Continuing Lecture on Neutron Multiplicity Analysis

THE UNIVERSITY OF
TENNESSEE
KNOXVILLE

Remaining Steps

- Detection Efficiency ε
- Neutron moments for spontaneous fission
- Neutron moments for (α,n) reactions
- Combined neutron moment equations

Second Advantage of PGFs

- First useful property is the ability to write out explicit moment equations
- Second is the composability of PGFs with another
- Say we have a PGF for a process $B(s)$ and want to modify it with a second process $C(s)$ that occurs after $B(s)$
  - Combined PGF is simply $B\big(C(s)\big)$
  - For a starting process $A(s)$, the combined process is $A\big(B(s)\big)$
  - For process A, then B, then C, we have PGF $A\Big(B\big(C(s)\big)\Big)$

Detection Efficiency

- Assuming one neutron has probability ε of being detected, the PGF for one neutron is the coin flip PGF
  - $\eta(s) = (1-\epsilon) + s\,\epsilon$
- The detection process occurs *after* neutrons are emitted from a fission chain, so the PGF of detecting fission chain neutrons is $h\big(\eta(s)\big)$
- Writing out moments is just the chain rule
  - $\phi_{1,detect} = \left.\frac{d\,h(\eta(s))}{ds}\right|_{s=1} = \Big(h'\big(\eta(s)\big)\eta'(s)\Big)_{s=1} = \epsilon\phi_1$
  - $\phi_{2,detect} = \left.\frac{d^2\,h(\eta(s))}{ds^2}\right|_{s=1} = \Big(h''\big(\eta(s)\big)\big(\eta'(s)\big)^2\Big)_{s=1} + \cdots = \epsilon^2\phi_2$
  - $\phi_{3,detect} = \left.\frac{d^3\,h(\eta(s))}{ds^3}\right|_{s=1} = \Big(h'''\big(\eta(s)\big)\big(\eta'(s)\big)^3\Big)_{s=1} + \cdots = \epsilon^3\phi_3$
- ☑

Spontaneous Fission

- Now instead of starting with one neutron, we start with a spontaneous fission which has a separate neutron multiplicity distribution
- Using $f_i(s)$ as the induced neutron PGF and $f_{sf}(s)$ for spontaneous, the Bohnel equation becomes
  - $h_{sf}(s) = f_{sf}\big(h_i(s)\big) = f_{sf}\left((1-p_F)s + f_i\big(h_i(s)\big)\right)$
- Writing out moments is just the chain rule (again)
  - $\boldsymbol{\phi_{1,sf}} = \left.\frac{df_{sf}(h_i(s))}{ds}\right|_{s=1} = \left(\frac{df_{sf}(h_i(s))}{dh_i(s)}\frac{dh_i(s)}{ds}\right)_{s=1} = \boldsymbol{\nu_{sf,1}\,\phi_{1,i}}$
  - $\boldsymbol{\phi_{2,sf}} = \left.\frac{d^2f_{sf}(h_i(s))}{ds^2}\right|_{s=1} = \left(\frac{d^2f_{sf}(h_i(s))}{dh_i(s)^2}\left(\frac{dh_i(s)}{ds}\right)^2\right)_{s=1} + \cdots = \boldsymbol{\nu_{sf,2}\,\phi_{1,i}^2 + \nu_{sf,1}\phi_{2,i}}$
  - $\boldsymbol{\phi_{3,sf}} = \left.\frac{d^3f_{sf}(h_i(s))}{ds^3}\right|_{s=1} = \left(\frac{d^3f_{sf}(h_i(s))}{dh_i(s)^3}\left(\frac{dh_i(s)}{ds}\right)^3\right)_{s=1} + \cdots$
    $= \boldsymbol{\nu_{sf,3}\,\phi_{1,i}^3 + 3\,\nu_{sf,2}\phi_{2,i}\phi_{1,i} + \nu_{sf,1}\phi_{3,i}}$
- ☑

