

MANTRA A NUCLEAR DATA EXPERIMENT

Presented by Gilles Youinou
NSUF Annual Meeting
March 1-3, 2016



Idaho National Laboratory



The Team

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- ❑ ATR Irradiation: B. Horkley
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Background

Two Complementary Sources of Information for Nuclear Data Evaluators:

1) Differential measurements

→ neutron source = particle accelerator

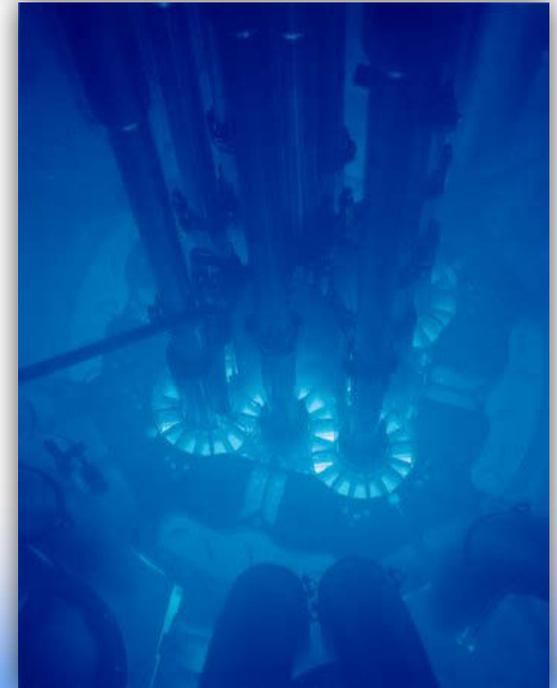
→ outcome = $\sigma(E)$

2) Integral measurements

→ neutron source = nuclear reactor

→ outcome = $\bar{\sigma} = \int_0^{\infty} \sigma(E) \varphi(E) dE = \underline{\text{effective cross section}}$

Even though theoretical progress has been made as far as the predictability of neutron cross-section models, **measurements are still indispensable** to meet tight design requirements for reduced uncertainties.

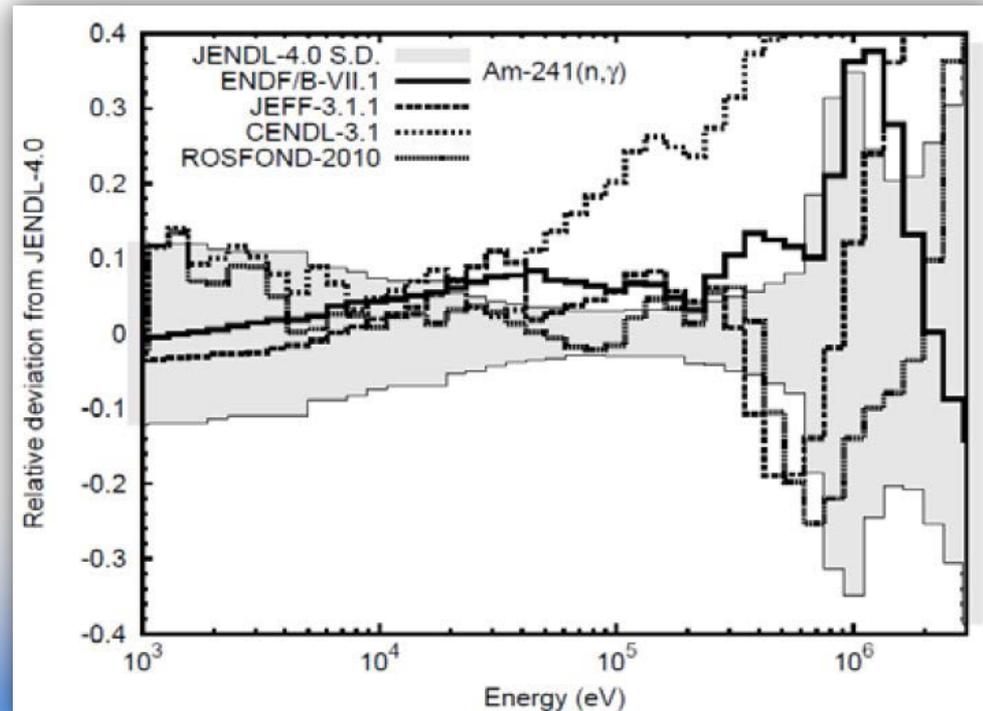
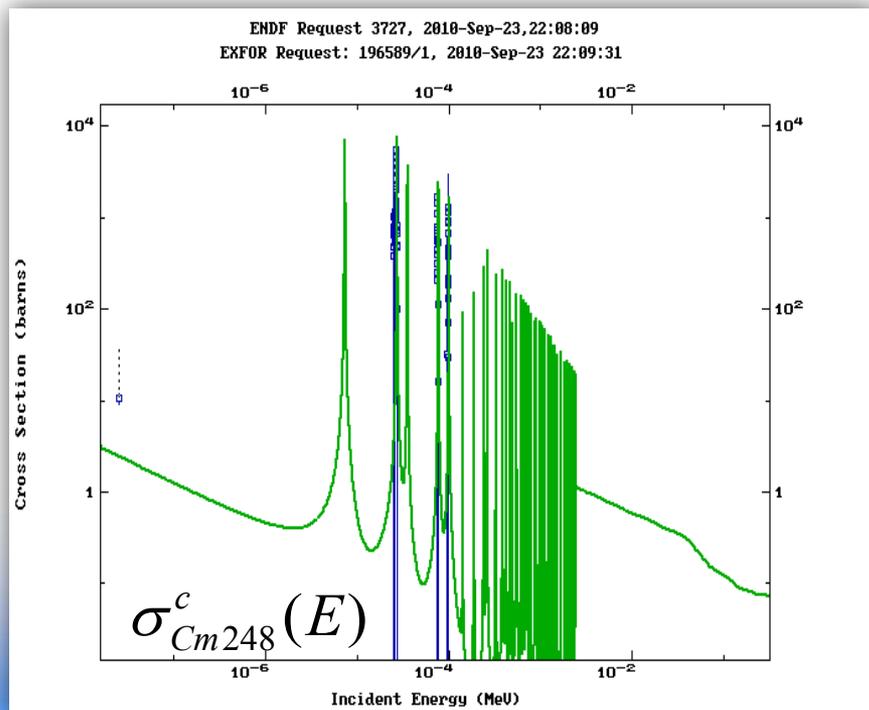


Objective of the MANTRA experiment:

Infer effective neutron capture cross-sections of most actinides of interest for reactor physics in fast and epithermal spectra (a few fission products are also considered)

Why such measurements?

