LA-UR-14-26562

The Importance of Nuclear Fission Data to Various Applications

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This lecture is based on my (and my colleagues') applied experiences

Topics to be covered :

(1) Time scales for super-critical systems.

(2) Initiation issues (very briefly).

(3) Neutron multiplicities and energy spectra for well counter design and simulation (used for nuclear safeguards).

(4) Prompt gamma-ray multiplicities and energy spectra including multiplicity-energy correlations, and beta-delayed fission neutrons for second line of defense and border security.

(5) Fission fragment yield curves and UGT yield assessments.

(6) Total kinetic energy (TKE) versus inducing neutron energy for (n,*f*) up to 20 MeV (Energy release).

(7) n-n correlations (just for the fun of it).

(8) Fission-fragment-neutron correlations for accurate determination of Prompt-Fission Neutron-Spectra (PFNS) at some accelerator based experiments.

(9) Nuclear explosions used to get nuclear physics data.

I will not discuss nuclear reactor applications.

Nuclear fission is a complex process

Neutron induced binary fission

$$\sigma_f(\varepsilon_\mathrm{n}), Y_{\mathrm{pre,post,delayed}}(Z, A, \varepsilon_\mathrm{n}), \mathrm{TKE}_{\mathrm{pre,post}}(\varepsilon_\mathrm{n}, A), \sigma_{TKE}(\varepsilon_\mathrm{n}, A),$$

$$P_\nu(\varepsilon_\mathrm{n}, A, \mathrm{TKE}), \mathrm{PFNS}(E_\mathrm{n}, \varepsilon_\mathrm{n}, \nu, A, \mathrm{TKE}, \theta_{\mathrm{LF}}),$$

similar quantities for γ–ray emission, n-n correlations, correlations between neutrons and γ–rays, the number and time scales of β–delayed neutron emission, etc…

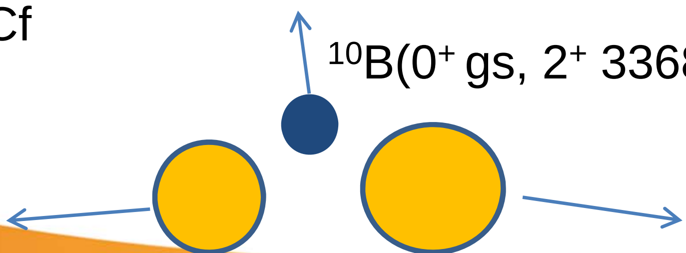
Similar quantities for spontaneous fission

Similar and additional quantities for ternary and quaternary fission for both SF and (*n*,f).
Gamma-ray and charged particle induced fission.
At high excitation energies binary fission is associated with p, α, d, t, ... emission.

There is a very large number of fission properties that can be measured.

As an example :

^{10}B is generated as a third fragment once in every 60,000 spontaneous fissions of ^{252}Cf



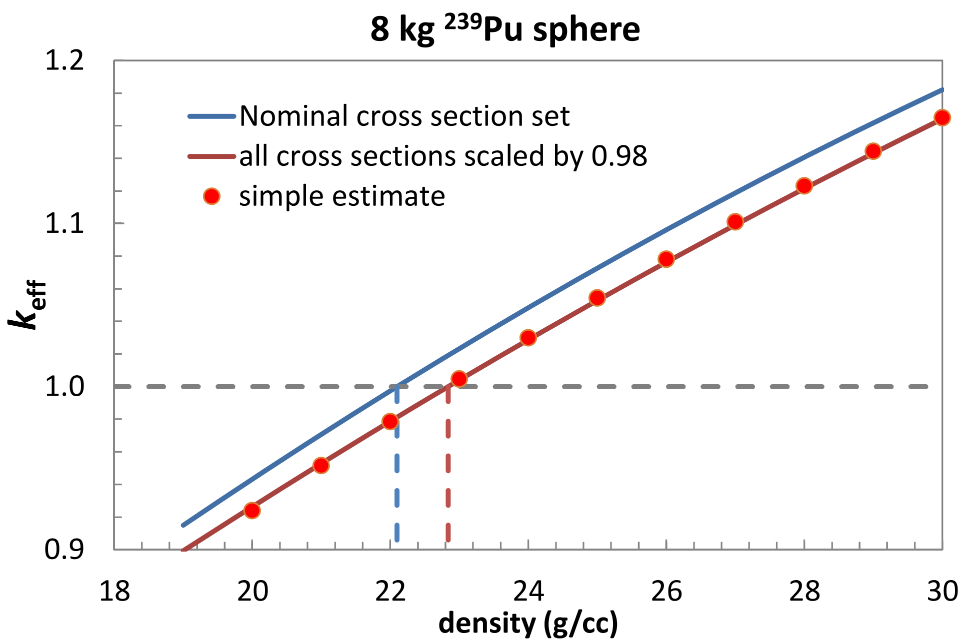
^{10}B(0^+ gs, 2^+ 3368 keV)

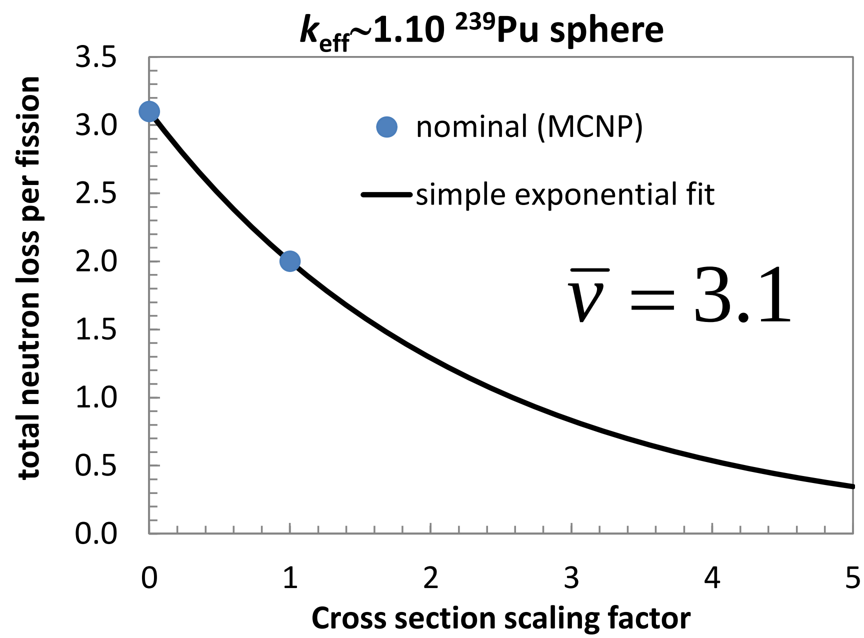
$N(2^+)/N(0^+) = 0.165 \pm 0.025$

A. V. Daniel *et al*., Phys Rev C 69 (2004)

(1) Time scales of super-critical systems (a little on sub-critical systems)

As a working example we shall consider a ^{239}Pu 8-kg bare sphere





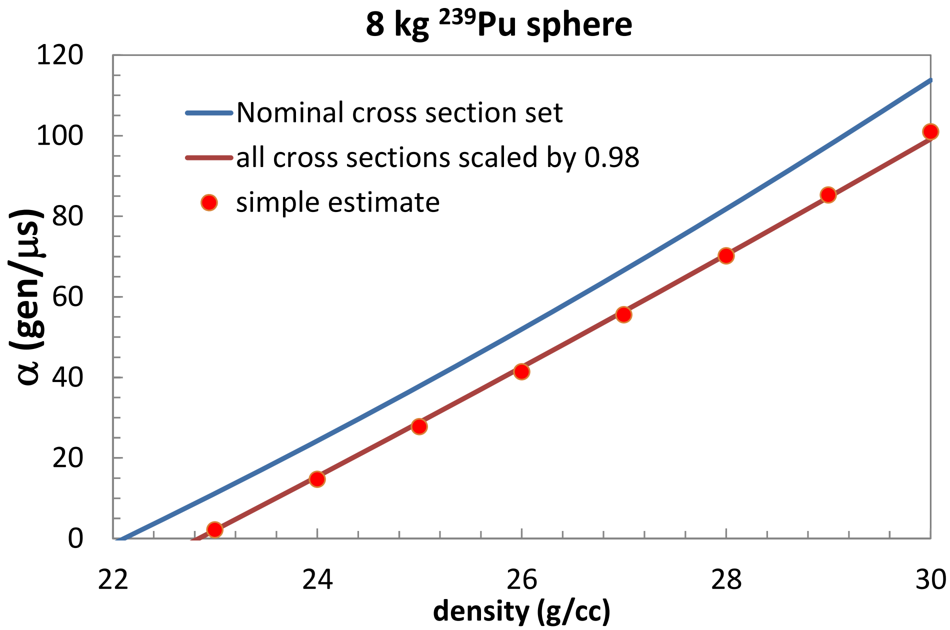
$$k_{\text{eff}} = \bar{\nu} - \ell$$

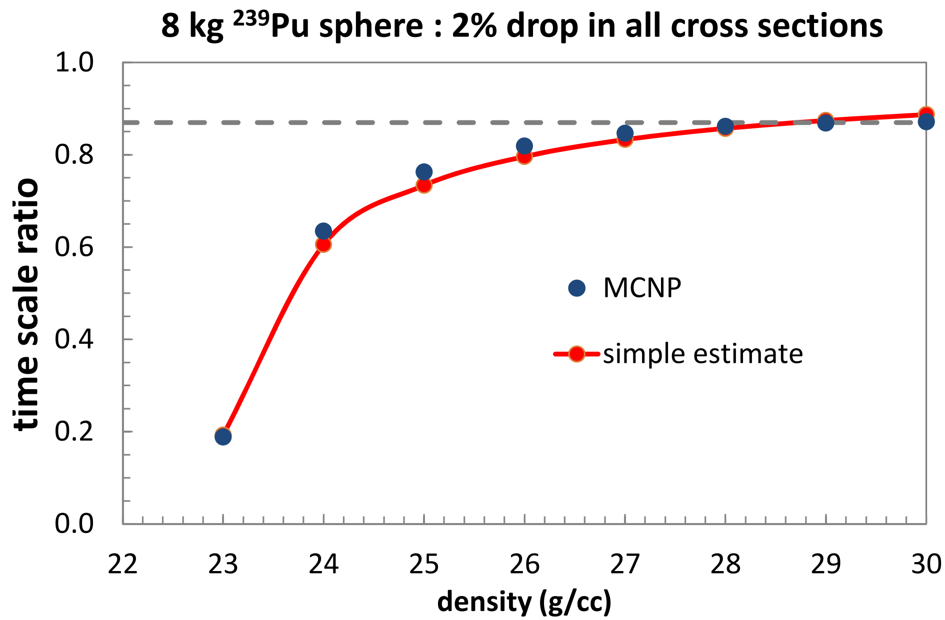
$$k^*_{\text{eff}} = \bar{\nu} - (\bar{\nu} - k_{\text{eff}}) \times 1.009$$
$$= k_{\text{eff}} \times 1.009 - \bar{\nu} \times 0.009$$

If all cross sections are scaled by 0.98 then the new total neutron loss per fission event is $\ell^* = \ell \times 1.009$

Good to 1:1000 from k_{eff}~0.9-1.2

$$\overline{N}(t) = N_o \exp(\alpha t) \qquad \alpha = \frac{k_{\text{eff}} - 1}{\tau_f}$$





$$\alpha^* \sim \alpha \left(\frac{k_{\text{eff}}^* - 1}{k_{\text{eff}} - 1} \right) \times 0.98$$

