SFR STATUS FOR ONGOING RESEARCH AND RESULTS: ADVANCED FUELS

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I. INTRODUCTION

The Advanced Fuel project in the framework of the SFR system development aims at investigating high burn-up minor actinide bearing fuels as well as claddings and wrappers withstanding high neutron doses (>200 dpa) and temperatures (core outlet temperatures: 550°C). The R&D topics of the Advanced Fuel project deal with fuel fabrication, fuel behavior under irradiation as well as pin clad and wrapper materials developments. (The feasibility demonstration of Uranium, Plutonium and Minor Actinides in the fuel cycle is addressed within the frame of a different project: Global Actinide Cycle International Demonstration (GACID) [1])

The Advanced Fuel project started on 21 March, 2007. It is conducted by CEA (France), DOE-INL (USA), JRC-TUI (Europe), JAEA (Japan) and KAERI (Korea).

II. CONTENT AND SCHEDULE OF THE AF PROJECT [2]

The main challenge for fuel developments for future SFR systems is the development and qualification of a nuclear fuel element (fuel, clad and wrapper types, compositions and designs) which meets the GIF goals. That means: achieving high burn-up (~20 at %), operating at high temperatures and recycling minor actinides into the fuel. High burn-ups will allow uninterrupted reactor operation over long periods of time and consequently, reduce spent fuel volumes, operation costs and eventually fuel cycle costs. High burn-ups are however associated with physical limitations, including dimensional stability of core materials, Fuel-Cladding Mechanical Interactions (FCMI) and/or Fuel-Cladding Chemical Interactions (FCCI), due to the swelling of the fuel. High temperatures will enhance the energy conversion ratio and consequently, the economic competitiveness of the reactor. High temperatures lead to further challenges for fuels and core materials developments too. Minor actinide incorporation aims at reducing the actinide content in the high level waste and consequently brings benefits in disposal requirements and potentially non-proliferation. Since americium is a strong gamma emitter, and curium a high neutron emitter, minor actinide incorporation in the fuels will necessitate shielding, remote operations by robots and simplification of the fuel fabrication process. Moreover the high volatility of Am components has to be managed during fuel fabrication and irradiation phases, where Am should be more

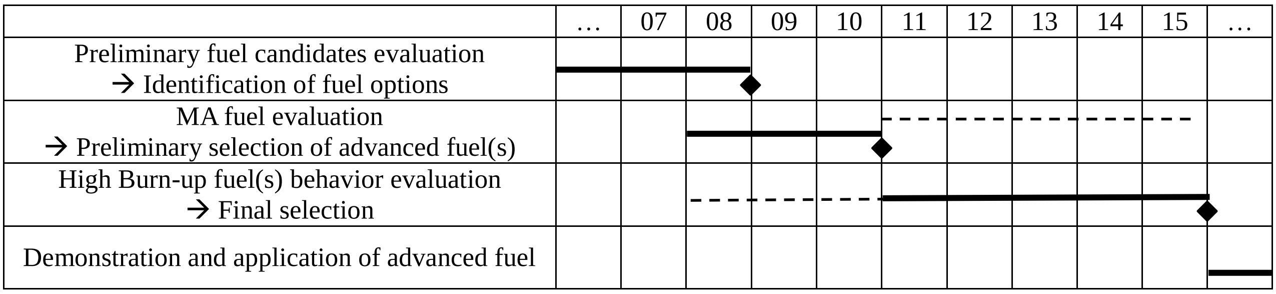

Figure 1: Main phases and milestones of the SFR Advanced Fuel project.

readily redistributed within the fuel than other actinides. Finally, the significant Helium production during fuel irradiation (related to ^{241}Am transformations) which can involve fuel swelling, degradation of the thermal properties and high pressurization of the pins is another major issue to be managed.

Based on background knowledge and past SFR experience, oxide, metal as well as nitride fuels (and more recently carbide[1]) are candidates under consideration. Oxide Dispersion Strengthened (ODS) Ferritic and Ferritic/Martensitic steels are the reference materials for the cladding and the sub-assembly wrapper.

The project consists of 3 major steps until 2015:

- The first phase deals with a preliminary evaluation of the candidate options (2007-2008) in order to identify the capability and the applicability of the fuels and the materials with respect to minor actinide incorporation therein, high temperature operation and high burn-up irradiation behavior. The evaluation concerns fabrication processes and behavior under irradiation. It aims at determining the next major steps to be undertaken for the use of these fuels too.

