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Experimental investigation of uranium release from spent nuclear fuels under conditions expected in a deep geological repository

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Experimental investigation of uranium release from spent nuclear fuels under conditions expected in a deep geological repository

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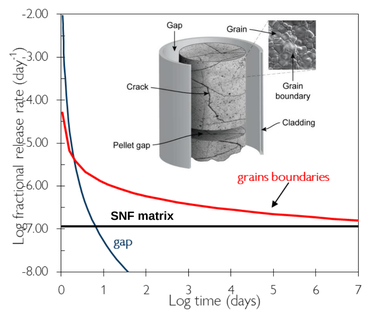
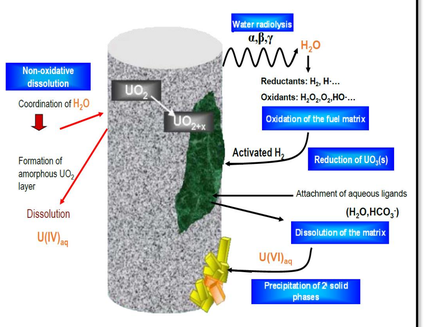
BACKGROUND AND OBJECTIVES

- Long-term management of highly radioactive waste in Germany foresees direct disposal of spent nuclear fuel (SNF) in a deep underground repository.
- Water access, consecutive failure of canisters and loss of fuel cladding integrity considered in long-term safety analyses of such facilities.
- Assessing performance of SNF in geological disposal systems requires process understanding of SNF dissolution and rates as well as the quantification of radionuclides release from SNF under reducing conditions of a breached container.





Objectives:

- Since most radionuclides produced during irradiation are trapped within the SNF matrix, understanding of the processes leading to the dissolution of the matrix and its dissolution rates are of particular interest for post-closure safety assessments.
- Behavior of SNF is studied under geochemical conditions representative of various repository concepts and SNF dissolution mechanisms are derived.

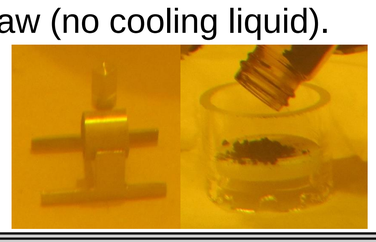
MATERIALS AND METHODS

SNF characteristics:

- Reactors: PWR Gösgen (CH) and PWR Obrigheim (D).
- Type of fuel: UO_2 and MOX.
- Average burn-up: 38 & 50.4 GWd/t_{HM}.

Preparation of SNF samples:

- Samples cut by low speed saw (no cooling liquid).
- Segments with cladding and fragments without cladding.



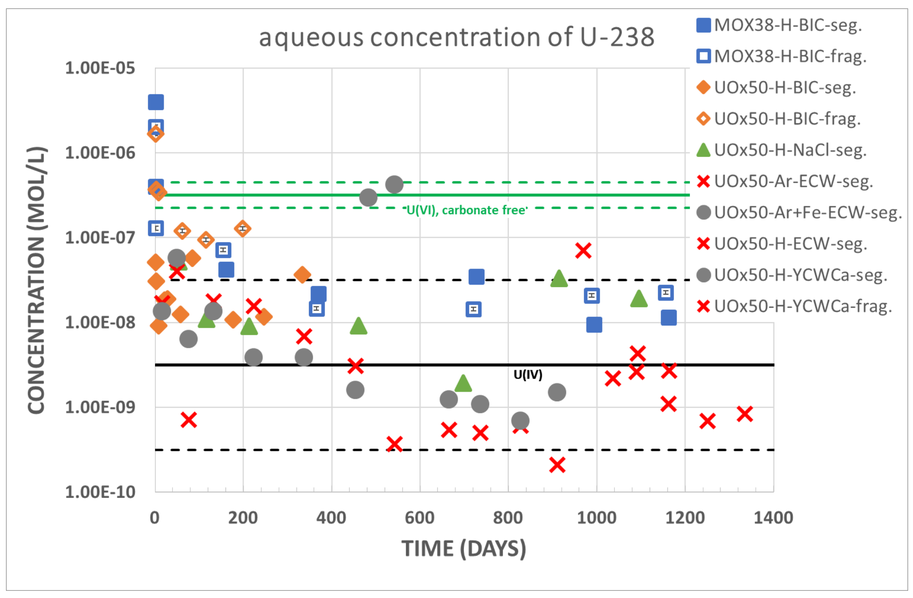
Leaching experiments:

- Ar or Ar/H_2 atmospheres.
- salt brines, granitic/bentonitic groundwater, cementitious groundwater.

RESULTS

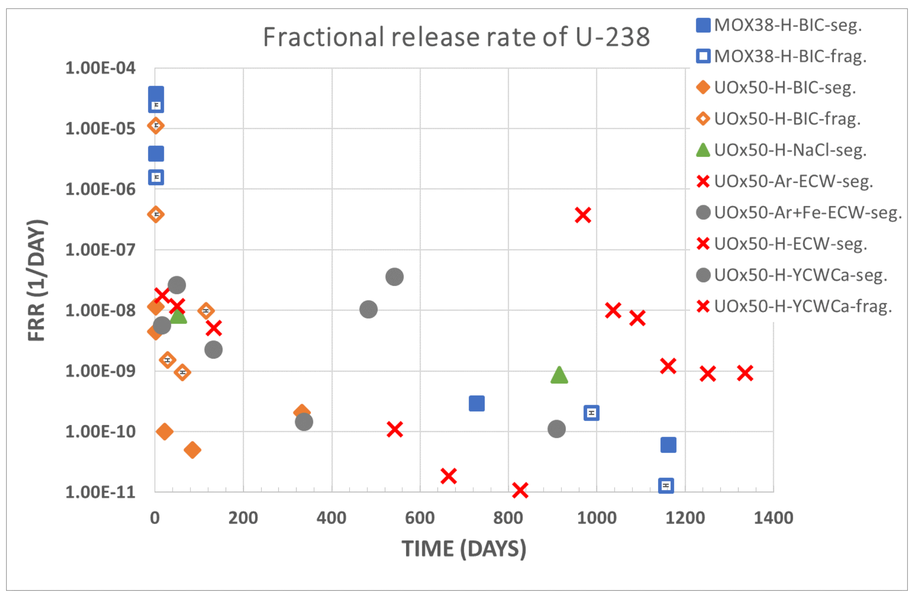
Uranium release to solution in molar concentration:

- Large scatter of [U] in initial stages of experiments.
- [U] approaches towards solubility limit of U(IV) after > 1 year independent of type of studied SNF (UO_2, MOX, segments or fragments).
- Similar behavior observed for other redox-sensitive elements (not shown).



Rates of uranium release to solution:

- Release rate of uranium decreases significantly in all experiments within first 400 days of leaching and seems to be constant within scatter between $1 \cdot 10^{-11}$ and $1 \cdot 10^{-9}$ per day.
- Although rate is very low, it is > 0 and continuous release is observed.
- In long-term safety assessments a fractional radionuclide release rate of 10^{-7} per year is often used (≈ $3 \cdot 10^{-10}$ to $2 \cdot 10^{-9}$ per day).



SUMMARY AND CONCLUSIONS

- Comparison of results from SNF experiments under various redox conditions demonstrate that oxidative matrix dissolution is inhibited due to presence of hydrogen.
- No significant difference in matrix dissolution of investigated spent UO_2 and MOX fuels.
- Composition and pH of simple, synthetic groundwater plays minor role in matrix dissolution.

References

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