2% drop of the cross sections causes a decrease in the neutron-population buildup time scale of ~13%

$$k_{\text{eff}} = \bar{\nu} - \ell$$

$$k_{\text{eff}}^{*} = k_{\text{eff}} + \frac{\Delta \bar{\nu}}{3}$$

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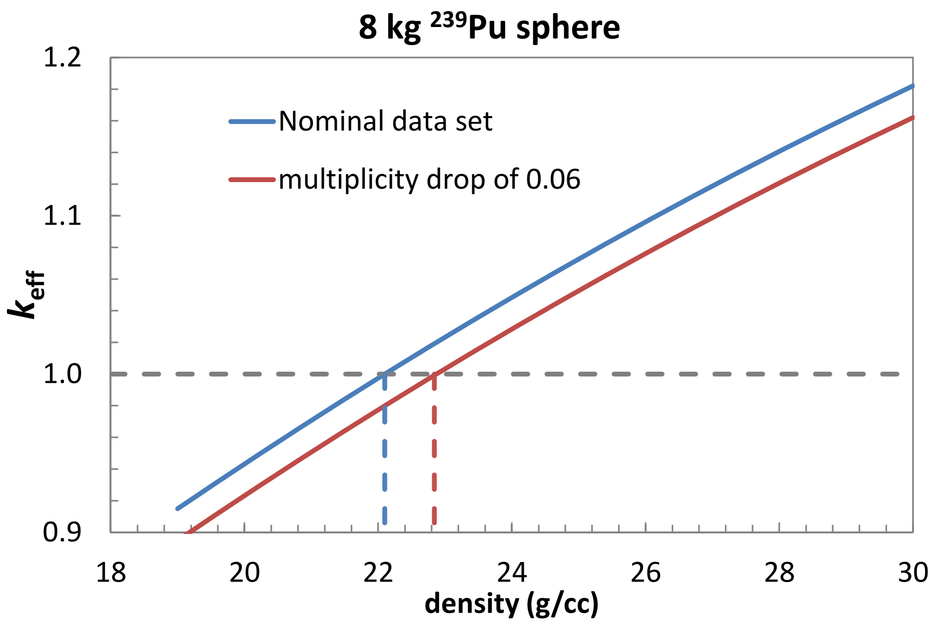
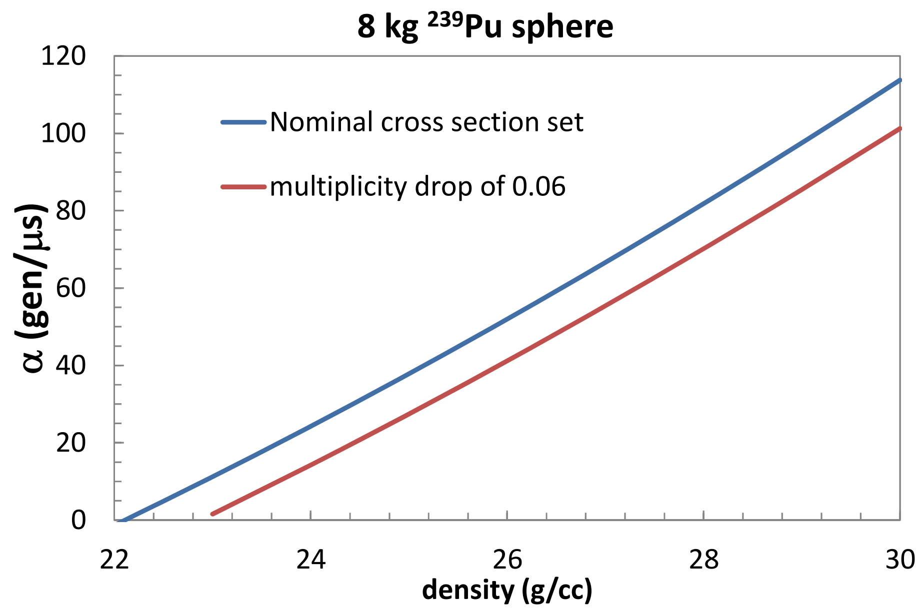
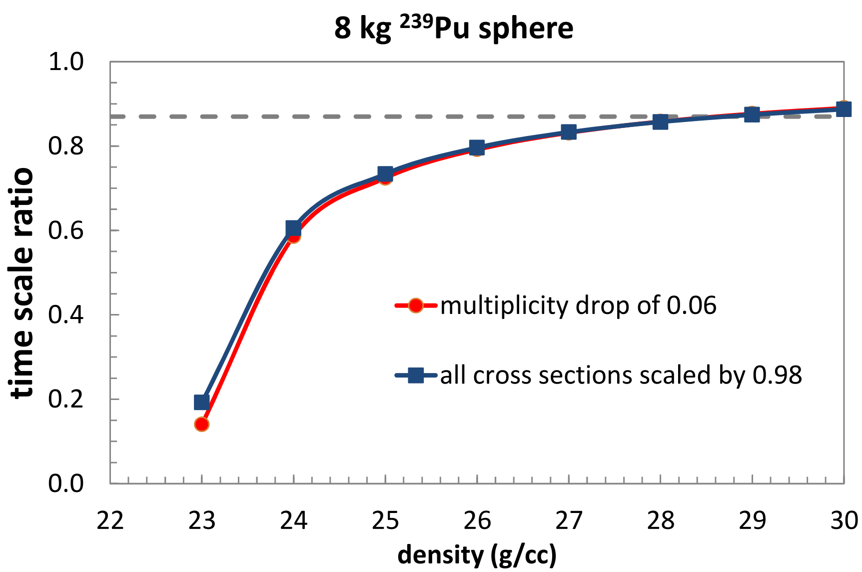
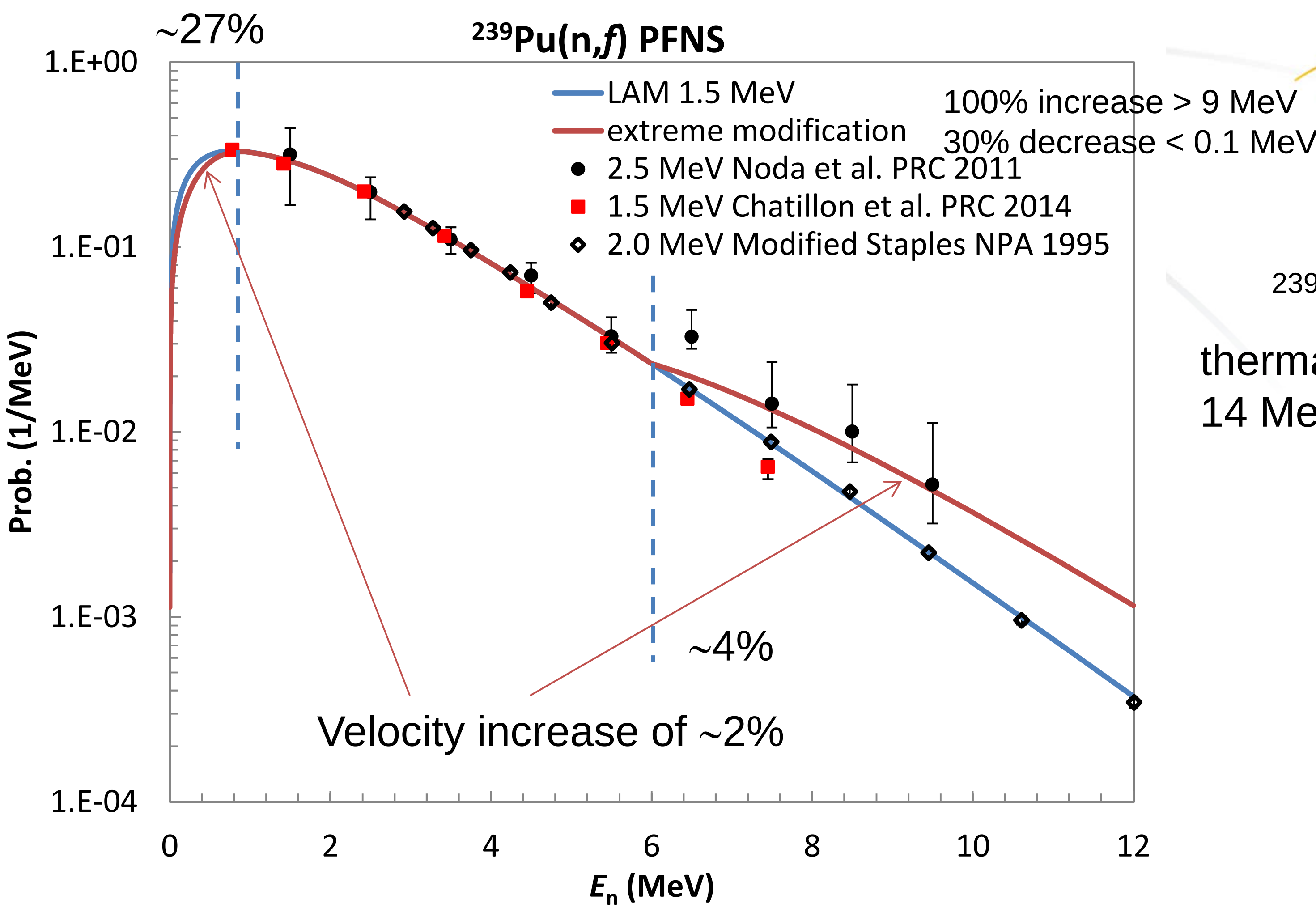






$$\alpha^{*} \sim \alpha \left(\frac{k_{\text{eff}}^{*} - 1}{k_{\text{eff}} - 1} \right)$$



^{239}Pu(n,*f*)

thermal <ν> ~2.9
14 MeV <ν> ~4.9

Low energy modification
<*E*> increases by ~ 0.05 MeV
<ν> increases by ~ 0.05/7~0.007
α increases by ~ 2% + 2% ~ 4%

High energy modification
<*E*> increases by ~ 0.10 MeV
<ν> increases by ~ 0.10 / 7 ~ 0.014
α increases by ~ 3% + 2% ~ 5%

Summary of important correlations between nuclear data and the neutron buildup time scales of fast density plutonium systems

(1) 2% increase in the Pu cross sections decreases neutron-population time scales by ~13% (for very super-critical systems)

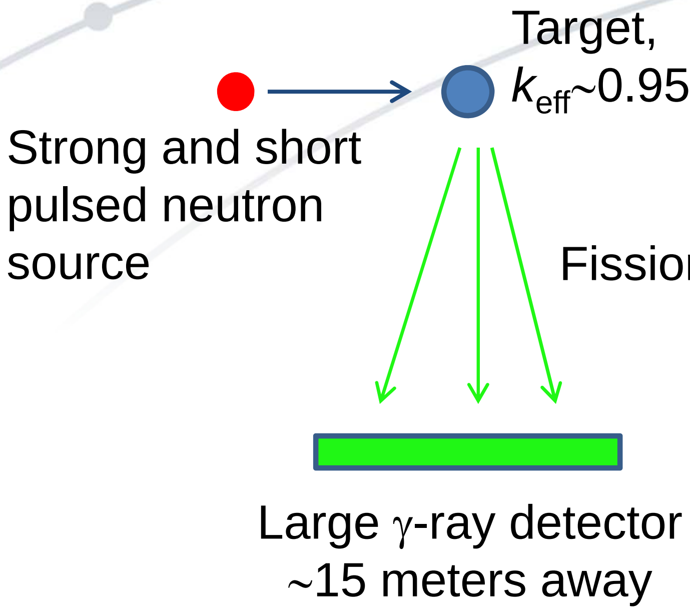
(2) 0.06 increase in the mean neutron multiplicity decreases neutron-population time scales by ~13% (for very super-critical systems)

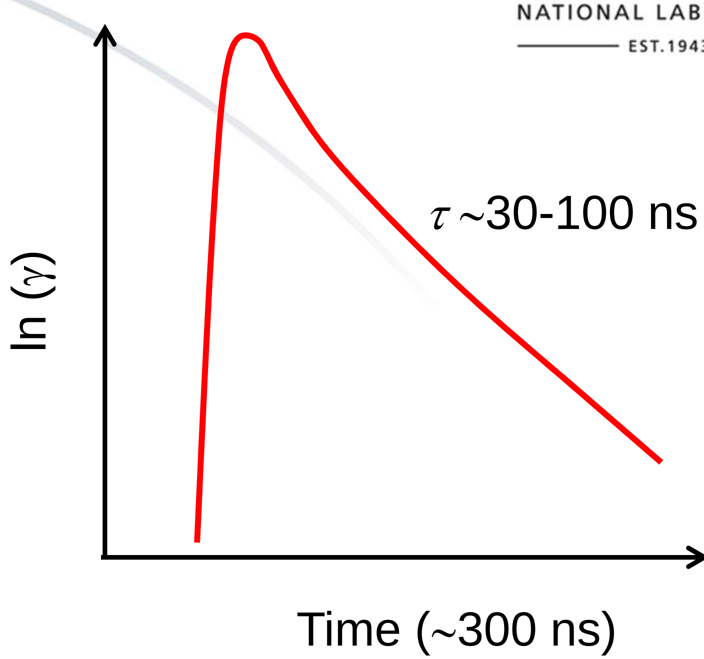
(3) 2% increase in the average velocity of the fission neutrons decreases neutron-population time scales by ~5%. Roughly 1/2 of this decrease is due to a 2% decrease in the mean time between fissions. The other 1/2 is due to a coupling between the mean neutron multiplicity and the energy of the incident neutrons.

