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Outcomes of the FP-7 project PELGRIMM investigating pelletized and sphere-packed oxide fuels for Minor-Actinides transmutation in a Sodium Fast Reactor.

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Abstract – *PELGRIMM that stands for "PELlets versus GRanulates: Irradiation, Manufacturing, Modelling", is a 4 years European project started in 2012, that investigates Minor Actinides - bearing fuels, shaped as pellets and beads, for Sodium Fact Reactors.*
Both Minor-Actinide transmutation options: the MA homogeneous recycle in driver fuels and the MA heterogeneous recycle on UO_2 *fuels located in radial core blankets, are under consideration. The objectives of the project are to capitalize on efforts made within the previous European projects: ACSEPT, FAIRFUELS, F-BRIDGE as well as CP-ESFR, and to take a new step in the long range qualification approach regarding fabrication process, irradiation behaviour, performance and safety, for pelletized and sphere-packed shaped Am-bearing fuels. The paper gives an overview of the PELGRIMM outcomes related to: first results on helium and fission gas behaviour in* $(Am,U)O_{2-x}$ *fuels irradiated in the separated effect test MARIOS; progress status of the semi-prototypic irradiation MARINE under preparation, that will provide a comparison between sphere-packed and pelletized* $(U,Am)O_{2-x}$ *fuel performances; investigations performed to extend Minor Actinide bearing fuel fabrication processes to alternative routes, that are the Weak Acid Resins and micro-wave technologies, in order to limit secondary waste streams; upgrades in calculation codes used to describe conventional fuels behaviour under irradiation, taking into account the specific issues related to MA-bearing fuels and sphere-packed fuels; and finally, progress related to the key issues of core physics, design and safety performance for sphere-packed MA-bearing driver fuels.*

I. INTRODUCTION

Minor Actinide (MA) incorporation into the fuel is a prerequisite for Gen-IV reactors to bring benefits in the disposal requirements by reducing the MA content in the high level wastes. Since americium displays a strong gamma emission (and curium a high neutron emission), 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 stage where Am would be more readily redistributed within the fuel than other actinides. Finally, harmful consequences of additional helium production during fuel irradiation (related to ^{241}Am transmutation) on fuel swelling, degradation of the thermal properties and high pressurization of the pins have to be prevented.

Based on historical experience and knowledge, oxide fuels have emerged in Europe as the shorter term solution to meet the Gen-IV Sodium Fast Reactors assigned performance and reliability goals and two main MA-recycle options are currently under consideration:

- the homogeneous mode, where small quantities (<3%) of MA-oxide are diluted in the $(U,Pu)O_2$ SFR standard driver fuel, in order to limit the impact of MA addition on SFR core safety parameters (sodium worth void, Doppler effects, delayed neutron fraction) and fuel cycle facilities;
- the heterogeneous mode, where MA-oxides are concentrated (contents from 10 to 20%) in UO_2 and MA-bearing sub-assemblies are located in the blanket of the core.

Investigations on both kinds of MA-bearing oxide fuels, started (more or less) recently and experimental knowledge remains mostly limited to laboratory-scale fabrication processes, small amount of characterization and out-of-pile testing, as well as scarce irradiation experiments [1].

The project PELGRIMM [2] that stands for PELlets versus GRAnulates: Irradiation, Manufacturing and Modelling, aims at investigating a new step in the long term process of the MA-bearing fuel qualification rationale, initiated within the European projects: ACSEPT (2008-2012), F-BRIDGE (2008-2012), CP-ESFR (2008-2013) and FAIRFUELS (2009-2015). 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, in order to:

- perform the Post-Irradiation Examinations (PIE) on MARIOS and SPHERE pins and fuels, providing the very first results respectively on the helium behaviour in $(Am,U)O_2$ fuels and a comparison between sphere-packed and pelletized $(U,Pu,Am)O_2$ fuel performances;
- take the next step in the $(Am,U)O_2$ fuel qualification rationale by performing a new irradiation 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 to describe the MA bearing fuel behaviour under irradiation;
- accomplish a preliminary design of a sodium cooled fast reactor core with spherepacked $(U,Pu,Am)O_2$ fuels and perform a preliminary safety assessment.

Finally, PELGRIMM aims at promoting the implication of European students and young researchers too, through: placements of internships in organisations involved in the project, travel grants to young scientists to attend conferences and workshops.

