Design and first operation of the DIAMINO ($U^{241}AmO_2$) experiment in OSIRIS MTR for Am-recycling program

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ABSTRACT

In the framework of actinide recycling in future GENIV Sodium Fast Reactors, the DIAMINO experiment has been developed to investigate gaseous release and swelling of Americium Bearing Blanket (AmBB) in neutron flux. The experiment takes place in the OSIRIS pool-type reactor (70 MW). Production of helium obtained in a thermalized spectrum will be close to that expected in a fast reactor.

The irradiation device consists in experimenting the behavior of the AmBB in different conditions, with two types of matrix microstructure (dense and porous), two temperature levels (600°C and 800°C) and two ^{241}Am compositions (7.5 % and 15%).

Before the irradiation of the real sample holder, a mock-up has been tested to optimize some input parameters used in thermal model, especially the level of gamma heating in the periphery of the OSIRIS core.

This article describes the design of the device based on accurate thermal and neutron calculations. It also details the control aspects to target the different samples temperatures and fissile power evolution during irradiation. Lastly, the simulation results with the first irradiation measurements are compared.

1. Introduction

Future GEN IV sodium-cooled fast reactors (SFR) may include Americium (Am) recycling in radial blankets positioned in the outer core regions. Heterogeneous Am-recycling on a UO_2 matrix would have the advantage of providing flexible breeding gains and Am burning with limited impact on core operating and safety (especially proliferation risk).

The behavior under irradiation of such blankets containing a significant quantity of Am in a $(U, Am)O_2$ solid solution must nevertheless be studied and characterized in greater detail. It is known in particular that a large quantity of helium will be produced under irradiation, mainly resulting from the transmutation chain of ^{241}Am. The impact of helium production on gas releases and pellets swelling must be identified for the design of Americium bearing blanket

(AmBB) assemblies, especially considering the expected moderated AmBB irradiation temperatures (that could lead to significant swelling).

The main goal of these experiments, with isothermal-type conditions in the experimental blankets, is to study Am-bearing blanket gaseous release and swelling as a function of temperature, blanket microstructure and gas production rates. The considered temperature range for the DIAMINO experiment (600°C - 800°C) in OSIRIS Material Testing Reactor is in the scope expected for AmBBs in an SFR, i.e. about 500 to 1500°C [1]. Two microstructures are being investigated : a standard dense one and an optimized one with a significant fraction of open porosity (in order to improve helium releases and then to limit gaseous swelling). Moreover, the transmutation rate is accelerated in MTRs (compared with SFRs). Two helium production rates, through 2 irradiation durations, are being studied in order to check the expected envelope effect of transmutation acceleration on AmBB swelling.

2. DIAMINO device description

2.1 Loading sample holder

The irradiated samples are $UAmO_2$ discs with a concentration of 7.5% and 15% (by mass) of ^{241}Am. These discs, splitted into 4 batches, are inserted into a TZM cup. The stack of six cups is then inserted in leaktight Inconel 600 cladding sealed by two laser-welded end plugs. An exploded view of the assembly, called needle is presented below. Two N type thermocouples are introduced in two thermowells on the top of the upper loading needle. The six needles containing the six batches are inserted in the DIAMINO sample holder which is itself inserted into a PHAETON irradiation furnace as shown on Figures 1 and 2.

