Calculation of Delayed-Photoneutron Production in Heavy-Water Reactors using Geant4

Evan Rand, Ph.D. - Canadian Nuclear Laboratories

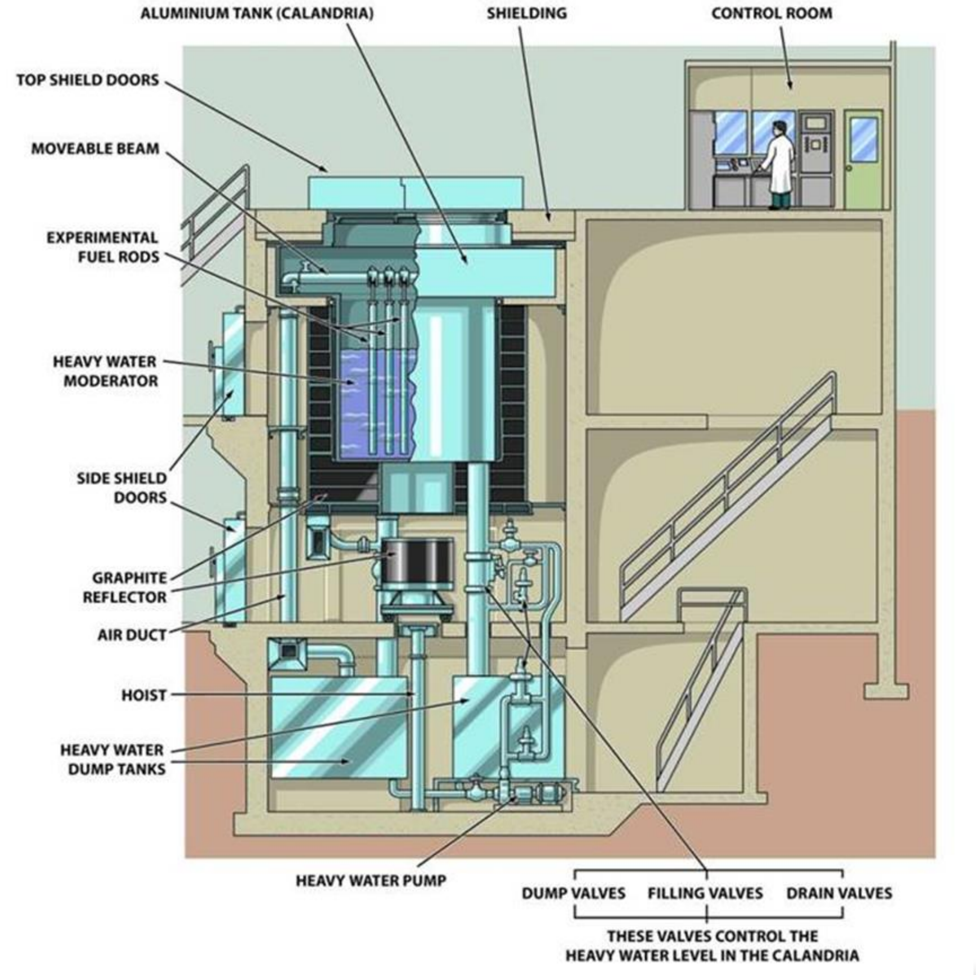
ENSAR2 Workshop **April 24th - 26th, 2019**

CW-01200-001-000

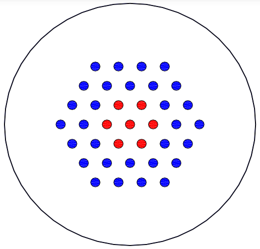
ZED-2 - Zero Energy Deuterium

58 years and counting

- Successor to **ZEEP** – **Z**ero **E**nergy **E**xperimental **P**ile
- First went critical on September 7th, 1960
- Tank type: reactor control via moderator level
- 2521 cores built, 190 of them unique, and counting!



Quick Facts



Power: up to ~200 W (thermal)

Peak Neutron Flux: 1×10^9 n/cm^2 s thermal, 5×10^8 n/cm^2 s fast

Calandria: 3.36 m in diameter, 3.35 m in height

Fuel: Various types and assemblies

Moderator: Heavy water (99.8 to 97.5 weight% D_2O), soluble poison capability, temperatures up to 90°C, and variable core height (criticality achieved by pumping moderator into calandria)

Fuel Arrangement: variable pitch square and hexagonal lattices

Coolant: Heavy water, light water, air, CO_2, organics, Pb-Bi, etc. (not active). Temperatures up to 300°C in some channels.

Flexibility: We can operate with new fuels/coolants/materials as required

Fuel

Natural UO_2 Bundles:

- 7, 19, 28, 37, and 43-element

Other Natural U flavours:

- Metal, Carbide, Silicide bundles
- ZEEP rods

Mixed oxides:

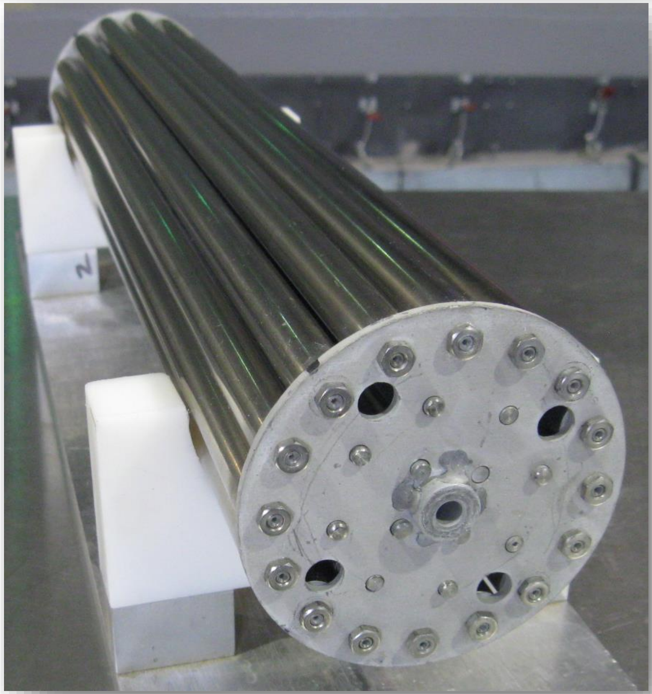
- Pu-U, ^{233}U-Th, Pu-Th, ^{235}U-Th

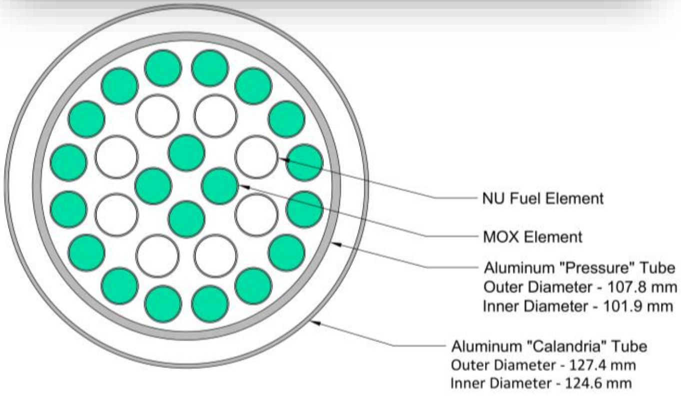
Bundles with burnable absorber
(Low Void Reactivity)

Enriched or reprocessed UO_2 bundles:

- LEU, RU

Assembly geometry: bundles in Pressure Tube/Calandria Tube, clad rods, etc.

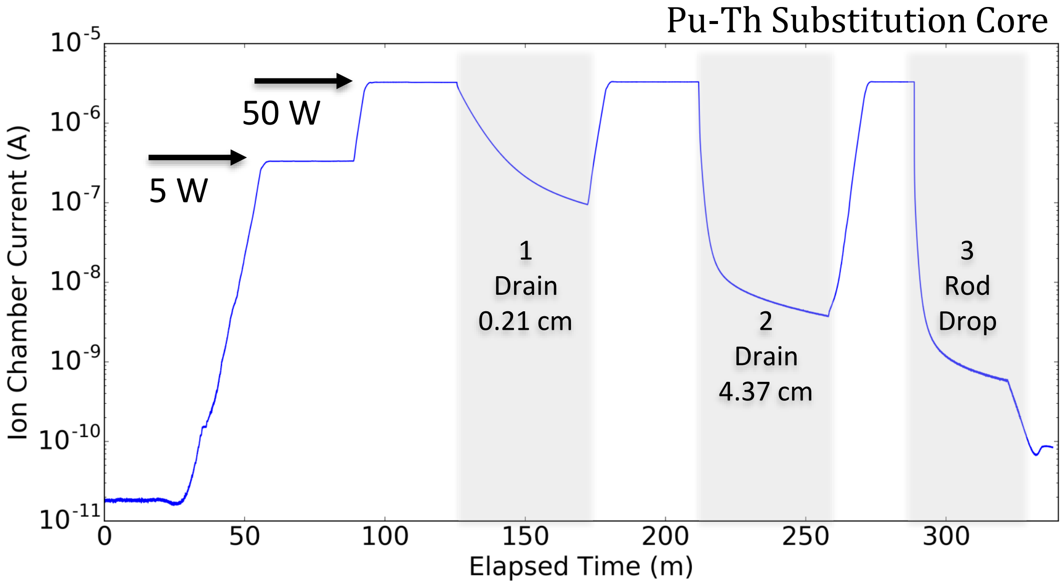




Reactor Transient Experiments in ZED-2

Understanding and predicting the transient behaviour of reactor systems is paramount for reactor safety and operation. Transients occur during startup/shutdown, accident scenarios (coolant loss), etc. The behaviour depends on a number of factors such as fuel type(s) (^{nat}U, LEU, MOX), reactor configuration, etc.

The study of these processes which control the time-dependent behavior of a nuclear reactor in known as ***Reactor kinetics.***



Reactor Kinetics

The energy- and time-dependent diffusion equation:

$$\frac{1}{v}\frac{\partial \phi}{\partial t} = (F_p - M)\phi + S_d(r,E,t) + S(r,E,t)$$

Prompt-neutron production

Neutron leakage/removal

Delayed neutrons

Neutron sources

$$F_p\phi = \chi_p(E)\int_0^\infty dE'\,\nu_p(E')\Sigma_f(r,E',t)\phi(r,E',t)$$

$$M\phi = -\vec{\nabla}\cdot D(r,E)\vec{\nabla}\,\phi(r,E,t) + \Sigma(r,E,t)\phi(r,E,t) - \int_0^\infty dE'\,\Sigma_s(r,E'\rightarrow E,t)\phi(r,E',t)$$

Almost impossible to solve in all practical scenarios, even when using today's most powerful computers. We are left with two approaches:

Deterministic (Point Kinetics) – An exact solution to an approximate problem.
Stochastic (Monte Carlo) – An approximate solution to the exact problem.

See: Introduction to Nuclear Reactor Kinetics, by Daniel Rozon

Point Kinetics ...assumes flux is separable:

$$\phi(r,E,t) = n(t)\,\psi(r,E,t)$$

Nuclear fission:

$$n + X \rightarrow A + B + \nu_p n_p + \nu_d n_d + \beta + \gamma$$

Total neutron yield:

$$\nu = \nu_d + \nu_p$$

Inverse Point Kinetics Solution:

$$\frac{dn(t)}{dt} = \left(\frac{\rho(t) - \sum_{i=1}^{ngroups} \beta_i}{\Lambda}\right) n(t) + \sum_{i=1}^{ngroups} \lambda_i C_i$$

$$\frac{dC_i(t)}{dt} = -\lambda_i C_i(t) + \frac{\beta_i}{\Lambda} n(t)$$

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$$\rho(t) = \frac{k_{eff}(t) - 1}{k_{eff}(t)}$$

Reactivity

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Effective multiplication factor

$$\rho(t) = \frac{k_{eff}(t) - 1}{k_{eff}(t)}$$

Reactivity

critical	$k_{eff} = 1$
supercritical	$k_{eff} > 1$
subcritical	$k_{eff} < 1$

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Prompt-neutron generation time

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Precursor concentrations

Prompt-neutron generation time

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Delayed-neutron fraction (β):

$$\beta = \frac{\nu_d}{\nu} \longrightarrow \beta_i = \frac{\nu_{d_i}}{\nu}$$

Delayed-neutron groups with differing time constants.

Inverse Point Kinetics Solution:

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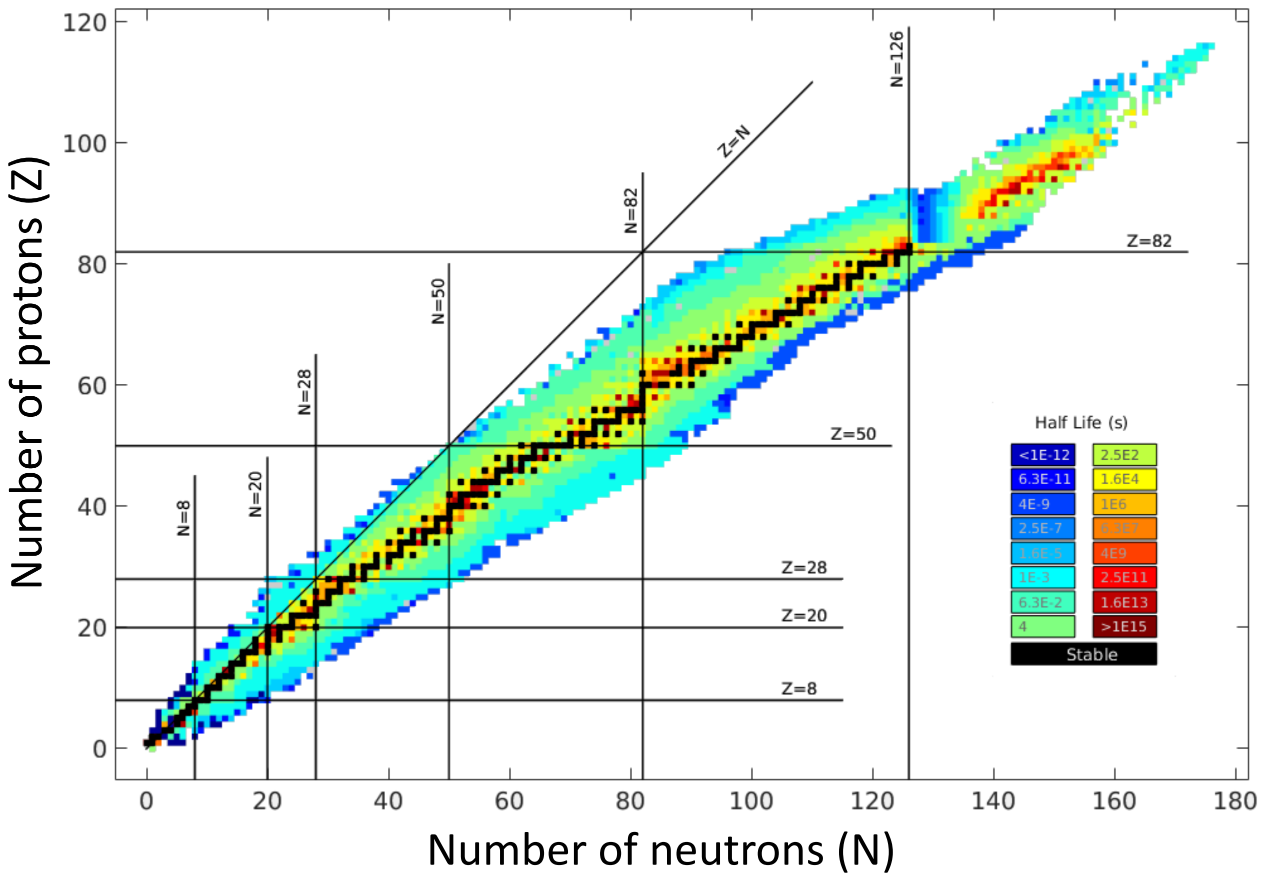
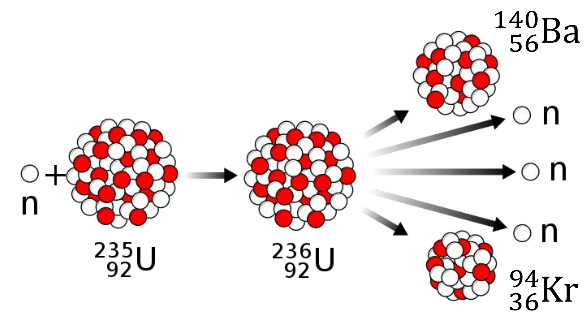
Nuclear Landscape



