Overview of French RandD on SFR MOX fuel fabrication

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The manufacturing of the (U,Pu)O_2 mixed fuel pellets designed for SFR are being conducted by the french actors CEA with support from ORANO and EDF, carrying out an approach covering all the manufacturing steps, from the implementation of the UO_2 and PuO_2 actinide oxide powders to the pellet sintering. A reference fabrication process has been proposed, considering the experience feedback from fuel manufacturing for Phénix and SuperPhénix reactors. The recent developments deal with the increase of the Pu content (up to 30%) and the effects of aged Pu (thermal and radiolytic effects) which are among the main challenges to sustain the closed fuel cycle. The R&D focuses firstly on experimental studies to deepen the understanding phenomena involved in the reference process. Further, a physical modelling is developed to build a simulation tool in order to improve the performance of each manufacturing steps in a lab-to-plant approach. Modelling and simulations are based on specific experiments (grinding and mixing process, mechanical measurement on tailored actinide powders, microscopic characterisation…).

Advances has been achieved in the understanding and modelling of physical phenomena describing the phenomenology of each steps. However, numerous challenges remain today in the modelling.

I. BACKGROUND

The French Atomic Energy Commission (CEA) is developing a large scale SFR simulation program which rationale is to maintain the closed fuel cycle (Ref. 1). Studies carried out during almost ten years, within the framework of the French Astrid Project (Refs. 2-6), led to the definition of reference process flowsheet for the fabrication of SFR fuel pellets. In particular, high Pu content fuel (up to 30% heavy metal) has been considered and the impact of degraded isotopic compositions leading to increased thermal constraints and radiolysis.

This paper presents a general overview of the French R & D program on the SFR fuel fabrication summed on the schema below.

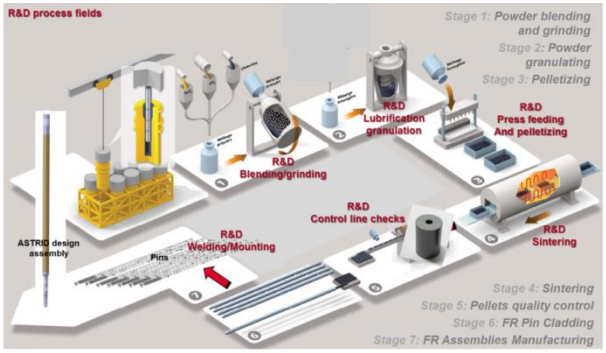

Fig. 1. SFR fuel fabrication process at a glance

II. FEEDBACK OF INDUSTRIAL FR FABRICATION (REFS 2-3)

The fuel fabrication plant of Cadarache (ATPu) was built to produce plutonium-based fuel within the scope of programs conducted by the CEA. Between 1962 and 2005, 25 tons of plutonium have been carried out to produce 450,000 fuel pins, corresponding to more than 110 tons of oxides for different SFR i.e. Rapsodie, Rapsodie-Fortissimo, Phenix and Superphenix, as well as a reload fuel for the Prototype Fast Reactor (PFR) and various experimental fuels. Since 1988, the ATPu manufactured also MOX fuels for LWRs at the same time, which required re-organising the process and the glove-boxes.

Two processes have been used in the ATPu from 1971 to 1999, the so-called 'RNR' process and the 'COCA' process distinguished by the granulation step: mechanical granulation / sieving for the RNR process ; size screening / forced sieving in the COCA process. The granulation step involved in the COCA process leads to a best compromise in term of microstructure homogeneity.

III. REFERENCE PROCESS FLOWSHEET

In the frame of the ASTRID program (Refs. 4, 6), led by the CEA, ORANO (ex-AREVA) committed a partnership with the CEA for the preliminary definition of industrial facilities for the future, to support fuel and core fabrication for ASTRID needs during the operational life of the reactor. These facilities designs lean on CEA and ORANO extensive experience in mixed oxide Fast

Reactor (FR) and LWR fuels manufacturing, at first in the ATPu, and at the MELOX Plant. Additional features are integrated to cope with the evolution of the raw materials, fuel design and regulatory requirements.
To design and define the process in term of capabilities and safety, the highest Pu content is 35% (Pu/(U+Pu)) with almost 4% of Pu238.