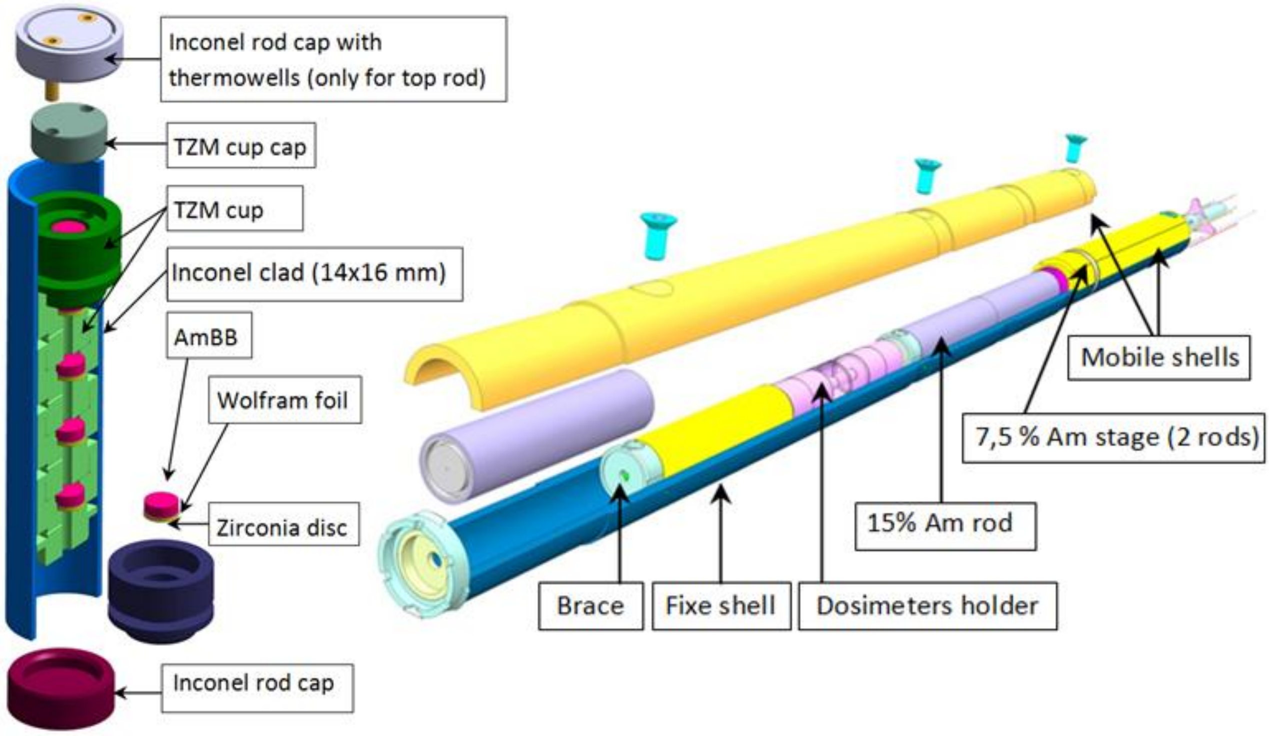

Fig. 1: Needle details and shell of the sample holder with needles and thermal separator

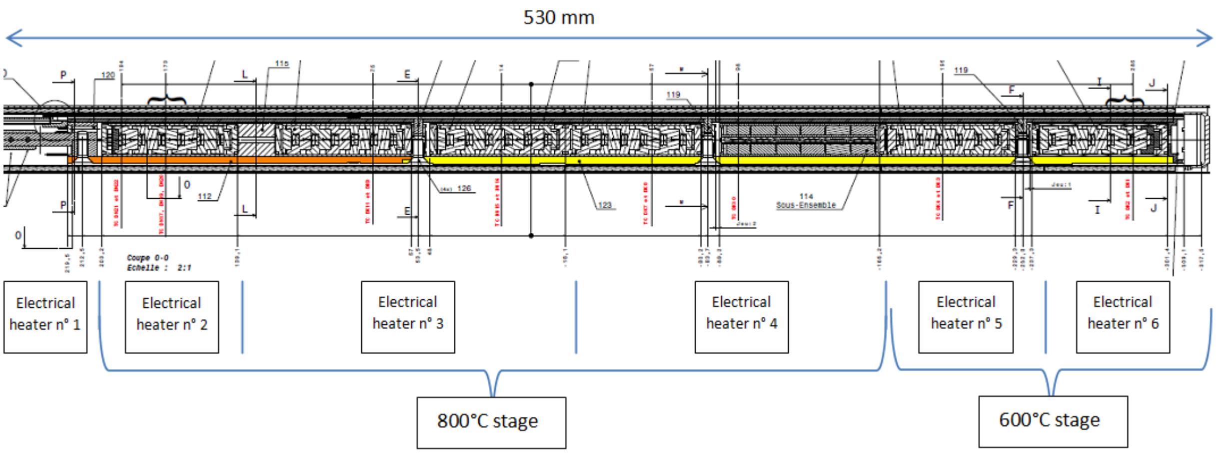

Fig. 2: Bottom of the sample holder in the safety double barriers PHAETON furnace

2.2 PHAETON furnace

The PHAETON furnace is a dual cylindrical container 4.5 m high for a useful internal diameter of 24 mm. The intermediary clearance is pressure-controlled to ensure the dual integrity of the containment barriers. The useful area containing the control heating elements covers about 600 mm of the lower extremity. The upper 4 meters are used to deport the leaktight and the handling connections outside the nuclear flux area and outside the reactor chimney. In the special case of DIAMINO irradiation, the selected position is outside the core as shown in the Figure 3 and 4. The cylindrical furnace with an external diameter of 32 mm is itself inserted in a water box that channels a cooling water flow.

