Abstract #1774, 224th ECS Meeting, © 2013 The Electrochemical Society

The Local Redox Conditions within Spent Nuclear Fuel inside a Failed Nuclear Waste Container – a Modelling Study

Linda Wu, Zack Qin, and David Shoesmith

Department of Chemistry and Surface Science Western
Western University, London, ON, N6A 5B7, Canada

The recommended approach for the long term management of used nuclear fuel in Canada is adaptive phased management which includes the option of isolation of the used fuel in centralized containment in a deep geological repository. Although it is not expected, in the event of container failure, two corrosion fronts would exist within the groundwater-flooded container: one on the fuel surface driven by radiolytic oxidants, and a second one on the carbon steel surface sustained by water reduction and producing the potential redox scavengers, Fe^{2+} and H_2. As illustrated in Fig. 1, a complex series of electrochemical, chemical and mass transport processes will determine the redox conditions at the fuel surface within the failed container, and will determine the fuel corrosion/radionuclide release rates. Of particular importance is the prospect that the Fe^{2+}/H_2 produced by steel corrosion could scavenge the radiolytic oxidants (predominantly H_2O_2) thereby limiting fuel corrosion.

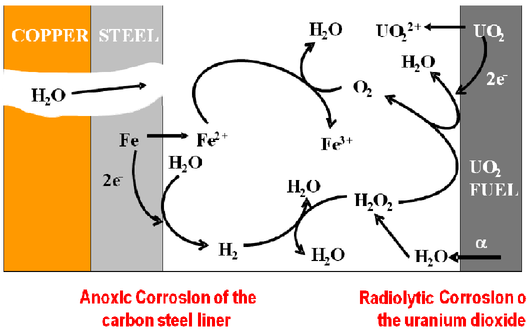

Fig 1. Schematic showing the corrosion scenario inside a failed nuclear waste container.

Due to the fractured nature of spent fuel, the local accumulation of radiolytic oxidants (H_2O_2) within the fuel and the limited transport of H_2/Fe^{2+} to these locations is expected. In addition, the microstructure of the spent fuel, in particular grain boundary tunnels caused by gas release in-reactor, can potentially provide internal corrosion sites within fuel matrix. Long term exposure tests have indicated such locations are particularly susceptible to radiolytic corrosion. A model is being developed to study the ability of steel corrosion products (Fe^{2+} and H_2) to suppress the α-radiolytic corrosion of the spent fuel. The model is based on experimentally-determined rate constants and finite element methods developed using COMSOL Multiphysics.

This model, while not yet fully developed, indicates that Fe^{2+} will have only a minor influence on fuel corrosion processes by scavenging H_2O_2 in the bulk solution via the Fenton reaction. By contrast, H_2 is found to be the dominant redox-control agent and able to significantly suppress fuel corrosion. The concentration of H_2 required to suppress corrosion varies with location within the fuel pellet. Presently, the influence of a breach in the Zircaloy cladding on the transportation of Fe^{2+}/H_2 to fuel corrosion sites is being investigated.

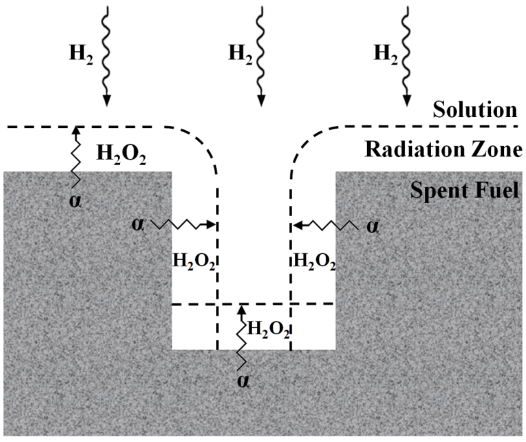

Fig. 2. Schematic showing the radiation zone (the depth of solution at the fuel surface within which radiolytic oxidants can be produced by alpha radiolysis) and the transport of H_2 from a remote steel location within the waste container. The geometry shown illustrates a short and wide fuel fracture.