Use of Irradiation Experiments for Validating ENDF/B-VII Data

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Introduction

- **The advanced nuclear systems and associated fuel cycles will need good quality cross section data to provide a reliable assessment of their performance.**
- **Closed fuel cycles with the objective of waste minimization imply, from a physics point of view:**
  - **A high content of minor actinides in the reactor core and in the fuel cycle**
  - **A high Fissile/Fertile isotope content in the core fuel**
  - **A variable, and potentially degraded, Pu isotopic vector in all the fuel cycle**
  - **Lower fuel density to achieve lower conversion ratios**
- **Basic data are available for TRU (transuranics) isotopes (up to Cf) but a validation is needed in order to quantify their reliability. The high amount of minor actinides (MA) foreseen in advanced fuel cycle systems requires a specific validation work especially for capture and fission cross sections of such isotopes.**

Introduction

- **The validation is traditionally done through the use of differential and integral experiments, and uncertainty assessment.**
- **The information that can be gathered on minor actinides (MA) from experiments comes mostly from small sample irradiation, reactivity oscillation, and fission and capture rates measurements. Separate isotope sample and fuel pin irradiation in power reactors provides a unique source of very useful measurements.**
- **The results of the analysis of such experimental data provide indications to nuclear data evaluators for improving the quality of basic files, and to assess their impact on advanced fuel cycles. These kinds of experiments belong to the category of elemental (separate) effects used in validation methodology.**

Introduction

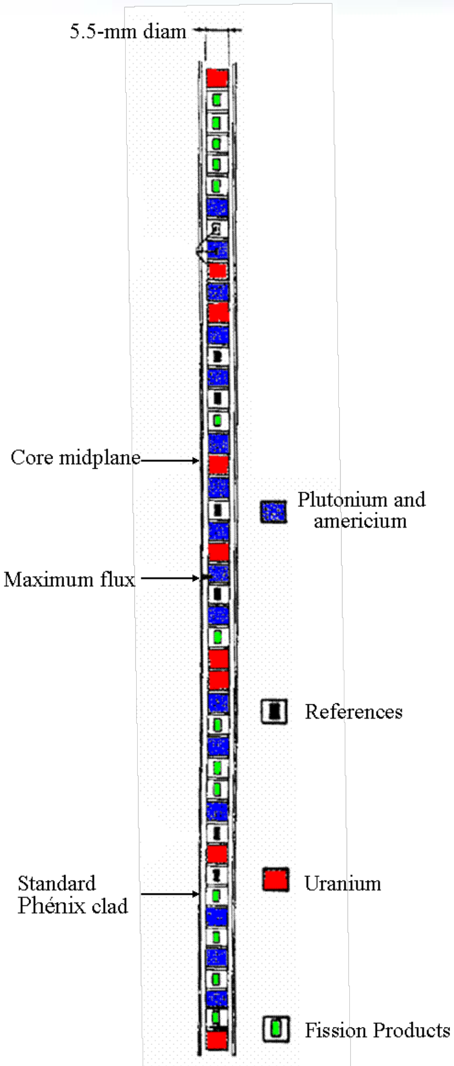
- **The experimental data of the PROFIL and TRAPU irradiation experiments , performed at the CEA fast reactor PHENIX, provide very clean and useful information on both cross section data and transmutation rates of actinides.**
- **These data are essential for the validation of the methods and data to be used in advanced fuel cycles where transmuter systems will be used to reduce the existing inventory of nuclear waste. In this presentation these irradiation experiments are used for validating ENDF/B-VII data.**
- **Moreover, in the validation process the use of sensitivity analysis allows to better gather information and indicate specific needs.**
- **A further question to be answered:**

Is there a need for further experiments or it is sufficient to have access to existing data and exploit them for future applications in advanced fuel cycle programs?

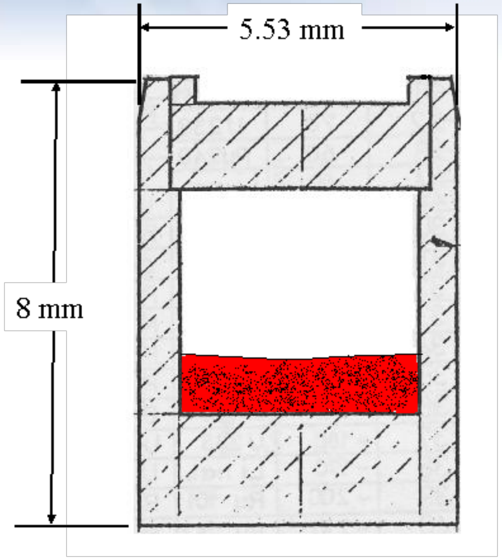
PROFIL-1 Experiment

- **During the PROFIL-1 experiment performed in 1974 a pin containing 46 samples of pure isotopes, including fission products, major and minor actinides (Uranium, Plutonium, and Americium isotopes) was irradiated in the PHENIX fast reactor for the first three cycles, corresponding to a total of 189.2 full-power days.**
- **The experimental pin was located in the central subassembly of the core, and in the third row of pins inside the subassembly. This location is far away from neutronic perturbations allowing clear irradiation conditions.**
- **Following the reactor irradiation, mass spectroscopy was then used, with simple or double isotopic dilution and well-characterized tracers to measure concentrations. The experimental uncertainty obtained with this method was relatively small (maximum of 3%).**

