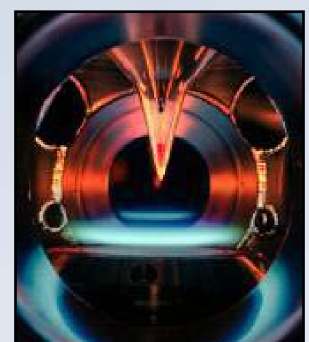
SAND2011-6984P

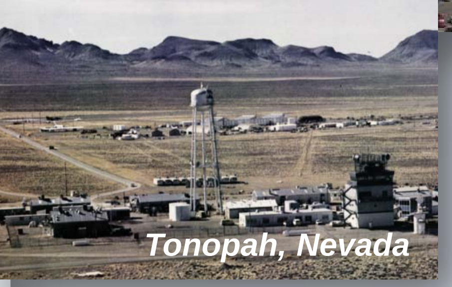
Sandia National Laboratories Overview

Key Mission Areas

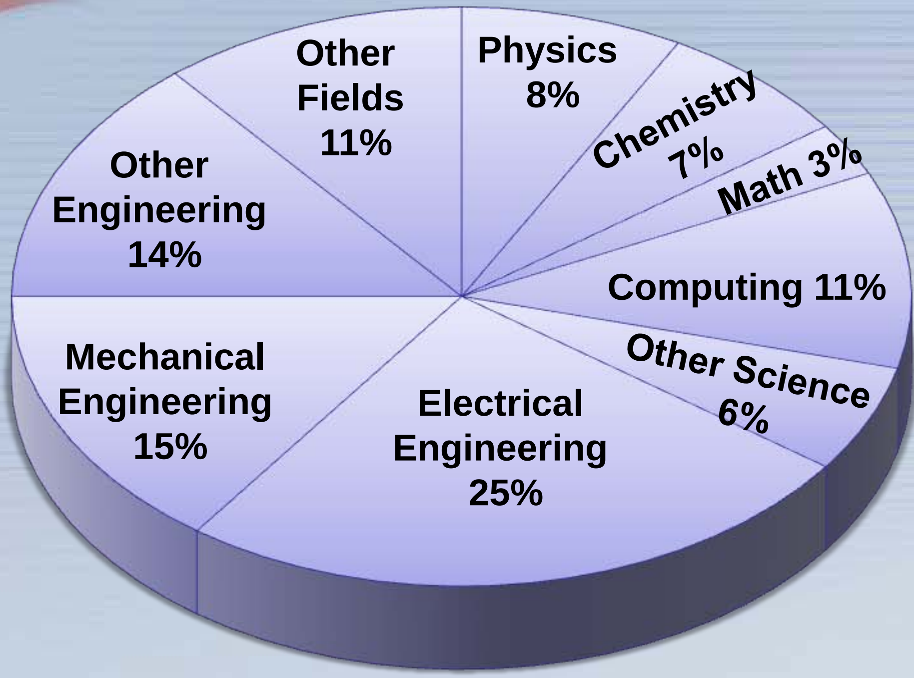
- Energy
- Homeland Security
- Defense Systems
- Nonproliferation
- Nuclear Weapons



Sandia Locations



Sandia has a Workforce of Over 10,000 Employees

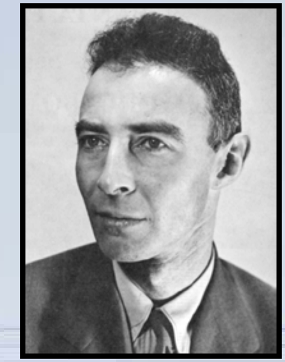


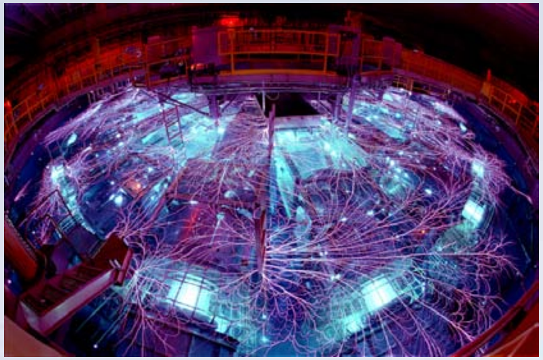
- 8,500 regular full-time employees
- 1,500 post docs, limited term, & contract employees
- 1,500 PhDs and 2,300 Masters
- $2+ billion operating budget

Work/life balance: eye candy

From Z (division) to Z (machine)

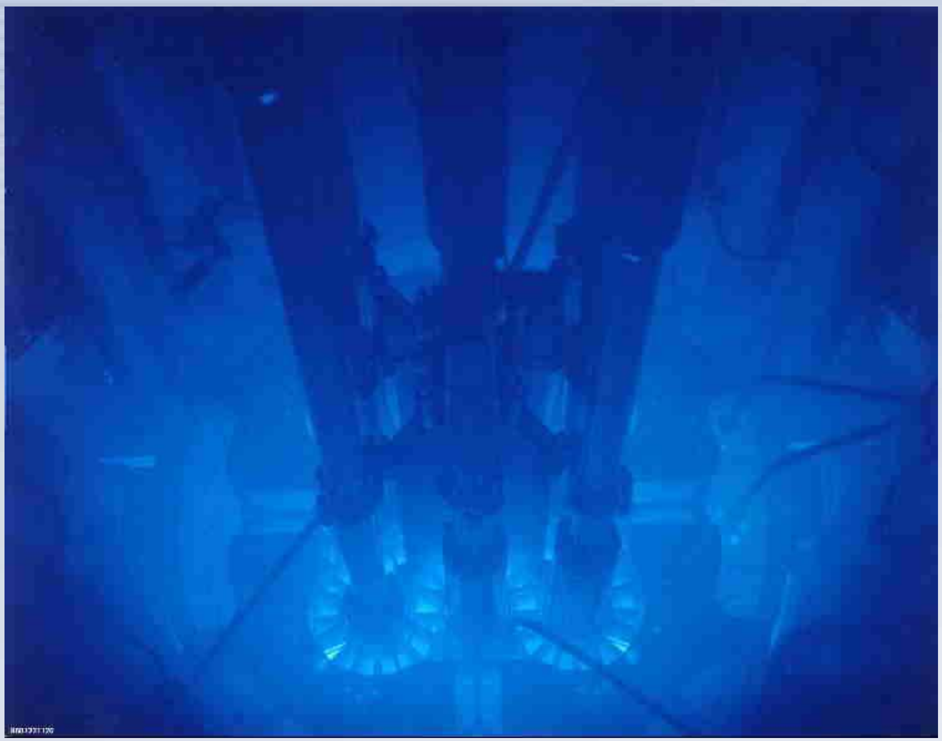
- 1945: Los Alamos Z Division moves to Albuquerque
- 1949: Sandia (Albuquerque) founded to turn physics packages into deployable weapons
- 1956: Livermore site opened
- 1974: technical advisor for WIPP
- 1997: started Z-pinch research for basic science and stockpile stewardship
- 2010: creation of Livermore Valley Open Campus





Some Sandia Opportunities for Nuclear Engineers

- **Pulsed power**
- **Nonproliferation**
- **Advanced fuel cycles**
- **Materials development**
- **Homeland security**
- **Waste transport/storage**



Sandia National Laboratories

Fast Neutron Signatures for Uranium Hexafluoride Enrichment Measurements

Scott Kiff

Radiation and Nuclear Detection Systems

September 20, 2011

Gas centrifuge enrichment plant, Piketon, OH

Sandia National Laboratories is a multi-program laboratory managed and operated by Sandia Corporation, a wholly owned subsidiary of Lockheed Martin Corporation, for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000.

LDRD LABORATORY DIRECTED RESEARCH & DEVELOPMENT

Sandia National Laboratories

Special thanks to…

- **Pete Marleau, Erik Brubaker**
  - Technical expertise
- **Mark Gerling**
  - Neutron Scatter Camera experiments, modeling
- **Michael Streicher (summer intern, Purdue)**
  - Neutron Scatter Camera experiments, modeling

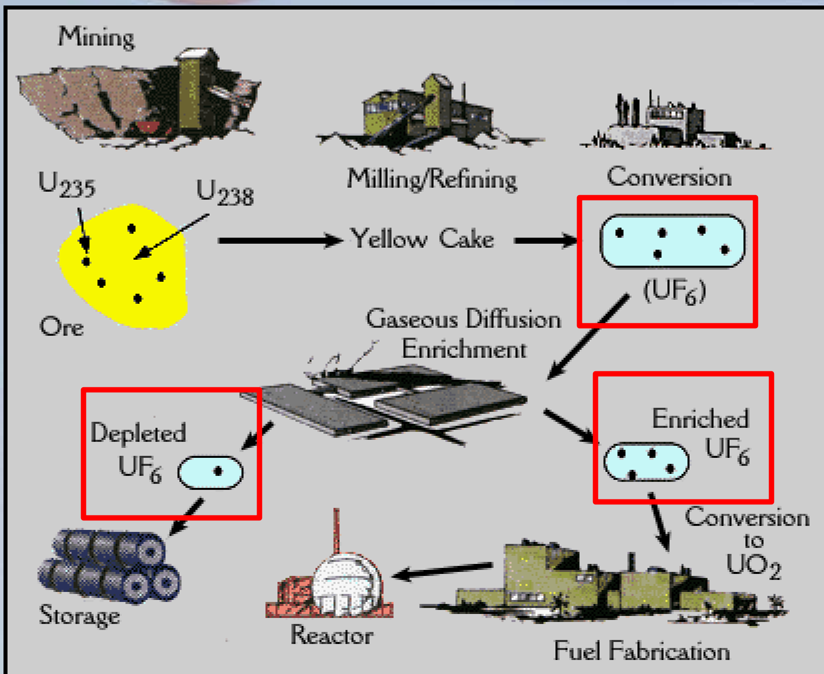
A Rough Presentation Outline

- Introduction to the problem
- How I got involved
- Using fast neutrons
- Initial calculations
- Some measurements
- Future plans
- Other possibilities
- Discussion

What is the problem?

