WP 8 - SAREC

Release of **sa**fety **re**levant radionuclides from spent nuclear fuel under deep disposal **c**onditions

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PARTNERS

26 Participants

CEA, Ciemat, Subatech (IMT Atlantique), ICSM, Uni Montpellier, Eurecat, UPC, FZJ, HZDR, IRSN*, ENSMP, Energorisk, KIT, Amphos 21, KTH, Ondraf/Niras, Raten, SCK CEN, SKB, Studsvik (subcontractor), Uni Helsinki, VTT, JRC-Karlsruhe, Lancaster, PSI, Bristol

End User Group

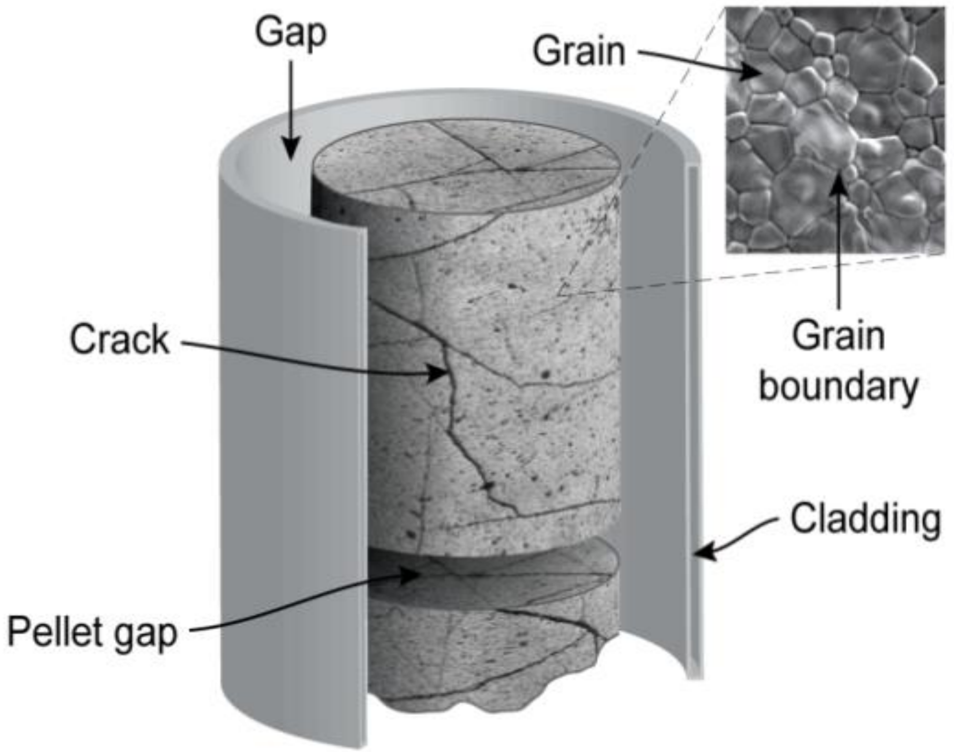
Andra, Enresa, Nagra, NWS, Posiva, Surao, SKB, BGE

Associated/Other interested parties

SSM, PNNL, LEI, UJV Rez, FTMC, ENUSA

BACKGROUND (I)

- **Spent Nuclear Fuel as High Level Waste**
  - Radionuclide inventory in spent fuel matrix, gap & grain boundaries, structural components (metals)
  - Radionuclide release: Matrix release, « Instant Release» , «Corrosion release »
  - Matrix release mainly via radiolytic oxidation of matrix UO_2
  - « Instant Release Fraction » = IRF, released as a pulse – relevant for some highly mobile fission products
  - « Corrosion Release Fraction » released at the rate of metal corrosion
  - IRF for Iodine and Cesium correlated to fission gas (eg Xe) release, FGR: assumed to behave similarly 1) during reactor operation 2) at water contact in repository
  - IRF & FGR important to quantify and understand in relation to overall radionuclide release rate



BACKGROUND (II)

- **Previous EU project with focus on IRF**
  - FP7 project FIRST Nuclides: Fast / Instant Release of Safety Relevant Radionuclides from Spent Nuclear Fuel, run between 2012-2014
  - Focus on leaching of high burnup fuel : 45 to 70 GWd/tHM
  - Importance of linear power rate (W/cm) as this relates to temperature and rate of diffusion
  - Results available in database for general use
- **Previous EU project with focus on matrix release**
  - Horizon 2020 project DisCo : Modern spent fuel dissolution and chemistry in failed container conditions
  - Focus on effects of dopants: Cr, Cr+Al, Gd, & MOX-fuel
  - Involved leaching studies on spent nuclear fuel, synthesis and leaching of model materials, and modelling
- **Previous EU project with focus on spent fuel characterization**
  - Horizon 2020 Eurad-1 WP SFC: Spent Fuel characterisation and evolution until disposal
  - Procedures to accurately determine the source term of irradiated spent fuels.
  - Characterisation techniques to understand the physiochemical evolution of irradiated spent fuels (pellets and cladding)