S,D,T of Detected Neutrons from Spontaneous Fission

- $S_{sf} = Q_{sf}\epsilon \cdot \phi_{1,sf}$

$$= Q_{sf}\epsilon \cdot \nu_{sf,1}\ \phi_{1,i}$$

- $D_{sf} = Q_{sf}\frac{\epsilon^2}{2} f_D \cdot \phi_{2,sf}$

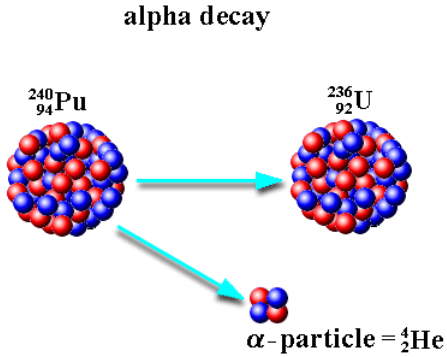
$$= Q_{sf}\frac{\epsilon^2}{2} f_D \cdot \left(\nu_{sf,2}\ \phi_{1,i}^2 + \nu_{sf,1}\phi_{2,i}\right)$$

- $T_{sf} = Q_{sf}\frac{\epsilon^3}{6} f_T \cdot \phi_{3,sf}$

$$= Q_{sf}\frac{\epsilon^3}{6} f_T\left(\nu_{sf,3}\ \phi_{1,i}^3 + 3\ \nu_{sf,2}\phi_{2,i}\phi_{1,i} + \nu_{sf,1}\phi_{3,i}\right)$$

S,D,T of Detected Neutrons from (α,n) Reactions

- $S_\alpha = Q_\alpha \epsilon \cdot \phi_{1,i}$
- $D_\alpha = Q_\alpha \frac{\epsilon^2}{2} f_D \cdot \phi_{2,i}$
- $T_\alpha = Q_\alpha \frac{\epsilon^3}{6} f_T \cdot \phi_{3,i}$



$$\alpha + {}^{18}O \rightarrow {}^{21}Ne + n$$

$$\alpha + {}^{19}F \rightarrow {}^{22}Na + n$$

- Instead of using Q_α as the source strength, it is convention to use the ratio $\boldsymbol{\alpha}$ of the number of neutrons produced from (α,n) to spontaneous fission

$$\boldsymbol{\alpha} = \frac{Q_\alpha}{Q_{sf} \nu_{1,sf}}$$

- Using $Q_\alpha = \boldsymbol{\alpha}\, Q_{sf} \nu_{1,sf}$
- $S_\alpha = \boldsymbol{\alpha}\, Q_{sf}\, \epsilon \cdot \phi_{1,i}$
- $D_\alpha = \boldsymbol{\alpha}\, Q_{sf}\, \frac{\epsilon^2}{2} f_D \cdot \phi_{2,i}$
- $T_\alpha = \boldsymbol{\alpha}\, Q_{sf} \frac{\epsilon^3}{6} f_T \cdot \phi_{3,i}$

S,D,T For Plutonium

- $S = S_{sf} + S_{\alpha} = Q_{sf} \epsilon \, \phi_{1,i} \, (1 + \boldsymbol{\alpha})$
- $D = D_{sf} + D_{\alpha} =$

$$Q_{sf} \frac{\epsilon^2}{2} f_D \cdot \left(\nu_{sf,2} \, \phi_{1,i}^2 + (1 + \boldsymbol{\alpha}) \nu_{sf,1} \phi_{2,i} \right)$$

- $T = T_{sf} + T_{\alpha} =$

$$Q_{sf} \frac{\epsilon^3}{6} f_T \left(\nu_{sf,3} \, \phi_{1,i}^3 + 3 \, \nu_{sf,2} \phi_{2,i} \phi_{1,i} + (1 + \boldsymbol{\alpha}) \nu_{sf,1} \phi_{3,i} \right)$$

- Using previously derived …

$$\phi_1 = \mathbf{M_L}$$

$$\phi_2 = \mathbf{M_L}^2 \left[\nu_2 \left(\frac{\mathbf{M_L} - 1}{\nu_1 - 1} \right) \right]$$

$$\phi_3 = \mathbf{M_L}^3 \left[3\nu_2^2 \left(\frac{\mathbf{M_L} - 1}{\nu_1 - 1} \right)^2 + \nu_3 \left(\frac{\mathbf{M_L} - 1}{\nu_1 - 1} \right) \right]$$

S,D,T For Passive Plutonium Assay

- We finally arrive at the S,D,T equations

$$S \equiv C_1 = Q_{sf}\varepsilon_n M_L \nu_{sf,1}(1+\alpha) \ ,$$

$$D \equiv C_2 = \frac{Q_{sf}\varepsilon_n^2 M_L^2}{2}\left[\nu_{sf,2} + \left(\frac{M_L - 1}{\nu_{if,1} - 1}\right)\nu_{sf,1}(1+\alpha)\nu_{if,2}\right] \ ,$$

and

$$T \equiv C_3 = \frac{Q_{sf}\varepsilon_n^3 M_L^3}{6}\left\{\nu_{sf,3} + \left(\frac{M_L - 1}{\nu_{if,1} - 1}\right)\left[3\nu_{sf,2}\nu_{if,2} + \nu_{sf,1}(1+\alpha)\nu_{if,3}\right] + 3\left(\frac{M_L - 1}{\nu_{if,1} - 1}\right)^2 \nu_{sf,1}(1+\alpha)\nu_{if,2}^2\right\} \ ,$$

