CONCEPTION AND FABRICATION OF INNOVATIVE AM-BASED TARGETS: THE CAMIX/COCHIX EXPERIMENT

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Abstract

A large experimental programme has been planned to be carried out in the French **PHÉNIX** reactor. The purpose is to evaluate the technical feasibility of minor actinide transmutation in fast reactors. Two major series of experiments have been designed for the heterogeneous transmutation mode. The first one, the **MATINA** (**MAT**rices for **IN**cineration of **A**ctinides) series, aims at testing both different inert matrices in a fast flux and different concepts. The study is generic and focuses on the material behaviour under representative irradiation conditions. Targets are free of minor actinides to make the fabrication and design steps easier and faster. The second one, **ECRIX**, **CAMIX** (**C**ompounds of **AM**ericium in PHÉN**IX**) and **COCHIX (C**oncept **O**ptimized mi**C**rostruture in P**H**ÉN**IX)**, is a further step in the demonstration phase of the "once-through" transmutation and deals with Am-bearing targets irradiated in a fast neutron spectrum "*locally*" moderated. The moderator materials tested will be calcium hydride CaH_{2-x} (cases of **ECRIX-H**, **CAMIX** and **COCHIX**) and boron carbide $^{11}B_4C$ (case of **ECRIX-B**) in order to accelerate the process of transmutation significantly.

Introduction

To evaluate the technical feasibility of minor actinides transmutation in the heterogeneous mode in fast reactors, an experimental programme is planned in the French **PHÉNIX** reactor. [1] The **CAMIX-COCHIX** irradiation experiment is one part of this programme and is more particularly intended to study the feasibility of the americium incineration. Irradiation conditions for **CAMIX-COCHIX** will be similar to the once used with **ECRIX-H** experiment [2] (AmO_{2-x} micro-dispersed in MgO produced by powder metallurgy) for testing alternative and/or optimised materials.

CAMIX-COCHIX irradiation will be carried out in neutron spectrum "*locally*" moderated by calcium hydride (CaH_{2-x}) so as to accelerate the process of transmutation significantly. The experiment will consist of the irradiation of a capsule which will contain three experimental pins with different fuels but containing the same amount of americium (0.7 g of Am per cm^3).

The innovations [3] are four. They consist in:

- inserting the americium compound in the cubic phase of **Y**ttria-**S**tabilised **Z**irconia **YSZ**;
- testing alternatives to **MgO** as host matrix;
- testing macromass concept;
- and also testing material produced by **SOL-GEL** techniques in comparison with powder metallurgy.

The **CAMIX-COCHIX** experiment will be loaded at the peripherical part of the fissile core. The irradiation conditions have been predicted by a computer code. The goal of the irradiation is to reach a cumulated extent of fission of 30at%, representative of an americium cumulated extent of transmutation of approximately 90%.

Target description

The **ECRIX** pins are representative of a simple concept, where AmO_{2-x} is micro-dispersed in **MgO**. The three **CAMIX/COCHIX** pins are representative of a concept, where AmO_{2-x} is replaced by an improved Am compound which makes up the pellet itself or is dispersed in magnesia. The three pins are filled as follows:

1. Single phase pellets: solid solution of americium and zirconium oxides **Am-YSZ** (**CAMIX n°1**). The Am content of the americium compound is ≈ 6 atomic%.
2. Composite pellets of americium particles **Am-YSZ** dispersed into a matrix of magnesia. The size range of the americium-bearing particles is 40-60 µm (representative of micro-dispersion). The americium concentration of the solid solution is ≈ 20 atomic% (**CAMIX n°2**).
3. The same as the previous one, except for the size of the americium particles which is in the range of 100-125 µm representative of macro-dispersion (**COCHIX n°3**).

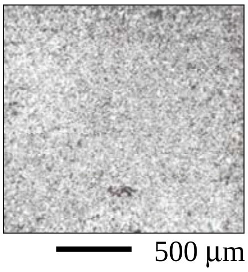
The **YSZ** (with 17 atomic% yttrium) has been selected as the americium host because it is highly resistant to irradiation damage, is refractory and has particularly stable physicochemical properties versus temperature. Moreover, this compound should offer a low oxygen potential compared to americium oxide (AmO_{2-x}).

