SPENT NUCLEAR FUEL MANAGEMENT, CHARACTERISATION, AND DISSOLUTION BEHAVIOUR: PROGRESS AND ACHIEVEMENT FROM SFC AND DISCO

Presented by Prof. Anders Sjöland, SKB, on behalf of the authors

EURADWASTE 2022, Lyon, France 2022-05-31

This project has received funding from the European Union's Horizon 2020 research and innovation programme 2014-2018 under grant agreement N°847593

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WP 8 : SFC – Spent Fuel Characterization

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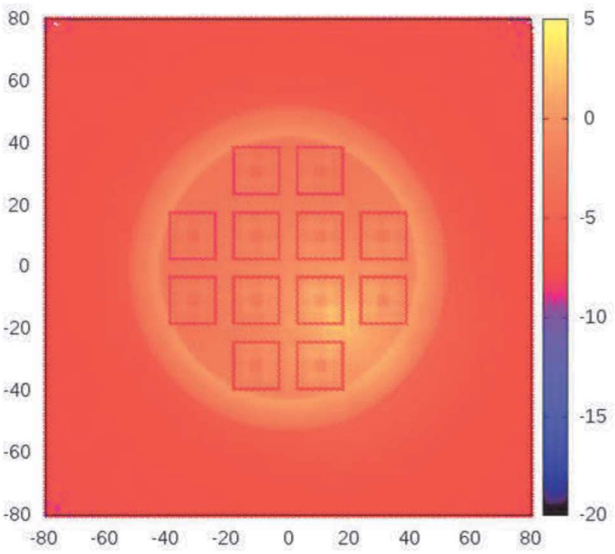
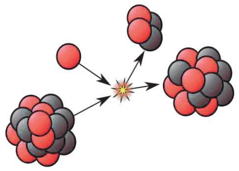
WHY SPENT FUEL CHARACTERIZATION?

- It is NUCLEAR MATERIAL we want to handle and dispose of
- All relevant properties of the fuel must be known in the different steps of the nuclear fuel cycle's back-end

- A geological repository is a 'difficult to access location'. The fuel cannot easily be inspected ever again.
- The requierment is that all nuclear material placed in a difficult to access location must be completely characterized before being put in such a place.
- No surprises in the future

- Long-term safety – e.g. fuel dissolution behavior (rate), fission gas release (FGR)





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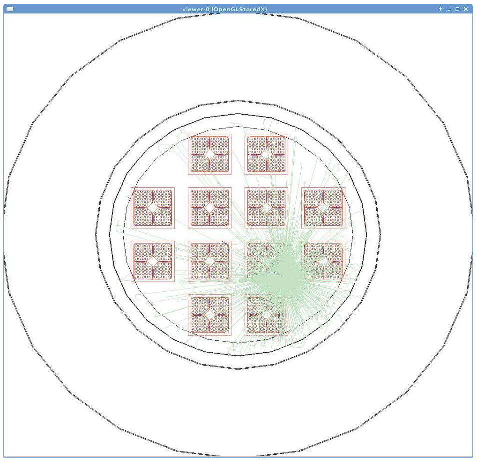
WHY SPENT FUEL CHARACTERIZATION?

- Calculations by nuclear codes are used to a large extent in the back-end.
- These must be validated experimentally.
- Little statistics, deep understanding necessary.



Parameters to characterize

- *Decay heat* – to fulfil temperature requirement on canister, bentonite, rock and fuel
- *Criticality* – multiplicity: to assure that criticality does not occur
- *Radiation doses* – both gamma and neutrons: For safety
- *Nuclide inventory: For safety analysis*
- *Safeguards verification*

Identify correct fuel, missing pins

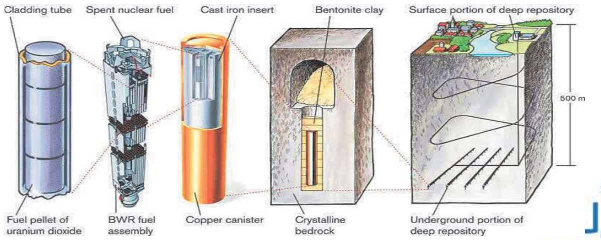
Contents of fuel – amount of fissile material- Burn-up (BU), Initial enrichment (IE), Cooling time (CT), weight

- Fuel integrity – mechanacaal

Damaged fuel

- Dissolution rate of fuel
- Fission gas release (FGR)

All parameters coupled



UNCERTAINTIES

- Uncertainty is a statistical term that is strictly defined.
- No fuzzy concept
- Used for technical DESIGN of systems such as casks, transport, repositories, etc.
- Initial state for safety analysis
- Little statistics, deep understanding necessary

- Thermal modeling
- Economical optimisation – required by law in Sweden for example

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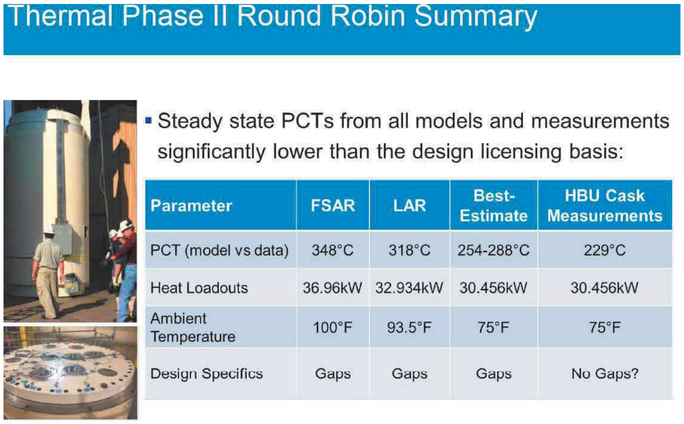
Uncertainties - required

- *Decay heat*: very high accuracy, order of few percent uncertainty *in reality perhaps 5-10 %*
- *Criticality*: very high accuracy, order of few percent uncertainty
- *Radiation doses*: high accuracy, order of 10 %
- *Nuclide inventory*: for most nuclides fairly low accuracy need; <100 % (for some nuclides higher accuracy needed)
- *Contents of fuel* – amount of fissile material - Burn-up (BU), Initial enrichment (IE), Cooling time (CT): intermediate accuracy

Temperature

- Often the requirement linked to decay power is temperatures
- Temperature modeling essential - often been considered ’easy’, but may not always be
- Often requirements and determinations contain large conservatisms



Thermal Phase II Round Robin Summary

- Steady state PCTs from all models and measurements significantly lower than the design licensing basis:

Parameter	FSAR	LAR	Best-Estimate	HBU Cask Measurements
PCT (model vs data)	348°C	318°C	254-288°C	229°C
Heat Loadouts	36.96kW	32.934kW	30.456kW	30.456kW
Ambient Temperature	100°F	93.5°F	75°F	75°F
Design Specifics	Gaps	Gaps	Gaps	No Gaps?