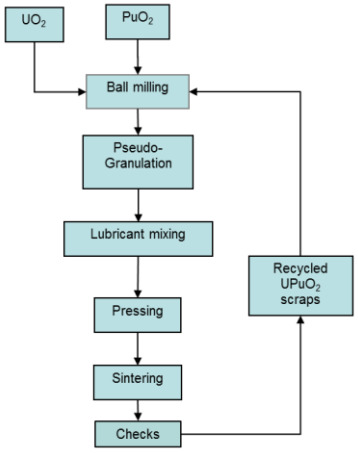

Fig. 2. Standard reference fabrication process of French SFR fuel

One of the objectives of R&D is to ensure that fuel pellets comply with manufacturing and control specifications, the main ones, the geometric and appearance characteristics given below :

- O/M ratio in the range]1.94-2.00[, with a target value of 1.97
- UO_2 agglomerate size smaller than 200 µm
- PuO_2 agglomerate size smaller than 30 µm

Density greater than 94% of theoretical density, with a target value of 97%.

III. R&D FIELDS

III.A. Raw oxide powders

The variability of raw materials UO_2 and PuO_2 is a first order parameter to assess the robustness of the process steps. Considering the diversification of the UO_2/PuO_2 powder supply, from dry or wet route, and the degradation of the fissile properties i.e. the evolution of the isotopic composition, it is worth studying the effect of various powders and the thermal load induced, especially on the organic product involved in the process.

III.B. Co-milling / mixing

In the fuel fabrication process for fast reactors, UO_2 and PuO_2 raw powders are together milled and mixed with recycled scraps (up to 20wt%) to make an intimate mixture and to obtain a specific area supposed to improve the efficiency of the subsequent sintering regarding the Pu homogeneity. This operation is classically performed in a uranium ball pebble mill. A methodology is developed to better understand the influence of the ball size on the characteristics of the powders whose properties determine in part the behavior of the granular medium in subsequent steps. The goal is to transfer lab results to the industrial scale using a transfer function. Modelling based on a discrete element approach (DEM) is developed by considering breakable cell bodies simulating rigid powder grains (Ref. 7) (see fig. 3). The motion and fragmentation of these cells is modeled and coupled in a rotating drum. Crushing modelling and simulation, the main issues, will help to understand and define targeted experiments using ball mills of different capacities, from 10^{-1} to 10^{0} L in the laboratory (see fig. 3), up to 10^{2} L in production at MELOX Plant in the framework of semi-industrial manufacturing runs in the MELOX Plant.

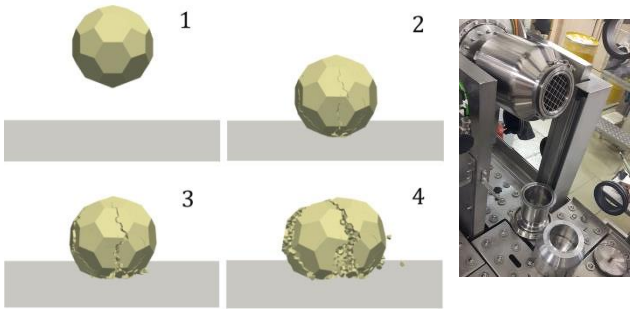

Fig. 3. A particle impacting a rigid plane, and the evolution of particle breakage. This test was performed with an impact velocity of 6 m/s.
15L capacity pebble mill (right side)

The simulation of grinding is obviously complex because of the number of parameters involved and the mechanisms to be simulated. In particular, the fragmentation of particles leads to a large amount of objects and developments are necessary to reach a sufficient level of physical description of the mechanical loading mode and, numerically, to perform parallel computing and increase the number of particles implemented. Furthermore, smart experiments have to be defined and carried out to build a reliable demonstration.

