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Spent fuel alteration 1D model integrating water radiolysis and reactive solute transport – Simulation of Cr/(Cr+Al)-doped UO_2 fuels leaching experiments.

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A 1D reactive transport model has been implemented in iCP [1] (interface COMSOL Multiphysics - PHREEQC) to assess the corrosion of Spent Nuclear Fuel (SF) matrix of a failed container in a deep geological repository. The model considers the SF as homogeneous UO_2(am,hyd) doped with Pd and couples transport by diffusion with the following processes: i) generation of water radiolysis species by alpha and beta radiation considering a complete radiolysis system with kinetic reactions involving: H^+, OH^-, O_2, H_2O_2, H_2, HO_2^-, $HO_2\cdot$, $O\cdot$, O^-, O_2^-,$H\cdot$, $\cdot OH$ and e^-, ii) kinetic oxidative dissolution of UO_2(am,hyd) and subsequent reduction of oxidized fuel, considering H_2 activation by Pd, occurring at the SF surface and iii) corrosion of Fe(s) in oxic and anoxic conditions. Processes i) were implemented in COMSOL (as well as transport by diffusion) and processes ii) and iii) were implemented in PHREEQC with their kinetic constants being calibrated with existing experimental data. Model development and its calibration is described in Riba et al. (2020) [2]. iCP couples COMSOL and PHREEQC into a unique model.

Several 1D simulations have been performed to quantify the effect of steel (Fe) next to the SF as well as the effect of pellet porosity on SF dissolution.

The UO_2(am) dissolution rate obtained with the model is 10^{-7} y^{-1} and lies in the range of values considered in safety assessments (10^{-8} – 10^{-6} y^{-1}) when taking into account the inhibiting (protecting) effect of H_2 on the long-term dissolution of SF matrix.

The model has been used to simulate different sets of independent experimental data (not used in the model calibration) of UO_{2x} spent fuel, Cr-doped UO_2 and Al-Cr-doped UO_2 leaching experiments under different experimental conditions. From these simulations the effect of Cr and Cr/Al used as dopants in “modern” types of light water reactors fuels on the spent fuel matrix dissolution is mechanistically investigated.

References

[1] Nardi et al. (2014). Interface COMSOL-PHREEQC (iCP), an efficient numerical framework for the solution of coupled multiphysics and geochemistry. Computers & Geosciences 69:10-21

[2] Riba et al. (2020). Spent fuel alteration model integrating processes of different time-scales. MRS Advances,5(3),159-166.