The effects of (1) and (2) and ~ 1/2 of (3) can be mitigated by tuning to k_{eff} measurements on criticality assemblies.

To mitigate the effects of (3) associated with the coupling between mean neutron velocity and the mean time between fissions we need more accurate measurements of fast-neutron induced PFNS, and/or the measurement of fast neutron-population time scales in super-critical or near-critical systems (either above or below critical).

Neutron Diagnosed Sub-critical Experiments (NDSE)





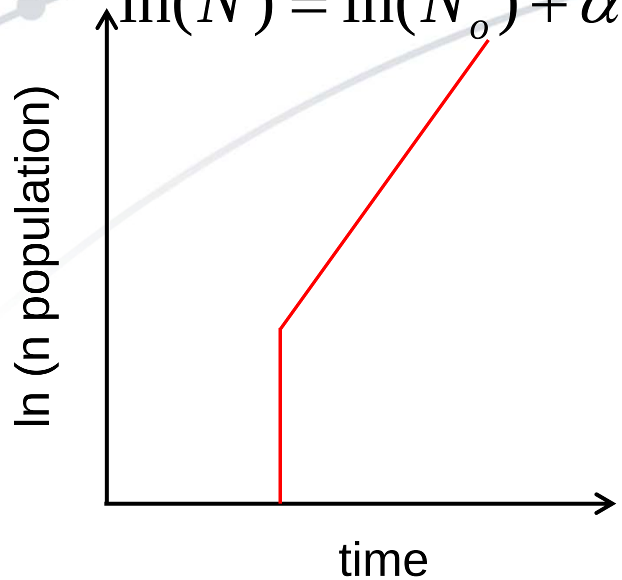
LANL recently funded a LDRD project to measure the time dependence of gamma emission from sub-critical static objects following the irradiation from a strong and short burst of neutrons.

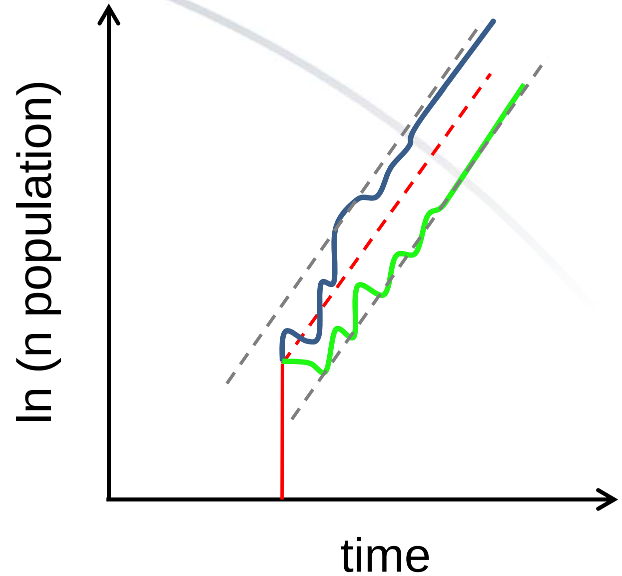
Other fission data of interest to this (and related) projects are :

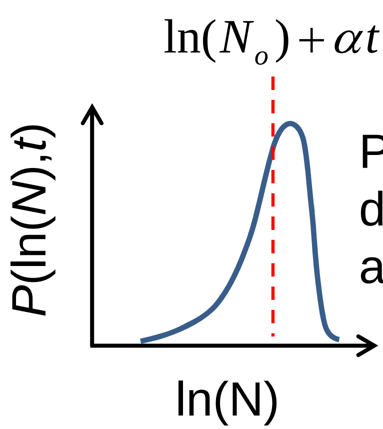
- The number and time scale of beta-delayed fission.
- The number and energy spectra of γ–rays emitted from fragments on the relevant time scales.
- Fission n and γ–ray multiplicity distributions, correlations between fission n and γ multiplicity and the corresponding energy spectra (for detailed noise analysis).

(2) Initiation issues (very briefly)

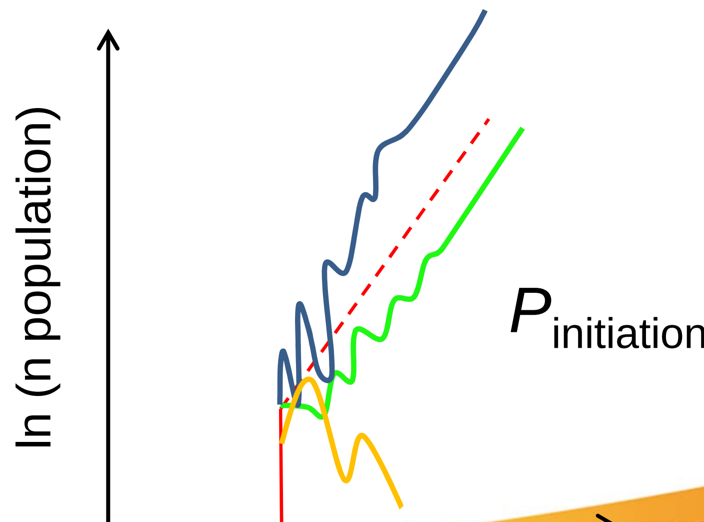
$$\ln(\bar{N}) = \ln(N_o) + \alpha t$$





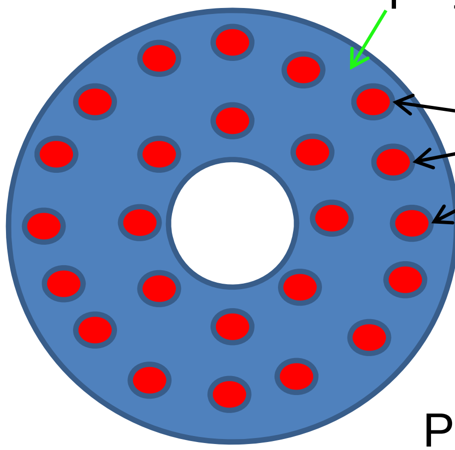


Probability distribution depends on $P_\nu(n, \varepsilon_n)$ and $P(E_n, \nu) \sim P(E_n)$



(3) Neutron multiplicities and energy spectra for well-counter design and simulation.

Safeguard of Pu separated from thermal reactor spent fuel, or Pu from breeder reactors.



Polyethylene slows the neutrons. Slow neutrons are detected via the n + ^{3}He $\rightarrow$ p + t reaction on a time scale of ~20-70 μs.

Height 30-60 cm, diameter 45-150 cm,
efficiency 10%-60%, tubes 18-200, rings 1-6,
pressure 4-10 atm., throat 12-24 cm, cost 10s $k – 100s $k.

Gamma rays can be used to infer the ratio of Pu isotopes (238-242).

If the only neutron emission was from the spontaneous fission of the even Pu isotopes then assay of Pu would be relative easy.

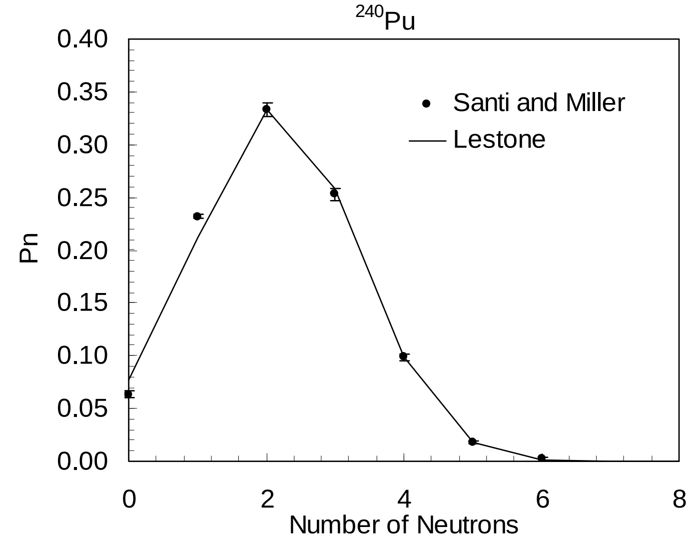
Three main sources of neutron emission from Pu samples.
(1) Neutron emission from the SF of the even isotopes.
(2) A common material at reprocessing and handling facilities is Pu oxide. This typically gives (α,n) rates comparable to the SF neutron rates. Small quantities of other low-Z materials can lead to higher (α,n) rates.
(3) For large Pu samples there is neutron multiplication via Pu(n,*f*) reactions.

Well counters can use the singles (S), doubles (D), and triples (T) rates to separate the SF, from the (α,n) in the presence of neutron multiplication.

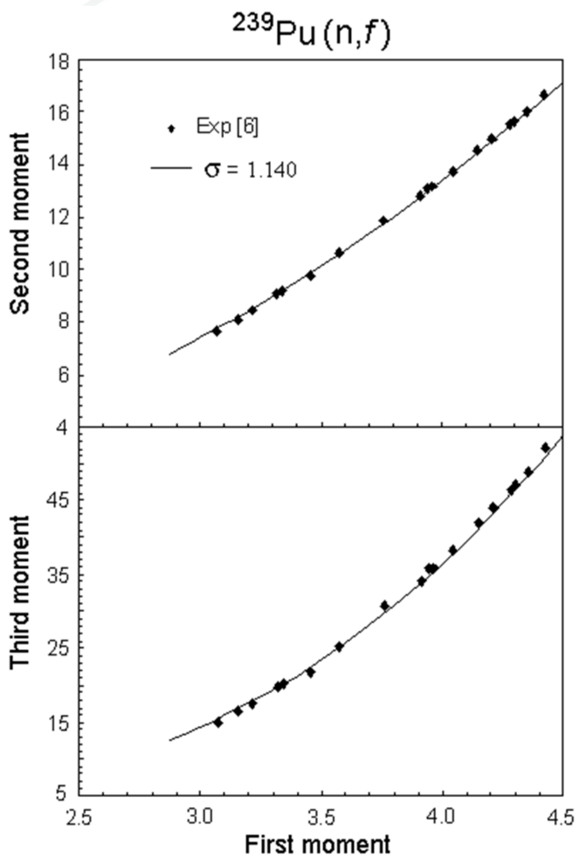
Pu assay at the 1% level is routine.

