PHENIX EXPERIMENT FACILITIES FOR FISSION PRODUCT DESTRUCTION AND ACTINIDE BURNING

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ABSTRACT

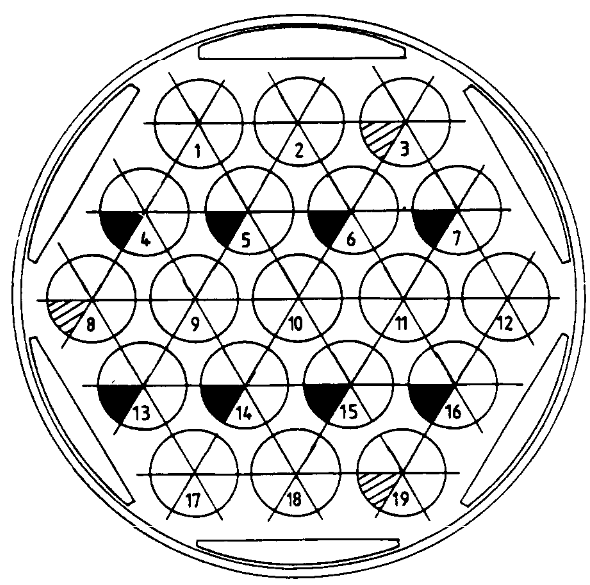
This paper reviews the experimental assessment of minor actinide and fission product transmutation in PHENIX. Already carried out tests are briefly described, such as the PROFIL experiment used to check the basic data files, and the SUPERFACT experiment aimed at proving the feasibility of minor actinide transmutation in fast reactors. The irradiation devices are described, especially the experimental moderated subassemblies used to produce C060. A calculation scheme of such moderated subassemblies is checked on experimental results of C060 production, and then applied to Pu238 production and Tc99 or 1129 destruction. The future experiments on minor actinides and fission product irradiation are described.

The PHENIX reactor is a powerful tool for experimental irradiations in fast reactor conditions. This paper will focus on the experiments, both performed and foreseen, about the transmutation of minor actinides and fission products.

I. REVIEW OF THE PERFORMED EXPERIMENTAL PROGRAM

A. In-tom experiments.

The PROFIL experiments, PROFIL-1 which took place during the first three cycles of PHENIX, and PROFIL-2, were made of 1 or 2 standard pins containing separate capsules (46 in PROFIL- 1 and 2x42 in PROFIL-2). A small sample (a few milligrams) of a pure element or isotope was placed in each capsule. These pins were put in a standard subassembly (S/A), in the first row of PHENIX inner tom, far away from neutronic perturbations. The post-irradiation analyses were compared to accurate evolution calculations (normalized to absolute fluence by the experimentally measured neodymium concentrations) using data from both the JEF-1 and the french CARNAVAL-IV data files. The C/E agreement is globally excellent, and this experiment helped to precise capture, fission and (n,2n) cress-sections (especially (n,2n) data for Pu239 and Np237), as well as branching ratios in the Am241 radiative capture decay chain and in the yield of Pu236 in Np237 irradiation.



4-16: 2% Am — 6-14: 20% Am + 20 % Np
5-15: 45% Np — 7-13: 2% Np

Figure 1- The SUPERFACT experimental cluster

The SUPERFACT experiment, performed in PHENIX from late 1986 to early 1988 during 5 irradiations cycles, was intended to demonstrate the feasibility of the transmutation of minor actinides (Np237 and Am241) in fast reactors. A S/A containing a 19-pin cluster (see figure 1 above) was irradiated, leading to an irradiation of 8.5 atoms%. 8 pins containing minor actinides mixed with uranium oxide were placed in the capsule: 2 with 2% Np237 each, 2 with 2% Am241 each, 2 with 45% Np237 each, and 2 with 20% Np237 and 20% Am241 each. The full interpretation of this experiment is still in progress; the transmutation rates for a 383 EFPD irradiation are between 25 and 30% for both Np237 and Am241[2].

