Investigation of pelletised and sphere-packed oxide fuels for minor actinide transmutation in sodium fast reactors, within the FP-7 European Project PELGRIMM

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Abstract

The FP-7 European Project PELGRIMM, which stands for "PELlets versus GRanulates: Irradiation, Manufacturing, Modelling" is a 4-year and 7.2 M € project, addressing minor actinide (MA) bearing oxide fuel developments for Generation IV – sodium fast reactor systems. Both options, MA homogeneous recycle in driver fuels with MA content at a few percent and heterogeneous recycle on UO_2 fuels bearing high MA contents located in the radial core blanket, are considered. Two fuel forms: pellet and spherepack, are under investigation.

A total of 12 partners from research laboratories, universities and industries, collaborate to share and leverage their skills, progress and achievements, covering a comprehensive set of investigations:

- *perform the post-irradiation examinations of MARIOS and SPHERE irradiated pins within the European Project FAIRFUELS, providing the very first results respectively on the helium behaviour in $(Am,U)O_2$ fuels and a comparison between sphere-packed and pelletised $(U, Pu, Am)O_2$ fuel performances;*
- *take the next step in the $(Am, U)O_2$ fuel qualification rationale by performing the semi-analytical test MARINE in HFR;*
- *extend minor actinide bearing fuel fabrication processes to alternative routes in order to limit secondary waste streams;*
- *extend the capabilities of existing calculation codes devoted to fuel behaviour under irradiation;*
- *accomplish a preliminary design of a sodium-cooled fast reactor core with sphere-packed $(U, Pu, Am)O_2$ fuels and perform a preliminary safety assessment.*

PELGRIMM promotes the implication of European students and young researchers too, through:

- *placements of trainees in organisations involved in the project;*
- *the development and delivery of training courses and workshops;*
- *contributions to research actions of the project by researchers preparing a doctoral thesis or employed in a post-doctoral position.*

The paper gives an overview of the PELGRIMM programme to be completed by December 2015.

Introduction

Minor actinide incorporation into the fuel is a pre-requisite for Generation-IV systems to bring benefits in the disposal requirements by reducing the MA content in the high-level wastes, and to potentially increase the proliferation resistance.

Since americium is a strong gamma emitter (and curium a high neutron emitter), the MA-bearing fuel fabrication process needs shielding, remote handling by robotic arms, simplification as well as implementation of relatively dust free steps. Moreover, the high volatility of some Am compounds has to be managed during fuel fabrication as well as during irradiation phases, where Am should be more readily redistributed within the fuel than other actinides. Finally, the harmful consequences of additional helium production during fuel irradiation (related to ^{241}Am transmutation) on fuel swelling, degradation of the thermal properties and the high pressurisation of the pins have to be prevented or managed in a reliable manner.

Regarding future SFR systems, based on historical experience and knowledge, MA-bearing oxide fuels emerge as a short-term solution to meet the performance and reliability goals [1]. Two main MA-recycle options are currently under consideration [2]:

- the homogeneous mode, where small quantities (<5%) of MA are mixed to the $(U,Pu)O_{2-x}$ driver fuel of the reactor;
- the heterogeneous mode, where high MA contents (10-20%) are added to an UO_2 support, MA-bearing sub-assemblies being located at the periphery of the core.

Both kinds of MA-bearing oxide fuel investigations have started (more or less) recently and experimental knowledge remains mostly limited to laboratory-scale fabrication processes, a small amount of characterisation and out-of-pile testing, as well as scarce irradiation experiments (see Table 1).

This paper gives an overview of the latest FP-7 European Project PELGRIMM (2012-2015), which takes a new step in the long-term process of the MA-bearing fuel qualification rationale, by investigating a wide range of items:

- from solid to sphere-pac fuel shapes;
- from MA- bearing driver fuels (MADF) to MA-bearing blanket fuels (MABB);
- from fuel fabrication and characterisation to behaviour and performance under irradiation;
- from experiments to modelling and simulation;
- from normal operating conditions in a sodium fast reactor to transients and severe accidents.

