

## WP 8 - SAREC

Release of **safety relevant** radionuclides from spent nuclear fuel under deep disposal **conditions**

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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

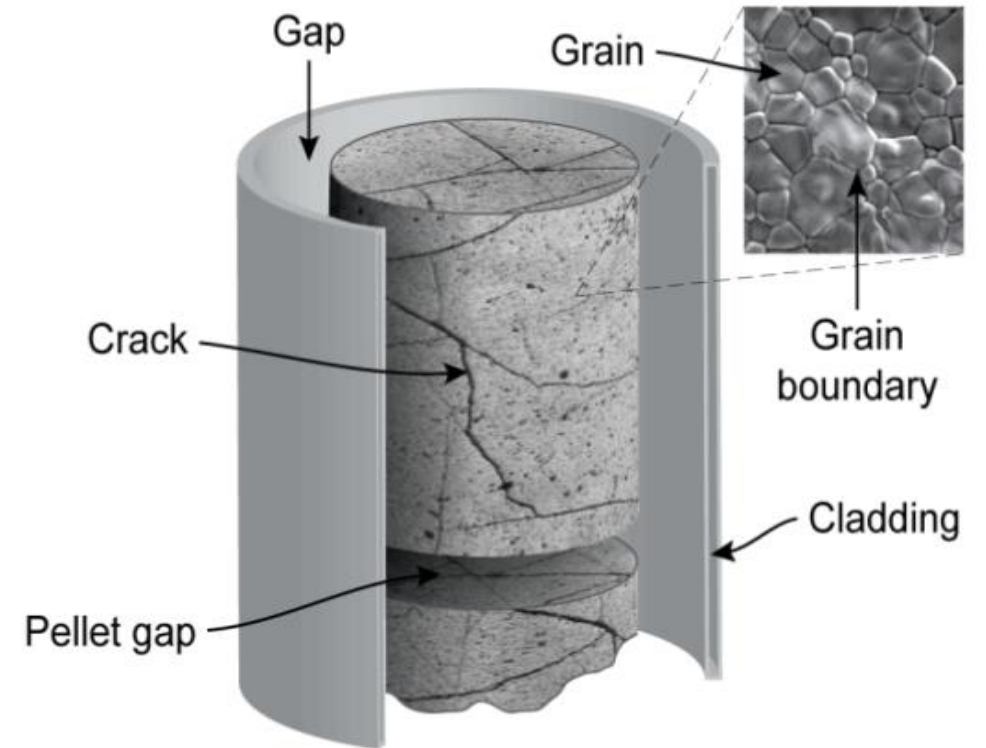
## Associated/Other interested parties

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)



# 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**

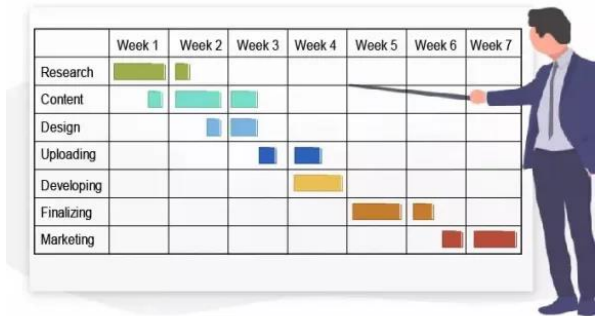
- Implementation Safety:  
Radionuclide release data that allows the needed re-evaluation of the current approaches to release of safety relevant radionuclides in post closure safety assessments
- Scientific Insight:  
Data for deriving an updated and improved mechanistic understanding of radionuclide release processes
- Knowledge Management:  
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                | <i>Lena Z Evins , SKB, WMO, Sweden</i>               |  |
| 2 | Knowledge Management                             | <i>Olga Riba, AMPHOS 21, RE, Spain</i>               |  |
| 3 | IRF/FGR Performance of Spent Nuclear Fuel        | <i>Michel Herm, KIT, RE, Germany</i>                 |  |
| 4 | Role of Grain Boundaries in Spent Fuel Corrosion | <i>Roberto Gaggiano, ONDRAF/NIRAS, WMO, Belgium</i>  |  |
| 5 | Studies on Model Materials                       | <i>Nieves Rodriguez-Villagra, CIEMAT, TSO, Spain</i> |  |
| 6 | Mechanistic modelling                            | <i>Mats Jonsson, KTH, RE, Sweden</i>                 | <i>6.1 Mats Jonsson, KTH, RE Sweden</i><br><i>6.2 Janne Heikinheimo, VTT, TSO, Finland</i> |