- Having estimates of (S, D, T) allows using the above equations to solve for (Q_{sf}, M_L, α)
- $Q_{sf} = m_{eff}^{240}\, Y^{240}$
- $Y^{240} = 473.5$ spontaneous fissions per second per gram
- Solving for Q_{sf} allows an estimate of m_{eff}^{240}

Modification for Active Assay of Uranium

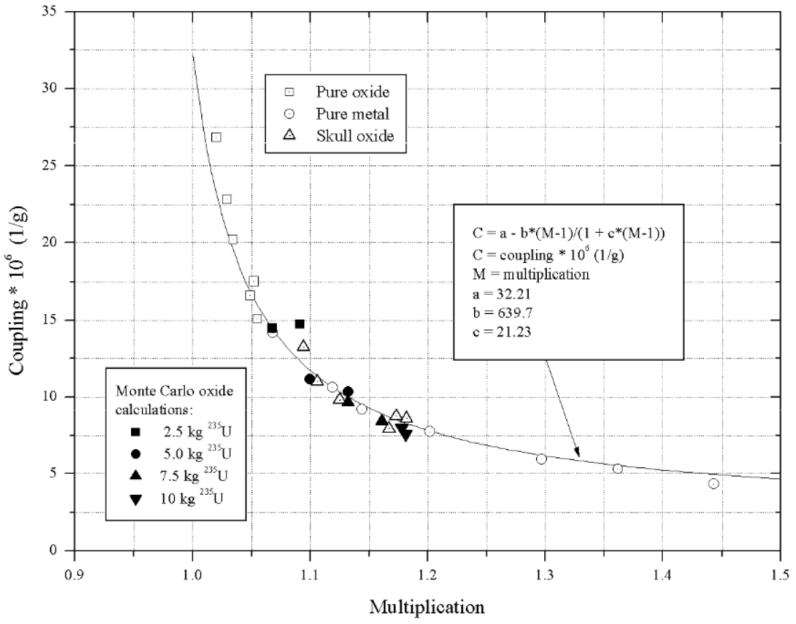
- For uranium and other materials with a negligible spontaneous fission rate, an external source of neutrons Y_s is required

$$Q_{active} = C\ Y\ m_{eff}^{235}$$

- Where C is a coupling term of the fraction of source neutrons which induce fission in the material
- ⚠ Because C and m_{eff}^{235} appear together, it is not possible to solve for m_{eff}^{235} alone
- However, the ratio T/D point model equations are dependent on M_L alone. Y^{240}

Modification for Active Assay of Uranium

- For getting m_{eff}^{235}, need to determine C
- One approach:
  - Use a heuristic functional form of C and perform measurements to fit
  - $C = a - \frac{b(M_L - 1)}{1 + c(M_L - 1)}$



Russo, P. *PASSIVE NONDESTRUCTIVE ASSAY OF NUCLEAR MATERIALS 2007 ADDENDUM*. (2007). Chapter 7

Modification for Active Assay of Uranium

- Second approach,

$$C = k\,\phi(\Omega)\,\psi(M_L)$$

  k = scaling factor
  $\phi(\Omega)$ is a function of sample geometry and source position
  $\psi(M_L)$ is reduction of source flux in sample
- Relationships can be determined through MC simulation
- Finally, using T/D to determine M_L and previous knowledge of C

$$m_{eff}^{235} = \frac{Q_{active}}{C\,Y_S}$$

Extensions, Limitations, and Alternatives of Point Model,

THE UNIVERSITY OF
TENNESSEE
KNOXVILLE

Point Model Extensions

- Add additional reactions such as neutron capture

$$h(s) = (1 - p_F - p_C)s + p_F f\big(h(s)\big)$$

- Matthew Tweardy (2018) developed a point model for uranium interrogated by a D-T 14.1 neutron source

$$h_f(s) = (1 - \beta)s + Q_S\big(h(s)\big)$$

β is coupling of generator to material
Q_S contains several reactions such as (n,2n) and (n,3n) present at high neutron energies