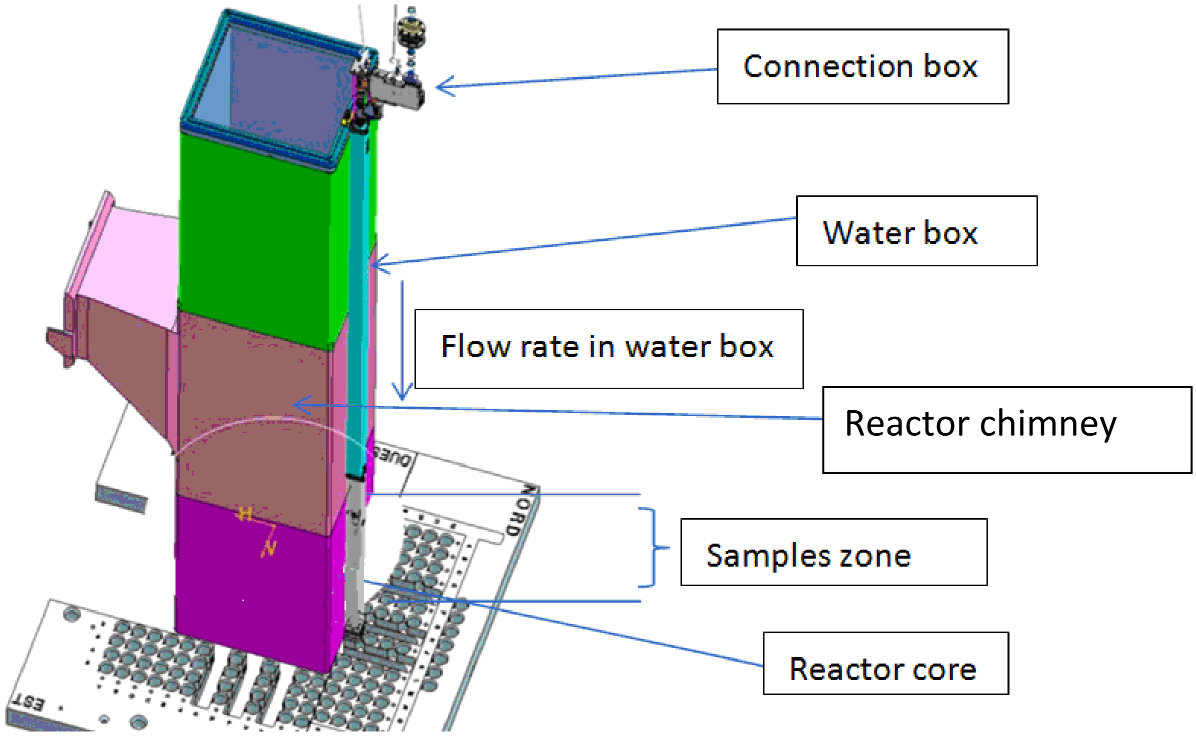

Fig. 3: Device in the water box on the reactor out-core

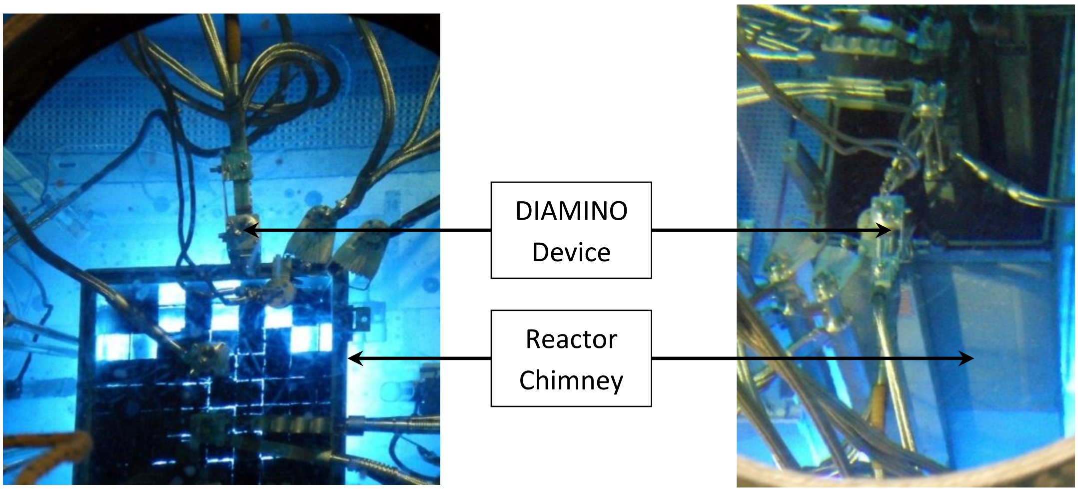

Fig. 4: Device on the reactor out-core

3. Technology components

3.1 Fuel discs and needles [1]

The properties of $UAmO_2$ discs as shown on Table I.

TABLE I : $UAmO_2$ discs properties.

	Batche 1 (600°C and 800°C)	Batche 2 (600°C and 800°C)	Batche 3 (800°C)	Batche 4 (800°C)
Diameter (mm)	$4{,}443^{+0.012}_{-0{,}017}$	$4{,}566^{+0.015}_{-0{,}032}$	$4{,}44^{+0.019}_{-0{,}028}$	$4{,}563^{+0.008}_{-0{,}020}$
Thickness (mm)	$1{,}557^{+0.021}_{-0{,}041}$	$1{,}548^{+0.019}_{-0{,}020}$	$1{,}531^{+0.008}_{-0{,}017}$	$1{,}530^{+0.013}_{-0{,}030}$
Relative Density (%)	$95{,}737^{+0.273}_{-0{,}387}$	$81{,}816^{+0.734}_{-0{,}346}$	$96{,}563^{+0.207}_{-0{,}233}$	$84{,}903^{+1.327}_{-0{,}593}$
Am mass content (Am/(U+Am)) (%)	15,4	13,7	7,1	7,4

The feedback from similar fabrications as SIROCCO with pulse laser welding resulted in the selection of Inconel 600 for the cladding, mainly because of its physico-chemical and mechanical properties at temperature (thermal creep). The behavior of pulsed laser welds proved to be very sensitive to certain impurities, found in the Inconel 600, and to the size of grains. A specific batch of Inconel 600 was suppliyed and underwent special treatment (tempering, hardening, etc.) to overcome the problem of hot cracking junctions.

The TZM (molybdenum alloy) cups were integrated with the needles to bring a significant gamma heating. Although the fission power density in the discs was high, their low volume made little contribution to the local heating, outside the gradient in the discs themselves. Channels were created in these cups to distribute the gases released by the $UAmO_2$ discs throughout the free volume of the needles.

Although the yttriated zirconia is particularly chemically stable, as a precaution, to ensure the chemical compatibility of contacts at high temperature, a wolfram sheet 0.2 mm thick was inserted between the zirconia and the $UAmO_2$ discs. Experience shows that the tungsten crucibles remain unaltered in contact with the $UAmO_2$ for long periods at temperatures far higher than the 800°C targeted on the discs.

In the last upper needle in the load, two thimbles were fitted in the top plug to house two intrusive thermocouples, to improve measurement representativeness in this area. In addition, needle integrity under pressure was tested in representative test samples.

The discs fabrication and needles assembly were performed in the ATALANTE facility in CEA MARCOULE site.