Slide courtesy of Al Csontos, Co-chair of EPRI ESCP Thermal Subcommittee

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TASK 2
FUEL PROPERTIES CHARACTERISATION AND RELATED UNCERTAINTY ANALYSIS

The project leading to this application has received funding from the European Union’s Horizon 2020 research and innovation programme under grant agreement n° 847593.

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Task 2: Fuel properties characterisation and related uncertainty analysis

- Subtask 2.1 “Theoretical study of SNF source terms” (Theory)
  co-ordinator PSI (D. Rochman)
  partners CIEMAT, JSI, JRC Geel, NAGRA, PSI, SCK CEN, VTT
- Subtask 2.2 “Develop, improve and demonstrate NDA methods/systems for SNF characterisation” (NDA)
  co-ordinator SCK CEN (M. Verwerft)
  partners CIEMAT, ENRESA (ENUSA), JRC Geel & Ispra, SCK CEN, SKB (UU)
- Subtask 2.3 “Calculate and determine experimentally the inventory of activation and FP in cladding” (Cladding)
  co-ordinator KIT (R. Dagan)
  partners CIEMAT, KIT, LEI, NAGRA, VTT, SURAO(CTU)
- Subtask 2.4 “Define and verify procedures to determine source terms of SNF with realistic confidence limits”
  co-ordinator PEL (KIT) (M. Seidl)
  partners CIEMAT, CPST, JRC Geel, ENRESA, JSI, PEL, SSTC NRS, KIT, SCK CEN, TUS, SKB (UU)

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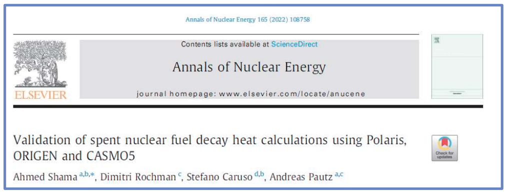
Status: 2020 - 2021

SUBTask 2.1 Theoretical study of SNF source terms

- cases to verify the performance of different codes and perform sensitivity and uncertainty analysis were identified
- results partly reported



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journal homepage: www.elsevier.com/locate/anucene

Validation of spent nuclear fuel decay heat calculations using Polaris, ORIGEN and CASMO5

Ahmed Shama [a,b,*], Dimitri Rochman [c], Stefano Caruso [d,b], Andreas Pautz [a,c]

PhD Thesis: A. Shama

Partner	Code(s)	Cases (reported / in progress)
CIEMAT	MCNP/EVOLCODE, MCNP/CINDER	SF95-5
ENRESA/ENUSA	POLARIS	*BWR (ENRESA)*
JSI (JRC)	POLARIS, SCALE (TRITON/NEWT), SERPENT2	S1.PWR, NPP Krško, SF95-5, REGAL, SKB-2006
SCK CEN	MCNP/ALEPH2, SERPENT2	S1.PWR, SF95-5, REGAL, SKB-2006
KIT	MCNP/CINDER, Nucleonica	SF95-5
NAGRA	SCALE (TRITON), POLARIS	SF95-5, BM1, Gundremmingen-7(B23), GE-Morris, SKB-2006
PSI	CASMO, CASMO/SIMULATE/SNF	GU1, GU3, BM1, BWR(ENRESA), GE-Morris, SKB-2006
VTT	SERPENT2	Gundremmingen-7 (B23)

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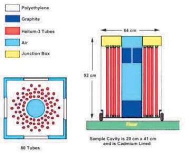
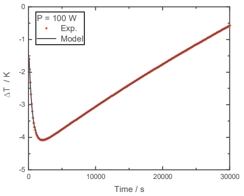
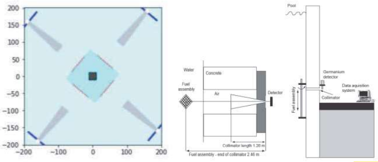
SUBTask 2.2: Develop and improve nda methods/systems



- Neutron emission rate of a SNF by NDA at SCK CEN
  - Experiments (see EUR30379 EN) and comparison with theoretical calculations finalised
    Journal paper in progress
  - Design of improved transfer container and detection system: finalised
    New experiments scheduled for autumn 2022



- Improve analysis procedure for calorimeter at CLAB (MSc Thesis: J. Ekman)
  - Analytical model to describe heat transfer
  - SERPENT model for γ-ray transport (estimate γ-ray escape)
- Improve analysis procedure for γ-ray measurements at CLAB (PhD Thesis: V. Solans
  - Uncertainty assessment of net peak area determination
  - Study of systematic effects (e.g. positioning of SNF assembly))
  - Decay heat prediction by Machine Learning



- Radiochemical analysis of a set of BWR samples: finalised, report in progress

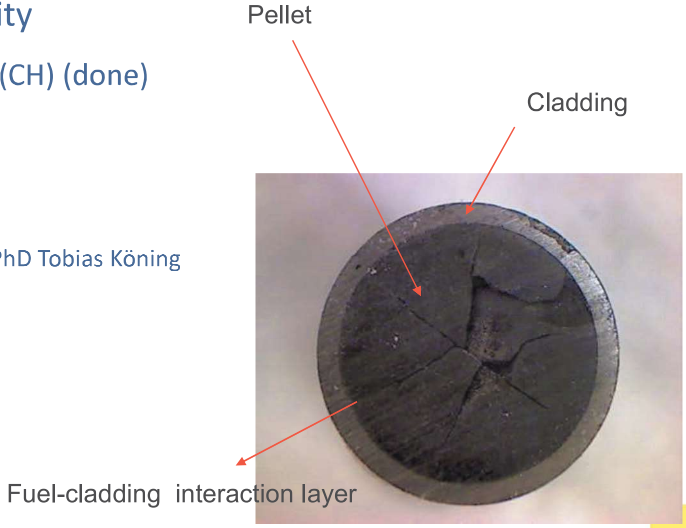
Status: 2020 - 2021

Subtask 2.3: calculate and determine the inventory in cladding

Cladding composition: important to study fuel integrity

- Selection of Zircaloy samples irradiated in a PWR Gösgen (CH) (done)
  (UO2, 50.4 GWd/t, 4 cycles, 3.8%, 32 years cooling time)
  - Zircaloy-4 of UO_x fuel rod segment
  - Zircaloy-4 plenum of UO_X fuel rod segment
  - UO_X fuel fragment
- Experiments using various techniques at KIT done, part of PhD Tobias Köning
- Code validation and comparison of results (in progress)

CIEMAT	MCNP/EVOLCODE (CIEMAT),
KIT	MCNP/CINDER
LEI, NAGRA	SCALE (TRITON)
VTT	SERPENT2



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Subtask 2.4: Define and verify procedures to determine source terms of SNF with realistic confidence limits

Planning was reviewed to account for delays due to COVID and signing of agreements

1. BWR SNF sample from task 2.2 (ENRESA/ENUSA)
   - PEL, CIEMAT
   - Report: September 2022
2. REGAL Sample (SCK CEN, JSI, JRC)
   - Neutron output: calculations with ALEPH2, SCALE, SERPENT2
   - Report: February 2023
3. SKB-50
   - Non-Disclosure agreements signed by partners
   - Distribution of data planned for January 2022
   - Report: December 2023

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TASK 3
BEHAVIOUR OF NUCLEAR FUEL AND CLADDING AFTER DISCHARGE

The project leading to this application has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement n° 847593.

