Nuclear Waste Management IEK-6
Institute of Energy and Climate Research

JÜLICH FORSCHUNGSZENTRUM

Studying corrosion mechanisms of modern LWR-fuels using UO_2-based model systems

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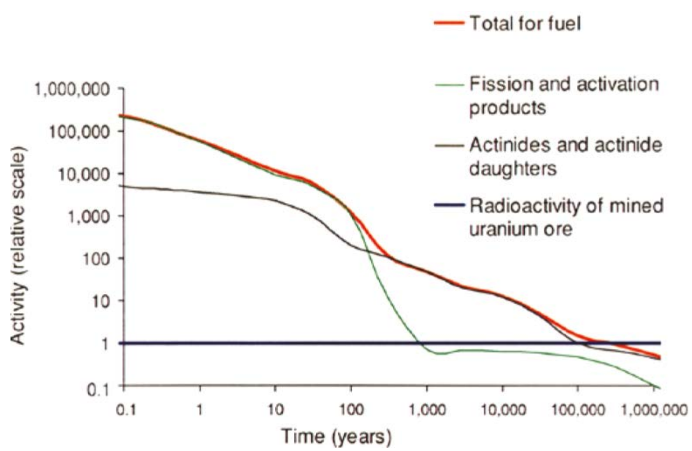
Spent nuclear fuel corrosion in a deep geological repository?

- SNF: high chemical & structural complexity
- “Fresh” SNF not representative for time scales >> 1,000 y
- doped SNF may show different corrosion behaviour than “standard” SNF
- Radionuclide release from spent nuclear fuel:
  - **Instant release fraction**
  - **Long-term matrix corrosion**: release of An, Ln, etc. matrix contains > 99% of the total actinide inventory

→ **Superposition of various effects**



Bruno & Ewing 1988

Approach: long-term matrix corrosion mechanisms under repository relevant conditions by **studying single effects of tailor-made UO_2 based model systems (formerly SIMFUEL)**

Our complementary approach

Our bottom-up approach, complementary to SNF studies:
Systematic exploration of UO_2 based model systems

- Fabrication of doped UO_2 model systems
- Characterization with state-of-the-art-techniques
- Corrosion studies

Improved & predictive understanding of spent nuclear fuel long-term matrix corrosion (> 10,000 y)

Our bottom-up approach to unravel mechanisms of long-term SNF matrix corrosion

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Fabricate UO_2 based model systems to individually study the following effects:

- **Effect of dopants from modern fuels, e.g. Cr_2O_3, Al_2O_3 or SiO_2**
- **Mimic α-radiation field of aged SNF by doping with e.g. U-233 or Pu-238**

Ongoing research at IEK-6:

- Effect of Ln doping of the UO_2 concerning the oxygen sublattice & effect on electrochemical potential?
- Effect of concentration & distribution of Ln dopants in the UO_2 system concerning corrosion?
- Effect of ε-particles as scavenging agents of radiolytical oxidants and corrosion suppression by H_2?
- Synergistic effects between Ln dopants & ε-particles on long-term matrix corrosion?

Matrix corrosion of doped fuel?

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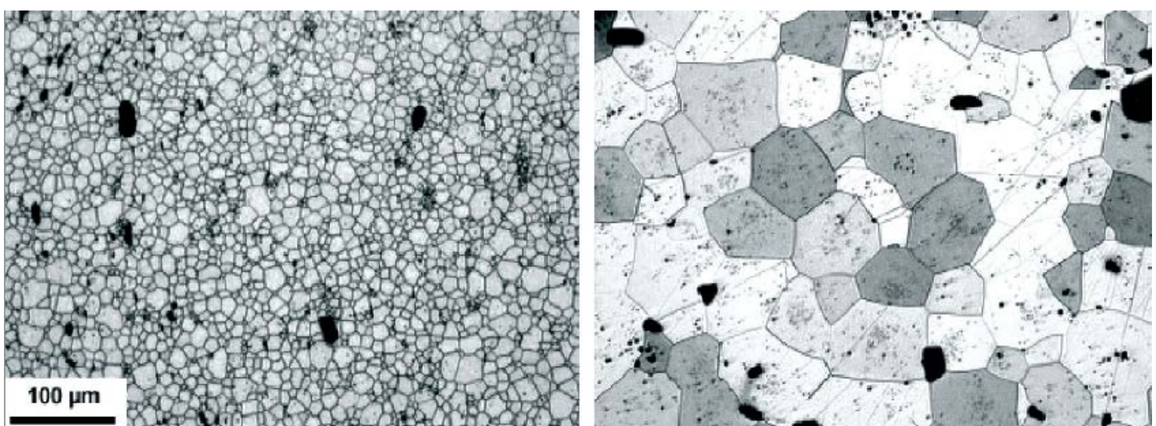
Doped fuel:

- Microstructural changes, e.g. larger grains
- Higher burn-up in comparison to undoped fuel

Direct comparison of UO_2 based model systems with results of doped SNF from ITU & Studsvik:

- Effect of grain size concerning long-term matrix corrosion
- Concentration influence on scavenging effects of dopants in H_2 atmosphere

→ **Transfer of results from UO_2 based model systems to “real” SNF**



Standard UO_2 | Cr_2O_3-doped UO_2

CEA, 2009.

Competences at FZJ - IEK-6

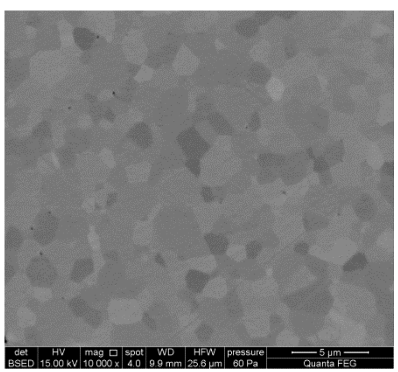
Highly experienced in fabrication of An-doped ceramic oxides:

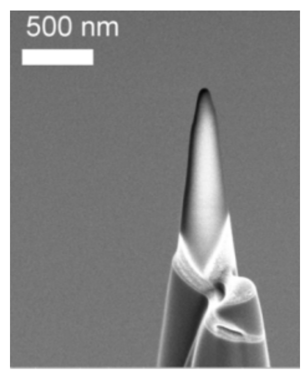
- Tailor-made synthesis routes for specific needs
  → focus: innovative wet-chemical approaches
- Hot press & piston cylinder press

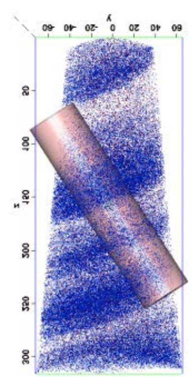
Advanced characterisation techniques/methods for a molecular level process understanding:

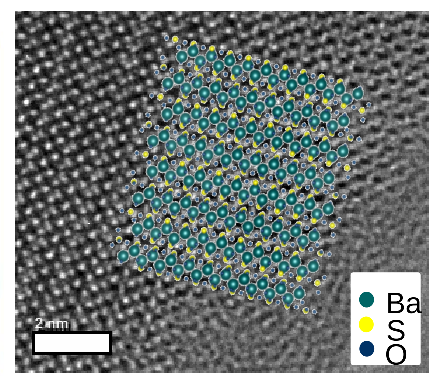
- Standard characterization equipment: XRD, SEM, Raman, IR
- FIB & TEM & EBSD: Microstructure & local structure of ceramics
- Atom Probe Tomography: Chemical composition at the atomic level