- Enriched uranium can be used to construct a nuclear weapon
- It is important to verify the enrichment of uranium as it exits the processing stream to detect material diversion efforts
- Diversions may be possible with current NDA technology

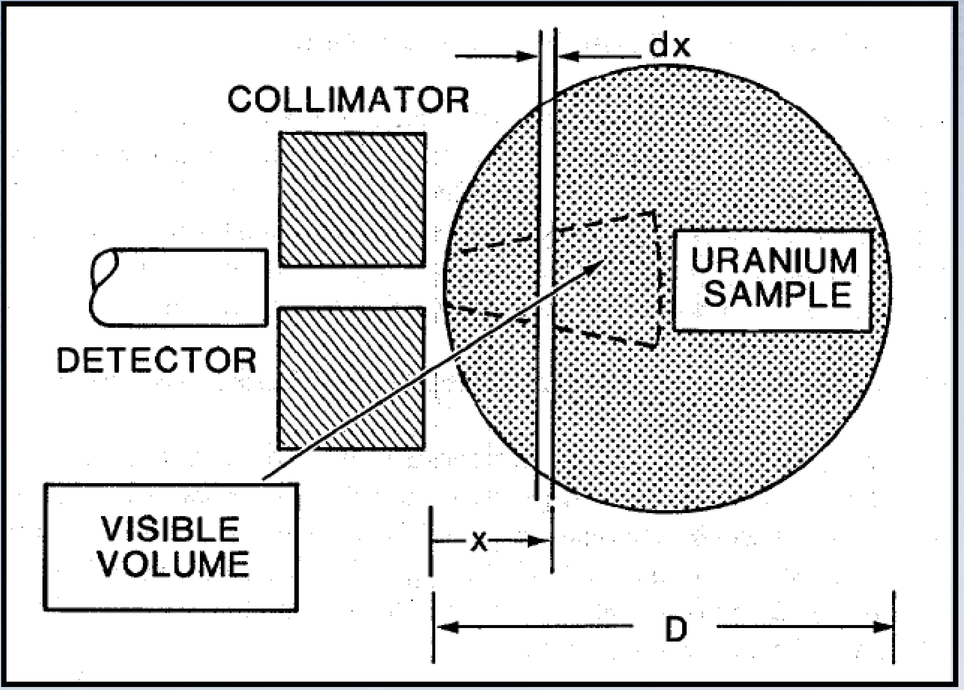
A quick review: the front end of the fuel cycle



		ial of uction	Minimum volume ft³	Minimum volume liters	Approximate tare weight (without valve protector) lb	Approximate tare weight (without valve protector) kg	Maximum, enrichment wt % uranium-235	Shipping Limit Maximum,[a] UF6 lb	Shipping Limit Maximum,[a] UF6 kg
[illegible]	[illegible]	kel	0.0053	0.15	1.75	0.79	100.00	1.0	0.45
[illegible]	[illegible]	Nickel	0.0254	0.72	4.2	1.91	100.00	4.9	2.22
5A	5	Monel	0.284	8.04	55	25	100.00	55	24.95
5B	5	Nickel	0.284	8.04	55	25	100.00	55	24.95
8A	8	Monel	1.319	37.35	120	54	12.5	255	115.67
12A	12	Nickel	2.38	67.4	185	84	5.0	460	208.7
12B	12	Monel	2.38	67.4	185	84	5.0	460	208.7
30A	30	Steel	25.65	726.0	1,400	635	5.0[b]	4,950	2,245
30B[c]	30	Steel	26.0	736.0	1,400	635	5.0[b]	5,020	2,277
48A	48	Steel	108.9	3,084	4,500	2,041	4.5[b]	21,030	9,539
48X[d]	48	Steel	108.9	3,084	4,500	2,041	4.5[b]	21,030	9,539
48F	48	Steel	140.0	3,964	5,200	2,359	4.5[b]	27,030	12,261
48G	48	Steel	139.0	3,936	2,600	1,179	1.0[f]	26,840[e]	12,174[e]
48Y[d]	48	Steel	142.7	4,041	5,200	2,359	4.5[b]	27,560	12,501
48H	48	Steel	140.0	3,964	3,170	1,438	1.0[f]	27,030	12,261
48HX	48	Steel	140.0	3,964	3,170	1,438	1.0[f]	27,030	12,261

Current technology is good, but…

- **“Enrichment meter” measures gamma emissions from the uranium hexafluoride (UF_6)**
  - Gives local enrichment, not total mass (need a scale for masses)
  - Sensitive to variations in container wall thickness
  - Not sensitive to material beyond outer skin of UF_6



The enrichment meter principle. From Reilly et al., *Passive Nondestructive Assay of Nuclear Materials*, Fig. 7.3

How I got involved…

Nuclear Instruments and Methods in Physics Research A 612 (2010) 309–319

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research A

journal homepage: www.elsevier.com/locate/nima

^{235}U enrichment or UF_6 mass determination on UF_6 cylinders with non-destructive analysis methods

R. Berndt [a], E. Franke [b], P. Mortreau [a,*]

[a] The European commission, Joint Research Centre, Institute for the Protection and Security of the Citizen, TP 800, Via Fermi, Ispra, Italy
[b] Consultant, Dresden, Germany

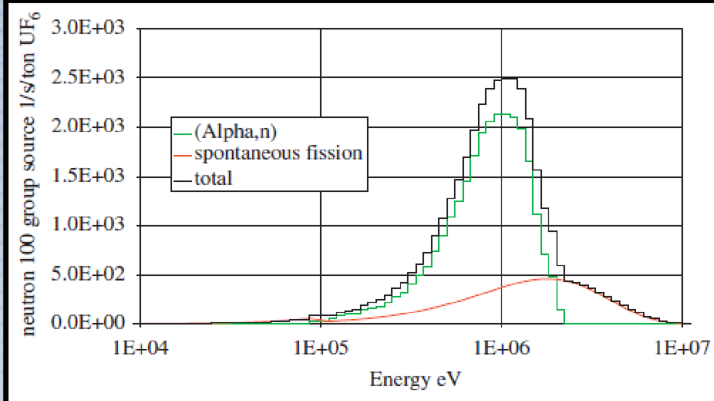


Fig. 4. Hundred group neutron spectra for natural UF_6 [7].

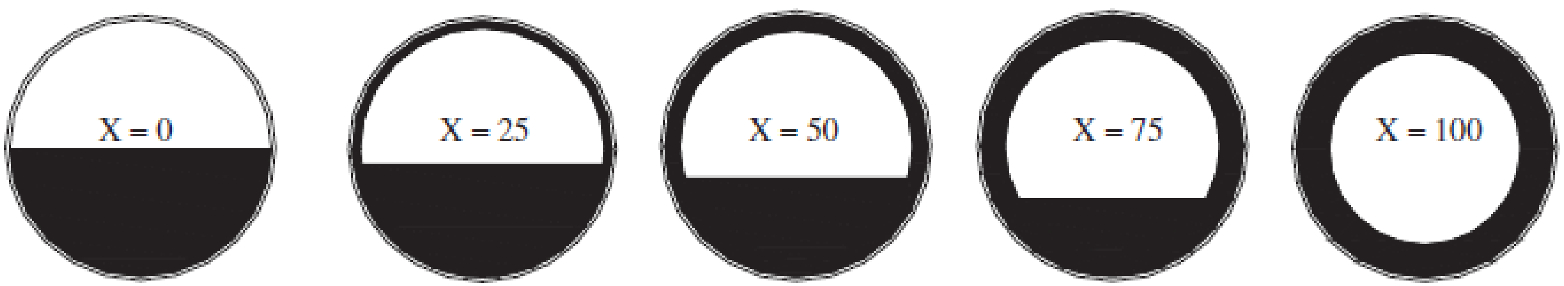
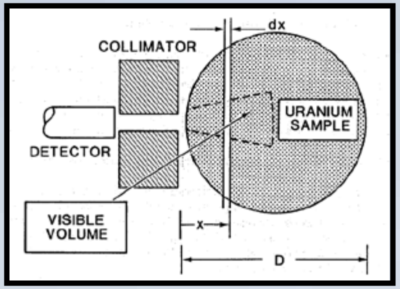


Fig. 7. UF_6 filling profiles.