Task 3: Behaviour of nuclear fuel and cladding after discharge

- Subtask 3.1 “Thermo-mechanical-chemical properties of the SNF rods and cladding”

  Coordinator UPM (J. Ruiz)

  Partners KIT (BAM), CIEMAT (UPM), EC-JRC KA, MTA-EK, NAGRA, PSI, TUS, VTT

- Subtask 3.2 “Behaviour of SNF pellets under interim storage conditions”

  Coordinator CIEMAT (N. Rodriguez)

  Partners CIEMAT, CNRS-CEMHTI, CNRS-ICSM (UMontpellier), FZJ (HZDR)

- Subtask 3.3 “Pellet-cladding interaction under conditions of extended storage, transport and handling of SNF rods”

  Coordinator MTA-EK (M. Király)

  Partners CIEMAT, KIT INE, MTA-EK, PSI

Subtask 3.1:
Thermo-mechanical-chemical properties of the SNF rods and cladding

EXPERIMENTAL

Conditions previous to accident

Optimization of hydrogen/hydrides visualization in irradiated cladding (PSI)

Creep tests on as-received, hydrogenated and heat-treated E110 samples (MTA)

New experimental setup for hydride reorientation (UPM)

Accident conditions

RC tests on unirradiated cladding with reoriented hydrides (UPM)

Mechanical properties of SNF rods from bending and RC tests (JRC KA)

MODELLING

Conditions previous to accident

Assessment/enhancement of FRAPCON-xt for rod internal pressure (CIEMAT)

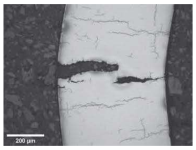
Doc

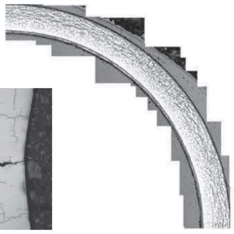
Preparation of BISON´s inputs, Implementation of dry storage creep model (VTT)

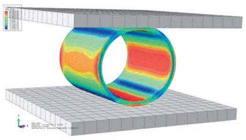
Accident conditions

Material characterization using Finite Element Analysis (BAM)

3D static ANSYS model representing a 3 point bending test (NAGRA)

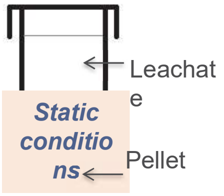




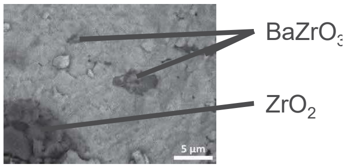


Subtask 3.2:
Behaviour of SNF pellets under interim storage conditions

- Impact of lanthanides and PGM elements on the chemical durability and surface modifications during leaching tests of UO_2 pellets, mimicking interim repository conditions (CNRS-ICSM)
  - Progress made last year: Synthesis and leaching tests of lanthanide bearing UO_2 ceramics and PGM (Platinum Group Metals) bearing UO_2 ceramics

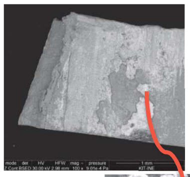


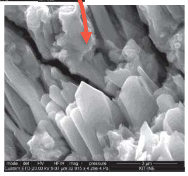
- Investigation of the effect of lanthanides on the behaviour of He and defects in doped uranium oxides using Nuclear Reaction Analysis and Positron Annihilation Spectroscopy (CNRS-CEMHTI)
  - Progress made last year: Sample reception from CNRS-ICSM, Sample characterisation (Raman, PAS, microscopy) & annealing, He release as function of the annealing temperature



- Study of the effect of the grain size on the oxidation behaviour of UO_2-based SIMFUEL doped with fission products (HZDR)
  - Synthesis of UO_2 and FP powders, Oxidative treatment, Characterization by XRD, …
- Characterization of the temporal evolution of SNF by improving the understanding of the ageing/degrading mechanisms in dry storage (CIEMAT)
  - Raw materials characterization (XRD, Raman, Electron microscopy, particle size, …)

Subtask 3.3: Pellet-cladding interaction under conditions of extended storage, transport and handling of SNF rods





- Chemical effects of activation and fission products on the cladding integrity under dry storage and subsequent transportation conditions (KIT INE)
  - Experiments completed in July 2020
  - Ongoing investigation of precipitates found at the fuel-cladding interface
- Investigation of intrusion of uranium into the Zr barrier (CIEMAT)
  - Samples prepared, Oxidation of UO_2-ZrO_2 pellets done
  - Delays in Postdoc recruitment campaign, 6 month delay due to the pandemic
- Mandrel ductility tests (MTA-EK)
  - All mandrel tests using E110 Zr1%Nb cladding completed
- Influence of the pellet on cladding stresses and the resulting hydride distril (PSI)
  - Delays concerning the commissioning of the hot lab FIB and Postdoc recruitment campaign

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WP8 : SPENT FUEL CHARACTERISATION AND EVOLUTION UNTIL DISPOSAL (SFC)

TASK4 – Accident scenario and consequence analysis

Luis E. Herranz /CIEMAT

The project leading to this application has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement n° 847593.

STRUCTURE AND PARTNERS

Task 4: Accident scenario and consequence analysis
Ciemat

ST.4.1: Accident scenario for fuel under dry interim storage conditions
nagra

ST.4.2:Consequence analysis of accident scenarios
Chornobyl R & D Institute

nagra
Ciemat
cea
IDOM
Technical University Sofia

Chornobyl R & D Institute
nagra
Ciemat
Technical University Sofia

MAJOR ACHIEVEMENT

Articulation of a consistent programme of work with strong links among partners and with others tasks (Task 3).