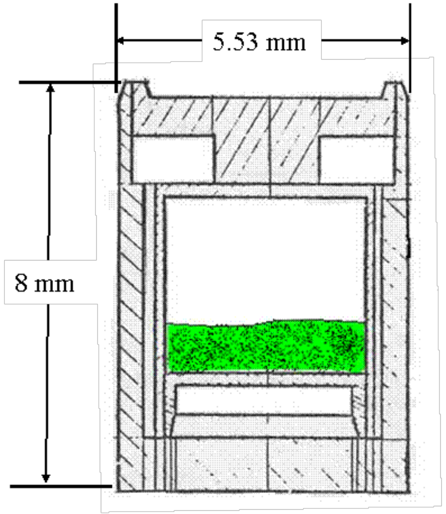
Isotope Sample Irradiation Pin Description



Axial distribution of isotope samples in irradiation pin



Single-wall capsule for Actinide samples



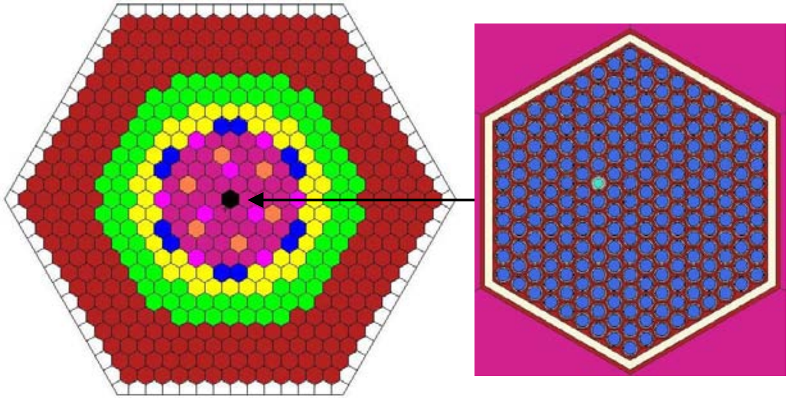
Double-wall capsule for fission product samples

PROFIL-2 Experiment

- **The second part of the PROFIL irradiation campaign took place in 1979. During the experiment two standard pins, each containing 42 separated capsules of fission products, major and minor actinides (Uranium, Plutonium, Americium and Neptunium isotopes), were irradiated for four cycles (from 17th to 20th) in the fast neutron spectrum reactor PHENIX in France.**
- **The experimental pins were located in the second row of the core and in the two experimental pins in the third row of the subassembly. Chemical and mass spectrometry analyses have been subsequently performed to determine the post-irradiation isotopic concentrations.**

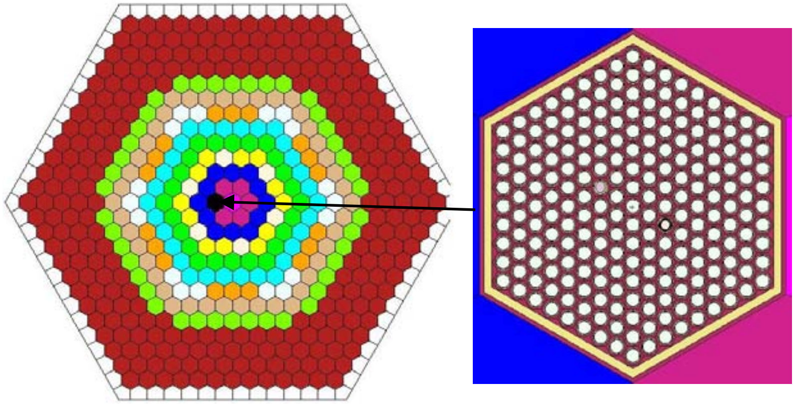
PROFIL-1 & 2 Analyses

- MCNP models for PROFIL-1&2 experiments have been developed.
- One group cross sections for PROFIL-1 samples were calculated by means of solutions obtained by taking the batch statistics of several runs with recorded surface sources.



