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ANALYSIS OF THE TRAPU AND DOUBLON IRRADIATIONS IN PHENIX FOR THE EXPERIMENTAL VALIDATION OF THE DARWIN PACKAGE FOR FAST REACTORS

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Abstract – *The prospect of reprocessing the fissile and fertile subassemblies of the PHENIX reactor, as well as the development of the 4th generation Fast Reactors make it essential to have a validated code for fuel inventory and fuel cycle calculations in Fast Reactors.*

This validation is based on irradiation experiments that were performed in PHENIX between 1977 and 1981. We have analyzed two of these experiments (i.e. TRAPU and DOUBLON) with the ERANOS-2.2 code system, associated with its nuclear data libraries JEFF-3.1.1 and the depletion module DARWIN-2.3.2. For each experiment, we have computed Calculation/Experiment (C/E) ratios for different isotopic ratios at the end of the irradiation. A fluence adjustment was performed in the calculations in order to match the ^{148}*Nd production.*

During the TRAPU experiment, ten well characterized fuel pins – of three different enrichment and Pu isotopic vectors – were placed in two subassemblies near the center of the PHENIX core between 1977 and 1979.

The analysis of this experiment shows a good C/E agreement for the main isotopes: ^{234}*U,* ^{235}*U,* ^{236}*U,* ^{239}*Pu,* ^{240}*Pu,* ^{242}*Pu and* ^{241}*Am. Discrepancies on some C/E's (*^{237}*Np,* ^{238}*Pu,* ^{241}*Pu, ...) are consistent with previous interpretations of TRAPU and with the analysis of separated samples irradiation experiments like PROFIL, that can be interpreted in terms of nuclear data.*

The purpose of the DOUBLON experiment was the validation of radial fertile blankets calculations, based on a detailed study of two subassemblies – of the first and second rows - that were irradiated in PHENIX between 1978 and 1981. Samples were taken at various heights in 9 fertile pins, chosen at increasing distances from the core.

The analysis of this experiment shows that the C/E's on the final amounts of ^{234}*U,* ^{235}*U,* ^{236}*U and* ^{239}*Pu are excellent. This is all the more important as they are the main isotopes in the final inventory. Concerning the* ^{240}*Pu - which is produced in much smaller quantities – the average C/E is also very good, but with a high uncertainty coming from the most peripheral pin. On the contrary, the C/E's of the* ^{238}*Pu,* ^{241}*Pu and* ^{242}*Pu are higher, with a very important pin-to-pin dispersion. These nuclei are produced in a very small amount, in total they represent less than one thousandth of the Pu produced. They are mainly produced by successive capture reactions on* ^{238}*U, which have significant cross-sections only in energy groups where the neutron flux is extremely low. Moreover, some of these resonant cross-sections are described in only one or two groups of our energy mesh, resulting in a large calculation uncertainty on the production rate of* ^{238}*Pu,* ^{241}*Pu and* ^{242}*Pu.*

I. INTRODUCTION

The DARWIN package [1] computes the evolution of the radioactive nuclides concentrations in a reactor, which is governed by the generalized Bateman differential equation. The code uses cross sections libraries [2] as well as a number of application libraries [3], in order to provide the following fuel cycle parameters: material balance, decay heat, activity, sources, etc… DARWIN can be coupled to either the ERANOS-2.2 or APOLLO2 neutronic calculation codes [4,5], in order to compute the material balance of a reactor subassembly after irradiation and cooling.

DARWIN is used intensively in an industrial context for LWR's and its validation for this type of reactor is very developed [5, 6]. For Fast Reactors, the validation of DARWIN needs to be improved. This situation has been pointed out recently, because of the perspective to reprocess both the fissile and fertile assemblies of the PHENIX reactor. Moreover, the prospect of the development of 4th generation fast Reactors - and primarily ASTRID [7] - now makes it essential to have a reliable validation of DARWIN for Fast Reactors.

For this purpose, we have analyzed irradiation experiments of fissile and fertile assemblies that were performed in PHENIX between 1977 and 1981:

- In the TRAPU experiment, fuel pins have been irradiated in a well characterized spectrum near the center of PHENIX. The main objective was to study the production of minor actinides (Np, Am, Cm), starting with different isotopic compositions of Plutonium.