- After the identification of the advanced fuel options, major R&D efforts will be focused on fabrication feasibility and irradiation behavior of Minor Actinides bearing fuels (2008-2010). A preliminary selection of advanced fuel(s) will then be made.

- The third phase (2011-2015) will consist of the assessment of the high burn-up capability of advanced fuel(s) and materials.

The culmination of this path of research and development will lead to the demonstration and application of the advanced fuel design(s) in the SFR.

The steps and the schedule of the Advanced Fuel project are summarized in Figure 1.

Remark: Fuels under consideration up to now have been mixed Uranium-Plutonium based fuels as SFR driver fuel with MA incorporation up to a few percent, in accordance with the so-called homogeneous MA recycling in nuclear systems. The heterogeneous route for MA transmutation, for which MA will be concentrated in the fuel of the radial blankets is now under discussion with the System Integration & Assessment project and could be included within the frame of the AF project in the future.

III. WORK PERFORMED AND RESULTS

Information from background knowledge, past SFR experience as well as ongoing national and collaborative SFR programs regarding cladding and wrapper material developments, non-MA-bearing fuel performance and fabrication as well as preliminary knowledge performance and fabrication technologies of MA-bearing fuel have been collected and shared between the AF

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[1] Although the carbide option was initially discarded of the fuel candidates, it has been introduced since 2008 regarding the significant and globally positive experience on carbide behavior under irradiation.

project members. This enables a review of the capability of the fuel and material candidates, the identification of the issues and the selection of the advanced fuel options from oxide, metal and nitride fuels.

III.A. Oxide fuel evaluation [3]

Oxide-based fuel has been the choice of most development and demonstration SFR programs worldwide in the past. Oxide fuels have thus reached an industrial maturity. The MOX fuel database is well established and collected data from Post Irradiation Examinations (PIE) have been used to develop and validate fuel performance codes such as GERMINAL (CEA) and CEDAR (JAEA). Despite two major drawbacks: poor thermal conductivity and chemical reactivity with sodium, the uranium plutonium oxide fuels show major advantages including high melting point (>2 400°C) and excellent stability under irradiation within a broad range of temperatures and burn-ups (up to 10 at%). Moreover, the low thermal conductivity presents some benefits, as the fuel center temperatures and radial thermal gradients under irradiation are high, enabling a high fission gas release fraction in the pin plenum (80% for a 10 at% burn-up) and thus a low overall fuel swelling (0.6-0.7%/at%). Regarding sodium-fuel reactivity, experimental studies as well as operational feedback from cladding rupture in experimental reactors have shown solutions to manage this drawback.

Regarding high burn-up capability of oxide fuels, FCMI seems to be manageable through the smeared density (which is defined as the ratio of the cross-sectional area of the as-fabricated fuel to the cross-sectional area defined by the cladding inner diameter). On the opposite, FCCI is a major issue with two inner cladding corrosion phenomena occurring at high burn-up, *i.e*. fuel cladding reaction and the fissile-fertile interface reaction. Fuel cladding reaction results in cladding wastage on a rather large area in the upper part of the fissile stack. The latter FCCI type affects a much more localized area located near the interface between fissile and fertile stacks. As these FCCI phenomena are partially linked to high Oxygen to Metal ratios of the fuel (O/M), low O/M ratios have to be investigated to reduce the cladding depletion. Thus fuel fabrication technology has to be adapted to provide the appropriate O/M.

Regarding presence of MA in the fuel, main issues include:

- fuel fabrication with high Am retention due to americium oxide volatility;
- MA addition effects on fuel properties and fuel microstructure (restructuring as well as redistribution of oxygen, plutonium and americium under irradiation);
- impact on FCCI;
- and fuel behavior in transients.