Summarizing the problems

- **Typical NDA techniques measure enrichment, not isotope masses**
  - To obtain masses, a load cell (scale) measurement is necessary
- **The enrichment measurement relies upon weakly-penetrating particles**
  - Sensitive to container wall thickness
  - Sensitive to geometry
  - Cannot sample entire volume…what's in the center?



+

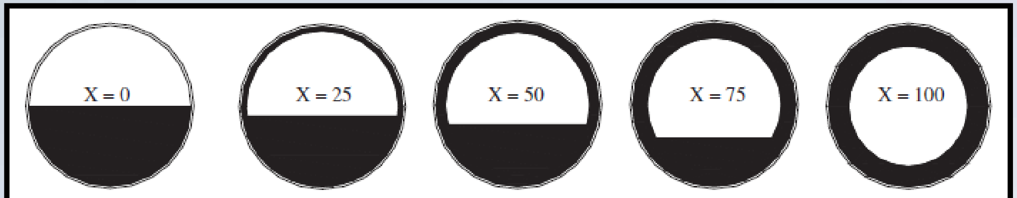
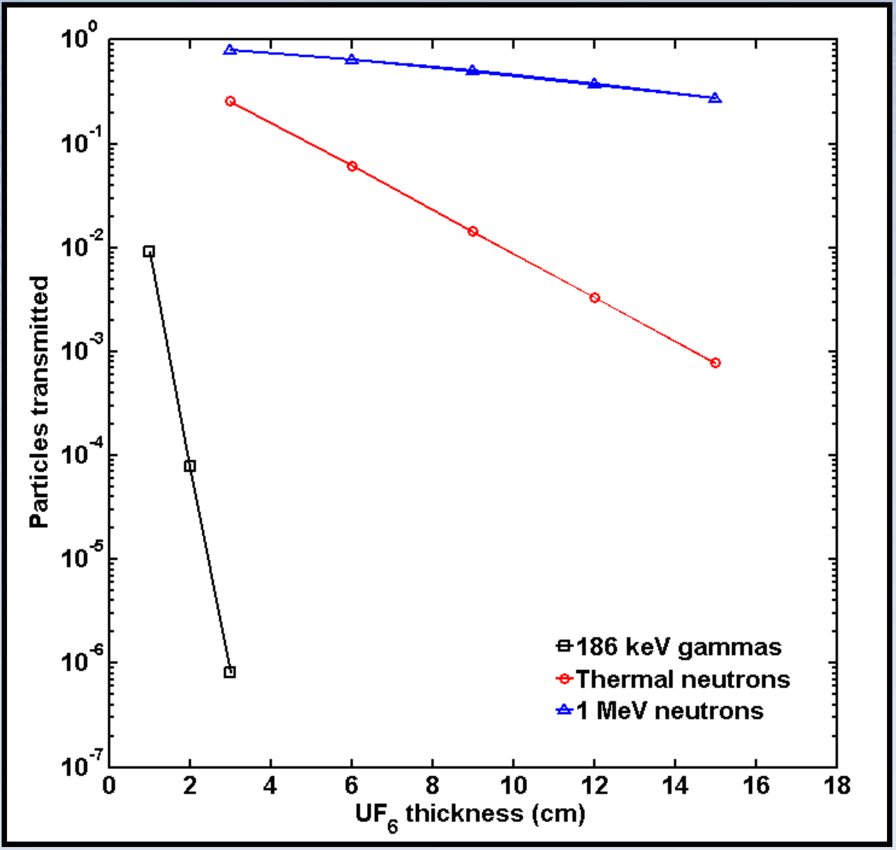


Fig. 7. UF_6 filling profiles.

= ?

Sandia's concept: directly measure fast neutron emissions

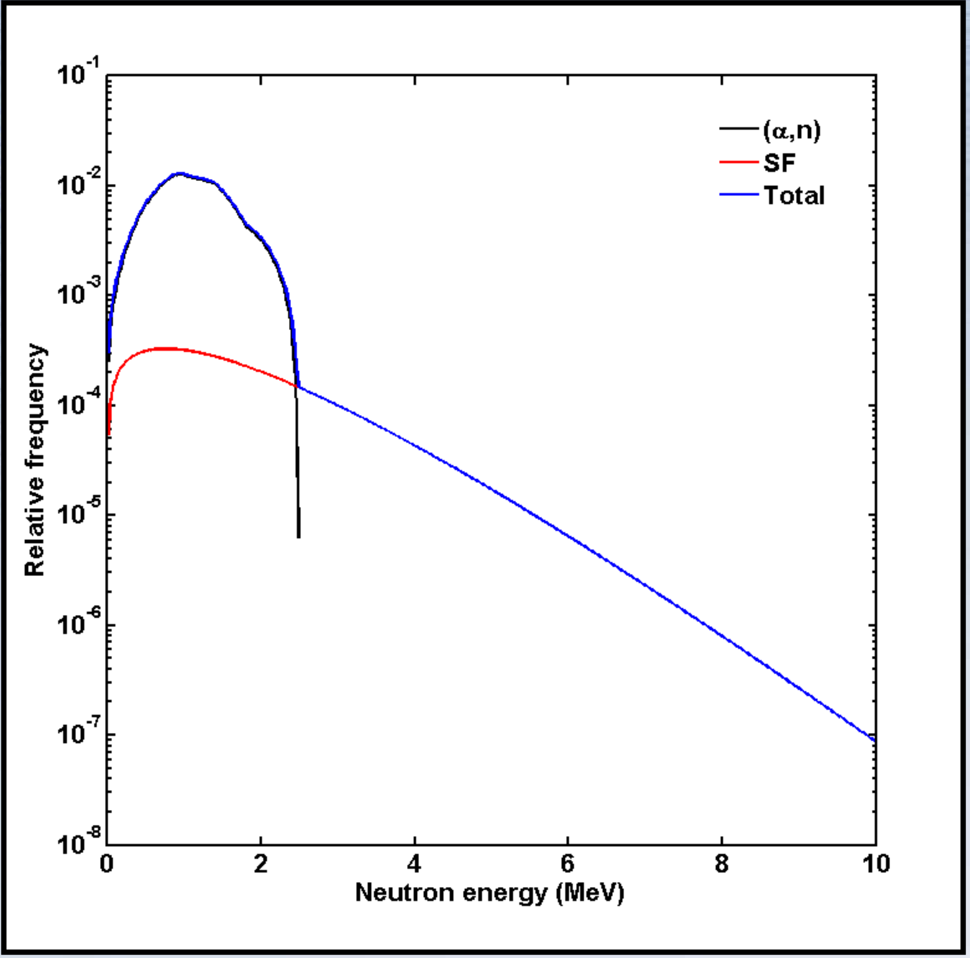
- **Fast neutrons generated by independent processes within the UF_6 can provide an independent enrichment measurement that samples the entire UF_6 volume**
- **Neutron imaging of the UF_6 distribution detects unexpected UF_6 geometries and applies necessary corrections**
- **Sandia has developed expertise in neutron imaging and spectroscopy that will enable success**



Transmission of particle beams through 5% enriched UF_6 (without container wall)

Neutron spectrometry can potentially be used to determine UF_6 enrichment and mass in a 30B

- ^{238}U: neutrons via spont. fission and (α,n) reaction on F atoms
- ^{234}U: neutrons via (α,n) reaction on F atoms
- The two processes have measurably different energy spectra
  - It should be possible to separate ^{234}U and ^{238}U contributions to the energy spectrum
  - Direct measurement of ^{234}U and ^{238}U masses
- ^{234}U content is proportional to ^{235}U content (proven by LANL for enrichment ≤ 5%)



SOURCES4C calculation of neutron spectrum for 5% enriched UF_6

Summarizing the concept…

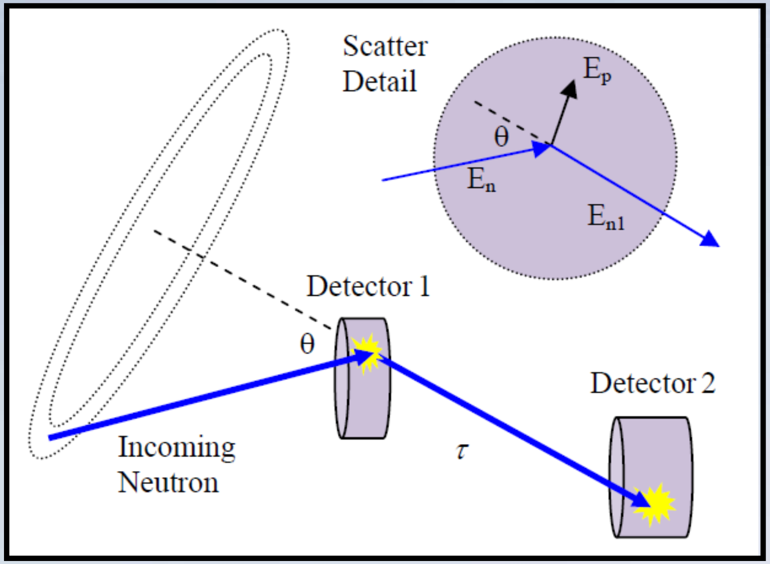
- **Spectral information**
  - The high-energy portion of the spectrum is purely from fission, and the magnitude is a function of the total ^{238}U mass
  - The low-energy portion of the spectrum is mostly from (α,n) on F, and the magnitude is a function of the total ^{234}U + ^{238}U mass
  - The sum of these components indicates the total sample mass
  - The ratio of these components is a function of uranium enrichment
- **Imaging information**
  - Imaging the material distribution may allow for geometry corrections to be applied (if necessary)
  - Imaging the total volume can provide confidence that diversions are not occurring (“smuggling” with an inner volume)