- ❑ Significant improvement in computer performances as well as in modeling and simulation (NEAMS, CASL, etc...); Monte Carlo is becoming more and more mainstream
- ❑ The most powerful computer coupled to the best code in the world is only as accurate as the data (nuclear or others) it uses
- ❑ For example, there are differences of 10 to 30% between Am-241 capture cross sections from different nuclear libraries. Similar situation for Np-237 and Am-243. Even larger deviation for other actinides.
- ❑ Scarce measurements available for some isotopes, e.g. Cm-248



Principle of the experiment:

1 – Irradiate very pure material (a few mg) in well-characterized neutron spectra in the Advanced Test Reactor (ATR) at INL

Major Actinides: Th-232, U-233, U-235, U-238, Pu-239, Pu-240, Pu-242,

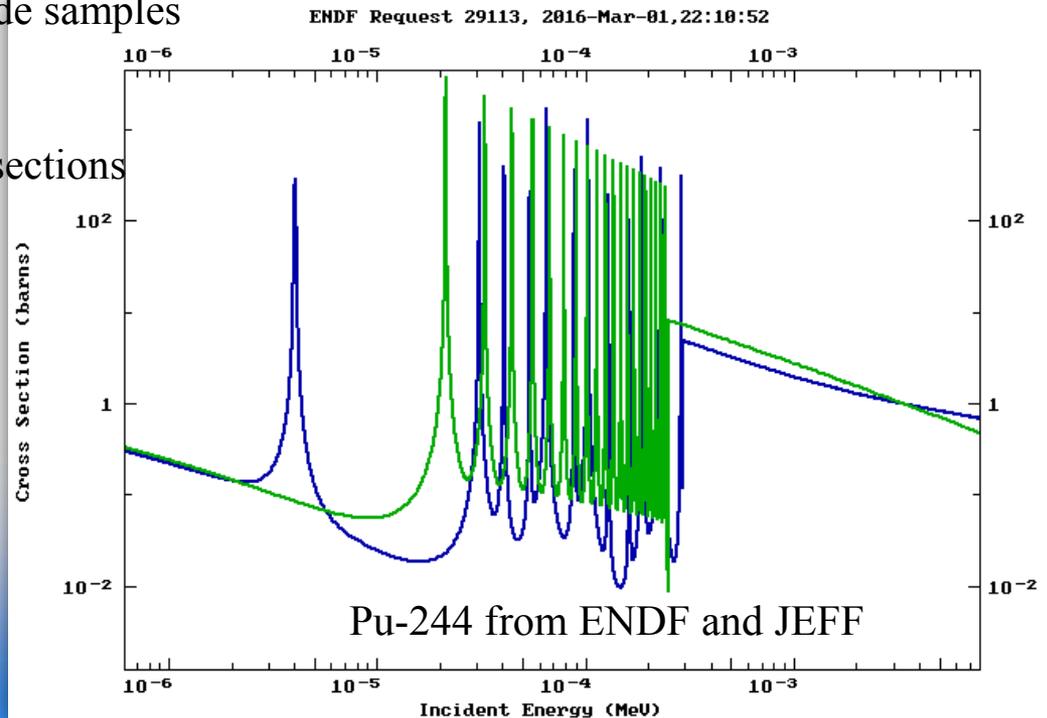
Minor Actinides: U-236, Np-237, *Pu-244*, Am-241, Am-243, Cm-244, Cm-248

Fission Products: Sm-149, Eu-153, Cs-133, Rh-103, Nd-143, Nd-145, Pd-105, Ru-101

2 – Determine the atom densities of the transmutation products using two independent sets of measurements → Multi-Collector Inductively Coupled Plasma Mass Spectrometry (MC-ICPMS)

3 – Measure the neutron fluence in the actinide samples
→ Nd-148 and Cs-137 in U-235 monitors

4 – Infer the effective neutron capture cross-sections
from results of step 2 and 3



When the neutron fluence is limited, the relation between measured and inferred quantities in a 100% pure sample of mass number A can be expressed to a good approximation as:

$$N_A(t) = N_A(0)[1 - \tilde{\sigma}_A^a \Gamma]$$

$$N_{A+n}(t) = \frac{1}{n!} N_A(0) \bar{\sigma}_A^c \bar{\sigma}_{A+1}^c \cdots \bar{\sigma}_{A+n-1}^c \Gamma^n$$

As long as the neutron fluence is small (less than 10^{21} n/cm²), only the captures matter and not absorptions

Very good approximation in most cases we are interested in. The correction factors will be taken into account in the actual analyses.

$$N_{A+1}(t) = N_A(0) \bar{\sigma}_A^c \Gamma$$

$$N_{A+2}(t) = \frac{1}{2} N_A(0) \bar{\sigma}_A^c \bar{\sigma}_{A+1}^c \Gamma^2$$

$$N_{A+3}(t) = \frac{1}{6} N_A(0) \bar{\sigma}_A^c \bar{\sigma}_{A+1}^c \bar{\sigma}_{A+2}^c \Gamma^3$$

□ inferred quantities $\rightarrow \bar{\sigma}_{A+n}^c = \int \sigma_{A+n}^c(E) dE$

□ measured quantities $\rightarrow N_{A+1} - N_{A+2} - N_{A+3} \dots$ and fluence $\Gamma = \int \varphi(E, t) dE dt$

Relation between measured and inferred quantities

Example of a very pure sample containing essentially an isotope of mass number A, but also a few tenths of a percent of impurities (let's say A+1, A+2 and A+3).

$$N_{A+1}(t) = N_A(0)\bar{\sigma}_A^c\Gamma + N_{A+1}(0)[1 - \tilde{\sigma}_{A+1}^a\Gamma]$$

$$N_{A+2}(t) = \underbrace{\frac{1}{2}N_A(0)\bar{\sigma}_A^c\bar{\sigma}_{A+1}^c\Gamma^2}_{\text{Contribution from A}} + \underbrace{N_{A+1}(0)\bar{\sigma}_{A+1}^c\Gamma}_{\text{Contribution from A+1}} + \underbrace{N_{A+2}(0)[1 - \tilde{\sigma}_{A+2}^a\Gamma]}_{\text{Left-over of the initial A+2}}$$

$$\left\{ \begin{array}{l} \bar{\sigma}_A^c = \frac{N_{A+1}^r(T) - N_{A+1}^r(0)}{\Gamma} \\ \bar{\sigma}_{A+1}^c = 2 \frac{N_{A+2}^r(T) - N_{A+2}^r(0)}{\Gamma \{N_{A+1}^r(T) + N_{A+1}^r(0)\}} \\ \bar{\sigma}_{A+2}^c = 3 \frac{\{N_{A+3}^r(T) - N_{A+3}^r(0)\}\{N_{A+1}^r(T) + N_{A+1}^r(0)\}}{\Gamma \{N_{A+1}^r(T)U_{A+2}(T) + N_{A+1}^r(0)V_{A+2}(T)\}} \end{array} \right.$$

$$U_{A+2}(T) = N_{A+2}^r(T) + 2N_{A+2}^r(0)$$

$$V_{A+2}(T) = N_{A+2}^r(T) + 2N_{A+2}^r(0)$$

Relation between measured and inferred quantities

If $N_{A+n}(0) \ll N_{A+n}(T)$ the expressions are simpler and the inference is less dependant on the precision of the measurement of the atom densities. Relatively easy with thermal or epithermal neutron spectra ($\bar{\sigma}^c \sim \text{few } 10 \text{ to few } 100 \text{ barns}$). More difficult with fast neutron spectra ($\bar{\sigma}^c \sim 1 \text{ barn}$)

→ High neutron fluence

→ High purity samples

$$\bar{\sigma}_A^c = \frac{N_{A+1}^r(T) - N_{A+1}^r(0)}{\Gamma}$$



$$\bar{\sigma}_A^c = \frac{N_{A+1}^r(T)}{\Gamma N_A^r(T)}$$

$$\bar{\sigma}_{A+1}^c = 2 \frac{N_{A+2}^r(T) - N_{A+2}^r(0)}{\Gamma \{N_{A+1}^r(T) + N_{A+1}^r(0)\}}$$



$$\bar{\sigma}_{A+1}^c = 2 \frac{N_{A+2}^r(T)}{\Gamma N_{A+1}^r(T)}$$

$$\bar{\sigma}_{A+2}^c = 3 \frac{\{N_{A+3}^r(T) - N_{A+3}^r(0)\} \{N_{A+1}^r(T) + N_{A+1}^r(0)\}}{\Gamma \{N_{A+1}^r(T) U_{A+2}(T) + N_{A+1}^r(0) V_{A+2}(T)\}}$$



$$\bar{\sigma}_{A+2}^c = 3 \frac{N_{A+3}^r(T)}{\Gamma N_{A+2}^r(T)}$$

$$U_{A+2}(T) = N_{A+2}^r(T) + 2N_{A+2}^r(0) \quad V_{A+2}(T) = N_{A+2}^r(T) + 2N_{A+2}^r(0)$$

