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Fast Neutron Signatures for Uranium Hexafluoride Enrichment Measurements

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Livermore, CA

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Gas centrifuge enrichment plant, Piketon, OH

U.S. DEPARTMENT OF ENERGY

NNSA National Nuclear Security Administration

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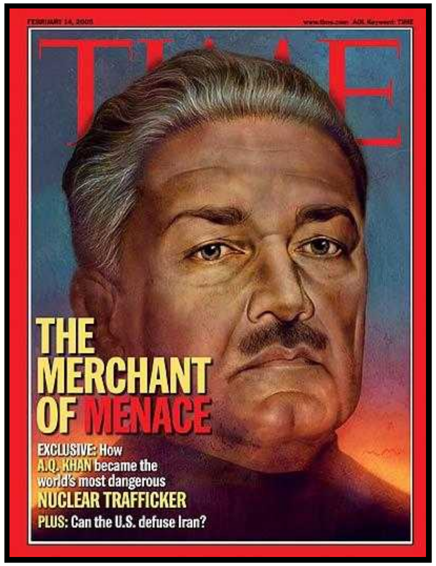
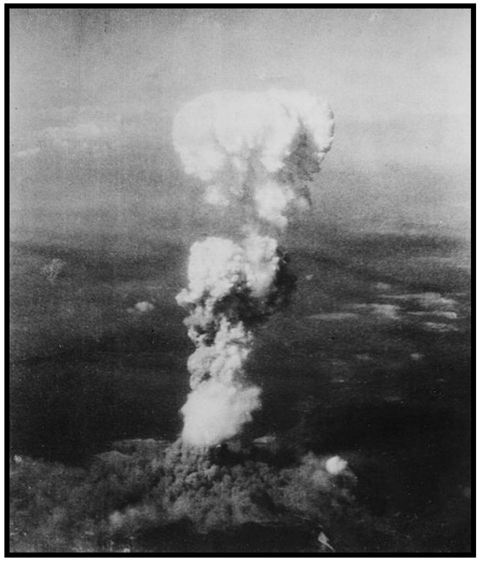
A Rough Presentation Outline

- Introduction to the problem
- How we got involved
- Using fast neutrons
- Initial calculations
- Measurements
- 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





A quick review: the front end of the fuel cycle

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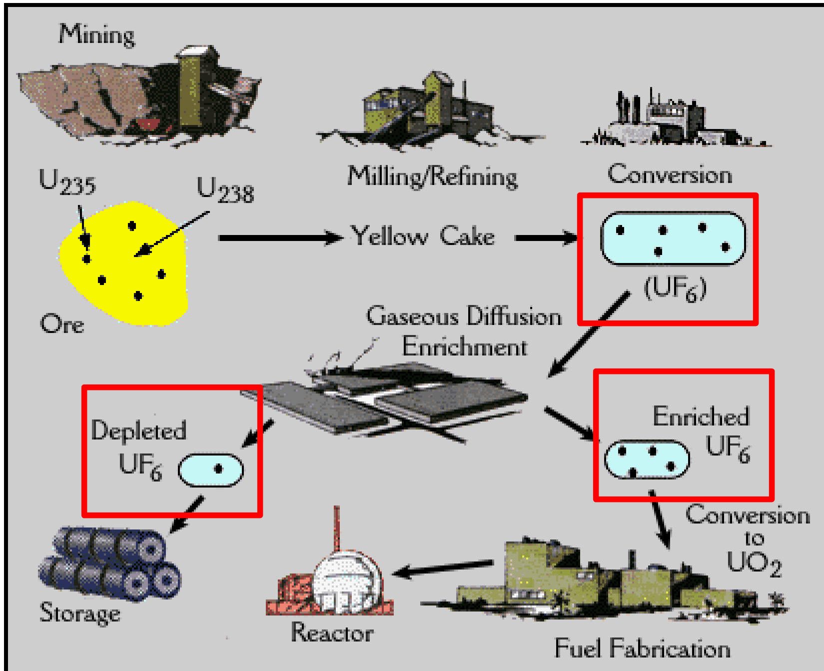
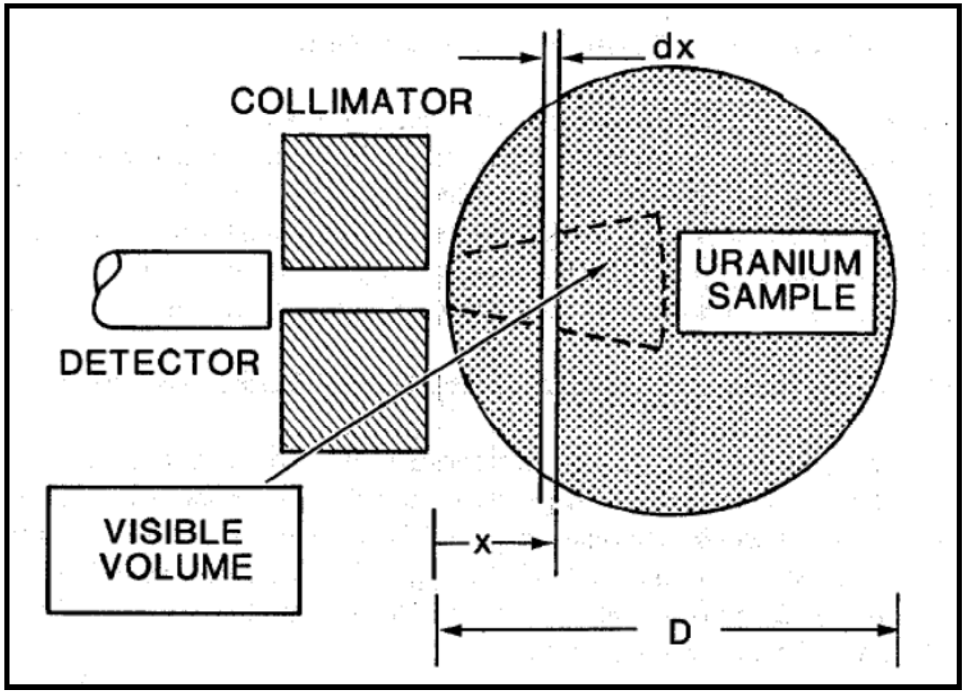