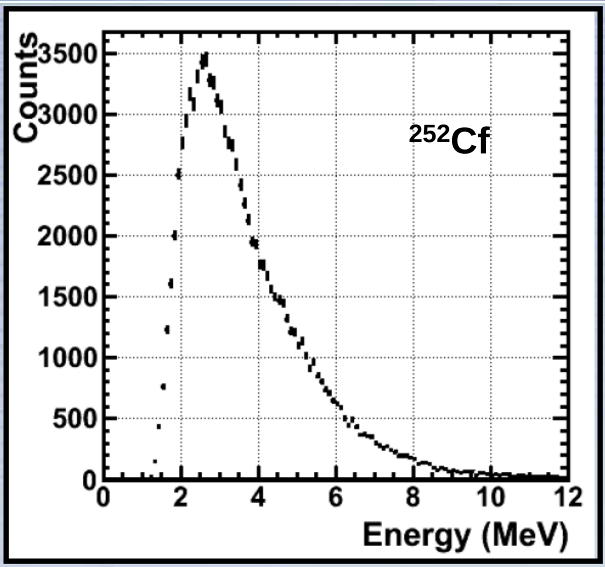
Advantages of this technique

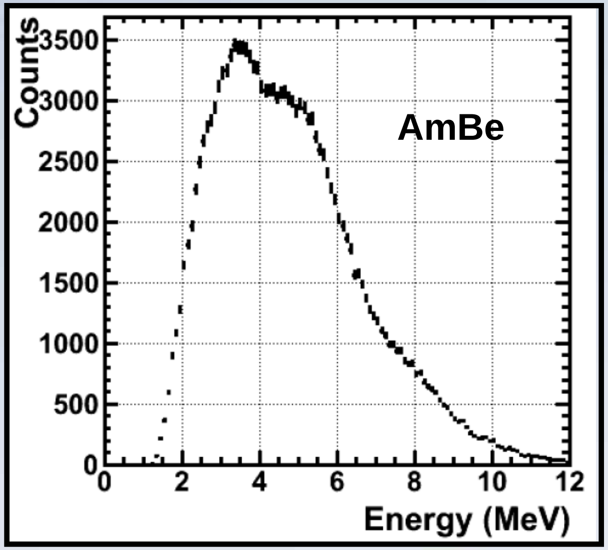
- **Independent verification of isotope ratios and masses**
- **Highly-penetrating particles are less sensitive to geometry perturbations**
- **No load cell necessary**
- **Imaging can be used to:**
  - Map material distribution
  - Reject natural backgrounds
  - Reject neutrons from nearby cylinders

Neutron spectrometry measurements can be performed with the Neutron Scatter Camera

- **The Neutron Scatter Camera is a mature system developed at Sandia for large-area search**
  - Multi-element system
  - Liquid scintillator for n/γ discrimination
  - Imaging capabilities (interaction cell locations, measured energies)
  - Spectrometry (deposited energy, time-of-flight)

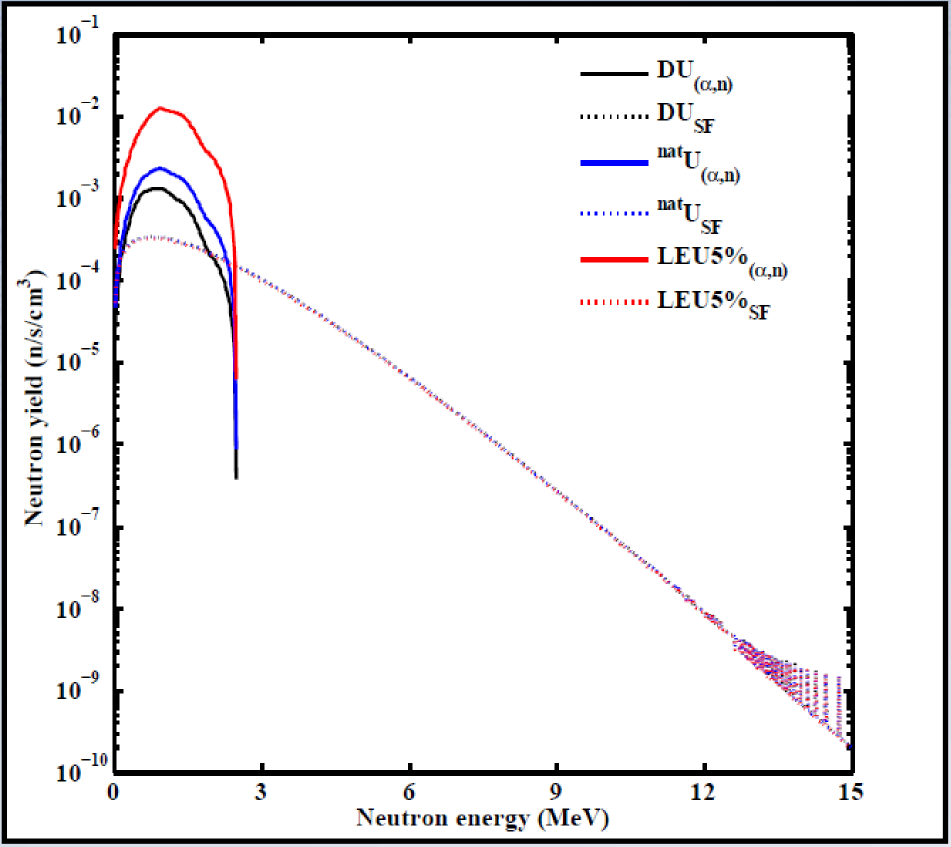






Initial calculations: emitted neutron spectra using SOURCES4C

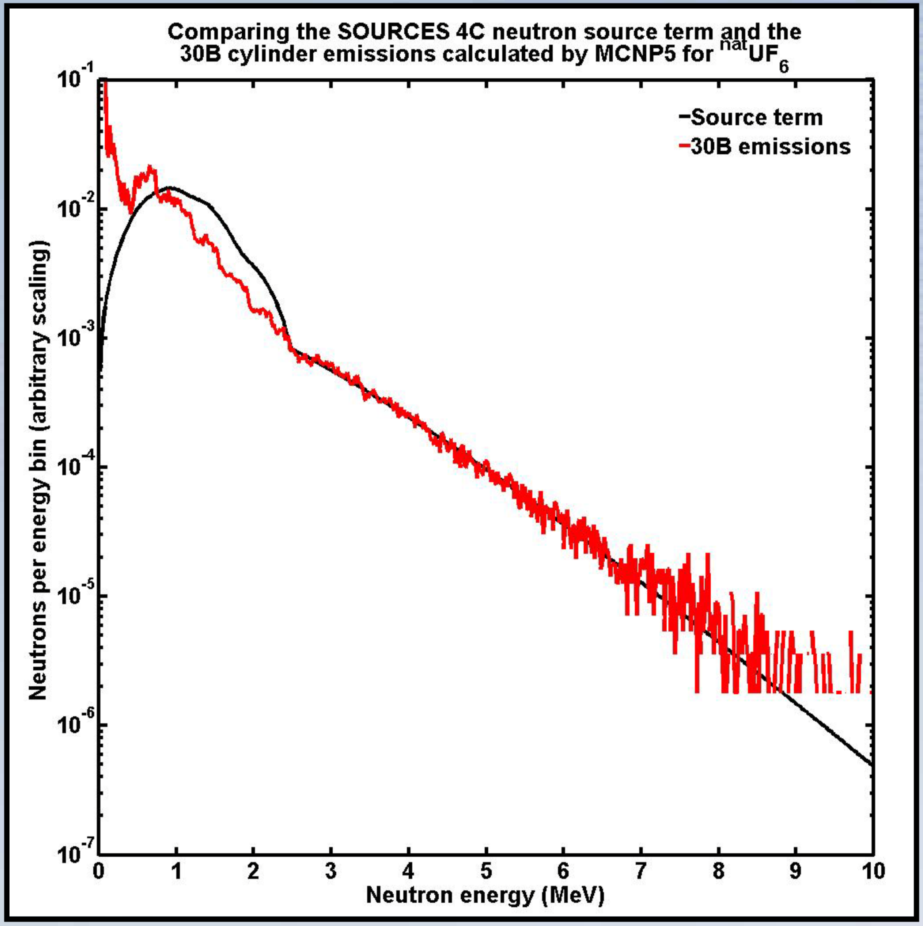
- **Calculate spectra for different enrichments to examine the dependence**
- **Use SOURCES4C**
  - Input: isotopics, density, energy bin boundaries
  - Output: (α,n), spontaneous fission rates (n/cm³s)
- **No detector response or transport physics in cylinder at this point**