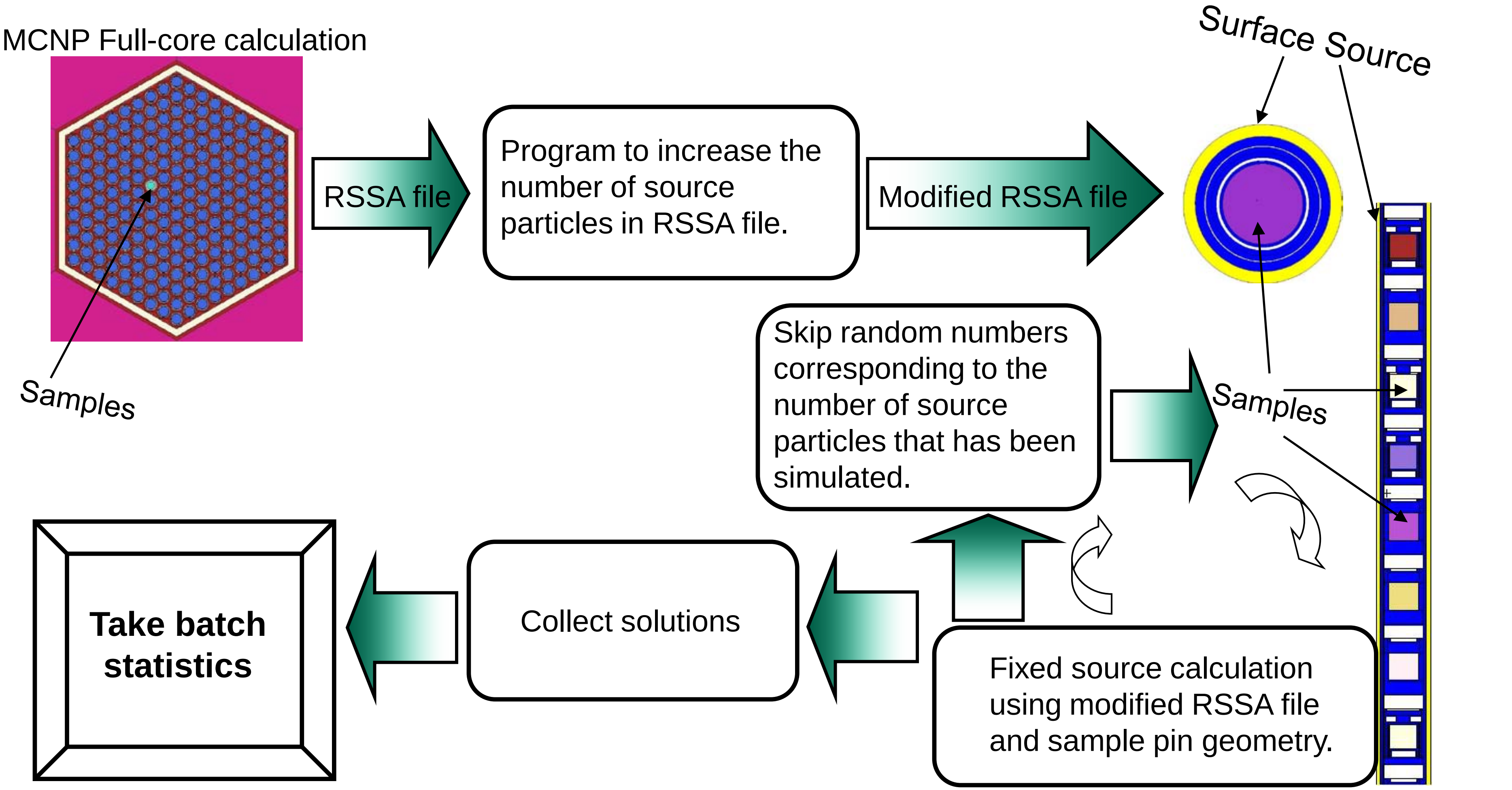
PROFIL-1

- For deterministic calculations a full-core VARIANT model and ECCO subassembly model for PROFIL-1 have been developed. The full-core VARIANT calculation has shown very close k_{eff} to that calculated by MCNP.