Sub-Task “domains”:

- **Sub-Task 4.1:** Fuel thermomechanics.
- **Sub-Task 4.2:** SNF accident management & consequences

Deliverables:

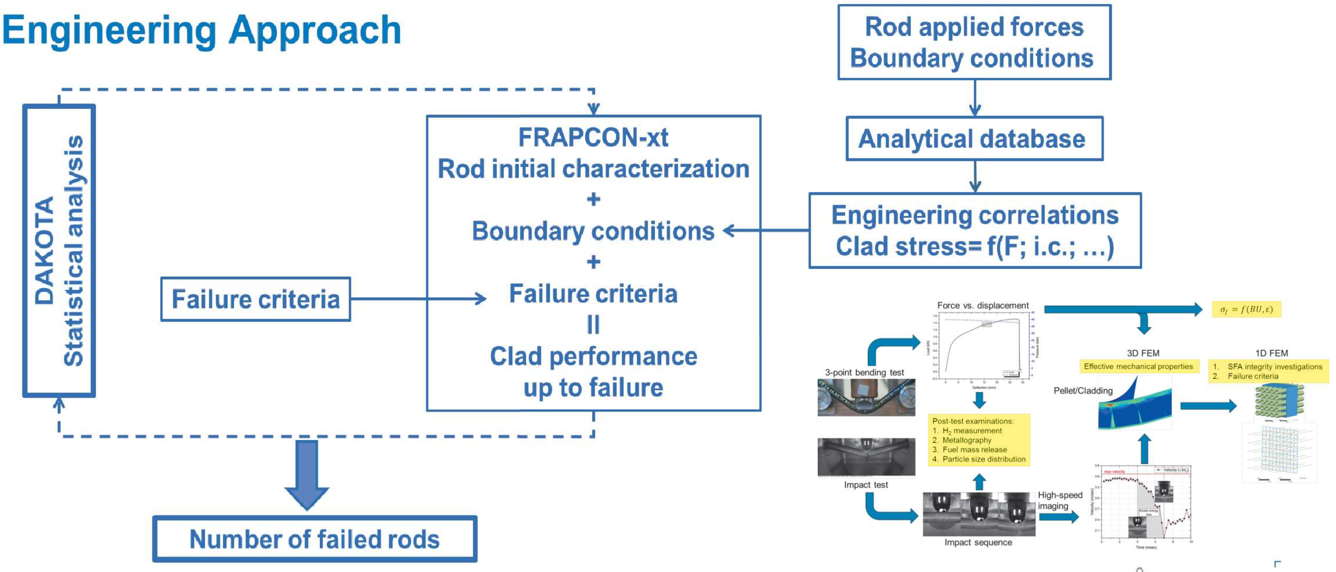
- **D8.11**: Identification and analysis of potential accident scenarios in an interim storage and/or packaging facility. Assessment of fuel performance (NAGRA - M48)
- **D8.12**: Analysis of the conditions of radioactive wastes packages contained SNF, FCM or HLW/LLW generated due to ChNPP accident (ChI – M36)

Technical Reports:

- Loading in SFA during Dry Cask Drop Accidents (IDOM- M36)
- On Source Term analysis from a vault system (CIEMAT – M42).
- On Criticality analysis of potential accidents (CIEMAT – M42).

SUB-TASK 4.1: FUEL THERMOMECHANICS

Engineering Approach



DAKOTA Statistical analysis

Failure criteria

FRAPCON-xt
Rod initial characterization
+
Boundary conditions
+
Failure criteria
||
Clad performance
up to failure

Number of failed rods

Rod applied forces
Boundary conditions

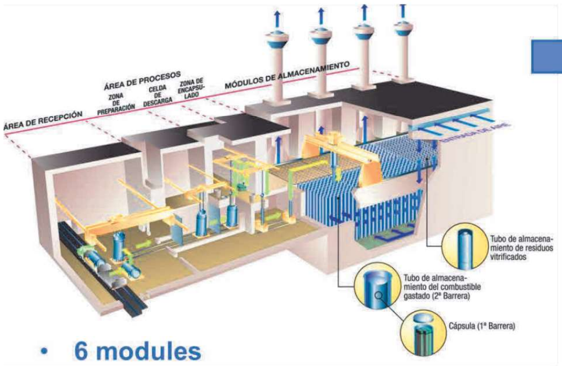
Analytical database

Engineering correlations
Clad stress= f(F; i.c.; …)

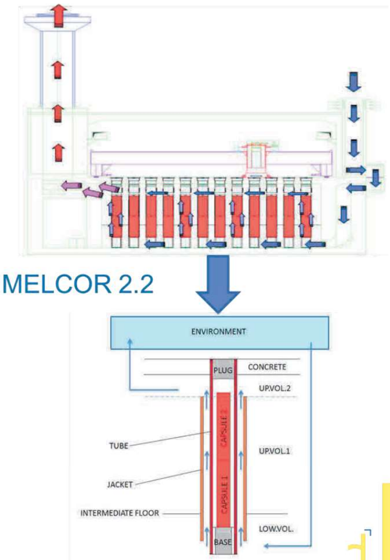
SUB-TASK 4.2: SNF ACCIDENT MANAGEMENT & CONS.

- Deliverable 8.12 drafted and under review.
- Source Term studies in vault systems initiated

Centralized Temporary Storage (CTS):



- **6 modules**
- **12 vaults**
- **120 wells per vault**



MELCOR 2.2

FUTURE OF THE WP

- **Expected added value**
  - Reduced uncertainty on decay heat ==> large economical savings using repository optimisation.
  - Reduced uncertainties on safety related parameters.
  - To contribute and further develop guidance on the operational safety of both interim storage and fuel packaging facilities.
  - Both fuel and cladding experience in time several degrading mechanisms that will inevitably affect their chemistries and hence alter their thermo-mechanical properties. The WP will provide new and meaningful insight on these mechanisms that are still not explained.
  - This WP will increase knowledge specific to the European case.
  - The combination of both numerical calculations and experimental methods will also provide a complete and thorough understanding of the mechanisms driving the behaviour of the SNF rods during its extended storage in both normal and accidental conditions.
  - Results from this WP will have a valuable impact from performance assessment and regulatory requirements point of view.

FUTURE OF THE WP (CONT.)

- **Work planned for the next year**
  - Finalise work of subtask 2.1.
  - Subtasks 2.2-2.4 continues as planned.
  - Task 3 to continue with experimental programs large delayed by the pandemic.
  - Subtask 4.1: Synthesis of the results which will contribute towards the deliverable(s).
  - Subtask 4.2: Finalisation of deliverable and reporting on performed accident vault analyses.