Initial isotopic compositions of the actinide samples (MC-ICPMS measurements)

U-233

Isotope Ratio	Ratio $\pm 1\sigma$
233/233	1.00E+00
234/233	(1.7383 \pm .0015) E-03
235/233	(6.5696 \pm 0.0083) E-04
236/233	(2.1510 \pm 0.0060) E-04
237/233	(7.2262 \pm 0.57) E-06
238/233	(2.8535 \pm 0.0072) E-03

U-235

Isotope Ratio	Ratio $\pm 1\sigma$
235/235	1.00E+00
236/235	(5.9100 \pm 0.012) E-04
237/235	< 6.7E-07
238/235	(8.4271 \pm 0.028) E-04

U-236

Isotope Ratio	Ratio $\pm U$
236/236	1.00E+00
237/236	< 7.40E-06
238/236	(1.4964 \pm .0015) E-02
239/236	(7.6 \pm 3.1) E-06
240/236	(8.3 \pm 3.3) E-06

Np-237

Isotope Ratio	Ratio $\pm U$
237/237	---
238/237	(5.455 \pm 0.014) E-05
239/237	(2.882 \pm 0.200) E-05
240/237	< 6.00 E-07
241/237	< 6.10 E-07

Pu-240

Isotope Ratio	Ratio $\pm U$
240/240	---
241/240	(3.2415 \pm 0.0061) E-05
242/240	(2.9433 \pm 0.0180) E-04
243/240	< 3.10E-07
244/240	< 6.60E-07

Pu-242

Isotope Ratio	Ratio $\pm U$
242/242	1.00E+00
243/242	(1.7824 \pm 0.0051) E-05
244/242	(2.2224 \pm 0.0690) E-05
245/242	< 7.60E-07
246/242	< 8.6 E-07

Am-241

Isotope Ratio	Ratio $\pm U$
238/241	(3.128 \pm 0.056) E-04
239/241	(3.45 \pm 0.65) E-05
240/241	< 3.8 E-06
241/241	1.00E+00
242/241	(3.601 \pm 0.047) E-05
243/241	(1.82 \pm 0.43) E-05
244/241	< 4.1 E-06

Am-243

Isotope Ratio	Ratio $\pm U$
243/243	1.00E+00
244/243	(4.9261 \pm 0.0180) E-05
245/243	< 7.0 E-06
246/243	< 6.1 E-06

Cm-248

Isotope Ratio	Ratio $\pm U$
248/248	1.00E+00
249/248	(3.1546 \pm 0.018) E-05
250/248	< 5.6 E-06
251/248	< 4.8 E-06
252/248	< 4.8 E-06

Measuring the neutron fluence using U-235 wires

The neutron fluence can be determined experimentally by measuring the amount of fission products such as Nd148 or Cs137 formed during irradiation

$$\bar{\sigma}_A^c = \frac{N_{A+1}^r(T) - N_{A+1}^r(0)}{\Gamma}$$

- ❑ it is not volatile and has no volatile precursors,
- ❑ it is stable or has a relatively long half-life,
- ❑ it has a low destruction cross-section and formation from adjacent mass chains can be corrected for,
- ❑ its fission yield is nearly the same for U-235 and Pu-239 and is essentially independent of neutron energy.

$$N_{Nd8}(T) = \gamma_{Nd8} \bar{\sigma}_{U5}^f \int_0^T N_{U5}(t) \phi(t) dt = \gamma_{Nd8} \bar{\sigma}_{U5}^f N_{U5}(0) \int_0^T e^{-\bar{\sigma}_{U5}^a \int_0^t \phi(x) dx} \phi(t) dt$$

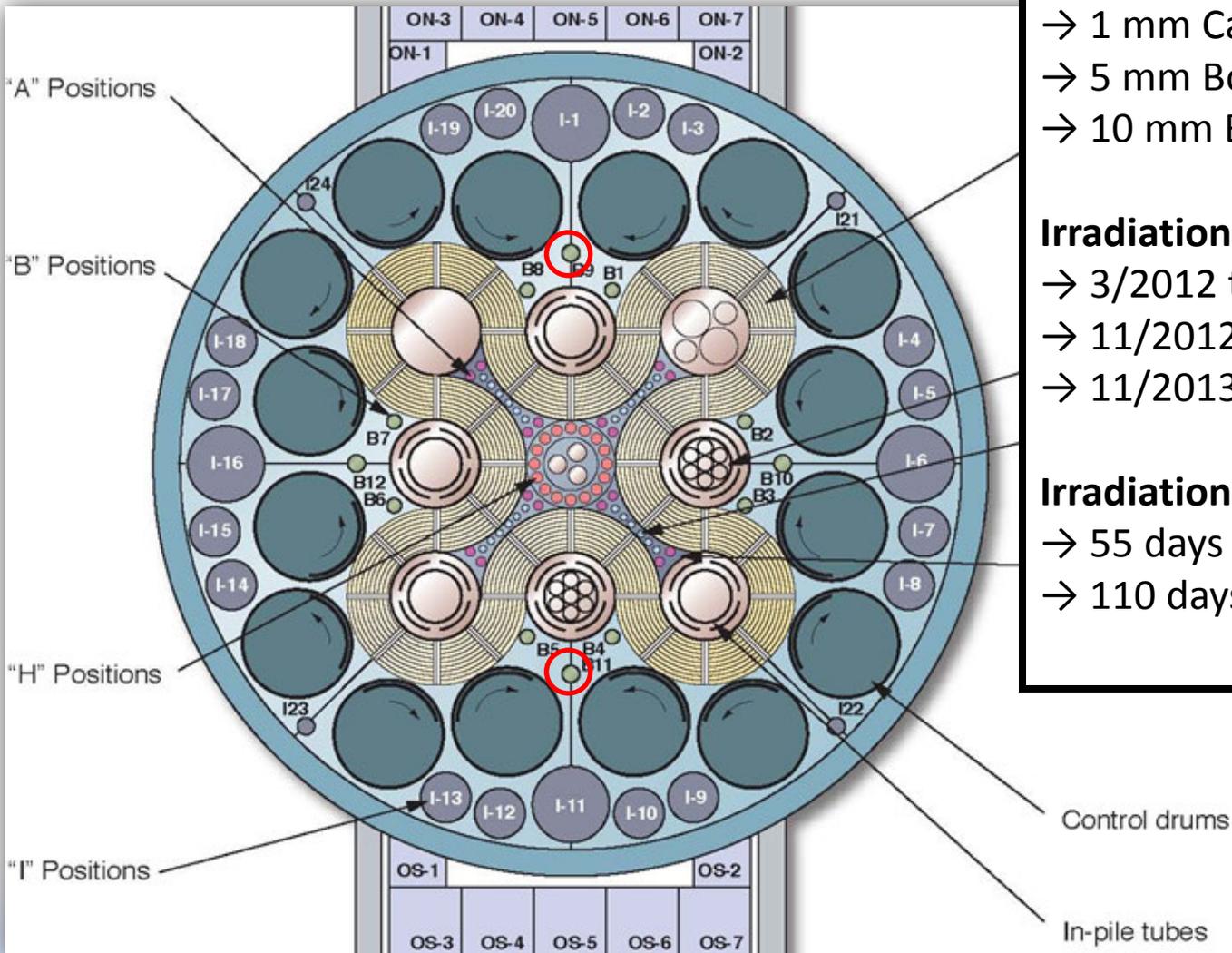
$$\bar{\phi T} \sim \left[\frac{N_{Nd8}(T)}{N_{U5}(T)} \right]_m \frac{1}{\gamma_{Nd8} \bar{\sigma}_{U5}^f}$$

Multi-Collector Inductively Coupled Plasma Mass Spectrometry (MC-ICPMS)

- ❑ Brought online in MFC's Analytical Lab in April 2013
- ❑ Leap above industry standard for accurate measurements of isotopic ratios, which relied on a technology called a Thermal Ionization Mass Spectrometer
- ❑ Provides a faster and more accurate method to determine how much of which elements and isotopes are present in any given sample.
- ❑ Much faster turnaround times for MC-ICPMS for a given sample (few hours) than for TIMS.