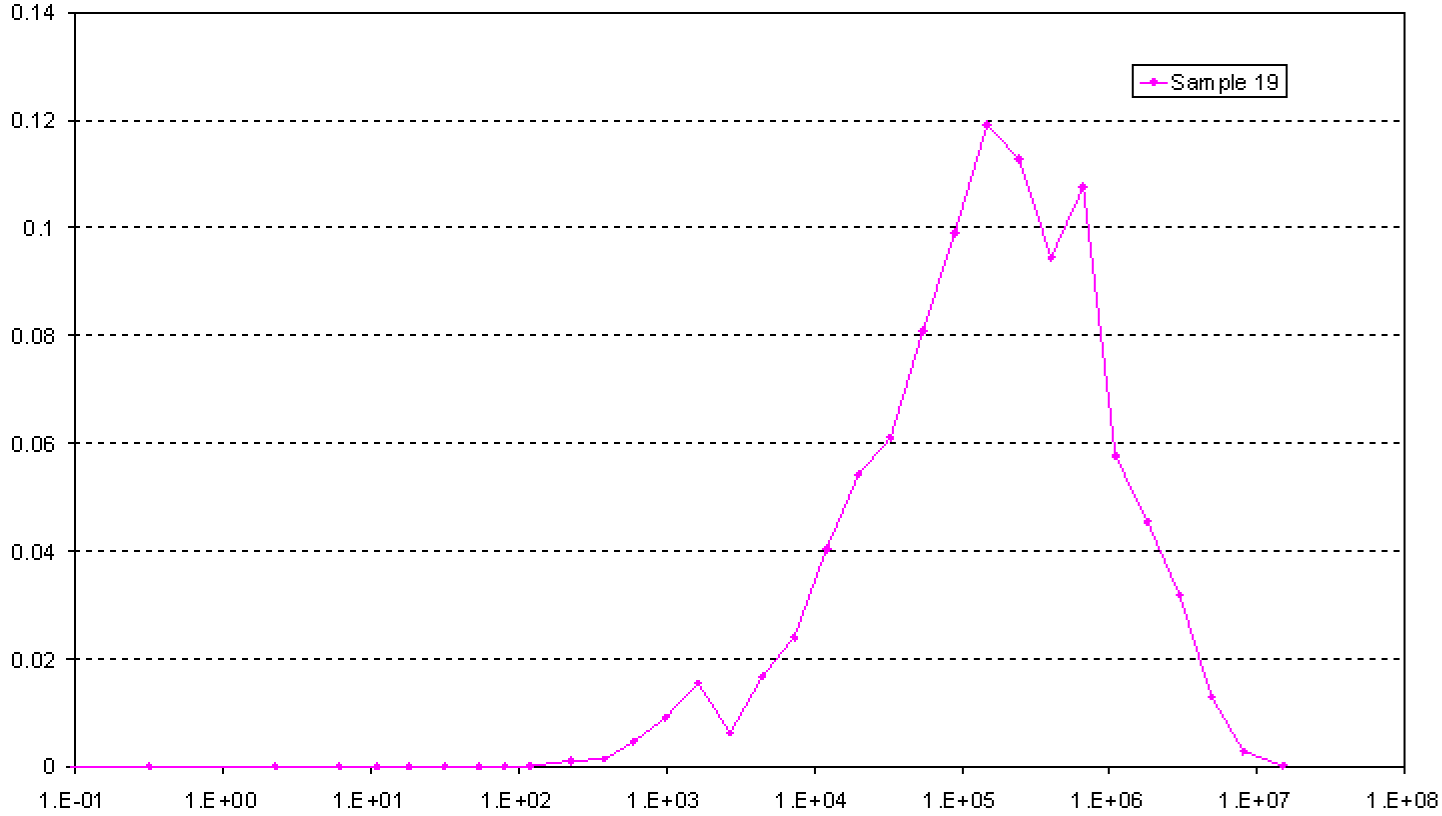
PROFIL-2

Procedure to calculate one-group cross sections over each sample with MCNP



Neutronics Calculation Results

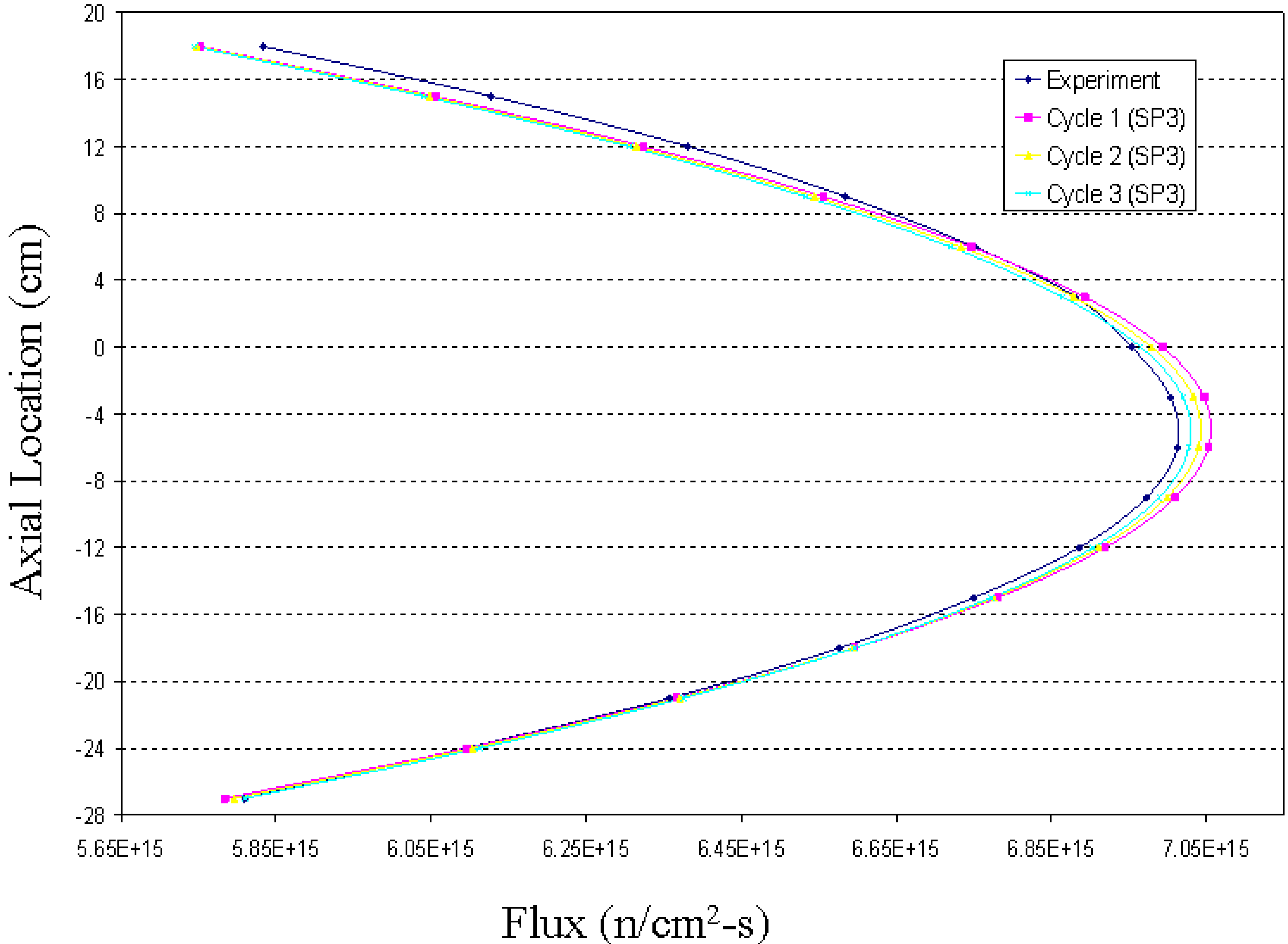
Normalized 33-group flux spectra at core midplane