PROGRESS OF THE WP

- **Actions taken from the previous review**
  - **Previous review had no comments on the WP.**
- **Analysis of the KPIs**
  - The number of publications in scientific peer-reviewed journals. Counting all types of publications:
    - Month 1-12:
      - Journal papers: 2
      - Conference contributions: 4
    - Month 13-24:
      - Journal papers: 5
      - Conference contributions: 2
      - Reports: 1
      - MSc theses: 1
    - Month 25-30 (half year):
      - Journal papers: 9
      - Conference contributions: 4
      - Reports: 1
      - Book chapters: 1
      - PhD theses: 2

Task 2: Education &Training

- ## Bachelor

William Lindberg (Uppsala Univ.) (JRC Geel)
    Subtask 2.2 Uncertainty evaluation of decay heat measurements at CLAB
Gašper Letnar (Univ. Ljubljana) (JSI)
    Subtask 2.1 and 2.2 Analysis of CLAB 2006 PWR and BWR cases

- ## MSc

Sonia Panizo Prieto (Univ. Compl. Madrid) (CIEMAT)
    Subtask 2.1 and 2.4 Compare results for the Takahama-3 reactor obtained with EVOLCODE and MCNP/CINDER
Aitor Bengoechea (Univ. Pol. Madrid) (SCK CEN)
    Subtask 2.1 SANDY application for the analysis of nuclear data covariance
Daan Houben (Univ. Hasselt) (SCK CEN)
    Subtask 2.1 and 2.2 Spent nuclear fuel sensitivity to model parameters
Johannes Ekman (Uppsala Univ.) (JRC Geel)
    Subtask 2.2 Performance assessment of CLAB calorimeter
Manuel Cabezas Navarro (Univ. Pol. Madrid) (SCK CEN)
    Subtask 2.2 Production of some important nuclides in spent UO_2 fuel

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Task 2: Education &Training

- PhD

Shama Ahmed (Ecole Polytechnique Féderale de Lausanne) (PSI, NAGRA) (29 October 2021)
    Subtask 2.1 Data-driven predictive models: calculational bias in characterisation of spent nuclear fuel
Sonia Panizo Prieto (Univ. Compl. Madrid) (CIEMAT)
    Subtask 2.1 Impact of sensitivity and uncertainty in nuclear data in reactors and fuel cycles
Julio Plaza del Olmo (Univ. Compl. Madrid) (CIEMAT)
    Subtask 2.2 Development of neutron monitoring systems for fundamental science and nuclear technologies
Virginie Solans (Uppsala University) (UU, SKB, JRC)
    Subtask 2.2 and 2.4 Improved data analysis and uncertainty quantification to better estimate source terms of SNF
Tobias Koenig (KIT)
    Subtask 2.3 Examination of the radionuclide inventory and chemical interactions on the interface between nuclear fuel and Zircaloy-4 cladding in irradiated LWR-fuel samples

Status: 2020 - 2021

Task 2: journal papers

- Fiorito et al., Ann. Nucl. Energy 161 (2021) 108415
  "On the use of criticality and depletion benchmarks for verification of nuclear data"
- Malec et al., EPJ Web Conf. 247 (2021) 12004
  "Development of fuel characterisation tool based on library interpolation"
- Shama et al. , Nucl. Eng. and Techn. 53 (2021) 2816
  "Uncertainty analysis of spent nuclear fuel decay heat calculations using SCALE modules"
- Rochman et al., Ann. Nucl. Energy 160 (2021) 108359
  "Analysis for the ARIANE GU1 sample: nuclide inventory and decay heat"
- Rochman et al., Eur. Phys. J. N 7 (2021) 14
  "Analysis for the ARIANE GU3 sample: nuclide inventory and decay heat"
- Rochman et al., Eur. Phys. J. N 7 (2021) 5
  "Fission yields and cross sections: correlated or not?"
- Ma et el., J. Anal. At. Spectrom. 35 (2020) 478
  "Non-destructive analysis of samples with a complex geometry by NRTA"
- Seidl et al., ATW - Int. J. Nucl. Power, 65 (2020) 353
  "On the potential to increase the accuracy of source term calculations for spent fuel from an industry perspective"

Status: 2020 - 2021

Task 2: Conferences and workshops

- Ahmed et al., Workshop on Machine Learning in Nuclear Science and Technology, Madrid, May 2021 (online)
  "Predictive models of computational bias in calculations of decay heats from spent nuclear fuel"
- Malec et al., PHYSOR 2020: Transition to a scalable nuclear future, Cambridge, April 2020
  "Development of Fuel characterisation tools based on library interpolation"
- Hernandez-Solis et al., PHYSOR 2020: Transition to a scalable nuclear future, Cambridge, April 2020
  "Boundary condition modelling effect on spent fuel characterisation and final decay heat prediction from a PWR assembly"
- Schillebeeckx et al., ESARDA Final Disposal Working Group Meeting, SCK CEN, Mol, February 2020
  "Fuel properties characterisation and related uncertainty analysis"
- Fiorito et al., Spent fuel workshop 2019, SCK CEN, Ghent, November 2019
  "ALEPH2 code for spent fuel characterisation"
- Schillebeeckx et al., Spent fuel workshop 2019, SCK CEN, Ghent, November 2019
  "EURAD: Task 2, Fuel properties characterisation and related uncertainty analysis"
- Kromar and Kurinčič, 28th Int. Conf. Nuclear Energy for New Europe, September 2019
  "Determination of the NPP Krško Spent Fuel Characteristics with the Serpent and SCALE Code Systems"
- Žerovnik et al., Int. Conf. on Mathematics Computational Methods to Applied Nucl. Sci. Eng., Portland, August 2019
  "Characterisation of spent PWR fuel for decay heat, neutron and gamma-ray emission: code comparison"

Status: 2020 - 2021

Task 2: others

- Reports
  - Schillebeeckx et al., JRC Technical Reports, EUR (EN) (2020)

    “A non-destructive method to determine the neutron production rate of a sample of spent nuclear fuel under standard controlled area conditions”
- Contribution to training courses
  - Schillebeeckx, at ANNETTE School on “Final stage of the nuclear lifecycle”, organised by KIT, Karlsruhe December 2019

    “Characterisation of SNF for intermediate storage and final disposal”
- Meetings
  - Shama, 1st EURAD Annual Event, March 2021

    “Validation and Uncertainty Analyses of SNF Characterization based on SCALE Code System”
  - Schillebeeckx, 1st EURAD Annual Event, March 2021

    “Thermal power produced by Spent Nuclear Fuel”
  - Schillebeeckx, 1st EURAD Annual Event, March 2021

    “Neutrons as a signature for the characterisation of spent nuclear fuel”
  - Rochman, Technical meeting on Spent Fuel Characterisation, IAEA, Vienna, November 2019