Table 1: List of irradiation experiments implemented with MA-bearing fuels

	MADF (MA-bearing Driver Fuels)				**MABB** (MA-Bearing Blanket fuels)		
Test	SUPERFACT [3,4]	AM1 [5]	AFC-2C&2D [6]	SPHERE [7]	SUPERFACT [3,4]	MARIOS [8]	DIAMINO [9]
date	80's	2008	2008-2010	2012	80's	2011	2012
participants	CEA/JRC-ITU	JAEA	INL-DOE	FAIRFUELS	CEA/JRC-ITU	FAIRFUELS	CEA
reactor	PHENIX	JOYO	ATR	HFR	PHENIX	HFR	OSIRIS
Fuel form	pellets			Pellets & Spherepac	Pellets	Disks	
MA content	2% Am or 2% Np	2-5% Am	2%Am	4%Am	20%Am 20%Np	15%Am	
MA compounds synthesis process	- Powder metallurgy - gelation & MA infiltration	Powder metallurgy		gelation & Am infiltration	gelation & MA infiltration	Powder metallurgy	
Burn-up	~7at%	10 min & 24h	10-24at%	under preparation	~6at%	305 EFPD	Under preparation
Linear Heat rate ($W.m^{-1}$)	30-35	43	<30		17-27		

PELGRIMM work plan

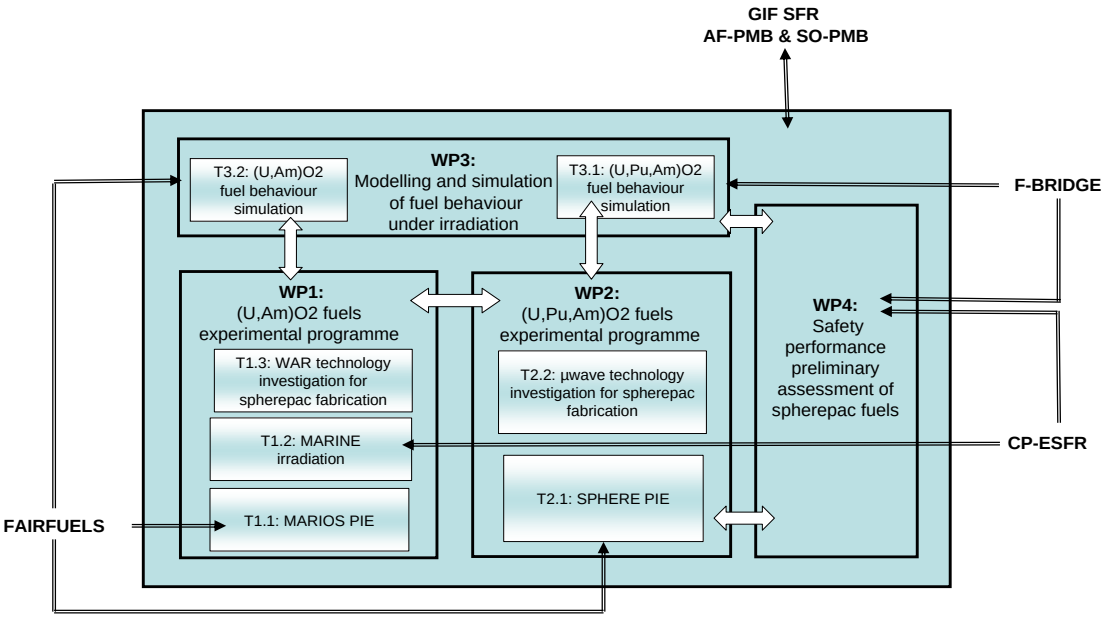
PELGRIMM, which stands for "PELlets versus GRanulates: Irradiation, Manufacturing, Modelling" is a FP-7 European Project of 7M€, which:

- capitalises on the efforts made within the previous European projects: ACSEPT (2008-2012)[10], FAIRFUELS (2009-2013) [11], F-BRIDGE (2008-2012) [12] and CP-ESFR (2008-2012) [13];
- takes a new step in the development of both MA-bearing fuel options: MADF and MABB, related to MA-bearing fuel fabrication processes, irradiation behaviour and core safety performance, by including a comparison of fuels shaped as pellets and beads.

