Risk-Informed Assessment of Radionuclide Release from Dissolution of Spent Nuclear Fuel

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- Note:

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Abstract

This paper presents radionuclide release behavior from dissolution of spent nuclear fuel (SNF) based on the assessment of representative literature and independent analysis. The presentation focuses on risk-informed release by considering a system performance assessment (PA) from the SNF dissolution behavior in a generic SNF disposal system.

Introduction

Three distributions of SNF dissolution rates were considered and exercised in a stylized probabilistic PA Code, *Scoping of Options and Analyzing Risk (SOAR*) under oxidizing, reducing and mixed groundwater redox conditions. The probabilistic approach captures uncertainties associated with models and data; and the stylized assessment uses a simplified approach compared to implementing full features, events and processes in a code. Cases are considered for commercial SNF including spent MOX; instant release fraction and slow matrix dissolution; coupling to container failure rate or to cladding protection; sorption and precipitation; insoluble colloids or dissolved multiple radionuclide species (i.e., poly-radionuclei); and radionuclide redistribution after SNF dissolution.

Radionuclide Release Behavior

The radionuclide release from the matrix dissolution could occur from initial emplacement time after container failure. We can assume that for 1 million year durability of SNF an average 10 % will release in 100,000 years and 1 % will release in 10,000 years. An earlier release rate could be higher compared with mean (i.e., average) values over the time for total dissolution. Initially higher surface areas of SNF fragment geometry or higher radionuclide inventory in rims will contribute to more radionuclide release. In the reducing environment, the SNF dissolution is generally considered to be inhibited by hydrogen produced during iron corrosion. However, there are uncertainties associated with this consideration. For example, the carbon steel can corrode at a similar rate to the hydrogen diffusion rate. For faster corrosion rate in the

distribution, the hydrogen diffusion rate will be faster as well before reaching a steady state. Therefore, hydrogen may diffuse out without accumulation. Low radiolysis levels in the mixed groundwater redox condition at a later time may contribute to appreciable solubility-controlled or oxidative release.

The rate of fast instant release or oxidative release will be coupled with a container failure rate. If the time interval of a container failure and the next container failure is longer than the time of instant release or oxidative release, the container failure rate will control the radionuclide release from SNF and container package. Therefore, the net release will not be cumulative with all container failures. The release will be dispersed over time. Later the net release will be coupled with a radionuclide transport rate in the host rock. Similar to this coupling, the release from SNF and container package can be coupled with cladding failure one more time in a similar manner. Various mechanisms for crack formation in cladding are being studied, including initial cracks, hydrogen-induced cracks and unzipping by oxidation of the SNF matrix or cladding materials.

If uranium and other actinides are precipitated, form surface complexation, or are sorbed in backfill or host rock, they need to be counted in assessing the SNF dissolution on the SNF surface. Activated and fission products have high solubility. Therefore, the congruent dissolution on the SNF surface is assumed and these high solubility radionuclides will not be retarded during the transport. In addition, the stability of precipitation, complex formation or sorption needs to be examined over a long period of time. Colloid or poly-radionuclei may facilitate the radionuclide release. The size of poly-radionuclei is sometime of molecular level and, therefore, their potential transport needs to be considered, including effects of failed container to retard the transport. As the exercise involves dissolved or sorbed species, precipitates and colloids, a mass balance was also exercised for the redistribution of radionuclides in the disposal system.

Summary and Future Work

Risk-informed release behavior of radionuclides is discussed from SNF dissolution. Confirmatory testing and model elaboration are under way.

References

T. Ahn, “Risk-Informed Assessment of Radionuclide Release from Dissolution of Spent Nuclear Fuel,” Presentation at Spent Fuel Workshop 2014, Karlsruhe, Germany, September 2014, NRC ADAMS, www.nrc.gov, ML14211A538, U.S. Nuclear Regulatory Commission, 2014