Nuclear Fission

$$ {}^{1}_{0}n + {}^{A_X}_{Z_X}X \rightarrow {}^{A_X+1}_{Z_X}X $$

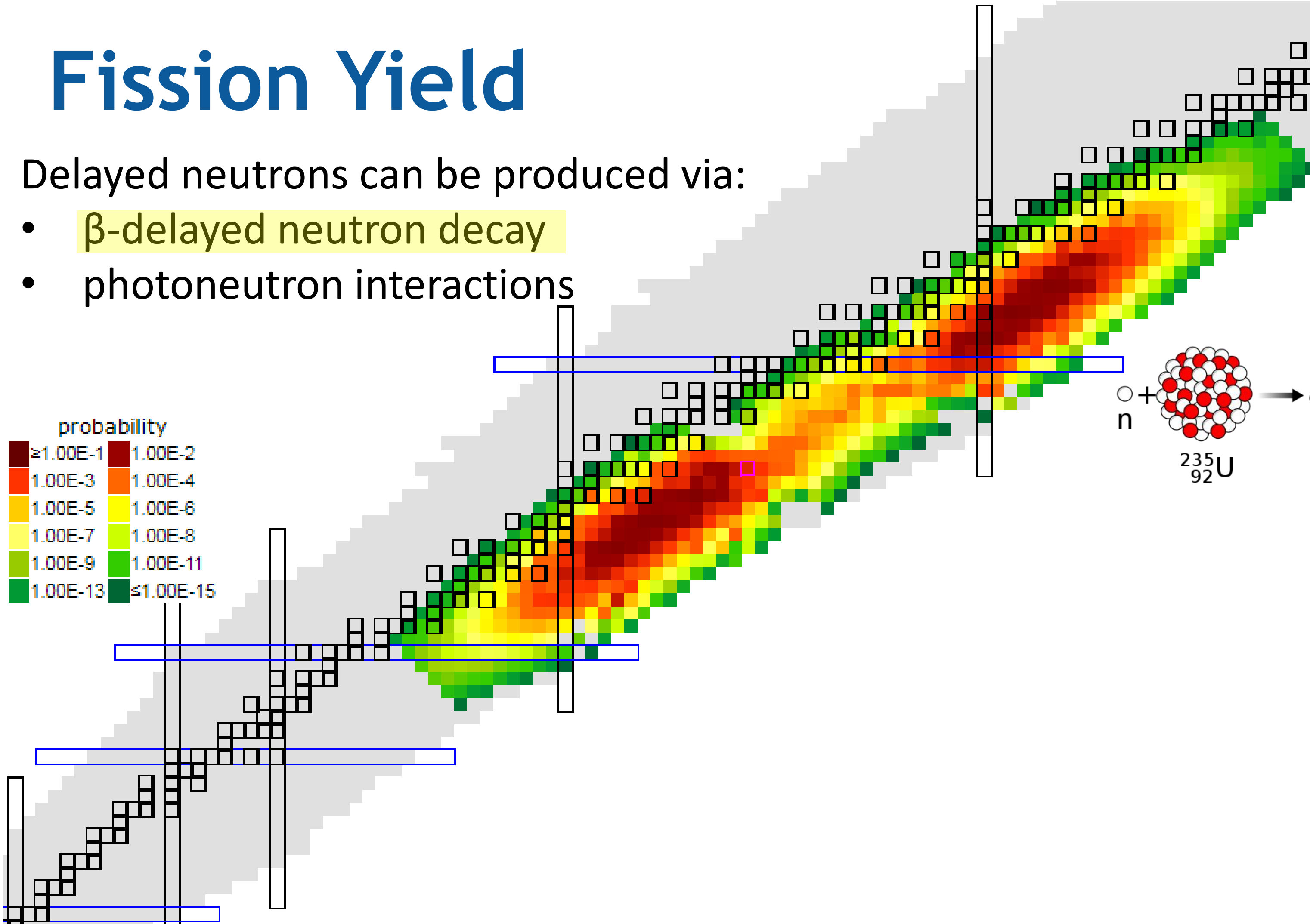


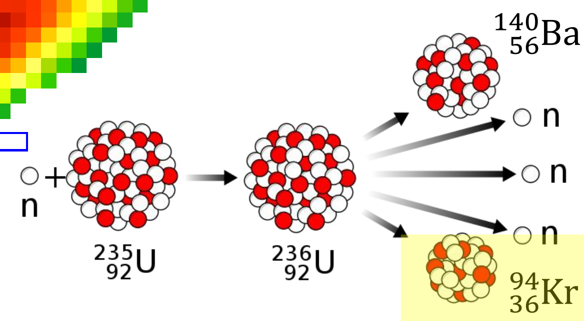
The presence of delayed neutrons is perhaps most important aspect of the fission process from the viewpoint of reactor control!

Fission Yield

Delayed neutrons can be produced via:

- β-delayed neutron decay
- photoneutron interactions





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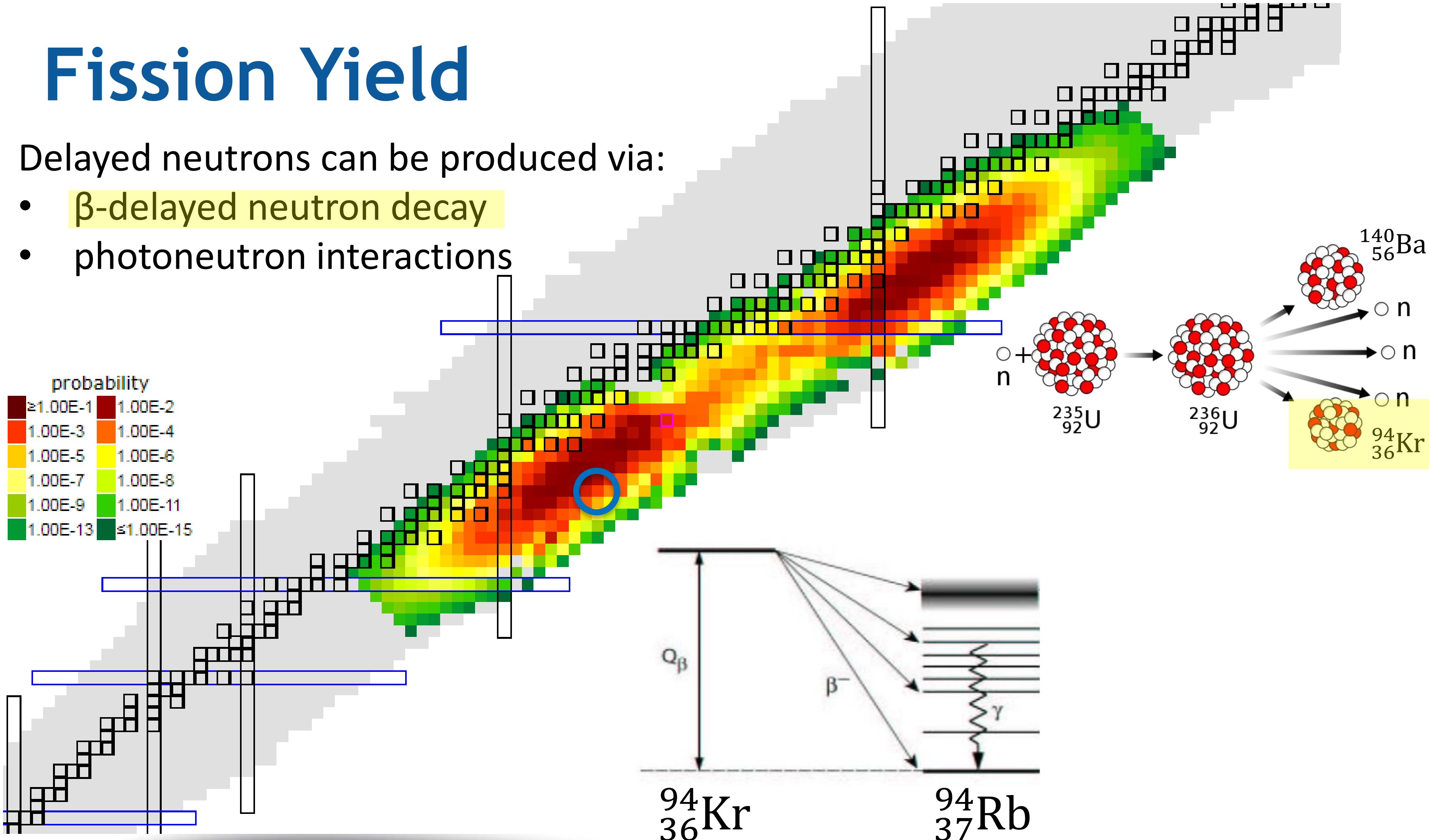

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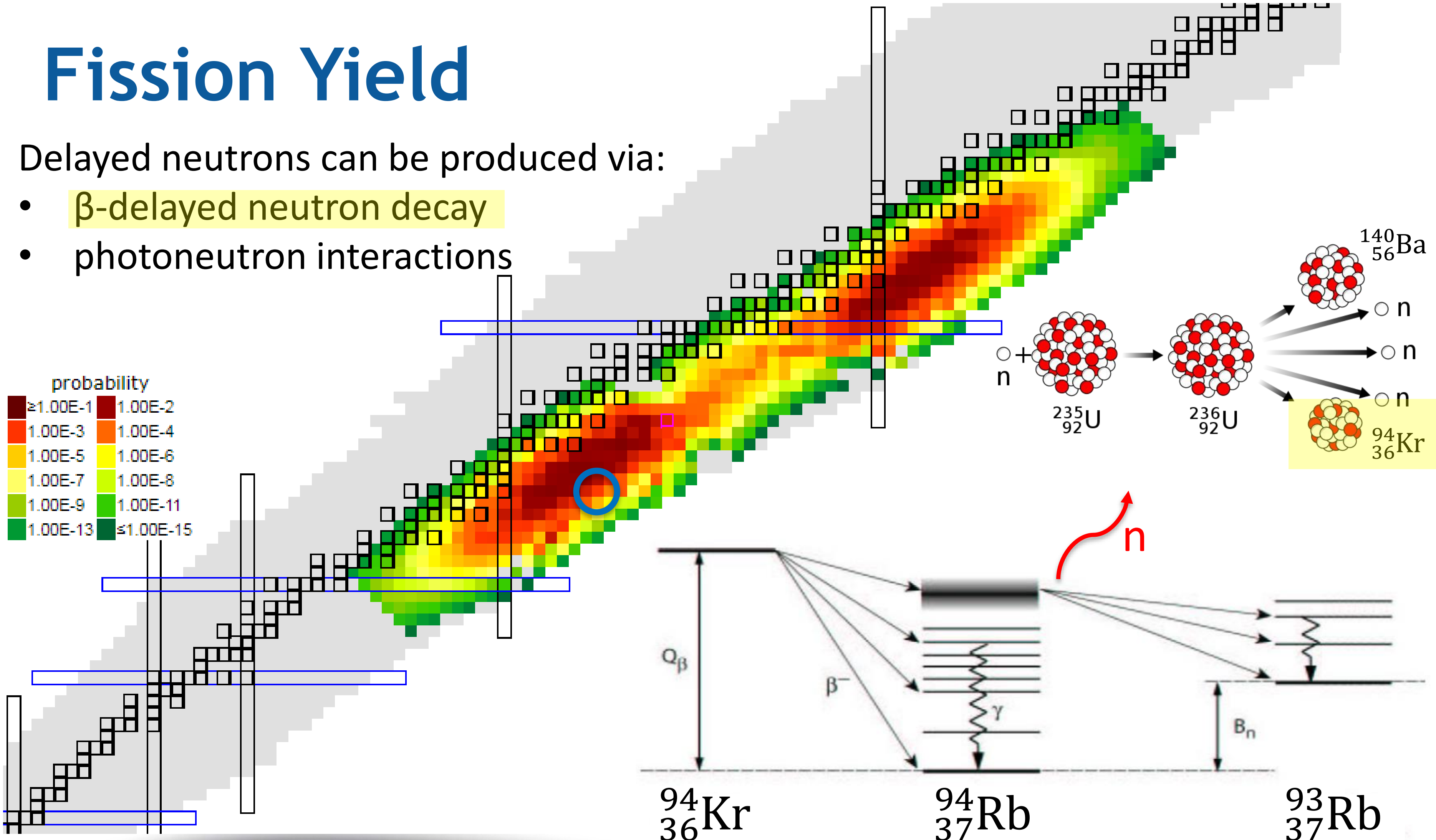

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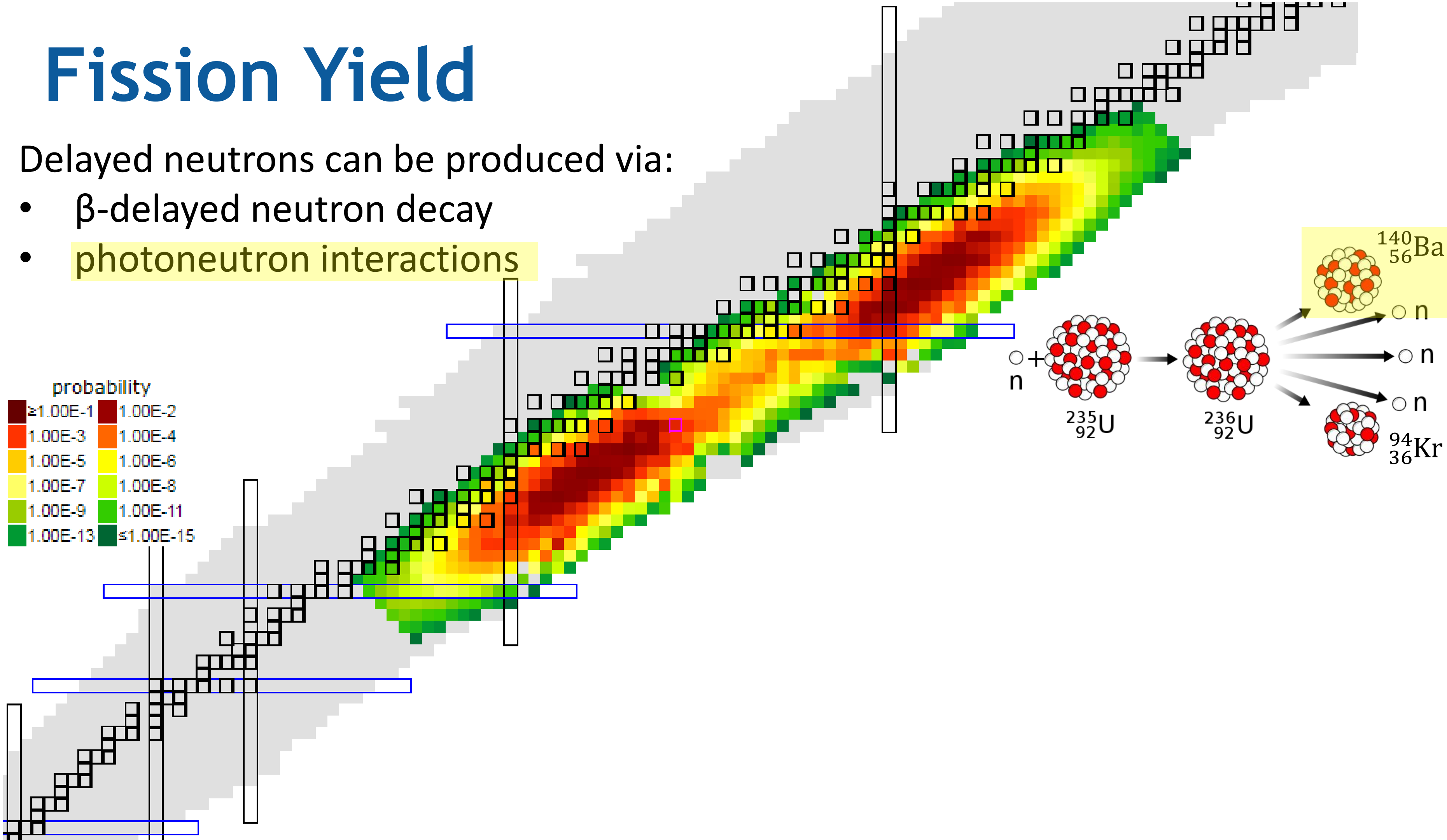

