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**Resume :** The assessment of the radiolytically induced gas generation during plutonium bearing materials handling is useful for transportation and storage risk management. The most stable phase for plutonium handling is PuO<sub>2</sub>. The H<sub>2</sub> generation by radiolysis of the water adsorbed on PuO<sub>2</sub> appears to be surface related since different surface states imply different kinetics. The inverse gas chromatography (IGC) is an analytic method dedicated to surface analysis which is based on the surface energy measurement. Roughly, this method consists in making chromatographic column filled with the material under study and then to elute specific probe molecules to track the surface evolution. IGC enables also the construction of adsorption isotherm by coupling a humid gas generator to the IGC apparatus. A preliminary study has been performed on CeO<sub>2</sub> (a non-radioactive material classically used as a surrogate for PuO<sub>2</sub>) and shows interesting results on the surface modification during storage under humid conditions. The literature shows no evidence of reactivity of ceria toward H<sub>2</sub>O. However the results suggest that the oxide undergoes some alteration by the adsorption of several monolayers of H<sub>2</sub>O. A thermal treatment performed following the "humid storage" seems to enable to restore at least partially the initial feature of the surface. These results support the hypothesis that the different H<sub>2</sub> generation kinetics on PuO<sub>2</sub> may be related to a modification of the surface induced by water adsorption.

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#### **Corrosion of spent nuclear fuel segment in presence of Fe (II)/Fe(III) oxide**

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**Resume :** The actinides source term for a spent nuclear fuel (SNF) final disposal system depends mainly on the SNF alteration and actinides interactions with Fe corrosion products of breached canisters. To study the fate of actinides released from SNF in presence of Fe oxide, a long-term corrosion experiment of 3562 days was performed by immersing a cladded SNF segment into 5 M NaCl solution in presence of Fe(II)/Fe(III) oxide powder. The powder consists of 80 wt.% Fe<sub>3</sub>O<sub>4</sub> and 20 wt.% Fe<sub>2</sub>O<sub>3</sub>, present as magnetite grains covered with layers of maghemite and hematite. The experiment was carried out in a glass vessel under Ar atmosphere at near neutral pH and room temperature. Solution and gas samplings were done at specific time intervals. The presence of CO<sub>2</sub> in the gas phase was detected and increased through the whole experimental duration. This phenomenon is related to initial C impurities present in the Fe oxide powder. In parallel, aqueous concentrations of actinides continuously increased over the entire observation period (e.g.: [U] measured at the end of experiment was 10<sup>-4</sup>M). The SNF sample and Fe oxide powder were characterised prior to and after the experiment. Radiochemical analyses and XPS showed that a significant amount of the actinides released from the SNF was retained onto the Fe oxide. U sorbed onto the Fe oxide was found to be in the hexavalent state.

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#### **Immobilization of actinides in the cementitious near field of a repository for radioactive waste**

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**Resume :** Cement stabilization of radioactive waste is a common solidification-stabilization (S/S) technique used prior to near-surface or deep geological disposal. Furthermore, current disposal concepts for low- and short-lived intermediate level radioactive waste foresee the use of cementitious materials for the construction of the engineered barrier system (container, backfill and liners). In the Swiss concept for a cement-based deep geological repository, cementitious materials are considered to control the release rate of radionuclides from the near field into the far field. The cementitious near field undergoes chemical alteration processes caused by the interaction of groundwater infiltrating from the host rock with hardened cement paste (HCP) of the near field. Hence, the mineral composition of HCP

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