- Tony Shin extended the PGF of neutron detection for scatter based neutron detection systems, which may count the same neutron once, two, or three times with probabilities ϵ_0, ϵ_1, ϵ_2

$$\eta(s) = (1 - \epsilon_0) + \epsilon_0(1 - \epsilon_1)s + \epsilon_0\epsilon_1(1 - \epsilon_2)s^2$$

- Liquid scintillator detectors can detect gammas as well, which are emitted during each fission. A joint PGF of neutrons and gammas can be written as

$$u(s,t) = (1 - p_F)s + p_f m(t) f\big(u(s,t)\big)$$

Where $m(t)$ is the multiplicity distribution of fission gammas. The joint moments can then be analyzed

$$\phi_{i,j} = \frac{1}{i!\,j!}\left(\frac{\partial^i}{\partial s^i}\frac{\partial^j}{\partial s^j}u(s,t)\right)_{s=1,t=1}$$

Point Model Limitations

- The Bohnel equation

$$h(s) = (1 - p_F)s + p_F f\big(h(s)\big)$$

makes several assumptions and simplifications to describe fission chains

- Time: Assumes all neutrons in a fission chain emitted simultaneously at once
- Energy: All neutrons have one averaged energy, additionally detector efficiency is averaged over energy
- Space: The probability of a neutron to induce fission p_F is independent of the object geometry and where the fission occurs
- Data: The material dependent coefficients of the multiplicity distribution $f(s)$ and the moments ν_1, ν_2, ν_3 are known.

Time

- LLNL has developed point model extensions which include the time dependence of neutron population, which can be measured using fast liquid scintillator based detectors

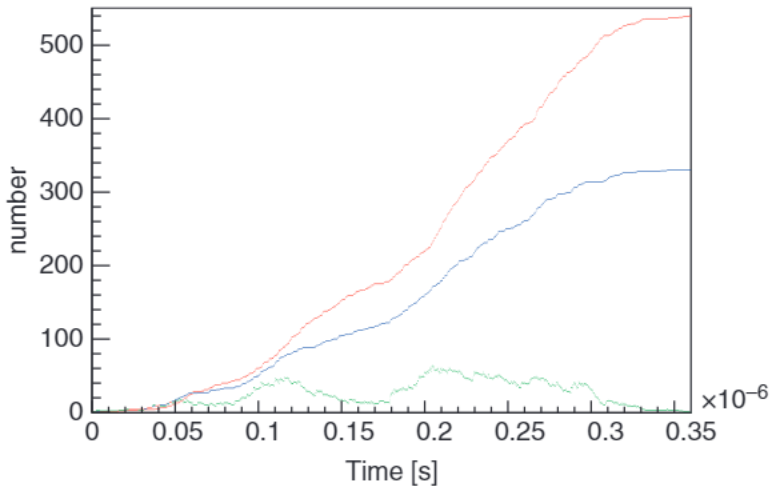


Fig. 9. Time evolution of the correlated fission chain populations for a single simulated fission chain from $M = 10$ HEU, starting from a single neutron; internal neutrons (green), accumulation of induced fissions (blue), and accumulation of leaked neutrons (red).

Kim, K. S., Nakae, L. F., Prasad, M. K., Snyderman, N. J. & Verbeke, J. M. Time Evolving Fission Chain Theory and Fast Neutron and Gamma-Ray Counting Distributions. *Nucl Sci Eng* **181**, 225–271 (2015).

Appendix B. Here, we give the results for the first three combinatorial moments of the internal neutron population:

$$
\begin{cases}
\left.\dfrac{\partial f}{\partial x}\right|_{x=1} = \displaystyle\sum_{i=1}^{\infty} i P_i^{int}(t) = e^{-\alpha t} \\
\left.\dfrac{1}{2!}\dfrac{\partial^2 f}{\partial x^2}\right|_{x=1} = \displaystyle\sum_{i=2}^{\infty} \dfrac{i(i-1)}{2} P_i^{int}(t) \\
\qquad = \dfrac{M-1}{\bar{\nu}} \nu_2 e^{-\alpha t}(1 - e^{-\alpha t}) \\
\left.\dfrac{1}{3!}\dfrac{\partial^3 f}{\partial x^3}\right|_{x=1} = \displaystyle\sum_{i=3}^{\infty} \dfrac{i(i-1)(i-2)}{3!} P_i^{int}(t) \\
\qquad = \left(\dfrac{M-1}{\bar{\nu}} \nu_2\right)^2 e^{-\alpha t}(1 - 2e^{-\alpha t} + e^{-2\alpha t}) \\
\qquad + \dfrac{1}{2}\dfrac{M-1}{\bar{\nu}} \nu_3 e^{-\alpha t}(1 - e^{-2\alpha t}) \; . \qquad (49)
\end{cases}
$$