[illegible]	[illegible]	ial of uction	Minimum volume ft[3]	Minimum volume liters	Approximate tare weight (without valve protector) lb	Approximate tare weight (without valve protector) kg	Maximum, enrichment wt % uranium-235	Shipping Limit Maximum,[a] UF_6 lb	Shipping Limit Maximum,[a] UF_6 kg
[illegible]	[illegible]	kel	0.0053	0.15	1.75	0.79	100.00	1.0	0.45
[illegible]	[illegible]	[illegible]kel	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

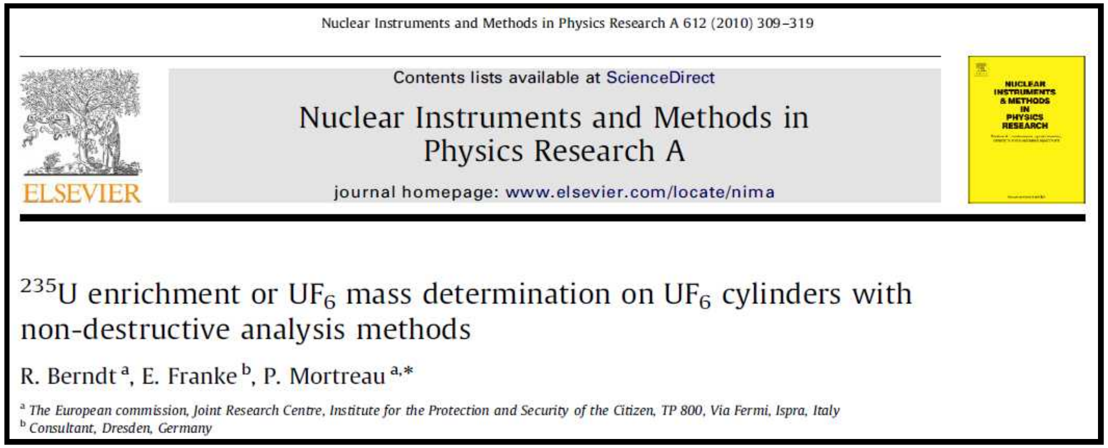
Current technology is good, but…

- "Enrichment meter" measures gamma emissions from the uranium hexafluoride (UF6)
  - Sensitive to variations in container wall thickness
  - Not sensitive to material beyond outer skin of UF6
  - Gives local enrichment, not total mass (need a scale for masses)



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

How we got involved…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ELSEVIER