The keys to understanding well counters in detail are:

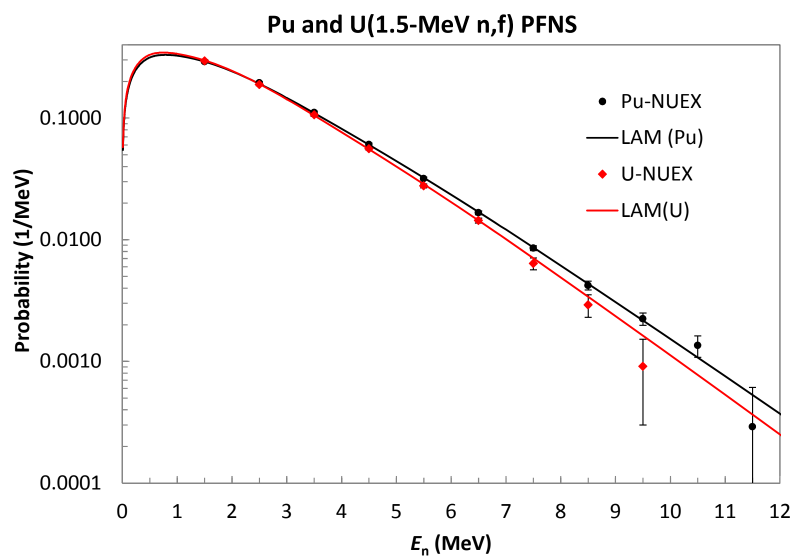
1. (1) Good Monte Carlo transport codes (e.g. MCNP)
2. (2) P_ν(n) for SF of ^{240}Pu, ^{238}Pu, and ^{242}Pu
3. (3) P_ν(n,ε_n) for ^{239}Pu(n,*f*) for multiplying samples
4. (4) PFNS for SF and Pu(n,*f*)



Lestone, LA-UR-08-1084 (2008)



Lestone, LA-UR-05-0288 (2005)
or arXiv:1409.5346



Lestone and Shores, LA-UR-14-24087 (2014)

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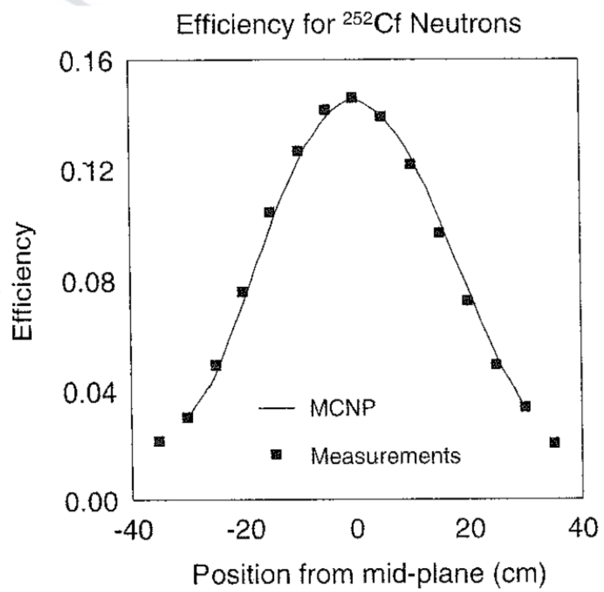
Example for a success with a well counter.

Kazakhstan spent fuel coincidence counter (SFCC) campaign 1998-2002.



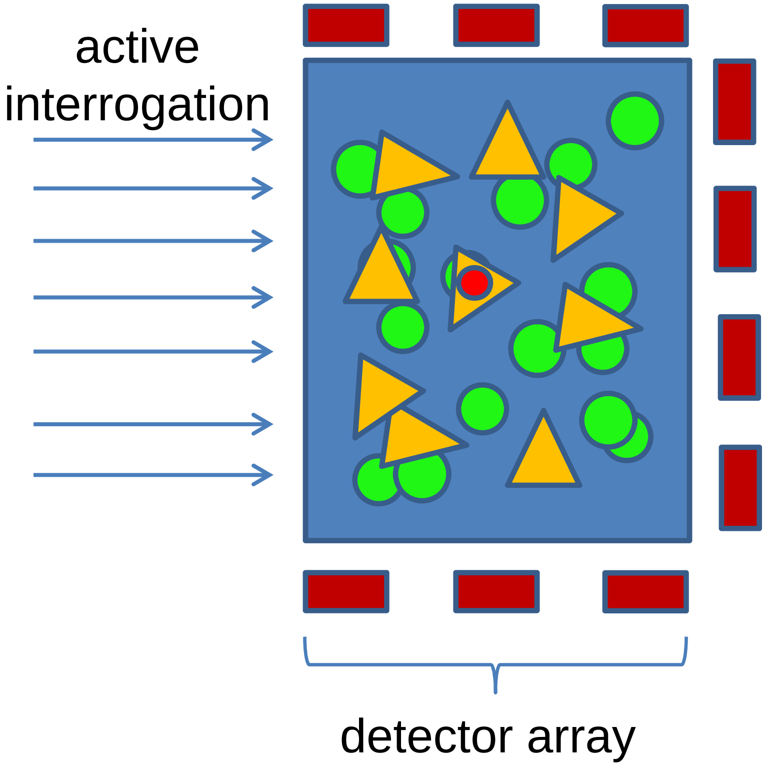
SFCC could assay the Pu content of breeder reactor spent fuel assembles (underwater) with radiation levels up to 10^5 rad/hour on contact.

After measuring ~1600 assemblies the measured Pu content was 0.4% higher than the state declared Pu content of those items.

Lestone *et al*., The Passive nondestructive assay of the plutonium content of spent-fuel assemblies from the BN-350 fast-breeder reactor in the city of Aqtau, Kazakhstan, NIMA **490**, 409 (2002).

(4) Gamma-ray multiplicities and energy spectra including multiplicity-energy correlations, and beta-delayed fission neutrons for second line of defense and border security.

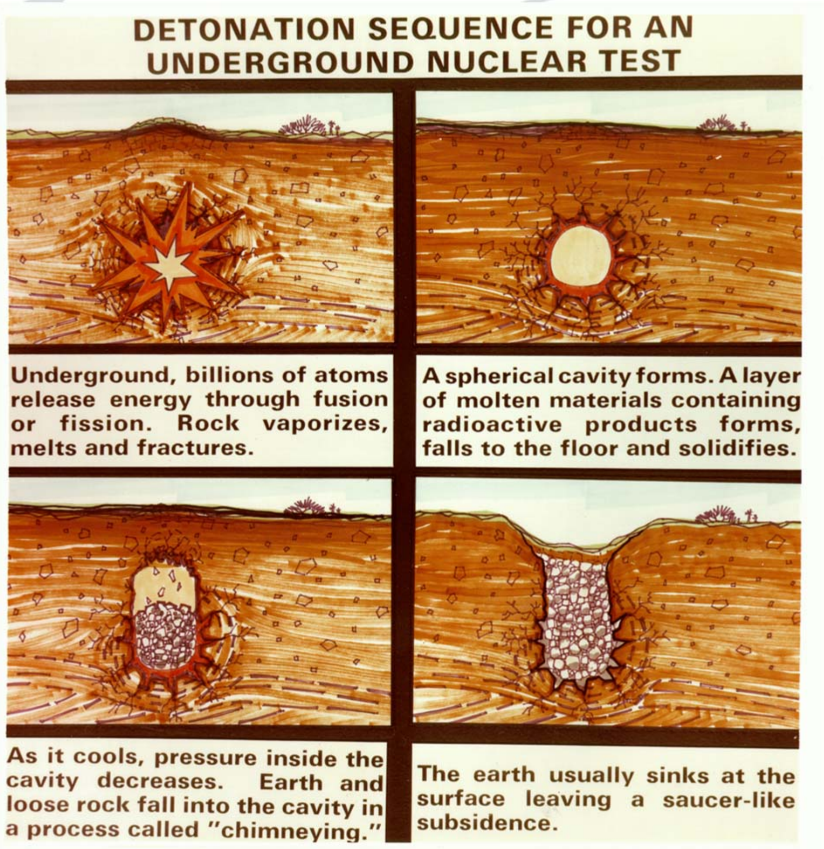
Many possible methods for detecting Special Nuclear Material (SNM) exist. Requirements : Fast (<10-30 s), simple go/no-go response, very low false-positive rate, and all of these on nearly every type of container.



Method A : Active interrogation with neutrons and look for high-multiplicity prompt fission gamma-ray coincidences.

Method B : Active interrogation with short pulse of Bremsstrahlung photons and look for beta-delayed fission neutrons between the beam pulses.

(5) Fission fragment yield curves and UGT yield assessments.



UGT radiochemistry in its simplest form

$$Y(\text{fission only Pu device}) \sim \frac{^{147}Nd}{^{239}Pu} \times \frac{1}{Y(147)} \times N_{239}(i) \times \left(\text{energy release per fission}\right)$$

^{147}Nd half life is 11 days

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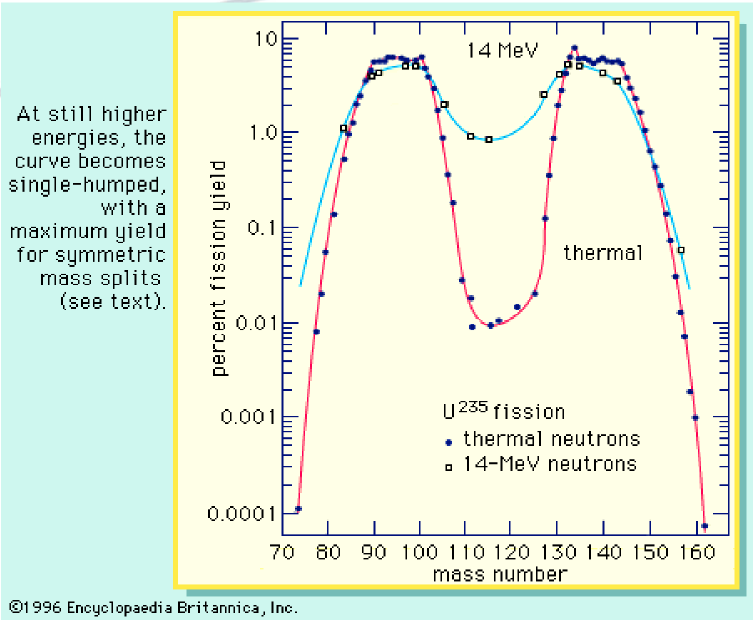
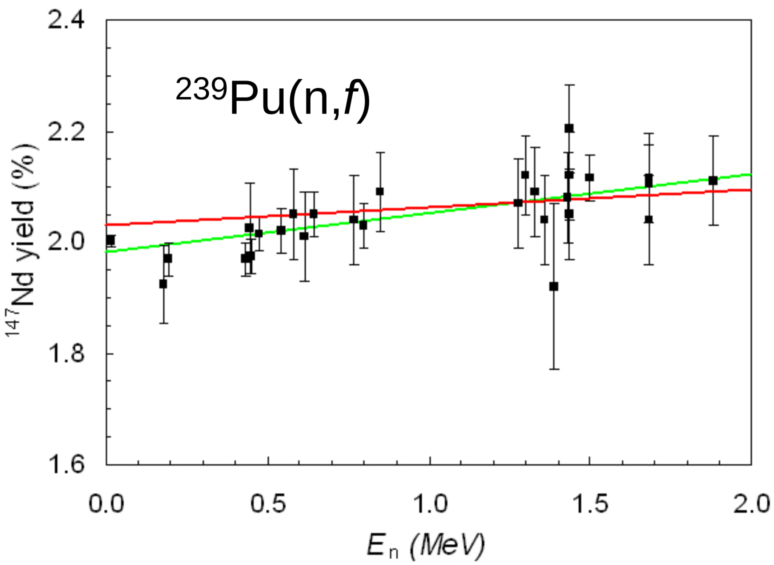
Fission fragment yield curves depend of the incident neutron energy



Until recently is was common practice in the nuclear weapons radiochemistry community to assume that the dependence of the fission fragment yield curves on incident neutron energy was small enough in the relevant energy region that it could be ignored.

This is no longer the case.
Chadwick *et al*., Nuclear Data Sheets, **111**, 2923 (2010).



Solid squares are from the Chadwick *et al*. (2010) and give an energy dependence of the ^{147}Nd yield of 4.3±0.9 % per MeV.

