DISSOLUTION OF UNIRRADIATED MOX FUEL IN THE PRESENCE OF METALLIC IRON

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In all European deep repository concepts for spent nuclear fuel, massive canisters made of or containing large amounts of iron are designed to prevent any early spent fuel contact with groundwater. In the case of canister failure, the corrosion of spent nuclear fuel will occur at repository depth, several hundred meters underground, where the groundwaters are typically anoxic. Spent nuclear fuel corrosion is therefore expected to occur simultaneously with anoxic iron corrosion. In this study the corrosion of an unirradiated MOX fuel pellet containing 10% Pu (specific alpha activity 1.7 GBq/g) was investigated under Ar atmosphere in carbonated water (2 mM $NaHCO_3$, 10 mM NaCl) and in simulated granitic groundwater from Forsmark, Sweden in the presence of metallic iron foils and iron powder. The anoxic conditions were simulated by degassing the solutions and by carrying out the tests in an autoclave pressurized with argon. The results of the test show that the anoxic corrosion products of iron neutralized completely the high alpha field of the MOX pellet. The production of hydrogen continued during the whole test with a linear rate of 8.5 10^{-5} mol H_2/day, reaching about 2 bars H_2 pressure at test termination. The Fe(II) concentrations increased from 5.6 10^{-4} M after 101 days and MOX pellet insertion to 9.2 10^{-4} M at test termination. No traces of oxidized uranium were observed in the autoclave during the whole duration (407 days) of the test; the uranium concentrations (from 2.9 10^{-9} M at start to 1.2 10^{-9} at the end) are in good agreement with the lower range of UO_2(am) solubility. Massive Fe(III) precipitation was observed on the surface of the MOX pellet, but not in the bulk solution. The Pu concentrations were below the detection limit of alpha spectrometry. We have reported very low Pu concentrations only at the initial stage of the leaching as determined by ICP-MS, later Pu was under detection limit. The analysis of the iron foils, iron powder and vessel surfaces showed that the majority of U(IV) was sorbed on iron corrosion products and much less in the glass surfaces of the vessel. Anoxic iron corrosion products such as ankerite and probably chukanovite were detected on the iron foils while several Fe(III) compounds including goethite, lepidocrocite and akageneite were formed on the surface of the MOX pellet. This study shows that the anoxic corrosion of iron in granitic Forsmark groundwaters completely inhibits the oxidative dissolution of a MOX pellet with a very high specific alpha activity. The findings emphasize the critical role of iron corrosion and its corrosion products in mitigating actinide solubility and mobility under repository conditions, thereby enhancing the stability of spent nuclear fuel under repository conditions.

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