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

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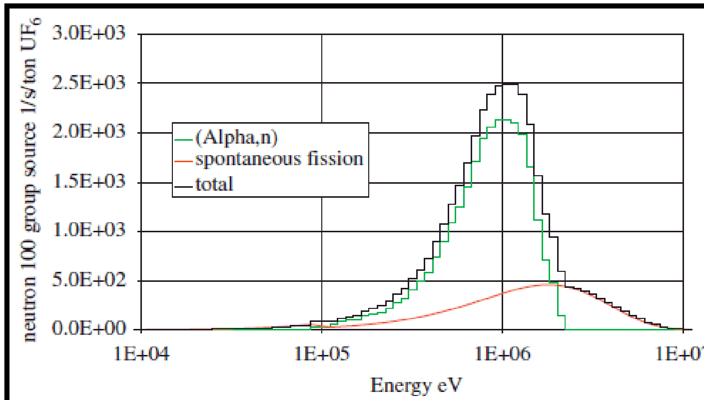


Fig. 4. Hundred group neutron spectra for natural UF_6 [7].

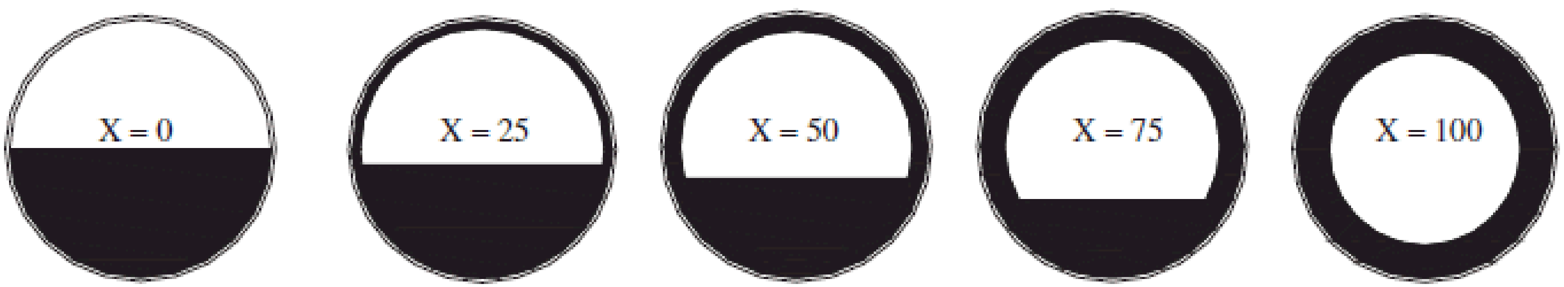
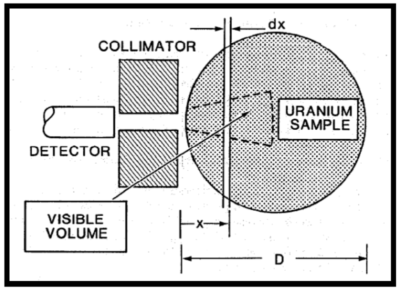


Fig. 7. UF_6 filling profiles.

Summarizing the problems

- 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?
- Typical NDA techniques measure enrichment, not isotope masses
  - To obtain masses, a load cell (scale) measurement is necessary