Samples irradiation in ATR



Two Positions: B9 and B11

Three Neutron Filters:

- 1 mm Cadmium
- 5 mm Boron (70% ^{10}B)
- 10 mm Boron (70% ^{10}B)

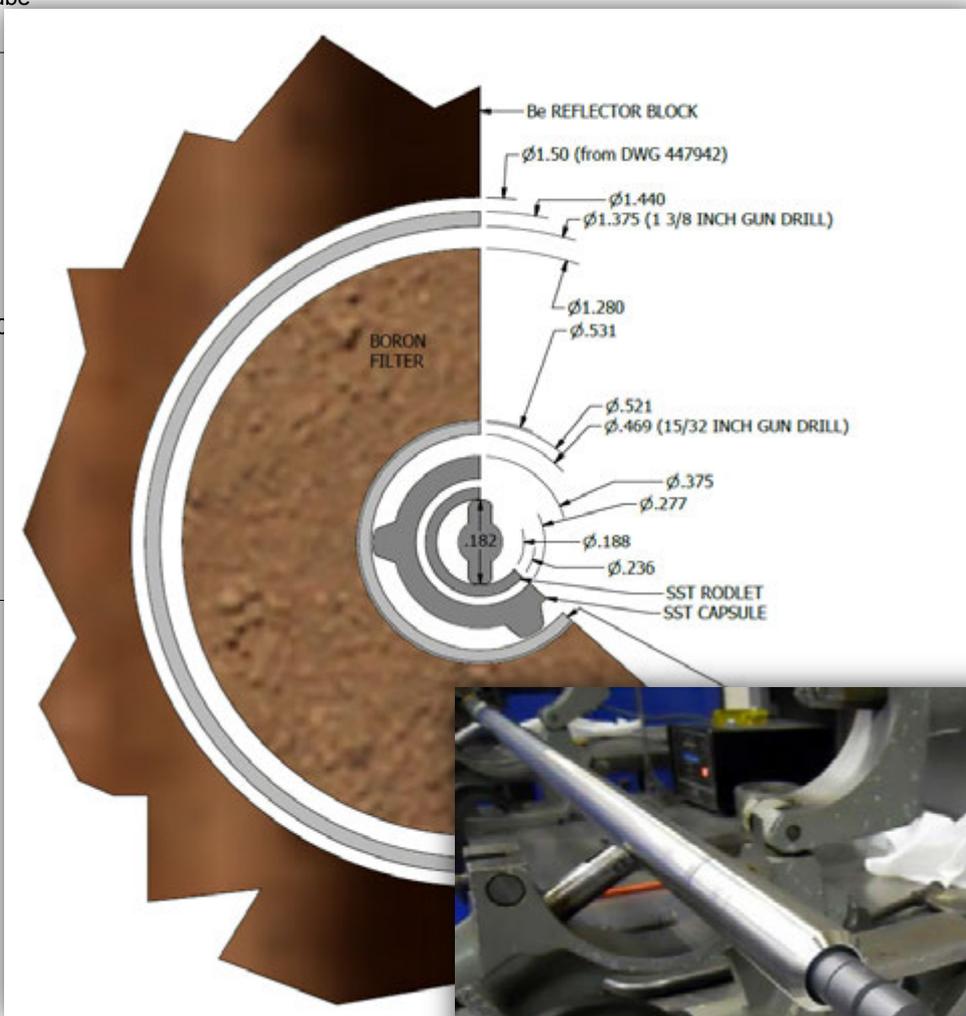
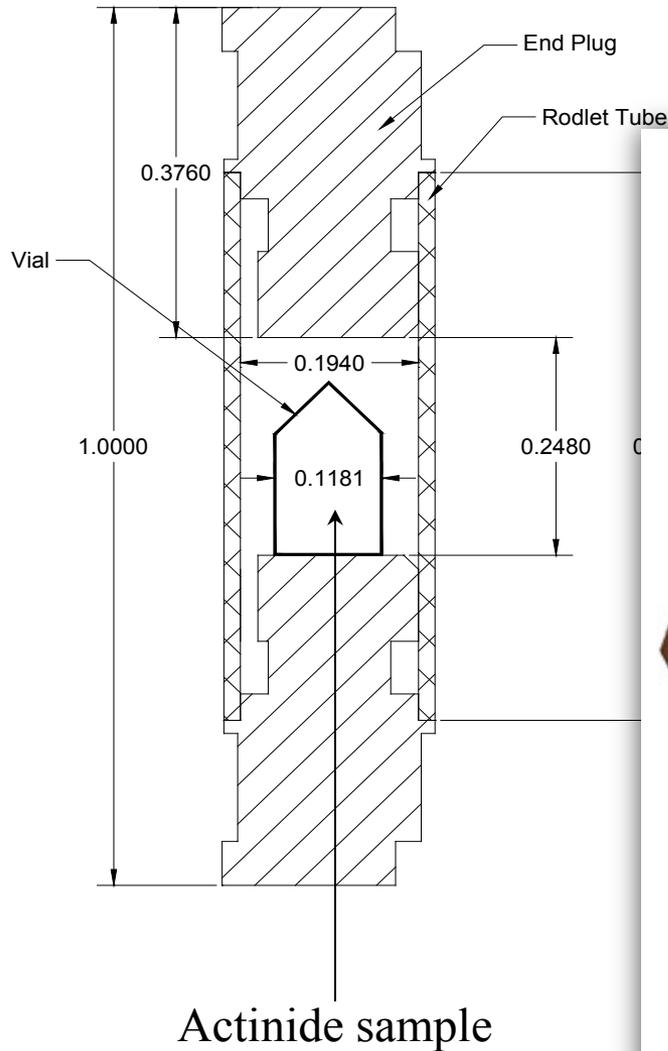
Irradiation in ATR:

- 3/2012 to 1/2013 for 5-mm B
- 11/2012 to 1/2013 for Cd
- 11/2013 to 1/2014 for 10-mm B