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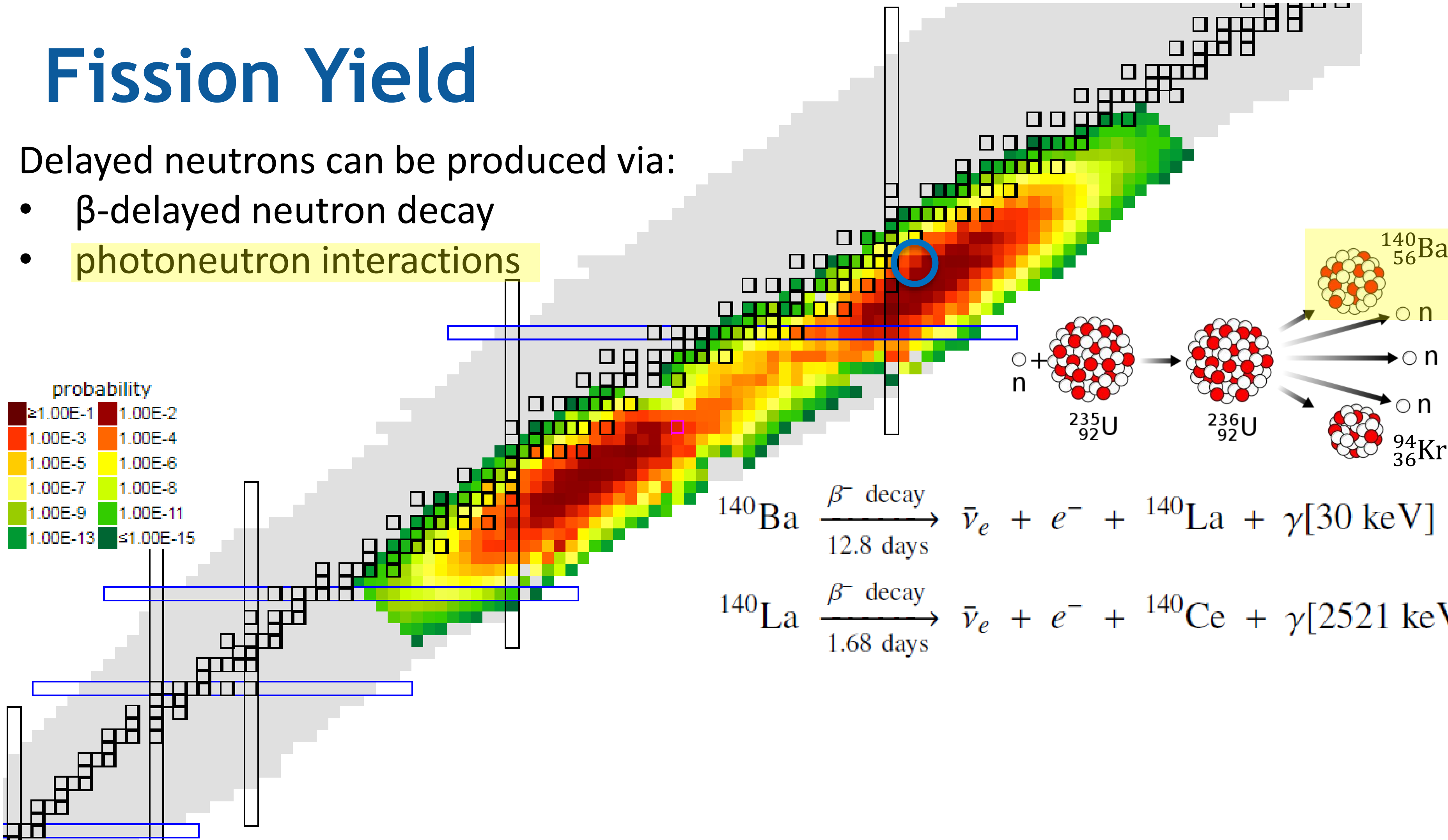

$$^{140}\mathrm{Ba} \xrightarrow[12.8\ \mathrm{days}]{\beta^-\ \mathrm{decay}} \bar{\nu}_e + e^- + {}^{140}\mathrm{La} + \gamma[30\ \mathrm{keV}]$$

$$^{140}\mathrm{La} \xrightarrow[1.68\ \mathrm{days}]{\beta^-\ \mathrm{decay}} \bar{\nu}_e + e^- + {}^{140}\mathrm{Ce} + \gamma[2521\ \mathrm{keV}]$$

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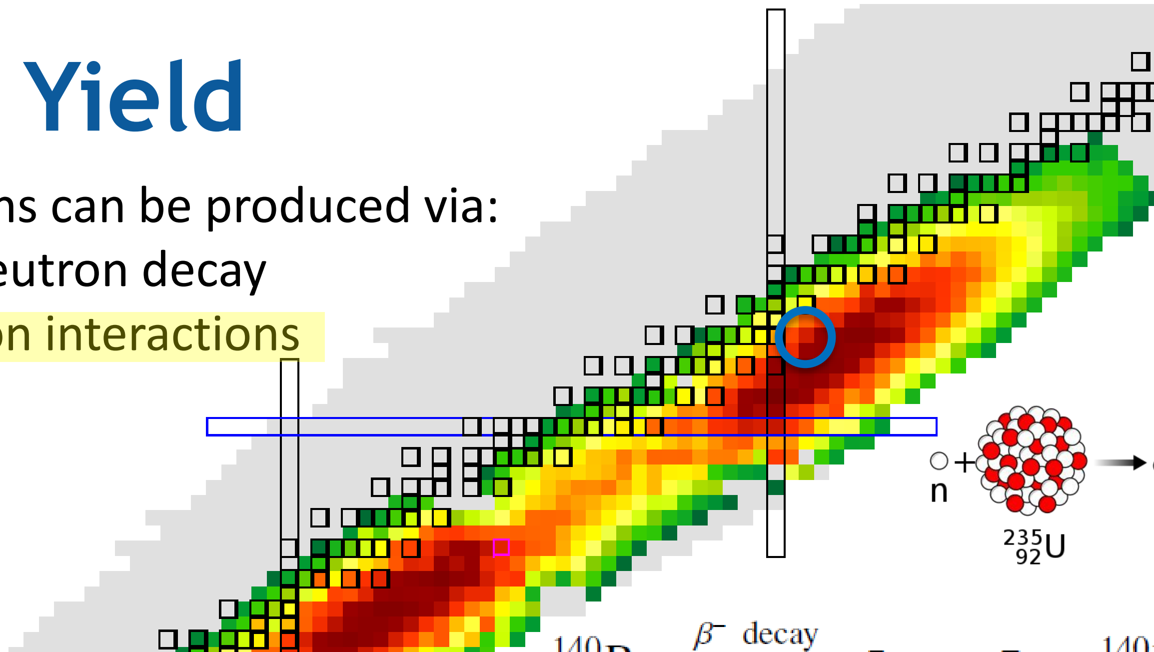

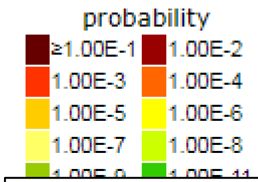

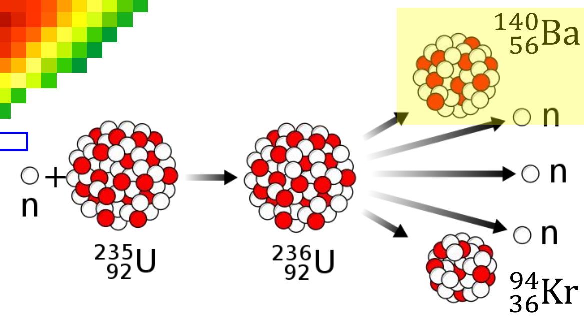

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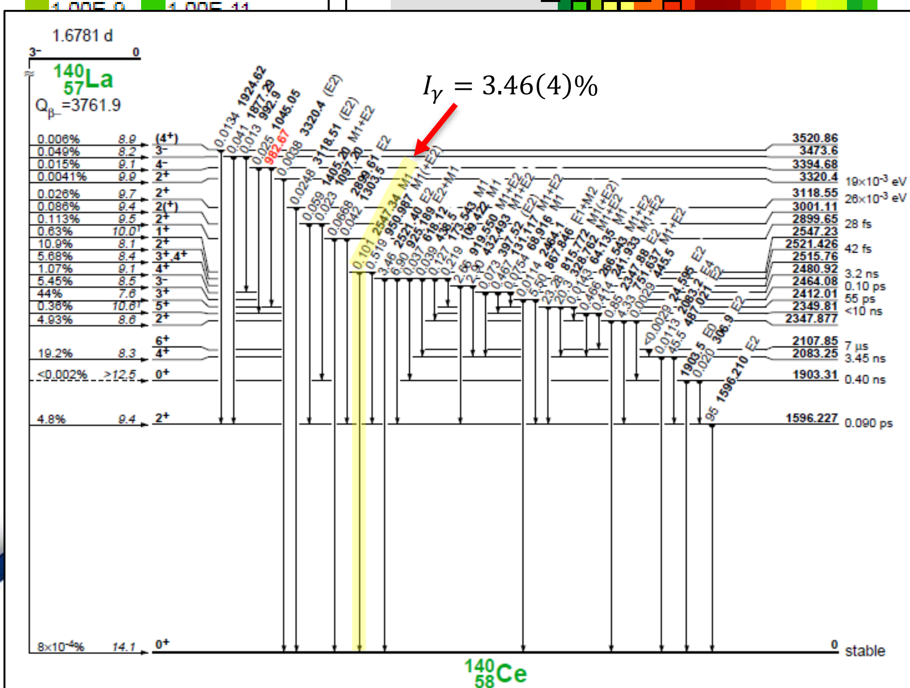

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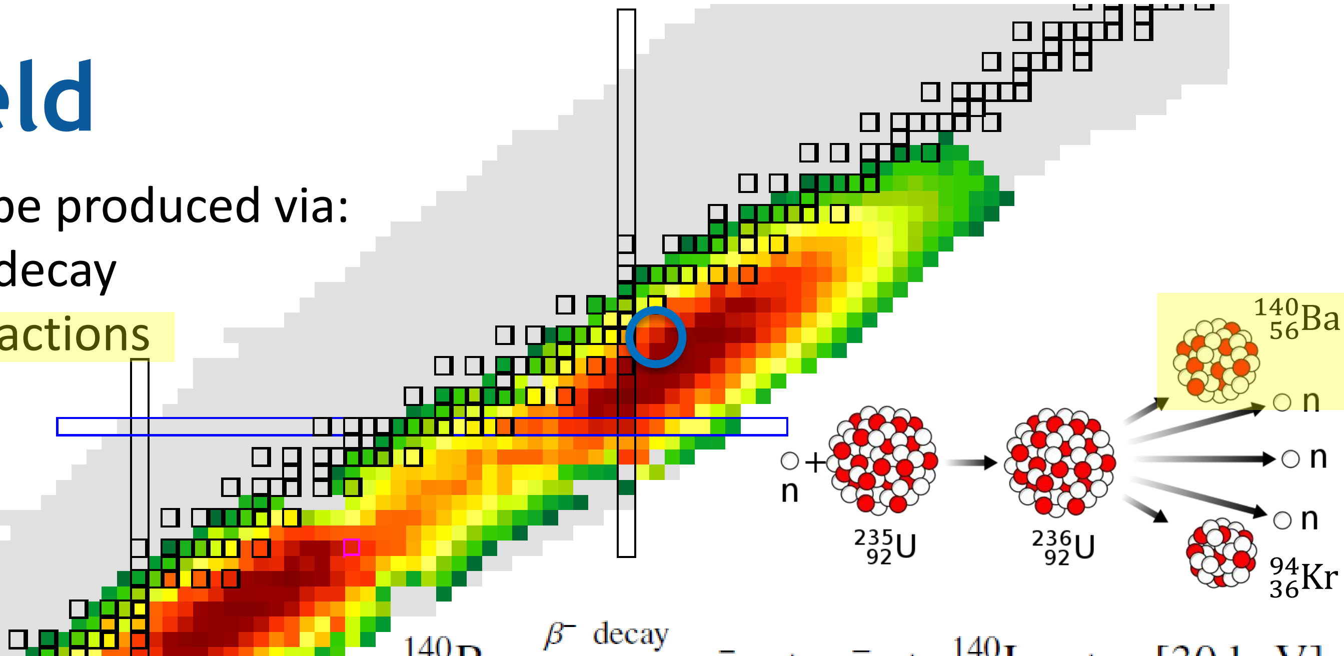

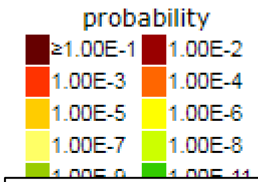

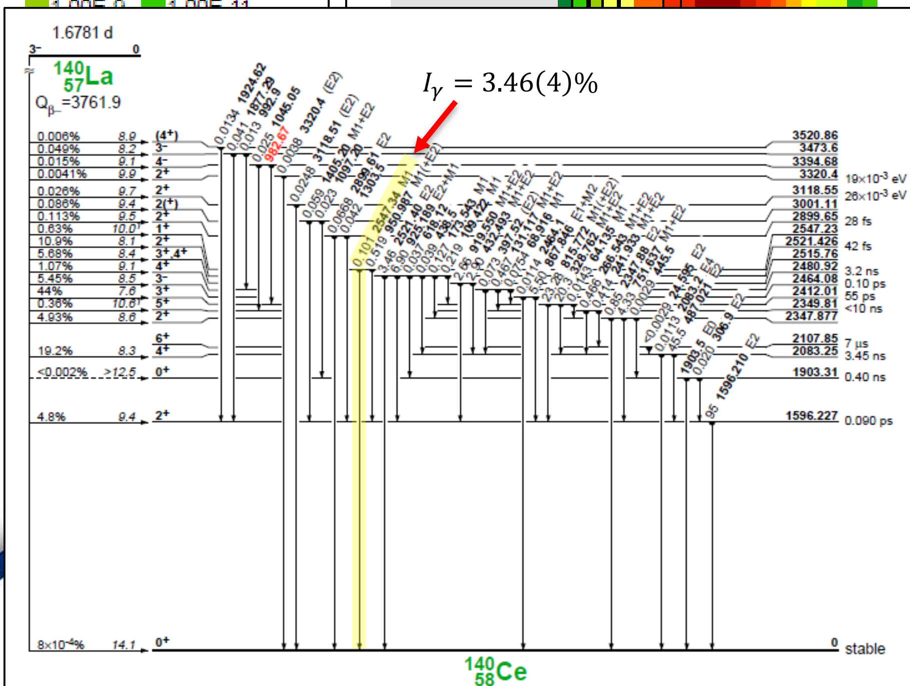

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Deuterium breakup reaction:

$$BE\left({}^{A}_{Z}X\right) = Z \cdot M_p c^2 + N \cdot M_n c^2 - M'\left({}^{A}_{Z}X\right)c^2$$

$$BE\left({}^{2}H\right) = 2.225\ MeV$$

$$\gamma[2521\ keV] + {}^{2}H \rightarrow p + n$$

Delayed Neutrons

$t_{1/2} \sim 13$ days

$t_{1/2}\ (s)$	1	2	3	4	5	6	7	8	9		10	11		← group number	
Photo-neutron	1,107,360	190,800	15,840	5,924	1,620	462.1	144.1	55.7	22.7	-	6.22	2.3	-	-	-
Direct-delayed	-	-	-	-	-	-	-	55.6	24.5	16.3	5.21	2.37	1.04	0.424	0.195
								1	2	3	4	5	6	7	8

$t_{1/2} \sim 1$ min

A set of parameters covering the all photoneutron timescales was recommended for **CANDU** analyses*. Interestingly, the data was stitched together from three different sources (Bernstein et al., Ergen, and Baumann), using experimental ^{235}U data from as far back as the 1940s.

