

Barrier function of a corroding iron based container

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Thick-walled cast-iron containers encapsulate highly radioactive waste in a deep geological repository and provide mechanical stabilization against lithostatic pressure and shear forces. Intrusion of water and corrosion, however, limits container integrity to a certain time period of several hundreds (BMU, 2010) to several thousands of years (NAGRA, 2002). Repository concepts in crystalline rock therefore include an additional copper coating, providing chemical resistance against water corrosion.

Safety analyses usually do not consider chemical barrier functions of corroding cast iron containers. Anaerobic iron corrosion establishes strongly reducing conditions with redox potentials at the water stability limit (see Fig.). H_2 produced upon anaerobic iron corrosion strongly inhibits the radiolysis driven corrosion of used nuclear fuel (Spahiu et al., 2000). Simultaneously, iron converts into secondary corrosion phases such as $Fe(OH)_2$, green rust phases, magnetite etc. In a disposal concept as discussed in Germany for horizontal waste emplacement of heat producing radioactive waste that involves thick-walled (e.g. POLLUX®) containers the Fe-inventory (ca. 270.000 Mg) is by far exceeding the forecasted inventory of uranium, the main component of used nuclear fuel (ca. 15.000 Mg) (Bollingerfehr et al., 2011). It is obvious that the corroding container must be considered as a potential additional chemical barrier in nuclear waste disposal concepts, that retards migration or even causes immobilization of dissolved radionuclides.

We have investigated the interactions of various radionuclides representative for heat producing radioactive waste with various iron mineral phases. According to our results and in agreement with published studies radionuclides do react with secondary iron corrosion products via various mechanisms:

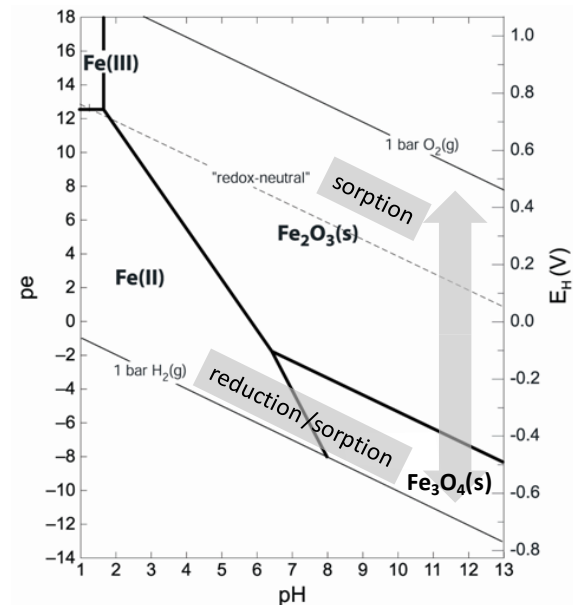


Fig.: Possible radionuclide retention processes schematically indicated in a simplified redox diagram of iron (10^{-4} mol L^{-1} total Fe concentration)(Metz et al., 2012)

- surface induced reduction of actinides (e.g. U, Np, Pu) and fission products (e.g. Tc, Se) by which they transform to reduced and poorly mobile species
- surface complexation of redox inactive radionuclides (e.g. Am) at reactive surfaces by which migration is retarded
- formation of secondary solid phases (e.g. $FeSe_x$)

Recent experimental findings on radionuclide retention on iron corrosion products will be summarized and their relevance for safety case consideration will be discussed.

References

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