B. Experiments in the radial blanket.

Various experiments took place in the radial blanket in order to optimize the production of radio-isotopes at a competitive price. All these experiments involved cobalt (but any other element could be irradiated) placed in moderated S/As in order to increase cross-sections and to reach high reaction rates, as the previous experiments without moderator (CARAPHE in the core and POSTILLON in the radial blanket) led to very low measured activities. The presence of a moderator induces power peaks near the target S/A, so the placement at the periphery of the radial blanket. In a series of experiments, called COMMODORE, two kinds of moderator have been tested (see figure 2 next page), beryllium and calcium hydride; the result was the choice of calcium hydride, more efficient than beryllium, with an optimized design of the S/A. Five carriers containing calcium hydride have been examined after a 218 EFPD irradiation to check for leaks; no leak was evidenced and the calcium hydride was found undamaged.

II. DESCRIPTION OF IRRADIATION DEVICES

The target S/As are constituted of a carrier containing a capsule at its center. In this capsule are placed the isotopes to be transmuted. The nature of the carrier depends on the irradiation zone: fuel pins in the core (as in the SUPERFACT experiment), steel pins or moderator tapes at the periphery of the core (see figure 2 next page). Inert earners can be reused from an irradiation to another. The use of a marker gas for the pins of such S/As, whereas standard pins are not so marked (the detection of clad failure being made by fission product or delayed neutrons detection) allows to track clad failures. A leakage of calcium hydride containers could be detected by the presence of Ar37 (Ca40 + n $\rightarrow$ Ar37 + α).

III. CALCULATION RESULTS

We present here the model designed to simuate transmutation in thermalized fast reactor S/As placed in the PHENIX radial blanket region. The results obtained in experimental Co59$\rightarrow$Co60 irradiations are used as a validation basis for this scheme, which is then applied to a heavy nuclide transmutation example (Np237$\rightarrow$Pu238) and finally to the transmutation of fission products (Tc99 and 1129), in order to estimate the expected performances of foreseen irradiations of such nuclides in PHENIX.

A. Modelling thermalized S/As.

All calculations are performed using the french code system for fast reactor core calculations, CCRR. The cell calculations are made with the CCRR HETARED module, using the standard CARNAVAL-IV library for fast reactors. However, more recent (JEF-2) 25-group capture and total cross-sections are used instead of CARNAVAL-IV ones for Tc99, 1129 and C059, while transfer data remain the same; when necessary, Np237 (n,2n) cross-sections are adjusted to Pu236 production with the branching values resulting from the interpretation of the PROFIL experiments. For spatial calculations, a 2D cylindrical (RZ) transport module is used, with the standard CCRR 25-group energy mesh.