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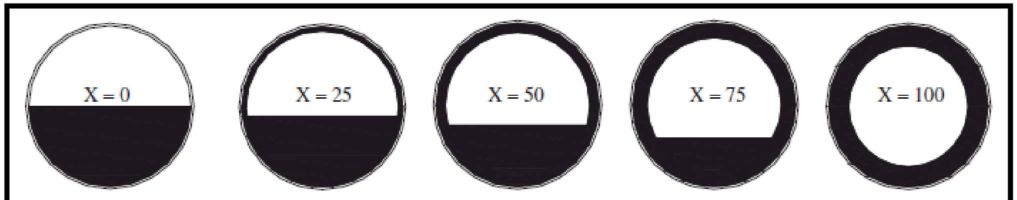
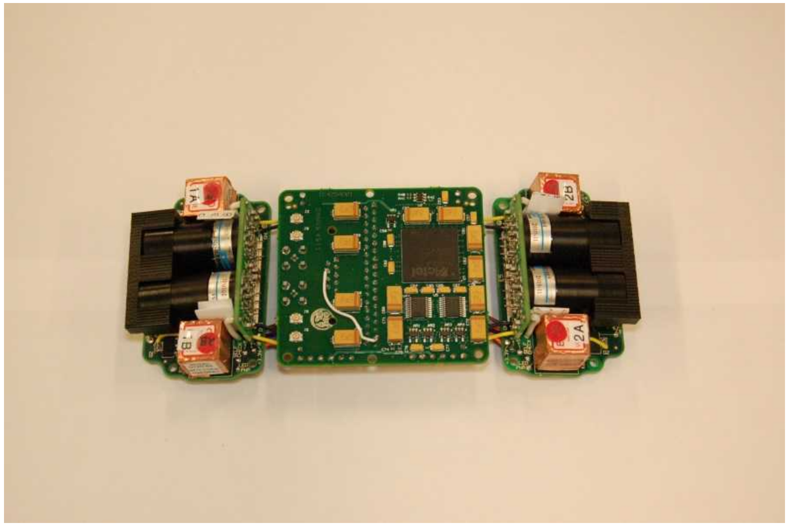
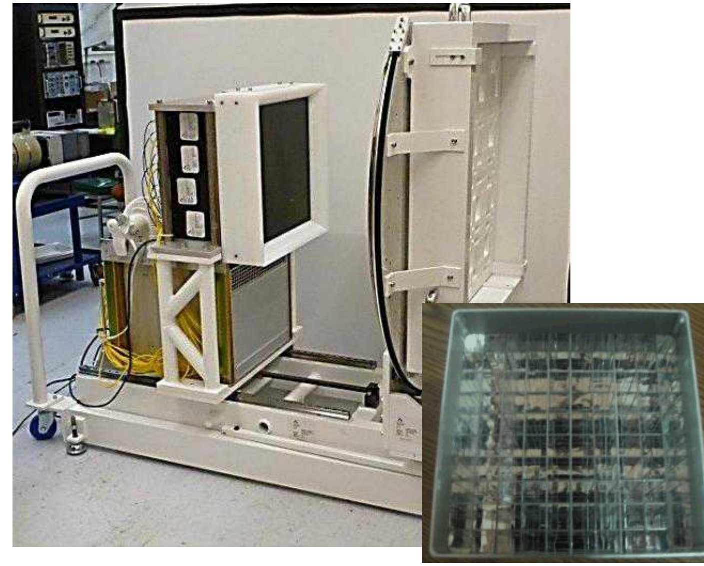


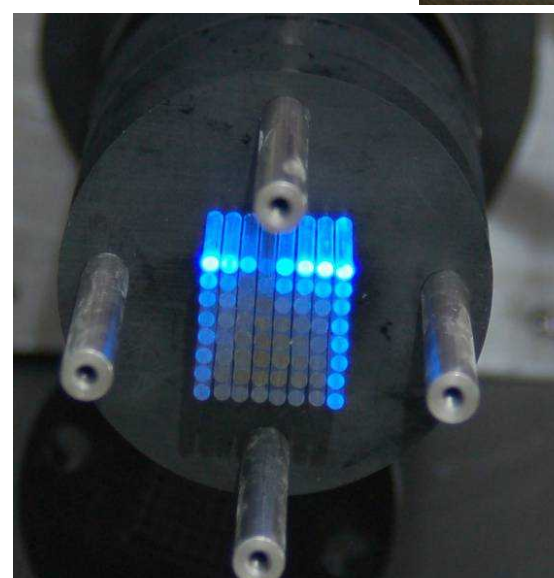
Fig. 7. UF_6 filling profiles.

= ?

