EUROPEAN COMMISSION

Community research

FIRST-Nuclides

(Contract Number: 295722)

DELIVERABLE (D-N°:5.12)

Presentation of the project at the Spent Fuel Workshop and preparation of a full paper

Author(s): **KIT-INE, AMPHOS 21**

Reporting period: e.g. 01/01/12 – 30/06/13

Date of issue of this report: **30/12/12**

Start date of project: **01/01/11** Duration: 36 Months

FIRST Nuclides

EURATOM

DISTRIBUTION LIST

Name	Number of copies	Comments
Mr. Christophe Davies (European Commission)	One electronic copy submitted via participant portal	
All consortium members and European Commission	One electronic copy available on the public area of the FIRST-Nuclides webportal	

Project co-funded by the European Commission under the Seventh Euratom Framework Programme for Nuclear Research &Training Activities (2007-2011)		
Dissemination Level		
PU	Public	X
RE	Restricted to a group specified by the partners of the FIRST-Nuclides	
CO	Confidential, only for partners of the FIRST-Nuclides project	

1. Introduction

One of the objectives of WP5 is the communication and dissemination of knowledge generated within the project via different channels. Presentation of the project and its results in international conferences is one of the main dissemination channels.

This deliverable includes three contributions at different international conferences:

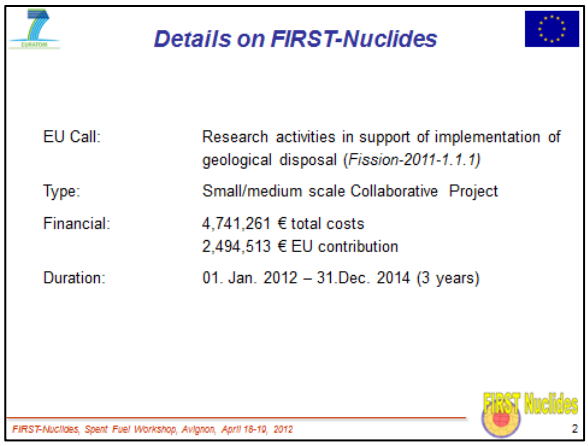
i. Oral presentation at the Spent Fuel Workshop (April 2012, Avignon, France)

ii. Oral presentation at ATALANTE 2012 (September 2012, Montpellier, France)

iii. Paper submitted at the EURADISS Workshop (October 2012, Montpellier, France) that will be published as Proceedings.

Both oral presentations were given by the coordinator of the project Bernhard Kienzler. An overall view of the organization and the work performed in the project are presented in all the contributions.

2. Oral presentation at the Spent Fuel Workshop



Definition of instant / fast release

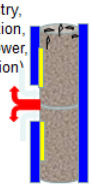
Project related definition or PA related definition

RN release by matrix corrosion controlled by radiolysis, hydrogen, Cl-....

↔

initial (fast) release of RN controlled by chemistry, spatial distribution, burn-up, lin. power, reactor operation

- gap
- large cracks
- grain boundaries
  - close to surface
  - rim
  - grain boundaries along cracks



Further aspects:

- **PA term:** „instant" release
- **Experimental / lab related:** fast release (for a specific fuel sample) controlled by: sample *preparation, fragment size, presence of cladding, duration of experiments, ...*

Rationale of FIRST-Nuclides

- Previous EU projects (MICADO, NF-Pro, SFS, ...)
  - Improvement in understanding.
    - high *burn-up UO_2, linear power, temperature, ramping, ...*
  - Relationship FGR and release of non-gaseous FPs
    - *gases, ^{135}Cs, ^{129}I, ^{14}C compounds, ^{79}Se, ^{99}Tc and ^{126}Sn.*
  - Grain boundary effects.
  - Chemical speciation of relevant elements.
- Vision Report and SRA of the IGD-TP
  - Key topic: Waste forms behaviour, in particular high burn-up spent UO_2 fuels.
- Recommendations by the End Users (kick off meeting)
  - Definition of instant – rapid – fast release and delineation to long-term RN release processes.
  - Up-scaling from lab sample to rod/assembly.
  - Effect of geochemical conditions.

The Consortium (1)

1. Partners / Beneficiaries

KIT, AMPHOS 21, ITU, JÜLICH, PSI, SCK•CEN, Subatech, CTM, Hungarian Academy of Sciences, Centre for Energy Research, Studsvik

2. Associated Groups (AG)

Groups participating at their own costs with specific RTD contributions or particular information exchange functions, or mobility measures (for European AGs only)

Los Alamos National Laboratory, Sandia National Laboratories, CEA, National Nuclear Laboratory, NDA, POSIVA

The Consortium (2)

3. End-user group:

Representatives:

ANDRA, Bundesamt für Strahlenschutz, enresa, nagra, ONDRAF/NIRAS, SKB

Roles and Tasks within CP FIRST-Nuclides:

- Review the work program
- Review the scientific-technical progress and contributions to the proceedings
- Assess the project status and provide recommendations for the work program
- Review the plan for dissemination of the final results.