Average Group Energy (eV)

Neutronics Calculation Results

Burn-up dependent axial flux profiles in irradiated pin



Calculational Scheme

- **One group cross sections were generated with MCNP and with ECCO/VARIANT using ENDF/B-VII files.**
- **In order to correctly normalize the results to the actual value of the flux (and hence eliminate the uncertainty in the total burnup), the production of Nd in the ^{235}U samples has been calculated and compared with the correspondent experimental values. Correcting factors have been obtained and applied to the values of the fluxes used in the time-dependent calculations .**
- **Time dependent calculations were subsequently performed with the NUTS code in order to obtain isotope concentrations at the end of irradiation.**

Calculational Scheme

- **In the past it was used:**

$$\sigma_{(c),A} \cdot \tau \cdot f(\tau) \cong \frac{\Delta N_{A+1}}{N_A} = \frac{N_{A+1}(\tau)}{N_A(\tau)} - \frac{N_{A+1}(0)}{N_A(0)}$$

$$f(\tau) = \left[\frac{N_{A+1}^{(\tau)}}{N_A^{(\tau)}} - \frac{N_{A+1}^{(0)}}{N_A^{(0)}} \right] \times \frac{1}{\sigma_{CA} \cdot \tau_{calc.}}$$

- **In a more accurate approach we correct the experimental density variation by a calculated quantity that takes out the variation due to all the phenomena other than the reaction rate that we are considering**:

$$\sigma_{(c),A} \cdot \tau \cdot \cong \frac{{}^{corr}\Delta N_{A+1}}{N_A} = \frac{{}^{\mathbf{exp}}\Delta N_{A+1}(\tau) - ({}^{calc}\Delta N_{A+1} - N_A^{(0)} e^{-\sigma\tau})}{N_A}$$

PROFIL 1 C/E

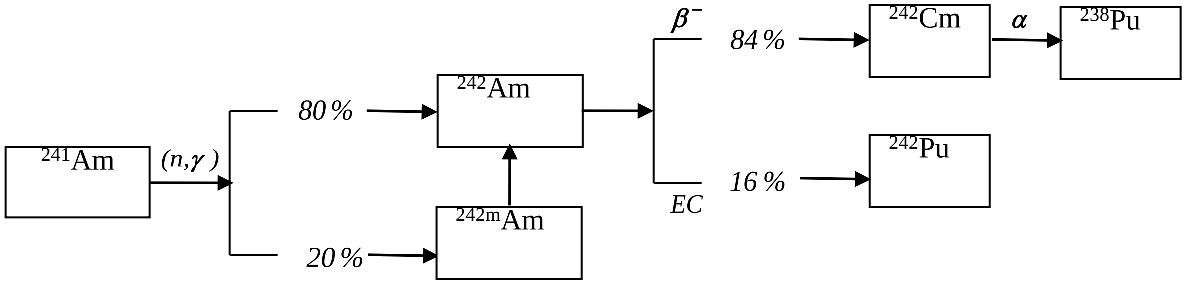
C/E's: Pu242 sample

Samples	Am243	Cm244	Pu242 σ_c	Am243 σ_c
15	1.058	0.826	1.058	0.768
32	1.044	0.933	1.045	0.890
40	1.078	0.925	1.079	0.852

C/E's: Am241 samples

Samples	Pu238	Pu242	Am242	a)Am241 σ_c	b)Am241 σ_c	c)Am241 σ_c
11	0.949	0.965	0.972	0.969	0.965	0.944
44	0.949	0.988	1.001	1.001	0.988	0.945

a) Calculated from Am242 build up. b) Calculated from Pu242 build up. c) Calculated from Pu238 build up