FIRST Nuclides

DisCo
Modern Spent Fuel Dissolution and
Chemistry in Failed Container Conditions

eurad2

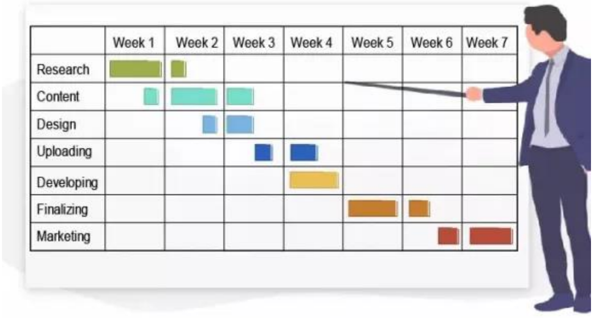
OBJECTIVES, DRIVERS AND EXPECTED OUTCOMES

- **State-of-knowledge**
  - Knowledge and data for the Spent Nuclear Fuel (Domain 3.1.1), builds on many previous EU projects and years of research, documented in Eurad-1 SoK
    (State of the Knowledge Report - Spent Nuclear Fuel (Domain 3.1.1) (DOI 10.5281/zenodo.7024752)
  - IN-CAN Processes 2000-2003, SFS 2001-2004, NF-PRO 2004-2007, MICADO 2006-2009, SKIN 2011-2013, REDUPP 2011-2014 ...
- **Objectives**
  - Improved quantification and mechanistic understanding of the release of safety relevant radionuclides covering most representative types of spent nuclear fuel (SNF)
  - Fuel evolution both prior and posterior to contact with groundwater to better predict the radionuclide source term for post-closure safety assessment
- **Strategic Research Agenda - Drivers & expected outcomes**
  - <u>Implementation Safety</u>:
    Radionuclide release data that allows the needed re-evaluation of the current approaches to release of safety relevant radionuclides in post closure safety assessments
  - <u>Scientific Insight:</u>
    Data for deriving an updated and improved mechanistic understanding of radionuclide release processes
  - <u>Knowledge Management:</u>
    Completion of SNF dissolution database, thus enhancing knowledge management and transfer between the participating organisations (and beyond)

OVERVIEW OF TASKS

	Task title	Task leaders	
		Main Task leader	Co Leader if applicable
1	Management/coordination of the WP	*Lena Z Evins , SKB, WMO, Sweden*	
2	Knowledge Management	*Olga Riba, AMPHOS 21, RE, Spain*	
3	IRF/FGR Performance of Spent Nuclear Fuel	*Michel Herm, KIT, RE, Germany*	
4	Role of Grain Boundaries in Spent Fuel Corrosion	*Roberto Gaggiano, ONDRAF/NIRAS, WMO, Belgium*	
5	Studies on Model Materials	*Nieves Rodriguez-Villagra, CIEMAT, TSO, Spain*	
6	Mechanistic modelling	*Mats Jonsson, KTH, RE, Sweden*	*6.1 Mats Jonsson, KTH, RE Sweden* *6.2 Janne Heikinheimo, VTT, TSO, Finland*

TASK 1 – MANAGEMENT / COORDINATION OF THE WP



- **Lead: SKB**
- **Overall management of the WP**
  - Scientific-technical coordination, monitoring, reviewing progress
  - Lead: SKB, WP Board consisting of Task leaders
- **Subtask 1.1 S/T coordination**
  - Check progress against planned milestones and deliverables
  - WP Board will meet regularly and communicate progress to PMO and other stakeholders
- **Subtask 1.2 Dissemination/outreach/impact**
  - Organise annual WP meetings
  - Contribute to EURAD-2 Newsletters and website
- **Subtask 1.3 Quality control**
  - Assessing work against key performance indicators (KPI)
  - Data management, open access requirements, and risk management