III.C. Granulation

The poor flowability of actinide powders, which are generally cohesive, define one of the main challenge we have to face in the process route, e.g. to fill the press molds in a reproducible and homogeneous manner.
The aim of granulation, in fact covering deagglomeration, reagglomeration, internal lubrication and spheroidization,

is to improve of the flowability of powders obtaining a targeted granular medium.
The flow kinetics of the powder embed static and dynamic aspects. Conventional characterization techniques give easy access to the physical characteristics of agglomerates such as their shape or particle size distribution. Deepening characterization of the rheological properties of the grains is necessary to define the properties of the targeted granular medium.
Parametric rheometer tests carried out on tailored UO_2 powders batches gave flow functions and, further, the bulk level cohesion of the powder. These data must be known to control the flow parameters of a powder, more or less consolidated in a stationary state.

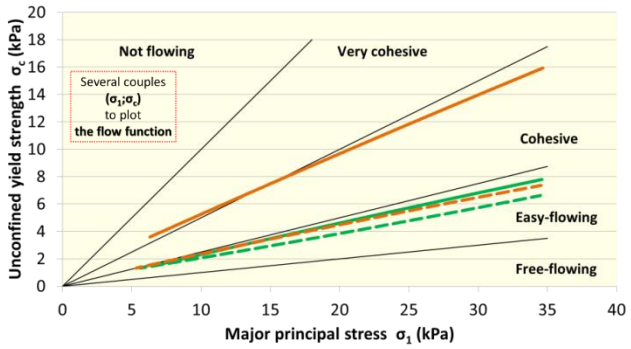

Fig. 4. Flow functions and bulk level cohesion (Ref. 8)

To characterize the dynamic flows, among required properties to be known, permeability measurement, based on the energy necessary to stir the powder at a given speed (Ref. 8).
A second objective is to go towards the simulation of the flows, integrating physics involved.
Macroscopic studies are also of interest considering that the handling and the flow of very angular particles is limited by strong friction effects which can be overcome by the help of transverse vibration.

III.D. Pelletizing

An efficient sintering step imposes the specification of incoming green pellets which have to be homogeneous, dense and without any density gradient. The understanding of the embedded physics during the pelletizing must be better understood, from the filling of the molds to the compaction / compression of the powders before the ejection of the green compact.
The pressing conditions (mechanical strain loading) determine the resistance of the green pellet and its behavior during ejection and handling until sintering. The performance of the pressing step must therefore be put in perspective with the sintering conditions on the other hand. The evolution of the granular structure during mechanical loading is still poorly known and difficult to explore by experimental means. Simulation of powder compaction is an alternative way to study the phenomena but the properties of the powders have to be integrated (fragmentation of the particles, and their change of shape and size). Simulations, based on FEM or DEM codes have then been conducted (Ref. 9,10).

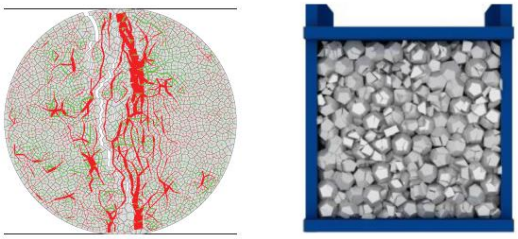

Fig. 5 : DEM Pelletizing simulations

Further work will deal with the possibility to define the pelletizing parameters according to the powder properties and the set of mechanical properties of the green pellets such as three- and four-point bending tests and diametral compression tests, used to measure the mechanical strength of green compacts. Finally, to predict the sintering behavior, distribution of U and Pu atoms have to be assessed.
For the shaping and its technological aspects, the weaknesses and strengths of the hydraulic and rotary presses were compared. The rotary press option was chosen because of the production rate, a better reproducibility of the pressing cycles and the reduction of the dispersion of the diameters of the pellets. The rectification step (grinding diameter correction) is thus not needed.
Another issue has to be addressed: the lubrication. Indeed, the thermal load of the powder due to the degradation of the isotopic composition will alter the lubricant properties during the pelletizing step and then increase the friction between the grains and between green pellet and the press mold wall.
To conclude, both a technological and a scientific approach are then required to describe the compaction and further the compression, and establish the link between the flowing properties of the granular medium.

