

Reactivity Change of UO₂-Based Nuclear Fuel Upon Exposure To Radiation In Groundwater

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Abstract

Several countries that utilize nuclear power have decided to place their used nuclear fuel in geological repositories in order to isolate this highly radiotoxic material from the biosphere for 10^5 – 10^6 years, i.e., until the radiotoxicity has declined to acceptably low levels. The existing repository concepts are all based on multiple barriers designed to prevent the intrusion of groundwater and migration of radionuclides. Political decisions to build a geological repository and to take it into use rely on very thorough safety assessments. One of the scenarios in such assessment is a complete barrier failure where groundwater comes into contact with the spent nuclear fuel. The fuel matrix of the majority of the existing commercial nuclear fuels is UO₂. After use in the reactor, the spent nuclear is still mainly composed of UO₂ (> 95 %) and the majority of the fission products and neutron activation products are incorporated into the UO₂-matrix. These radiotoxic species are assumed to dissolve congruently with the UO₂-matrix in groundwater. Hence, the stability of the UO₂-matrix in groundwater is a key-feature of the safety assessment. In general, UO₂ has very low solubility in the reducing groundwater expected at most potential repository sites. However, the inherent radioactivity of the spent nuclear fuel will lead to radiolysis of the groundwater in contact with the spent nuclear fuel. This will alter the redox conditions and considerably enhance the fuel matrix solubility. It has been shown that the radiolytic oxidant that dominates the oxidative dissolution process is H₂O₂.¹ For this reason, there are numerous studies focusing on the reactivity of H₂O₂ have been conducted. Very recently, it was demonstrated that the redox reactivity (and thereby the dissolution) of UO₂ towards H₂O₂ is significantly reduced with increasing exposure to H₂O₂.² This was quite unexpected considering that HCO₃[−] present in the groundwater (and in the lab experiments) was expected to efficiently dissolve U(VI) from the oxidized UO₂-surface. As a consequence, the expected rate of matrix dissolution and thereby also the risk of radionuclide migration from the repository need to be revised. As H₂O₂ is not the only radiolytic oxidant capable of oxidizing UO₂ under repository conditions, we have performed experimental studies in order to assess the possible change in UO₂ reactivity upon exposure to O₂ and CO₃[−] (since groundwater normally contains HCO₃[−]). The results will be discussed in view of the nature of the oxidants and the experimentally observed alterations of the UO₂-matrix.

References

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