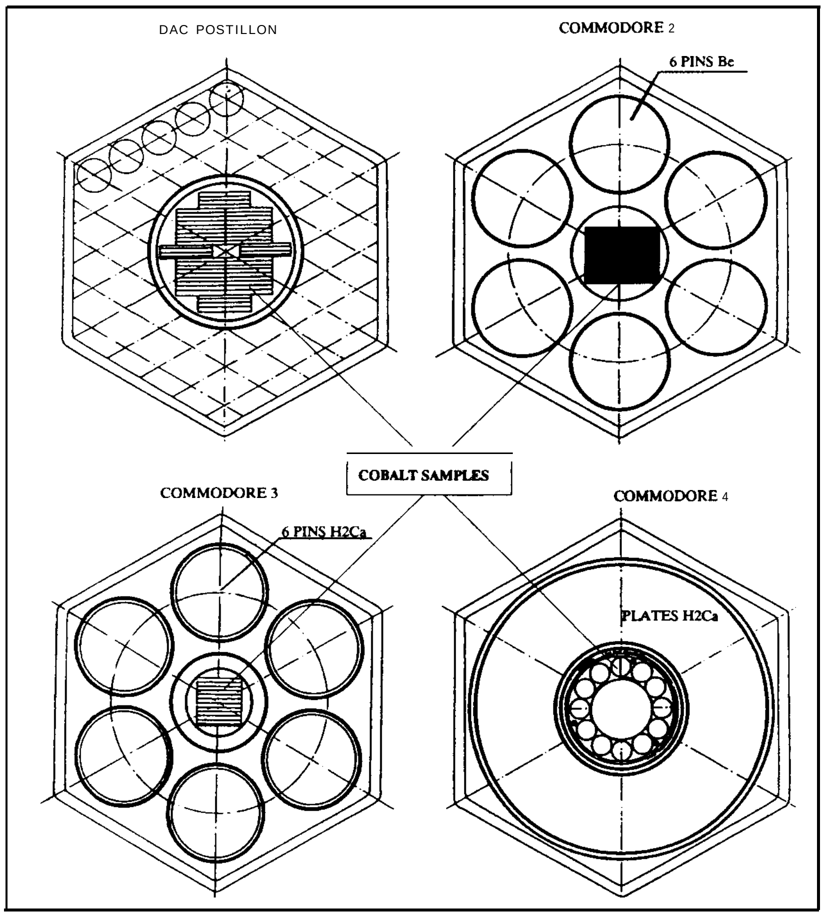


Figure 2- Various carrier and rig cross sections

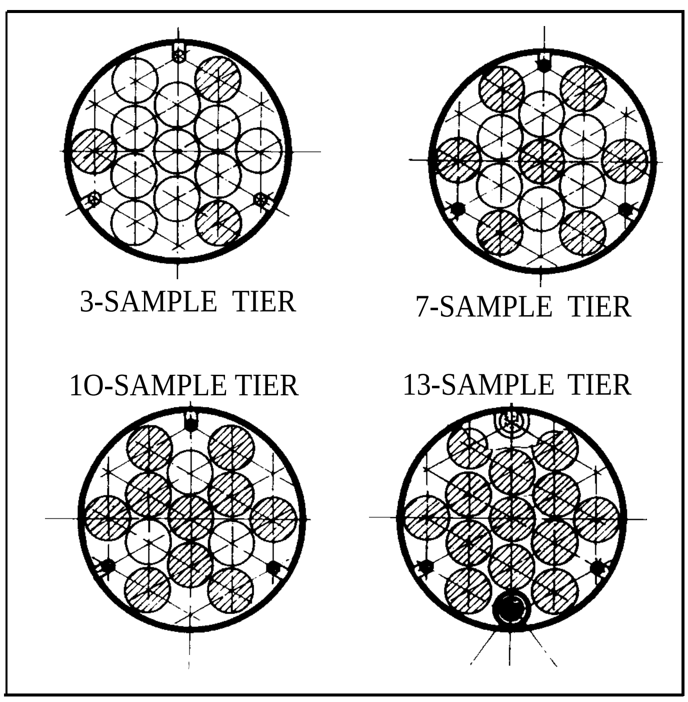


Figure **3-** Example of tiers in the COMMODORE-4 experiment (shaded = cobalt pin, blank = empty pin)

In order to model correctly a single target S/A placed in the radial blanket, far away from core 'center, a target thermalized S/A is placed at core center and surrounded by enough fertile S/As to simulate its position in the radial blanket (that is, in order to feed it with a correct incoming spectrum). A correction is then made on the flux level between this central position and the real one. Simple self-shielding corrections, using a dilution cross-section model with self-shielding coefficients to apply on multigroup cross-sections, have been taken into account. The validity of such an amount of approximations must then be checked on experimental measurements.

B. Comparisons with C059 irradiations.

The COMMODORE-4 experiment, with an annular carrier of calcium hydride and a complicated 8-tier structure in the capsule (see figure 3 on the previous page) with variable radial pin disposition in central (C), intermediate (I), and peripheral (P) rings (designed to check spatial shielding effects), has been modelled according to the preceding scheme. The C059 multigroup capture cross-sections, depending on the self-shielding level, are shown in figure 4 below. The results are summarized in the following table, showing calculated values, mean deviations to experiment and to infinite dilution calculation:

Ring	Calc. activity Ci/g	c/E (%)	C/C_∞ (%)
P	7.77	1.31	0.89
I	4.74	1.03	1.06
C	4.20	0.98	1.06

The discrepancies in calculations for the peripheral ring and the top and bottom tiers, nearest of the incoming flux from the moderator, may suggest that CARNAVAL-IV hydrogen slowing-down, while validated in FBR spectra, is not so reliable at low energies. This point will be discussed in the next section. As shown in the last column of the table, the energetic self-shielding effects are relatively low, while the spatial (radial) effects are larger.