Starting from a single neutron at $t = 0$, the first moment

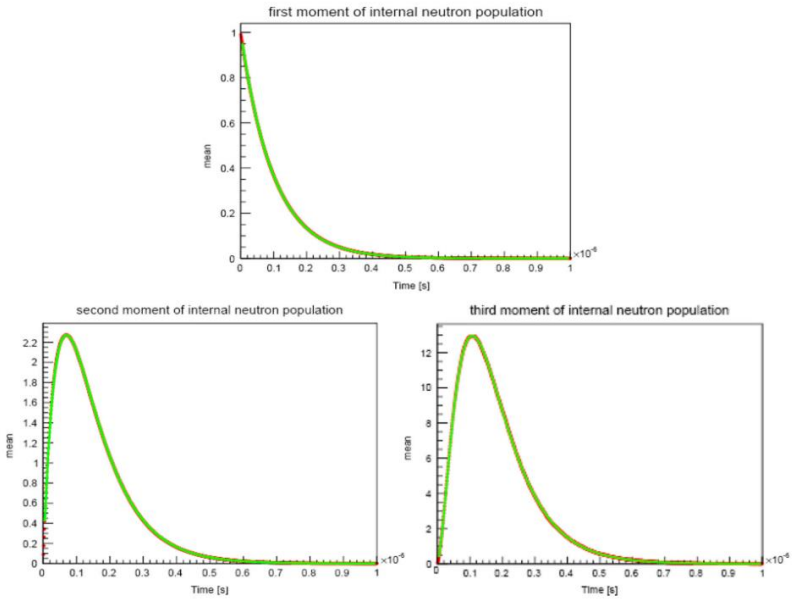


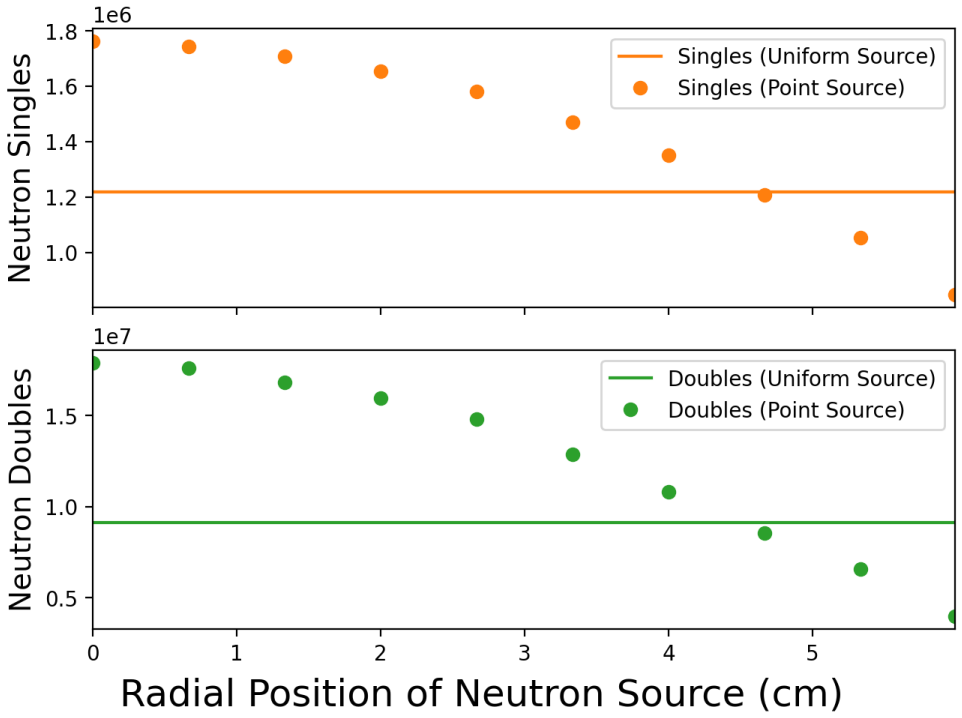
Fig. 10. Time evolution of the first three moments of the internal neutron population. The formulas of Eq. (49) are green, and the simulation results are red. The fission chain Monte Carlo data from which the moments are computed are from Fig. 4.