3.2 Sample holder

The thermal calculations showed that the temperature of the sample holder could reach 670°C at the beginning of irradiation, for maintaining the fuel discs at 800°C and 600°C. Inconel 600 was selected for the lower section of the sample holder. The common and upper parts of the sample holder remained at lower temperature and stainless steel 316L was deemed sufficient. PORAL filters were arranged below the biological protection to contain the fission products that would be released in case of a needle rupture.

In the lower section, two half-shells, as presented in Figures 1 and 2, encircle the needles and the dosimeter holder. The clearance gas gap between the sample holder and the internal furnace tube was optimized for each needle as shown on Figure 5.

The structure of the dosimeter holder was identical to that of the needs so that the dose integrators were in a neutron environment as close as possible to that of the fuel discs. The neutron flux maps calculated in the device could then be used to refit the fluence values measured in the dosimeter holder to deduce the fluence received in the fuel discs. Aluminum-cobalt and Iron dose integrators were chosen.

The thermocouples were inserted in grooves into the fixed half shell of the sample holder, with some also placed in the upper movable shell.

Gas circulation is maintained around and inside the sample holder shells during irradiation. This circulation enables to detect a needle rupture by feeding back activity to a detector located in a shielded glove box. This circulation is also used to change the gas composition (He/Ne mix) to adjust the temperatures, in addition to the action of electric furnaces.

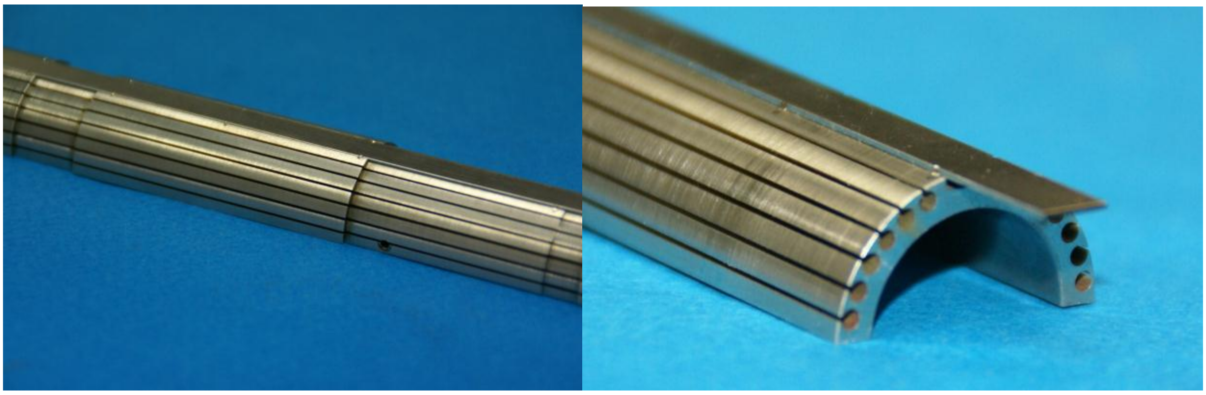

Fig. 5: The half shell which contains thermocouples during processing

3.3 Furnace electrical heaters

The PHAETON furnace was already used for the FURIOSO experiment [2] (High temperature testing material for ITER program). The lower section of the furnace has six heating elements whose height distribution means the needles can be operated individually except for one heating elements which in fact controls two needles (Figure 2). The electric flux of a maximum linear power of 330 W/cm passes through the helium gas gap in the furnace shown by the clearance between the copper sprayed containing the thermocouples and the heating elements and the external stainless steel tube. Any change in the electric power has a local impact on the temperature of the structures. The furnaces are operated by an automatic control with a response time of several ten seconds.

3.4 Device design studies

The design of the device is based on studies achieving the desired experimental conditions and showing sufficient dimensioning margins in relation to the safety reference frameworks. These studies relate to the physico-chemical, neutronical, thermo-mechanical, thermal, thermal-hydraulic and radiological aspects. These studies have among other things determined the appropriate materials, the thicknesses of the different components to achieve specified temperatures.

The neutron and photon calculations [3] are needed to produce a part of the other calculations input parameters. TRIPOLI-4® Monte-Carlo code [4] produced local power, neutron, gamma flux and gamma heating distributions inside the different materials of the device, as an initial time reference. This study is needed to optimize fuel discs geometry in APOLLO2 2D model. APOLLO2 calculations [5] produced the temporal evolution power of each fuel disc. The power increases rapidly during the first irradiation cycle (^{242m}Am creation with a highly cross section in the thermal domain) followed by a slow and continuous increase (Figure 6). The time power profile shows a set of peak, corresponding to the end of the OSIRIS irradiation cycle. APOLLO2 calculations provided also a 172 energy groups flux neutron distribution needed for radiological inventory evolution (including helium) with the DARWIN/PEPIN2 code [6].