MgO is still considered as the reference inert matrix because of its good thermal properties and its good behaviour under irradiation at weak fluence (**MATINA1** results. [4] However, its behaviour under irradiation at high fluence still remains to be tested.

The size range of the Am-based particles is related to the risk of the matrix degradation by the irradiation effects. Unlike with *"micro dispersion", "macro dispersion"* should allow us to limit the damage of the matrix by fission fragments.

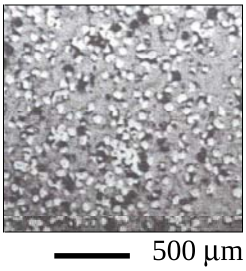
The main characteristics of the composition and form of the **CAMIX-COCHIX** targets are described below.

Figure 1. **Optical micrograph of CAMIX n°1**



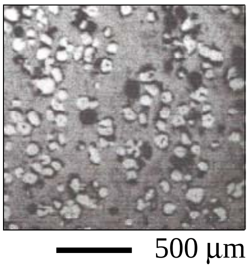
CAMIX n°1 (see Figure 1):
Target type: Solid solution
Composition: $(Am_{0.06},Zr_{0.78},Y_{0.16})O_{2-x}$
Americium isotopy: 100% ^{241}Am
Outer diameter: 5.42 mm
Density: ≥ 90% TD

Figure 2. **Optical micrograph of CAMIX n°2**



CAMIX n°2 (see Figure 2):
Target type: CERCER Micromass
Fuel Particles (F.P.): $(Am_{0.2},Zr_{0.66},Y_{0.14})O_{2-x}$
Americium isotopy: 100% ^{241}Am
F.P. size: $40 \le \varnothing_{particle} \le 60$ µm
Inert matrix: MgO
Volumic% of F.P: 30%
Outer diameter: 5.25 mm
Density: ≥ 85% TD

Figure 3. **Optical micrograph of COCHIX n°3**



COCHIX n°3 (see Figure 3)
Target type: CERCER Macromass
Fuel Particles (F.P.): $(Am_{0.2},Zr_{0.66},Y_{0.14})O_{2-x}$
Americium isotopy: 100% ^{241}Am
F.P. size: $100 \le \varnothing_{particle} \le 125$ µm
Inert matrix: MgO
Volumic% of F.P.: 30%
Outer diameter: 5.25 mm
Density: ≥ 85% TD

The expected volumic fraction of the damaged matrix should be close to 100% for the **CAMIX n°1** and **n°2** targets and close to 30% for the **COCHIX n°3** target.

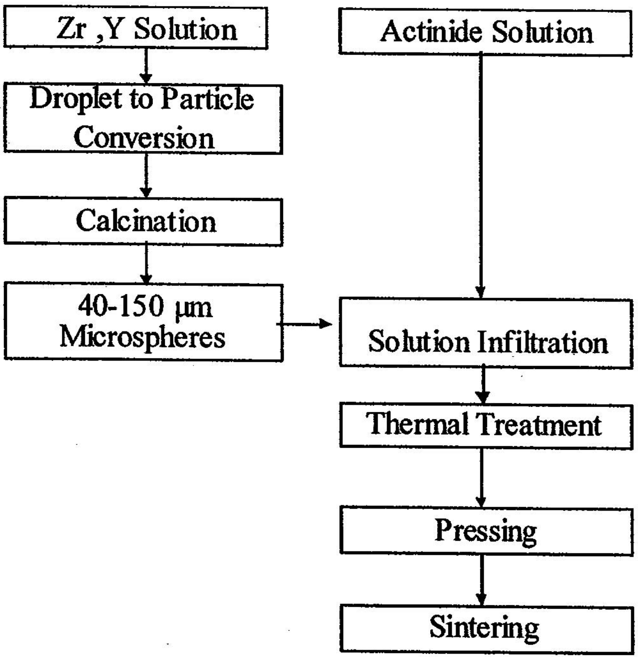
Lastly, the volumic swelling of the pellet should be lower than 2% for target **CAMIX n° 1** using **YSZ** and lower than 21% for targets **CAMIX n°2** and **COCHIX n°3** using **MgO**.