Figure 5 next page gives an idea of the spectra in different regions. The reaction rate (RR) distribution at low energy for each kind of pin is given in the following table; the l-group averaged Co cross-sections range from 2.3 to 4.0 hams, depending on the position of the pin.

E (eV) <	0.42	3.1	23	101
RR in P ring (%)	75	86	92	96
RR in I&C rings (%)	62	77	88	94

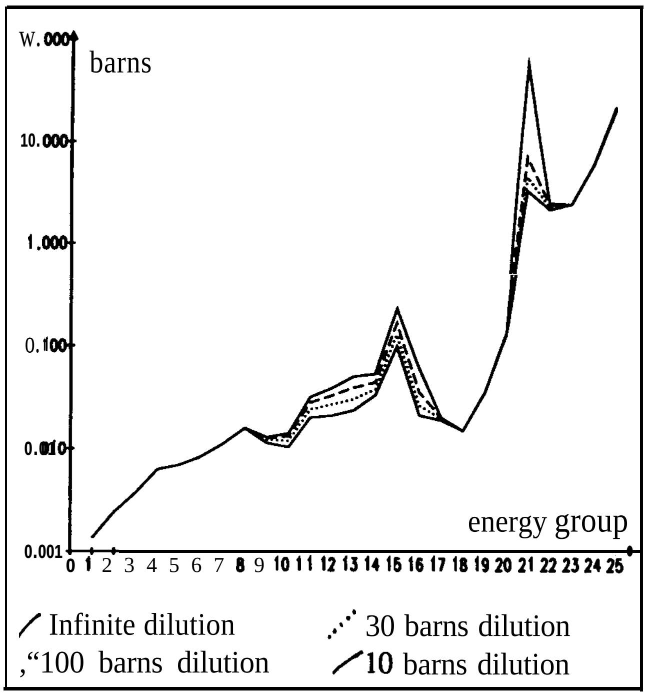


Figure 4- Self-shielded cross-sections of C059

C. Np237 → Pu238 transmutation.

Np237 target pins are irradiated to produce Pu238, useful in isotopic energy generators (spatial and medical research). These pins are placed in the optimized thermalized S/A, with only one ring of 12 target pins placed close to the moderator. The figures 6 and 7 next page show Np237 capture cross-sections and the spectrum in the Np237 region. The RR distribution at low energy is given in the following table. Energy self-shielding effects amount to a - 16% correction on reaction rates.