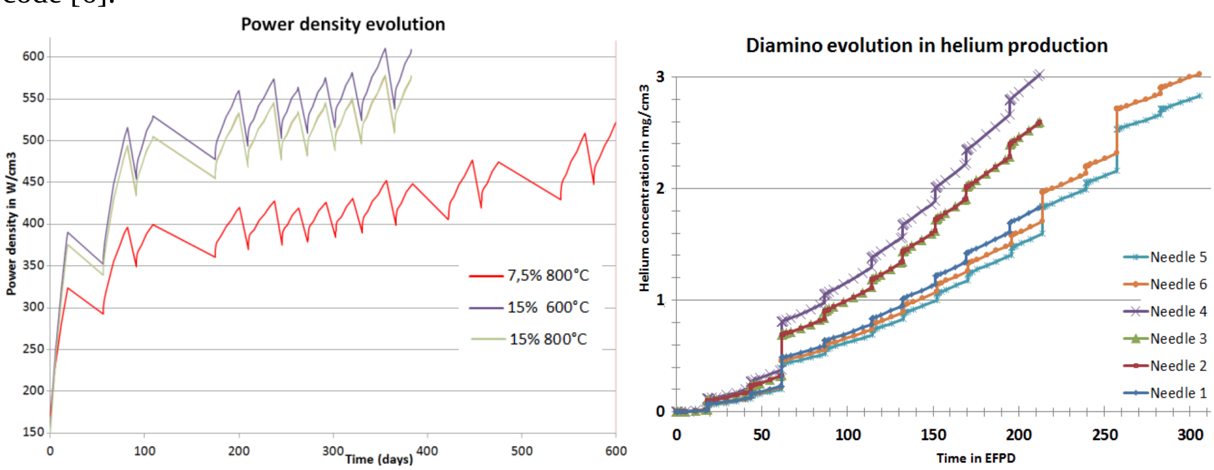

Fig. 6: Power density and helium evolution of $UAmO_2$ in each needle

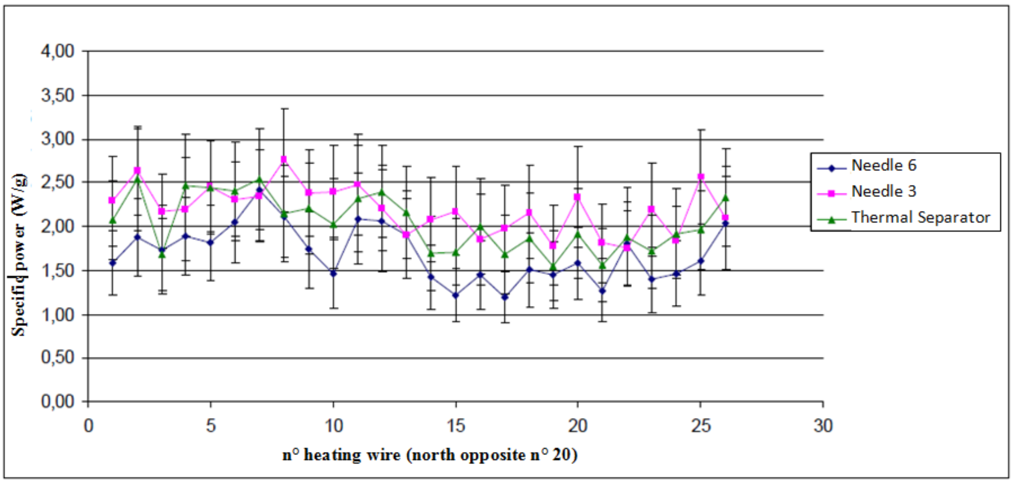

Fig. 7: Azimuthal distribution of gamma heating in heater wires at the level of needles 3 and 6 and thermal separator (x 1.42 W/g)

The thermal studies showed that the azimuthal variations of gamma heating obtained by the transport calculations (Figure 7) had little influence on the temperature gradients in the structures (this effect is strongly reduced by good thermal conductivity of metals), this effect was ignored subsequently.

In the vicinity of the median plane, the gamma heating peak calculated by TRIPOLI-4® reaches 1.4x2.8 = 3.9 W/g in the heating wires. This value is very over-estimated compared with the measurements taken by calorimetry in this irradiation position (2.8 W/g by applying the maximum uncertainty by excess). The measurements in this location are usually between 1.6 - 2 W/g; these variations come from a different core loading. Even when taking measurement uncertainties into account, the calculated and measured intervals remain disjointed.

In the same way, the TRIPOLI-4® calculations at the needle cladding give 2.42*1.42 = 3.4 W/g of Inconel around the maximum flux plane. This value is still over-estimated by 25% compared with the calorimetric measurements. The radial distribution was considered as an attenuation factor in the structures. This factor has been adjusted to find the radial distribution of temperatures measured in a reactor, in the thermal mock-up of the device.

The thermal calculations, performed using the LICOS code [7], were used to:

- Design the various gas clearances in order to maintain the nominal temperature on the fuel discs throughout irradiation with the capacities of the PHAETON furnace (given the major change in power of fuel discs).
- Check that the maximum use temperature was not reached on any component, especially the inner furnace tube and the sample holder structures for all operating situations.
- Optimize the hot junction position of thermocouples (areas with low thermal gradient).
- Determine the temperature refitting values to be applied to the thermocouples to find out the average temperature of the fuel discs.
- Assess and limit the temperature gradient in each fuel disc (~130°C). Special care was given to equalizing the thermal resistance above the discs (gas clearance with radiation taken into account) with that below the discs (contact with yttriated zirconia disc).

- Producing significant concentration of He in fuel with an acceptable acceleration factor compared with the SFR reference design.
- Choose the best irradiation position.

Thermo-mechanical studies (NASTRAN and LICOS codes) have demonstrated the mechanical resistance of structures (including the $UAmO_2$ discs) in all plausible normal and accidental situations according to a probabilistic classification of occurrence.

Thermal-hydraulic studies have shown that the device was cooled for any failure of a single item of equipment, like the shut down of the cooling pump of the water box, for example.

Figure 8 shows the temperatures, taken from preliminary calculations, of various components of the device at the beginning and the end of irradiation. Note a fare greater temperature difference between the fuel discs and the needle cladding between the beginning and the end of irradiation; this gradient comes from the increased power of fuel during irradiation. The heating power component drops to maintain the fuel at constant temperature.