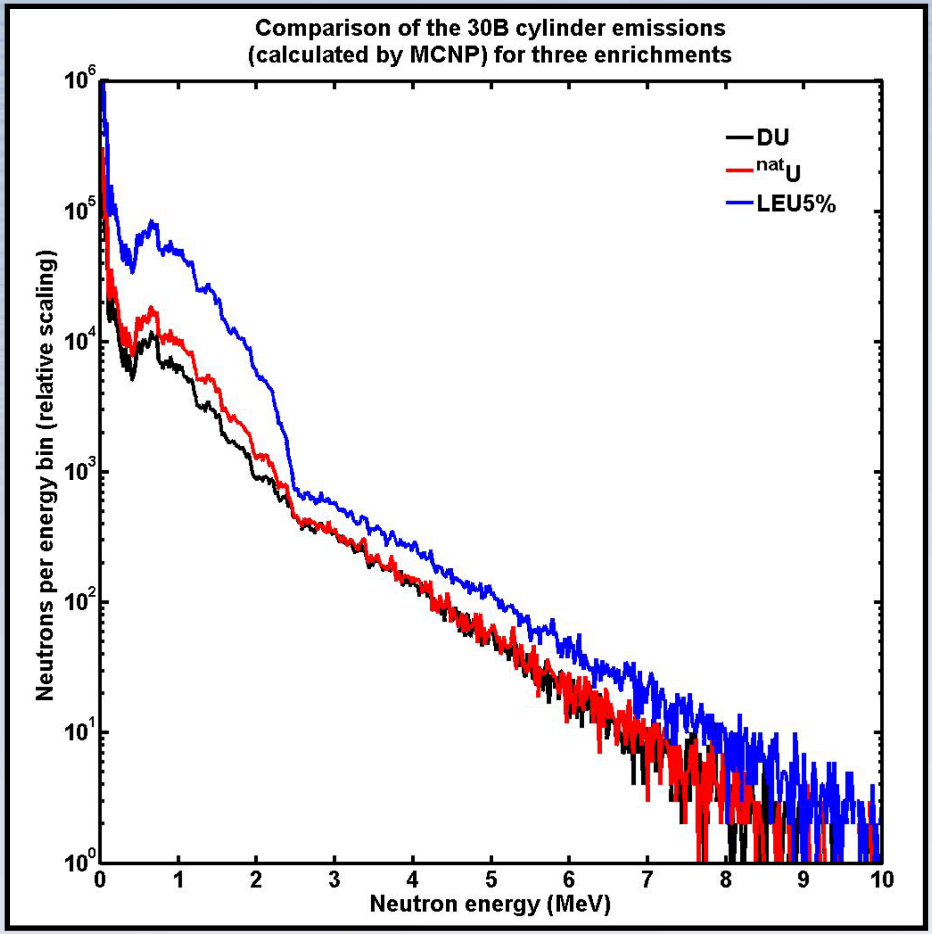


Calculations of the cylinder emissions imply manageable spectral perturbations

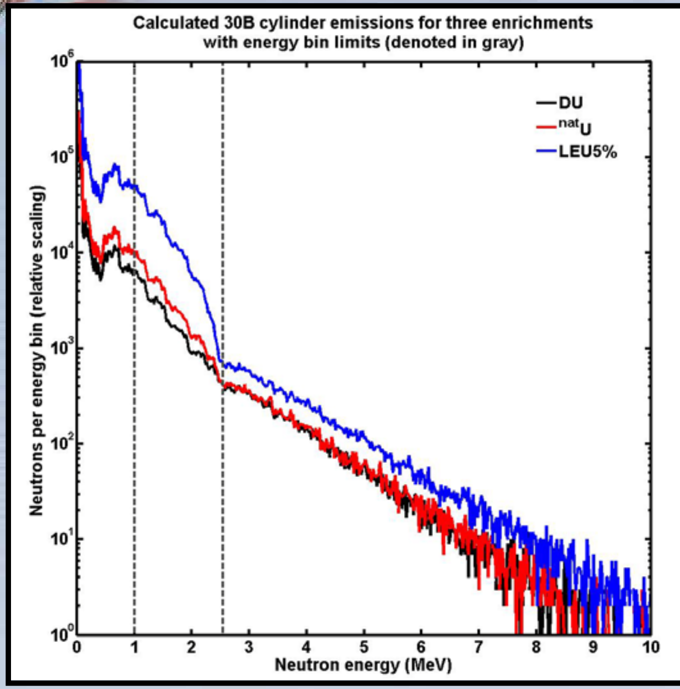
- **The source term is perturbed in a large mass of UF_6.**
  - Scattering
  - Induced fission
  - Absorption
- **A 30B cylinder was modeled in MCNP5.**
  - Enrichments: DU, ^{nat}U, 5% enriched ^{235}U
  - Maximum fill mass
- **Spectra appear to maintain enough structure for the measurement concept to work.**

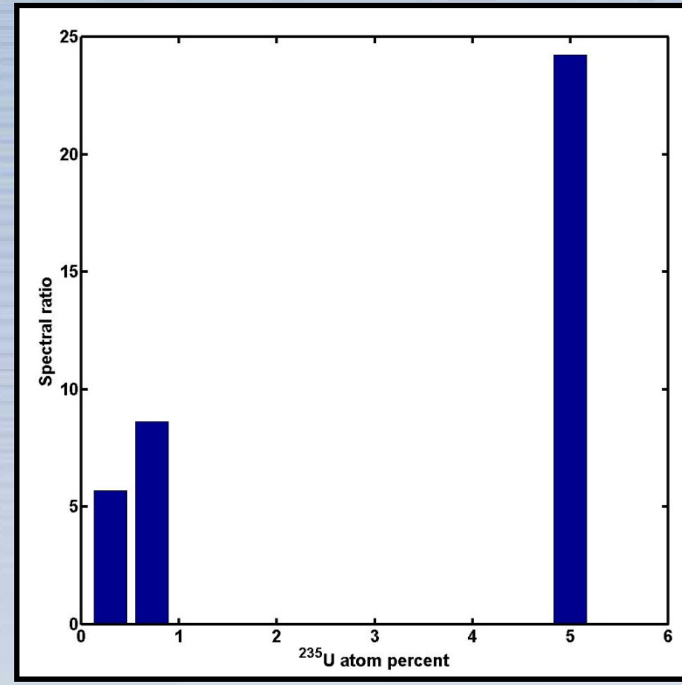
Results of the MCNP5 calculations





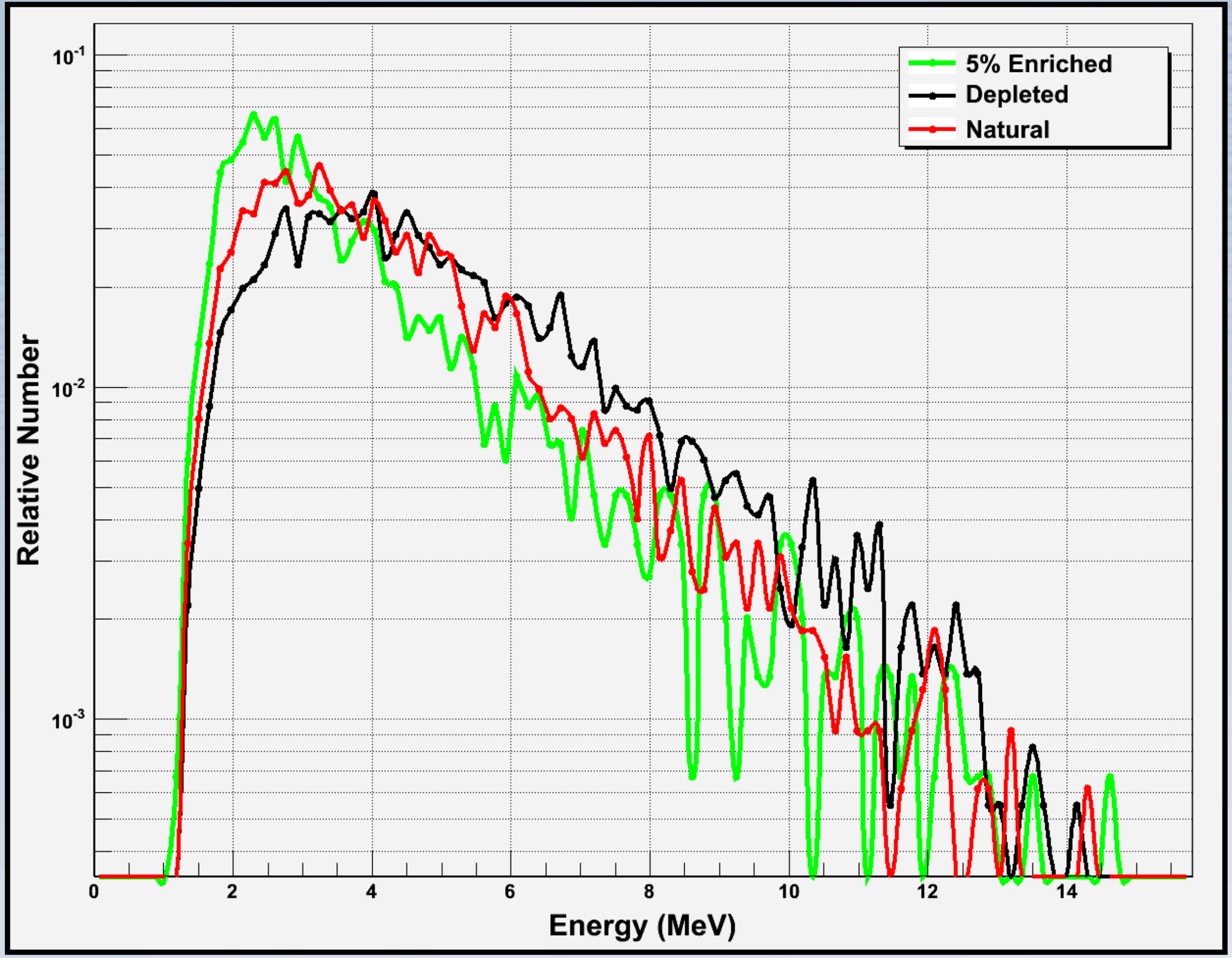
The enrichment can be inferred from the neutron energy spectrum