Some issues have partially been addressed with the preparation or the examination of MA bearing fuels irradiated within the frame of SUPERFACT-1 [4] and Am1 [5] experiments. SUPERFACT-1 performed in the 80's has provided the first results on the incineration of minor actinides in the homogeneous mode and has demonstrated the general good behavior of the MA-bearing fuel up to 6.5 at% at low linear power (~40 W.m^{-1}). The pellets were manufactured by sol-gel processes and were composed of solid solutions: $(U_{0.74}Pu_{0.24}Am_{0.02})O_2$ and $(U_{0.74}Pu_{0.24}Np_{0.02})O_2$. Post irradiation examinations have shown that the fuel underwent a similar microstructure evolution as standard fuels and neither Pu nor Am redistribution was found. Finally, a large helium release was measured in the americium bearing fuel (4 times greater than standard fuel). Additional irradiation data have been provided on Am (3 and 5 wt%) and Am/Np (2%/2%) bearing fuels by the Am1 irradiation test performed in Joyo: remote fabrication technology has been established at laboratory scale; out of pile measurements (melting point, O/M ratio, oxygen potentials, …) have been performed; PIE have shown that structural changes such as formation of lenticular pores and central void occurred within the first 10 minutes of the irradiation; no signs of fuel melting were found.

Finally, regarding the fabrication of MA bearing fuels for industrial applications, the

Powder Metallurgy standard process which generates dust has to be modified and/or simplified to limit the steps of powder handling. Prospects for the production of MA oxide fuels can be based on co precipitation or sol-gel methods. Pin manufacturing could be simplified too, using for instance a spherepac type process.

III.B. Metal fuel evaluation [6]

U-Zr and U-Pu-Zr alloy fuels with a sodium bonded fuel pin, were selected for many of the first SFR studies in the U.S.. Advantages of metallic fuels are their high thermal conductivity, high fissile density and available experience on metallurgy fabrication processes. An extensive database of their performances was generated [7], burn-ups of 10 at% at steady state were reached in reactors. Metal fuels capabilities have also been demonstrated up to 19 at% burn-up with Ferritic/Martensitic (F/M) steel clad. Moreover, metal fuels have been shown to exhibit sufficient margins to failure under transient conditions, despite their low melting temperature (<1 000°C).

One major drawback for U-Pu-Zr fuels is FCCI due to high fuel swelling especially at high burn-ups, which can lead to inter-diffusion phenomena between fuel and clad components, and then to the development of low temperature melting phases. As a consequence, fuel smeared densities and peak clad temperatures, have to be managed in order to prevent FCCI. FCMI seems not to be a major issue due to the high plasticity of metal fuels, if a large fuel-to-cladding gap as well as a large pin plenum, are available to accommodate fission gas. Other issues are sodium bond requirement and difficulties in modeling metal fuel behavior under irradiation due to the complexity of the U-Pu-Zr phase diagram.

The main issues for MA-bearing metal fuels are the same as for oxide fuels, with in addition, demonstration of:

- MA-bearing oxide feedstock reduction to metal alloy feedstock;
- an acceptable level of FCCI.

Some issues have already been partially addressed in the framework of the preparation or examination of MA bearing fuels, irradiated in the frame of X-501 [8], AFC1 [9] and METAPHIX-1 [10,11] experiments. The X-501 experiment has demonstrated the acceptable behavior up to ~6.5 at% of a U-20Pu-10Zr fuel containing 1.2 wt% Am and 1.3 wt% Np. The microstructure of the irradiated fuel is similar to U-Pu-Zr and FCCI of the HT-9 F/M clad is not strongly affected by small amounts of Am and Np. AFC1 metal compositions have shown excellent performance up to 8-10 at%. Non Destructive Examinations of METAPHIX-1 have shown neither crucial damage nor excessive deformation for metal fuel rods containing 5wt% or less MA, irradiated up to 2.5at%. In contrast, these experiments have shown that mechanical and thermal properties have not been seriously degraded by the addition of MA elements.

Finally, regarding fabrication of MA bearing fuel for industrial applications, a promising technique could be precision injection casting, which would provide a fuel slug without textured structure and which require a relatively short fabrication sequence easy-to-build and easy-to-use equipment.

III.C. Nitride fuel evaluation

Nitride fuels were identified as candidates for SFR, nearly three decades back, on the basis of their attractive physical and chemical properties *e.g.* a high heavy metal density, a strong thermal conductivity connected with a high melting temperature (>2 700°C) as well as a good compatibility with stainless steels, sodium, water (T≤60°), air (T≤25°C) and hydro-reprocessing. So, improved performances of nitride fuelled core (in comparison to oxide) such as a larger breeding ratio, higher linear heat rates and an improved safety have been expected. Nevertheless, fuel dissociation of (U,Pu)N fuels at a temperature substantially lower (~1 730°C) than the melting point if nitrogen overpressure is not maintained, has been identified as a critical issue in case of severe accidents. Another key issue consists of the ^{15}N enrichment requirement to prevent mostly the generation of the radiotoxic long lived ^{14}C from $^{14}N(n,p)$ reaction and additional He production from $^{14}N(n,\alpha)$ reaction.