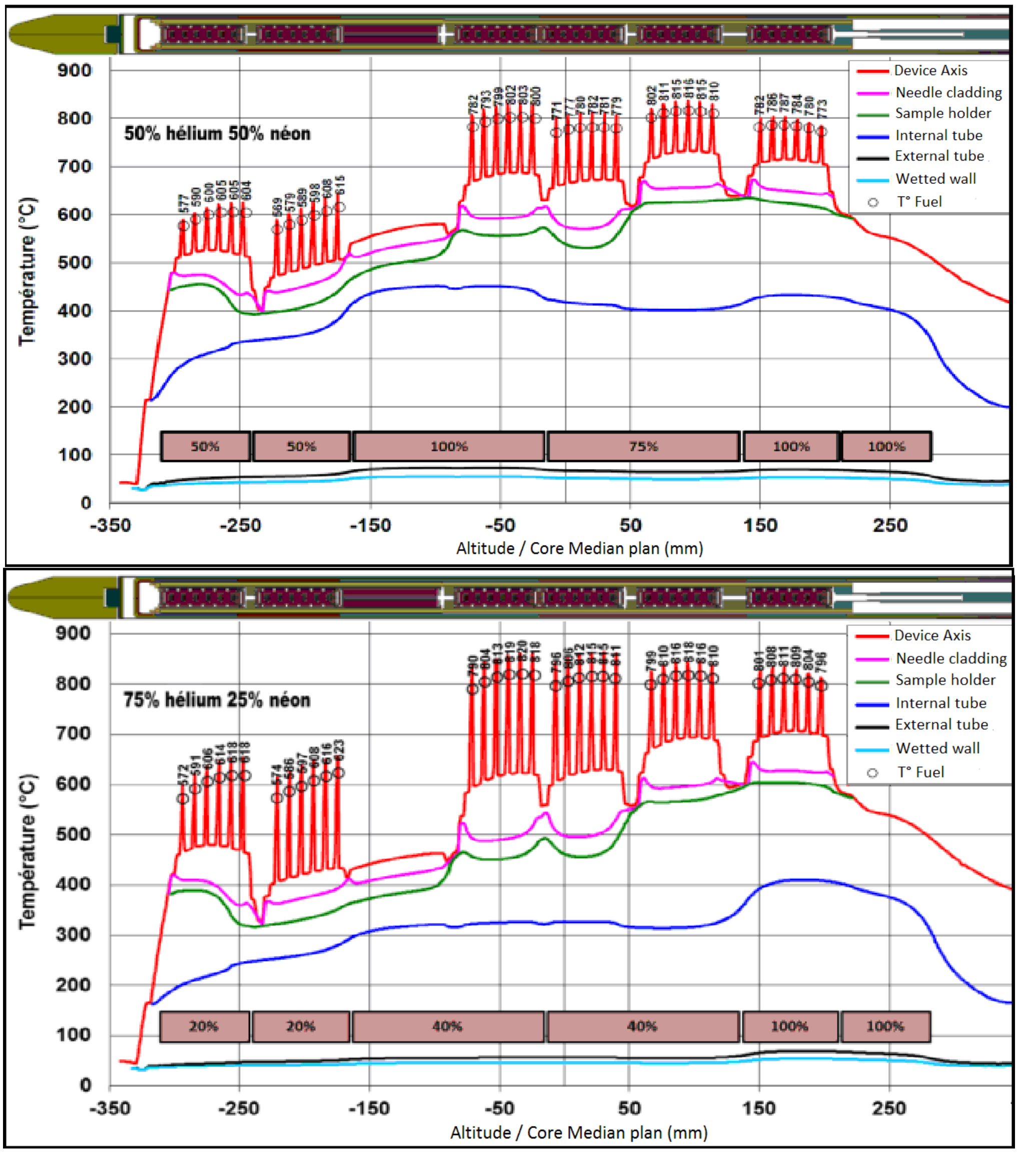

Fig. 8: Forecast temperatures at the beginning (up) and at the end (down) of irradiation

The compatibility of materials in contact at high temperature was verified in normal and accidental situations. In normal situations, the criteria to be respected must allow the irradiation to be run until the end without damage the component functions (alignment, insulation, leak tightness, geometry, etc.) and without altering the irradiation conditions (flux, temperature and burn-up). In an accident situation, interactions between materials, including the cooling water, must not result in structural fusion (eutectic, overheating…).

4. Model qualifications

4.1 PHAETON furnace

To qualify the numeric model of the PHAETON furnace, we have carried out a thermal test campaign with the empty furnace, on a hydraulic bench out of the flux. Several series at different electrical powers have been performed with various gases in the thermal barrier (helium, neon and nitrogen). The thermal calculations with the corresponding furnace model have shown that the code behaves well in these configurations without nuclear heating. After correcting a few geometric dimensions in the order of magnitude of uncertainties of dimensional measurements on the gas clearance, the residual differences have been limited to fifteen or so °C on measurements around 320°C on the inner furnace tube.

4.2 Refitting the model with the thermal mock-up

A mock-up of the device with the complete needles but without the $UAmO_2$ discs was produced in order to validate the irradiation device model. Most of the nuclear power heating in the device comes mainly from gamma heating. The lack of fissile load had very small impact on the thermal behavior of the device, above all at the start of irradiation when the density of the fissile power of discs is still modest. The mock-up took place at the same position in OSIRIS water reflector in the same PHAETON furnace. The sample holder was virtually identical to the real one, except a slightly modification of the temperature measurements distribution. Thus the thermal behavior at load end, where the boundary conditions are less well known in the numeric model (axial thermal leakages), will be best characterized.