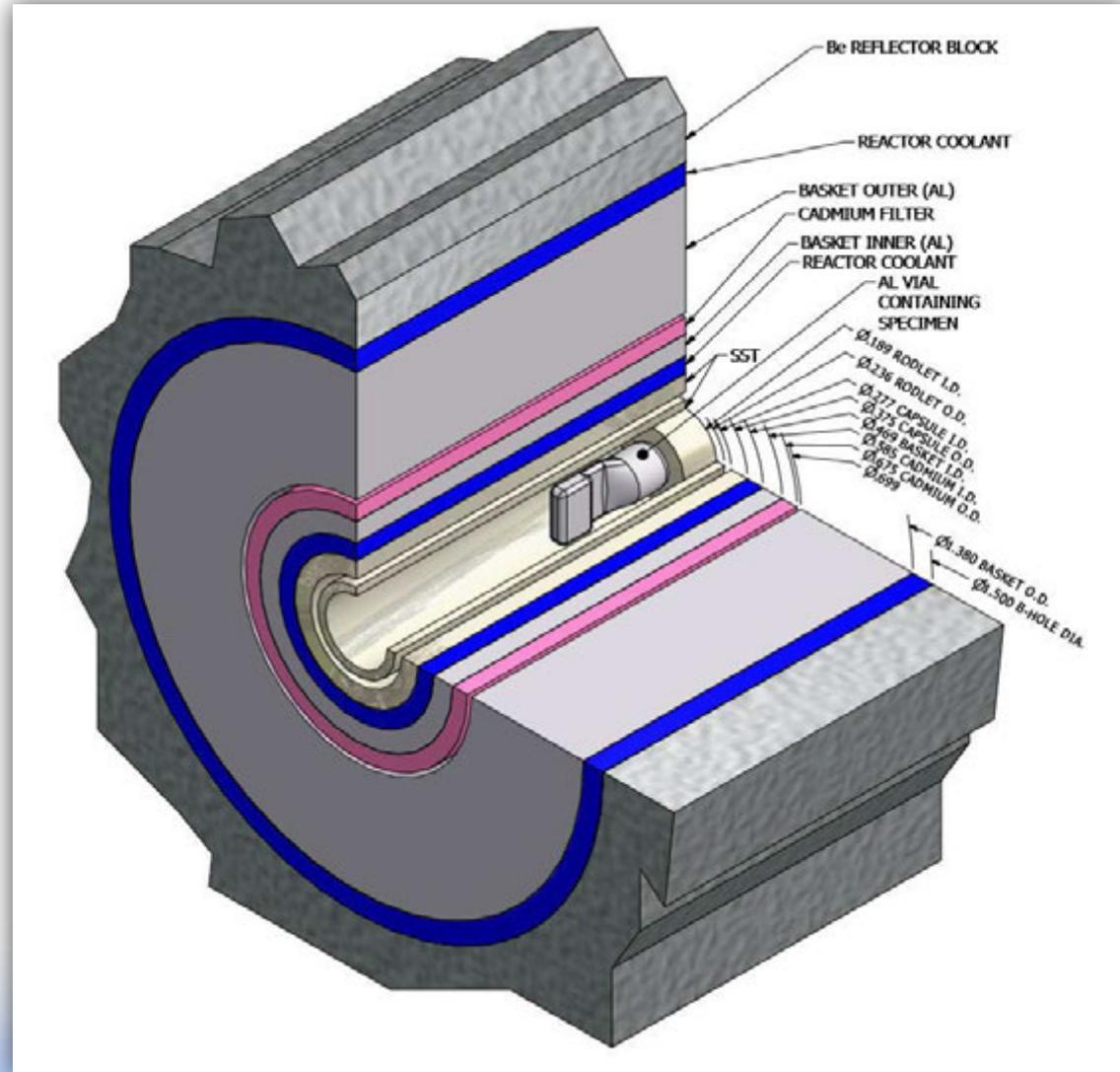
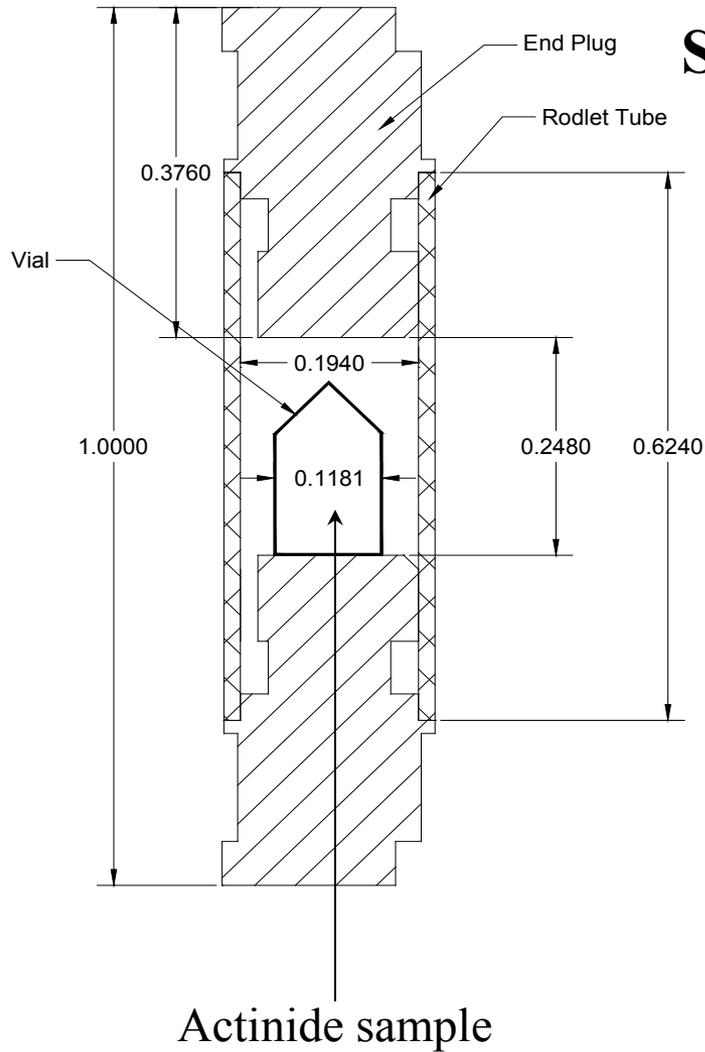
Irradiation Time:

- 55 days for Cd-filtered
- 110 days for B-filtered

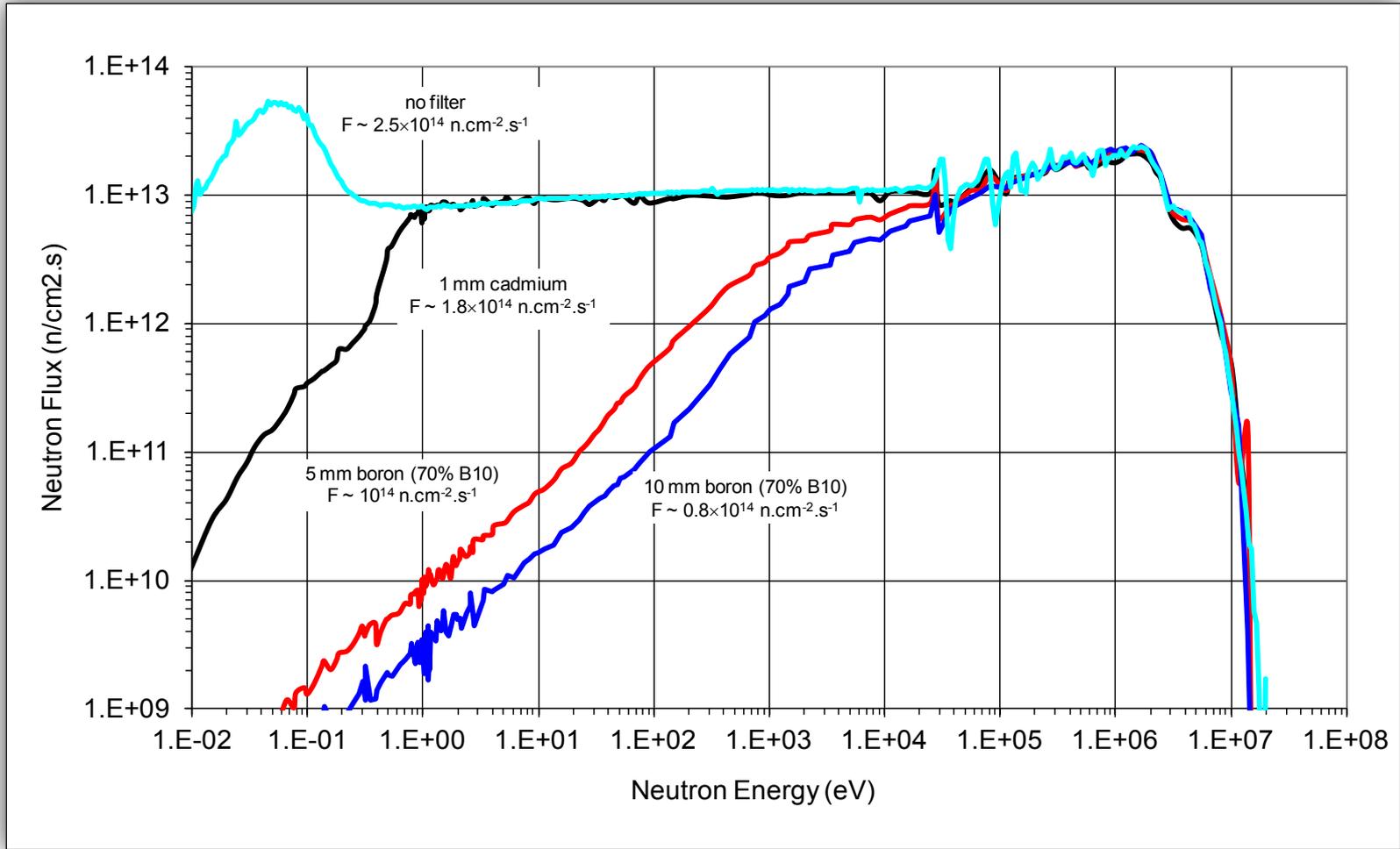
Samples irradiation in ATR: Boron filter