We performed detailed "microscopic" Geant4 calculations based on nuclear data to extract the photoneutron group structure, which had never been performed in the past.

Calculations performed on ^{235}U, ^{233}U, and ^{239}Pu.

*P. J. Laughton, Revised delayed photoneutron data for use in CANDU-reactor analysis, in: Proc. Ann. Conf. Canadian Nucl. Soc., no. 2, 1998, pp. 376–388

Simulation

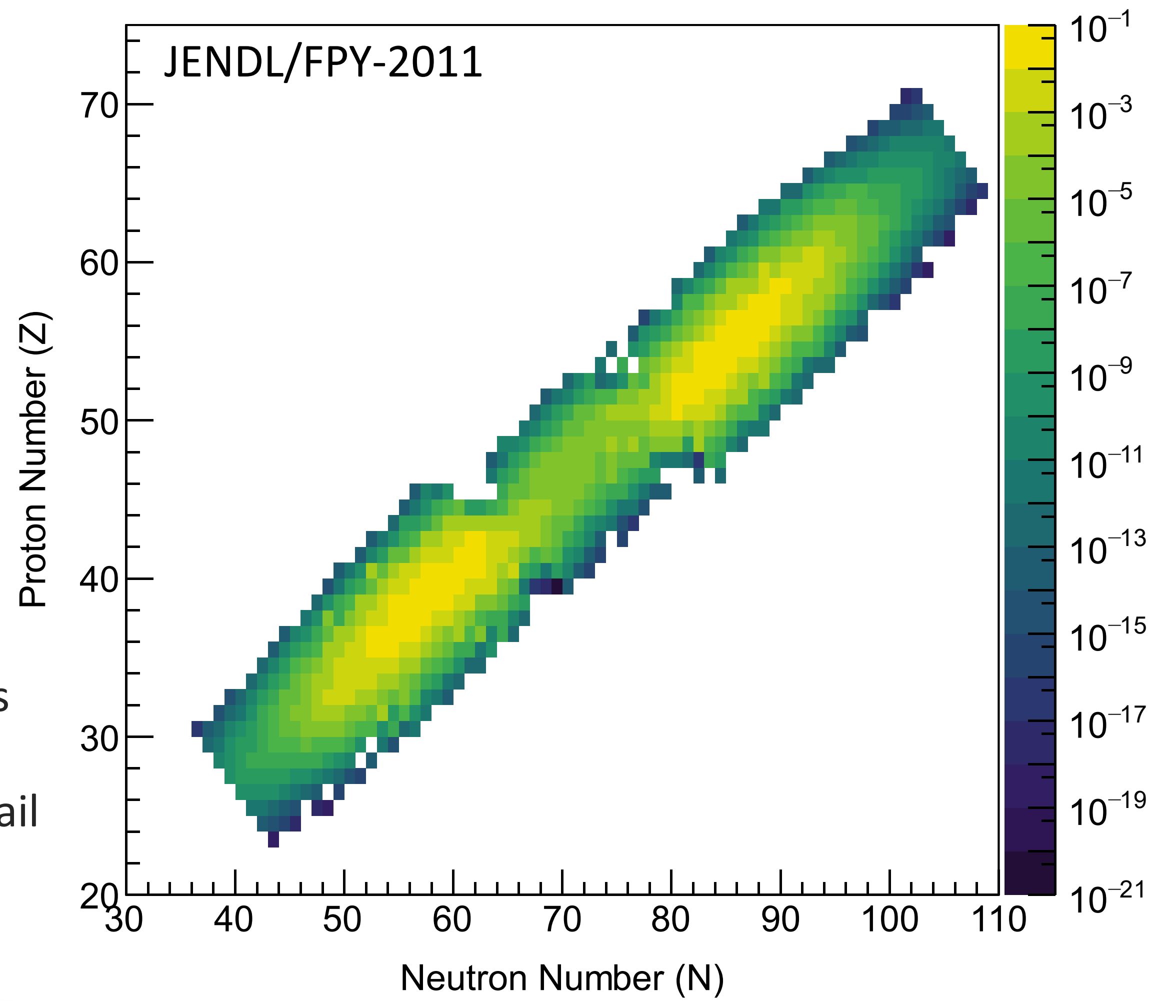
The simulation was built using Geant4 (*10.03.p02*), and applied radioactive decay and photon emission physics:

G4RadioactiveDecay5.1.1

G4PhotonEvaporation4.3.2

G4ENSDFSTATE2.1.

No geometry was simulated, rather, the emission of β particles and γ-rays from the radioactive decay process was tracked in detail from input isotope fission yields.



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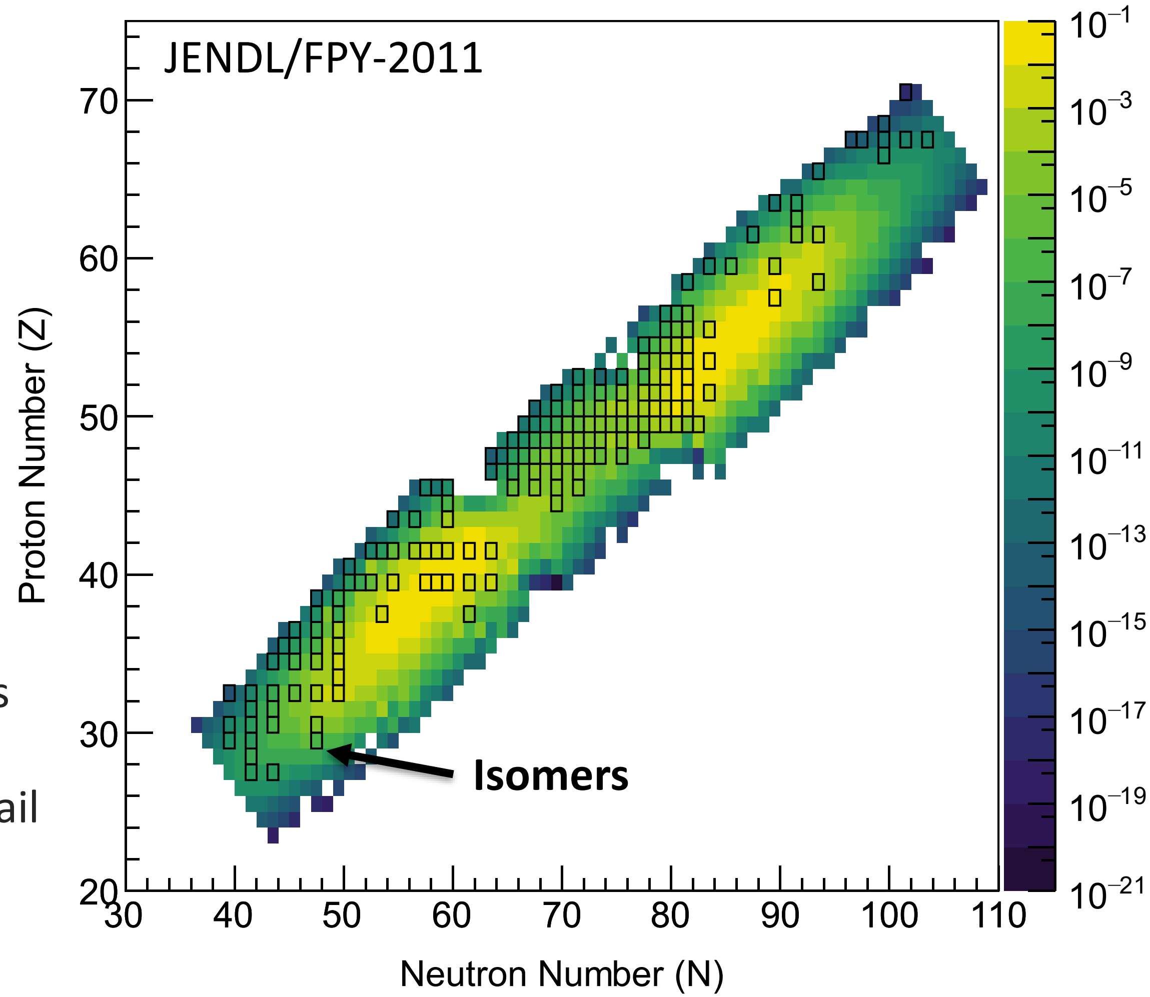

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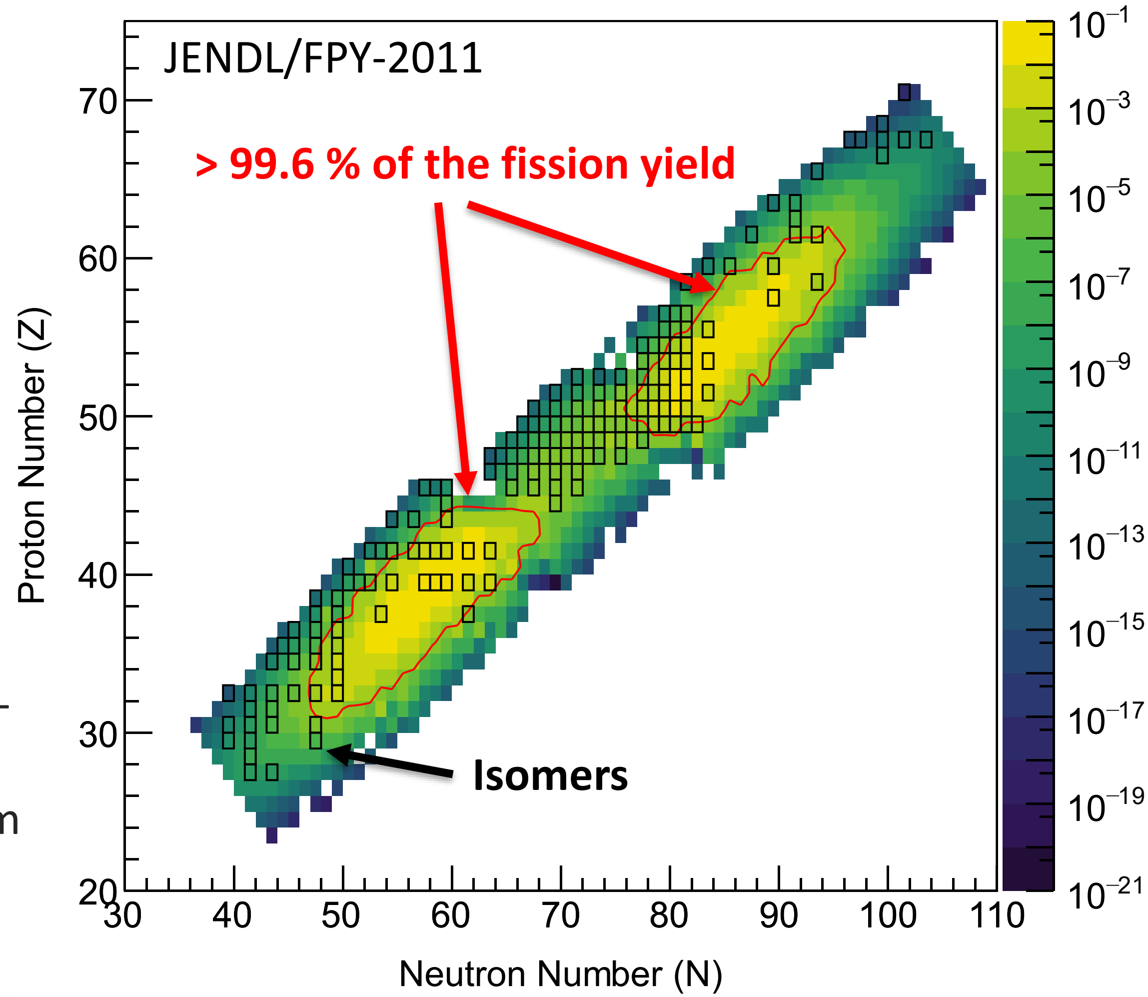
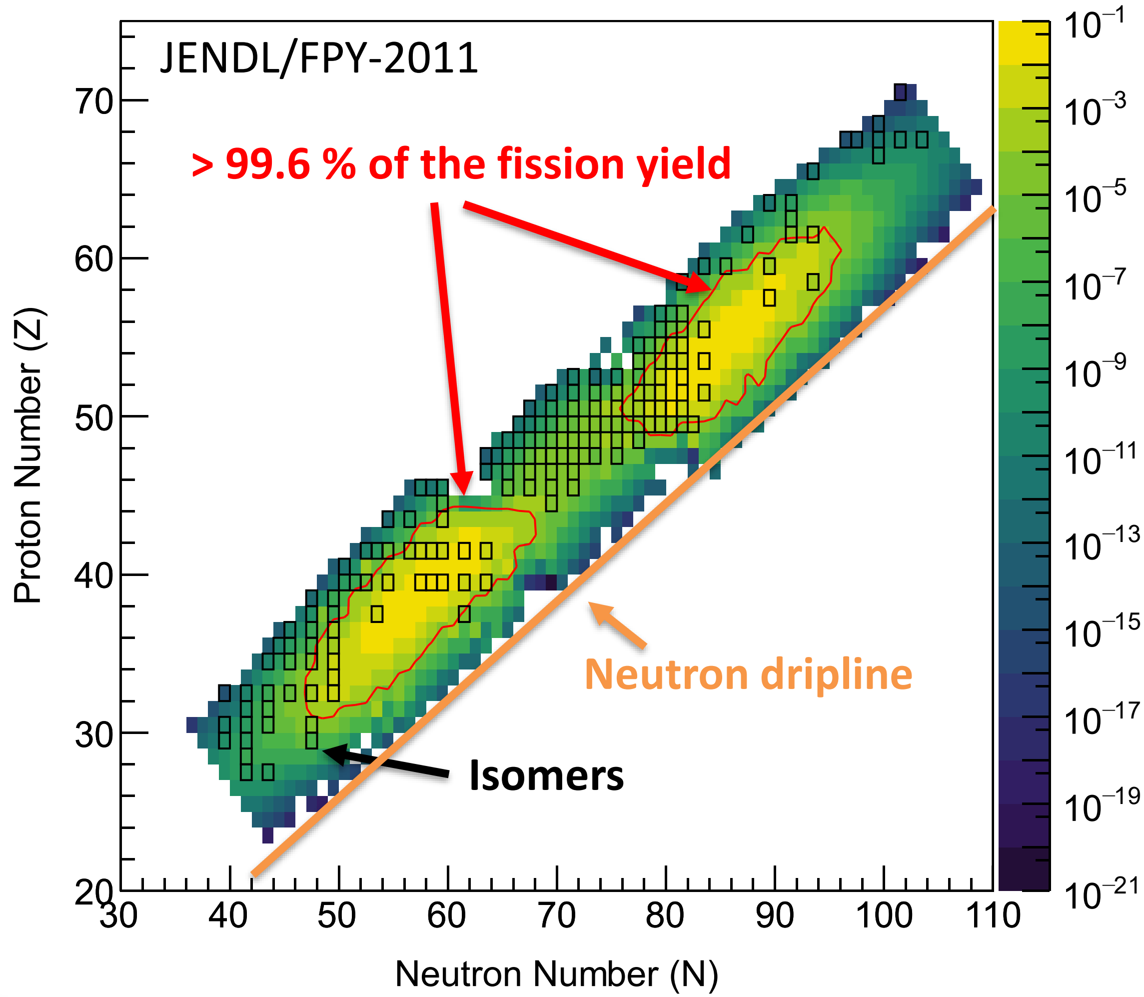


Simulation

Approximations

1. Nuclei near the neutron dripline with no known nuclear data information were input into the simulation using theoretical β-decay half-lives*. These calculations used a proton-neutron quasiparticle random-phase approximation (pn-QRPA) and applied mass formula predictions from Hilf et al.† which are thought to be the most reliable far from stability.