# 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

- **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**

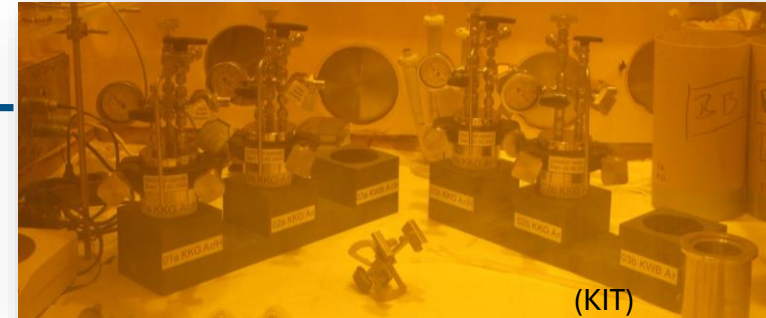
- Completion and rationalization of an updated, traceable and versatile database of SNF dissolution
- Implementation of a web interactive interface for the database
- Training materials (and conduct the training, if needed) on modeling accumulation of gaseous fission products during burnup and storage involving for example SCALE calculation suite (and/or MCNP)

| 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-Pi-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-Pi-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-Pi-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-Pi-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-Pi-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-Pi-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-Pi-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 | 0,00333 | Forsyth(1997) |



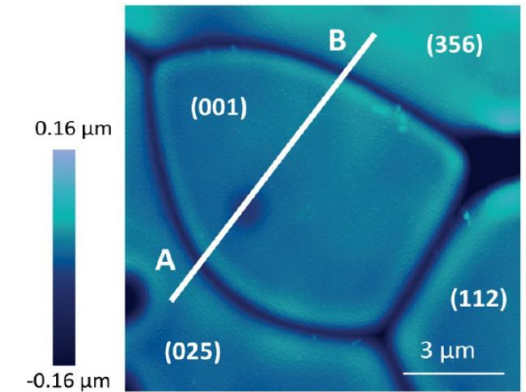
## TASK 3 - IRF/FGR PERFORMANCE OF SPENT NUCLEAR FUEL

- **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<sub>HM</sub> & FGR 2.5 %-13 %
  - CANDU fuel with burnup 7-8 GWd/t<sub>HM</sub> & FGR 3-7 %
- **Experimental conditions**
  - Initial oxygen free, anoxic or reducing conditions in autoclaves with pure Ar or Ar/H<sub>2</sub> or pure H<sub>2</sub> 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

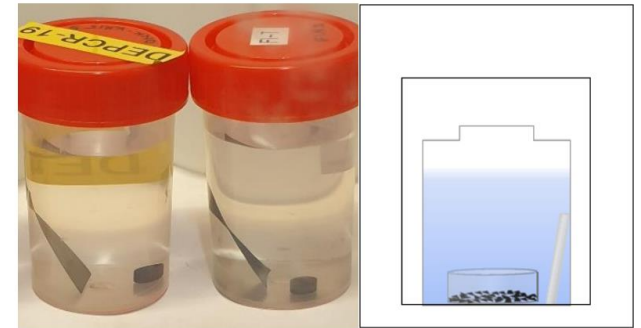
- **Lead: Ondraf/Niras**
- **Spent nuclear fuel and model materials studied**
  - SNF leached as it is smashed using sonolysis
  - Model materials doped with fission products
- **Leaching, heating and solid state analyses**
  - SNF leached as it is smashed – releasing radionuclides from grain boundaries
  - 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 ( $\mu$ -Raman), and synchrotron based and laboratory-based methods (XRD, XAS, XES), AFM and XPS



(Corkhill et al 2014)

## TASK 5 - STUDIES ON MODEL MATERIALS

- **Lead Ciemat**
- Studies on model materials simulating UOX fuel
  - Simfuel, FP-doped  $\text{UO}_2$  and Zr-doped  $\text{UO}_2$ , and ATF\*s: Cr-doped  $\text{UO}_2$  and (Cr,FP)- $\text{UO}_2$
  - 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  $\text{H}_2\text{O}_2$