The curves are based on different simple theoretical assumptions. Lestone, NDS, **112**, 3120 (2011). My best model based estimate of the energy dependence of the ^{147}Nd yield is 3.6±1.0 % per MeV.

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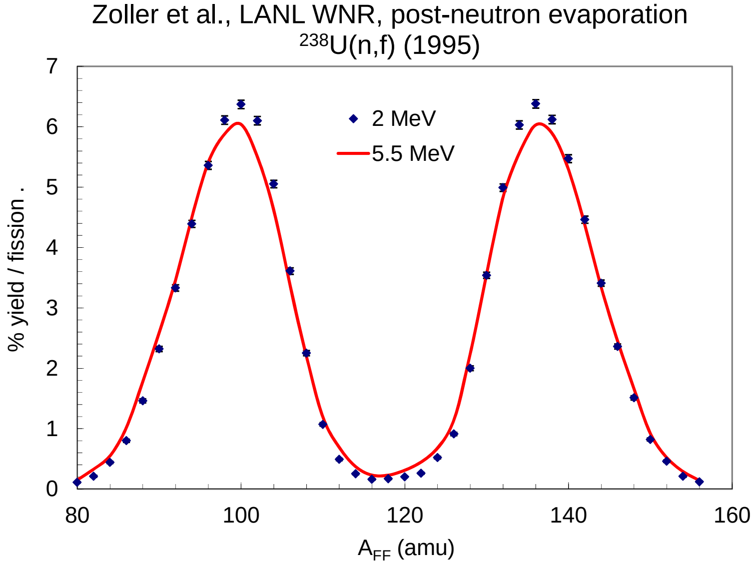
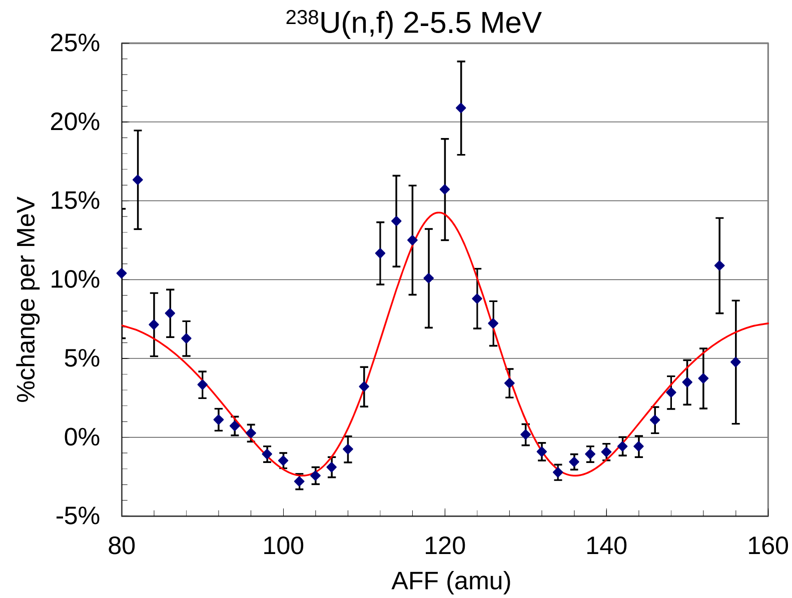
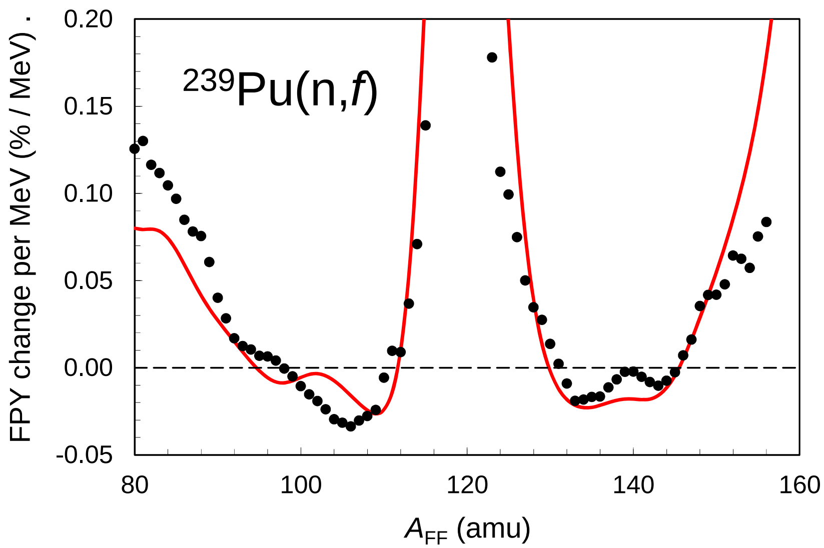
Accurate, complete, and verified yield curves for fast-neutron-induced fission of ^{239}Pu to not exist.





Zoller *et al*. mass resolution was ~3-4 amu.



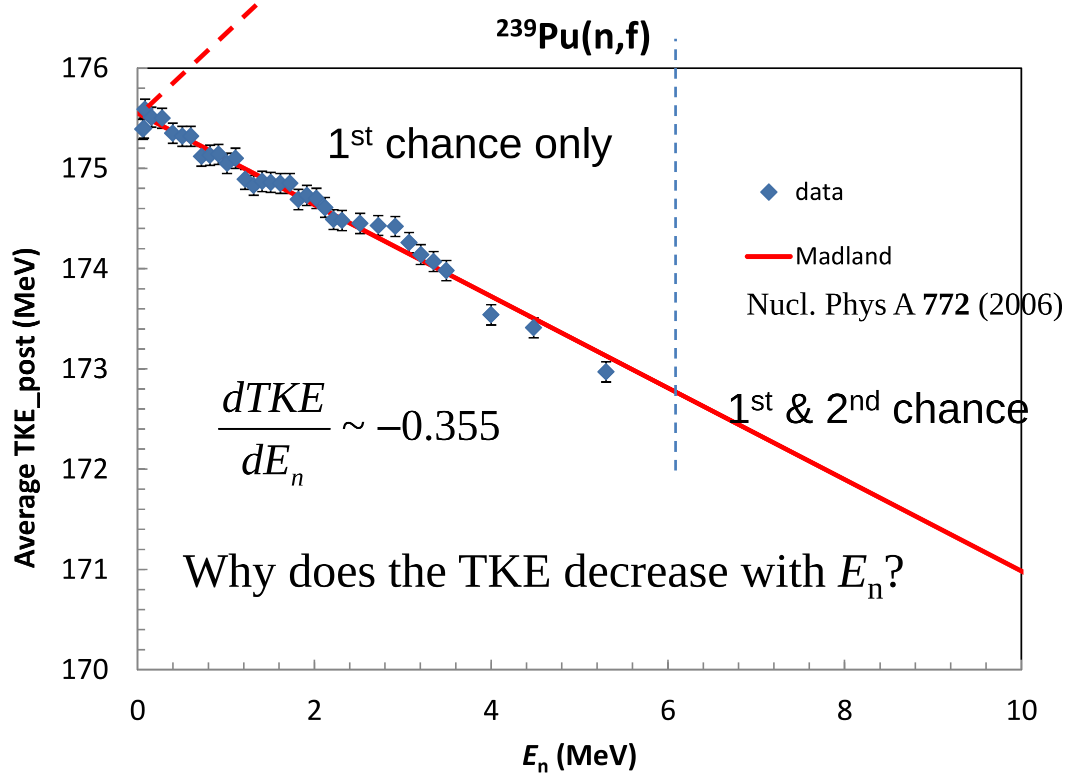
Curve is ENDF/B-VII.1 estimate.

Solid circles are from a model.
Lestone, NDS, **112**, 3120 (2011).

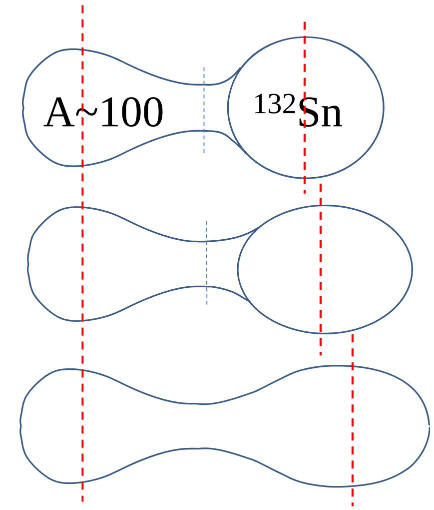
(6) Total kinetic energy (TKE) versus energy for (n,f) up to 20 MeV (Energy release).

^{6}Li(n,*f*)α,t TKE = ε_n + 4.8 MeV (fragments are in their ground states)

^{239}Pu(n,*f*) TKE $\neq$ ε_n + 176 MeV (fragments have internal heat)



A possible hypothesis



Increasing excitation energy

$\Downarrow$

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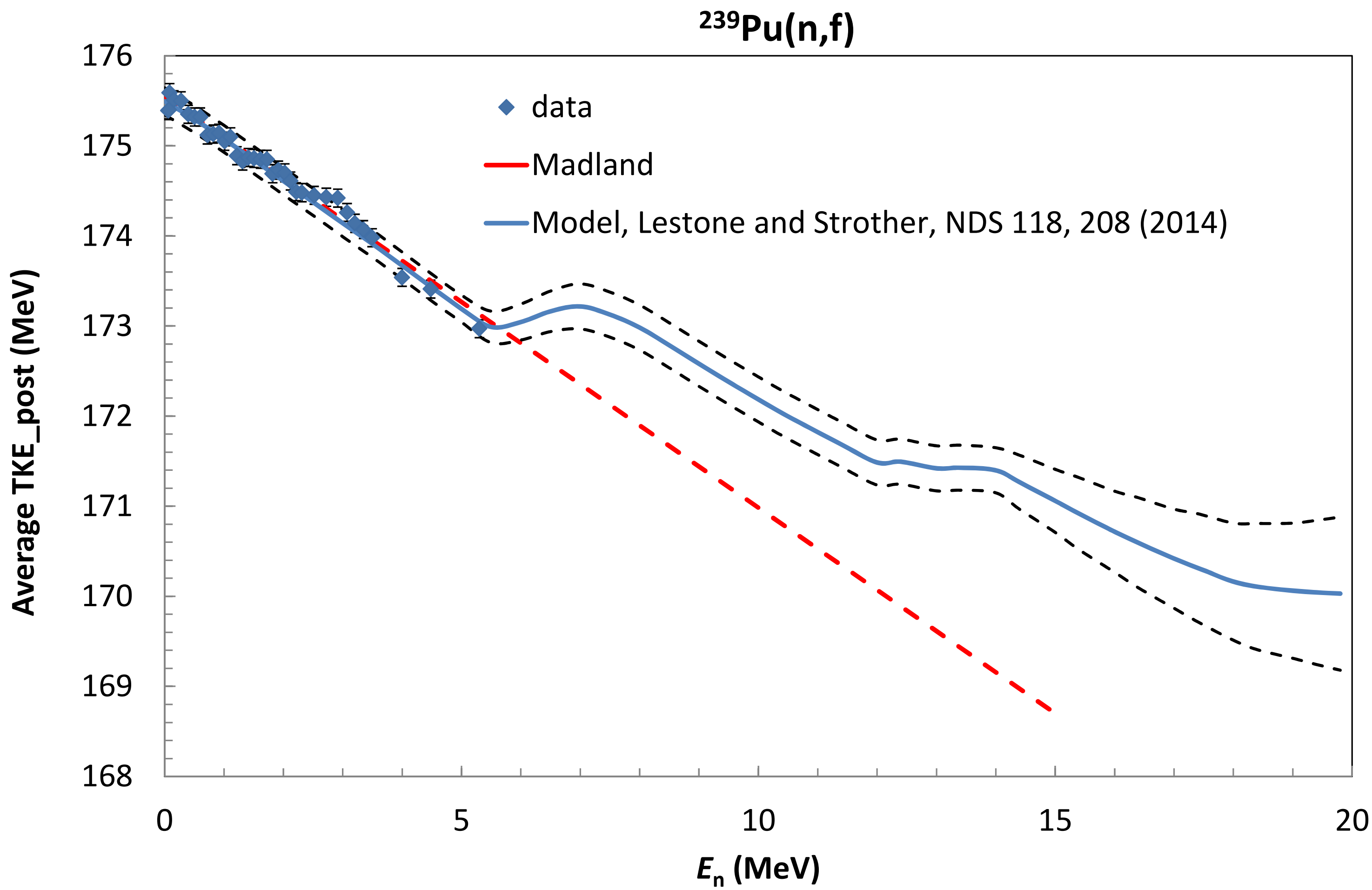