    “EURAD WP8, Spent fuel characterisation with the SERPENT and SCALE Code Systems”
  - Schillebeeckx, Technical meeting on Spent Fuel Characterisation, IAEA, Vienna, November 2019

    “EURAD WP8, Task 2: Spent fuel characterisation and related uncertainties”

Task 3: Education AND Training

- PhD

Efstathios Vlassopoulos (NAGRA) (École polytechnique fédérale de Lausanne, Thèse No 7619, 12 July 2021)
Subtask 3.1 Structural performance and mechanical properties investigation of spent nuclear fuel rods under static and dynamic bending loads

Miguel Cristóbal-Beneyto (UPM)
Subtask 3.1 Effect of hydrides on the mechanical behavior of nuclear fuel cladding

Okan Yetik (PSI)
Subtask 3.1 NEU-HERA: NEUtron radiography investigation of HydridEs Redistribution in Active zirconium claddings

Task 3: journal papers and reports

- Duarte et al., J. of Nucl. Mat. 557 (2021) 153284
  “Effect of the inner liner on the hydrogen distribution of Zircaloy-2 nuclear fuel claddings”
- Ruiz-Hervias et al., J. of Nucl. Mat. 544 (2021) 152668
  “Failure mechanisms in unirradiated ZIRLO® cladding with radial hydrides”
- Papaioannou et al., JRC Technical Reports, JRC122564, European Commission (2020)
  “Progress report, Part-A: Mechanical properties of spent nuclear fuel rods”

Status: 2020 - 2021

Task 3: Conferences and workshops

- Aguado et al., TopFuel Conference, Santander, Oct 2021
  “Assessment of FRAPCON predictive capabilities for spent fuel characterisation”
- Duarte et al., TopFuel Conference, Santander, Oct 2021
  “Neutron radiography imaging: A tool for determination of hydrogen distribution in unirradiated and irradiated fuel claddings”
- Yetik et al., TopFuel Conference, Santander, Oct 2021
  “Influence of scattering correction on quantitative hydrogen analysis of fuel claddings using high resolution neutron radiography”
- Ruiz-Hervias et al., TopFuel Conference, Santander, Oct 2021
  “Effect of radial hydrides on failure mechanisms in pre-hydrided cladding”
- Cristobal-Beneyto et al., Spanish Nuclear Society Annual Meeting, Granada, Oct 2021
  “Hydride reorientation of nuclear fuel cladding with internal pressure”
- Vlassopoulos et al., Safety of Nuclear Waste Disposal, Berlin, Nov 2021
  “Structural integrity investigations of spent nuclear fuel with finite element modeling”
- Zencker et al., IAEA CM on the CRP on Spent Fuel Characterization, virtual, Jun 2021
  “EURAD WP 8 (SFC) – Task 3: Behaviour of nuclear fuel and cladding after discharge”
- Zencker et al., IAEA First Research Coordination Meeting on Spent Fuel Characterization, virtual, Dec 2021
  “EURAD WP 8 (SFC) Task 3 – Update: Behaviour of nuclear fuel and cladding after discharge”

Status: 2020 - 2021

DisCo Project consortium and associated groups

16 beneficiares
Grant Agreement: 755443

4,5 years (1 June 2017-30 Nov 2021)

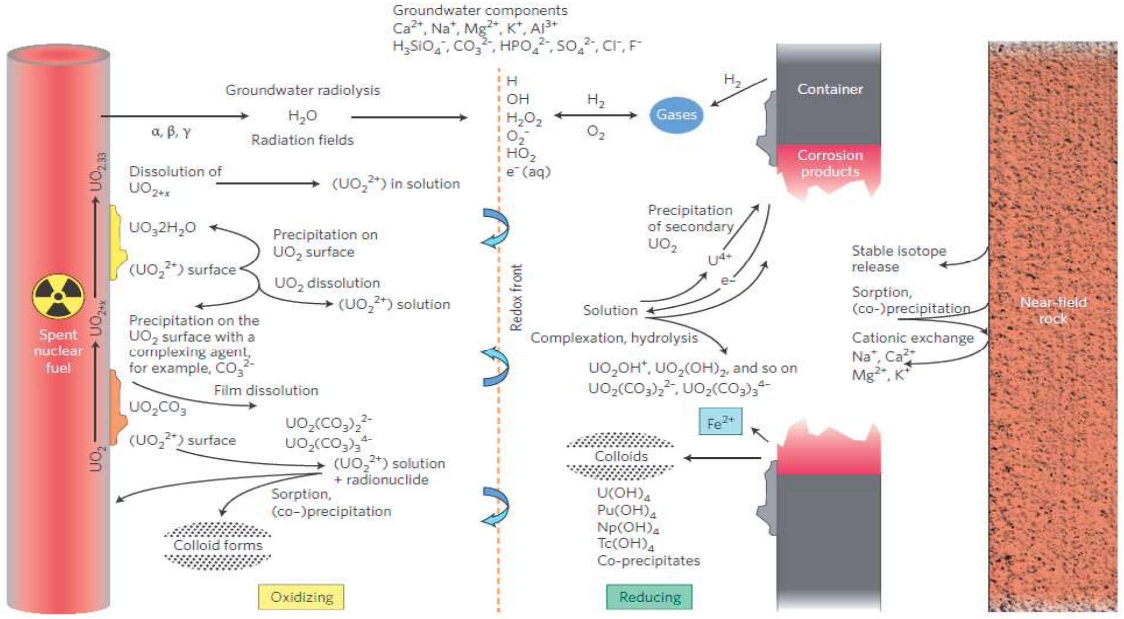
EUG: Sweden (SSM&SKB), Finland (Posiva), UK (RWM), Germany (BASE), Belgium (Ondraf/Niras & FANC), France (Andra), Spain (Enresa & CSN), Switzerland (Nagra& ENSI)

Associated Group: (green dots on map)
CV Rez (CH), LEI (LT),
MTA EK (HU), ICHTJ (PL),
EIMV (SI), Subatech(FR)

SKB
AMPHOS 21
KIT Karlsruher Institut für Technologie
JÜLICH FORSCHUNGSZENTRUM
Gobierno de España Ministerio de Ciencia e Innovación
Ciemat Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas
The University Of Sheffield.
SCK•CEN STUDIECENTRUM VOOR KERNENERGIE CENTRE D'ETUDE DE L'ENERGIE NUCLEAIRE
VTT
Studsvik
NATIONAL NUCLEAR LABORATORY
UNIVERSITY OF CAMBRIDGE
PAUL SCHERRER INSTITUT PSI
cea
eurecat Centre Tecnològic de Catalunya
ARMINES
European Commission