TASK 2 - KNOWLEDGE MANAGEMENT

Author ID	Database ID	Temperature (°C)	Solution composition	measured FGR (%)	Type of data	Origin of data	Cs (%)	Sr (%)	I (%)	Rb (%)	Tc (%)	Mo (%	Ref.
ATM-105	ATM105-Pl-34-B	25	borate buffer		IRF	graph	1,5		2,5				Gray (1999)
ATM-105	ATM105-Pw-34-B	25	borate buffer		IRF	graph	1,0		5				Gray (1999)
ATM-106	ATM106-Pl-43-DIW	25	DIW		IRF	graph	2	0,11			0,13		Gray(1995)
ATM-106	ATM106-Pw-43-HCl	25	0.1 M HCl		IRF	graph	0,5	0,03					Gray(1995)
ATM-106	ATM106-Pl-43-B	25	borate buffer		IRF	graph	2,0		0,1				Gray (1999)
ATM-106	ATM106-Pw-43-B	25	borate buffer		IRF	graph	0,5		8,5				Gray (1999)
ATM-106	ATM106-Pl-46-DIW	25	DIW		IRF	graph	2,5	0,02			0,01		Gray(1995)
ATM-106	ATM106-Pw-46-HCl	25	0.1 M HCl		IRF	graph	1,0	0,13			0,01		Gray(1995)
ATM-106	ATM106-Pl-46-B	25	borate buffer		IRF	graph	2,5		1,2				Gray (1999)
ATM-106	ATM106-Pw-46-B	25	borate buffer		IRF	graph	1,0		8,0				Gray (1999)
ATM-106	ATM106-Pl-50-DIW	25	DIW		IRF	graph	6,5	0,1			0,05		Gray(1995)
ATM-106	ATM106-Pw-50-HCl	25	0.1 M HCl		IRF	graph	1	0,07			0,12		Gray(1995)
ATM-106	ATM106-Pl-50-B	25	borate buffer		IRF	graph	6,5		15				Gray (1999)
ATM-106	ATM106-Pw-50-B	25	borate buffer		IRF	graph	1,0		7,6				Gray (1999)
11-01	BWR-27	20-25	GW		IRF	table	0,233	0,00566		0,0647	0,00489	0,00623	Forsyth(1997)
11-02	BWR-30,1	20-25	GW		IRF	table	0,00296	0,00499		0,0794	0,00502	0,00622	Forsyth(1997)
11-03	BWR-32,7	20-25	GW		IRF	table	0,329	0,00841		0,0732	0,0029	0,00357	Forsyth(1997)
11-04	BWR-34,9	20-25	GW		IRF	table	0,488	0,0132		0,119	0,00426	0,00533	Forsyth(1997)

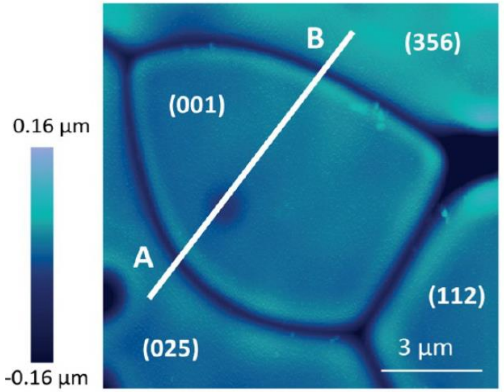
- **Lead: Amphos 21**
  - Knowledge transfer to the EURAD-2 community and beyond through the EURAD-2 KM programme.
- **Subtask 2.1: Knowledge capture**
  - Knowledge relevant to the WP, gained prior to EURAD-2 and extended during this WPs progress
  - Initial version of the State-of-the-Art report prepared at the beginning of the WP
  - At the end this report will be updated integrating the key findings
- **Subtask 2.2: Knowledge transfer**
  - Specific activities to transfer knowledge to interested parties
  - Online training, face-to face-training, e-learning materials, workshops, posts for social media, summary sheets, videos, guidance …
- **Subtask 2.3: Database and Training Materials**
  - Completion and rationalization of an updated, traceable and versatile database of SNF dissolution
  - Implementation of a web interactive interface for the database
  - Training materials (and conduct the training, if needed) on modeling accumulation of gaseous fission products during burnup and storage involving for example SCALE calculation suite (and/or MCNP)