Target fabrication process

The fabrication of **CAMIX n°1**, **CAMIX n°2** and **COCHIX n°3** targets is performed in the hot cells of the **M**inor **A**ctinide **LAB**oratory (**MALAB**) of the Institute for Transuranium Elements at Karlsruhe **ITU**. [5]

The fabrication process for the fuel material of **CAMIX** n°1 is shown schematically in Figure 4. **YSZ** spheres are produced by a **SOL-GEL** process and have polydisperse size distribution in the 40 to 150 μm range. The calcined spheres are infiltrated with a calculated amount of americium nitrate solution by **IN**filtration of **RA**dioactive **M**aterials process (**INRAM**). After the infiltration step, the spheres are thermally treated at 1 073 K to convert the infiltrated americium nitrate into the corresponding oxide. They are then pressed into the shape of pellets and sintered at 1 920 K under argon.

Figure 4. **CAMIX n°1 fabrication process**



The fabrication process for the fuel material of **CAMIX** n°2 and **COCHIX** n°3 is shown schematically in Figure 5. **YSZ** spheres are produced as described above. The spheres are then sieved and specific size fractions, 40-60 μm for **CAMIX** n°2 and 100-125 μm for **COCHIX** n°3, are selected. The spheres are then infiltrated into consecutive steps with an americium nitrate solution by **INRAM** process. After each infiltration, they are thermally treated at 1 073 K. For composite fabrication, the matrix MgO is mixed with calibrated spheres and compacted into pellets. Finally, sintering is performed at 1 920 K under argon in order to obtain the targets.

Conceptual studies – Irradiation device and target pin

The three **CAMIX-COCHIX** pins will be irradiated together in an irradiation capsule loaded into a special carrier sub-assembly (S/A) with a moderator material so as to accelerate the process of transmutation significantly (see Figure 6).

Figure 5. **CAMIX n°2 & COCHIX n°3 fabrication process**

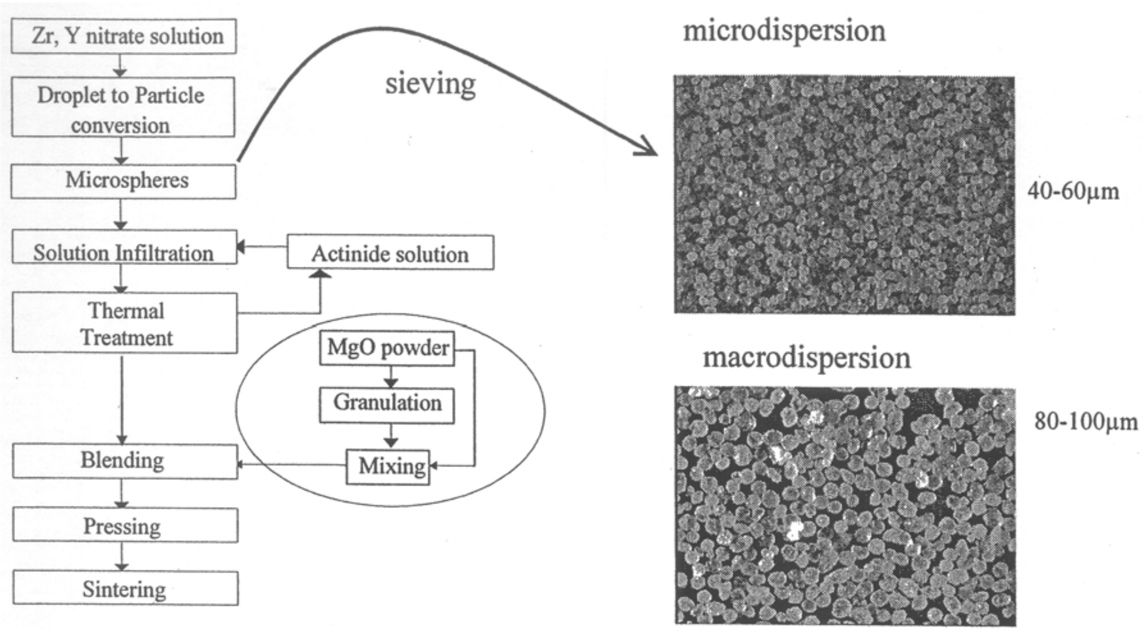
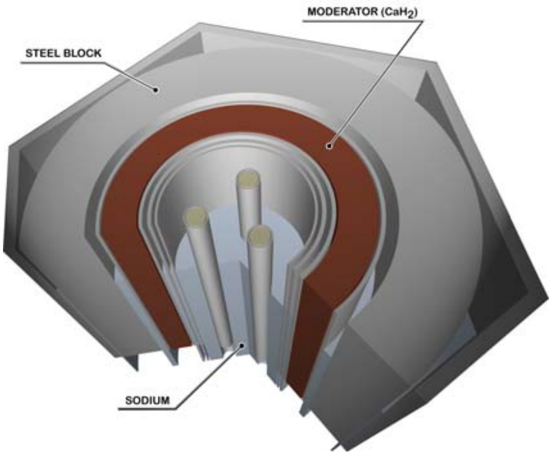