Work Programme:

➔ 6 Workpackages

WP 1: Samples and tools

Selection, characterization and preparation of and set-up of tools for handling and transportation of the highly radioactive material

WP leader: Volker Metz (KIT)

Fuel data under investigation

		PWR	BWR	THTR / VVER
Discharge		1989 -2008	2005 – 2008	
Manufacturer		AREVA	Areva/ Westinghouse	
Cladding	Material	Zry-4 – M5	Zyr 2	Graphite / Zr1%Nb
	∅	9.50 - 10.75 mm	9.84 - 10.2 mm	
	Thickness	0.62 - 0.73 mm		
Pellet	Enrichment	3.80 – 4.94 %	3.30 -4.25 %	2.4 -16.8%
	Grain size	5-40 µm	6 - 25 µm	20 -80 µm
	Density	10.41 g cm^{-3}	10.52 g cm^{-3}	10.8 g cm^{-3}
	Specifics	standard, NIKUSI production	standard and Al/Cr addition	
Irradiation	Burn-up	50.4 – 70.2 GWd/t	48.3 – 57.5 GWd/tU	
	Cycles	2 - 14	5 – 7	
lin. Power	average	186 -330 W/cm	160 W/cm	130 – 228 W/cm
FGR		4.9 – 23 %	1.2 – 3.1 %	

(D-N°:5.12) – Presentation of the project at the Spent Fuel Workshop and preparation of a full paper
Dissemination level: PU
Date of issue of this report: **30/12/12**

WP 2: Gas release and rim and grain boundary diffusion

WP leader: Detlef Wegen (JRC-ITU)

- Experimental determination of fission gas release

KIT: Set-up /calibrate analytical methods.
Application to hot gas samples from a segment

STUDSVIK: Laser-Ablation Mass Spectroscopy for determination of radial fission gas distribution.

JRC-ITU: Annealing up to 1600 K to determine venting/volatilization from the grain boundary. Knudsen cell effusion test

WP 2: Gas release and rim and grain boundary diffusion

WP leader: Detlef Wegen (JRC-ITU)

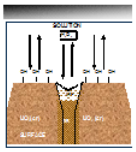
- Experimental investigation of rim / grain boundary diffusion

JRC-ITU: Static experiments with ^{18}O water, incl. specific analysis.
Grain boundaries, pores and µ-cracks for reaction with and intrusion of water.

JÜLICH : Determination of microstructure and element distributions in fuel samples before and after leaching.

CNRS: Dissolution experiments with in-situ µ-Raman spectroscopy.
α-irradiation of grain boundaries under Ar and Ar/H_2.

α beam irradiation of grain boundaries



WP 3: Dissolution based release

WP leader: Karel Lemmens (SCK·CEN)

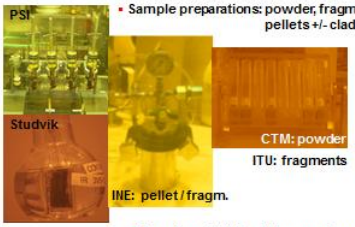
- Dissolution based radionuclide release and to the extent possible the chemical speciation of the relevant isotopes.

Partners: **KIT-INE, JRC-ITU / CTM, PSI, Studsvik, EK (MTA), SCK•CEN**
Variations: - Fuel types (WP 1)
- Experimental details (leachants / replacements)
- Test durations (100 days – 1 year)
- Analyses

Specific Analyses: Before and after leach experiments by beam line techniques (e.g. for redox state)
Tc (L_1-edge) => EXAFS + XANES, **Se** (K-edge) => XANES
I (L_1-edge) => XANES

WP 3: Experimental details

- Sample preparations: powder, fragments, pellets +/- cladding



- Atmospheric conditions

Conditions	
PSI	anoxic/oxic
KIT	anoxic
ITU/CTM	oxic
SCK·CEN	oxic
Studsvik	oxic

- Leach media (simple to complex or buffer solution)

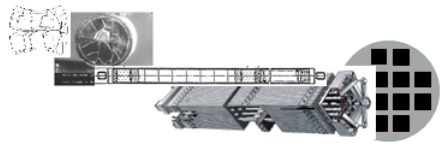
	borate	Na^+	I^-	Cl^-	HCO_3^-	Ca^{2+}	Mg^{2+}	SO_4^{2-}	pH
PSI	28	>0.13	0.13		1.1	-	-	-	8.5
KIT	-	10	-	10	-	-	-	-	
ITU/CTM	-	19	-	19	1	-	-	-	7.4
SCK·CEN									
Studsvik	-	2.26	-	1.4	1.8	0.464	0.19	0.1	8.1-8.2

WP4: Modelling

WP leader: Joan de Pablo (CTM/UPC)

Aims of the conceptual and numerical modelling

- Quantify the fast/instant release fraction of fission products.
  - *FP migration along the grain boundaries and fractures,*
  - *effects of defects in the cladding on the fast release.*
- Up-scaling from fragments/pellets to fuel assemblies.



- Modeling the chemical state of relevant elements.
- Transition between fast release and the significantly slower release processes due to radiolytically driven matrix dissolution.
- Assessing the potential impact on the Safety Case.