The irradiation of the thermal mock-up has allowed to assess a few parameters tainted with huge uncertainties. These include the gamma heating in the various structures or the axial leaks in the upper needle. The water box used is the one where the device is the closest to the reactor core. The differences between the mock-up measurements and the results of the calculation arise out of imperfections in the meshing and in the physical model (radiation activation, contact resistors, connection thermal bridges). By adjusting the gamma heating distribution, assumed homogeneous throughout the section, and its absolute value, very variable according to the core loading and the material of the structure in question (new or worn reactor fuel elements near the DIAMINO device), we have been able to find the entire furnace and sample holder temperature chart in the range of temperatures targeted in all the irradiation configurations. Nevertheless an air pollution of few % in the furnace gas was considered to explain its thermal behavior.

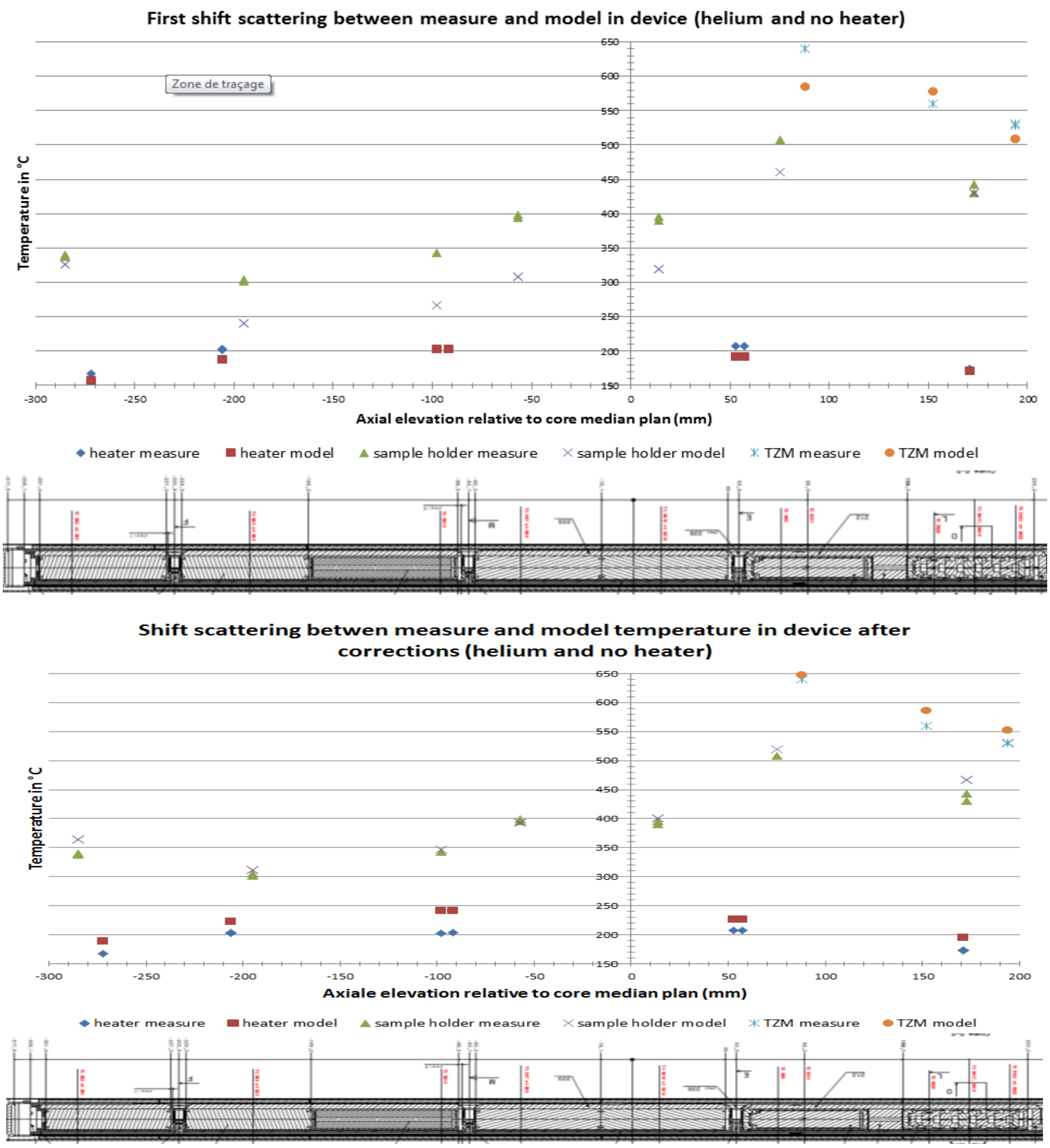

Fig. 9 & 10: First shift scattering between measure and model before and after corrections

These figures compare the temperatures measured and those calculated in the helium configuration without electric heating before (Figure 9) and after (Figure 10) corrections. Temperature differences between the model and the tests as shown on Table II:

TABLE II: Temperature differences between the model and the tests for other used configurations.