*A. Staudt et al., Atomic Data and Nuclear Data Tables 44 (1) (1990) 79-132.
†E. Hilf et al., Gross theory of nuclear masses and radii, in: the 3rd International Conference on Nuclei far from Stability (1976)

Simulation

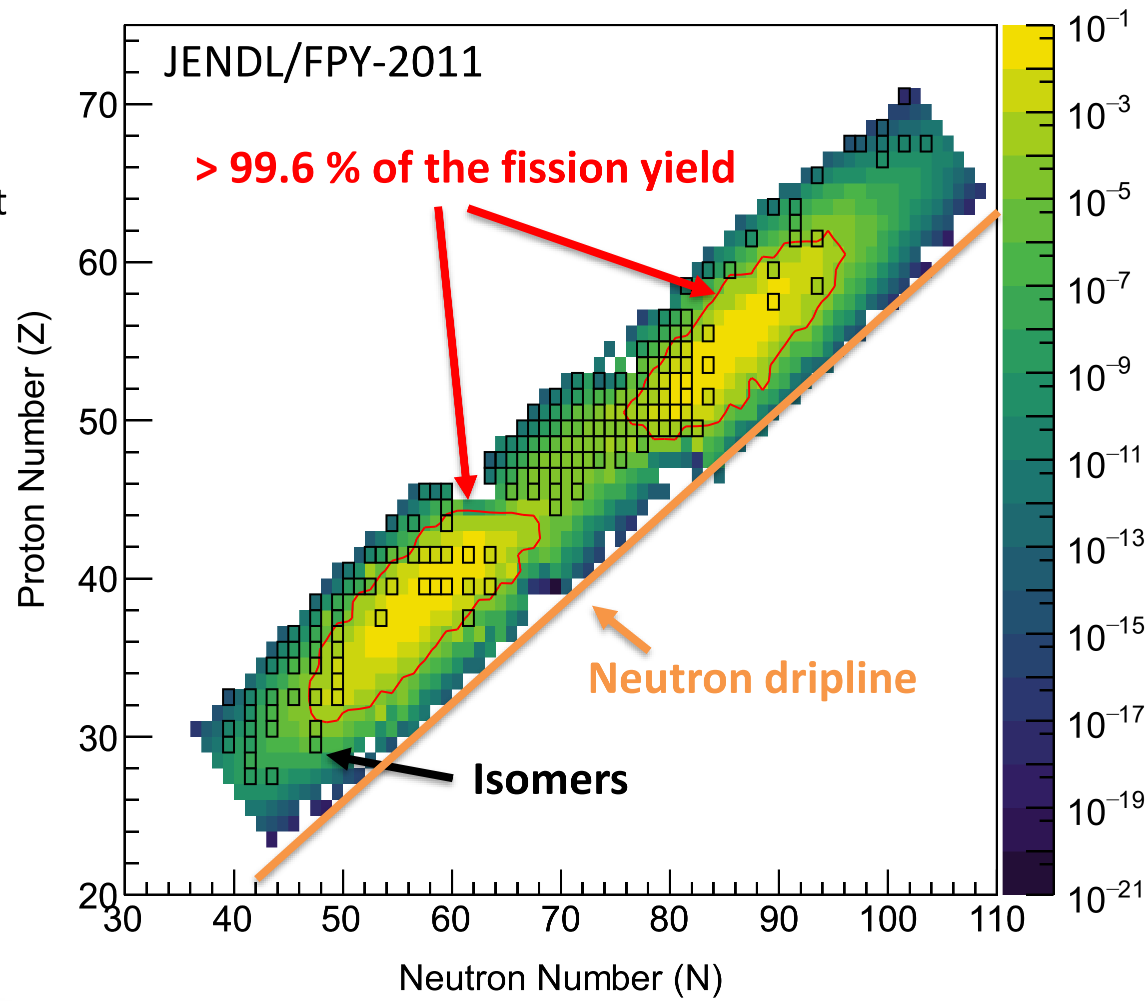
Approximations

2. Geant4 currently does not support β-delayed neutron decay.

2019 Planned Feature in Geant4!

Fortunately, we calculated < 1% of the total fission yield directly populates β-delayed neutron branches in ^{235}U, determined from measured branching ratios and predictions using an updated empirical Kratz Herrmann formula (KHF)* where data was lacking.

* B. Pfeiffer et al., Progress in Nuclear Energy 41 (1) (2002) 39-69.



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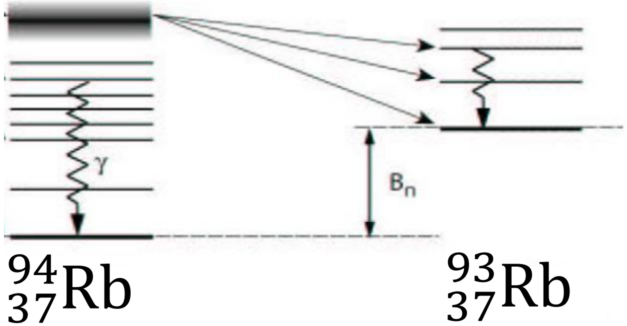
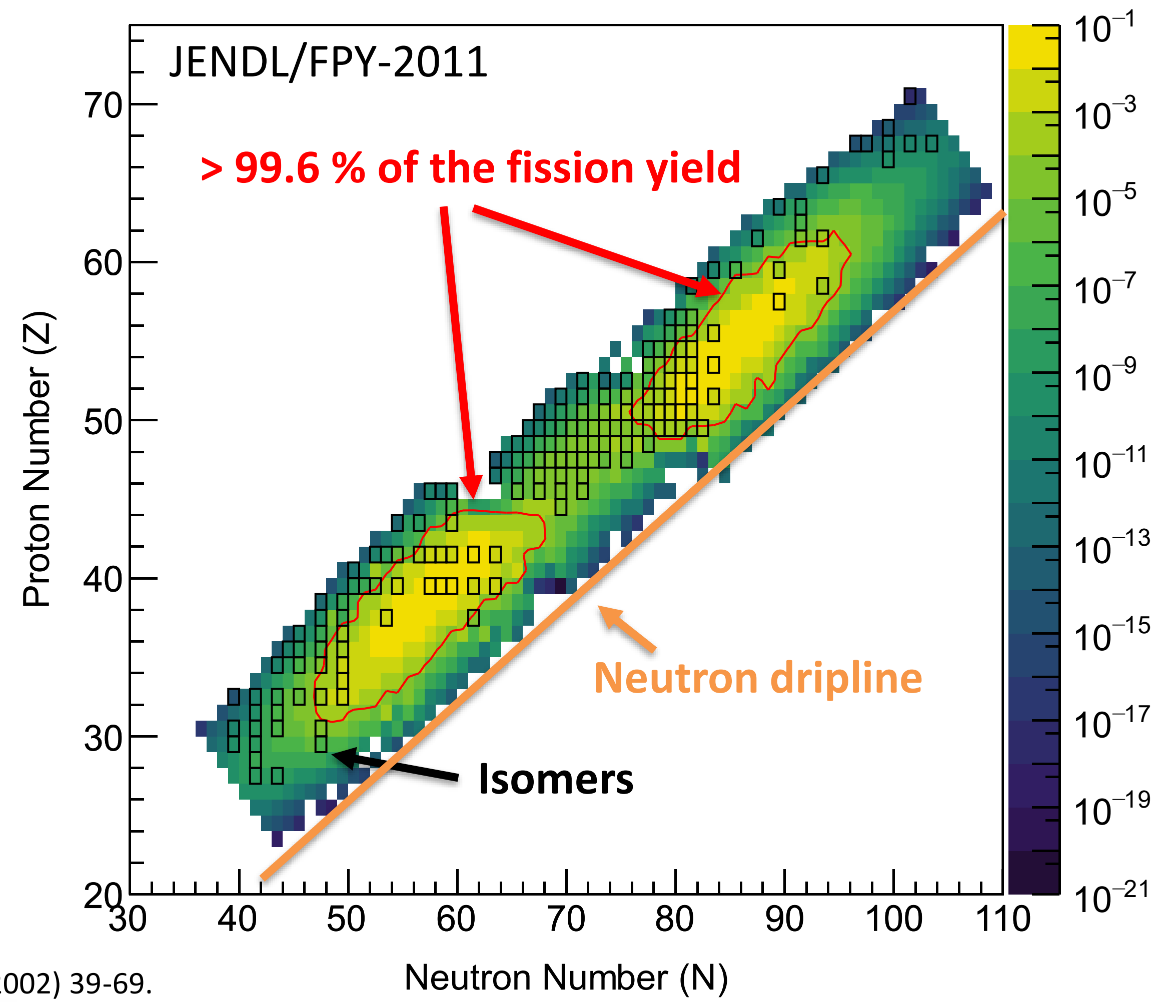


Correlations between β-delayed neutron emission and high-energy γ decay was also found to be small. Determined by conservatively assuming N_γ to be reduced by P_n for each isotope. In total, the number of high-energy γ-rays emitted per fission was only reduced by < 1.5%.



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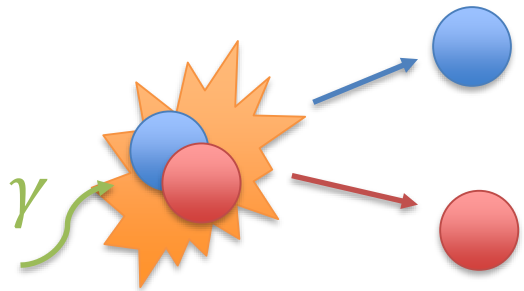
Simulation

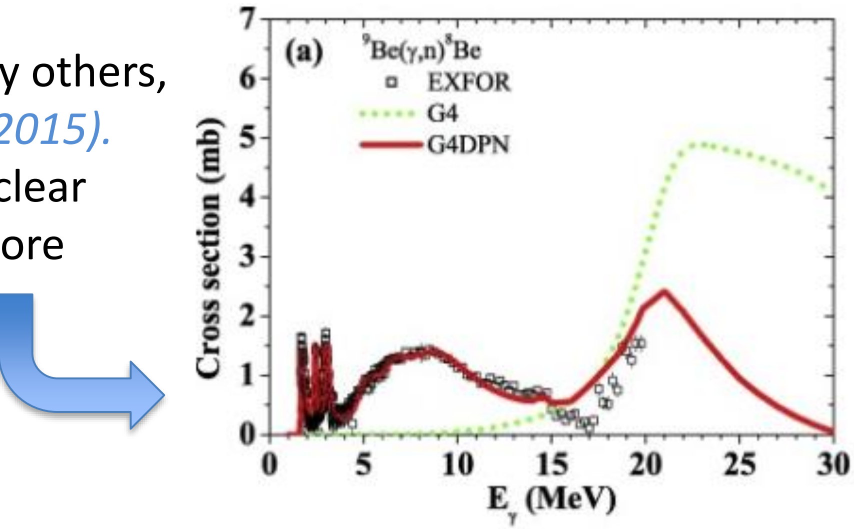
Modifications to Geant4 Physics

Photonuclear interactions with nuclei are handled by the **`G4PhotoNuclearProcess`** class in Geant4 (using a Bertini-style cascade). The model has only been validated at much higher energies (**> 60 MeV**) and fails to produce neutrons from ^{2}H in our energy regime.



$$\sigma_{\mathrm{Geant4}} = 0$$
$$^{2}H(\gamma, n)\ ^{1}H$$

Large discrepancies for other isotopes observed by others, *J. W. Shin, Nucl Instrum Methods Phys Res B 358 (2015).*
In this work a data-based (ENDF/B-VII.1) photonuclear reaction model for Geant4 was developed for a more accurate simulation of photon-induced neutron production < 30 MeV.



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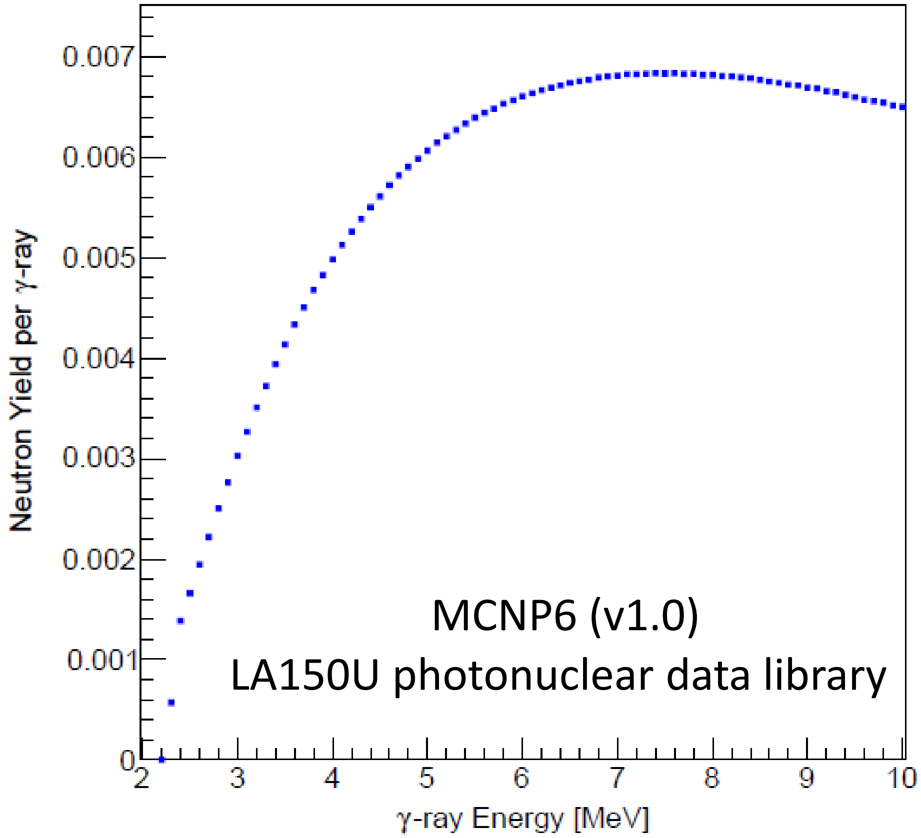
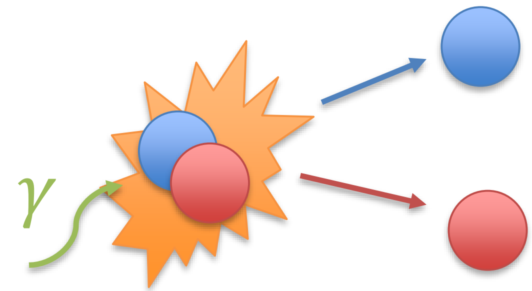