Spent fuel dissolution in repository conditions

- Controls the slow release of the majority of radionuclides from the fuel, which are retained in the UO2 matrix



(From Ewing 2015)

DisCo objectives, concept & methodology

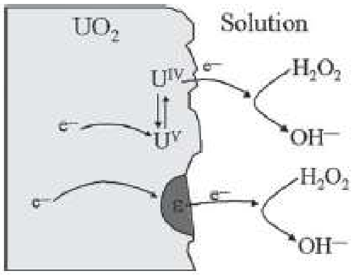
1) Improve **understanding of spent fuel matrix dissolution** in repository conditions

2) Test **modern nuclear fuel types (doped & MOX)** for comparison with conventional fuels:

- Both real spent fuel and synthesized model materials

+ Disseminate knowledge through training and mobility measures

- Long-term matrix dissolution in reducing environments
- Alpha radiolysis, hydrogen peroxide and the hydrogen effect
- Deeper understandning of mechanisms for improved modelling
- Adding elements other than U to the fuel matrix will affect the electrochemical properties of solid: how important is this for the overall dissolution process and radionuclide release rates?
- The project rests on three pillars:
  1) dissolution experiments with spent nuclear fuel
  2) dissolution experiments with synthesized model materials and
  3) chemical modelling of the systems



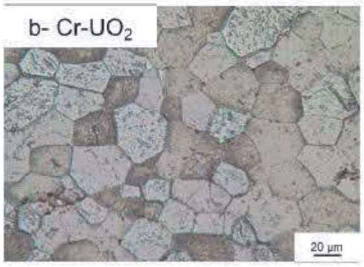
DisCo WP 2 - Samples and experimental systems

Spent fuel, doped UO_2, and UO_2

- Hot cells:
  Cr- & Cr+Al-doped fuel (Adopt), MOX and standard fuel. BU ~40 -60 MWd/kgHM
  +Failed fuel from UK
- Alpha labs: UO2 with ^{238}Pu, ^{238}Pu+Cr
- U/Th Laboratories:
  UO_2
  UO_2+Cr
  UO_2+Cr+Al
  UO_2+Gd
  $(U, Th)O_2$

Results

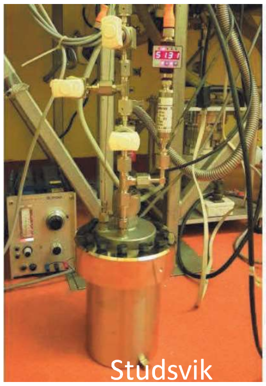
- Prepared and characterized samples for dissolution experiments
- Sample synthesis procedures developed
- Enhanced understanding of the UO_2 solid state - effects of additives on crystal structure, U valence, oxide lattice defects and microstructure



Example of Cr-doped UO_2 prepared by SCK·CEN.

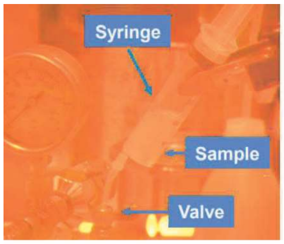
DisCo WP 3 – Dissolution of spent fuel

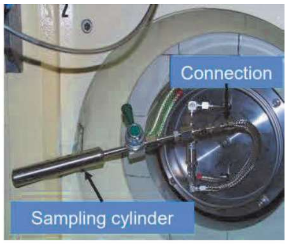
Experimental set-ups		
Water type (reductant):	Bicarbonate Water (H2)	Young Cement Water with Ca (H2)
Reducing (H2, Ar/N2/H2 mix, or anoxic with corroding Fe)		
UO2 spent fuel	√	√
UO2 spent fuel w Cr+Al	√	
UO2 spent fuel w Cr	√	
MOX spent fuel	√	
Oxidizing/Anoxic(Ar), H2O2, or Air		
UO2 spent fuel, Air		√
MOX spent fuel, Ar	√	



Controlled atmosphere in autoclaves
Hot cells - remote handling via manipulators - Sampling procedures

Results from experiments in bold given here (next)

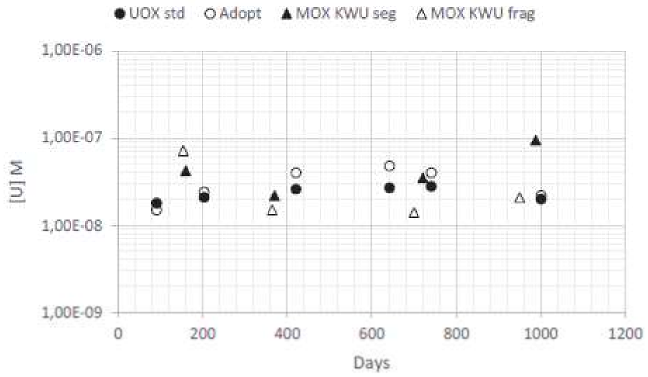


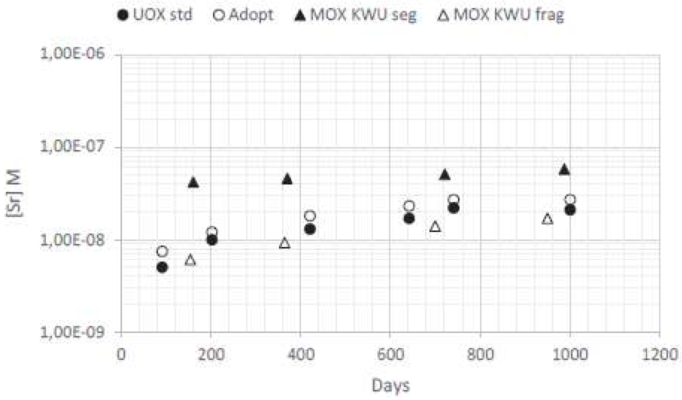


DisCo WP 3 – Selected Results

Reducing environment with hydrogen in the gas phase – matrix dissolution

Only data after 90 days are shown – system stabilizes after ca 1 year – long term experiments required for matrix dissolution studies. Data from Studsvik (UOX and Adopt) and KIT (MOX).

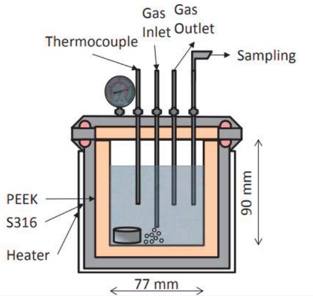




U concentration fairly stable but a bit higher than expected. Sr release continues slowly, approches zero (Studsvik)
NO significant difference in matrix dissolution behaviour and radionuclide release for UOX , Adopt, and MOX.