E (eV) <	0.42	3.1	23	101
RR (%)	**33**	**47**	**68**	**82**

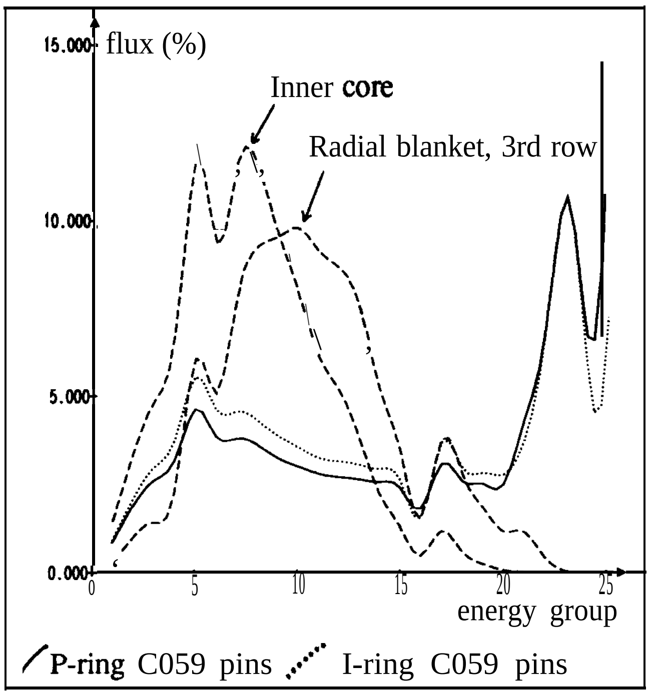


Figure 5- Flux spectra in inner core, radial blanket and cobalt pins

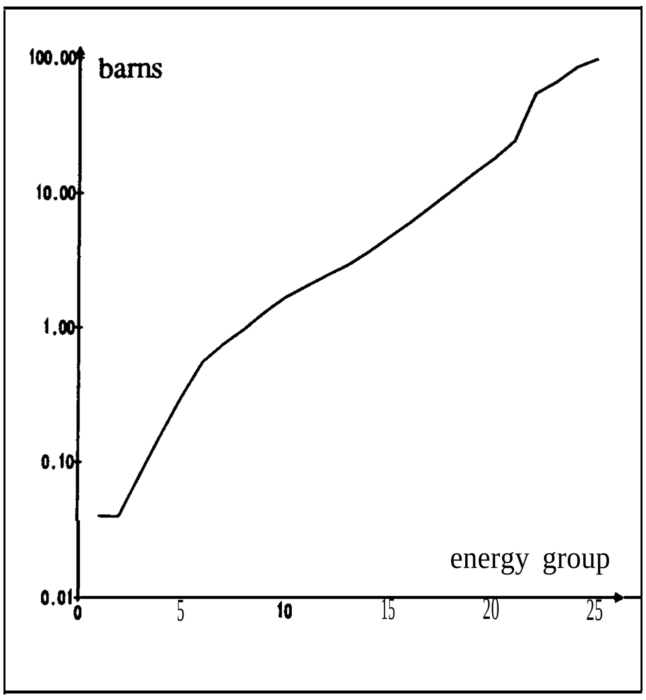


Figure 6- Self-shielded cross-sections of Np237

A rough estimate of the influence of hydrogen slowing-down has been made using a 7-group comparison between data issued **from** HETARED and APOLLO, a **PWR-validated** cell code (up to 7 energy groups could be obtained by overlapping the energetic meshes of the two codes). This lead to a further correction of -14% on reaction rates. The final l-group **capture** cross-sections for Np237 are then about 16 barns. Speaking in terms of **"half-**life" ($\ln 2 / \sigma \Phi$) of Np237 under irradiation, the calculated values **are** 2.7 years in the second **row** of radial **blanket,** and 5.2 years in the third one. The use of the branching ratios leading to **Pu236** formation during Np237 irradiation (drawn from the interpretation of the **PROFIL** experiments) shows that the content of **Pu236** in the plutonium **produced** should not exceed 3 ppm.