WP5: Knowledge, reporting and training:

WP leader: Vanessa Montoya (AMPHOS21)

- Knowledge management and documentation of the State-of-the-Art with regular up-dating.
- Stepwise build-up of scientific-technical reporting
- Dissemination and Communication,
- Training

All the information is included in the project webpage

www.firstnuclides.es

Dissemination and Communication

- FIRST-Nuclides Project Workshops (after 10, 24 and 33 months)
  - Mandatory for beneficiaries
  - 1st Annual Workshop: October 9-11, 2012 in Budapest, HU
- Web portal: www.firstnuclides.eu
- Generic poster
- Presentations, e.g. in "EURATOM FP7 Research Projects and Training Activities Vol. 3"
- Newsletter
- Reporting
  - Deliverables
  - Scientific-technical results in peer reviewed publications
  - Proceedings of Annual Workshops: KIT Scientific Report Series
- Presentation of FIRST-Nuclides, results and conclusions at international workshops, conferences, etc.

WP6: Coordination and management

WP leader: Bernhard Kienzler (KIT)

➔ Thanks to the organizers of the Spent Fuel Workshop 2012.

➔ Thank you for listening.

2. Oral presentation at the ATALANTE 2012

Euratom 7th Framework Programme
Collaborative Project "Fast / Instant Release of Safety Relevant Radionuclides from Spent Nuclear Fuel"

FIRST-Nuclides

ATALANTE 2012, Montpellier, F, September 2-7, 2012
Session: Geological repository nuclear chemistry

Bernhard Kienzler, KIT-INE, Karlsruhe, D
Alba Valls, AMPHOS21, Barcelona, E
Volker Metz, KIT-INE, Karlsruhe, D

Acknowledgement: The research leading to these results has received funding from the European Atomic Energy Community's Seventh Framework Programme (FP7/2007-2011) under grant agreement no. 295722, the FIRST-Nuclides project.

Details on the CP FIRST-Nuclides

EU Call:	Research activities in support of implementation of geological disposal (*Fission-2011-1.1.1*)
Type:	Small/medium scale Collaborative Project
Financial:	4,741,261 € total costs 2,494,513 € EU contribution
Duration:	01. Jan. 2012 – 31.Dec. 2014 (3 years)

Definition of instant / fast release

Project related definition or PA related definition

RN release by matrix corrosion controlled by radiolysis, hydrogen, Cl-.... ↔ initial (fast) release of RN controlled by chemistry, spatial distribution, burn-up, lin. power, reactor operation

- gap
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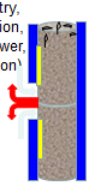

Definitions:

- **PA term:** „instant" release
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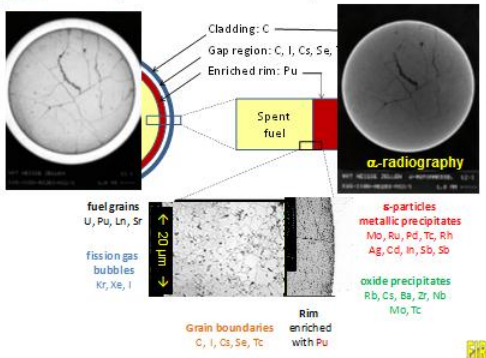
Power and temperature history

Pellet	KKG	GKN II	KKL
Burn-up$_{av.}$ [GWd/tHM]	64	58	58
∅ [mm]	9.3	8.05	8.5
Lin power [W/cm]	228	167	184
ΔT [K]	725	528	585
$T_{coolant}$ [K]	325	305	263
T_{center} [K]	1050	833	848

$\lambda_{irrad\ fuel}$ = 2.5 W m^{-1} K^{-1} (Lucuta 1996)
ΔT = lin. power/(4π·λ)

Power ramp	
Lin. Power Change [W/cm]	100
ΔT [K]	318

Complex structure of spent fuel



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Rim Zone

Porosity: $\varepsilon = BU_{av} \cdot d_1 \cdot \exp\left(-d_2 + d_3 \cdot \frac{r}{r_0}\right)$

Rim burn-up: $BU_R = 1.33 \cdot BU_{av.pellet}$

Rim Thickness: $R_t = 5.44\ BU_R - 281$

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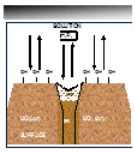
α beam irradiation of grain boundaries



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Partners: **KIT-INE, JRC-ITU / CTM, PSI, Studsvik, EK (MTA), SCK•CEN**

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WP 3: Experimental details

- Sample preparations: powder, fragments, pellets +/- cladding

- Atmospheric conditions

	Conditions
PSI	anoxic/oxic
KIT	anoxic
ITU/CTM	oxic
SCK·CEN	oxic
Studsvik	oxic

- Leach media (simple to complex or buffer solution)

	borate	Na^+	I^-	Cl^-	HCO_3^-	Ca^{2+}	Mg^{2+}	SO_4^{2-}	pH
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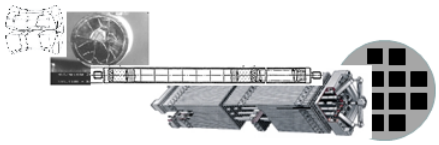

- Modeling the chemical state of relevant elements.
- Delineation from matrix dissolution process.