DisCo WP 4 – Dissolution of model materials

Experimental set-ups				
Water type (reductant):	Bicarbonate Water (H2)	Young Cement Water with Ca (H2)	Natural Groundwater (Fe)	Synth. Callovo-Oxfordian water (+/-Fe)
Reducing (H2, Ar/N2/H2 mix, or anoxic with corroding Fe)				
UO2	**√**	√	√	
UO2 w Gd	**√**	√		
UO2 w Cr	**√**	√	√	
UO2 w Cr+Al	**√**	√		
UO2 w 238Pu/233U	**√**	√	√	
UO2+Cr w 238 Pu	**√**	√	√	
(Pu,U)O2				√
Oxidizing/Anoxic(Ar), H2O2, or Air				
UO2	√			
UO2 w Cr	√			
(U,Th)O2	√			√

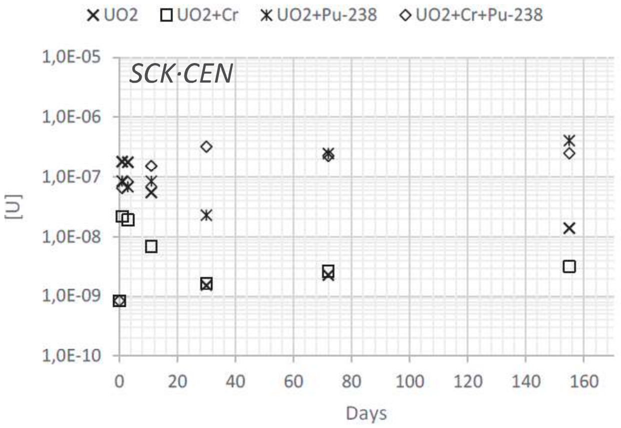


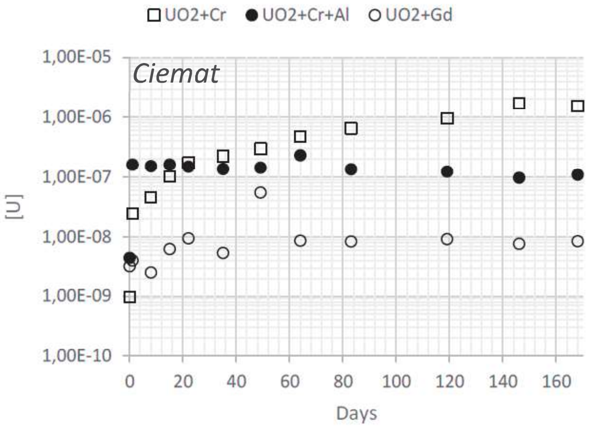
Autoclaves in glove box, Ciemat

Controlled atmosphere in glove boxes & autoclaves
Samples simpler than spent fuel allows increased complexity in aqueous phase
Sampling & analysis of aqueous phase, & ”post mortem” analyses of solid phase
Results from experiments in bold here (next)

DisCo WP 4 – Selected Results

Uranium concentration in aqueous solution vs time for undoped and doped UO2 in experiments with hydrogen in the gas phase. Some air contamination is noted for Ciemat data.





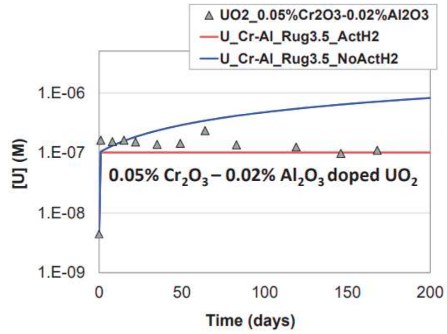
Effects of alpha doping seen clearly in the SCK CEN data, but no clear effect of the Cr in the long run. Of the different dopants, a clear beneficial effect of Gd is seen, the combination of Cr and Al may also be inhibiting U oxidation, while the effect of Cr appears to be insignificant in that respect.

DisCo WP 5 – Selected Modelling Results

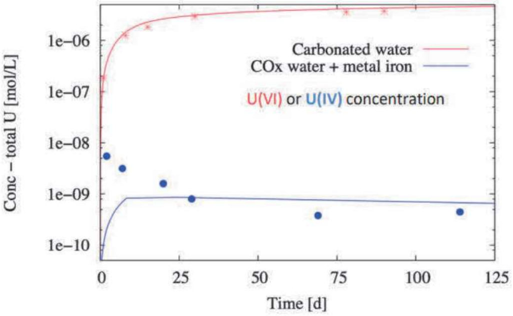
Cr-doping does not affect the oxygen potential of the irradiated fuel above burnups of ca 20 MWd/kgU . Oxygen potential in Cr-doped fuel should therefore not cause change in segregation of fission products.

Improvements of the Matrix-Alteration-Model (MAM): now implemented with the iCP (Comsol-PhreeqC coupling interface), able to reproduce data from DisCo experimental systems,
for example effect of Cr+Al (Ciemat data)

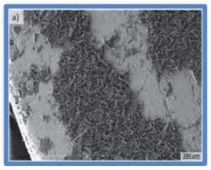
The MOX experimental system was modelled using the CHESS-HYTEC approach. Results show that Fe keeps U and Pu concentrations very low: The corroding iron in this groundwater system appears enough to keep actinides reduced, keeping the dissolution rate low.



MAM with iCP & data Ciemat



$$\frac{1}{2}H_2O_2 + Fe^{2+} \rightarrow Fe^{3+} + OH^-$$



DisCo Conclusions & Outlook

- Similar matrix dissolution behaviour for MOX, doped fuels (with Cr and Al as dopants), and standard fuel types.

→ Confirms they can be included in the plans and safety assessments for spent nuclear fuel repositories.

- Reducing environment (hydrogen overpressure) & presence of corroding iron in the repository can keep the dissolution rate of spent nuclear fuel low.
- Improved understanding of
  1) mineral chemistry and microstructure of the uranium dioxide matrix
  2) repository systems, from thermodynamics of the spent fuel to the effect of a redox front on the radionuclide transport in the repository.

- Examples of remaining knowledge gaps :
  Hydrogen – Some remaining questions on how hydrogen acts on the system;
  Uranium (IV) - Impact of recrystallization of the fuel surface when at the solubility level of U(IV)
  Iron - Effect of Fe (II)-concentrations over time and different repository settings
  Chromium: valence of Cr in the uranium oxide needs to clarified. Expected valence +3 **–** but some results indicate +2 or even +1.

- Outlook:
  - Other additives: New fuel types under the ATF umbrella need investigation (UO_2 with oxide/metallic/ceramic additives)
  - Doping effect from four-valent elements: How do eg Th, Pu influence the oxidative dissolution?
  - Grain boundaries & IRF : How do the grain boundaries participate in the dissolution process?