PROFIL-1 one-group cross sections C/E

σ	C/E		
	MCNP	Deter.	Exp. Unc.
σ_{capt} U-235	0.948	0.958	1.7 %
σ_{capt} U-238	0.972	0.967	2.3 %
σ_{capt} Pu-238	1.299	1.348	4.0 %
σ_{capt} Pu-239	0.906	0.922	3.0 %
$\sigma_{n,2n}$ Pu-239	0.745	0.663	15.0 %
σ_{capt} Pu-240	0.964	0.976	2.2 %
$\sigma_{n,2n}$ Pu-240	0.779	0.715	15.0 %
σ_{capt} Pu-241	0.950	0.956	4.1 %
σ_{capt} Pu-242	1.061	1.062	3.5 %
σ_{capt} Am-241	0.968	0.975	1.7 %
σ_{capt} Am-243	0.834	0.845	5.0 %
σ_{capt} Mo-95	1.032	1.025	3.8 %
σ_{capt} Mo-97	0.968	0.993	4.4 %
σ_{capt} Ru-101	1.101	1.124	3.6 %
σ_{capt} Pd-105	0.852	0.834	4.0 %
σ_{capt} Cs-133	0.878	0.905	4.7 %
σ_{capt} Nd-145	0.955	1.033	3.8 %
σ_{capt} Sm-149	0.915	0.924	3.1 %

TRAPU Experiment

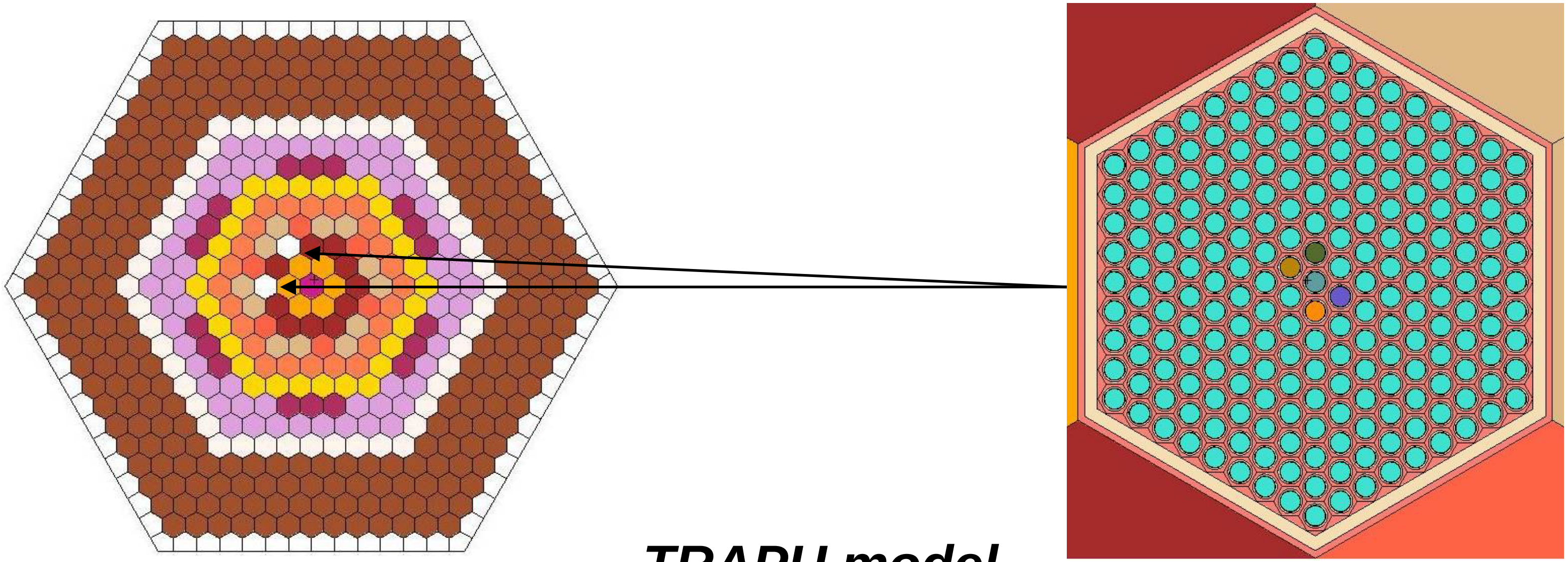
- **The TRAPU experiment consisted of a six-cycle irradiation (10th to 15th) of mixed-oxide pins that contained plutonium of different isotopic compositions but heavily charged in the higher isotopes (Pu240, Pu241 and Pu242) compared to typical PHENIX fuel. Standard pins were placed in regular PHENIX subassemblies located in the third row of the reactor. Three types of plutonium containing pins were used.**
- **After irradiation, small samples (20 mm high) were cut from the experimental pins (both fuel and clad) and put into a solution in order to determine the fuel composition by nuclide. Neodymium-148 was used as burn up indicator since it is a stable fission product with a small capture cross section, and it enables determination of the number of fission reactions that have taken place in the sample. Mass spectrometry was then used, with simple or double isotopic dilution and well-characterized tracers.**

Plutonium Isotopic Compositions of the Three TRAPU Fuel Pins

Pin	Isotope Composition [%]				
	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242
TRAPU-I	0.1	73.3	21.9	4.0	0.7
TRAPU-II	0.8	71.4	18.5	7.4	1.9
TRAPU-III	0.2	34.0	49.4	10.0	6.4

TRAPU Modeling

- ERANOS input for TRAPU developed by CEA has been converted to MCNP model.
- TRAPU has a larger sample volume than PROFIL-1&2. (make MC calculation easier).