Samples irradiation in ATR: Cadmium filter



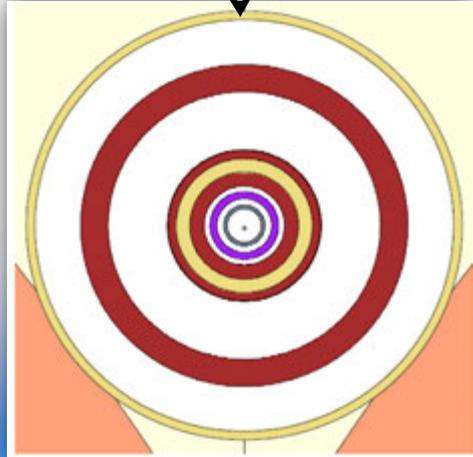
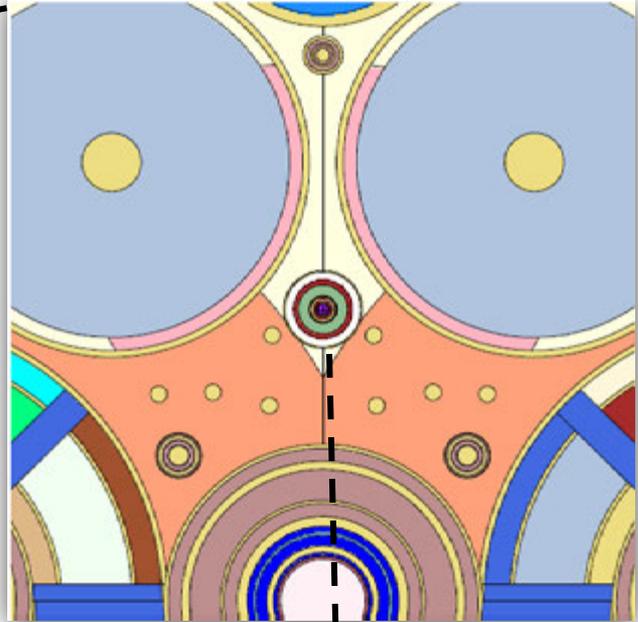
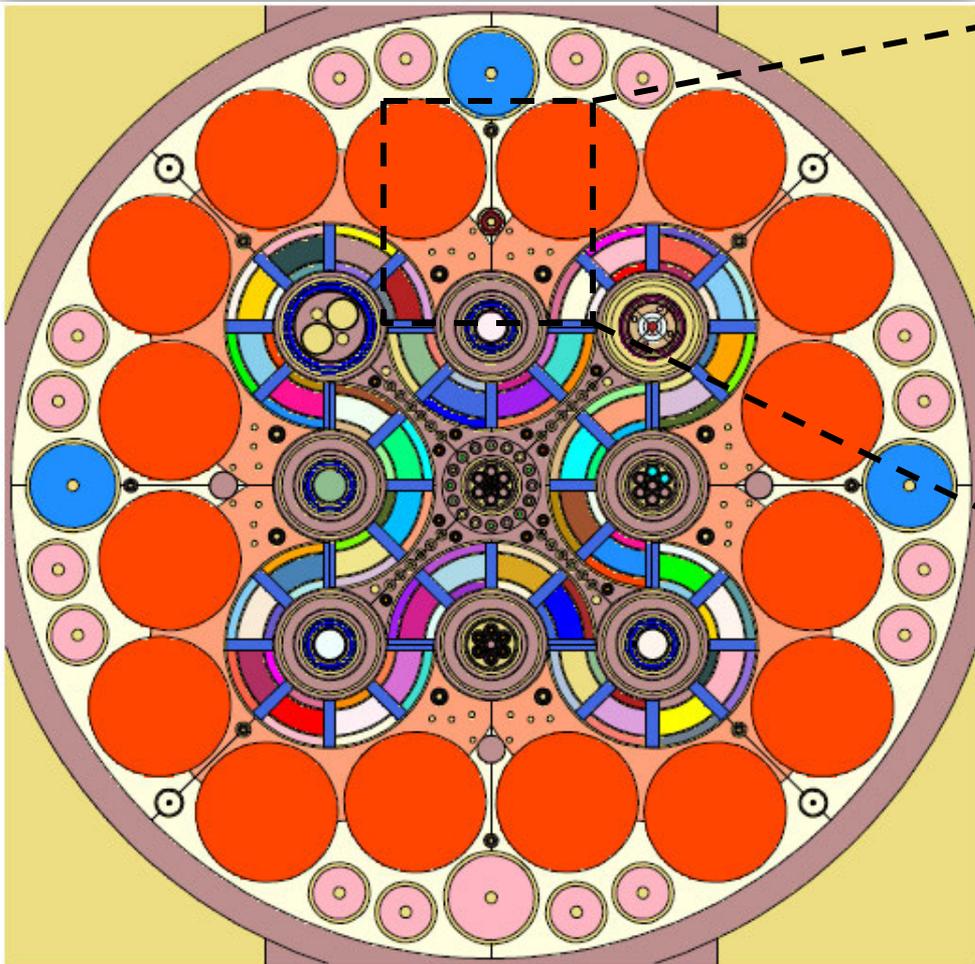
Effect of neutron filters on the neutron spectrum in the samples



σ_{U238}^c depends strongly on $\varphi(E)$ $\bar{\sigma}_{U238}^c = 14 \text{ barns}$ with Cd filter and 0.6 barn with 5-mm B filter

❑ Detailed ATR as-run MCNP calculations to determine the irradiated sample compositions and compare with experiment

❑ 100's of Billions of particles necessary to obtain good statistical convergence



Use of the Rapid Turnaround Experiment Opportunity

❑ Investigation of the self-shielding effect on the effective cross section made possible by MC-ICPMS precision and accuracy (half a percent down to relative concentrations of about 10^{-5})