- The purpose of the DOUBLON experiment was the qualification of radial fertile blankets calculations, in order to determine the external regeneration gain. The experiment performs a detailed study of two fertile subassemblies of the first and second rows.

We have used the JEFF-3.1.1 / ERANOS-2.2 / DARWIN-2.3.2 package to compute Calculation / Experiment ratios (C/E's) for some isotopic ratios in both experiments.

II. Analysis of the TRAPU Experiment

II.A. Experimental Setup

This irradiation experiment ran for 6 cycles (1977-1979) in the central area of the PHENIX reactor.

The experimental setup consisted of ten fuel pins - with standard geometrical characteristics - placed in the central part of two subassemblies near the core center. Table I shows the three very different plutonium enrichment and isotopic vectors that were used, the diluent being natural Uranium.

TABLE I

Compositions of the TRAPU pins

	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	$(PuO_2)/(U,Pu)O_2$ (% in mass)
TRAPU1	0.12	73.26	21.92	3.99	0.71	19.60
TRAPU2	0.77	71.37	18.54	7.42	1.90	19.25
TRAPU3	0.22	33.97	49.40	10.03	6.38	28.04

Figure 1 shows the positions of the TRAPU pins in the core. They are located in two subassemblies in the core center, near a control rod. However, the absorbing column remains well above the experimental samples, so we can expect a shift in the average flux during the irradiation but the spectrum effect should be negligible.

The pins remained 649 days in the reactor, which corresponds to 364.5 Equivalent Full Power Days (EFPD's). It corresponds to a fission rate of about 6 to 7 heavy nuclides per 100 initial heavy nuclides (6-7 at%).

Once the irradiation was finished, 10mm samples were cut in each pin in an axial position corresponding to the core midplane; these samples were analyzed by mass spectroscopy, after an isotopic dilution. The Curium was analyzed by both α and mass spectroscopy.

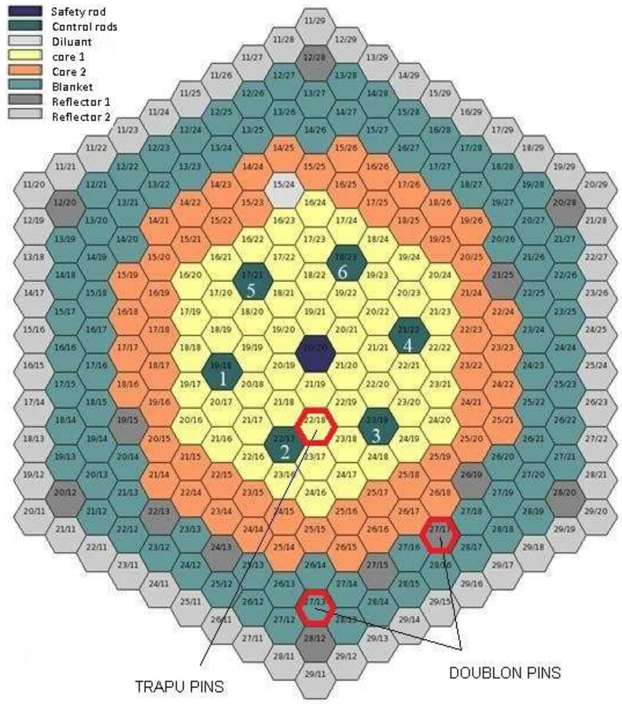


Fig. 1. Positions of the TRAPU and DOUBLON pins in the PHENIX reactor.

II.B. Calculation Route

Our calculations are performed with the ERANOS-2.2 code system [4]. It uses the JEFF-3.1.1 libraries [2], which contain anisotropic transfer sections (Legendre order 1) and probability tables for many nuclides, in order to take into account the self-shielding effects.

The cell calculations are performed by the ECCO [8] code, which treats simultaneously the self-shielding effects (by the sub-groups method) and the determination of flux and current (using collision probabilities). The heterogeneous subassembly is calculated in 1968 energy groups. It is described as a hexagonal array of pins, surrounded by a stainless steel wrapper. Each cylindrical pin is described as two concentric cylinders, representing the fuel pellet and the cladding.

Concerning the control rods, the B_4C density is simply adjusted using the multiplication factor used in the reactor monitoring calculations.