Figure 6. **Schematic view of the three pins in the irradiation device**



The carrier S/A is designed to be compatible with the core environment: in particular, the neutronic impact on the surrounding S/A has to be evaluated and minimised at the design stage (choice of the moderator and steel block thickness).

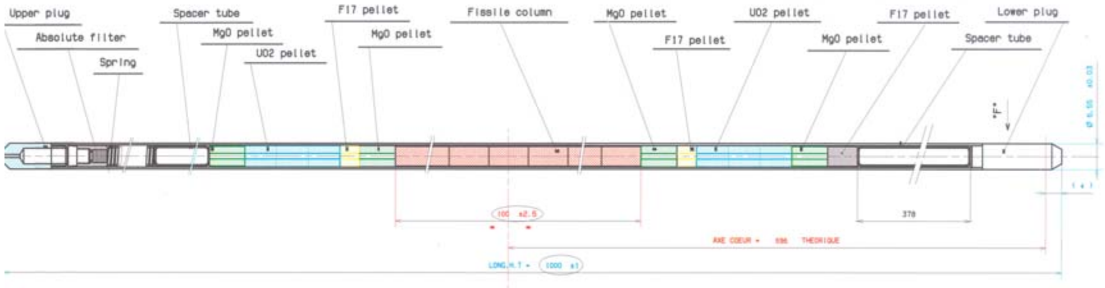
The carrier S/A containing the moderator blocks (CaH_{2-x}) and the capsule containing the target pins are cooled with sodium. The cooling flow in the capsule is oversized for the limited power generated, so that the clad temperature of the experimental pin will be close to the sodium temperature at the core entry (380°C).

The design is similar for all the pins (see Figure 7):

- cladding: 15-15Ti ε (5.65×6.55 mm)
- length: 100 cm

- target column: length 10 cm
- plenum: 16 cm^3
- filling gas: helium (95 vol% - 1 atm)
- clad failure detection system: UO_2 (^{235}U - 4 wt%)

Figure 7. **CAMIX-COCHIX experimental pin**



Operating conditions

The operating conditions of the **PHÉNIX** reactor are characterised by a total power of 350 MWth. For **CAMIX-COCHIX**, the irradiation parameters will be the same than as those used for the ECRIX-H experiment. They are recalled below:

- irradiation position (see Figure 8): 1st row of radial blanket
- irradiation duration: 340 Equivalent Full Power Days EFPD
- fast fluence (E>0,1 MeV): 2 10 26 n/m^2
- dose 10 dpa

The previsional irradiation conditions were determined by neutronic calculations. The detailed geometry and the properties of the materials were taken into account.