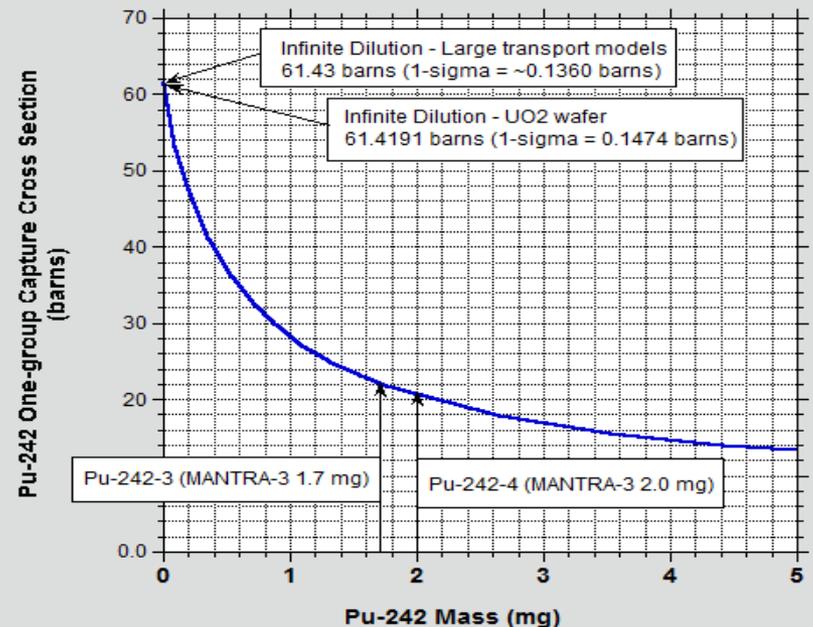
❑ Example: in a Pu240 sample, Pu240 is A, whereas in a Pu239 sample it is A+1 and in a U238 sample it is A+2

➤ In Pu240 sample, Pu240 capture cross section is determined as $\bar{\sigma}_A^c = \frac{N_{A+1}^r(T)}{\Gamma N_A^r(T)}$

➤ In Pu239 sample, Pu240 capture cross section is determined as $\bar{\sigma}_{A+1}^c = 2 \frac{N_{A+2}^r(T)}{\Gamma N_{A+1}^r(T)}$

Pu240 capture cross section may be self-shielded the Pu240 and significantly less or not at all and U238 samples

Figure 3. MANTRA-3 Pu-242 capture cross section (barns) versus Pu-242 sample mass. [PuO2 wafer at TD density]



Preliminary results for some cadmium filtered samples

Isotope	Sample Position	Type	σ_g C/E	Experimental Uncertainty	C/E Uncertainty
^{241}Am	15	A	0.84	0.52%	2.51%
	27	A	0.83	0.52%	2.47%
^{242}Pu	9	A	1.01	0.70%	4.87%
	25	A	0.95	0.70%	4.87%
^{243}Am	9	A+1	0.89	0.19%	2.73%
	25	A+1	0.91	0.19%	2.73%
	5	A	1.00	0.52%	3.12%
	21	A	0.84	0.52%	3.76%
^{244}Pu	10	A	2.74	0.67%	3.40%
	17	A	2.91	0.65%	3.41%
^{244}Cm	5	A+1	1.09	0.92%	4.22%
	21	A+1	1.03	0.78%	4.36%
	12	A	1.11	5.09%	6.86%
	22	A	1.12	5.09%	6.85%
^{248}Cm	8	A	0.98	0.39%	4.23%
	18	A	0.92	0.40%	4.20%
^{237}Np	3	A	0.93	0.22%	2.41%
	16	A	0.97	0.23%	2.42%

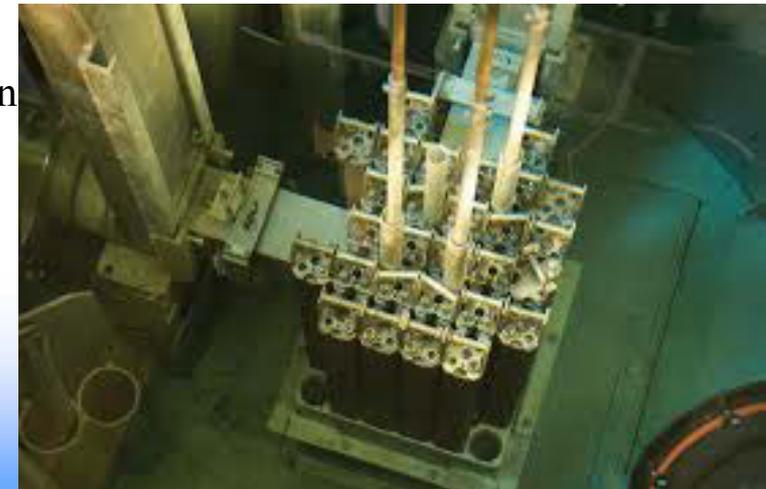
International exposure

- ❑ MANTRA has been presented several times to the OECD/NEA Expert Group on Integral Experiments for Minor Actinide Management
- ❑ Mentioned several times in 2015 NEA report “Review of Integral Experiments for Minor Actinide Management”
- ❑ Potential collaboration was discussed and “the participants agreed that the MANTRA-2 campaign will be one of the key components of the international collaboration proposed by this EG.”; This will be discussed in more detail at the NEA Nuclear Science Committee meeting in June 2016
- ❑ Presented at the International Nuclear Data Conference in 2013; 2-3 articles will be submitted in 2016 in Nuclear Science & Engineering peer-review journal



New follow up ideas

- ❑ Complementary measurements in NRAD. Reactivity oscillation measurements in a multi-spectra environment.
- ❑ New capability for NSUF (not available elsewhere in the US; only a few in the world)



Backup slides

Principle of a sample irradiation experiment

- As an isotope is irradiated in a neutron field, new isotopes are created
- The relation linking these new isotopes is given by the Bateman equations

$$\frac{dN_{238}^{92}(\vec{r}_s, t)}{dt} = -N_{238}^{92}(\vec{r}_s, t) \tilde{\sigma}_{238,92}^a(\vec{r}_s, t) \phi(\vec{r}_s, t)$$

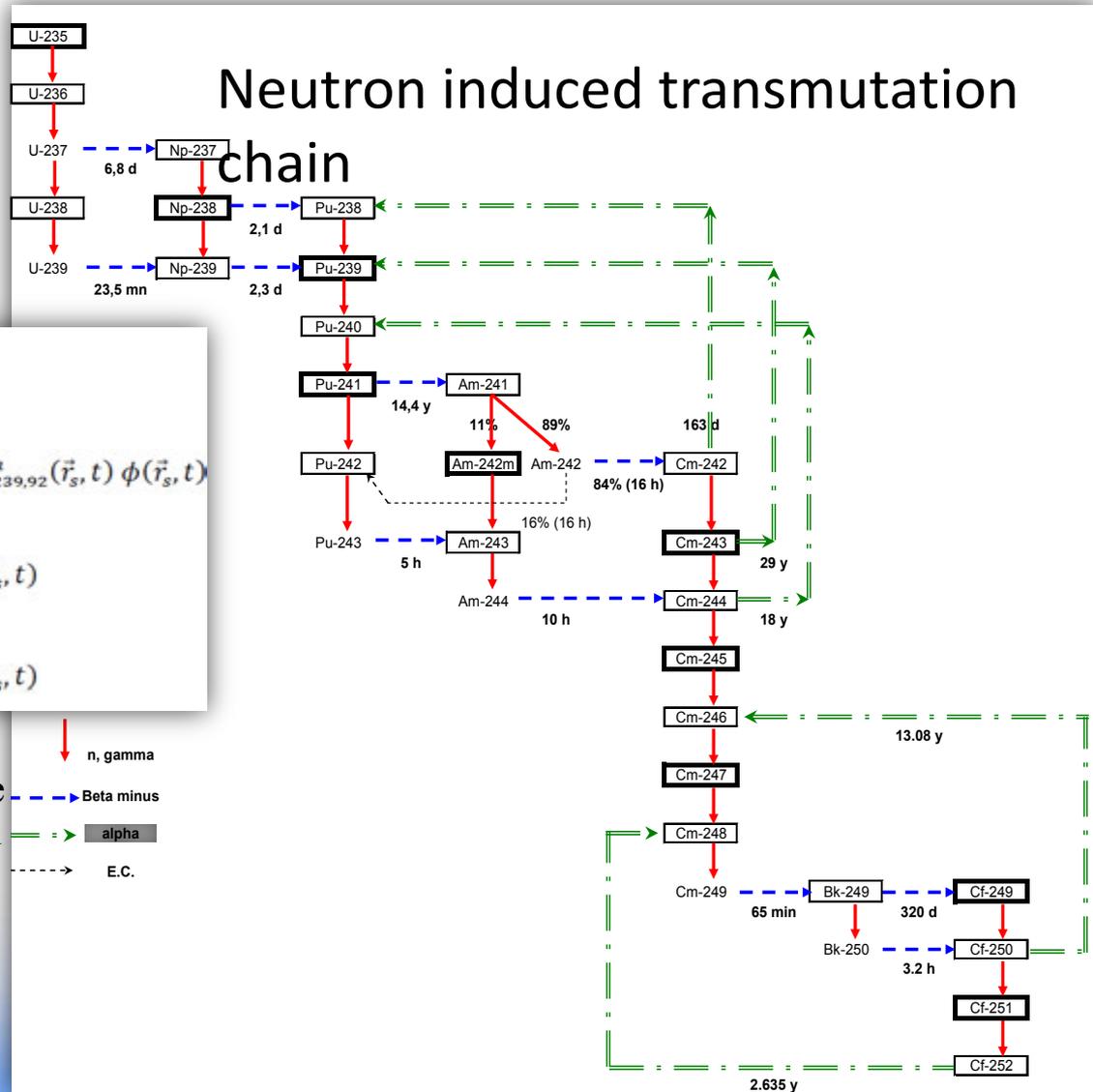
$$\frac{dN_{239}^{92}(\vec{r}_s, t)}{dt} = +N_{238}^{92}(\vec{r}_s, t) \tilde{\sigma}_{238,92}^c(\vec{r}_s, t) \phi(\vec{r}_s, t) - N_{239}^{92}(\vec{r}_s, t) \tilde{\sigma}_{239,92}^a(\vec{r}_s, t) \phi(\vec{r}_s, t)$$