III.E. Sintering

The sintering step is defined as a choosen high temperature treatment used to consolidate green pellet, densifies the material (target density: 97% of theoretical density), forms the solid solution $(U,Pu)O_2$ (up to 30% Pu) and reaches the O/M ratio target. The sintering cycle generally comprises a temperature rise with a linear ramp, a constant temperature plateau and finally a controlled cooling, and, advantageously, with an adaptation of the oxygen potential through the H_2/H_2O ratio. The sintering of the SFR fuels is performed in a more reducing condition than that of the PWRs and requires a precise adjustment of the oxygen partial pressure of the sintering gas, in contrast to the PWR fuel for which the stoichiometry 2.00 is reached for a large range of oxygen potential.

The SFR fuels will have Pu contents up to 30% Pu, higher than those of SPX and those specified for the last core of PX. Increasing the Pu content of the fuels requires adaptations of the sintering process to reach the specified O/M ratio and avoid problems due to phase change or stoichiometric effects.

Beyond the intrinsic complexity of the sintering mechanisms, essentially multi-physics and based on strong couplings, a sharp physical description have to be taken into account in the models. The definition of the REV (Representative Elementary Volume) is a key issue to perform scalable simulations (from the grain set to the pellet) with a known level of uncertainty. The interdependence of the simulated physical phenomena and the large range of order of magnitude in terms of time and specific distance represents difficulties to overcome.

An important objective for sintering modelling is to establish a link with the data set resulting from the simulation of the input data sintering (residual density and stress gradient, U / Pu distribution and distances between these elements) assumed to be validated by an *ad hoc* experiment.

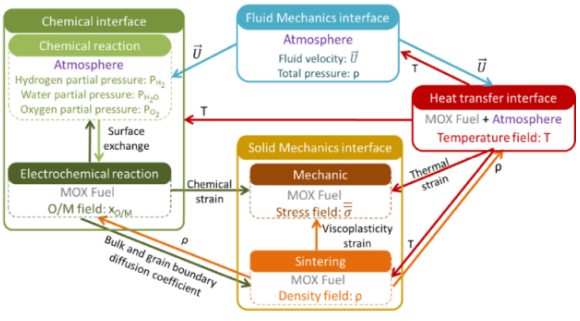

Figure 6 : Multi-physic modelling of the sintering step

The experiment will also focus on the mesoscopic scale (10^{-7} to 10^{-3} m) to study sintering and transport mechanisms, and therefore the defects that allow this diffusion, and then build the microstructure, to feed the modelling and to increase the predictive character of the codes.

It is also important to highlight the importance of looking for information to calibrate the models at the nanoscale using means that are available today, such as the environmental SEM or the heating TEM, to visualize, in the proper sense, the formation and evolution of sintering necks, and, more precisely, the evolution of crystalline orientation in the flux zone. The comparison of these data with those obtained from the self-diffusion and interdiffusion tests will be important in order to better control the sintering thermal cycle (ie heating rate and temperature) and, consequently, the microstructure of the sintered ceramics. Sintering Curves or MSC (Master Sintering Curves) will be established to feed the modelling.

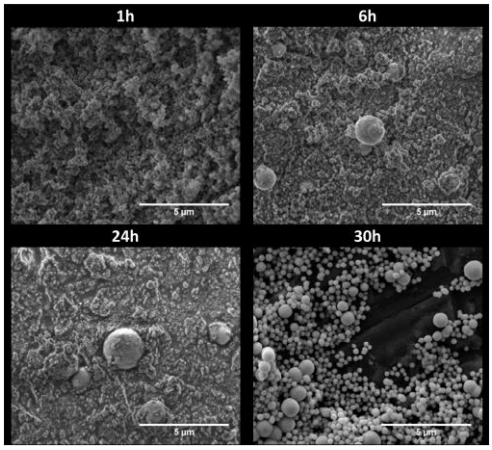

Fig 7. SEM micrographs of the powders obtained after hydrothermal conversion of uranium IV aspartate at 160 °C for different durations (Ref. 11)

IV. INDUSTRIAL SCALE FABRICATION TESTS (REFS. 5,6)

IV.A. Background

As part of ASTRID's first core qualification, ORANO and CEA teams early launch tests in the MELOX Plant to prepare the manufacturing of SFR fuel. Industrial manufacturing campaigns were carried out at MELOX in 2015 and 2016.

First campaign (2015) of tests was dedicated to demonstrate the feasibility of this manufacturing at half industrial scale. Specified analyses results were performed in MELOX plant laboratory, completed by more specific characterizations at CEA.