→ Identification of dopant localisation & microstructure



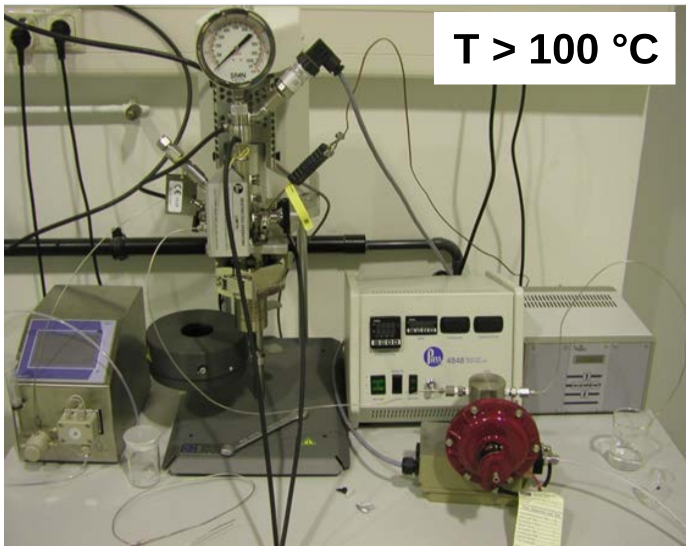


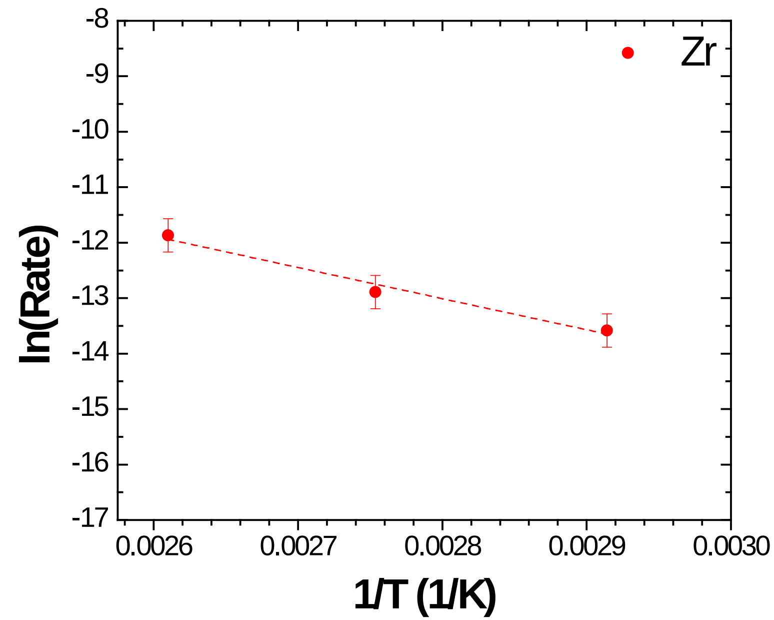


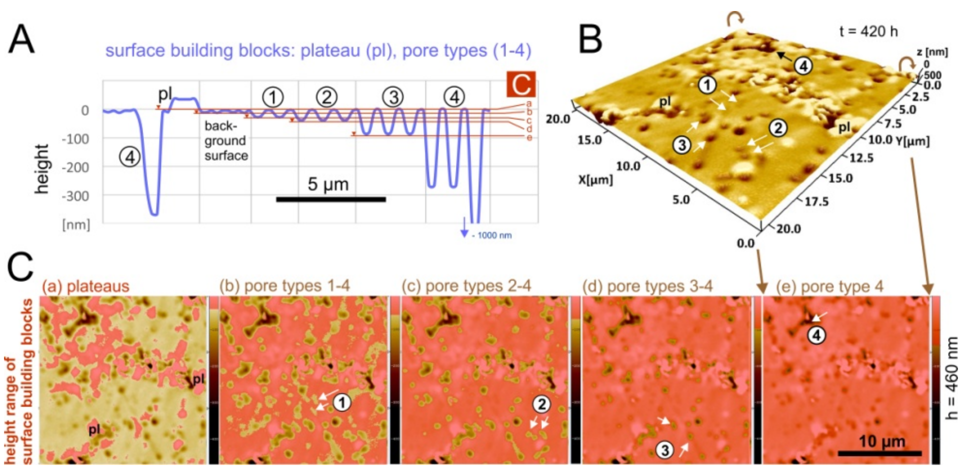


Competences at FZJ - IEK-6

Corrosion experiments in autoclaves & glove boxes:







Summary

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Innovative bottom-up approach, complementary to SNF research

- Fabrication of tailor-made e.g. Cr_2O_3 doped UO_2 model systems containing Ln (FP product surrogates) and ε-particles
- Mimic α-radiation field of aged SNF by doping with e.g. U-233 or Pu-238
- Microstructural & electrochemical characterisation
- Oxidative (radiolytic) corrosion studies with groundwater simulants (representative for repositories in crystalline rocks and clay systems) under reducing atmosphere
- Transfer of mechanistic insights from UO_2-based model systems to spent nuclear fuel

→ **Predictive & improved molecular understanding of key processes relevant for long-term matrix corrosion**