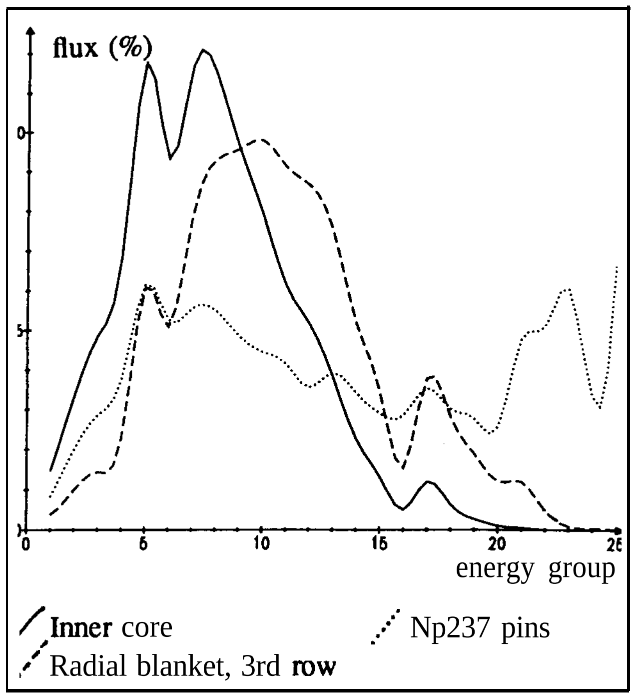


Figure 7- Flux spectra in inner core, radial blanket and Np237 pins

D. Tc99 and 1129 destruction.

Pellets made of **pure** Tc99 or 1129 (quite **unrealistic** for practical applications in **this** latter case, as the physical properties of iodine require to compound it with other elements in order to be able to withstand in-core conditions) **are** grouped in pins placed in the same fashion than in Np237 irradiations. The S/As are placed either in the second or third row of the radial blanket. The figures 8 to 10 show the Tc99 and 1129 capture cross-sections as well as the spectrum in the corresponding pins.

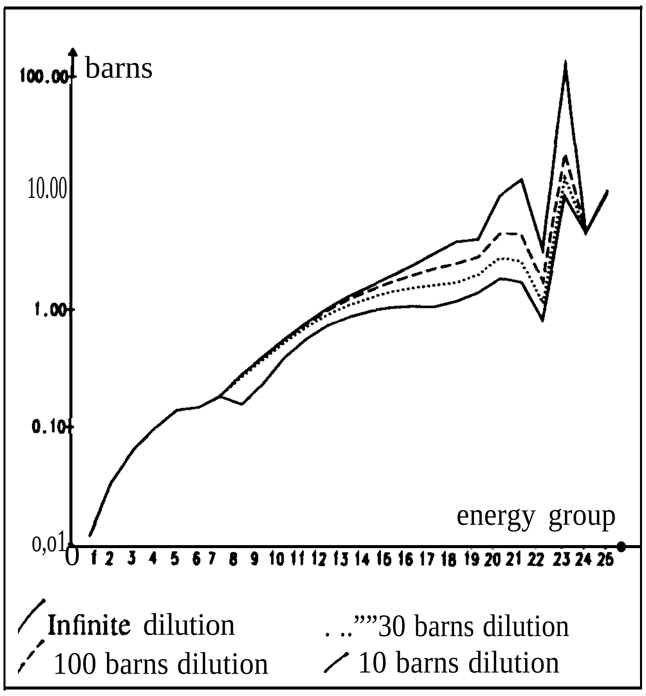


Figure 8- Self-shielded cress-sections of Tc99

Energy self-shielding effects amount to 30 % for Tc99 and 15 % for 1129. The RR distribution at low energy as well as l-group capture cross-sections (σ_c) and fluxes (Φ), together with the equivalent half-life ($T_{1/2}$) are given hereafter.

E (eV) <	0.42	3.1	23	101
Tc99 RR (%)	56	65	87	88
I129 RR (%)	86	92	94	95

For Tc99:

Position	2nd row	3rd row
Φ (10^{14}n/cm^2/s)	6.0	2.9
σ_c (barns)	3.5	3.7
$T_{1/2}$ (years)	10	21

For 1129:

Position	2nd row	3rd row
Φ (10^{14}n/cm^2/s)	6.9	3.3
σ_c (barns)	4.7	4.8
$T_{1/2}$ (years)	6.7	14

A study of a large-scale irradiation of fission products in fast reactors is also presented at this conference[3].