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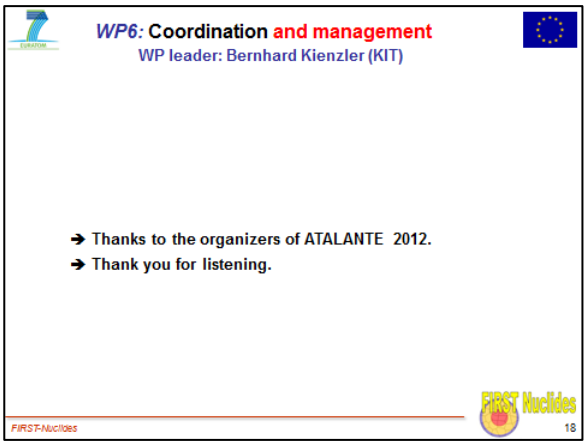

3. Paper contribution at the EURADISS Workshop

EURATOM 7TH FRAMEWORK PROGRAMME: COLLABORATIVE PROJECT "FAST / INSTANT RELEASE OF SAFETY RELEVANT RADIONUCLIDES FROM SPENT NUCLEAR FUEL" (FIRST-Nuclides)

Bernhard Kienzler[1*], Volker Metz[1], Alba Valls[2]

[1]Karlsruhe Institute of Technology, Institute for Nuclear Waste Disposal (DE)
[2]Amphos21 (ES)

* bernhard.kienzler@kit.edu

Abstract

The EURATOM FP7 Collaborative Project "FIRST-Nuclides" is established with the overall objective to provide for improved understanding of the fast / instantly released radionuclides from disposed high burn-up UO_2 spent nuclear fuel. This issue is given high priority by the European Implementing Geological Disposal Technology Platform and the outcome of the project is relevant for all types of host rocks in Europe.
The project provides for experiments combined with modelling studies applying the different results as well as for up-scaling from experimental conditions to entire Light Water Reactor fuel rods. Spent fuel materials of known initial enrichment, burn-up and irradiation histories, are selected and characterized. Experiments and modelling studies address the correlation between the fast release of fission gases and non-gaseous fission products.

In this paper, the objectives of the Collaborative Project, the implementation and organization are presented. The integration and communication within CP FIRST-Nuclides is demonstrated in the context of the kick-off meeting and the 1st Annual Workshop held in February and October, 2012, respectively. An overview on the scientific and technical achievements is given as well as the efforts for harmonization and adjustments of the experimental work.

Introduction and Objectives

In the Vision Report and the Strategic Research Agenda (SRA) of the "Implementing Geological Disposal – Technology Platform"(IGD-TP), a key topic deals with "waste forms and their behaviour". On this background, the EURATOM FP7 Collaborative Project "Fast / Instant Release of Safety Relevant Radionuclides from Spent Nuclear Fuel (CP FIRST-Nuclides)" was granted and started on 1st January 2012. It is planned for three years, and the experimental period will span the first 30 months: Spring 2012– Fall 2014. The total costs of CP FIRST-Nuclides amount to 4,741,261 € and the financial contribution by EU to 2,494,513 €. Karlsruhe Institute of Technology, Institute for Nuclear Waste Disposal (KIT-INE) is the coordinator of this Collaborative Project (CP).
The CP FIRST-Nuclides deals with understanding the behaviour of high burn-up spent uranium oxide (UO_2) fuels in geological repositories. This waste type represents one of the sources for the release of radionuclides after loss of the disposed canister integrity.Therefore, the time-dependent release of radionuclides from spent high burn-up UO_2 fuel is also required for safety analyses. The first release fraction consists of radionuclides (1) in gaseous form, and (2) those showing a high solubility in groundwater.
With respect to the fast / instant release of radionuclides (RN) from spent nuclear fuel elements under deep underground repository conditions, a number of questions are still open. Consequently, the CP FIRST-Nuclides aims on the determination of the "instant release fraction (IRF)" values of iodine, chlorine, carbon and selenium that are still largely unknown. The elements I, C, Cl and Se tend to form anionic species. Such anions are hardly chemically retained in the repository barrier system.

Organization of FIRST-Nuclides

The CP is organized in six workpackages (WP) (see Figure 1). WP 1: "Samples and tools" deals with the selection, characterization and preparation of the materials to be studied and the set-up of tools. One of the essential requirements of the project is that typical and sufficiently well characterized spent fuel is being used for the experiments and modelling studies. WP 2 covers the "Gas release and rim and grain boundary diffusion experiments" and WP 3 includes "Dissolution based release studies". These WP include the determination of the chemical form of released radionuclides, fission gases, ^{135}Cs, ^{129}I, ^{14}C, ^{79}Se, ^{99}Tc and ^{126}Sn. WP 4 "Modelling" deals with modelling of migration/retention processes of fission products in the spent fuel structure. Special attention is attributed to model the fission product migration along the grain boundaries, the effects of fractures in the pellets and of holes/fractures in the cladding. The modelling work within FIRST-Nuclides will help to

clarify which geometric scales dominate the fast/instant release. WP 5 "Knowledge, reporting and training" is responsible for the knowledge management generated within the project, the state-of-the-art report, the general reporting, keeping the documentation up-to-date and organizing training measures. The management of the Collaborative Project is included in WP 6.