Simulation of neutronic conditions

The objective of neutronic calculations is twofold:

- to determine the performances of the experiment required for thermal calculations (cumulated extent of transmutation, heat rating of experimental pins...);
- to determine the impact of the carrier on the surrounding S/A (local and total power, max linear heat rating...).

Standard calculations were performed with the ERANOS system [6] associated with the adjusted library ERALIB1 [7] which allow all the neutronic characterisations to be performed. The adopted method is a heterogeneous description for the cell calculation in fine groups and a space calculation in transport method which takes into account photon propagation.

Figure 8. **CAMIX-COCHIX irradiation position**

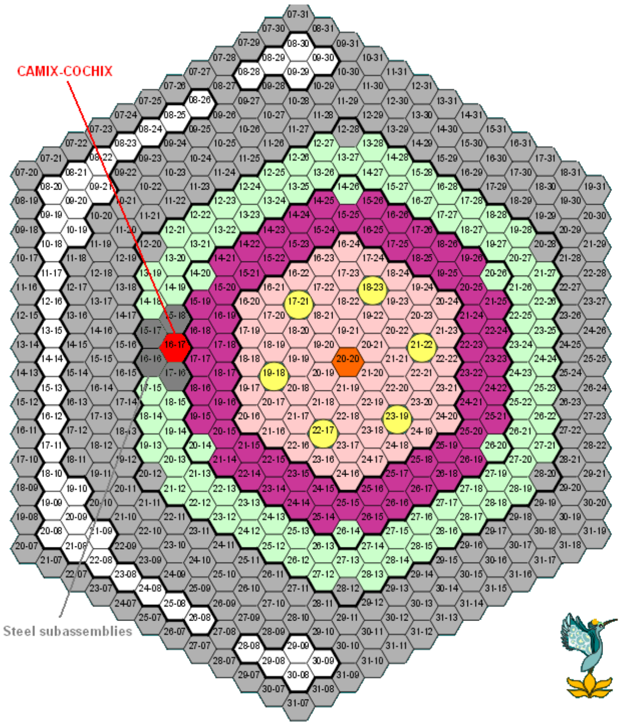
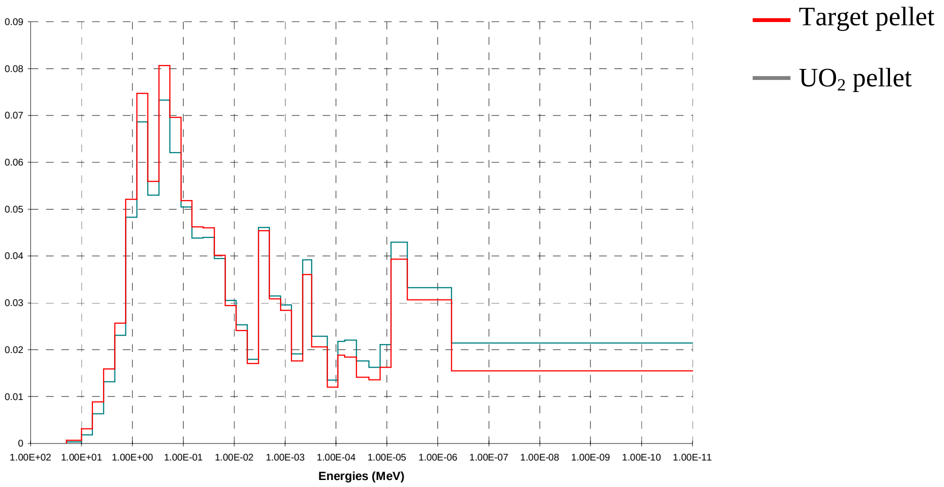