This paper gives an overview of the technical outcomes gained within the project.

II. PROGRESS STATUS OF IRRADIATION TESTS

As time required for an irradiation campaign on Am-bearing fuels, including the design of the test, the manufacturing and assembly of the components, the implementation in a reactor and the Post-Irradiation Examination program, is longer than the standard duration of an European project (3-4 years), irradiation campaigns have been regularly split in steps that are distributed in projects that follow each other.

The PELGRIMM project hosts the PIE activities for the irradiation tests SPHERE and MARIOS which were designed, manufactured and implemented in HFR within the FP-7 FAIRFUELS project (2009-2015). The design, manufacturing and implementation in HFR of the new irradiation test : MARINE (that stands for MA heterogeneous Recycle semi-INtegral Experiment) is part of the FP-7 PELGRIMM too. (MARINE PIE program will be included in a future European project.)

SPHERE [3] consists of two experimental pins containing an $(U,Pu,Am_{0.03})O_{2-x}$ fuel, shaped as pellets in one mini-pin and a stack of beads of 2 size fractions (50 & 800µm) in the other mini-pin. Indeed, if MA-bearing fuel pelletized stacks have been preferred so far, the implementation of the spherepacked fuel stacks could lead to a significant simplification of the fabrication process with the elimination of the steps that involve fuel powders (and dust). So, the SPHERE test emerges as a first of a kind since irradiation behaviour of spherepacked and pelletized stacks of an Am-bearing driver type fuel can be compared. The irradiation was performed (within the frame of FAIRFUELS) in HFR up to April 2015, leading to a maximum burnup of ~5at% and a maximum Linear Heat Rate around $300W.cm^{-1}$. Non Destructive Examinations will start soon after a mandatory cooling period of ~3 months.

MARIOS [4] is the first separate-effect irradiation performed in HFR (within FAIRFUELS) aiming at studying temperature- and porosity- dependent helium release and fuel swelling for tailored porosity disks of $Am_{0.85}U_{0.15}O_{2-x}$ fuels [5], using an astute experimental design that leads to flat temperature profiles in the samples. Thus, four mini-pins, each containing 6 small disks (diameter: 4.5mm and thickness: 1.5mm) of fuels with 2 open-porosity ratios (7.7% and 12.5%), were irradiated during 304 Equivalent Fuel Power Days at constant temperatures that ranged from 980 to 1370°C [6].

Neutron radiography performed after the irradiation has shown an overall good shape of the irradiated disks that has been confirmed for the pin 2 at the dismantling stage completed in HFR hot cells, as most the disks were recovered in 2 fragments of equal size.

Pins puncturing results have shown that helium release was total whatever irradiation temperatures and disk porosity contents. Fission gas releases have displayed temperature dependences regardless porosity ratios (see Figure 1). Finally, unlike axial distributions per disks of the non-volatile fission product ^{95}Nb, gamma spectrometry results for ^{137}Cs seems to match (qualitatively) the Fission Gas release rates (see Figures 2 and 3), showing that the temperature threshold for Cs release lies near the investigated temperature range.

This very first results on $(U,Am)O_2$ fuels will be completed in a near future by the Destructive Examination stage to be performed on pins 1 and 3, in the LECA facility at CEA-Cadarache.