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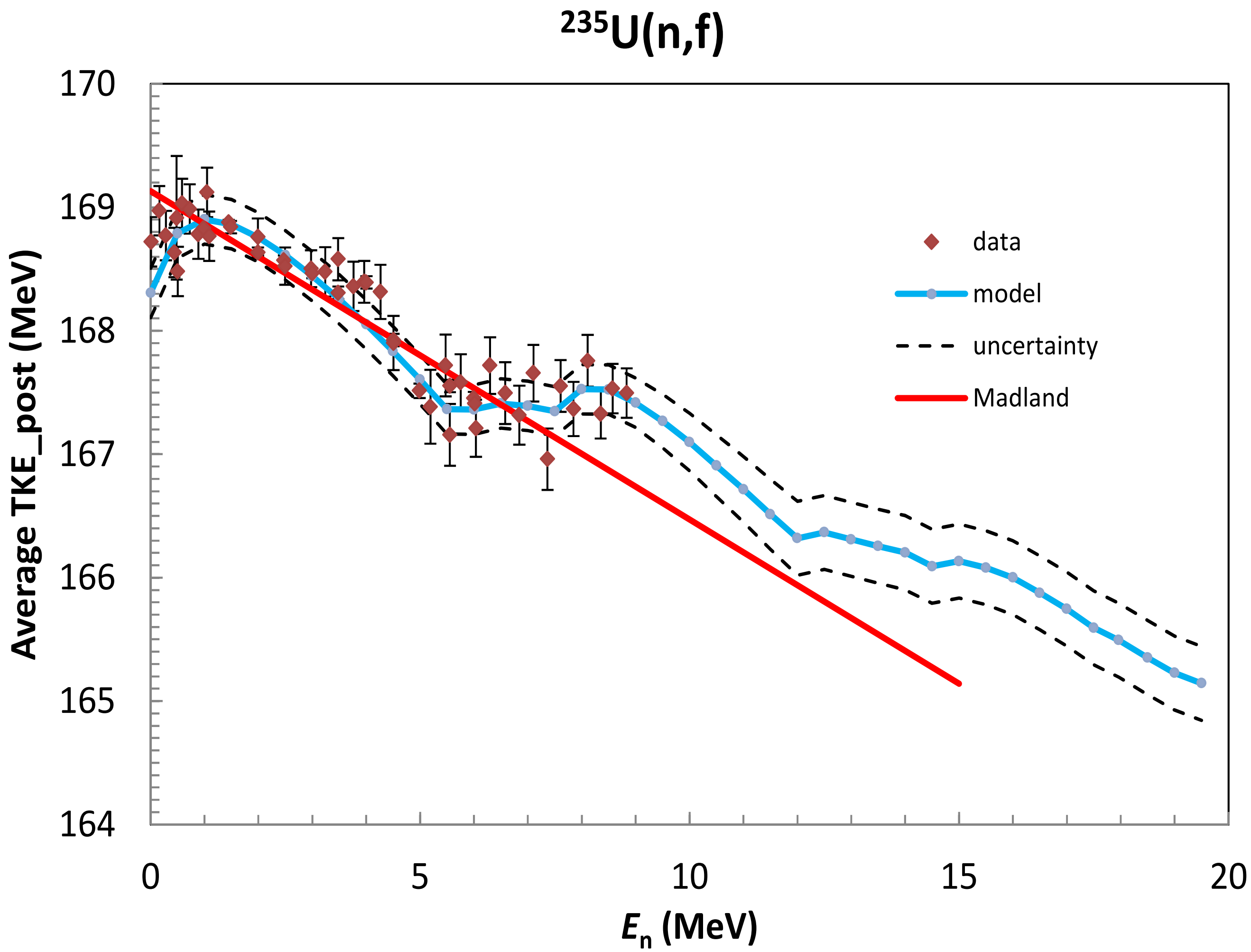
Lestone and Strother, NDS **118**, 208 (2014)



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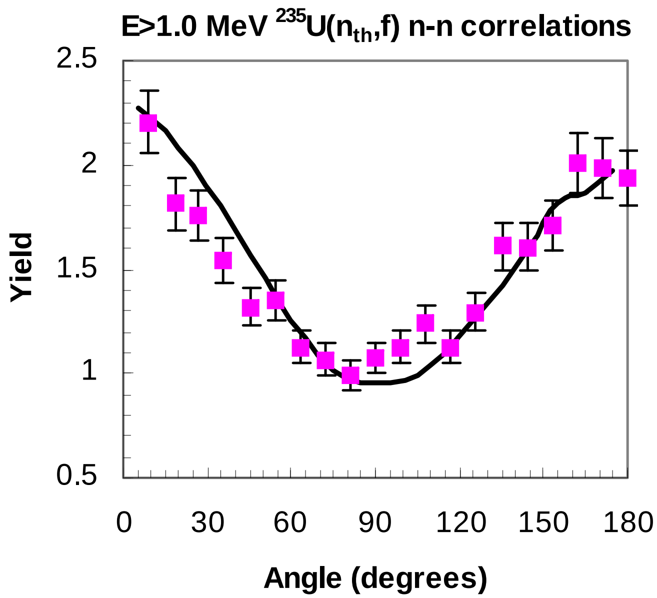
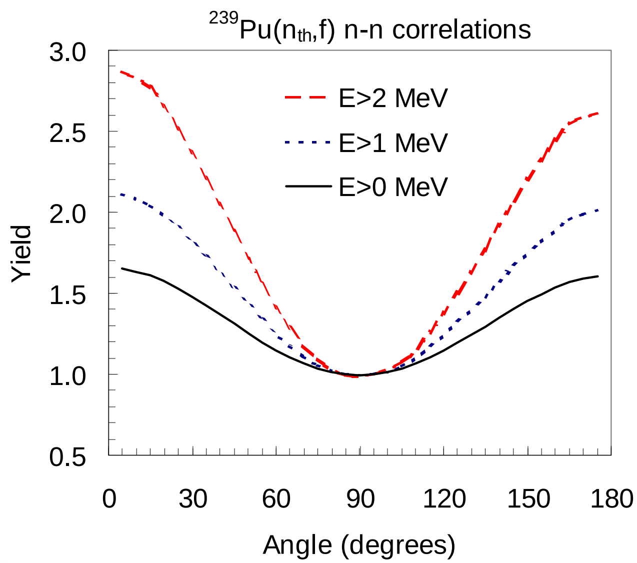
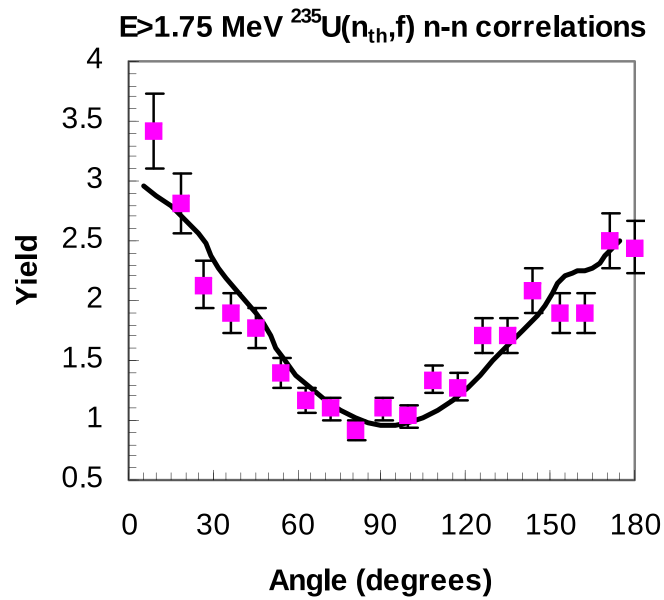
(7) n-n correlations (just for the fun of it)



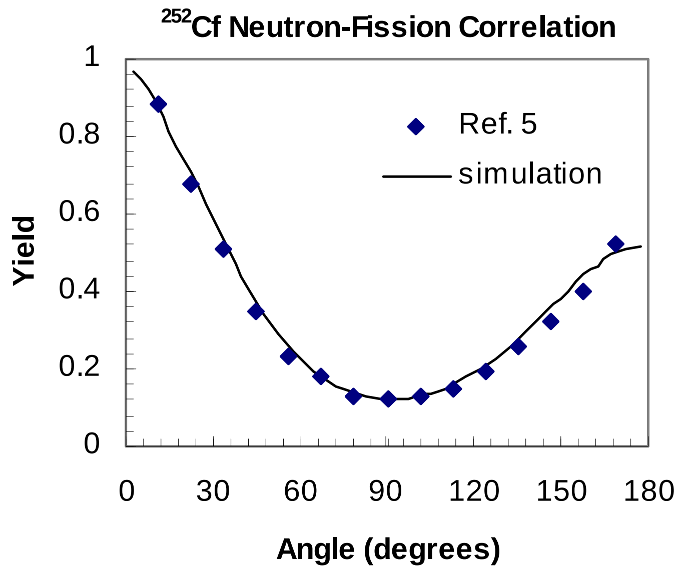


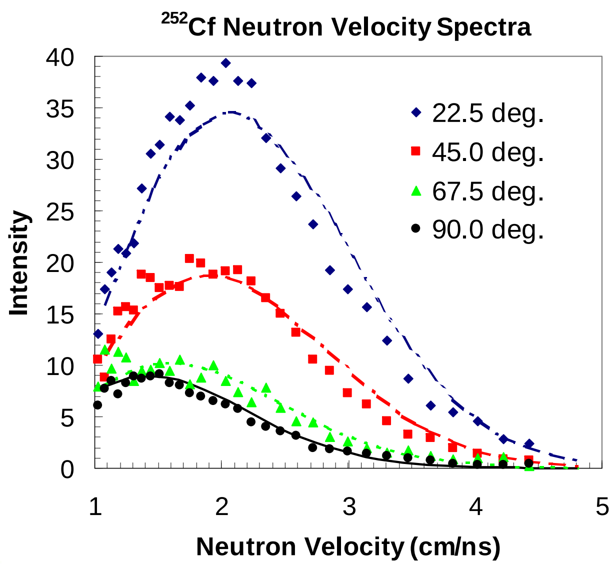
Lestone, LA-UR-99-5444 (1999)
or arXiv:1409.5345



The highest energy fission neutrons are the ones more correlated to the fission fragment directions

(8) Fission-fragment-neutron correlations for accurate determination of Prompt Fission-Neutron-Spectra (PFNS) at some accelerator based experiments.





Lestone, LA-UR-99-5444 (1999)

The highest energy part of the PFNS is strongly influenced by neutrons within 45° of the fission fragment directions.

