that often prevail in potential repository sites. However, the inherent radioactivity of the fuel (originating from the heavier actinides and the fission products) changes the redox conditions due to radiolytic decomposition of water. Among the radiolysis products, both strong oxidants and reductants can be found. Initially, the oxidizing radiolysis products will dominate the surface chemistry for kinetic reasons. It has been demonstrated that the most important radiolytic oxidant is H_2O_2. Interestingly, the reaction between H_2O_2 and UO_2 involves the formation of surface-bound hydroxyl radicals.

In this presentation, the mechanism and kinetics of radiation-induced dissolution of UO_2-based spent nuclear fuel will be discussed in view of recent findings. In addition, the mechanism and impact of reactions counteracting the dissolution process will be discussed in order to give a comprehensive picture of the intriguing interfacial radiation chemistry of spent nuclear in contact with water.