The composition of the different bodies involved in the project structure is described below. The project is implemented by a consortium with ten beneficiaries (AEKI/MTA, AMPHOS21, CNRS, CTM, JÜLICH, JRC-ITU, KIT, PSI, SCK•CEN and STUDSVIK). *The Coordination Team* consists of KIT (Coordinator) and AMPHOS21 (Coordination Secretariat) which are responsible for project management, knowledge management, documentation, dissemination and training. Their responsibilities include further the coordination of the project work and activities, communication between the Project Consortium and the Commission, monitoring the use of resources and transferring financial resources, communication between different project beneficiaries and bodies, documentation of the project outcome and its dissemination and communication to interested parties. *Workpackage leaders* head the individual workpackages (i.e. WP1 and WP6 (KIT), WP2 (JRC-ITU), WP3 (SCK•CEN), WP4 (CTM) and WP5 (AMPHOS21).

Several organisations from France (CEA), USA (LANL, SANDIA), UK (NDA, NNL and a consortium coordinated by the University Cambridge), Finland (Posiva, TVO), Germany (GRS) contribute to the project without any funding as *Associated Groups (AG)* with particular information exchange functions. Finally, a group of six implementation and regulatory oriented organizations (SKB, NAGRA, ONDRAF/NIRAS, ANDRA, BfS, ENRESA) are participating as an *"End-User Group (EUG)"*. This group ensures that end-user interests are reflected in the project work, and reviews the project work and scientific-technical outcome.

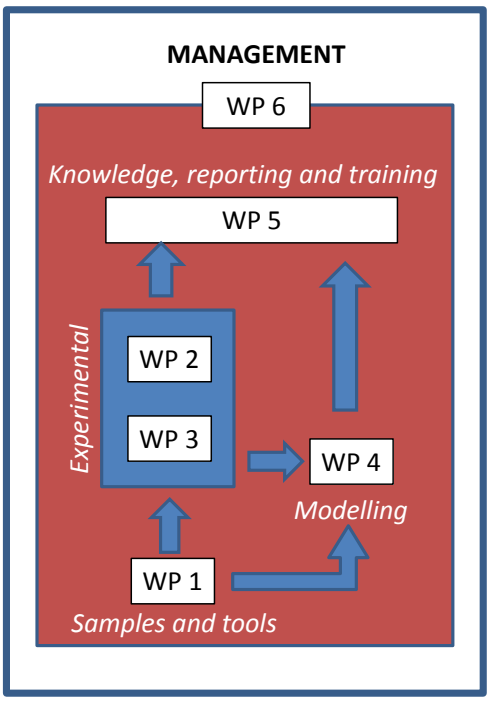

Figure 1. Project Workpackages interdependencies

Integration and Communication within CP FIRST-Nuclides

Integration, communication, discussion of scientific and technical results and dissemination of the project's achievements are fundamental for the project. For this reason, annual project workshops are organized for presenting the status of the work programme. Moreover, these events give the project beneficiaries the opportunity to discuss administrative and future implementation issues. These project workshops additionally include meetings of the ExCom (Executive Committee), and of the EUG members, as well as special meetings where the AGs have the opportunity to present their work and ideas. Furthermore, as training and education component, "Topical Sessions" are included in the workshop programs dealing with general scientific-technical aspects relevant to the project.
The Project Workshops contribute to the integration within the project and communication with a broader interested community. This is done both by invitation of external groups to the workshops and by documentation of the progress in public Annual Workshop proceedings. These proceedings include i) a summary of the project and the ongoing research and technical activities, ii) abstracts of the Topical Session presentations and presentations of the AGs. Moreover, the details of the work are described in scientific/technical contributions (S&T) written by the beneficiaries and reviewed by the EUG.

Kick-off Meeting
The kick-off meeting was held in Barcelona, Spain, 9th – 10th February 2012. The participants came from the beneficiary organisations (32), Associated Groups (8) and 5 from the organizations representing the End User Group. Besides an intensive discussion of the work programs of the workpackages, key note lectures were given on "Impact of the irradiation history of nuclear fuels on the corrosion behaviour in a disposal environment" by Daniel Serrano-Purroy (JRC-ITU) and on "Thermodynamics of fission products in spent nuclear fuel" by Ondrej Benes (JRC-ITU). Furthermore, David Sassani, as representative of the AG SANDIA gave an "Overview of Used Fuel Degradation & Radionuclide Mobilization Activities within the Used Fuel Disposition Campaign".
Several remarks by the End User Group were of high importance for the project: The CP FIRST-Nuclides fits in the overall view of WMOs (Waste Management Organizations) including their engagement in the SRA of IGD-TP. A series of general comments of EUG on FIRST-Nuclides Description of Work (DoW) were provided. It was positively acknowledged that the CP FIRST-Nuclides covers a broad scope and overall approach. It was also highlighted that the structure is expected to lead to useful results.
For the WMOs it is important that the key project results will be published in the scientific literature. The EUG acknowledged also that efforts have been made to get background data on spent fuels (irradiation history, etc.) that will be studied in the project. They pointed out that the variety of fuels used in the experimental program need to be representative for power reactors under typical operating conditions, including the range of burn-up values and the present trend to higher burn-ups. For fuels irradiated under different conditions, the details need to be documented as well. Some specific comments of members of the EUG covered inter alia the effects of solution chemistry and of temperature on rapid release processes. The effects of reducing conditions are of great interest as well as the fast radionuclide release from