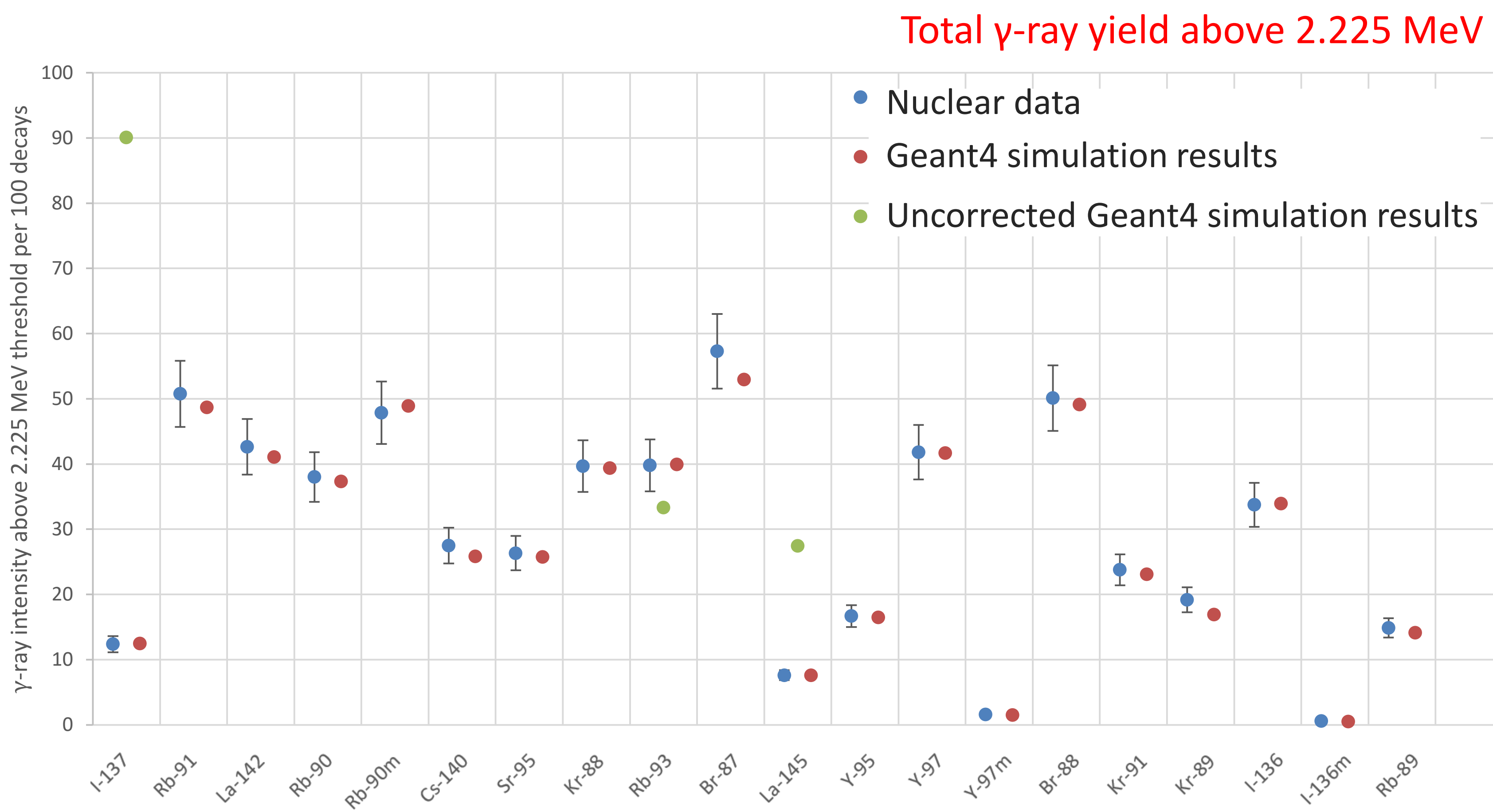


MCNP6 photonuclear physics* for deuterium was hardcoded into our Geant4 simulation.

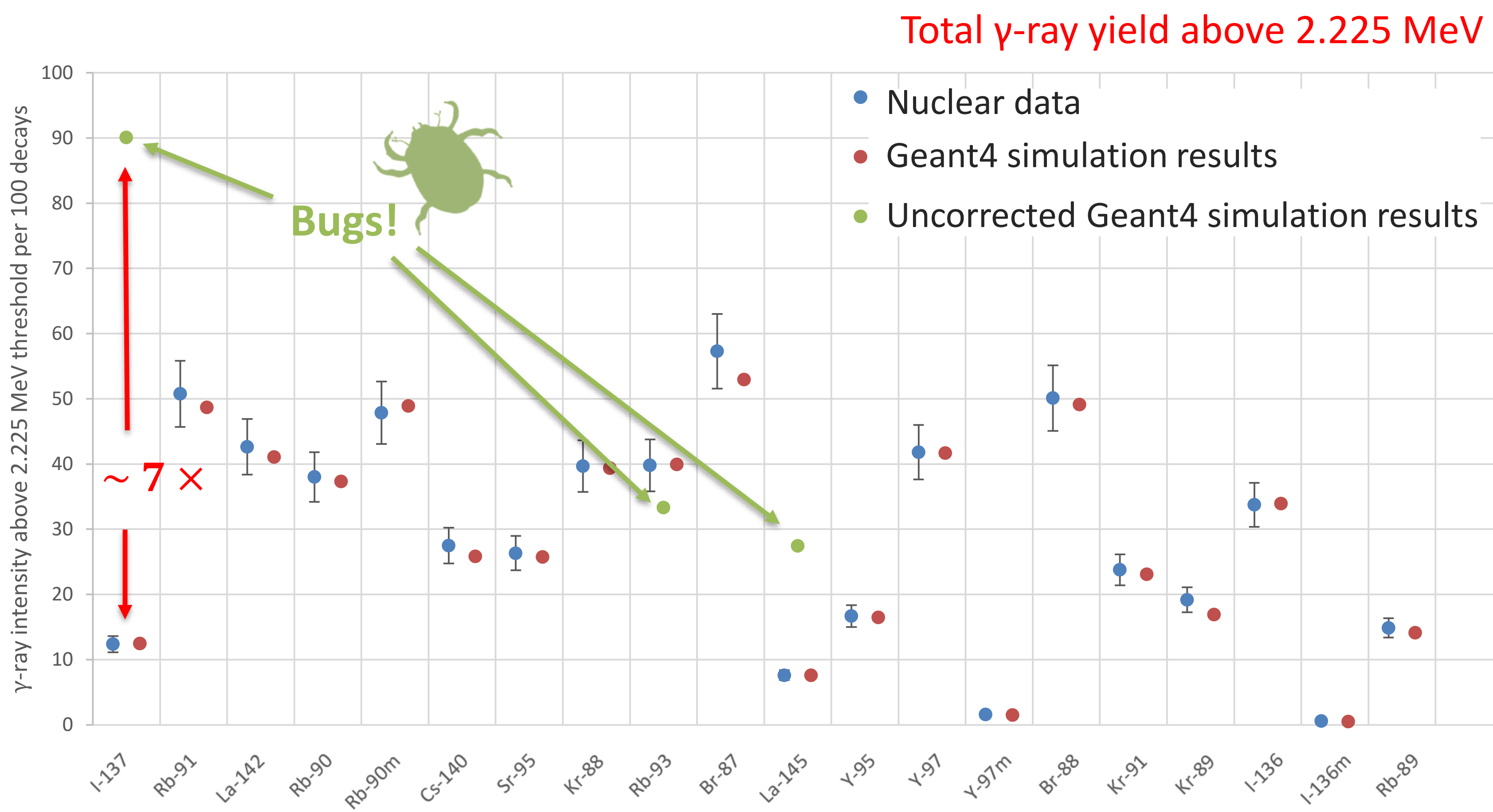
MCNP6 photoneutron production simulation from a point-source of monoenergetic γ-rays within a 10 m radius homogeneous sphere of heavy water, which effectively mimics an infinite heavy water bath for γ-ray energies < 10 MeV.

*M. C. White, Release of the LA150U photonuclear data library, Tech. Rep. memo X-5:MCW-00-87(U), Los Alamos National Lab (2000)

Validation Work

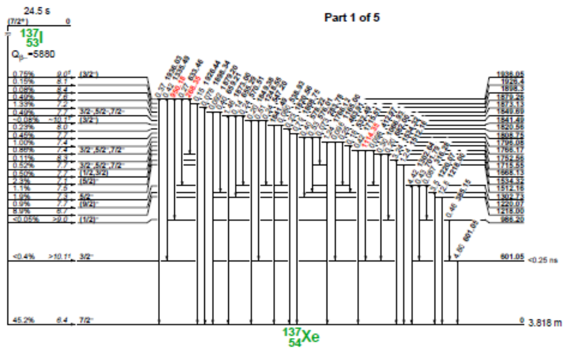


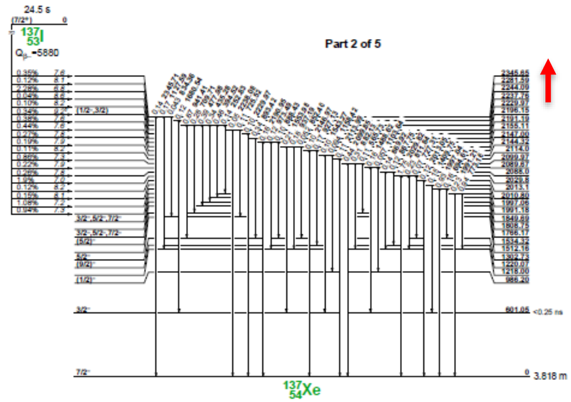
Validation Work



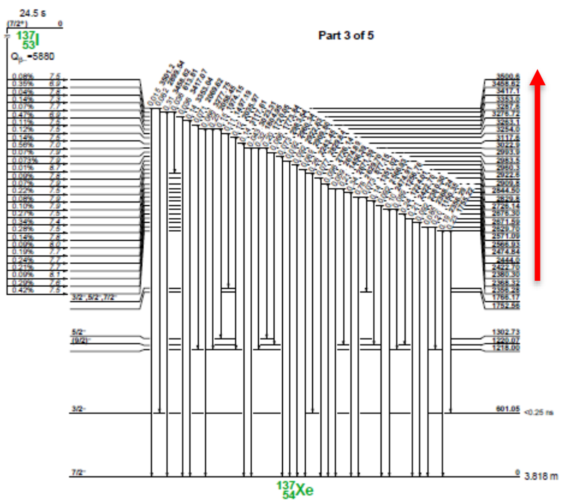
Validation Work

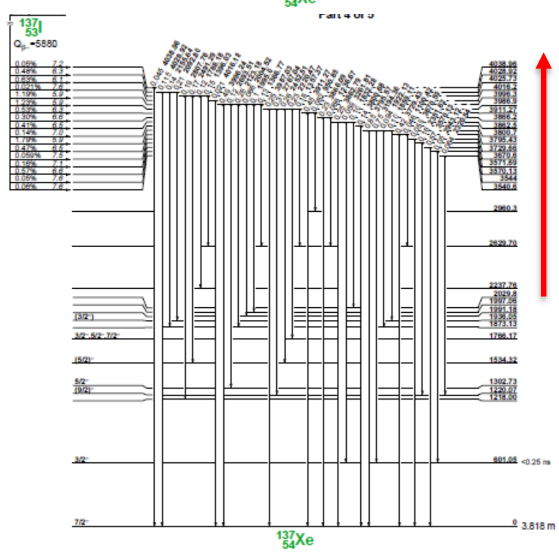
$$^{137}\mathrm{I} \xrightarrow{\beta^-} {}^{137}\mathrm{Xe}$$

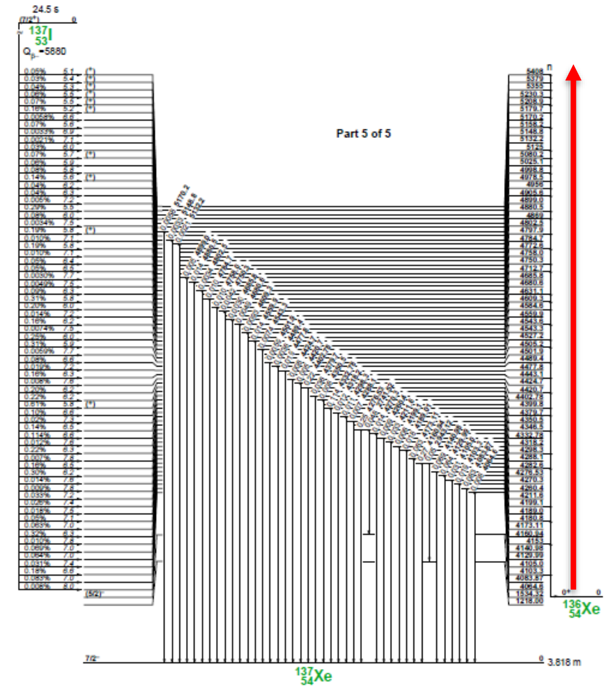




As the complexity of the decay increases the absolute intensities derived from Geant4 deviate from literature values (nuclear data).



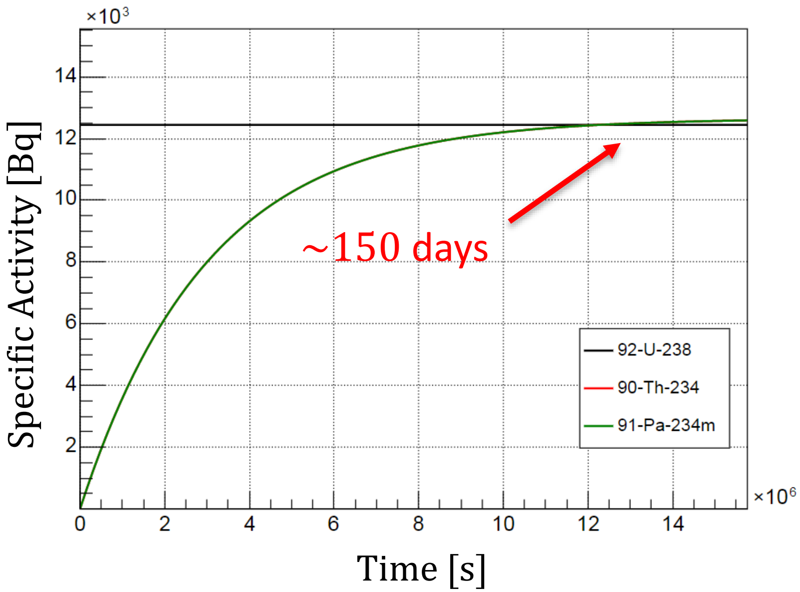




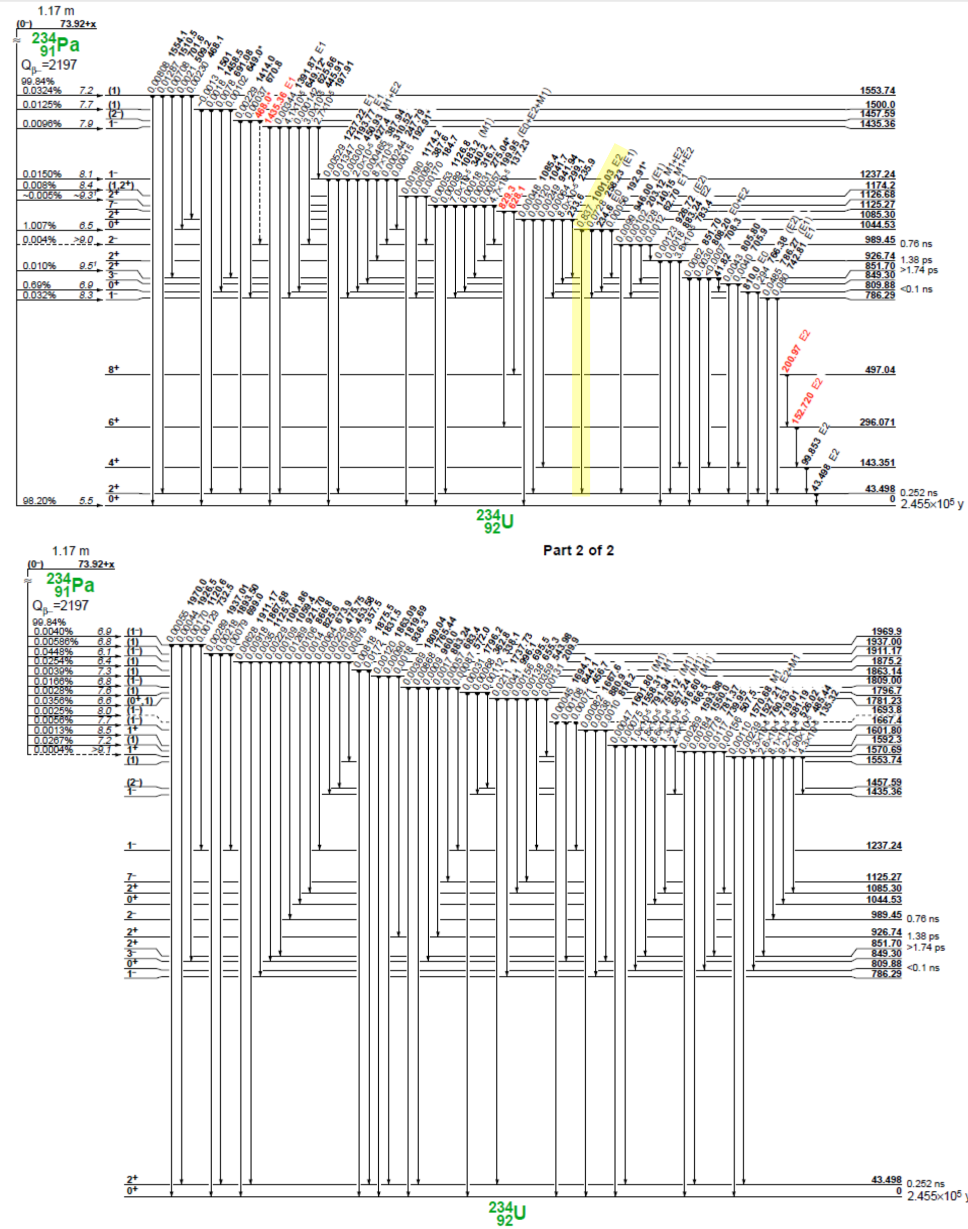
Data from: R. B. Firestone, C. M. Baglin & S. Y. F. Chu, *Table of Isotopes, 8th ed.* (Wiley, 1996).