TASK 3 - IRF/FGR PERFORMANCE OF SPENT NUCLEAR FUEL

- **Lead: KIT**
- **Leaching and post-leaching characterization experiments at hot cell facilities**
  - EURECAT (Spain), FZJ (Germany), JRC-Karlsruhe (EC), KIT (Germany), RATEN (Romania), SCK·CEN (Belgium), Studsvik Nuclear (Sweden)
  - Segments/fragments from UOX and MOX fuel rods with burnup of 29-58 GWd/t_{HM} & FGR 2.5 %-13 %
  - CANDU fuel with burnup 7-8 GWd/t_{HM} & FGR 3-7 %
- **Experimental conditions**
  - Initial oxygen free, anoxic or reducing conditions in autoclaves with pure Ar or Ar/H_2 or pure H_2 in the gas phase
  - Aqueous solutions mimic groundwaters for granitic environments (« BIC ») or young cement water with Ca (« YCWCa »).
  - Sampling of gas and liquid phases at regular intervals during the leaching time
- **Data collected**
  - Evolution of radionuclide concentrations in aqueous solution
  - Fission gases released during leaching
  - Allows study of IRF in connection to released fission gases, matrix dissolution, and influence of aqueous solution composition
  - When possible, analyses of the solid before and after leaching will allow evaluation of effect of leaching on solid state characteristics

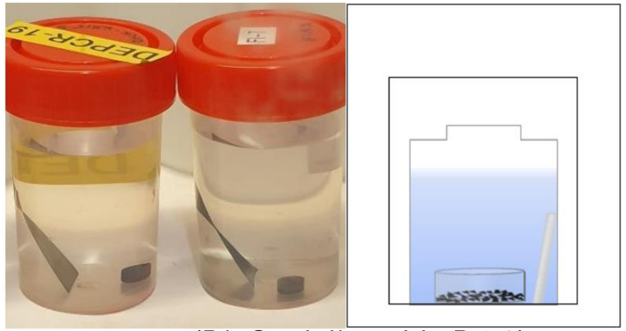
TASK 4 - ROLE OF GRAIN BOUNDARIES IN SPENT FUEL CORROSION

- **Lead: Ondraf/Niras**
- **Spent nuclear fuel and model materials studied**
  - SNF leached as it is smashed using sonolysis
  - Model materials doped with fission products
- **Leaching, heating and solid state analyses**
  - SNF leached as it is smashed – releasing radionuclides from grain boundaries
  - Model materials heated and studied for release of I & Cs
  - Evolution of microstructures and radionuclide distribution
  - Solid state analyses using various techniques: SEM & ESEM, TEM, EBSD, EDS, EPMA, Raman microscopy (µ-Raman), and synchrotron based and laboratory-based methods (XRD, XAS, XES), AFM and XPS



(Corkhill et al 2014)

TASK 5 - STUDIES ON MODEL MATERIALS



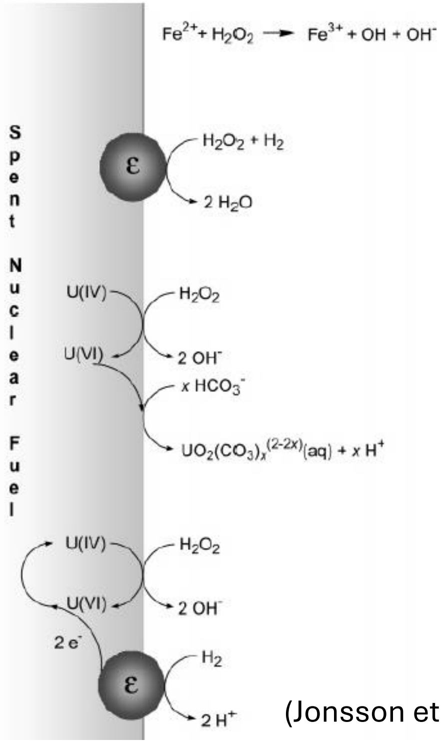
(DisCo deliverable D4.3)

- **Lead Ciemat**
- Studies on model materials simulating UOX fuel
  - Simfuel, FP-doped UO_2 and Zr-doped UO_2, and ATF*s: Cr-doped UO_2 and (Cr,FP)-UO_2)
  - Most model materials will be synthesized specifically for the project
- Leaching experiments will be performed under reducing, anoxic and oxic conditions
  - Evolution under presence of water (leaching), and under radiation (radiolysis).
  - Static dissolution & complementary electrochemically accelerated dissolution
  - « BIC » & « YCWCa » & Finnish synthetic ground waters (« FIN »)
  - Radiation effects either by external radiation or mimicked by addition of H_2O_2

*Accident Tolerant Fuels /Advanced Technology Fuels

TASK 6 - MECHANISTIC MODELLING

- **Lead: KTH**
- **Subtask 6.1: Radionuclide release modelling (Lead KTH)**
  - Radiation-induced dissolution of UO2 based on mechanistic descriptions of surface reactions
  - Semi-empirical modeling of matrix dissolution and IRF,
    to explore effects of varying leaching conditions and SNF structures
  - Upscaling & benchmarking