- **The ratio of neutrons in the (α,n) and S.F. regions is a function of enrichment**
  - Cut data at the end of the (α,n) spectrum (~2.54 MeV)
  - A realistic detector will have a detection threshold (choose 1 MeV)
- **For the simulated data, the ratio is a monotonic function of enrichment**

The (simulated) 30B emissions with full detector response



These spectra are normalized to more easily compare the distributions

Challenge: filling profile differences may require imaging

- **Imaging may be necessary to couple with spectrometry**

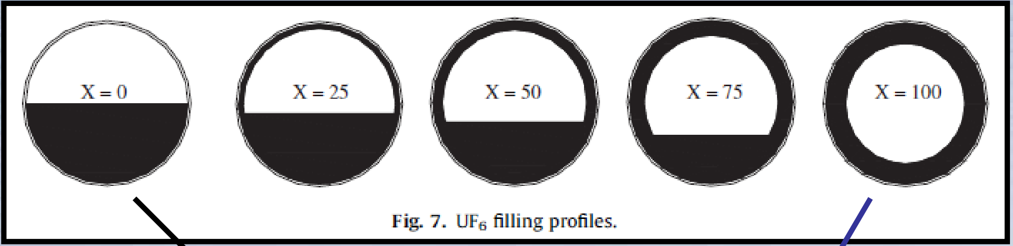
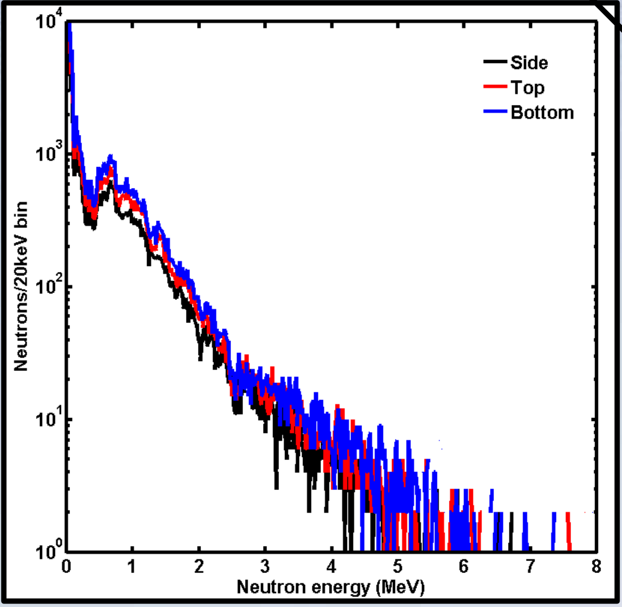
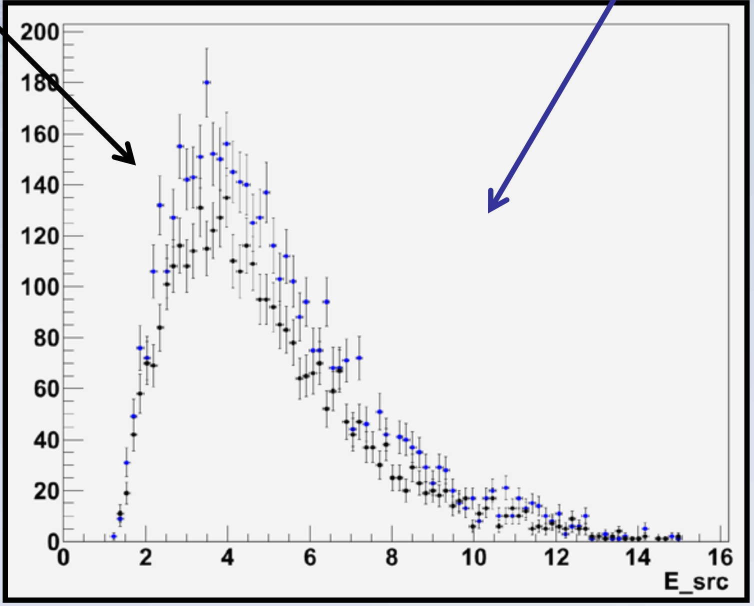


Fig. 7. UF_6 filling profiles.





Three measurement locations for X=0, $^{nat}UF_6$

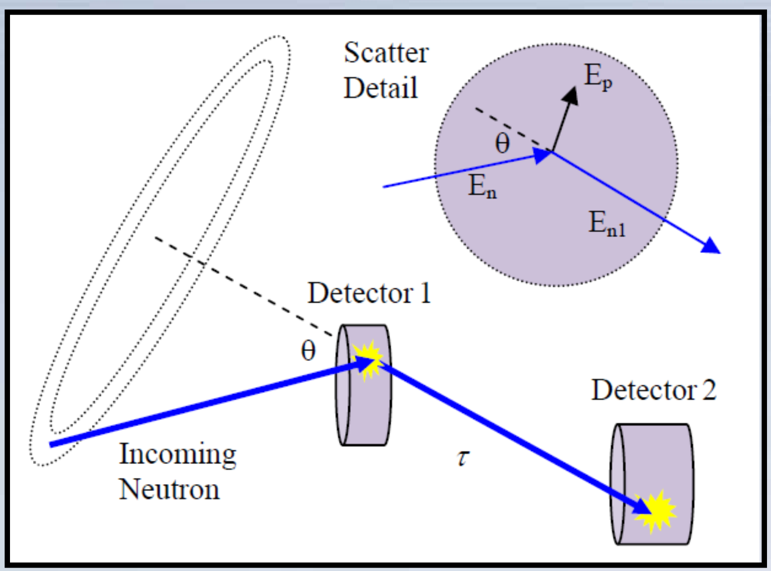
DUF_6: X=0 (black), X=100 (blue)

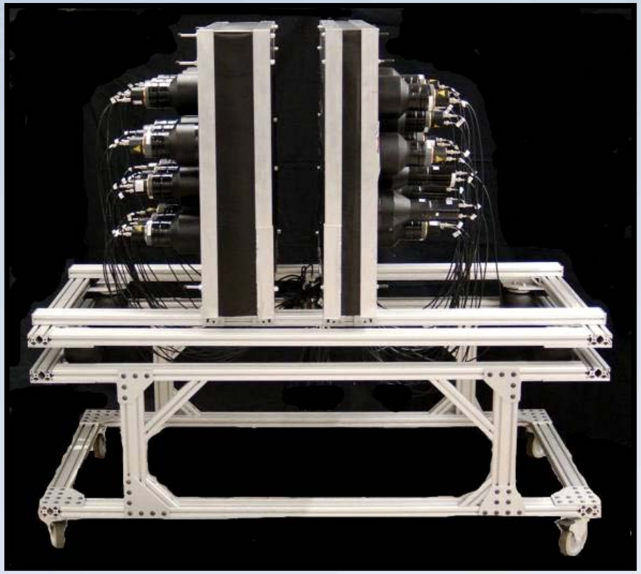
LDRD LABORATORY DIRECTED RESEARCH & DEVELOPMENT

Sandia National Laboratories

Experiments to characterize the Neutron Scatter Camera

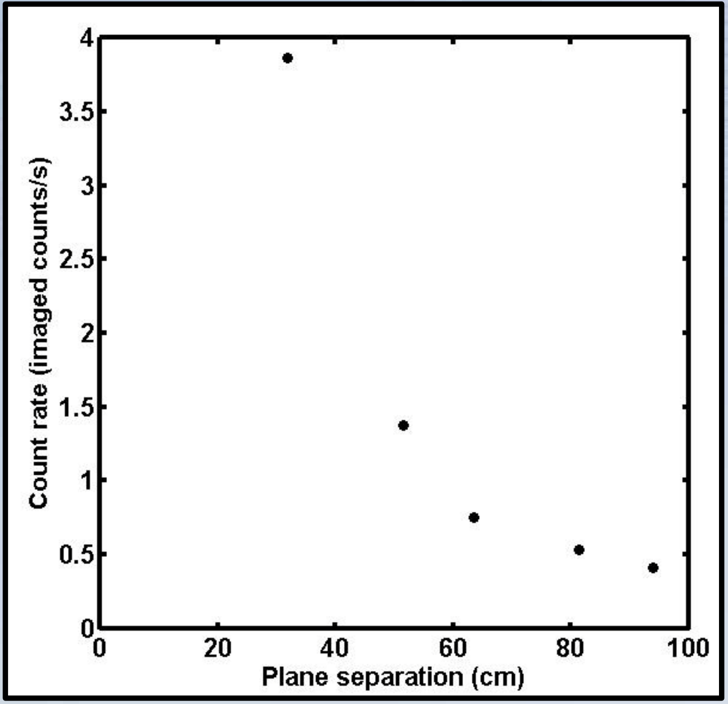
- **We want to understand the tradeoff between energy resolution and double scatter rates as a function of plane spacing for the Neutron Scatter Camera**
  - Close planes = high rate, degraded spectrum
  - Extended planes = low rate, best spectrum