The core calculations have been performed with the AVNM [9] module (Advanced Variational Nodal Method), which solves the Boltzmann equation with a 33 energy groups mesh, using a Variational Nodal method for the spatial processing and a decomposition of the spherical harmonics (PN) for the angular treatment. In our case, the angular treatment is limited to the SP3 order, using the simplified approximation of the spherical harmonics.

Once the reaction rates and spectrum have been calculated for each cycle and experimental position, we have used the DARWIN-2.3.2 [1] code in order to evaluate accurately the evolution of the concentrations of each isotope in the samples. In DARWIN, the Bateman equations are solved either by an analytical method or by a Runge-Kutta fourth order numerical method. The code uses cross section libraries as well as application libraries [3].

II.C. Comparison with the Experiment

Table II shows the Calculation/Experiment ratio (C/E) for different isotopic ratios after the irradiation. We have made an average of the samples corresponding to pins of the same kind (one for TRAPU-1, two for TRAPU-2 and TRAPU-3). The uncertainty is the maximum between the individual C/E's dispersion and the quadratic sum of the experimental uncertainties for the samples of the same kind (which are not reported in the table). In fact, the observed dispersion can significantly exceed the estimated experimental uncertainty.

TABLE II

Calculation / Experiment on the final amount of the nuclides measured in TRAPU

		TRAPU - 1	TRAPU - 2	TRAPU - 3	average	σ
234U /	238U	1,000	1,015	1,044	**1,019**	0,024
235U /	238U	1,004	1,024	1,021	**1,016**	0,012
236U /	238U	0,958	0,978	0,981	**0,973**	0,014
237Np /	238U	0,868	0,860	0,819	**0,849**	0,030
239Pu /	238U	1,016	1,000	0,998	**1,004**	0,011
238Pu /	239Pu	0,952	0,972	0,955	**0,959**	0,012
240Pu /	239Pu	0,986	0,982	1,003	**0,991**	0,012
241Pu /	239Pu	0,962	0,971	0,975	**0,969**	0,007
242Pu /	239Pu	1,025	1,011	1,006	**1,014**	0,011
241Am /	239Pu	0,960	0,996	0,994	**0,983**	0,024
242Amm /	241Am	1,048	1,050	1,008	**1,035**	0,027
243Am /	241Am	1,068	1,021	1,064	**1,051**	0,030
244Cm /	239Pu	0,895	1,016	1,037	**0,983**	0,086
242Cm /	244Cm	1,168	1,023	0,994	**1,061**	0,106
243Cm /	244Cm	-	0,638	0,622	**0,630**	0,008
245Cm /	244Cm	-	1,204	1,490	**1,347**	0,143
148Nd /	238U	1,009	1,002	0,988	**1,000**	0,012
143Nd /	148Nd	0,965	0,972	0,973	**0,970**	0,005
144Nd /	148Nd	1,046	1,049	1,072	**1,056**	0,016
145Nd /	148Nd	0,985	0,991	0,991	**0,989**	0,004
146Nd /	148Nd	0,992	0,999	0,994	**0,995**	0,004
150Nd /	148Nd	0,983	0,978	0,976	**0,979**	0,004

A +1.5 % fluence adjustment was necessary in order to match the ^{148}Nd production, which was used as a burnup indicator. Indeed, the ^{148}Nd is a stable fission product with a small capture cross section and therefore enables the determination of the number of fission reactions in the samples.

The accuracy of the fluence calculation might be limited for the following reasons:

- there is an uncertainty of 2 to 3% on the core nominal power,
- in our calculations, we have used average compositions for each row of subassemblies,
- we have also used an average control rod position for each reactor cycle,
- the neutronic weight of the control rods was simulated by adjusting the boron concentration rather than by using a reactivity equivalence method.