Our view of the world: fast neutrons







Sandia's concept: directly measure fast neutron emissions

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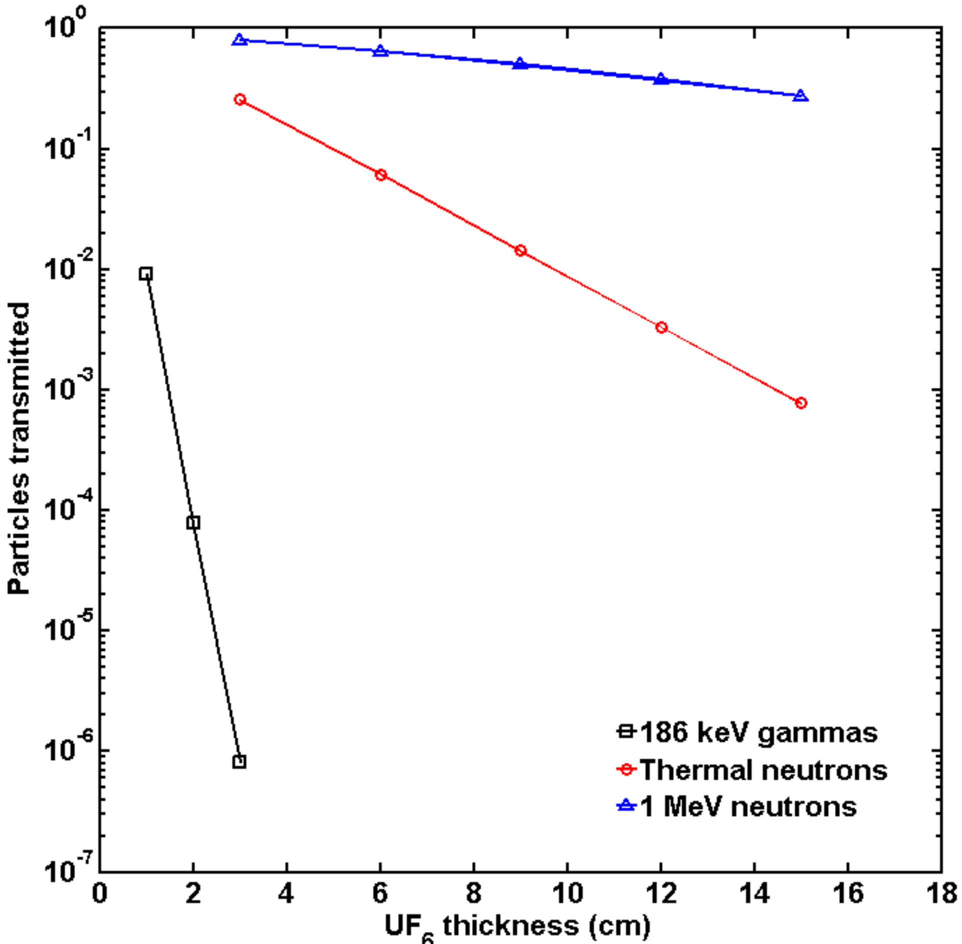
- Fast neutrons generated by independent processes within the UF_6 can provide an independent enrichment measurement that samples the entire UF_6 volume
- Neutron imaging of the UF_6 distribution detects unexpected UF_6 geometries and applies necessary corrections
- Sandia has developed expertise in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



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

Neutron spectrometry may be used to determine UF_6 enrichment & mass in a 30B

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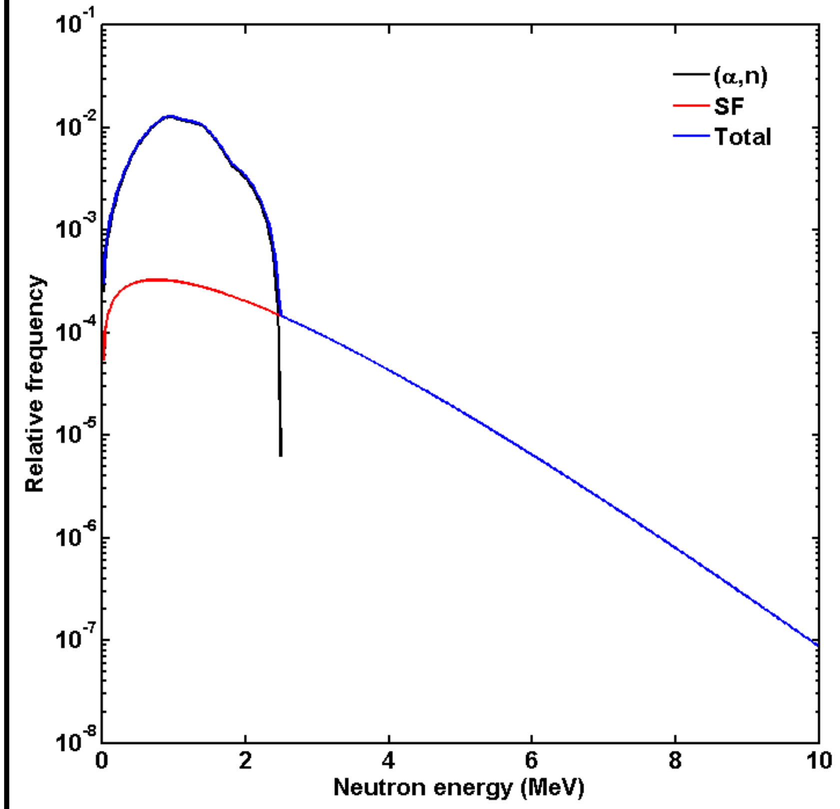
- ^{238}U: neutrons via spont. fission and (α,n) reaction on F atoms
- ^{234}U: neutrons via (α,n) reaction on F atoms
- The two processes have measurably different energy spectra
  - It should be possible to separate ^{234}U and ^{238}U contributions to the energy spectrum
  - Direct measurement of ^{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 the container geometry is consistent with expectations