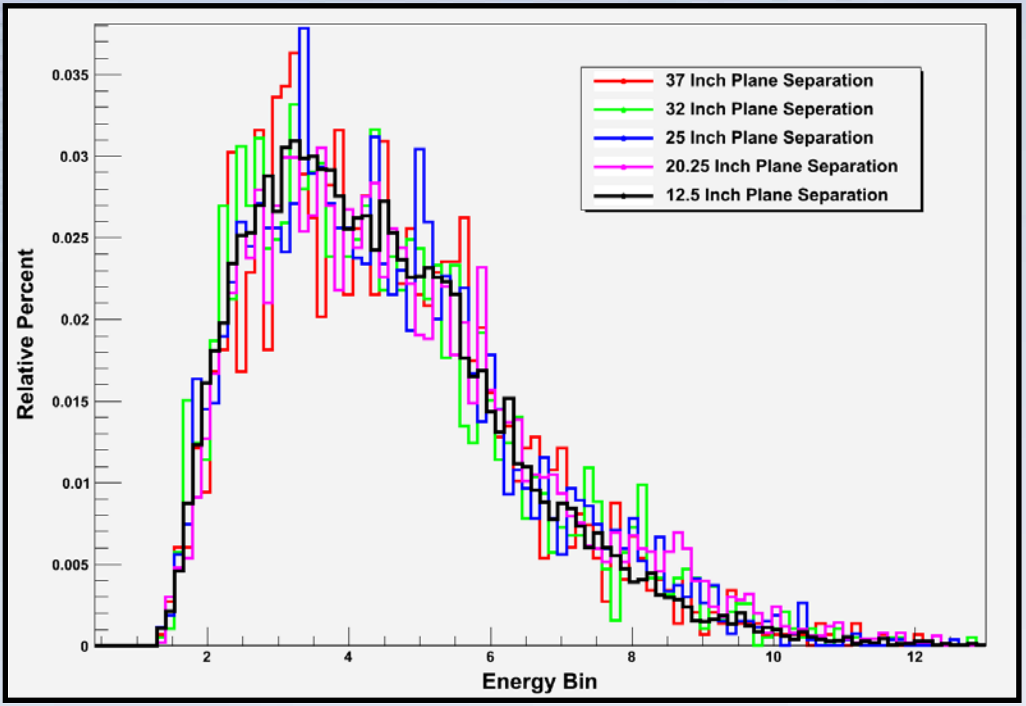




Experiment: rates vs. plane spacing

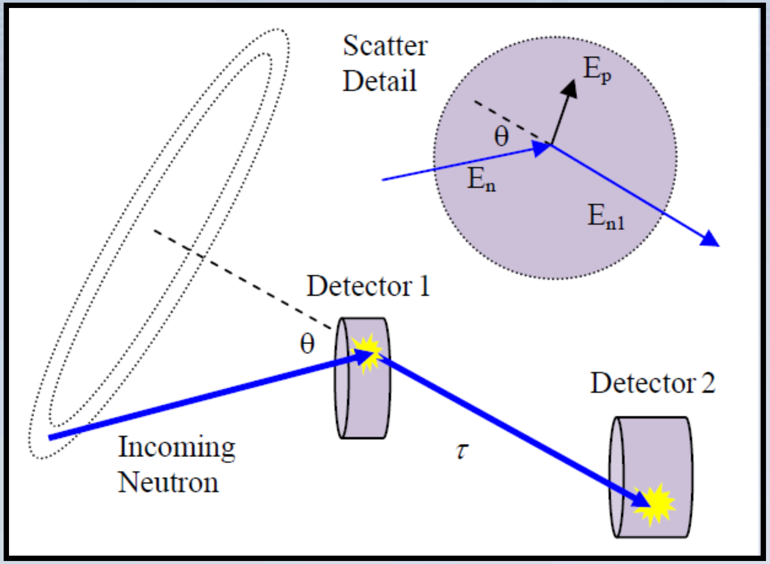
- Used an AmBe source in the B942 high bay
- DTRA camera with 4 rear cells missing (12+8)

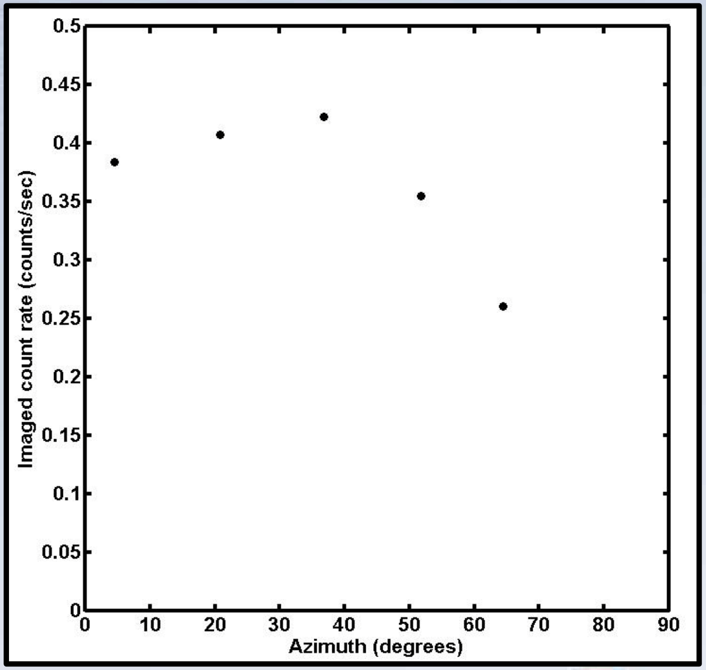




Experiments: rates vs. camera angle

- **The rate of imaged events will vary with the angle of the Neutron Scatter Camera relative to the source**
  - Angles between cell pairs changes
  - Alters the energy partitioning between cells for identical neutrons