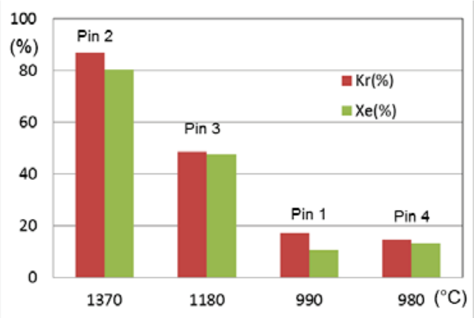


Fig. 1. Fission Gas release results versus temperature for MARIOS pins

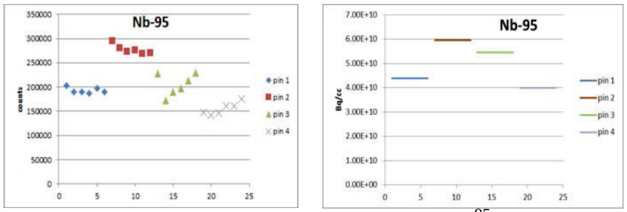


Fig. 2. Measured and calculated distributions ^{95}Nb per disk

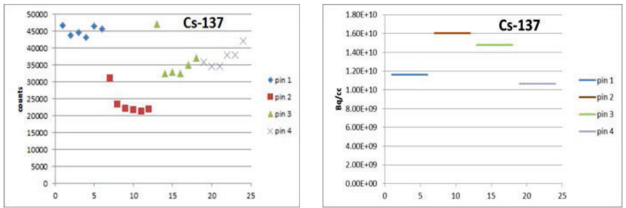


Fig. 3. Measured and calculated distributions ^{137}Cs per disk

The semi-integral irradiation test MARINE that is currently under preparation within PELGRIMM for an irradiation start by the end of 2015 for 336 EFPD, is part of the second stage in the long-range qualification approach of $(U,Am)O_2$ fuels (separated-effect tests like MARIOS belonging to the first step). MARINE is the SPHERE matching piece as it includes 2 mini-pins of pelletized and spherepacked fuel stacks of $Am_{0.85}U_{0.15}O_{2-x}$. Moreover, the MARINE mini-pins are instrumented with pressure transducers follow on-line gaseous release from the fuels during the irradiation.

The synthesis conditions of the fuels are summarized in section III. The design of the experimental device and the irradiation conditions (described in paper 5221 of these proceedings [7]) would lead at the end of the irradiation (336 EFPD) to fuel central temperatures of 850 and 990°C for the sphere-packed and pelletized pins respectively. So, outcomes from MARIOS at ~1000°C could directly be used for the interpretation of MARINE PIE results. Moreover, the helium production in MARINE (around $5 mg.cm^{-3}$of fuel) would be close to the $~4 mg.cm^{-3}$ foreseen in recent scenarios [8] for the heterogeneous recycling of Am in sub-assemblies close to the core of an industrial SFR of 3600MWth. Nevertheless, helium production rates in MARINE would not be representative as the MARINE duration would be significantly lower (336EFPD) compared the irradiation time in an industrial reactor (~2000 EFPD).

III. FUEL SYNTHESIS ROUTES INVESTIGATED

Even if powder metallurgy flowsheets, used to supply $(U,Pu)O_2$ standard fuels at industrial scale, can be used at lab-scale to prepare Am-bearing fuel samples (MARIOS disks for example), dust-free routes and simplified flowsheets, are essential to scale-up the Am-bearing fuel fabrication processes. Several options have been investigated within the project: some are based on sol-gel processes that lead to dense or porous spherical particles of homogeneous compounds as components are mixed in solution; another one consist on the adaptation to oxide fuels of the Weak Acid Resin technology initially implemented in the 1970's for the production of uranium carbide kernels.

Already, within FAIRFUELS, the SPHERE $(U,Pu)AmO_{2-x}$ beads were prepared by infiltration of porous $(U,Pu)O_2$ precursor beads (prepared by sol-gel gelation), with americium nitrate solutions. Two sizes of Am-bearing beads (50 and 800µm) were synthesized and heat treated. One batch fraction of small sized beads was then transformed to sintered pellets, which were then loaded in one mini-pin. The other batch fraction of small sized beads and the batch of bigger beads were packed by vibrations in the other mini-pin. For MARINE [7] the synthesis procedure was almost similar to SPHERE for the preparation of pellets, using UO_2 as precursor instead of $(U,Pu)O_2$. So, UO_2 porous beads were infiltrated by a low acid Am nitrate solution to prevent UO_2 dissolution as much as possible. The resulting granulates were heat treated, pressed and sintered. The dense (95%) and defect-free $U_{0.87}Am_{0.13}O_{1.93}$ pellets (see Figure 4) were then stacked in the MARINE 1 mini-pin.

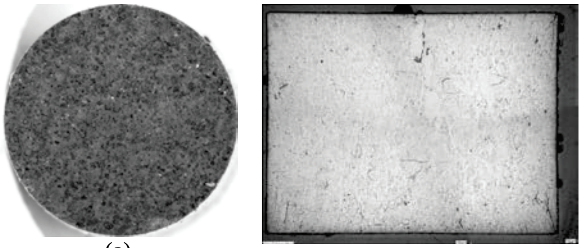

(a) (b)

Fig. 4. Visual aspect (a) and ceramography (b) of a MARINE pellet

The synthesis of the 2 diameter size beads (50 and 800µm) for the MARINE 2 mini-pin was much more challenging than expected and the two fractions were prepared by the external gelation method. $(U,Am)O_2$ sol-gel broth solutions were prepared and dispersed in droplets. After calcination, the beads for the large fraction (Figure 5) were sieved, while beads for the small fraction were crushed before sieving. After a sintering steps, granulates were vibro-packed to fill in in the second mini-pin and the smeared density is ~60%.