Advantages of this technique

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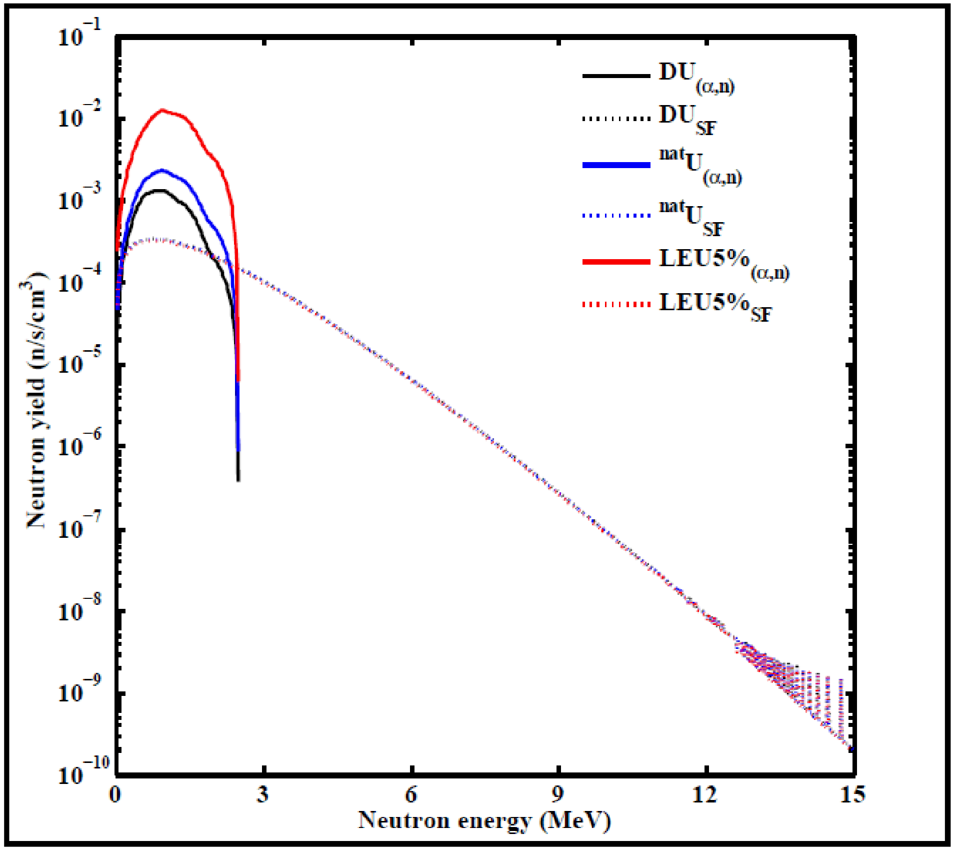
- Independent verification of isotope ratios and masses
- Highly-penetrating particles are less sensitive to geometry perturbations
- No load cell necessary
- Imaging can be used to:
  - Map material distribution
  - Reject natural backgrounds
  - Reject neutrons from nearby cylinders