Second campaign (2016) of tests, included a powder preparation step at industrial scale on one of the blender of the MELOX plant, in order to prepare the industrial

manufacturing of MOX pellets for one fuel bundle, designed for a prototypical irradiation.

IV.A. Main results and lessons learnt

A first test was held at MELOX in 2015 to study of the effect of pelletizing conditions, the thermal sintering cycle (carried out under Ar-H2 (4%v) humidified at 360 vpm H2O) and the type of recycled scraps (2h grinded sintered pellets) coming from LWR or SFR type fuel. The first batch meets almost all the requirements including the plutonium distribution. Optimized sintering treatment, comprising a sharp humidification cycle, have been studied and performed to avoid any defects in the microstructure and obtain the specified O/M ratio.
The incentive of the 2016 test was to study the influence of manufacturing parameters such as grinding on a larger scale (on the production mill) and the robustness of sintering treatment. The pore spectra obtained from the microstructure characterization range between 1 to a few tens of μm. Dense islands or porosity have been observed, similar to those observed in the pellets of the 1999 Phenix campaign of ATPu. The Pu distribution meets the requirement whatever is the sintering conditions. However, over-milled type microstructure have been sometimes observed due to an excess of energy released in the powder in the production mill.
A CEA-MELOX working group was also set up to optimize the conditions for preparation and characterization of these samples, which focus on difficulties on the industrial ground to check and control the SFR pellets which include the O/M ratio, the Pu rich agglomerate size, ceramography. The quality of ceramographic preparation has been improved between 2015 and 2016 tests.

V. FUEL FABRICATION SIMULATION AT A GLANCE

There is no qualified simulation tool for the fabrication of nuclear fuel. The objective is to model and then simulate all the manufacturing steps, which will be chained together, to describe and further predict the output. Finally, the simulation tool will be part of the *PLEIADES* multi-scale simulation platform (Ref. 12) dedicated to fuel behavior for the specific application *GERMINAL* (in-pile SFR fuel pin behaviour).
Building this simulation tool demands to address many challenges :

- The first challenge relates to the level of understanding of each manufacturing steps.
- The second challenge deals with the possibility to build a reliable multi-scale model able to describe the phenomena from the atomic level to the pellet scale (10^{-10} to 10^{-3} m), and further, from the laboratory to the manufacturing plant. This challenge represents a key point to go towards a rigorous simulation of fuel fabrication. The notion of multi-scale modelling covers indeed a particular reality : each step, spatio-temporally dissociated from the previous, provides output data to be integrated for the following step.

For the future, smart experiments have to be defined and carried out to develop reliable modelling on specific surrogate systems. Incoming results and further insight into modelling will then fill the gaps and the lack of knowledge for each step.
HPC issues have to be address regarding the performance of simulation tools, in terms of accuracy (uncertainties propagation) as well as the calculation time involving parallelization and coupling DEM and FEM approaches.

VI. CONCLUSION AND FUTURE OUTLOOK

This paper gives an overview of the french SFR fuel fabrication R&D program, an integrated part of the french SFR simulation program currently defined at CEA.
Efforts will focus on the comprehension of physical or chemical phenomena, and, further, on the modelling in order to perform reliable simulations in a multi-scale and/or lab-to-plant approaches for each fabrication steps and for the entire manufacturing scheme. Industrial feedback is then of great importance.
The main scientific issues to be addressed are listed below :

- Characterizations of flowing properties of actinides powders (PuO_2, UO_2 and $(U, Pu)O_2$).
- Discrete modelling of the powder front end of the process that is to say grinding, granulation, and filling of press molds as well as the transitic and mechanical stability of powders in the plant.
- Multi-scale and multi-physic modelling of each steps assumed to be coupled.
- Validation of simulation through designed and specific smart experiments.

Simulation tools will be used to evaluate the robustness and the flexibility of the process steps focusing in particular on the variability of the inputs (UO_2/PuO_2 powder supply, from dry or wet route) and the thermal load due to the evolution of the isotopic composition (aged Pu). Modelling and simulation will also provide elements to predict and optimize manufacturing parameters.

ACKNOWLEDGMENTS

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