Validation Work

Another important example in the detection of ^{238}U



The absolute intensity of the 1 MeV γ-ray from ^{234m}Pa decay was found to be 0.9125% using the Geant4 data libraries, which is approximately 10% larger than what is found in literature.



Data from: R. B. Firestone, C. M. Baglin & S. Y. F. Chu, *Table of Isotopes, 8th ed.* (Wiley, 1996).

Results

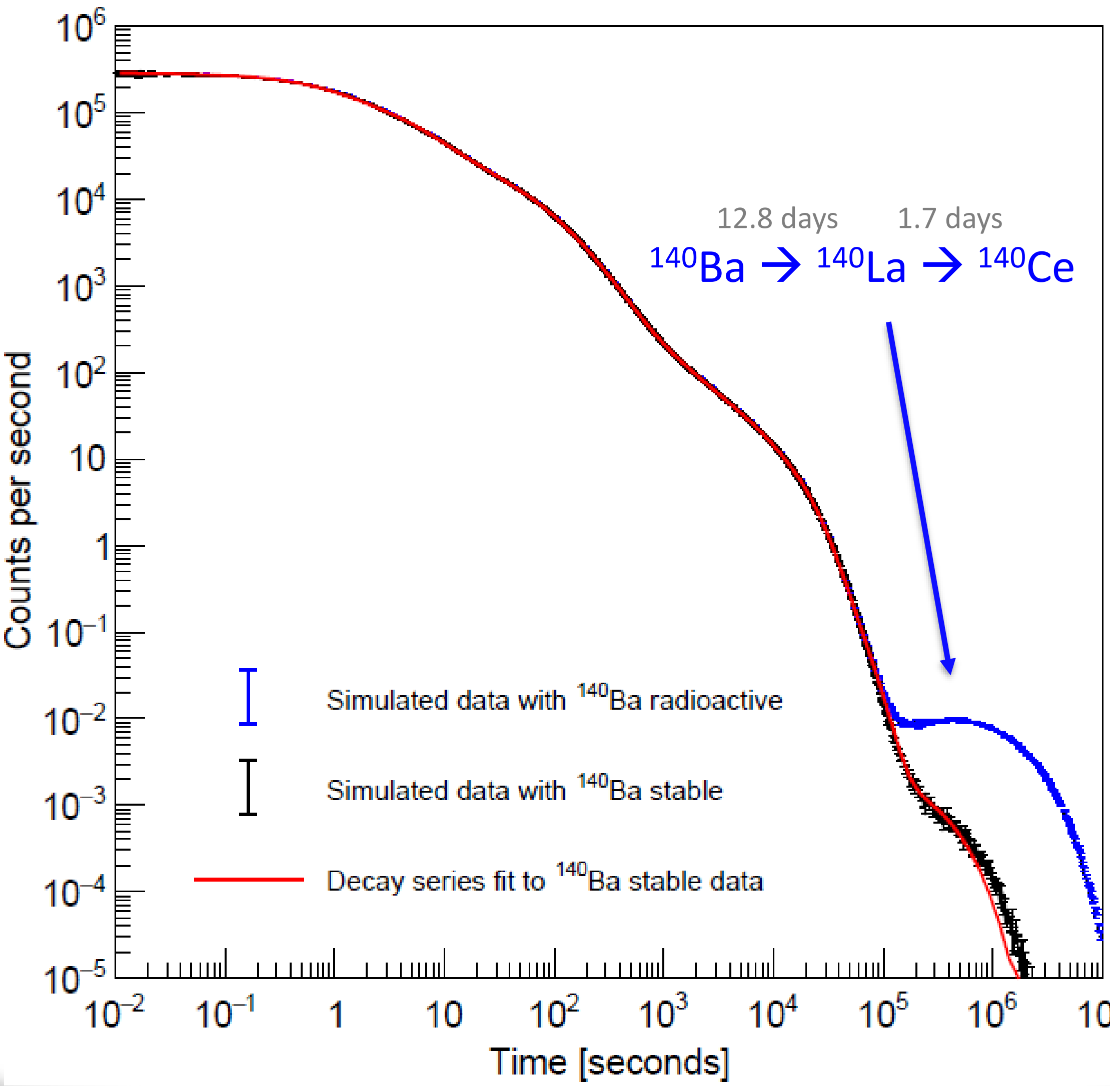
One billion $^{233,\ 235}U$ and ^{239}Pu fission fragment events were simulated. For **computational efficiency** the pn prod. curve was increased 145-fold. Thus, each simulation represented approximately 72.5 billion effective fission events.

Similar to past experimental work, data was fit to a series of exponential terms,

$$f(t) = \sum_{i=1}^{12} A_i e^{-\lambda_i t},$$

using ROOT and the MINUIT2 χ^2 minimization toolkit. Relative pn yields for each group was extracted:

$$\frac{\beta_{\gamma_i}}{\beta_\gamma} = \frac{A_i/\lambda_i}{\sum_{i=1}^{12} A_i/\lambda_i}.$$



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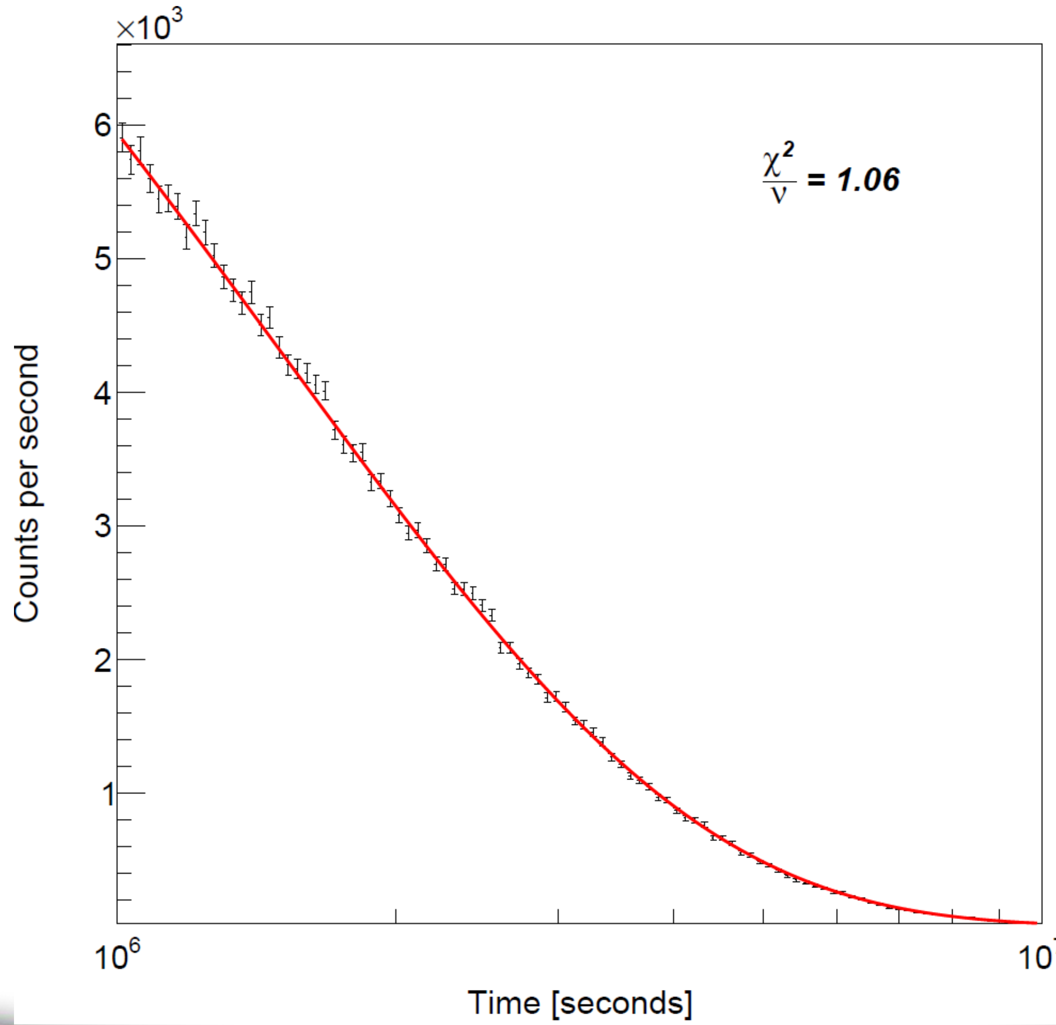
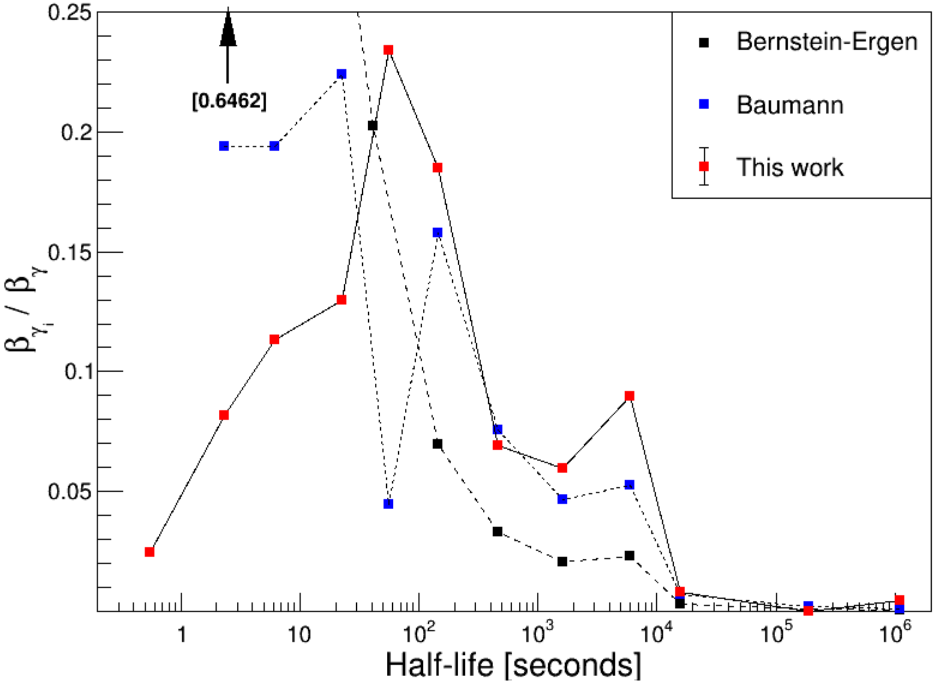


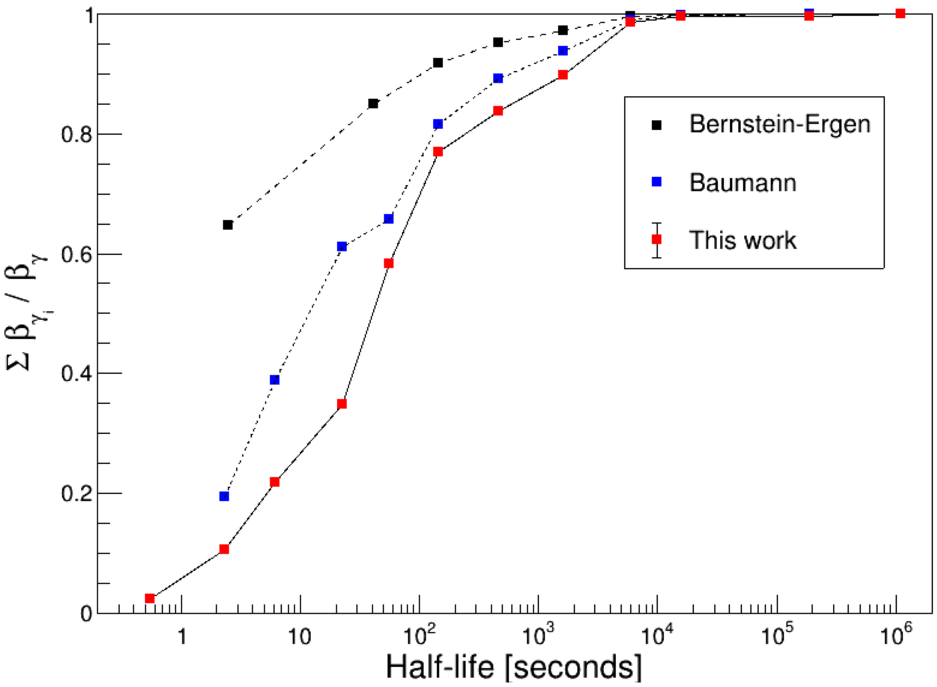
Results

Relative photoneutron group yields for $^{233, 235}$U and ^{239}Pu derived from simulation. The errors are purely statistical resulting from the χ^2 minimization fitting and do not include uncertainties persisting in the nuclear data.

