

Spent fuel evolution in deep geological disposal site

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A short introduction in repository science, discussing the so-called "pillars of safety" i.e. the factors that contribute to reduce substantially the dose from the repository as compared to the toxicity of the high level waste forms [1]. Then a discussion of dose relevant nuclides and how to select them will be given [2]. The processes which occur in a nuclear reactor, with emphasis on the chemical state of fission products in the mixed oxide fuel matrix will be discussed, emphasizing their influence in the oxidative dissolution of the fuel matrix [3, 4].

The radiolysis promoted dissolution of spent fuel and the factors that affect both matrix dissolution and the fast released fraction of radionuclides (the so called Instant Release Fraction, IRF) will be discussed afterwards [5, 6]. The influence of fuel burnup on matrix dissolution rates will be discussed based on data with spent fuel leaching [7] and the dissolution kinetics of model systems such as UO₂ doped with various amounts of other cations. The redox equilibria with some examples of redox kinetics for important redox processes that occur in a repository will be also discussed.

Recent data on spent fuel leaching under expected deep repository conditions created by the massive iron containers and their anoxic corrosion products will be discussed with emphasis on the importance of process understanding. Both batch autoclave experiments with several fuel types and flow through tests will be discussed, as well as investigations with alpha doped UO₂ simulating "old" fuel [8]. The importance of the oxidative fuel dissolution under repository conditions will be discussed and compared to the non-oxidative fuel dissolution. Fuel dissolution models and the mechanistic understanding of the fuel dissolution process under reducing conditions will be presented, as well as the need for further investigations in field of interfacial radiolysis [9].

Finally radionuclide speciation and the importance of reliable thermodynamic databases with emphasis on NEA-TDB will be discussed, together with radionuclide solubilities, co-precipitation and sorption. Co-precipitation will be discussed by looking in more detail to the example of Ra-Ba sulfate co-precipitation under repository conditions [10].

References

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