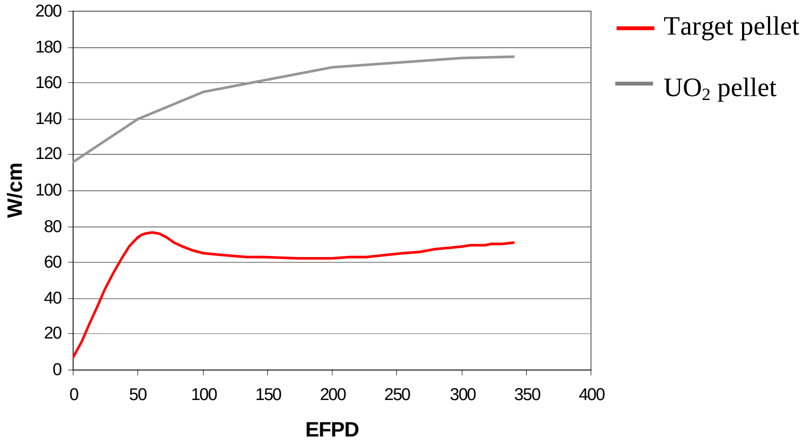
Figure 9 presents the neutronic spectra in the **CAMIX-COCHIX** pins. It is noted that a thermal component appears, resulting from the effect of CaH_{2-x} around the pins.

Figure 9. **Neutronic spectra (33 energy groups) in the CAMIX-COCHIX pellets**



The power in the **CAMIX-COCHIX** pellets is shown Figure 10.

Figure 10. **Evolution of the linear power in the CAMIX-COCHIX pellets**



For the targets, at the beginning of life, the **CAMIX-COCHIX** pins provide energy by capture mainly on ^{241}Am due to the moderate flux. The thermal capture of this isotope is dominant. There is then formation of the very fissile isotope ^{242m}Am and the power in the target increases rapidly. With the extension of the irradiation, fissile Pu and Cm isotopes appear, and the fission reaction is dominant but the power level does not change significantly. The maximum linear power 75 W/cm occurs just at 50 EFPD after the beginning of the irradiation and becomes almost stable until the end (70 W/cm at 340 EFPD).

For the UO_2 pellets, the linear power increases all along the irradiation and reaches 175 W/cm at 340 EFPD.

The other major characteristics predicted for **CAMIX-COCHIX** are gas production (see Table 1):

Table 1. **Gas production**

	Target pellets (340 EFPD)	**UO_2 pellets** (340 EFPD)
He product (NTP)[1]	48 cm^3	0.10 cm^3
Xe+Kr product (NTP)	8 cm^3	13 cm^3

Concerning the effect on the fissile sub-assemblies close to the carrier S/A, the power increase comes from the backscattering effect of the thermal neutrons. By design, this impact is minimised by a reduced thickness of the CaH_{2-x} moderator and adding of steel shell. This was found to be a very local phenomenon, limited to the outer fuel pins. The increase in the total S/A power remains low all along the irradiation. Beside the carrier S/A, fertile subassemblies were replaced by steel subassemblies in order to avoid high over-power on UO_2 pellets.

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[1] NTP (273.15 K, 0.1013 MPa).

Simulation of the thermal and mechanical behaviours

The operating temperature of the target material is an important parameter of its behaviour under irradiation. The heating of the pin depends on the pin geometry, thermal properties as well as loading and irradiation effects. The main characteristics needed for thermal calculations are given below:

Geometry:

- Inner and outer diameters of the cladding.
- Nature of the gas inside gap between pellet and cladding.
- Outer diameter of pellet.
- Pellet density.

Physical properties:

- Coefficient of the thermal expansion.
- Thermal conductivity.
- Melting temperature of target material.

Loading:

- Linear power of pellet.
- External temperature of the pin (coolant temperature).

Irradiation effects:

- Swelling.
- Degradation of thermal conductivity.
- Gas composition inside gap between pellet and cladding.

The geometry is defined by calculations so that the pellet temperature remains lower than its melting temperature. Concerning temperature results, we only show here results without uncertainties.

The physical properties of magnesia and (**Am-YSZ)** were taken from literature and the loading values are given by neutronics studies. The thermal conductivity of (**Am-YSZ**) was temporarily chosen equal to 2 W/(m.K) but the thermal properties of the targets will be measured at the **ITU** for the final design. The estimated melting temperatures considered are 2 400°C for **CAMIX** n°1 and 2 154°C for **CAMIX** n°2 and **COCHIX** n°3 (eutectic temperature of ZrO_2-MgO system).