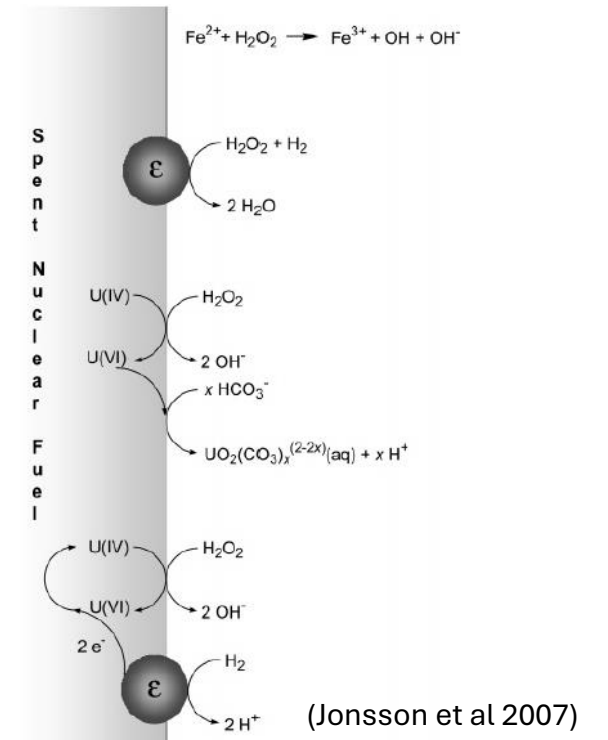


(DisCo deliverable D4.3)

\*Accident Tolerant Fuels /Advanced Technology Fuels

## TASK 6 - MECHANISTIC MODELLING

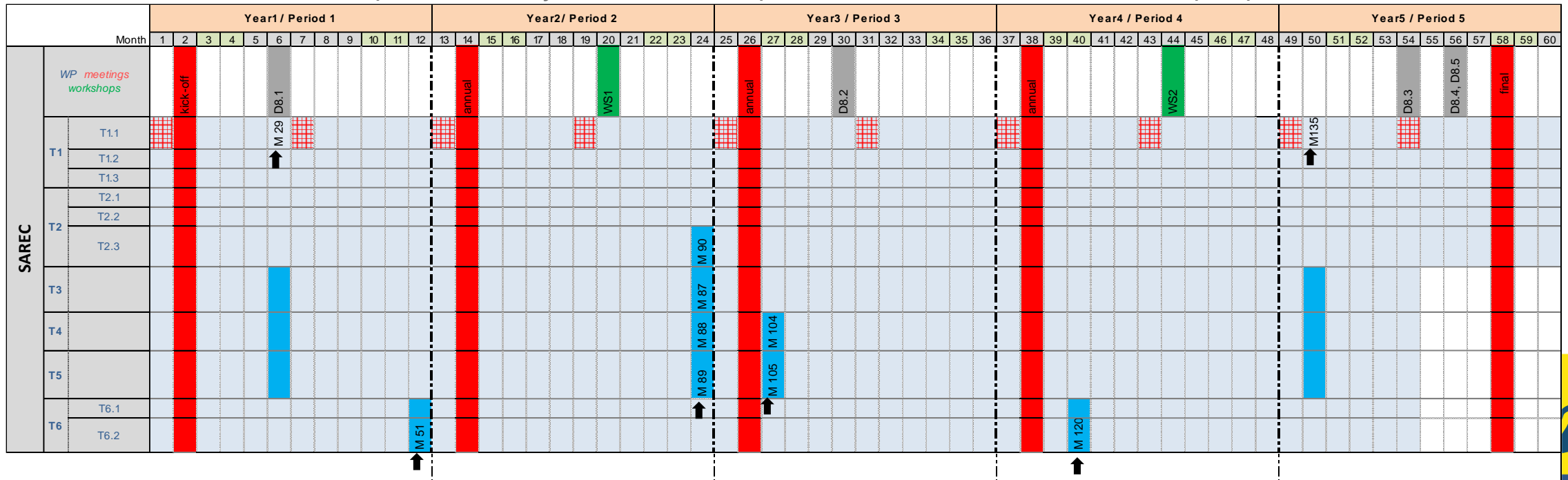
- **Lead: KTH**
- **Subtask 6.1: Radionuclide release modelling (Lead KTH)**
  - Radiation-induced dissolution of UO<sub>2</sub> based on mechanistic descriptions of surface reactions
  - Semi-empirical modeling of matrix dissolution and IRF, to explore effects of varying leaching conditions and SNF structures
  - Upscaling & benchmarking
- **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 UO<sub>2</sub> 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.