Group (i)	Half-life (s)	$\beta_{\gamma_i}/\beta_\gamma$ ($\times 10^3$) ^{235}U	^{233}U	^{239}Pu	Baumann	Bernstein-Ergen
1	1107360	4.49 ± 0.03	5.27 ± 0.03	6.83 ± 0.04	1.1	0.5
2	190800	0.205 ± 0.002	0.261 ± 0.003	0.568 ± 0.005	2.3	1.0
3	15840	8.04 ± 0.03	12.75 ± 0.04	6.77 ± 0.04	7.3	3.2
4	5924	89.48 ± 0.08	128.2 ± 0.1	109.6 ± 0.1	52.7	23.2
5	1620	59.6 ± 0.1	65.7 ± 0.1	99.3 ± 0.2	46.6	20.5
6	462.1	69.0 ± 0.2	94.3 ± 0.2	80.5 ± 0.2	75.7	33.4
7	144.1	185.5 ± 0.3	202.7 ± 0.3	98.9 ± 0.4	157.6	69.5
8	55.7	234.4 ± 0.4	245.9 ± 0.4	263.0 ± 0.5	44.8	-
-	41	-	-	-	-	202.5
9	22.7	129.6 ± 0.3	115.9 ± 0.4	172.4 ± 0.5	223.9	-
10	6.22	113.3 ± 0.3	75.9 ± 0.3	61.2 ± 0.3	194.0	-
-	2.5	-	-	-	-	646.2
11	2.3	81.6 ± 0.2	44.6 ± 0.2	78.5 ± 0.2	194.0	-
12	0.548	24.81 ± 0.06	8.41 ± 0.05	22.48 ± 0.08	0	-

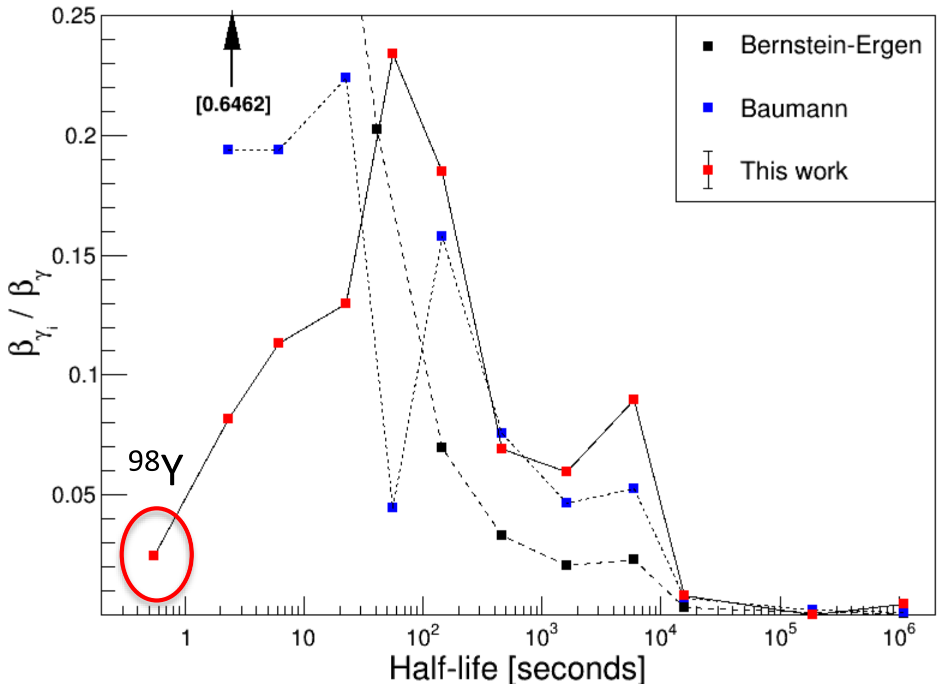
Results

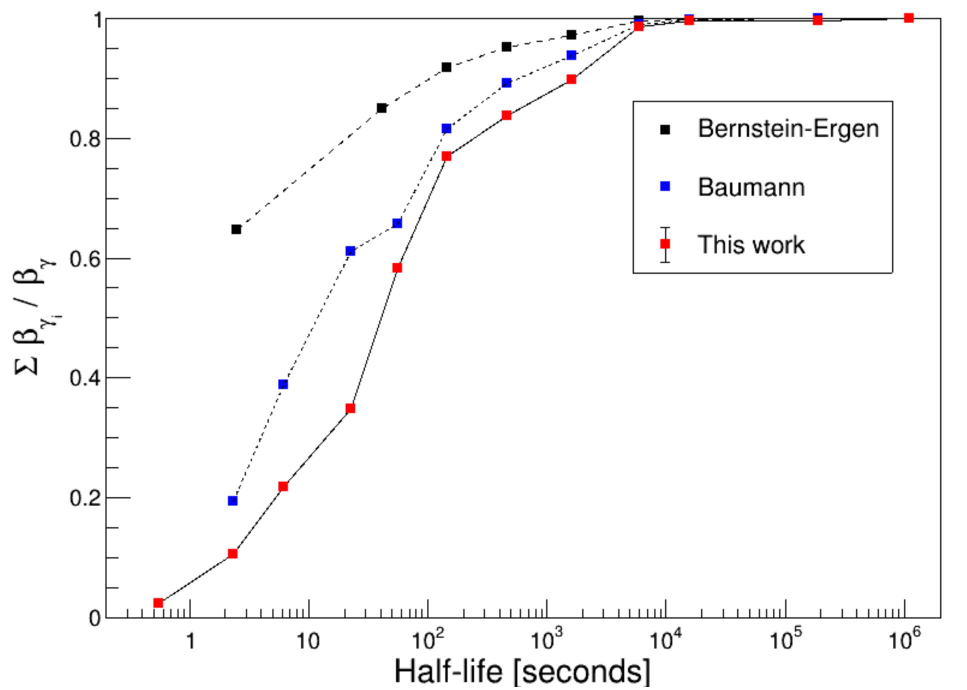




Relative photoneutron group yields (a) and running total (b) for the thermal fission of ^{235}U calculated using Geant4. The current recommended values based on the ^{235}U experimental work of Baumann and others are presented under the label “Baumann”.

Results





Relative photoneutron group yields (a) and running total (b) for the thermal fission of ^{235}U calculated using Geant4. The current recommended values based on the ^{235}U experimental work of Baumann and others are presented under the label "Baumann".

Conclusions

- Large discrepancies between the simulation results and the experimentally derived data were found, most significantly for the shortest and longest half-life groups.
- These results are **inconsistent** with the ("stitched together") experimentally derived group data.

For example, significantly more yield was found in simulation for the 55.7 sec half-life group. We can perform a sanity check!

Major contributor of this group: ^{91}Rb, using the

- Fission yield [JENDL-FPY 2011]
- Photoneutron production yield [MCNP6]
- γ-ray energies from nuclear data [Nuclear Data Sheets 114 (10) (2013)]
- Total neutron yield of $\bar{\nu} = 2.437 \pm 0.017$ [ENDF/B-VII.1]

We calculate a nuclide specific photoneutron yield of $\beta_{\gamma}^{^{91}\mathrm{Rb}} = (4.0 \pm 0.2) \times 10^{-5}$
Baumann reports 3×10^{-5} for the entire group, under-predicting its importance.
Other contributors with half-lives near 55.7 seconds: ^{86,87}Br, ^{139}Xe, ^{140}Cs, and ^{144}La

E.T. Rand, J.E. Atfield, L.R. Yaraskavitch, "*Microscopic calculation of delayed-photoneutron production in D_2O using Geant4*", Annals of Nuclear Energy **129**, 390-398 (2019)

Limitations and Future Improvement

1. **β-delayed neutron decay physics in Geant4.**
   - Current status? Nuclear data driven or model driven?

2. **Large discrepancies are observed in (γ, n) interactions with nuclei < 30 MeV. Cross section on deuterium was found to be zero, thus we ported MCNP6 physics into our simulation.**
   - A nuclear data driven photonuclear reaction model for Geant4 was developed by others (J. W. Shin, Nucl Instrum Methods Phys Res B 358, 2015).
   - Possibility to validate and incorporate this reaction model?

3. **Absolute γ-ray yields from the Geant4 radioactive process deviate from literature values for many isotopes.**
   - There is no “easy fix”, left to the users to verify and check.
   - Using the per-decay sampling approach has its advantages (physically correct, event particle coincidences, angular correlations, etc.) However, there exists a community of users who only need to reproduce absolute γ-ray intensities. Currently users can hack radioactive decay and photonevaporation data files, write their own generator, or use GPS.
   - A supported and simplistic γ-decay model would meet the needs of these users and help identify issues in the radioactive decay module.

Thank you!

Questions?

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Canadian Nuclear Laboratories
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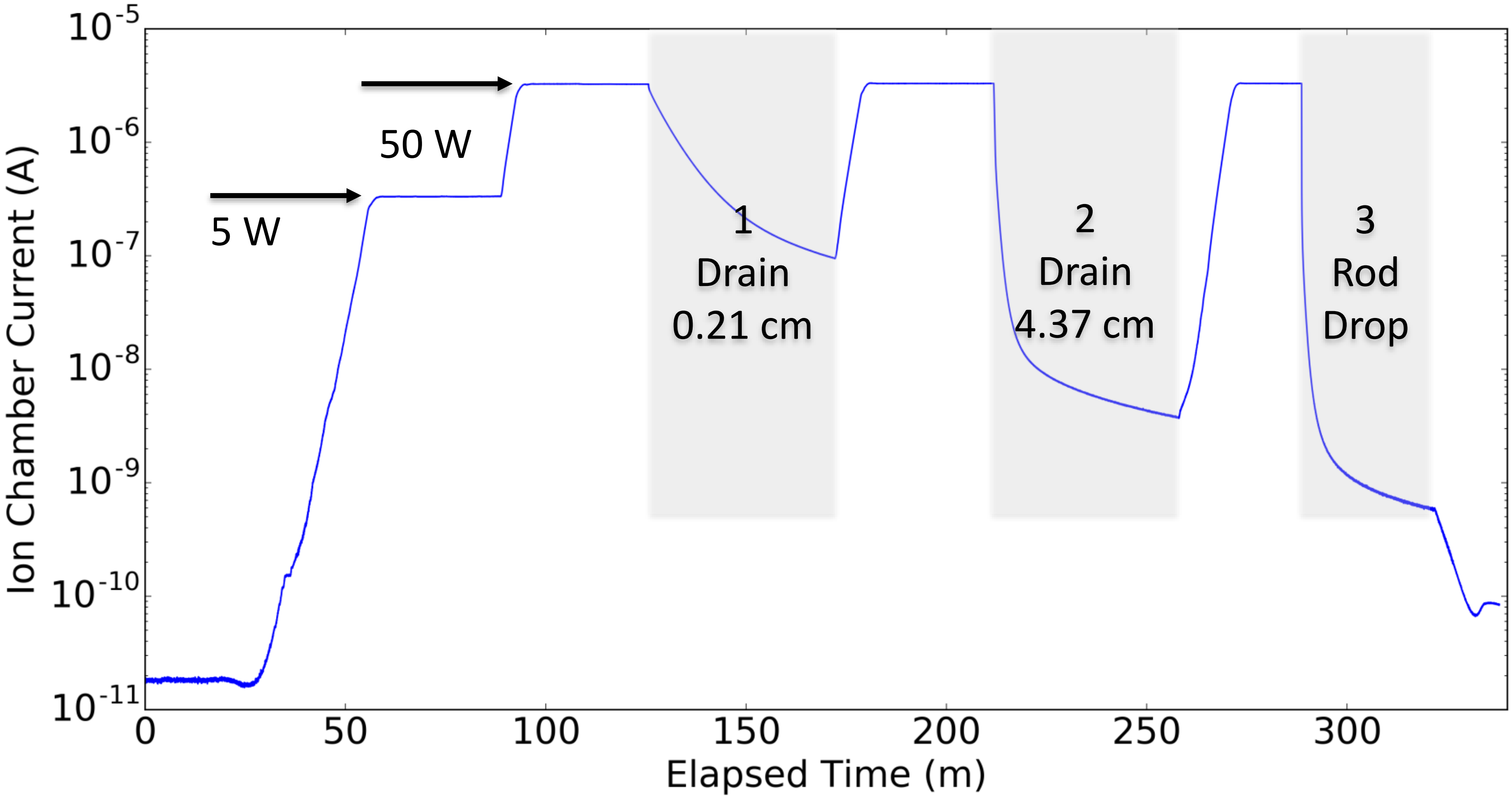
Backup Slides

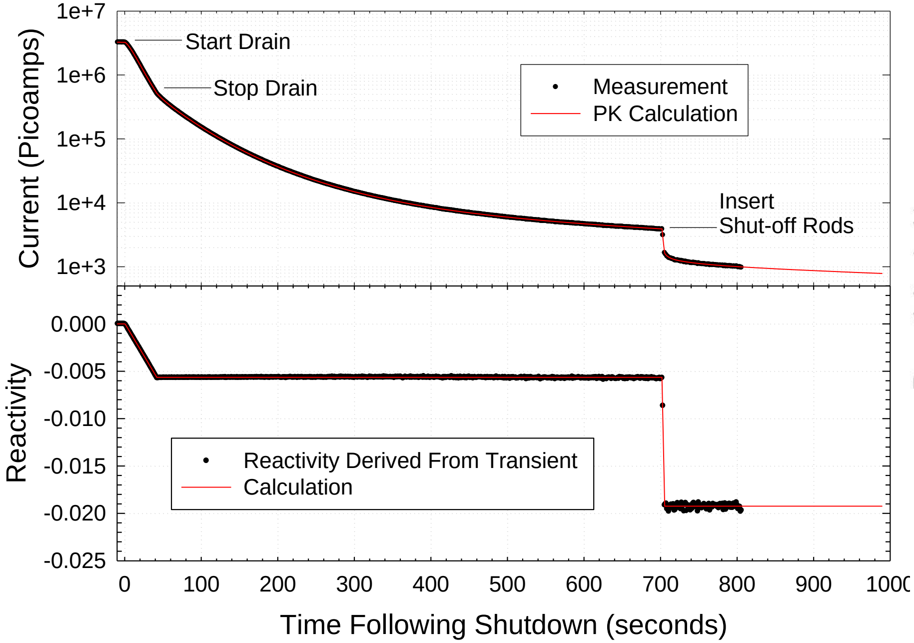
Pu-Th Substitution Core

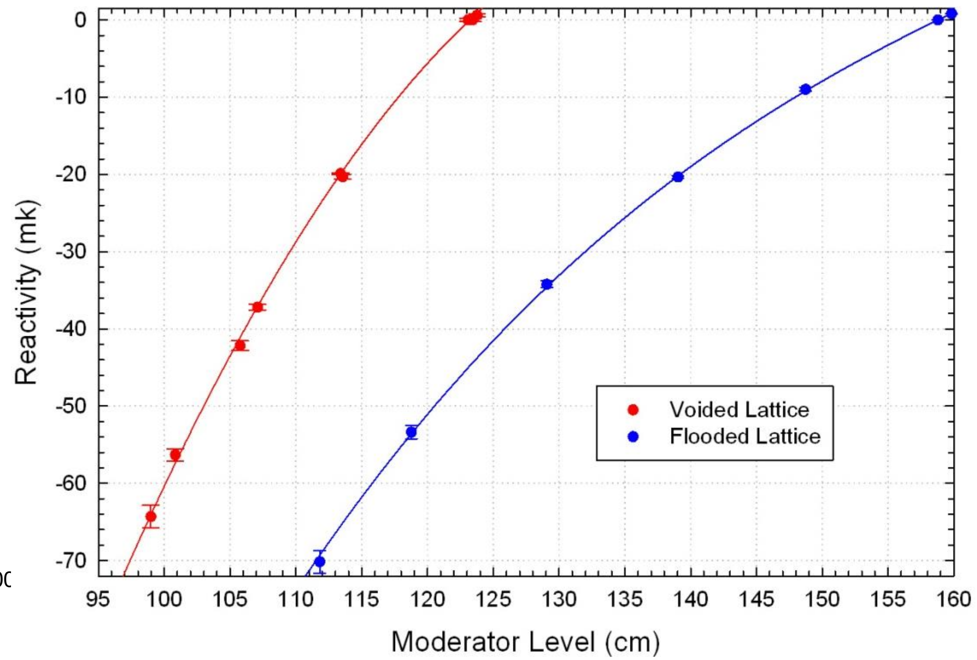
- Pu-Th Test Core



Existing Capabilities

- Direct Measurement of Reactivity Effect on Voiding





Zeller and Atfield, "Transient experiments in ZED-2 to investigate the impact of leakage on reactor physics phenomena", AECL Nuclear Review, 4 (2015) 67-73.

Other recent work: Atfield, Yaraskavitch, and Rand, "Kinetics experiments in ZED-2 using heterogeneous cores of advanced nuclear fuels", Annals of Nuclear Energy, 121 (2018) 36-49.