MOX fuel which is not covered in the project. Other questions concerned the representativeness of the results for a fuel rod or fuel assembly obtained by leaching individual rod sections where discussed during the meeting.

First Annual Workshop

The First Annual Workshop was held already in Budapest, Hungary, 9th – 11th October, 2012. At this workshop, a Topical Session was organized as part of training activities within the project. The first presentation in this session was given by Christoph Gebhardt and Wolfgang Goll, (AREVA) on "Characteristics of Spent Nuclear Fuel". This talk provided information on fuel management for Pressurized Water Reactor (PWR) and Boiling Water Reactor (BWR) fuels, including neutron-physical aspects of the different modern fuel types and the burn-up distributions as well as the power/temperature histories. The manufacturing processes and inventory of nuclides, additives and impurities of fuel pellets were explained as well as the types, element inventory and corrosion and hydrogen uptake of the fuel rod cladding. Finally, the defect mechanisms during storage of fuel under various conditions were discussed.

The 2nd talk of the Topical Session was given by Paul Van Uffelen (JRC-ITU) on "The potential of the TRANSURANUS code for source term calculation of spent nuclear fuel". TRANSURANUS is a computer program for the thermal and mechanical analysis of fuel rods during various operating conditions, generally referred to as a fuel performance code. Emphasis was given to assessing both the temperature and the stress levels in a fuel pin, since they are linked to the crucial safety criteria, set by the regulators, to be fulfilled during the entire in-pile lifetime of the fuel rod. The lecture treated the main equations and the associated limitations with the focus on fission product behaviour. Following mechanisms were considered in greater detail: recoil, knock-out & sputtering, lattice diffusion, trapping, irradiation re-solution, thermal re-solution, thermal diffusion, grain boundary diffusion, grain boundary sweeping, bubble migration, bubble interconnection and sublimation or vaporisation. For each of these mechanisms, a brief description was provided along with a discussion of its domain of application. An explanation of the main relevant mechanisms was described and, the various models for fission gas release and swelling available in the open literature were reviewed.

Both presentations of this Topical Session provided knowledge which was new to most of the participants and which will help to keep a common level of understanding of fuel, fuel rods and the modern modelling tools throughout the CP FIRST-Nuclides.

Communication to international audiences

The CP FIRST-Nuclides was presented orally at the "Spent Fuel Workshop" organized by CEA, Avignon, April 18-19, 2012. The generic poster was shown at the E-MRS 2012 SPRING MEETING, Strasbourg, May 14-18, 2012. Furthermore a talk on the project was given at the ATALANTE Conference, September 2-7, 2012 at Montpellier. The coordinator is invited to give a talk on FIRST-Nuclides at the International High-Level Radioactive Waste Management Conference, April 28 - May 2, 2013, Albuquerque, New Mexico, USA.

Scientific and Technical Achievements

Deliverables

CP FIRST-Nuclides has been running for 10 months (January-October 2012). In this period, two main deliverables have been submitted. The first one covers the "*State-of-the-Art and Rationale for Experimental Investigation*" within FIRST-Nuclides. This deliverable presents the State-of-the-Art with respect to the fast / instant release of safety relevant radionuclides from spent nuclear fuel. In the first part of the report basic information is given, such as the characterization of nuclear fuel, irradiation and temperature-induced processes in UO_2 during its use in reactors, and disposal concepts for spent nuclear fuel in different countries. The second part of the report documents, the State-of-the-Art on fast release summarizing the results obtained from more than 100 published experiments with different samples, experimental techniques, and durations. The evaluated publications covered the gap and the grain boundary contribution to the release from 80 experiments using PWR fuel of different burn-up, 20 experiments with BWR fuel and 8 MOX fuel experiments. All authors refer to a definition of the fast / instant release as a fraction of the inventory of radionuclides that may be rapidly released from the fuel and fuel assembly materials at the time of canister breaching. In the context of safety analysis, the time of mobilization of this fraction can be considered as an instantaneous release of some radionuclides at the time of containment failure. The report will be updated regularly according to the results obtained in the 7th Framework Programme Collaborative Project FIRST-Nuclides.