Indeed, even if fuel pelletised forms have been preferred so far in all irradiation tests, except in the SPHERE experiment where granulates are under investigation (see Table 1), sphere-pac technology (leading to the production of beads) is attractive regarding MA-bearing fuels as it would lead to a significant simplification of the fabrication process thanks to the elimination of some process steps as milling, pressing and grinding, which involve fuel powders (and dust). Moreover, the compactness of the fabrication process would be increased. Finally, the sphere-pac fuel performance under irradiation could provide significant advantages, thanks to a better accommodation of solid swelling (compared to pellets) through the re-arrangement of the free inter-particular areas, and ultimately in better management of the helium generated during irradiation.

The Pert Chart drawn in Figure 1 gives an overview of the PELGRIMM technical content as well as the dependencies and the two-way links within the project.

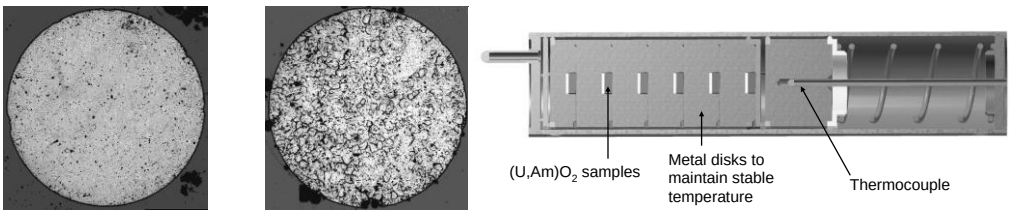
Figure 1: PELGRIMM technical activities



Work-Package 1 (WP1) addresses experimental investigations on minor actinide bearing blanket –MABB- fuel developments in order to:

- leverage the efforts made within the FAIRFUELS (and ACSEPT) projects to perform the MARIOS separate-effect irradiation [8], by executing an extended Post-Irradiation Examination (PIE) Programme (Task 1.1). PIE will provide the very first results on helium behaviour and accommodation concerning two $U_{0.85}Am_{0.15}O_{2-x}$ fuel microstructures (Figure 2a) shaped as discs and irradiated in dedicated mini-pins (Figure 2b) [14] at two temperatures of interest regarding MA-heterogeneous recycle implementation [15]: 1 000 and 1 200°C.

Figure 2: MABB microstructures and pin used for the MARIOS irradiation

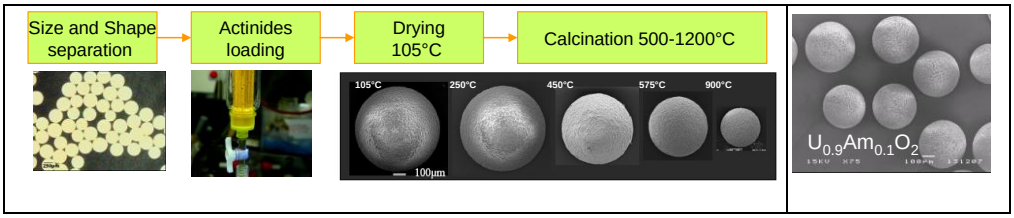


(a): Macrograph of high density (93%)and low density (88%) discs (diameter of 4.5 mm) [16].