Table II shows C/E behaviors that are consistent with previous interpretations of TRAPU and with interpretation of separate sample irradiation experiments like PROFIL, that can be interpreted in terms of nuclear data [10, 11]. To be more specific:

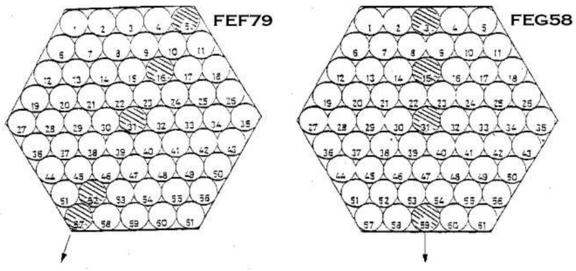
- There is a minor overestimation of the ^{235}U/^{238}U ratio, especially sensitive to the fission of ^{235}U.
- The ^{236}U/^{238}U ratio - especially sensitive to the capture of ^{235}U - is slightly underestimated.
- There is a significant underestimation of the final amount of ^{237}Np, which comes from an underestimation of the ^{238}U(n,2n) cross-section. Again, this is consistent with the analysis of PROFIL.
- The final quantities of ^{239}Pu and ^{240}Pu – which are the main isotopes in the initial isotopic composition - depend little on the fluence and are well predicted.
- The underestimation of ^{238}Pu may come from its sensitivity to the exact reactor operation, via the decay of ^{242}Cm.
- The underestimation of ^{241}Pu is related to an underestimation of the integral capture of ^{240}Pu.
- There is a good prediction of the final amount of ^{241}Am, ^{242}Am and ^{243}Am, given the associated dispersions (see Table II).
- The final amounts of ^{242}Cm and ^{244}Cm are relatively well predicted - with significant uncertainties - unlike those of ^{243}Cm and ^{245}Cm.

III. Analysis of the DOUBLON Experiment

III.A. Experimental Setup

The purpose of the DOUBLON experiment was the validation of radial fertile blankets calculations, based on a detailed study of two standard subassemblies – of the first and second row - that were irradiated in PHENIX between 1978 and 1981. In each subassembly, the experimental pins – shown on Figure 2 - have been selected along an axis from the core center. These experiments had not been analyzed before.

Fig. 2. Position of the DOUBLON pins in the first (FEF79) and second (FEG58) row subassemblies. The arrow indicates the direction of the core center and the experimental pins are shaded



- The first row subassembly (FEF79, with 0.44% of ^{235}U) was irradiated in position 27-17 (see Figure 1) during 639.6 EFPD’s.

- The second row subassembly (FEG58, with 0.47% of ^{235}U) was irradiated in position 27-13 (see Figure 1) during 758.5 EFPD’s. The environment of this subassembly has changed during the irradiation, which was taken into account in our core calculations.

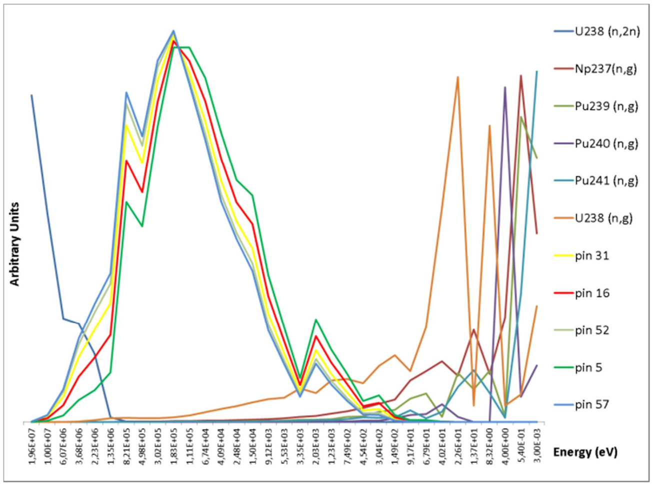
In each pin, a 20mm sample was cut at the core midplane (0mm) after the irradiation. For some pins, additional samples were cut in a low position (-300 mm), a very low position (- 500mm, i.e. at the level of the lower axial blanket) and a high position (+300 mm).

The samples were cut at the LAMA in Grenoble and transferred to the SEN/COMIR in Cadarache for dissolution. The analyses were then performed at the DCAEA / SEA / SEACC in Fontenay aux Roses concerning the following isotopes: ^{234}U, ^{235}U, ^{236}U, ^{238}U, ^{238}Pu ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu and the isotopes of neodymium (used for fluence adjustment).

III.B. Calculation Route

The calculation scheme is the same as for the analysis of TRAPU. We observe on the left side of Figure 3 – which shows the energy spectrum in the various pins of the first row subassembly FEF79 - a strong spatial variation of the neutron spectrum during the penetration in the blanket. It is therefore essential to use the "local" spectrum of the experimental pin during the cross section condensation from the ERANOS 33 groups mesh to the one group mesh used in DARWIN. Indeed, ERANOS also provides a one-group cross section, but it is condensed on an average spectrum of the blanket and using it would introduce a significant bias in the reaction rates calculations.