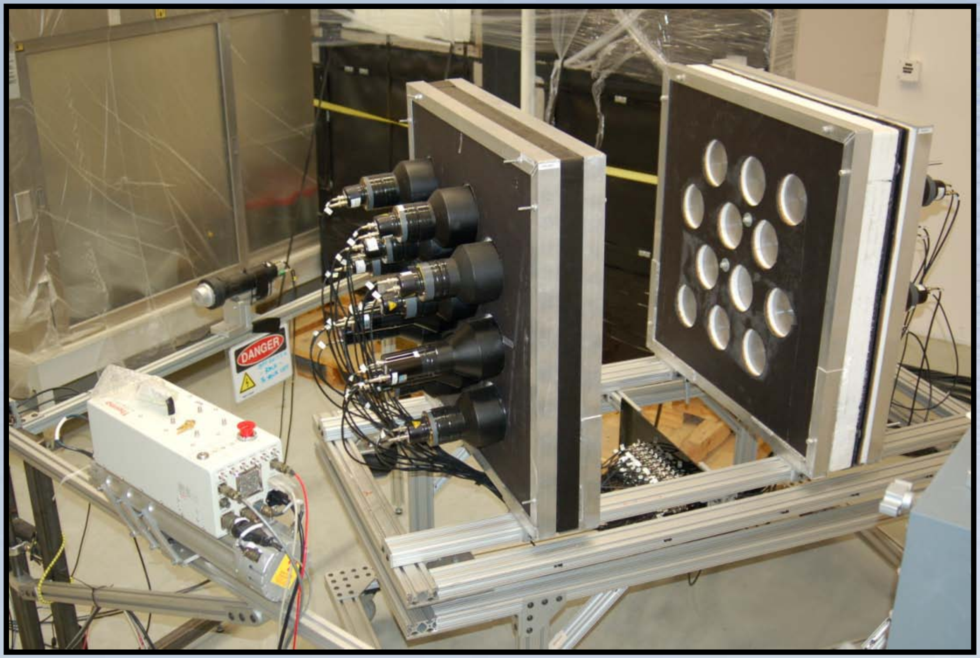


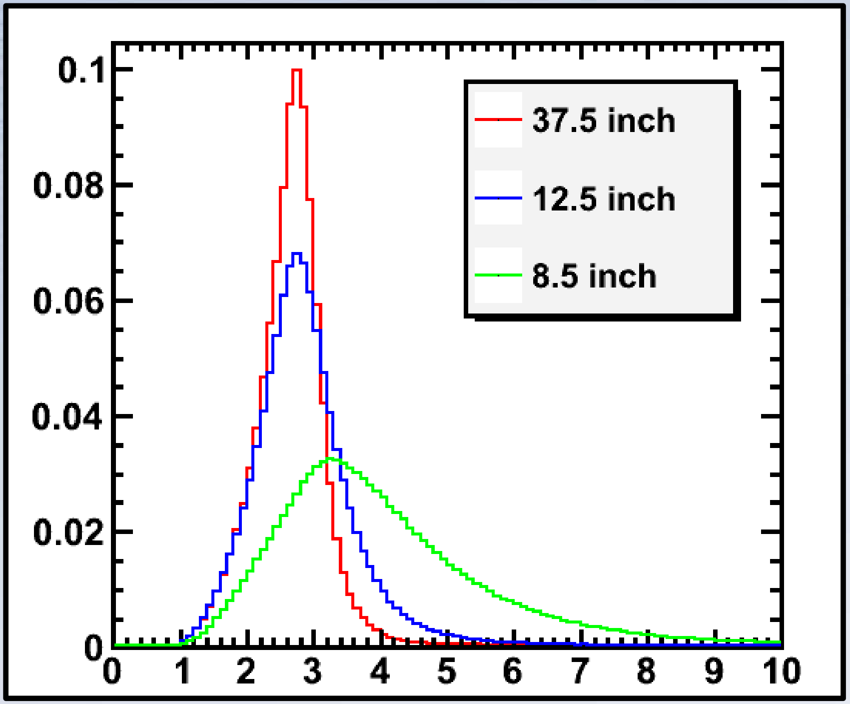
Quantifying the change in energy resolution

- **The AmBe experiments are fine for rate studies, but it is difficult to quantify the energy resolution vs. plane spacing**
- **Use a D+D neutron generator**
  - Monoenergetic neutrons of an appropriate energy: 2.45 MeV
  - Over time, also build in 14.1 MeV neutrons

$$D + D \rightarrow \begin{cases} {}^{3}He + n \\ T + p \end{cases}$$

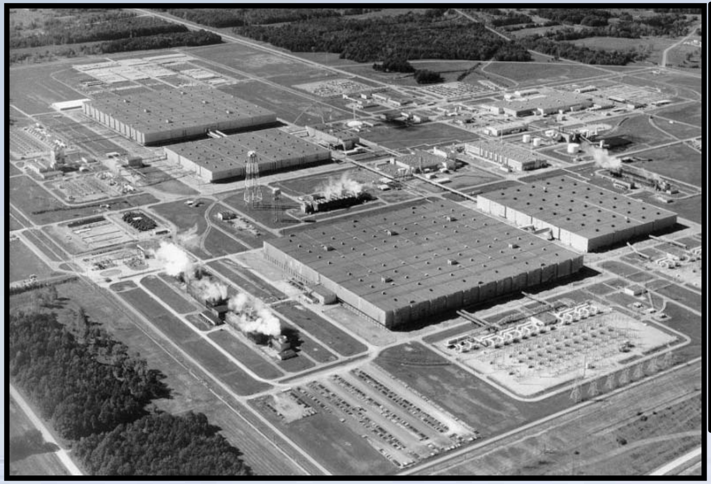
D+D source experiments





Planning: field measurements

- **It is important to measure filled 30B cylinders with the Neutron Scatter Camera to collect data with**
  - The appropriate (α,n) source term
  - A complex source with multiplication, scattering
  - Appropriate rates (neutrons and gammas)
  - Realistic backgrounds
- **Paducah Gaseous Diffusion Plant may be the place**

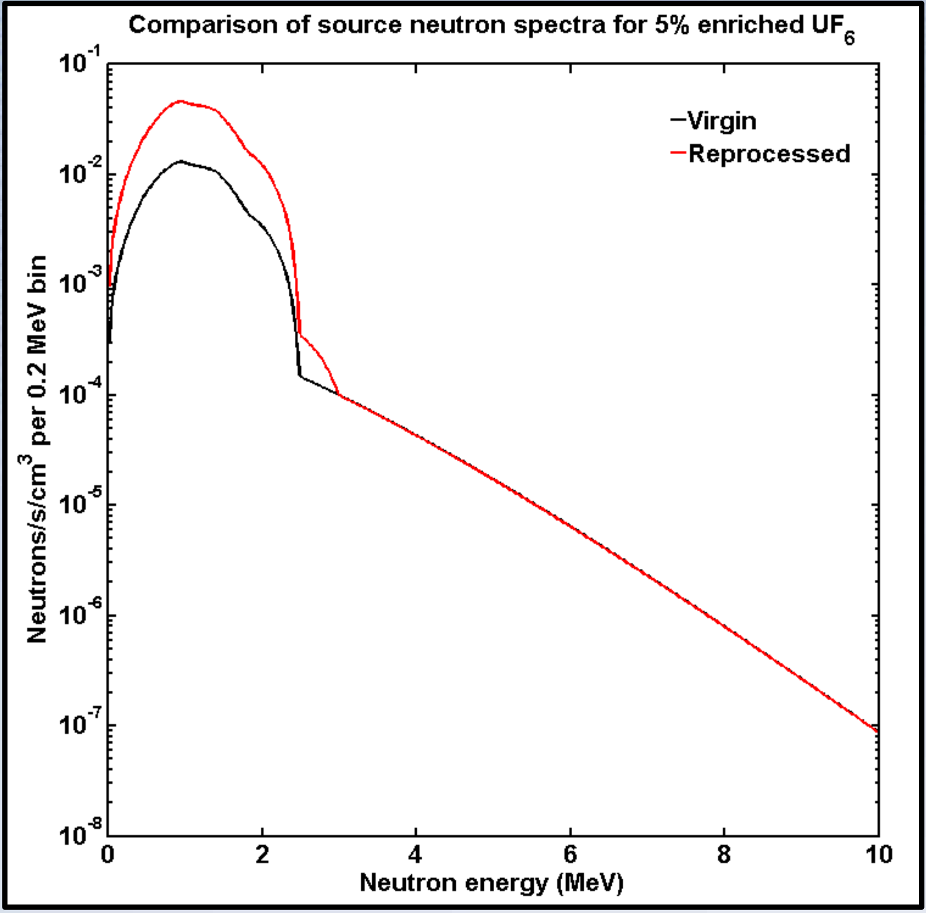


Discussion and Summary

- **Direct measurement of neutron signatures for UF_6 material accountancy appears to be a useful safeguards technique**
  - Two physical processes create neutrons with different energy spectra
  - Simulations indicate enrichment can be extracted from emitted neutrons, even after full transport
- **Advantages:**
  - Sensitive to entire cylinder volume (detect diversion attempts)
  - Imaging suppresses backgrounds from nearby cylinders, allows one to map material distribution within a cylinder (if important)
- **Accomplishments:**
  - Simulations show spectrum changes with enrichment (transport, detector response included)
  - Basic lab experiments to characterize Neutron Scatter Camera

Challenge: virgin vs. reprocessed UF_6

Isotopes	Amount (%)	Neutron source 1/s/ton UF_6 Spontaneous fission	Neutron source 1/s/ton UF_6 $UF_6(\alpha,n)$	Relative yield (%)
Depleted U				
^{232}U	0	0	0	0
^{234}U	0.00234	0	7.85E+03	28.9
^{235}U	0.3	0	1.86E+03	6.9
^{236}U	0	0	0	0
^{238}U	99.6977	9.96E+03	7.53E+03	64.3
Natural U				
^{232}U	0	0	0	0
^{234}U	0.0056	0	1.88E+04	51.3
^{235}U	0.718	0	4.45E+02	1.2
^{236}U	0	0	0	0
^{238}U	99.2764	9.92E+03	7.49E+03	47.5
LEU 5%				
^{232}U	0	0	0	0
^{234}U	0.039	0	1.31E+05	86.9
^{235}U	5	0	3.10E+03	2.1
^{236}U	0	0	0	0
^{238}U	94.961	9.50E+03	7.17E+03	11.1
LEU 5% repr.				
^{232}U	7.60E−07	0	1.66E+04	3.26
^{234}U	0.133	0	4.46E+05	87.48
^{235}U	5	0	3.10E+03	0.61
^{236}U	1.55	0	2.76E+04	5.41
^{238}U	93.317	9.50E+03	7.04E+03	3.24



Considering PuO_2 holdup accountancy

- **Consider PuO_2 holdup in a reprocessing facility**
  - Gloveboxes
  - Pipes, components
- **Geometry is variable, materials self-absorbing**
  - Gamma-based measurements are not accurate
- **Can spectrometry and imaging be used to locate and quantify material?**

La Hague reprocessing facility

Plutonium vs. uranium measurements

- **Consider PuO_2. How do the proposed measurements compare to UF_6?**

- **PuO_2 advantages:**
  - Spectrum: (α,n) on O produces higher-energy neutrons
  - Rates: There are >1000x more neutrons emitted from PuO_2 per cm^3
- **PuO_2 disadvantage:**
  - Isotopics are messy…f(initial enrichment, burnup, cooling time)

Considering the PuO_2 neutron spectrum

- Each isotopic mix produces unique (α, n) and SF neutron spectra
- Ideal: use spectrum to estimate each isotope's individual contributions and mass
- Reality: not enough information; must use (α,n), SF components only (for pure neutron measurements)