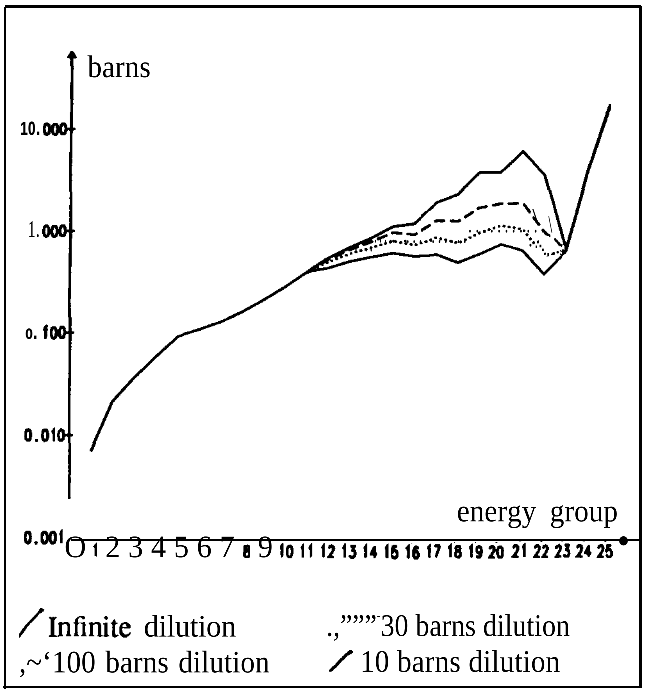


Figure 9- Self-shielded cross-sections of 1129

IV. REVIEW OF FORESEEN IRRADIATIONS

Independently of other irradiations, a bulk of some 20 moderated S/As placed in the radial blanket is scheduled to produce 1.5106 Ci/year (5.6 10^{16} Bq/year) of Co60.

An irradiation of Np237 and Am241 in a metallic fuel is scheduled (METAPHIX experiment) in collaboration with CRIEPI and TUI. It will be the first irradiation of metallic fuel in PHENIX; an analogous irradiation with oxide fuel is foreseen in support to SUPERPHENIX.

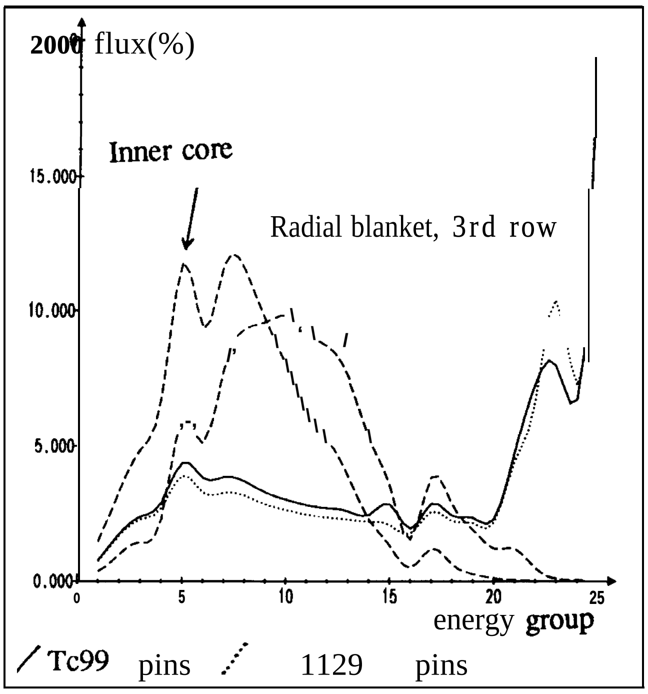


Figure 10 - Flux spectra in inner core, radial blanket and Tc99 and 1129 pins

An irradiation of Np237 in a moderated S/A placed in the radial blanket is also scheduled (NEPTUNIX experiment, with a 25-cm long pin containing 25 g of neptunium). Its aim is to determine the reaction rates on Np237, and especially the amount of Pu236 in the plutonium produced. Instead of pure NpO_2, a mixture NpO_2–MgO will be used (50 % $Np0_2$ in mass), in order to decrease the linear rating of the pins, in which Pu238 and small amounts of Pu239 will be created during irradiation. The pin diameter will be 7.44 mm, with a stainless steel cladding; an empty pin made of zircalloy, less absorbing at low energies, will be irradiated simultaneously to check its behavior in such temperature conditions. The coolant is allowed to flow inside the capsule in order to remove heat directly from the pins.

This program will be complemented with the irradiation of fission products, and especially a small sample of Tc99 in oxide form. Inert matrices such as MgO, $A1_20_3$ and $MgA1_20_4$ will also be irradiated, in order to check their behavior in reactor conditions.

REFERENCES

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2. C. PRUNIER et al., “Some Specific Aspects of Homogeneous Am- and Np-based Fuel Transmutation through the Outcome of the SUPERFACT Experiment in PHENIX”, this conference.

3. J. TOMMASI et al., “Long-Lived Waste Transmutation in Reactors”, this conference.