	Configurations			
	100% He Furnaces turn-off	100% He 25% furnaces	100% He 47% furnaces	50% Neon Furnace turn-off
Target temperature on sample holder	300 to 640 °C	400 to 710 °C	480 to 760 °C	340 to 840 °C
Max. (Tmeasures – Tmodel) in the sample holder	-30 / +4°C	-8 / +23°C	+1 / +29°C	-38 / + 26 °C
Target temperature on furnace	100 to 210 °C	220 to 310 °C	310 to 400 °C	10 to 210 °C
Max. (Tmeasures – Tmodel) in the furnace	-38 / - 18°C	-23 / - 8°C	-9 / + 18°C	-34 / +12 °C

As the average temperatures of $UAmO_2$ discs are obtained by applying a calculated correction in the measured sample holder temperatures, only a fraction of the differences above will have an impact when determining disc temperatures. For the real irradiation the calculations / measurements comparisons on temperature are used to detect a change in the irradiation conditions that would make the refitting function obsolete. Note that the value of this calculated correction increases over time, as the power density in the discs also increases, above all during the first irradiation cycle.

5. Results on first DIAMINO irradiation cycles

During the first irradiation cycle with fuel, the actual irradiation device was found to be hotter than expected. The reason was the change in the experiment environment. During the calibration cycle of the thermal mock-up, the fuel assemblies near the device were spent assemblies, whereas the OSIRIS load for the first cycle of the experiment, had fresh assemblies. This increased the gamma heating. This increase taken into account in the calculations returned the value of calculation-measurement differences to the vicinity of that optimized in the mock-up.

The average disc temperatures (600°C - 800°C) were quickly achieved and even exceeded for a needle with an all-helium configuration and furnaces turn-off. Between the 1st and the 2nd irradiation cycle the water box has been changed to reduce significantly nuclear powers (fission + gamma) and to give a little operating margin (this box has a furthest position to the core). The calculations of changes in the fissile power and the production of helium were also updated to take into account irradiation conditions with the new water box.

Model was also used to determine the temperature of all 36 fuel discs by measuring the temperature of the opposite sample holder. The axial temperature distribution depends on the distribution of neutron flux, the enrichment of discs, their porosity, axial heat leaks (gas layer and thermal bridges connected to the supports). Figure 11 shows this distribution at the beginning of the third irradiation cycle.

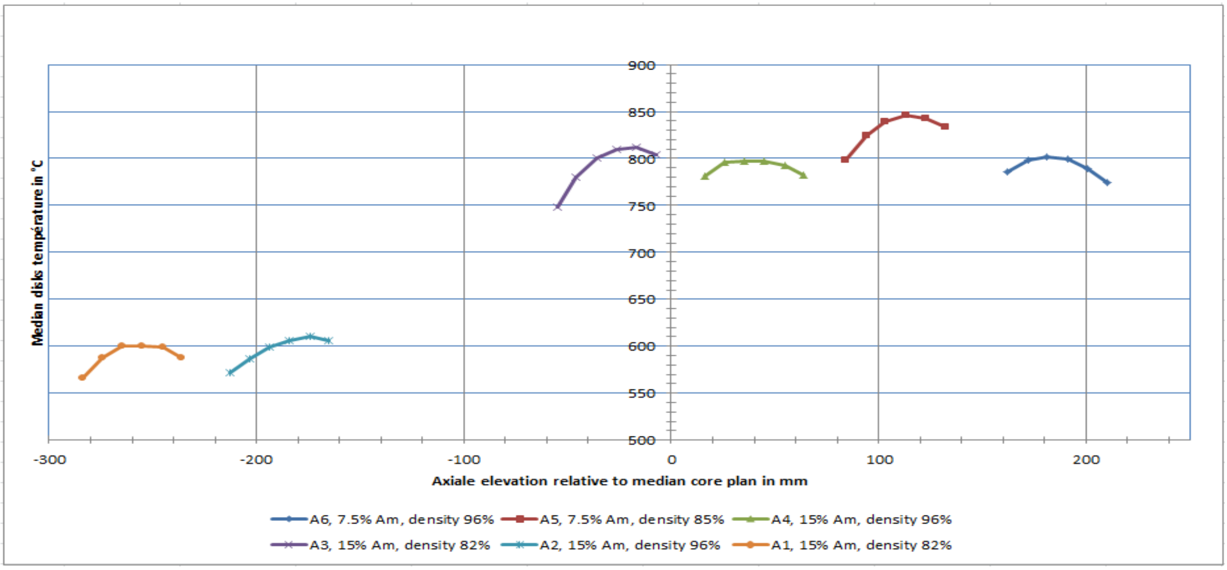

Fig. 11: Temperature distribution in fuel discs at the beginning of the third irradiation cycle

6. Conclusion

After four cycles of irradiation (about 80 EFPD) the DIAMINO irradiation presents interesting results, with two different temperature levels at the loads in the same device, despite the major dispersions of gamma heating according to the reactor cycles. Where it was impossible to maintain all the temperatures at their set point value with the available control means, the maximum levels obtained remain acceptable to meet the goals of the experiment.

The temperature calculation takes into account the power reactor variations, the burn-up of the AmBB fuel and the gaseous composition variation.

At the end of the irradiation (315 EFPD is expected for low Am concentration pellets and about 212 EFPD for high Am concentration pellets), post-irradiation examinations will especially quantify the effects of helium production in the matrix.

The examination campaign will include:
- A comparison between neutronographs before and after irradiation to ensure the general evolution of discs during irradiation.
- The general appearance of fuels (cracking, open porosity, swelling, gas bubbles).
- The metrology of needles and discs, the density measurement of discs.
- Thermal treatment with gas release analysis (helium and fission gas).
- Dissolution for chemical and isotopic analysis (fission and transmutation rate).
- Microanalyses (DRX, MEB, Microprobe, SIMS) and ceramographs.

After the irradiation, the dosimeters analysis will allow refitting neutron calculations by integrating the fast and thermal flux during irradiation as to reach the best estimate $UAmO_2$ discs burn-up.

7. References

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