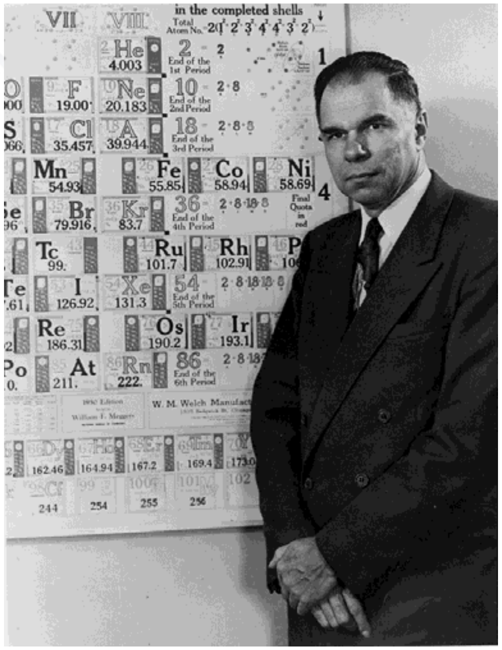
What does this mean for measurements of PFNS based on fission fragment neutron coincidences?
If there is a beam direction and the fission fragment and neutron coverages are not uniformly sampled over 4π then one must be careful that biases are not introduced into the inferred PFNS. If not done correctly the biases will grow with increasing outgoing neutron energy.

A measurement of fast-neutron induced fission of Pu PFNS where the fission trigger samples all fissions with no reference to a beam direction is Lestone and Shores, LA-UR-14-24087 (2014).

(9) Nuclear explosions used to get nuclear physics data.

The analysis of US Nuclear explosion and/or debris has provided data to the nuclear physics community.

The new elements Einsteinium (Es, Z=99) and Fermium (Fm, Z=100), new isotopes of Pu, their half-lives and decay mechanisms, were obtained via an analysis of the debris of Mike in 1952. Mike was a 10 Mt surface blast at Enewetak (1952). The team included G.T. Seaborg.

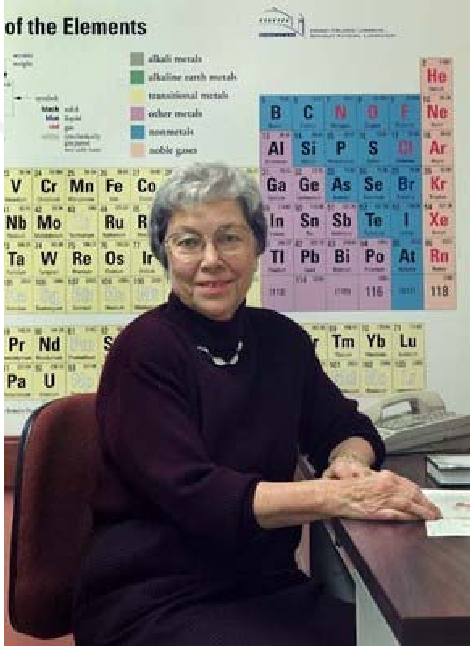


The discovery of the new elements, and the new data on neutron capture, was initially kept secret on the orders of the U.S. military until 1955. Nevertheless, the Berkeley team were able to produce Es, and Fm by civilian means, through the neutron bombardment of ^{239}Pu, and published this work in 1954 with the disclaimer that it was not the first study that had been carried out on the elements. The 'Ivy Mike' studies were declassified and published in 1955.

A. Ghiorso, *et al*., New Elements Einsteinium and Fermium, Atomic Numbers 99 and 100. *Phys. Rev.* **99,** 1048 (1955).

Mass Symmetry in the Spontaneous Fission of ^{257}Fm.
J. P. Balagna *et al*. Phys. Rev. Lett. **26**, 145 (1971).
The team included Darleane C. Hoffman.

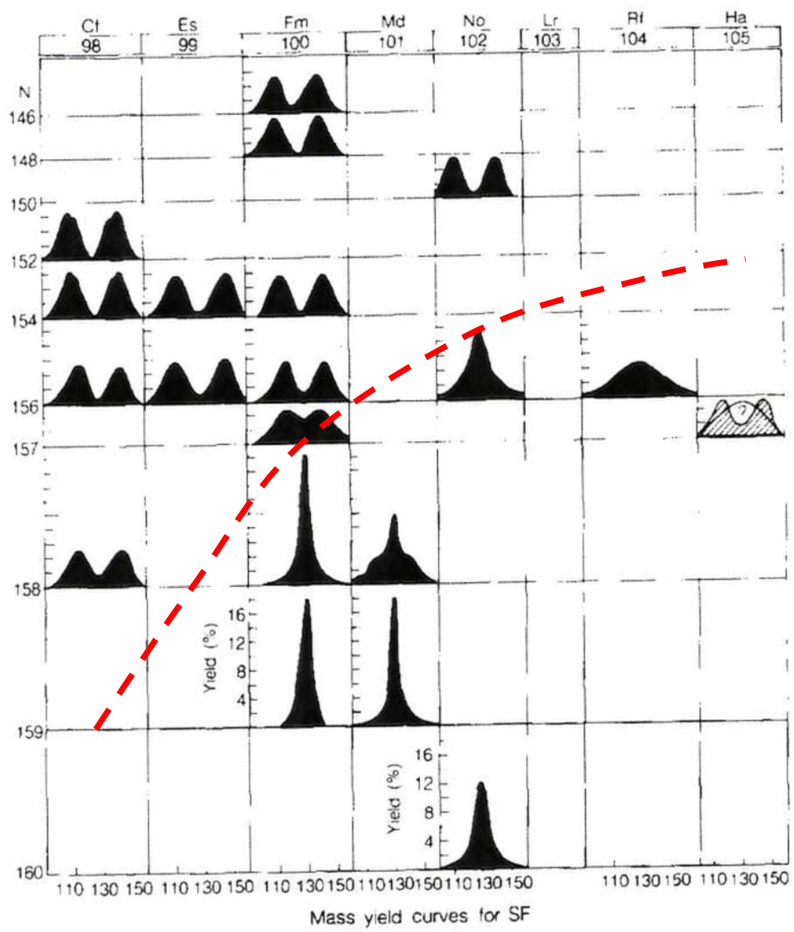
Two different source of ^{257}Fm were employed. The first was isolated from debris from the "Hutch" nuclear explosion. The second was produced by neutron capture in Curium targets in the high-flux isotope reactor (HFIR). Both sets of data showed substantially identical spectra; only results from the HFIR materials were presented in the open literature.



Hutch was a 20-200 kt underground nuclear explosion at NTS (1969). ^{257}Fm half life is 100.5 d.

The ^{257}Fm study confirmed speculation that fission-fragment shell effects should accelerate the trend towards symmetric fission with the approach towards ^{264}Fm = 2 x ^{132}Sn.

Darleane C. Hoffman, Spontaneous Fission Properties and Lifetime Systematics, Nucl. Phys. **A502**, 21c (1989).



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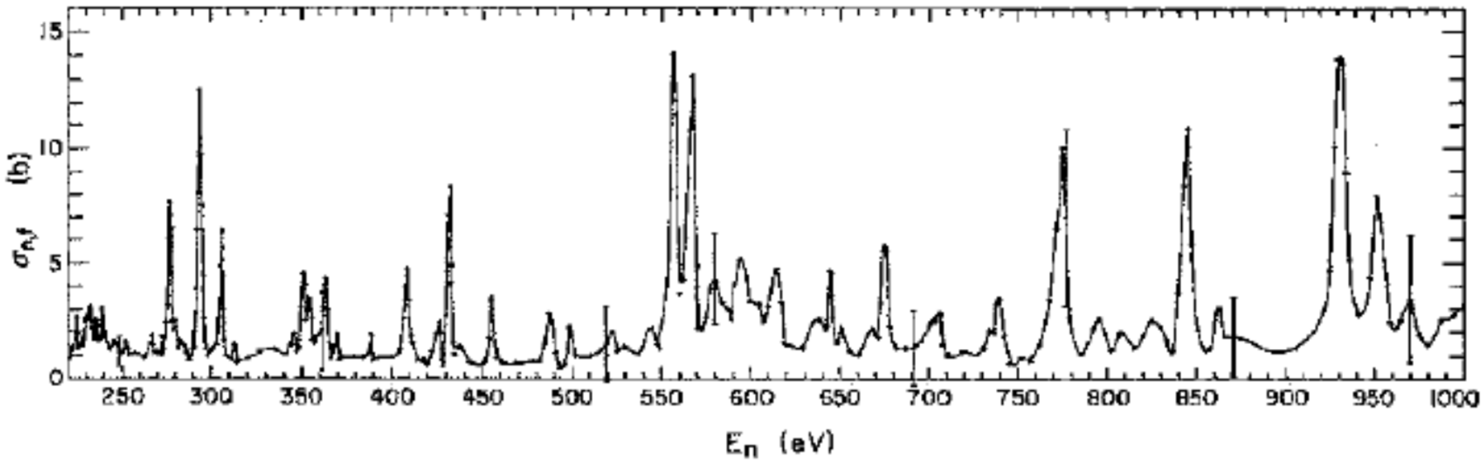
In the 60's and 70's there were several experiments to obtain cross sections for neutron inducing reactions via time-of-flight techniques using neutron explosives for the neutron source. Typically the targets contained nuclei with short half-lives that were only available in very small quantities. Cross Sections were typically measured at hundreds of meters from the explosive device and relative to ^{6}Li(n,αt) and/or ^{235}U(n,*f*).

P. A. Seeger *et al*., Fission Cross Sections for ^{241}Am and ^{242m}Am, Nucl. Phys. **A96**, 605 (1967). Petrel, 1.3 kt underground nuclear explosion at NTS (1965).

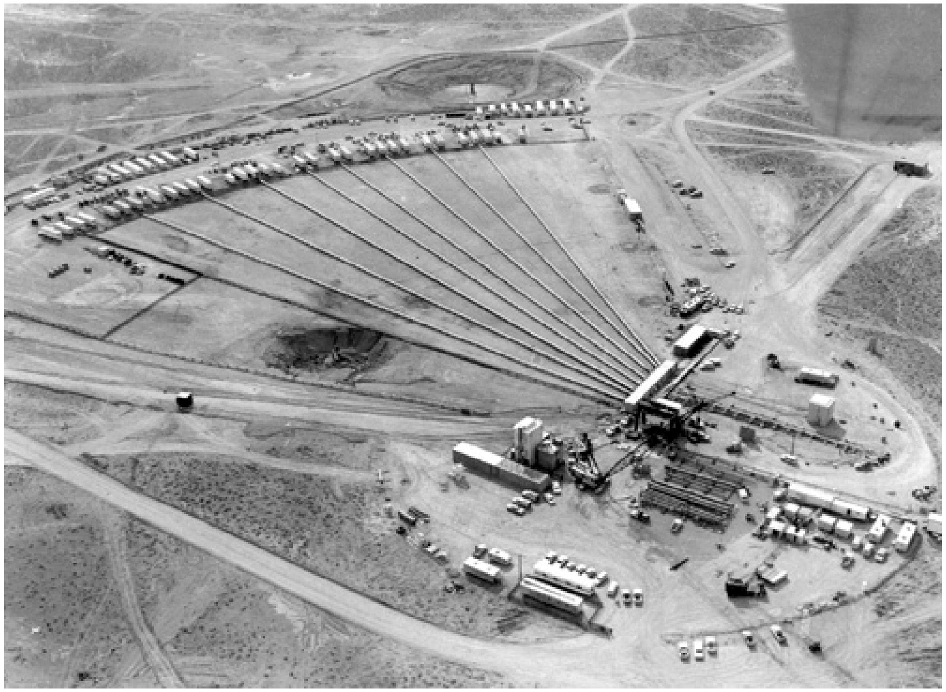
J. H. McNally *et al*., Neutron-Induced Fission Cross Section of ^{237}U, Phys. Rev. C **9**, 717 (1974). Pommard, 1.5 kt underground nuclear explosion at NTS (1968).