The 2nd deliverable is entitled "*Characterisation of spent nuclear fuel samples to be used in FIRST-Nuclides* and *relevance of the samples for the Safety Case"*. Data on fuel samples, which are available for the FIRST-Nuclides partners, are classified according to their data category with respect to the fuel / cladding characteristics. Moreover, these data-sets are classified according to three information levels: (i) essential information representing the minimum data that should be available for the fuel chosen for the study, (ii) parameters and data, which are not directly measured, but derived from calculations, and (iii) supplemental information referring to characteristics that may be needed depending on the studies to be performed. Critical parameters of the selected fuel samples are compared to those of high burn-up fuels which need to be disposed of in Europe, to assure the relevance of the samples for the Safety Case.

Spent fuel rods are owned mainly by the reactor operating utilities. Due to competition, it is not easy to get the complete information on the actual burn-up history of the fuel samples, as well as the right to publish the data. To get access required a lot of work and negotiations for many institutions. The following table summarizes the most important characteristics of the spent nuclear fuels to be used within FIRST-Nuclides.

Table 1: *Overview of the fuels to be investigated within CP FIRST-Nuclides*

		PWR	**BWR**	**THTR/VVER**
Discharge		1989 -2008	2005 – 2008	
Manufacturer		AREVA	Areva/ Westinghouse	
Cladding	Material	Zry-4 – M5	Zyr 2	Graphite / Zr1%Nb
	∅	9.50 - 10.75 mm	9.84 - 10.2 mm	
	Thickness	0.62 - 0.73 mm		
Pellet	Enrichment	3.80 – 4.94 %	3.30 -4.25 %	2.4 -16.8%
	Grain size	5-40 µm	6 - 25 µm	20 -80 µm
	Density	10.41 g cm^{-3}	10.52 g cm^{-3}	10.8 g cm^{-3}
	Specifics	standard, NIKUSI production	standard and Al/Cr addition	
Irradiation	Burn-up	**50.4 – 70.2 GWd/t**	**48.3 – 57.5 GWd/tU**	
	Cycles	2 - 14	5 – 7	
lin. Power	average	**186 -330 W/cm**	**160 W/cm**	130 – 228 W/cm
Fission Gas Rel.		**4.9 – 23 %**	**1.2 – 3.1 %**	

Harmonization within the R&D work

During the kick-off meeting, the questions posed by the EUG were mainly concerning the representativeness of rod sections for up-scaling to a fuel rod, rapid release under anoxic conditions and the effects of solution chemistry. This discussion was continued among the beneficiaries, especially in the context of WP 3 "Dissolution based release" i.e. leach tests to measure IRF. The results will certainly depend not only on burn-up, and other fuel-immanent characteristics, but also on some parameters such as sample sizes, sample preparation, leachant and atmospheric conditions. For this reason, the partners agreed upon the leaching conditions shown in Table 2.

Previous investigations showed no significant influence of salinity on the IRF (Kienzler et al. 2012). Therefore, a standard water with 19 mM NaCl and 1 mM $NaHCO_3$ was selected to be justified. The beneficiaries KIT, SCK·CEN, PSI, JRC-ITU /CTM and STUDSVIK agreed also upon the use of different fuel sample sizes, such as cladded pellets, fragments, powder with and without claddings. This selection guarantees that all relevant disposal conditions are covered. Additionally, this selection will provide data needed for modelling.

Table 2: *Overview on the leachants, atmospheric conditions and duration of the experiments*

	Leachant	Atmosphere	Duration
KIT	19 mM NaCl + 1 mM $NaHCO_3$	Argon/H_2	Total duration ~12 Months
SCK·CEN	19 mM NaCl + 1 mM $NaHCO_3$	Anoxic or slightly oxidizing	Total duration ~12 Months
PSI	19 mM NaCl + 1 mM $NaHCO_3$	Anoxic or slightly oxidizing	Total duration ~12 Months
JRC-ITU / CTM	19 mM NaCl + 1 mM $NaHCO_3$	Air (oxidizing)	Total duration 40-60 days
STUDSVIK	10 mM NaCl + 2 mM $NaHCO_3$	Air (oxidizing)	Total duration ~12 Months
EK	Storage pool water pH 4 and 7, boric acid concentration 15-21 g/(kg H_2O)	Oxidizing (open pool surface)	

S&T Contributions

Within the CP FIRST-Nuclides, the outcome of the project is documented by Scientific/Technical Contributions to be published in the Annual Workshop Proceedings. In the first 10 months of the project, most of the work load was on selection, characterization and preparation of materials and set-up of tools for handling and transportation of the highly radioactive material, including licensing issues. Nevertheless, 20 S&T contributions have been submitted for the 1st Annual Workshop Proceedings.

All partners having experimental programs contribute to WP 1: Selection, characterization and preparation of and set-up of tools for handling and transportation of the highly radioactive material. The contributions address the characterization and cutting schemas of fuel samples to be used for the planned leach tests. Results are presented of non-destructive analysis of fuel rods with respect to visual examination, γ-scanning, defect determination of the cladding, as well as the preparation of samples for IRF investigations, post irradiation examinations, and for combined XRF/XAS investigations at the Swiss beam line and ANKA facility at KIT. EK characterizes the spent VVER-440 with defects in the storage tank.