The worldwide experience on nitride fuels has been limited to 150-200 irradiated pins for maximum burn-ups of 9 at% and linear heat rates of 45-130 $W.m^{-1}$. The overall swelling of nitride fuels fits a linear rate of 1.1 %/at% below a critical temperature which decreases from ~1 200 to ~950°C with increasing burn-up. Beyond the critical temperature, the swelling rate increases exponentially before being restrained by the cladding. Because of the high swelling rate, fission gas release remains low (<50%) even at high burn-up.

For high burn-up applications, FCMI is the major issue to manage in order to prevent large clad deformation and clad breach. It could, nevertheless, be solved by acting on the smeared density (~70-80%) and by favouring fuel open porosity for gas release.

For MA bearing fuels, because of the limited thermal stability of MA nitrides, MA redistribution could occur for high linear power or during power/temperature excursions. Furthermore, moderate temperature fabrication techniques have to be found to prevent Am losses.

To partially address these issues, experimental data from irradiation experiments on (U,Pu)N fuels (NIMPHE-1, NIMPHE-2 [12] and BORA-BORA [13]) as well as on MA bearing fuels (AFC1 [14] and FUTURIX-FTA [15]), are under analysis.

III.D. core materials evaluation [16]

The extremely high flux of fast neutrons in a SFR core is a main source of damage to subassembly materials used for the pin cladding and wrapper tube. Increasing final burn-ups (~20 at%) and core outlet temperatures (550°C) in Generation IV SFRs, imply that fuel structures must support both extremely high irradiation doses (~200 dpa) and higher peak temperatures: ~580°C for the duct and 650-700°C for the cladding according to the fuel type.

Austenitic steels have excellent material properties at high temperature and an acceptable swelling capability up to ~160 dpa, satisfying the requirements for the cladding of the current SFR systems. To achieve a higher burn-up in the Generation IV SFR system, steels with superior swelling resistance characteristics need to be utilized. To this end, F/M steels have been considered as primary candidates for the cladding and duct materials. Past experience on these steels has shown an excellent swelling resistance up to 200 dpa. However, these steels don’t have sufficient creep strength above 650°C to meet GIF goals except for metal core designs. The HT9 F/M steel has then been selected as a promising candidate for ducts and metal fuel cladding.

To extend the range of F/M steels to temperatures well above 650°C, Oxide Dispersion Strengthened steels made of a fine distribution of oxide particles in a F/M steel are promising candidates because of their high temperature strength.

IV. CONCLUSION

International collaborative activities are performed on fuel and core material developments within the Advanced Fuel project for Generation IV SFR systems. As a first milestone of the project plan, the R&D outcomes of national and collaborative programs have been collected and shared between the AF project members in order to review the capability of oxide, metal and nitride fuels and core materials candidates, to identify the issues and select the viable options.

Based on historical experience and knowledge on fast fuel development, as well as specific fuel tests currently being conducted on MA bearing fuels, both oxide and metal fuels emerge as primary options to meet quickly the performance and the reliability goals of Generation IV SFR systems. As the irradiation performance database for nitride fuels is limited, even if their attractiveness is high, these fuels are at an early stage of development with longer term R&D activities still required.

The status of core materials such as cladding and duct has been reviewed. The promising candidates are F/M and ODS steels.

The next step for the AF-PMB consists in introducing carbide fuels in the assessment (2008-2009), gaining knowledge and solving issues regarding core materials, performance and fabrication technologies of MA bearing fuels, from national and collaborative programs. A primary selection of advanced fuel(s) is expected by 2010.

List of abbreviations

AF: Advanced Fuel

F/M: Ferritic / Martensitic

FCCI: Fuel-Cladding Chemical Interactions

FCMI: Fuel-Cladding Mechanical Interactions

MA: Minor Actinides

MOX: Mixed Oxide

O/M: Oxygen to Metal ratio

ODS: Oxide Dispersed Strengthened

SFR: Sodium Fast Reactor

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