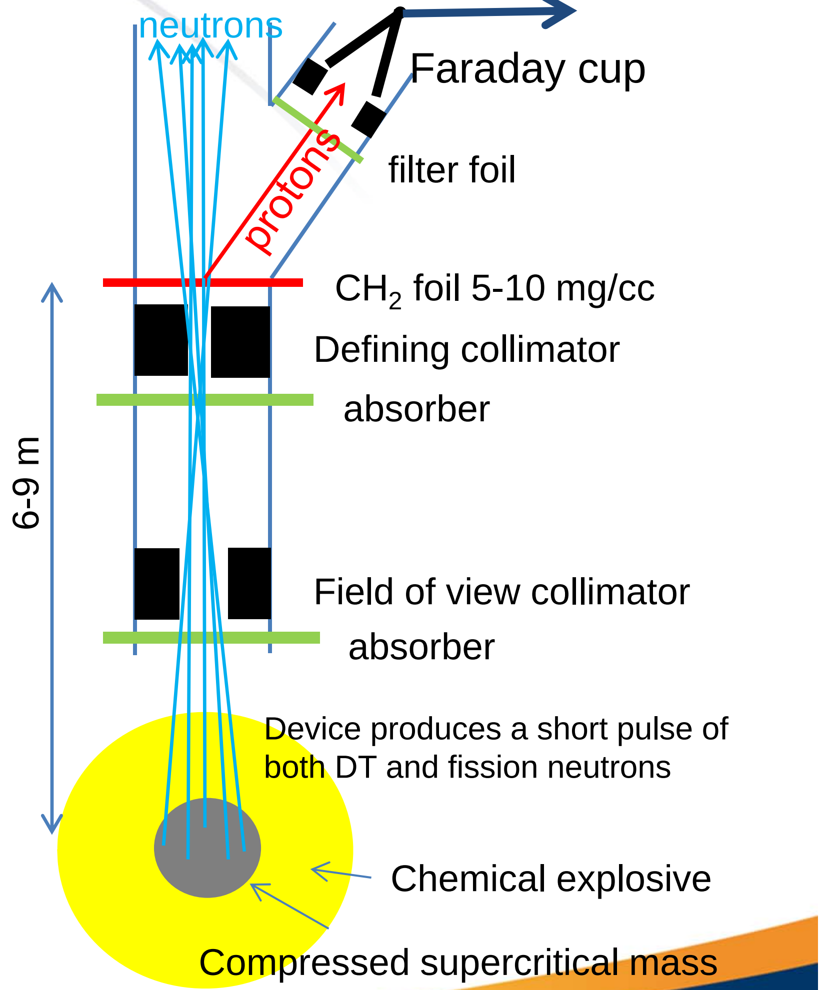


NeUtron EXperiment (NUEX)

NTS cross section measurements led to the use of ^{235}U fission foils to measure the neutron output from nuclear explosions (mid-70's).

These early NUEX led to the modern NUEX measurements where the neutron output were obtained by collecting protons scattered from a CH_2 foil in a Faraday cup (FC NUEX, late 70's to 1992).

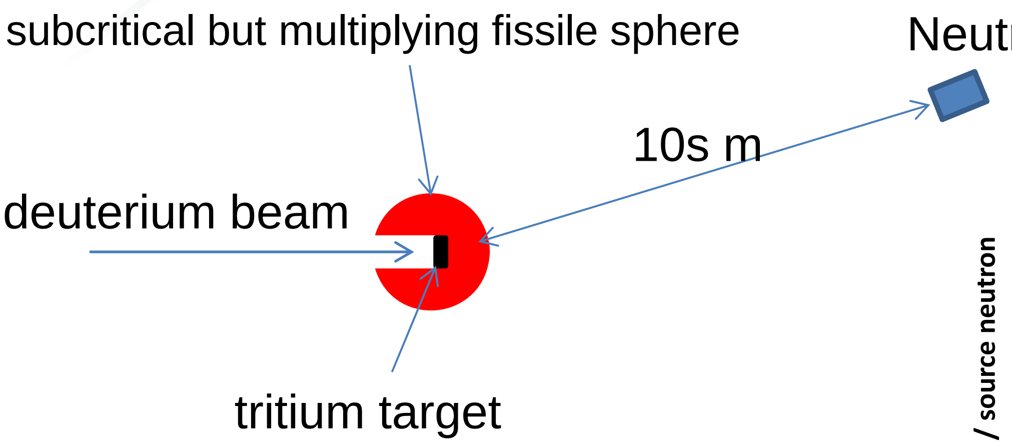


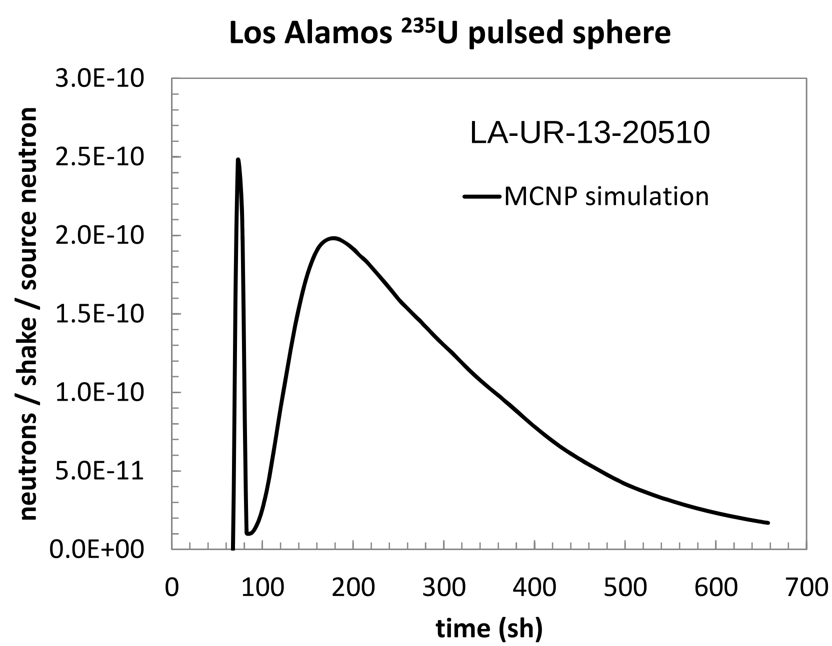


NUEX current curves from nuclear tests are classified. We will use the calculated neutron TOF spectrum of the ^{235}U pulsed sphere [LA-UR-13-2010] as a surrogate for discussion purposes.

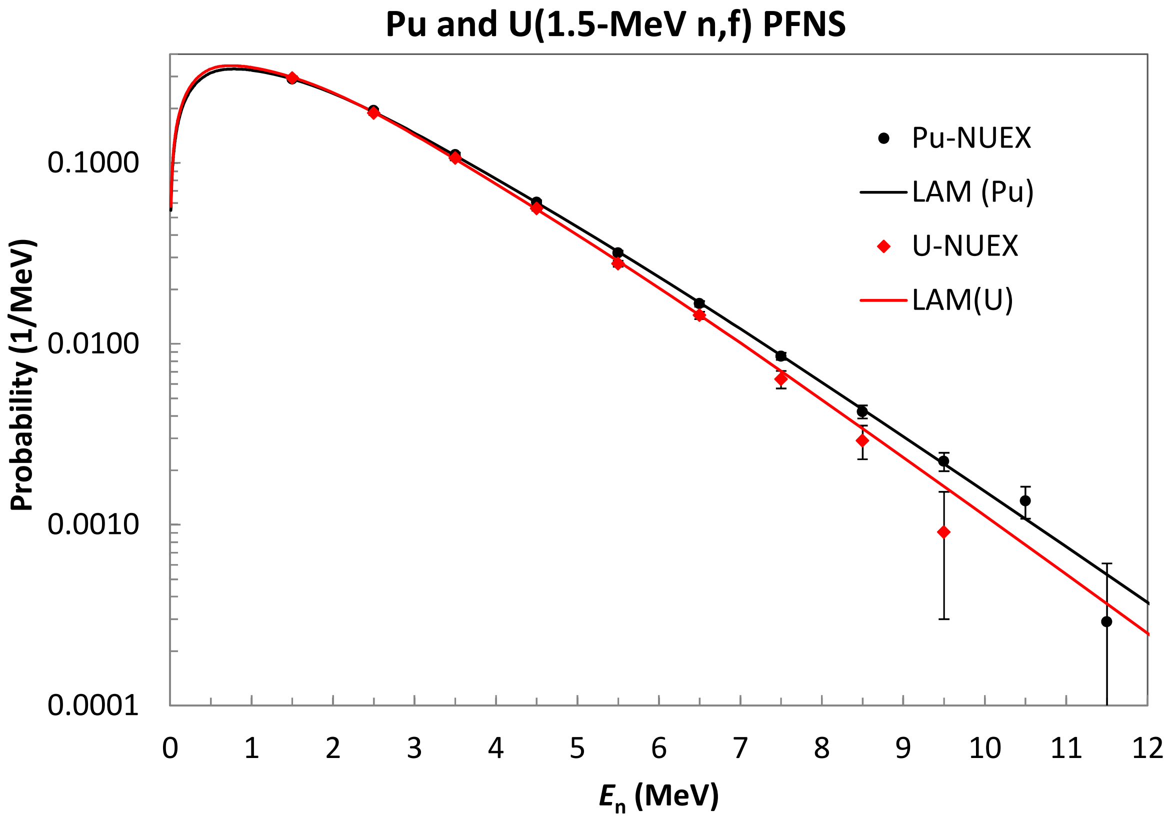
Los Alamos[1] and Lawrence Livermore[2] National Laboratory pulsed sphere programs

[1] Ragan *et al*., Nucl. Sci. and Eng. **61** (1976)   [2] Goldberg *et al*., Nucl. Sci. and Eng. **105** (1990)





Ragan *et al*. (Pulsed ^{235}U sphere : 1976) Calculations using the ENDF/B-IV uranium cross sections are in good agreement with the experiment, especially in the 1- to 6-MeV energy range where the uncertainties in both the experimental and calculated results are the smallest.



J. P. Lestone and E. F. Shores, Uranium and Plutonium Average Prompt-fission Neutron Energy Spectra (PFNS) from the Analysis of NTS NUEX Data,
Nucl. Data Sheets, **119**, 213 (2014).

For more information on the corresponding uncertainty budget see Lestone and Shores, LA-UR-14-24087.

- Prompt-fission-neutron-spectra have been inferred from Nevada-Test-Site nuclear tests from two plutonium devices, and one uranium device.
- The 1.5-MeV ^{239}Pu(n,f) PFNS is inferred from ~1 to 12 MeV. The 1-σ error bars are less than 3% up to fission-neutron energies of 6 MeV, less than 5% up to fission-neutron energies of 8 MeV, and less than 20% up to fission-neutron energies of 11 MeV.
- The 1.5-MeV ^{235}U(n,f) PFNS is inferred from ~ 1 to 10 MeV. The 1-σ error bars are less than 3% up to fission-neutron energies of 5 MeV, less than 5% up to fission-neutron energies of 7 MeV, and are less than ~ 20% up to fission-neutron energies of ~ 9 MeV.
- The ^{239}Pu-NUEX and ^{235}U-NUEX inferred PFNS, and their ratio, are in agreement with the LAM.

Summary

- Time scales of super- and sub-critical systems depend on cross sections (fission, scattering, absorption), mean neutron multiplicity per fission, and PFNS.
- Initiation : $P_\nu(n,\varepsilon_n)$.
- Well counters : SF and (n,f) neutron multiplicity distributions and PFNS.
- 2nd line of defense and border security : Detailed analysis of some detection schemes need $P_\gamma(n,\varepsilon_n,E_\gamma)$. Dependence on ε_n is weaker than for neutron emission.
- Accurate yield assessment of nuclear explosions : Accurate $Y(Z,A,\varepsilon_n)$
- Correct material heating due to the fission fragments : TKE(ε_n)
- n-n correlations remind us the high energy tail of PFNS is strongly influenced by events with small angles between the neutrons and the fragments.
- If there is a beam direction and the fission fragment and neutron coverages are not uniformly sampled over 4π then fission-neutron correlations can give biases to inferred PFNS that will increase with outgoing neutron energy.
- Explosions : New elements (Es, Fm).

  SF becomes symmetric as the system A approaches 264.

  Fission cross sections data for radioactive targets.

  PFNS from NUEX.