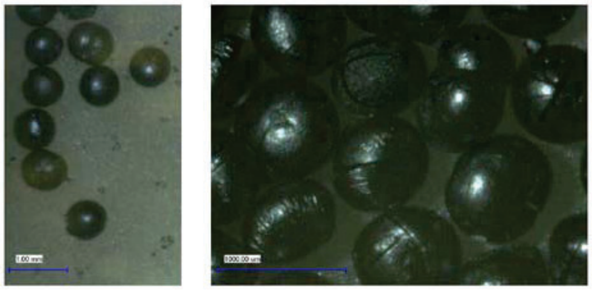


Fig. 6. Large size MARINE beads visual aspect

In parallel, a variant of the internal gelation route described in Figure 6, using for gelation of the drops, a microwave cavity instead of a silicon oil hot bath (that lead to secondary wastes) has been investigated on non-radioactive surrogates [9,10]. Promising results have been obtained (Figure 8) and implementation of the devices in glove-boxes started for validation on U-type compounds.

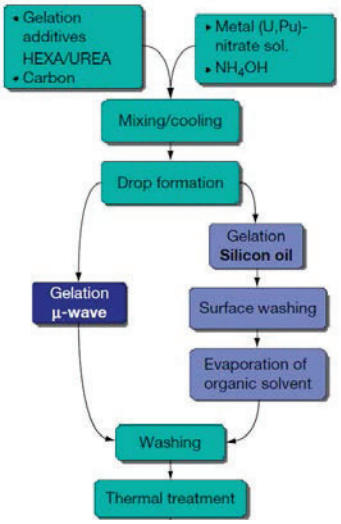
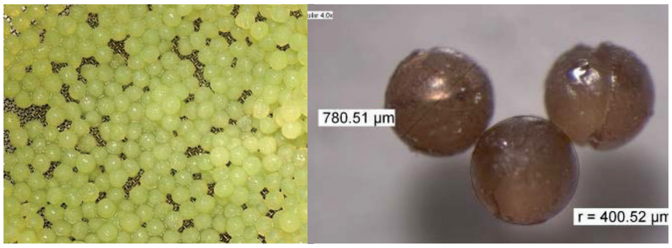


Fig. 7. Flowchart of the internal gelation route through a microwave cavity (right) compared to a silicon oil bath gelation step (left).



(a) (b)

Fig. 8. Spheres collected after microwaved gelation before (a) and after (b) drying.

Finally, the Weak Acid Resin flowchart has been revisited and adapted (Figure 11) to oxide fuels [11,12] up to the synthesis of (U,Am)O_2 beads and pellets[13].

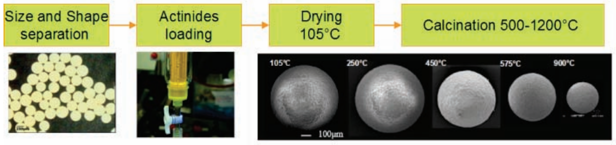


Fig. 11. Principle of the WAR process applied to Uranium dioxide based materials synthesis

A batch of 900mg of $U_{0.90}Am_{0.10}O_{2-x}$ microspheres was prepared by thermal treatments of ion exchange resins loaded with Am^{3+} and UO_2^{2+} cations. The degradation of the polymeric skeleton under air followed by reducing heat treatment led to the synthesis of spherical precursors with diameter around 400µm (see Figure 12) and apparent density of 24 % . A solid solution was formed during the reducing heat treatment and ensured a homogeneous distribution of uranium and americium atoms in the solid. Moreover, oxide microspheres were suitable for pressing and a dense pellet (density of 95%) was achieved after dynamic sintering under a reducing atmosphere up to 1800°C. This pellet meets the required specifications for dense pellet envisaged for Am transmutation on an UO_2 support in sodium fast reactors and proves the technical feasibility of this dust-free process.