(b): Sketch of a mini-pin with 6 disks

- take the next step in the fuel qualification rationale, by performing the semi-integral experiment MARINE in HFR (Task 1.2), i.e. by testing the behaviour of pelletised and sphere-packed $(U,Am)O_2$ fuels stacked in two instrumented (on-line pressure measurement) small-pins. A second stage in the long-range qualification approach of MABB will then be reached. Moreover, the promising sphere-pac concept that consists in filling pins with several diameter size microspheres (prepared here by the well established process consisting of an external gelation and MA infiltration [17]) will be tested for the first time on MABB compositions. Finally, the MARINE irradiation test is the matching piece to the SPHERE irradiation (Table 1) performed on minor actinide driver fuel (MADF) within the FAIRFUELS Project.
- extend the MABB bead synthesis route to be used for the MARINE fuel fabrication to an alternative using the Weak Acid Resin (WAR) technology [18] (T1.3). The latter is based on the loading of beads of ion exchange resin by actinides solutions and their mineralisation into oxides by heat treatment (see Figure 3). Both are wet technologies and consequently dust free. As less secondary process wastes are expected from the WAR process implementation, its industrial scalability could be more appropriate. Finally, both conversion routes fully address concerns on fuel cycle integration from reprocessing to fabrication, and, doing so, are being performed on minor actinide solutions.

Figure 3: Principle of the WAR process applied to the synthesis of uranium dioxide-based materials [19]



Work-Package 2 (WP2) addresses experimental investigations on minor actinide bearing driver fuel –MADF- developments in order to:

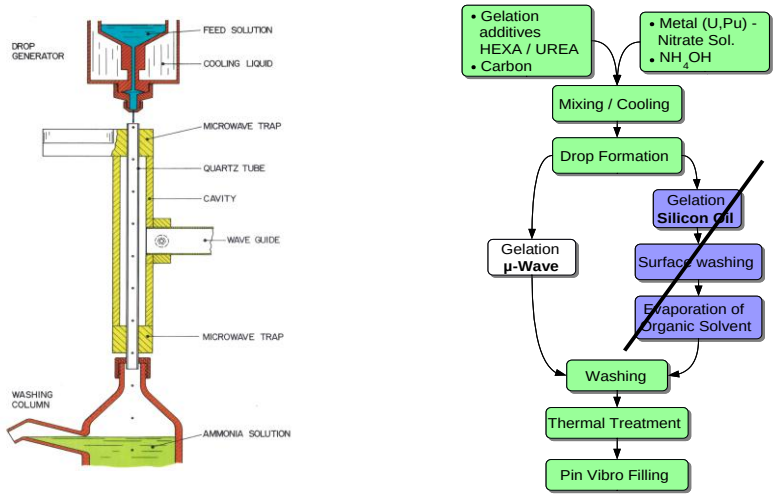
- exploit the efforts made and the results emanating from the FAIRFUELS Project to perform the SPHERE semi-integral irradiation (Table 1) in HFR, by executing the essential post-irradiation examinations. PIE will provide the very first results on MADF sphere-pac fuel behaviour under irradiation as well as the first comparison between sphere-packed and pelletised MADF fuel performances.
- extend and explore MADF bead synthesis routes in addition to that developed for SPHERE fuel fabrication (prepared by external gelation and MA infiltration). Hereby, it is planned to convert spherical drops produced by internal gelation, to green microspheres via the microwave technology [20] in order to limit secondary waste streams (see Figure 4).

Work-Package 3 (WP3) addresses modelling and simulation of fuel behaviour under irradiation regarding both MADF and MABB compositions as well as sphere-pac and pellets in order to establish reliable predictive capabilities of existing fuel performance codes to specific issues related to MADF/MABB fuels, namely:

- low MA content in $(U,Pu)O_2$ support matrix and high content of MA in UO_2 support matrix;
- high temperatures in SFR driver fuels (MADF) and moderate or even low temperatures (<1 500°C) as expected for MABB in dedicated blanket assemblies near the SFR core periphery;

- helium high release ratios in MADF fuels and potentially low release ratios in MABB fuels. The latter could lead to excessive MABB fuel swelling and necessitates its accommodation, possibly including microstructures making release easier;
- transition from pelletised to sphere-packed forms of MA-bearing fuels, leading to modification of thermal and mechanic property descriptions as well as changes in fuel/cladding mechanical Interactions due to the softer mechanical behaviour of sphere-packed fuels (specifically here experience gained from the F-BRIDGE Project).