Spatial Dependence of Neutron Moments

The multiplication of a subcritical assembly depends, of course, on the position in the active material at which the source of neutrons is introduced.

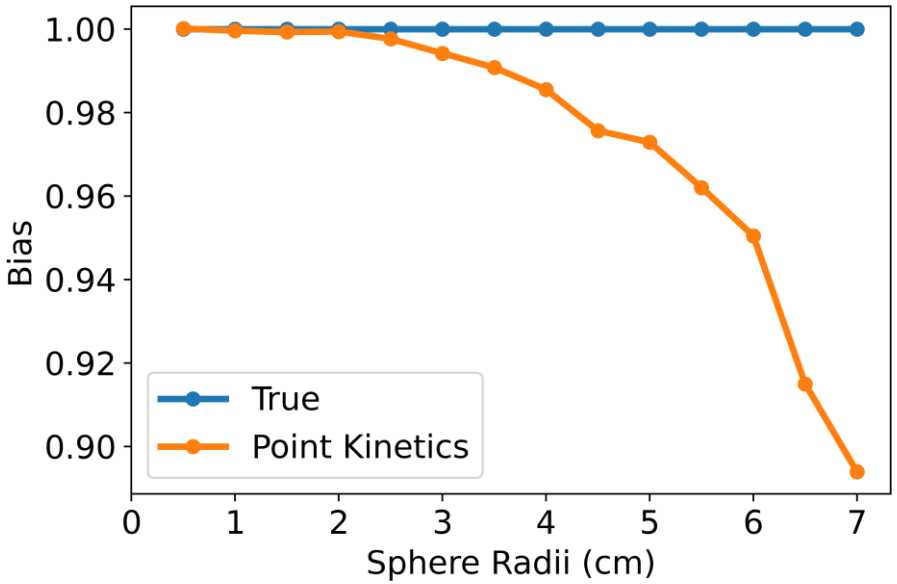
THE DEFINITION OF "NEUTRON MULTIPLICATION" R. Serber July 25, 1945

- Bias of “point” assumption observed in 1945, well before point kinetics
- Effect is clear using MCNP simulations by moving a neutron source within a 6 cm radius sphere of U235
- Variations of neutron singles and doubles compared to uniform source
- **Leads to underestimation of # starting events (F) and enrichment**



Bias of Point Kinetics

- Use MCNP to simulate number of expected neutron singles and doubles for spheres of U235 with varying radii
  - Source is uniform distribution of fission energy neutrons
- Use point kinetics to solve for number of starting events, F
- Results show bias of point kinetic model with increasing radii
  - Underestimation of F, assigning inspected materials with a lower enrichment
- As geometry increases in size, assumption of spatial uniformity breaks down and bias increases



Approaches for Relaxing Point Assumption

- Weighted Point Model (Geist, Kirck, Mayo (2004), LA-UR-04-0570)
  - Estimates spatial variation of p_f by measuring higher neutron moments (quads, quints, etc)
  - Not dependent on geometry, but requires measuring very difficult observables due to efficiency
- Göttsche, Kirchner (2015, doi:10.1016/j.nima.2015.07.007) adds correction factors g_n to point kinetic moments
  - g_n are determined beforehand for specific geometries in analytic calculation
  - Determined only if geometry **and** composition are known beforehand
- Pázsit, Pál (2021, doi:10.1016/j.anucene.2020.108119) have developed fission chain theory with spatial dependence at the start for an arbitrary **known** geometry and composition, can calculate moments of large objects, but again relies on analytic calculation of known geometry and composition
- Both methods require symmetric geometry (sphere, cylinder, etc)

$$g(z|\boldsymbol{r},\boldsymbol{\Omega}) = z e^{-\ell(\boldsymbol{r},\boldsymbol{\Omega})} + \int_0^{\ell(\boldsymbol{r},\boldsymbol{\Omega})} \mathrm{d}s\, e^{-s}\, q_r[g(z|\boldsymbol{r}\prime(s))]$$