TRAPU model

Experimental Concentrations for the TRAPU Program (U238=100).

	TRAPU-I		TRAPU-II		TRAPU-III	
Isotope	**Initial**	**Final**	**Initial**	**Final**	**Initial**	**Final**
U-234	**0.0060 ± 3.3 %**	**0.0062 ± 2.5 %**	**0.0106 ± 4.7 %**	**0.01278 ± 1.3 %**	**0.0088 ± 5.6 %**	**0.00995 ± 1.0%**
U-235	**0.7263 ± 0.3 %**	**0.4830 ± 0.3 %**	**0.7614 ± 0.3 %**	**0.4969 ± 0.2 %**	**0.7447 ± 0.3 %**	**0.4869 ± 0.2%**
U-236	**0.0015 ± 13.3 %**	**0.0664 ± 0.5 %**	**0.0025 ± 8.0**	**0.0688 ±0.4 %**	**0.0099 ± 5.0%**	**0.07801 ± 0.3%**
Np-237	**0.0001 ± (*)**	**0.0365 ± 6.8 %**	**0.0011 ± (*)**	**0.0390 ±3.3 %**	**0.0042 ± (*)**	**0.0473 ± 3.2%**
Pu-238	**0.0296 ± 1.3%**	**0.0455 ± 0.9 %**	**0.1804 ±0.5 %**	**0.2020 ±0.4 %**	**0.0852 ± 0.6%**	**0.2420 ± 0.4%**
Pu-239	**17.939 ± 0.5%**	**16.366 ± 0.4 %**	**16.780 ± 0.5 %**	**15.852 ±0.3 %**	**13.068 ± 0.5%**	**13.338 ± 0.3%**
Pu-240	**5.367 ± 0.5%**	**6.308 ± 0.4 %**	**4.359 ± 0.5 %**	**5.433 ± 0.3 %**	**19.006 ± 0.5%**	**18.197 ± 0.3%**
Pu-241	**0.9768 ± 0.5%**	**0.9449 ± 0.4 %**	**1.744 ± 0.5 %**	**1.304 ± 0.3 %**	**3.858 ± 0.5%**	**3.406 ± 0.3%**
Pu-242	**0.1744 ± 0.6%**	**0.2353 ± 0.5 %**	**0.4472 ± 0.5 %**	**0.5473 ±0.4 %**	**2.455 ± 0.5%**	**2.598 ± 0.3%**
Am-241	**0.0657 ± 2.0%**	**0.1410 ± 3.0 %**	**0.3432 ± 2.0 %**	**0.3915 ± 3.6 %**	**1.029 ± 2.1%**	**1.084 ± 2.1%**
Am-242	**-**	**0.0044 ± 3.6 %**	**-**	**0.01437 ± 4.0 %**	**-**	**0.0419 ± 12.5%**
Am-243	**-**	**0.0160 ±3.6 %**	**-**	**0.03945 ± 4.0 %**	**-**	**0.1888 ± 2.5%**
Cm-242	**-**	**0.00765 ± 2.4 %**	**-**	**0.02506 ± 2.6 %**	**-**	**0.07052 ± 2.1%**
Cm-243	**-**	**-**	**-**	**0.002475 ± 2.7 %**	**-**	**0.006985 ± 2.6%**
Cm-244	**-**	**0.00243 ± 2.4 %**	**-**	**0.005366 ± 2.2 %**	**-**	**0.02684 ± 1.7%**
Nd-148	**-**	**0.1464 ± 0.3 %**	**-**	**0.1468 ± 0.2 %**	**-**	**0.1720 ± 0.2%**

(*) This value has not been measured, but deduced from the Am-241 decay

C/E Values (preliminary) of Final Concentrations in the TRAPU Experiment Using ENDF/B-VII Data

Isotope	TRAPU-1	TRAPU-2	TRAPU-3
U-234	1.00± 3.9 %	1.02± 3.8 %	1.07± 4.6 %
U-235	1.00± 0.4%	1.02± 0.4 %	1.02± 0.4 %
U-236	0.98± 0.8 %	1.00± 1.0 %	1.00± 0.9 %
Np-237	0.94± 6.8 %	0.95± 3.3 %	0.89± 3.2 %
Pu-238	1.00± 1.5 %	0.99± 1.0 %	1.02± 1.6 %
Pu-239	1.03± 0.6 %	1.02± 0.5 %	1.02± 0.4 %
Pu-240	1.01± 0.6 %	0.99± 0.6 %	1.00± 0.6 %
Pu-241	1.01± 0.6 %	0.99± 0.6 %	1.01± 0.6 %
Pu-242	1.04± 0.8 %	1.01± 0.6 %	1.01± 0.6 %
Am-241	0.98± 3.2 %	0.99± 3.9 %	0.99± 2.6 %
Am242M	1.03± 3.8 %	1.04± 4.3 %	1.03± 3.1%
Am-243	0.98± 2.6 %	0.96± 3.1 %	1.01± 2.5 %
Cm-242	1.04± 3.9 %	1.02± 3.1 %	1.02± 2.7 %
Cm-243	-	0.49± 3.1 %	0.49± 3.2 %
Cm-244	0.85± 2.1 %	0.95± 2.3 %	0.97± 1.8 %