Modifications of thermal conductivity properties due to irradiation damages (neutron effect) were taken into account by a damage factor. This factor is applied since the beginning of the irradiation. Other possible damages were considered such as the decrease of thermal conductivity by the appearance of porosity.

The decay thermal conductivity is then equal to:

$$\lambda_{\text{decay}} = \lambda_{100\%\ \text{TD}} \times 0.5 \times \frac{1 - \text{P}}{1 + (2 \times \text{P})}$$

where $\lambda_{100\%\ \text{TD}}$ is the thermal conductivity of fully dense compound,

0.5 is the damage factor,
and P is the pores volume fraction.

Concerning the gap gas composition (between pellet and cladding) and the target swelling we were considered the following hypothesis:

CAMIX-COCHIX gas release:	50% of the production
CAMIX n°1 swelling:	no swelling
CAMIX n°2 et COCHIX n°3 swelling:	10% volumic swelling at 340 EFPD

Thermal design has been carried out by two models with the finite elements code CAST3M:

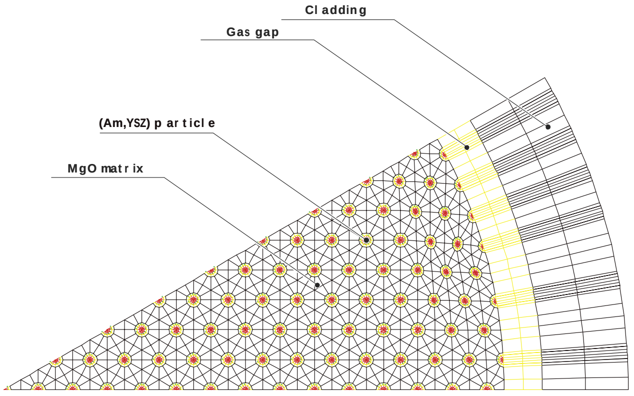
- the first one, allows calculation of CAMIX n°1 target temperature,
- the second one, [8] allows the calculation of CAMIX n°2 and COCHIX n°3 pellets. Its purpose is to calculate hot spots in the particles.

CAMIX n°1 model is an axysimetrical model (r,z) where thermal conductivity comes from measurements of the solid solution (see Figure 11). In this case, power is evenly distributed.

Figure 11. **CAMIX n°1 model – meshing**

The common CAMIX n°2 and **COCHIX** n°3 model is a 2D (r,θ) model where particles are homogeneously arranged in an hexagonal network (see Figure 12). In this case, power is only generated in (**Am-YSZ**) particles.

Figure 12. **CAMIX n°2 and COCHIX n°3 model – meshing**



For **CAMIX** n°1, the maximum temperature occurs at 340 EFPD and is equal to 1 380°C in the centre of the pellet. For **CAMIX** n°2 and **COCHIX** n°3, at 50 EFPD, temperature can reach 1 000°C in the centre of the pellet. In both cases, the target temperature remains lower than the melting temperature of the targets with large margins.

Concerning the cladding mechanical behaviour, the pin was designed so as to avoid a cladding rupture by internal pressure or by mechanical interaction with the pellet. [9]

Conclusions and perspectives

With the **MATINA**, **ECRIX** and **CAMIX-COCHIX** experiments, the first phase in the preparation of the transmutation programme in **Phénix** is now completed and successful. The experiment design studies were finalised and several target concepts were proposed:

ECRIX B&H:	AmOx microdispersed in MgO
CAMIX n°1:	(Am-YSZ) solid solution
CAMIX n°2:	(Am-YSZ) microdispersed in MgO
COCHIX n°3:	(Am-YSZ) macro masses in MgO

The duration to reach the objective for the testing material (cumulated extent of fission 30%) is respectively 670 EFPD for ECRIX-B and 340 EFPD for the other experiments. The irradiation of the **ECRIX** experiments will start next year and the starting point of the **CAMIX-COCHIX** experiment is scheduled at the beginning of 2004 for 340 EFPD in **Phénix**. At the end, we will have comparative elements which will allow us to choose the optimal concept for americium transmutation in fast reactor.

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