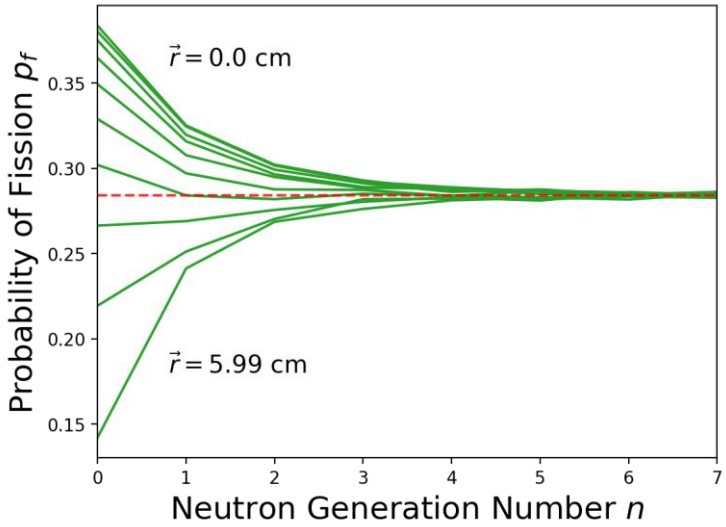
$$g(z|\boldsymbol{r}) = z g_0(z|\boldsymbol{r}) + \frac{1}{4\pi}\int_{4\pi} \mathrm{d}\boldsymbol{\Omega} \int_0^{\ell(\boldsymbol{r},\boldsymbol{\Omega})} \mathrm{d}s\, e^{-s}\, q_r[g(z|\boldsymbol{r}\prime)]$$

TRANSITION TO SETH MCCONCHIE'S MATERIALS

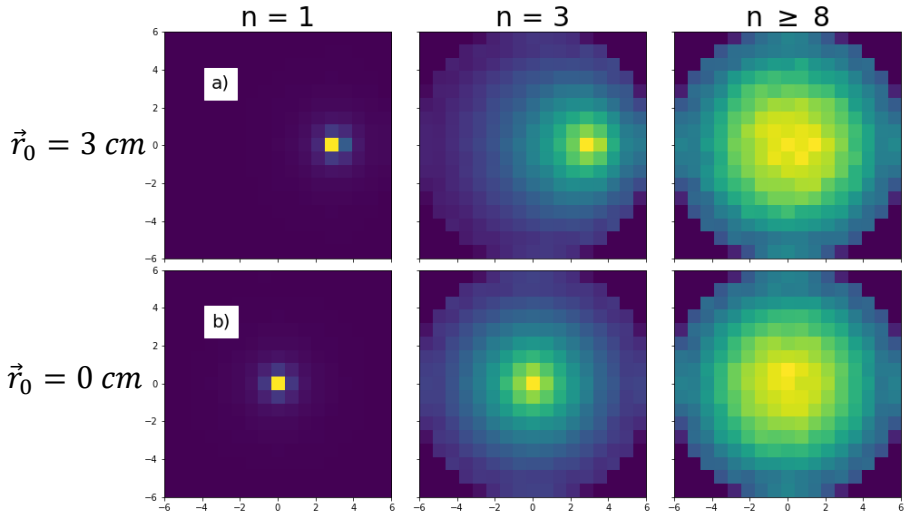
Factors of Spatial Dependence on API Images for Neutrons

Analysis of Monte Carlo simulations show two effects which violate point model in large geometries
- Left: The variation of p_F with both source position and the collision number of the neutron population
Shown output plots are for a pure U235 6 cm radius sphere