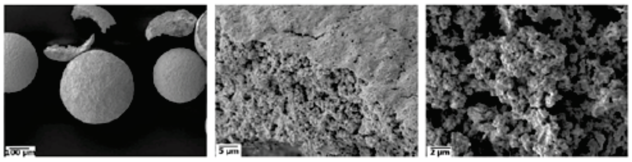


Fig. 12. FEG-SEM micrographs of $U_{0.90}Am_{0.10}O_{2-x}$ beads prepared by WAR technology [13]

IV. MODELLING AND SIMULATION OF FUEL BEHAVIOUR UNDER IRRADIATION

Most of fuel performance codes have been originally developed, verified and validated to model standard fuels for Light Water Reactor and Fast Reactor types. Upgrades of codes such as MACROS (developed by SCK•CEN), TRANSURANUS [14], SPHERE&SPACON [15] started within the frame of FP-6 EUROTRANS, FP-7 FAIRFUELS and FP-7 F-BRIDGE to consider some of the specificities for Am-bearing fuels and for spherepacked fuels. The objective of the work within PELGRIMM, is to capitalize on previous investments and to go further in the capabilities of the codes already mentioned plus GERMINAL [16], to describe the behaviour under irradiation of (U,Pu,Am)O_2 and (U,Am)O_2 fuels shaped as pellets or spherepacs.

Implementation of new compositions and fuel forms has been made by correction of existing models and correlations for standard fuels when appropriate. Such approach is practical; however, validity of it still shall be tested.

Moreover, the efforts have covered aspects of modeling such as helium production and release under a fast neutron spectrum; plutonium and oxygen redistribution [17]; melting temperatures [18]; evolution rate of fuel restructuring, columnar grain growth and central void formation; … For simulation of Helium release, a new model has been developed and the interpretation of some experiments by means of this model enabled to propose a new value for the diffusion coefficient of helium in uranium and uranium-plutonium oxides over a large and representative temperature range, which has been compared to the values in the open literature [19]. Furthermore, the interpretation of the data by means of the new model allowed to propose a value for the helium bubble diffusion coefficient, as well as for the trapping coefficient associated with intra-granular bubbles.

An assessment and a comparative evaluation of the fuel performance codes and their ability to model the behavior of fuel pins with either sphere-pac or pellets, has been initiated. Experimental data to use are issued from the SPHERE irradiation test for $(U,Pu,Am)O_2$ and SUPERFACT-1 [20] for $(U,MA)O_2$ fuel. First calculations with MACROS have shown a good agreement with PIE results for fission gas release, burnup and transmutation behaviour. Disagreements have been pointed out regarding fuel restructuring and central void formation phenomena that have been calculated whereas they are absent. Analysis of input data and models are underway.

V. SIMPLIFIED DESIGN AND SAFETY PERFORMANCE ASSESSMENT OF AN ADVANCED SPHERE-PAC $(U,Pu,MA)O_2$ SFR CORE

As sphere-packed fuels are foreseen to be good candidates for MA-bearing fuel concepts due to simplification of the fabrication process associated with the production of beads and due to potential good swelling behaviour under irradiation, the PELGRIMM project start linking the investigation of spherepacked fuel synthesis and behaviour under irradiation to problematics of core physics, design and safety performance.

This study is in continuity with the former FP-7 CP-ESFR project which aimed at designing and analyzing a 3600MWth sodium-cooled fast reactor loaded with a standard driver fuel shaped as pellets [21]. The so-called Working Horse (WH) aimed at a reduced sodium void worth, but still relatively high with ~1200 pcm at Beginning Of Life (BOL) and being a major cause for core disruption during an Unprotected Loss Of Flow accident (ULOF). Among the proposed solutions to optimize the core and reduce the positive void worth, the CONF2 core design with a large upper sodium plenum provided a reduced positive void worth [22]. So, the objective within the FP7-PELGRIMM project is to perform a safety assessment and a comparative evaluation of the CONF2 core loaded with either sphere-packed or pelletized $(U,Pu,Am)O_2$ driver fuel.