Sensitivity (%) to isotope build up: TRAPU-2

Basic Data	Isotope build-up						
	^{234}U	^{235}U	^{236}U	^{237}Np	^{238}Pu	^{239}Pu	^{240}Pu
$^{234}U\ \sigma_{cap}$	-10	0.2					
$^{234}U\ \sigma_{fis}$	-6						
$^{235}U\ \sigma_{cap}$		-10.8	90	9.7	0.2		
$^{235}U\ \sigma_{fis}$	-0.2	-36.2	-16.2	-1.2			
$^{236}U\ \sigma_{cap}$			-5.7	10.5	0.2		
$^{238}U\ \sigma_{cap}$				-2.3		26.7	3.9
$^{238}U\ \sigma_{fis}$				-0.4		-0.1	
$^{238}U\ \sigma_{(n,2n)}$	0.1			82.5	1.8		
$^{237}Np\ \sigma_{cap}$	0.2			-14.0	1.9		
$^{237}Np\ \sigma_{fis}$				-3.1			
$^{238}Pu\ \sigma_{cap}$	-1.1				-8.0		
$^{238}Pu\ \sigma_{fis}$	-2.1				-16.9		
$^{239}Pu\ \sigma_{cap}$			0.2			-8.1	26.2
$^{239}Pu\ \sigma fis$						-29.4	-4.3
$^{240}Pu\ \sigma_{cap}$			-0.1	0.1	0.4		-9.3
$^{240}Pu\ \sigma_{fis}$			-0.1				-6.7
$^{241}Pu\ \sigma_{cap}$					-0.1		
$^{241}Pu\ \sigma_{fis}$				-0.1	-0.6		
$^{241}Am\ \sigma_{cap}$	2.7			-0.7	26.1		

Sensitivity (%) to isotope build up: TRAPU-2

Basic Data	Isotope build-up							
	^{241}Pu	^{242}Pu	^{241}Am	^{242}Am	^{243}Am	^{242}Cm	^{243}Cm	^{244}Cm
^{238}U σ_{cap}	0.6							
^{239}Pu σ_{cap}	5.7	0.4	0.6	0.1	0.2	0.2		
^{239}Pu σfis	-0.7		-0.2					
^{240}Pu σ_{cap}	30.5	3.5	4.4	1.5	1.7	2.0	1.2	0.8
^{240}Pu σ_{fis}	-1.2	-0.1	-0.1					
^{241}Pu σ_{cap}	-8.8	24.2	-1.4	-0.5	15.5	-0.7	-0.4	10.7
^{241}Pu σ_{fis}	-40.7	-5.4	-6.7	-2.4	-2.5	-3.2	-1.8	-1.2
^{242}Pu σ_{cap}		-7.7			93.8		-0.1	95.4
^{242}Pu σ_{fis}		-4.6			-2.4			-1.8
^{241}Am σ_{cap}		2.8	-30.5	82.1	3.3	77.7	85.4	2.8
^{241}Am σ_{fis}			-4.7	-2.8	-0.1	-3.4	-2.2	
^{242}Am σ_{fis}				-27.1	-0.3			-0.2
^{243}Am σ_{cap}					-15.2		0.2	89.7
^{243}Am σ_{fis}					-1.8			-1.4
^{242}Cm σ_{cap}						-3.1	97.7	0.5
^{242}Cm σ_{fis}						-3.7	-2.6	
^{243}Cm σ_{fis}							-20.6	

Conclusions

- **Irradiation experiments are extremely useful for validating data related to minor actinides and fission products.**
- **The analysis of the PROFIL and TRAPU experiments has indicated that ENDF/B-VII data files need significant improvements in the fast spectrum range for capture cross sections of: Pu-238, Pu-239, Am-243, Cm-242, Ru-101, Pd-105, and Cs-133. Indications also for major improvement of (n,2n) cross sections of U-238, Pu-239, and Pu-240 have been also obtained.**
- **The PROFIL and TRAPU experiments are clean and relatively well documented; however, because they have been performed long time ago (more than 30 years), some information is missing (control rod position history, some experimental uncertainty, etc.). Moreover, some MA isotopes were not irradiated**
- **A new campaign of irradiation experiment is planned: MANTRA (Measurement of Actinide Neutron Transmutation Rates with Accelerator mass spectrometry). Information on transmutation rate up to Cf will be possible with irradiation at ATR and analysis at ATLAS. First experimental results will be available in late 2012.**