Fig. 3. 33 energy groups spectrum in the pins of subassembly FEF79, compared to some reaction cross sections (arbitrary units). The energy is in eV



III.C. Comparison with the Experiment

Table III shows the C/E for different isotopic ratios at the core midplane after the irradiation. In the table, the pins are ordered from the left to the right by their distance to the core, FEF79_57 being the nearest and FEG58_3 the furthest. We have not reported all the pins in the table in order to make it easier to read, but the average is made on all the pins available. The high uncertainties on some average ratios mainly reflect the pin-to-pin dispersion.

As our initial compositions of the blanket does not contain any ^{234}U or ^{236}U, we had to introduce an arbitrary amount of these isotopes – respectively 50ppm and 500ppm - in order to have a good C/E agreement after the irradiation.

For the same reason as previously, a fluence adjustment of +1% is necessary to match the ^{148}Nd production (the effect is an increase of 1.3 % of the ^{148}Nd/^{238}U ratio). The uncertainty is determined in the same way as previously.

TABLE III

Calculation / Experiment on the final amount of the nuclides measured in DOUBLON

C/E	FEF79_57	FEF79_5	FEG58_59	FEG58_3	…	average	σ
234U / 238U	1,06	1,02	1,05	1,02		**1,05**	-
235U / 238U	0,99	1,02	0,99	1,01		**1,00**	0,02
236U / 238U	1,05	1,02	1,13	1,10		**1,08**	-
239Pu / 238U	0,99	0,92	1,00	0,90		**0,98**	0,08
238Pu / 239Pu	2,53	0,58	0,93	0,45		**1,31**	1,2
240Pu / 239Pu	1,03	0,95	0,94	0,86		**0,97**	0,11
241Pu / 239Pu	1,01	0,74	0,83	0,46		**0,76**	0,3
242Pu / 239Pu	0,46	0,22	0,17	0,06		**0,26**	-
148Nd / 238U	1,04	0,74	1,09	0,87		**1,00**	0,26
143Nd / 148 Nd	0,95	0,96	0,92	0,93		**0,93**	0,03
144Nd / 148 Nd	1,08	1,08	1,01	1,00		**1,03**	0,05
145Nd / 148 Nd	1,00	1,00	0,98	0,98		**0,98**	0,02
146Nd / 148 Nd	1,02	0,99	1,00	0,98		**0,99**	0,03
150Nd / 148 Nd	0,93	0,96	0,97	0,97		**0,96**	0,03

Figure 4 shows the evolution of the concentrations of different isotopes in the central pin of subassembly FEG58. The final concentrations – normalized to the content in ^{238}U - are given in Table IV.

Fig. 4. Evolution of the nuclides concentrations in the central pin of subassembly FEG58. The time is in days, the concentration in arbitrary units, normalized to ^{238}U

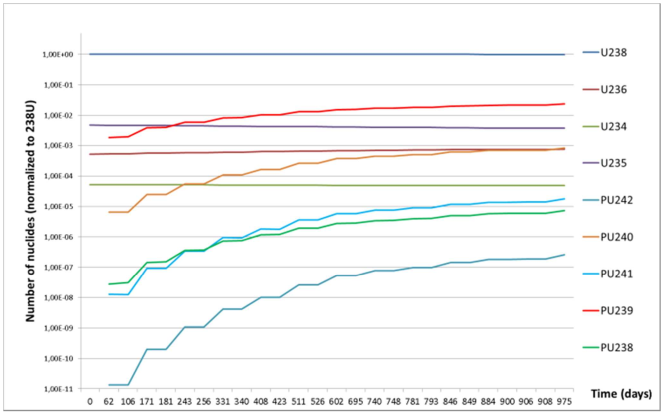


TABLE IV

Final concentrations - normalized to the content in ^{238}U – at the end of the DOUBLON irradiation

238U	**1,00E+00**
239Pu	2,37E-02
235U	3,79E-03
240Pu	8,26E-04
236U	7,54E-04
234U	4,78E-05
241Pu	1,79E-05
238Pu	7,37E-06
242Pu	2,61E-07

In Table III we observe that:

- There is a good C/E for the final amounts of ^{234}U and ^{236}U, which shows that we have introduced a reasonable amount of these isotopes to the initial composition.
- The C/E's on the final amounts of ^{235}U (that comes mainly from the initial ^{235}U) and ^{239}Pu (that is produced by capture on ^{238}U) are excellent. This is all the more important as Table IV shows that these are the main isotopes in the final inventory.
- For the ^{240}Pu the C/E is also very good, but with a rather high uncertainty. The ^{240}Pu is produced in small quantities – 30 times less than the ^{239}Pu - by successive captures on the ^{238}U and ^{239}Pu.
- The C/E's of the ^{238}Pu, ^{241}Pu and ^{242}Pu are high, with large pin-to-pin dispersions. Unlike in the fuel, these isotopes are not present in the initial composition and they are mainly produced by the following reactions:

238U(n,2n)→237U(β-)→**237Np(n,γ)** →238Np(β-)→238Pu

238U(n,γ) →239U(β-)→239Np(β-)→**239Pu(n,γ) →240Pu(n,γ)** →241Pu

238U(n,γ) →239U(β-)→239Np(β-)→2**39Pu(n,γ) →240Pu(n,γ) →241Pu(n,γ)** →242Pu

Figure 3 shows the cross sections of the reactions listed above (in bold). We observe that they are significant only in energy groups where the number of neutrons is extremely low. Moreover, some of these resonant cross-sections are significant in only one or two groups of our 33 groups mesh. We therefore understand that the calculation of the ^{238}Pu, ^{241}Pu and ^{242}Pu production is associated with a very large uncertainty. That being said, Table IV reminds us that these nuclides are produced almost as traces, totaling less than one thousandth of the Pu produced. Significant uncertainty on the prediction of the production of these nuclides is therefore of little importance in the final material balance calculation.

The C/E's for the samples in the other axial positions are not reported here, but they show the same trends as at the core midplane. In the high and low positions (resp. +/- 30cm) the fluence is about 30% lower than at the core midplane. In the very low position - located at the level of the lower axial blanket - it is approximately 60 % lower.

IV. Conclusion

We have analyzed irradiation experiments of fissile and fertile assemblies that were performed in PHENIX between 1977 and 1981. We have used the JEFF-3.1.1 / ERANOS-2.2 / DARWIN-2.3.2 package to compute Calculation / Experiment ratios (C/E's) for different isotopic ratios in both experiments.

- In the TRAPU experiment, fuel pins have been irradiated in a well characterized spectrum near the center of PHENIX. The main objective was to study the production of minor actinides (Np, Am, Cm), starting with different isotopic compositions of Plutonium. From the analysis of the experiment, we observe a good C/E agreement for the main isotopes: ^{234}U, ^{235}U, ^{236}U, ^{239}Pu, ^{240}Pu, ^{242}Pu and ^{241}Am. Discrepancies on some C/E's (^{237}Np, ^{238}Pu, ^{241}Pu, ...) are consistent with previous interpretations of TRAPU and with the analysis of separated samples irradiation experiments like PROFIL, that can be interpreted in terms of nuclear data.

- The purpose of the DOUBLON experiment was the validation of fertile blankets calculations, in order to determine the external regeneration gain. The experiment performs a detailed study of two fertile subassemblies of the first and second row. The average C/E's on the final amounts of ^{234}U, ^{235}U, ^{236}U and ^{239}Pu are excellent. This is all the more important as they are the main isotopes in the final inventory. Concerning the ^{240}Pu - which is produced in much smaller quantities – the average C/E is also very good, but with a rather high uncertainty coming from the most peripheral pin. On the contrary, the C/E's of the ^{238}Pu, ^{241}Pu and ^{242}Pu are high, with a very important pin-to-pin dispersion. These nuclei are produced in a very small amount, in total they represent less than one thousandth of ^{239}Pu produced. They are mainly produced by successive capture reactions on ^{238}U, which have significant cross-sections only in energy groups where the neutron flux is extremely low. Moreover, some of these resonant cross-sections are described in only on one or two groups of our energy mesh, resulting in a large calculation uncertainty on the production rate of ^{238}Pu, ^{241}Pu and ^{242}Pu.

Both experiments provide a good basis to validate the evolution calculations of fissile and radial blanket subassemblies.

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