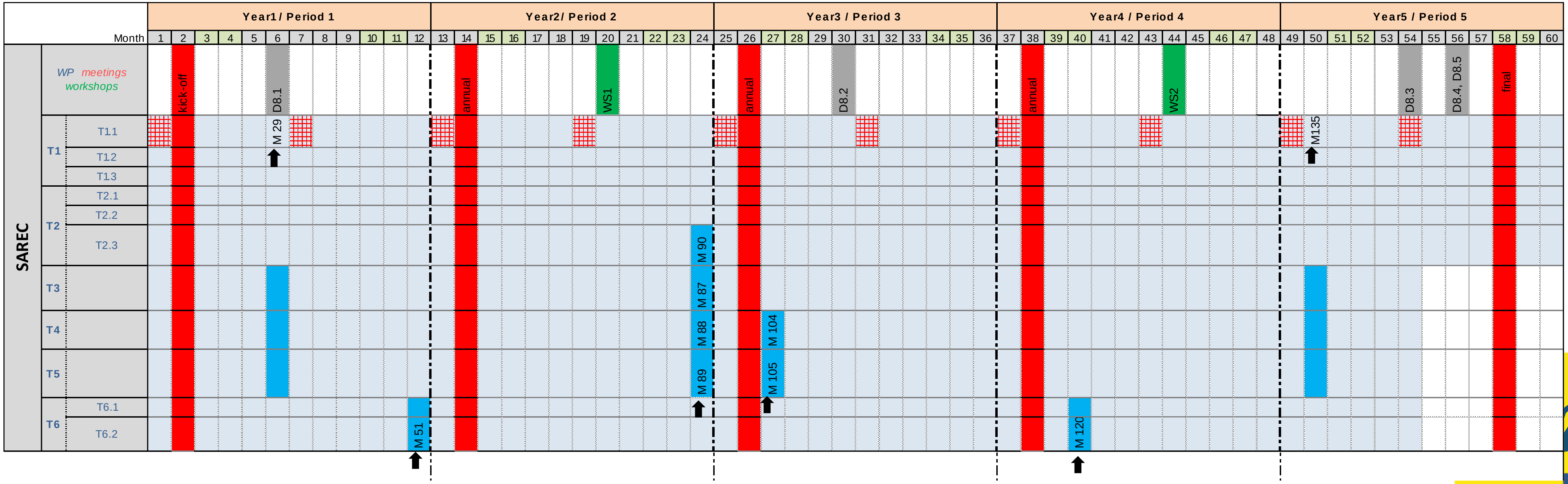


(Jonsson et al 2007)

- **Subtask 6.2: Source term and FGR modelling (Lead VTT)**
  - Simulations and statistical fuel performance analyses of FGR will be performed for VVER and/or EPR fuel
  - Development of iodine and cesium release correlation coupling with fission gas release
  - Radionuclide inventory calculations for UO2 and MOX supporting the IRF & matrix dissolution studies

GANTT CHART

- **Active during the five years of Eurad-2**
  - 1 kick-off & 4 annual WP meetings, separate Task workshops, biannual WP Board meetings, 2 WP workshops
  - 5 deliverables and 10 milestones. 1st year: Initial State-of-the-Art report, Sample data collected and confirmation of sample availability , Model development initiated and first set of in-put parameters defined



SUMMARY & IMPACTS

- **We aim to improve**
  - Quantification and mechanistic understanding of the release of safety relevant radionuclides
  - Insight into fuel evolution both prior and posterior to contact with ground water
  - Definition of initial state and radionuclide source term for post-closure safety assessment
- **Connecting experiments, modelling, and knowledge management**
  - Spent nuclear fuel leaching combined with studies of model systems for deeper process understanding
  - Modelling to implement current and new understanding of mechanisms
  - Database and updated State-of-the-Art Report to ensure knowledge capture and knowledge transfer
- **Impacts guided by drivers**
  - *Implementation Safety* :
    Contributing to long-term safety of deep geological repositories via improvement of current approaches to release of safety relevant radionuclides in post closure safety assessments.
  - *Scientific Insight* :
    Advancing state of the art with regards to spent nuclear fuel radionuclide inventory, microstructure and mechanistic understanding of radionuclide release processes
  - *Knowledge Management* :
    Enhancing knowledge management and transfer between organisations, Member States and generations via SNF dissolution database and documenting the advance of State-of-the-Art.