Initial calculations: emitted neutron spectra using SOURCES4C

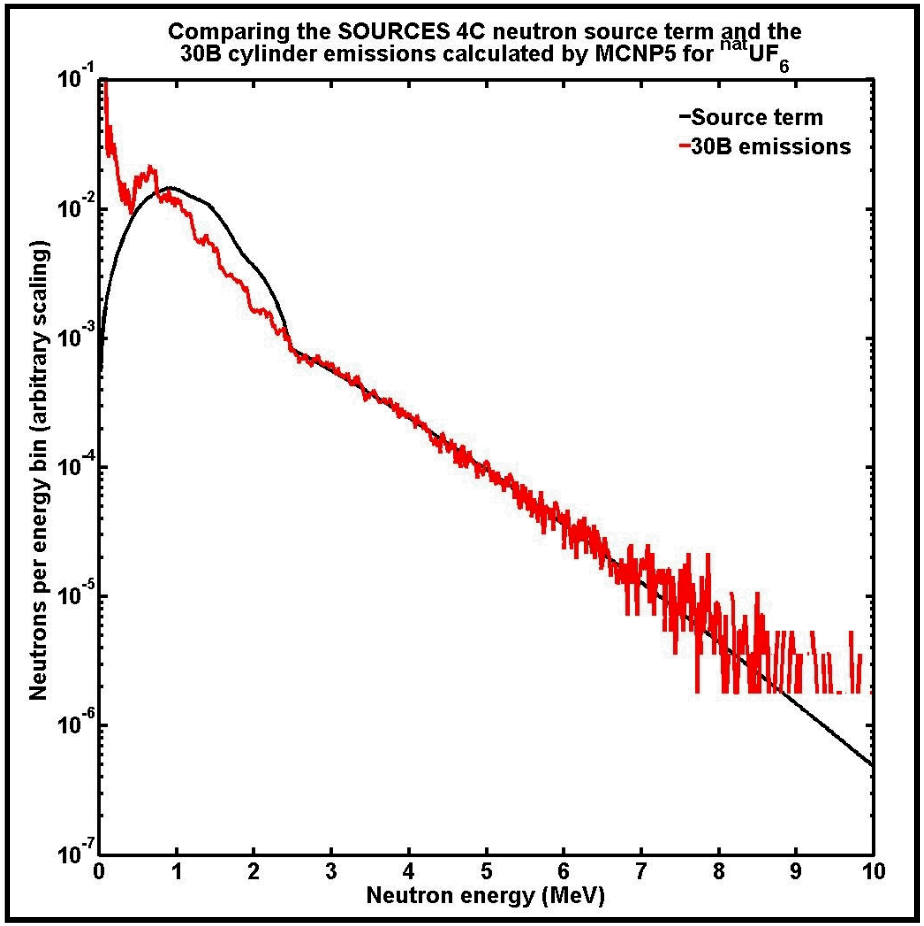
- Calculate spectra for different enrichments to examine the dependence
- Use SOURCES4C
  - Input: isotopics, density, energy bin boundaries
  - Output: (α,n), spontaneous fission rates (n/cm^{3}s)
- No detector response or transport physics in cylinder at this point

