

Corrosion Behaviour of AGR Simulated Fuels – Evolution of the Fuel Surface

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The vast proportion of spent fuel in the UK comes from Advanced Gas-Cooled Reactors (AGR). Current UK nuclear waste policy is that spent AGR fuel is to be contained in steel canisters and disposed of directly via burial in a geological repository. It is expected that after 1000 years, once the thermal pulse has occurred, that the steel canisters in which spent AGR fuel pellets are stored in will come into contact with groundwater, corrode, and subsequently fail. The failure of these steel canisters will lead to the corrosion of the spent uranium dioxide fuel inside the canister under environmental conditions. Alpha radiation is the only type of radiation that is expected to prevail after the thermal pulse. However, when groundwater comes into contact with alpha radiation produced by UO₂ fuel pellets, alpha radiolysis of water will take place, resulting in the formation of oxidising species such as hydroxyl radicals and hydrogen peroxide.

When H₂O₂ reacts with UO₂ under groundwater conditions, secondary uranyl peroxide mineral phases are produced, in particular studtites and metastudtites, on the surface of UO₂. The stability of these minerals has not hitherto been studied on UK specific spent fuel, especially in the presence of high carbonate concentrations, which are expected to be present in UK repository groundwater compositions. Thus, it is currently unknown whether the presence of uranyl peroxides will retard the migration of radionuclides from the spent fuel matrix.

Here we present for the first time electrochemical/alpha spectroscopy experiments into the effect of deliberately added H₂O₂ on electrodeposited uranium oxide steel planchets acting as simple model fuel pin assemblies. At [H₂O₂] ≤ 100 μmol dm⁻³, dissolution of the electrodeposited uranium oxide surface proceeds rapidly, unimpeded by any corrosion product layer. At H₂O₂ concentrations between 1 and 100 mmol dm⁻³ the electrodeposited film is covered by a studtite corrosion product layer and uranium oxide dissolution is slow. Finally, at a H₂O₂ concentration > 100 mmol dm⁻³ the uranium oxide film again begins to dissolve, uninhibited by the presence of a corrosion product layer. Such corrosion processes have been described through the formation of a novel pseudo-polargram, Figure 1.

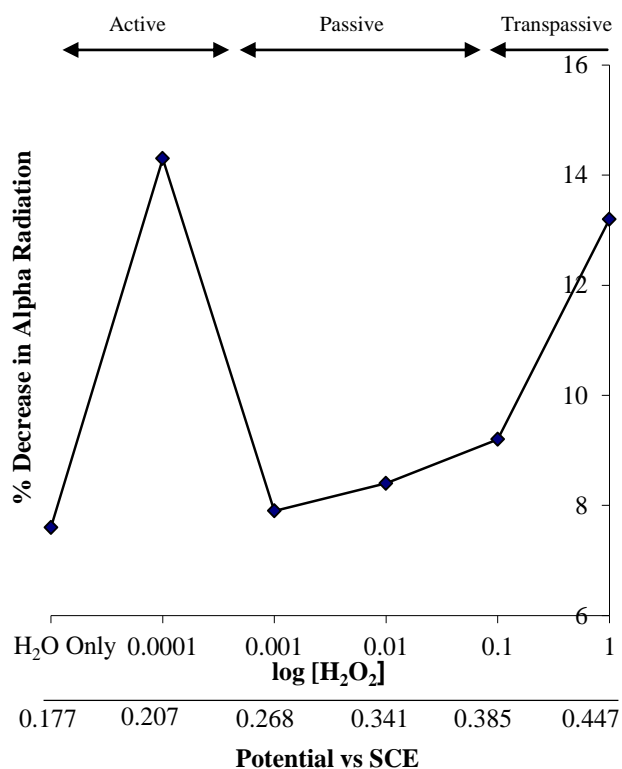


Figure 1- Pseudo current vs. potential plot showing the effect of hydrogen peroxide concentration on a electrodeposited uranium oxide planchet in pH 6.1 distilled water

We also discuss here preliminary cyclic voltammetric and impedance spectroscopy results for more realistic SimFuel rotating disc electrode (RDE) samples in increasing H₂O₂ concentrations. Future experiments will aim to measure SimFuel RDE corrosion rates under equivalent UK repository groundwater conditions.