The PELGRIMM investigations started with the design of CONF2 loaded with sphere-packed fuel and the determination of core safety parameters and burn-up behavior. The neutronic analyses were performed with the MCNPX code. Variants of the CONF2 core contain Am fractions from 0 to 4 wt% in the driver fuel. This CONF2 design features a relatively low sodium void worth of ~ 500 pcm at BOL compared to the ~ 1200 pcm of the WH core. Results show the expected worsening of the safety parameters with burn-up and MA content. The Doppler coefficient is decreasing while Na void coefficient is increasing again up to 2100 pcm for EOC3.

The accident simulations are performed with the MAT5DYN code for the accident initiation phase and with the SIMMER-III code for the complete accident scenario. The codes had to be adapted to the specifics of the sphere-packed fuel. SIMMER analyses for the CONF2 core with pellet fuel have been performed first, assessing the claimed improvement by introducing the large upper sodium plenum. Results [23] showed that the sodium plenum seems to effectively prevent positive reactivity surges by voiding and subsequent power excursions. The low transient power allows the rewetting of structures and the disruption process remains limited. Differently than in the WH core after a first mild power excursion no recriticality appeared. For sphere-pac fuel under BOL conditions one might discern between BOL ‘green fuel’ and BOL restructured fuel after some hours of operation. The interest in the ‘green fuel’ case is because of the low thermal conductivity and consequently possible high fuel centerline temperatures. SIMMER analyses reveal that indeed under these conditions locally centerline temperatures above the melting point are reached. This shows that a starting procedure of the reactor is necessary to achieve a restructured fuel which is in its thermal conditions closer to a pellet fuel of same density. Both for the pellet and sphere-pac core, the plenum effect is active and prevents a scenario with multiple recriticalities. In the CONF2 core with BOL restructured fuel one experiences even a total re-wetting of the core in the last phase of the ULOF. These results reveal a new phenomenology not seen before for such large cores. For EOC3 cores the safety parameters are deteriorated and a ULOF in the CONF2 core again ends in whole core melting and core destruction.

No sound experimental data base on transient behavior of sphere-pac fuels exists. The safety analyses therefore represent first scoping studies. Based on the current analyses, the implementation of sphere-pac fuel does not cause any specific design problems for the CP-ESFR. For

BOL conditions the ULOF simulation shows a very mild transient for the sphere-pac CONF2 core both with un-restructured and restructured fuel. The first safety analyses also indicate that sphere-pac fuels do not seem to cause any specific safety problems, if introduced in an SFR.

V. CONCLUSIONS

The PELGRIMM project addresses MA-bearing oxide fuels for Generation IV – SFR Systems. Two options, MA homogeneous recycle in driver fuels and heterogeneous recycle in high MA content bearing UO_2 fuels are considered. The project aims at taking 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-packed fuel shape, from fuel fabrication and characterization to behaviour under irradiation; from experiments to modelling and simulation, as well as a spherepacked loaded core physics pre-assessment from normal operating conditions to transients and severe accidents.

The results on helium and Fission Gas behaviour in $(Am,U)O_{2-x}$ pins irradiated in the separated effect test MARIOS are currently available and show that: helium was totally released from the fuels. Fission Gas release rates are strongly temperature dependent and Cs release follows FG trend.

The semi-prototypic irradiation MARINE including pelletized and spherepacked $(U,AM)O_2$ fuels, to be implemented in HFR is almost ready to start.

The investigations to extend Minor Actinide bearing fuel fabrication processes to alternative routes, that are the Weak Acid Resins and micro-wave technologies, have given promising results.

Upgrades in models and correlations used to describe conventional fuels behaviour under irradiation, taking into account the specific issues related to MA-bearing fuels and sphere-packed fuels, are underway. Recalculations with several codes of the SPHERE and SUPERFACT-1 irradiation have been initiated.

Key issues of core physics, design and safety performance for sphere-packed MA-bearing driver fuels have been addressed.

At the end of the project that could be later than initially planned due to unexpected difficulties met in experiments, a new stage will have been reached in the long-term fuel qualification approach thanks to the skills, the involvement and the staunch determination of the PELGRIMM consortium.

ACKNOWLEDGMENTS

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NOMENCLATURE

BOL: Beginning Of Life
DE : Destructive Examinations
EFPD : Equivalent Full Power Days
FG: Fission Gas
MA : Minor Actinide
NDE : Non Destructive Examinations
PIE: Post Irradiation Examinations
SFR: Sodium cooled Fast Reactor
ULOF: Unprotected Loss of Flow
WH: Working Horse

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