Figure 4: Microwave internal gelation route and set-up



Most of the codes under consideration have already been upgraded within the framework of previous and on-going European projects. These codes are:

- MACROS, upgraded in FP-5 FUTURE, FP-6 EUROTRANS and currently FP-7 FAIRFUELS projects to predict ADS type fuel behaviour under irradiation [21];
- TRANSURANUS [22] upgraded within the F-BRIDGE (FP7) project [23] [24] to integrate sphere-pac fuel geometry as well as some of the mesoscopic information already gained from fundamental research activities, which govern macroscopic Generation-IV fuel behaviour at a macroscopic scale.
- SPHERE/SPACON codes [25] [26] dedicated to sphere-packed fuel behaviour under irradiation, upgraded within F-BRIDGE to be able to simulate the behaviour of advanced sphere-packed fuel in Generation-IV systems.

The latest code to be used is a dedicated version of GERMINAL [27] adapted for the description of the MABB fuel behaviour under irradiation.

Thus, PELGRIMM capitalises on previous investments and provides a lean and efficient means to provide high-quality modelling needed for this project.

Work-Package 4 (WP4) addresses, in continuity with F-BRIDGE and CP-ESFR projects, a preliminary safety performance assessment of sphere-packed MADF fuels in order to start linking sphere-packed fuel fabrication and irradiation behaviour developments to the problematics of core physics, design and safety performance. Indeed, F-BRIDGE has to provide some evaluations of the impact and suitability of sphere-packed fuels for Generation-IV systems, based on literature review and partner experiences, as well as

economic assessments of sphere-pac versus pelletised conventional fuels and possible schemes related to MA recycle in fast reactor. CP-ESFR includes, among others, activities related to SFR core designs loaded by pelletised MADF fuel elements, burning performances and core safety characteristics [28] The approach will be as follows:

- selection of a reference core design (pelletised fuel) from information provided by CP-ESFR;
- design of a core loaded with a MADF sphere-packed fuel;
- determination of core safety parameters and burn-up behaviour;
- simulation of fuel behaviour under normal operation and design basis conditions with special view to the impact of helium pressurisation and fuel expansion on transient behaviour. For design extension conditions up to severe accidents, pin failure and fuel micro-sphere motion effects will be investigated. The impact of fuel micro-sphere and gas releases will be determined and safety margins will be assessed.

Finally, beyond links between tasks and work-packages in PELGRIMM, as well as with other projects, synergies will be sought with activities on SFR systems performed within the Generation-IV International Forum, exchanging with the Advanced Fuel Project Management Board and the Safety and Operation Project Management Board [29]. Moreover, education and training activities with the placement of trainees of Master's degree during ~6 months in organisations involved in the project will foster the PELGRIMM dynamism, contribute to knowledge dissemination and provide incentive to join the project.

Conclusion

The FP-7 European Project PELGRIMM contributes to the sodium fast reactor fuel investigations that aim to reduce the actinide content in high-level waste and consequently limit disposal requirements. The project deals with minor actinide-bearing oxide fuel developments within two current minor actinide transmutation options: homogeneous and heterogeneous recycling, as well as two kinds of fuel shapes: pellets and beads. 12 partners (CEA, AREVA, EDF, ENEA, ENEN, JRC, KIT, KTH, LGI, NRG, PSI, SCK-CEN) will:

- make the most of the ground made in the MARIOS and SPHERE irradiation tests carried out as part of FAIRFUELS, providing the very first results respectively on the helium behaviour in $(Am,U)O_2$ fuels and a comparison between sphere-packed and pelletised $(U,Pu,Am)O_2$ fuel performances;
- take the next step in the $(Am,U)O_2$ fuel qualification rationale by performing the semi-integral test MARINE in HFR;
- extend minor actinide-bearing fuel fabrication processes to alternative routes in order to limit secondary waste streams;
- extend the capabilities of existing calculation codes of fuel behaviour under irradiation;
- address a preliminary safety performance assessment of sphere-packed $(U,Pu,Am)O_2$ fuels in the core of a sodium fast reactor.

PELGRIMM will also promote involvement among European students and young researchers through trainee placements in organisations involved in the project, the development and delivery of training courses and workshops, and contributions to research actions of the project by researchers preparing a doctoral thesis or employed at a post-doctoral position.

Acknowledgments

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