$$\frac{dN_{239}^{93}(\vec{r}_s, t)}{dt} = +\lambda_{239,92}^\beta N_{239}^{92}(\vec{r}_s, t) - N_{239}^{93}(\vec{r}_s, t) \tilde{\sigma}_{239,93}^a(\vec{r}_s, t) \phi(\vec{r}_s, t)$$

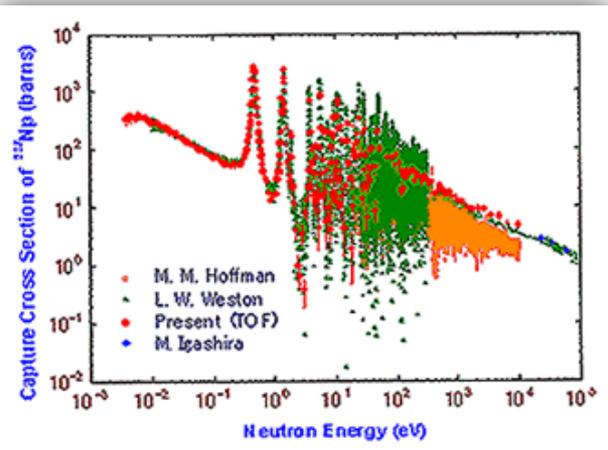
$$\frac{dN_{239}^{94}(\vec{r}_s, t)}{dt} = +\lambda_{239,93}^\beta N_{239}^{93}(\vec{r}_s, t) - N_{239}^{94}(\vec{r}_s, t) \tilde{\sigma}_{239,94}^a(\vec{r}_s, t) \phi(\vec{r}_s, t)$$

- Measurement of the atom densities before and after irradiation allows inferring neutron capture cross sections

Neutron induced transmutation chain



Using differential and integral measurements to improve nuclear data libraries



Differential measurements



Nuclear Data Evaluators

Modify ENDF data library & uncertainties if consistent



Reactor Physicists



Analyses of C/E from **integral experiments**
Sensitivity and uncertainty quantification



Suggest modifications to nuclear data & uncertainties



□ Iterative process between reactor physicists and nuclear data evaluators ends when both sides are satisfied with the end result, i.e. an improved nuclear data library.

Sm149	1
Np237	2
flux wires	3
Am243	4
U233	5
U235	6
Cm248	7
Pu242	8
Pu244	9
flux wires	10
Pu240	11
Th232	12
Am241	13
Np237	14
Pu244	15
Cm248	16
flux wires	17
U238	18
Am243	19
Pu239	20
U236	21
Pu242	22
flux wires	23
Am241	24
Eu-Cs-Rh	25

Loading plan for thin-boron-filtered samples (same for thick-boron filtered samples)

21 capsules for samples: 19 for actinides + 2 for FP

Th-232, U-233, U-235, U-236, U-238, Np-237 (2), Pu-239, Pu-240, Pu-242 (2), Pu-244 (2), Am-241 (2), Am-243 (2) and Cm-248 (2)

NB: Cm-244 will be used only for cadmium-filtered irradiation

Sm-149, Eu-153, Cs-133, Rh-103

4 extra-capsules for U-235 wires to measure the total fluence + spectrum wires

Loading plan for cadmium-filtered samples = boron-filtered samples + 3 capsules

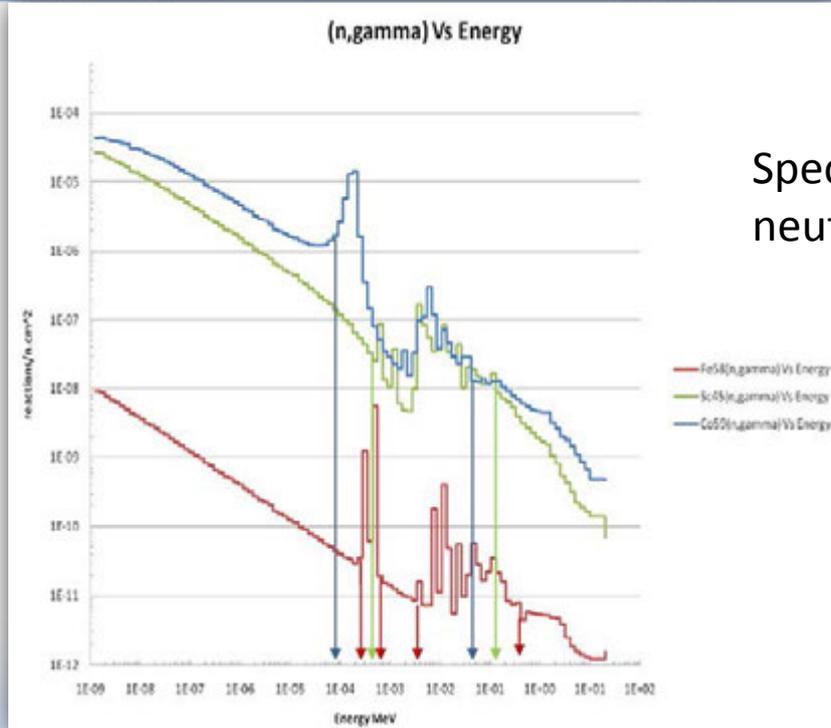
2 capsules for Cm-244

1 capsules for Nd-145 and Pd-105 (Ru-101 and Nd-143 to be loaded in the Sm-149 capsule)

Characterization of the neutron spectrum by measuring the activity of specific wires



Reactions	Energy (MeV) Groups
Sc45 (n, gamma)	4E-4 to 1E-1
Fe58 (n, gamma)	2E-4 to 6E-4 & 2E-3 to 3E-1
Co59 (n, gamma)	1E-4 to 3E-2
S32 (n, p)	1 to 7
Ti46 (n, p)	2.5 to 7.5
Fe54 (n, p)	1 to 7
Ni58 (n, p)	5E-1 to 7
Cu63 (n, alpha)	1.5 to 10



Spectral unfolding techniques are available to infer the neutron spectrum from the activity of the various wires