$p_f(\mathrm{n}|\vec{r}_0)$ vs n For Varying $\vec{r}_0$

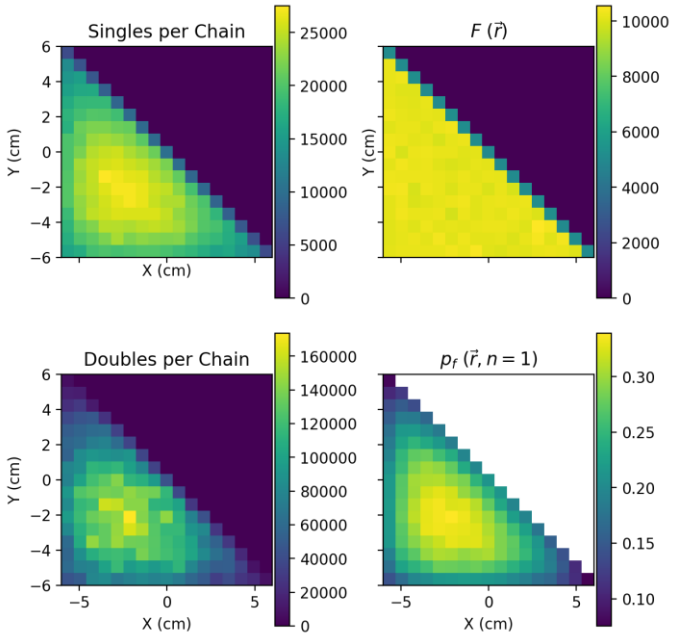


$\Psi(\vec{r}, n|\vec{r}_0)$ vs n For Varying $\vec{r}_0$

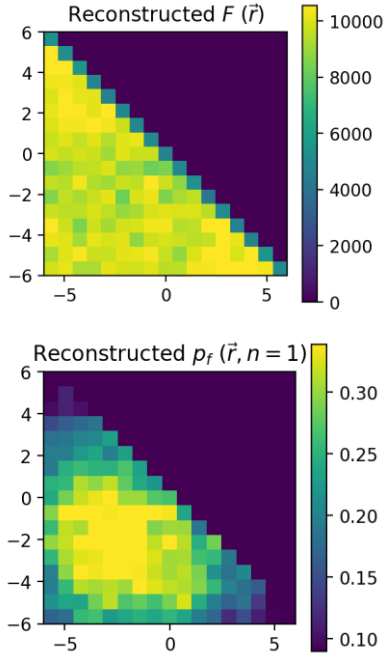


Results - Square Wedge of U235

MCNP (Simulation)



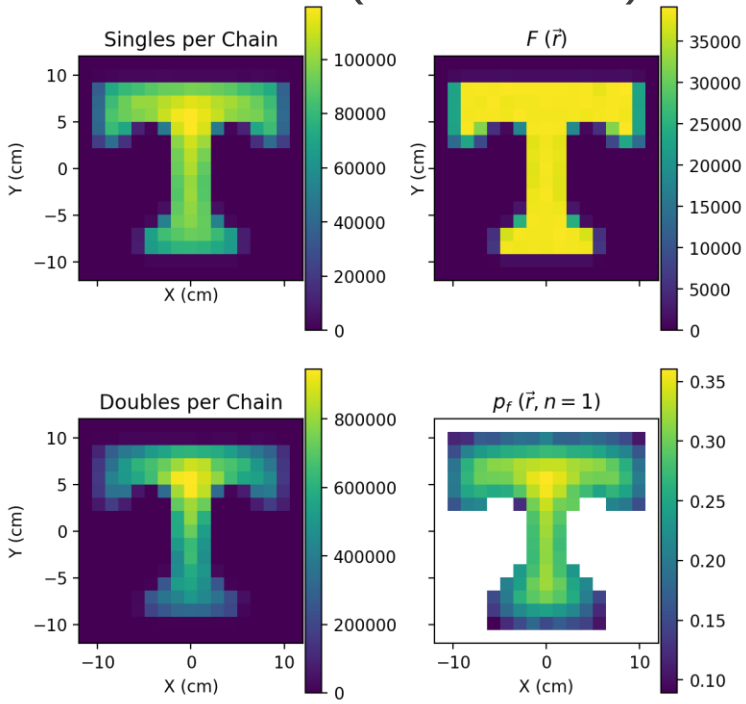
VFP Model



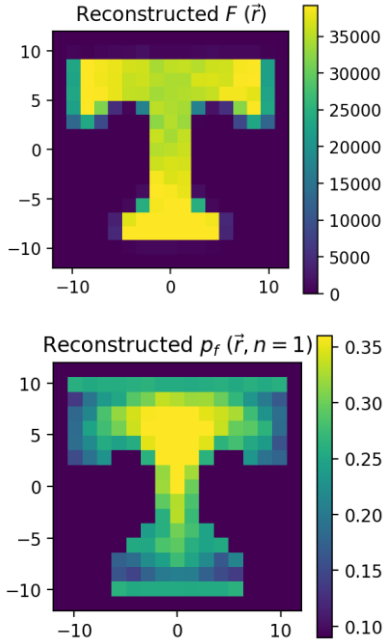
- 6x6x6cm cube of U235 cut on diagonal
- True: F = 2.5e7
- Point Kinetics: F = 2.37e7, %Error = -5.2 %
- VFP Model: F = 2.49e7, %Error = -0.4 %

Results - UTK “Power T” Logo

MCNP (Simulation)



VFP Model



- 20cm diameter, 10cm depth of U235 UTK logo
- True: F = 2.5e7
- Point Kinetics: F = 2.37e7, %Error = -5.2 %
- VFP Model: F = 2.45e7, %Error = -2.0 %