WP 2 covers gas release studies and investigation of rim and grain boundary diffusion processes. Contributions by JRC-ITU include a description of puncturing the rod segment at the plenum, collecting the internal gas in a standard volume and determining the pressure from KIT's 50.4 GWd/t_{HM} fuel rod segment. Analyses of gas composition and concentrations were performed and described by KIT.

Once the puncturing of the fuel rod segment was performed at ITU, the pressure was measured and gases were collected in stainless steel single-ended miniature sampling cylinders (50 ml). The cylinders were carried back to KIT-INE, where the gases were analysed by means of a quadrupole gas mass spectrometer (gas MS). The gas MS is optimized for low gas sample consumption. Within the system, the total gas pressure is controlled and

three different expansion-volumes allow to charge relatively low gas contents in the desired pressure range. By this method, the c*oncentrations of the* gases and absolute gas volumes sampled during the puncturing test of the 50.4 GWd/tHM fuel rod segment are determined and the FGR of Kr is calculated to 5.3% and of 6.3% for Xe. JÜLICH delivered a thorough microstructure analysis of their HTR spent fuel, as well as explanations, why this material can be used as one extreme for the spectrum of high burn-up fuels under investigation within FIRST-Nuclides. Radiolytic corrosion of grain boundaries of unirradiated UO_2 fuel is investigated by CNRS aiming on the understanding of the corrosion mechanisms at the solid/solution interface and taking into account irradiation effects at a µ-metric scale. For valuation of the effects, Raman spectroscopy is applied and preliminary results are reported. STUDSVIK presents the puncturing tests, and determination of the internal free volume of a rod. Furthermore, a new ablation equipment is described which will be used to study the radial distribution of I, Xe and Cs and to explore any correlation to the FGR and instant release leach rates of the corresponding fuel samples. It consists of a New-Wave UP-213 floating Nd:YAG laser in connection to an ablation chamber that is housed in a hot cell. The transport gas (He or Ar) from the ablation cell is injected into a Perkin Elmer Elan 6100 DRC II Inductively Coupled Plasma Mass Spectrometer (ICP-MS), installed in a glove box. The sample preparation technique is established with the cross sections to be analysed taken at mid pellet position one pellet away from the leaching samples.

For the investigations of the dissolution based release (WP3), the different concepts for leach tests are elaborated. To receive a consistent picture in FIRST-Nuclides, the dissolution based releases are investigated on different scales of the spent fuel material, pellets, de-cladded pellets, fragments and powder. PSI and SCK·CEN will use equivalent equipments. KIT-INE has started three static leaching experiments with a pellet-sized cladded segment and two decladded SNF fragments. The sampling procedures for gases and liquids are presented. These experiments will be conducted in autoclaves under reducing conditions performing regular sampling of gases and solutions. The partners ITU, KIT-INE, SCK·CEN and STUDSVIK will perform the experiments with the specified water composition given in Table 2. The experiments will cover different redox regimes from oxidizing / anaerobic to reducing.

The modeling approach presented by CTM (WP4) aims on establishing the leaching time to determine the IRF. In the experiments this time is arbitrary, ranging from 10 to 60 days or even longer. In published papers, the release of the selected radionuclides was measured for more than one year of leaching time, however showing a relatively low rate. The work will help to understand if this lower release rate is controlled by the availability of the water to contact grain boundaries or by dissolution of the matrix. KIT presented modelling of boundary and initial conditions required for up-scaling of migration / retention processes of fission products in the spent nuclear fuel structure. Calculations covered the temperature history of the fuel segment, the rim structure with respect to burn-up, porosity and Xenon content. Amphos21 developed a very promising approach to model water penetration into the inner parts of fuel pellets on the basis of models used for geo matrices.

Conclusions and Future work

A project dealing experimentally with spent nuclear fuel requires significant preoperational works by each beneficiary. In WP 1, the selection, characterization and preparation of materials process succeeded to receive the materials as well as the clearance by the fuel owners. Fuel rod characterisation and first gas measurements were reported. The dissolution based Instant Release experiments will start in the beginning of 2013, as scheduled in the work plan. As the experiments will have a maximum duration of one year, no delay of the project is expected. Also the modelling approaches show a significant progress, especially for the planning of the duration of dissolution based experiments.

The profile of the CP FIRST-Nuclides is established with the high number of Associated Groups, which have got access to the achievements of the project and contribute to the CP with own presentations. During the first 10 month of the project three additional AGs joined the Consortium.

The comment and recommendations by the End User Group during the kick-off meeting have mainly been addressed and answered during the presentations during the 1st Annual Workshop and by the S&T contributions of the workshop proceedings. During the workshop, the members of the EUG asked many relevant and constructive questions which were answered by the beneficiaries. The members of the EUG provided for lively discussions. Overall responses on the objectives and the status of the CP FIRST-Nuclides were positive, and any recommendations for changes to the work program were not given. However, the need of similar investigations for MOX fuel was raised by several participants.

Due to the positive feedback, the project work will continued as planned. Responses to the general comments and questions of the EUG during the kick-off meeting will be provided at the end of the project. However, the need of similar investigations for MOX fuel is raised by several participants.

Details on CP FIRST-Nuclides may be found on the project's homepage:
www.firstnuclides.eu .

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