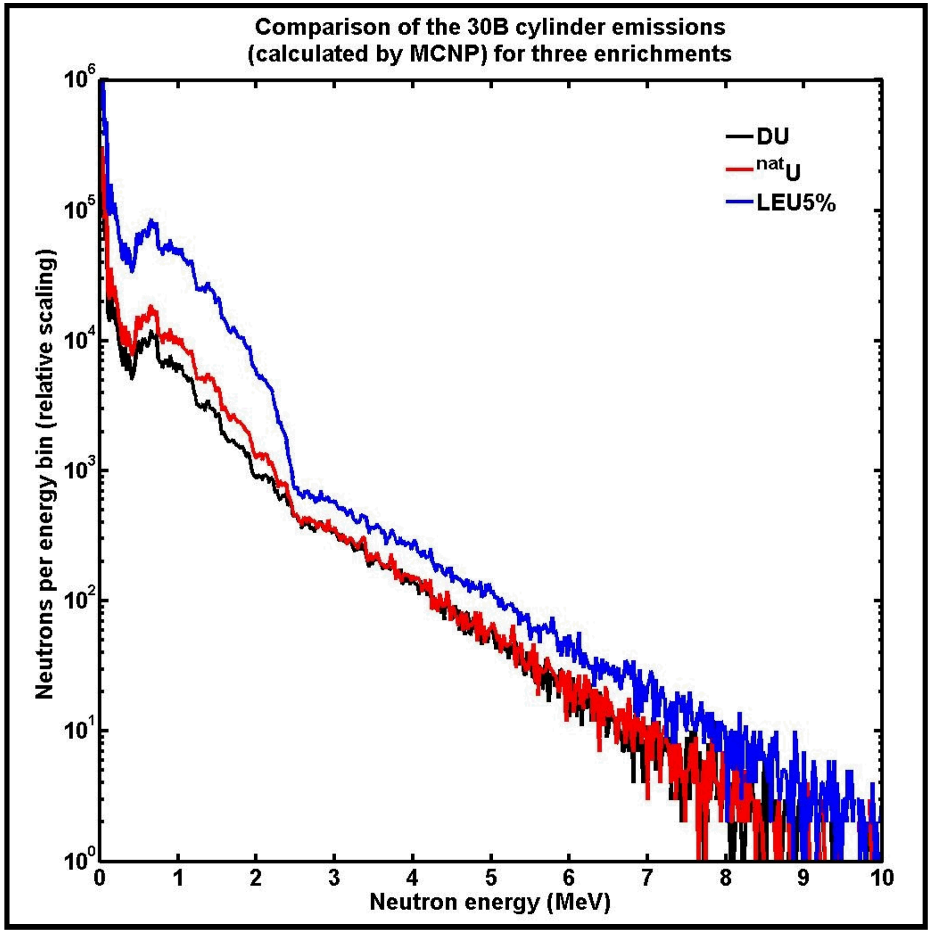


Full transport calculations imply manageable spectral perturbations

- 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.

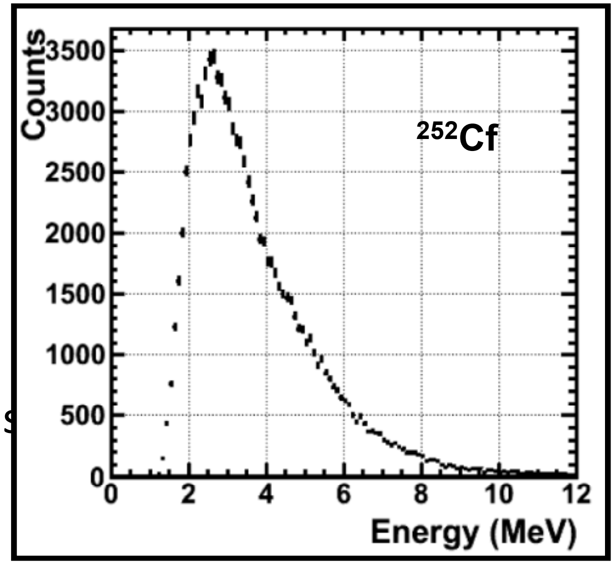
Results of the MCNP5 calculations

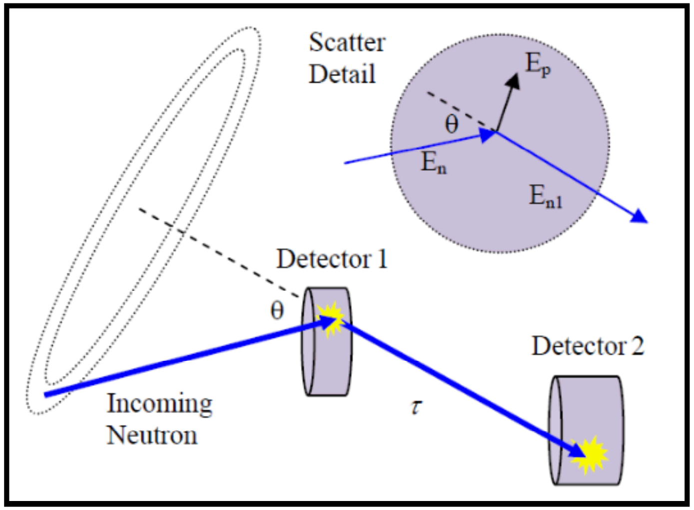


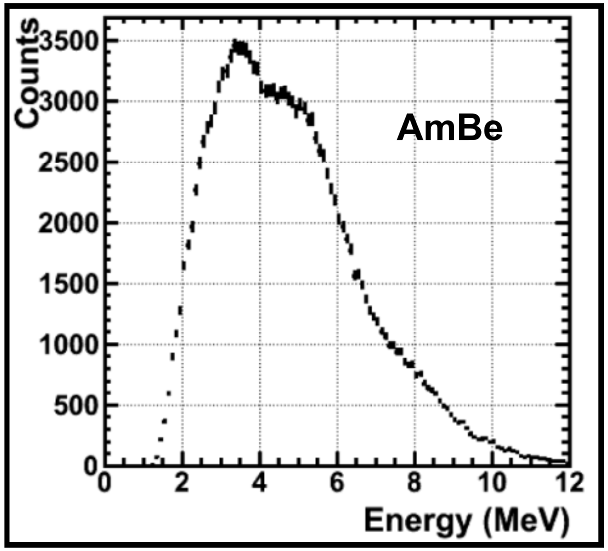


Neutron spectrometry can be performed with the Neutron Scatter Camera

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







The (simulated) 30B emissions with full detector response

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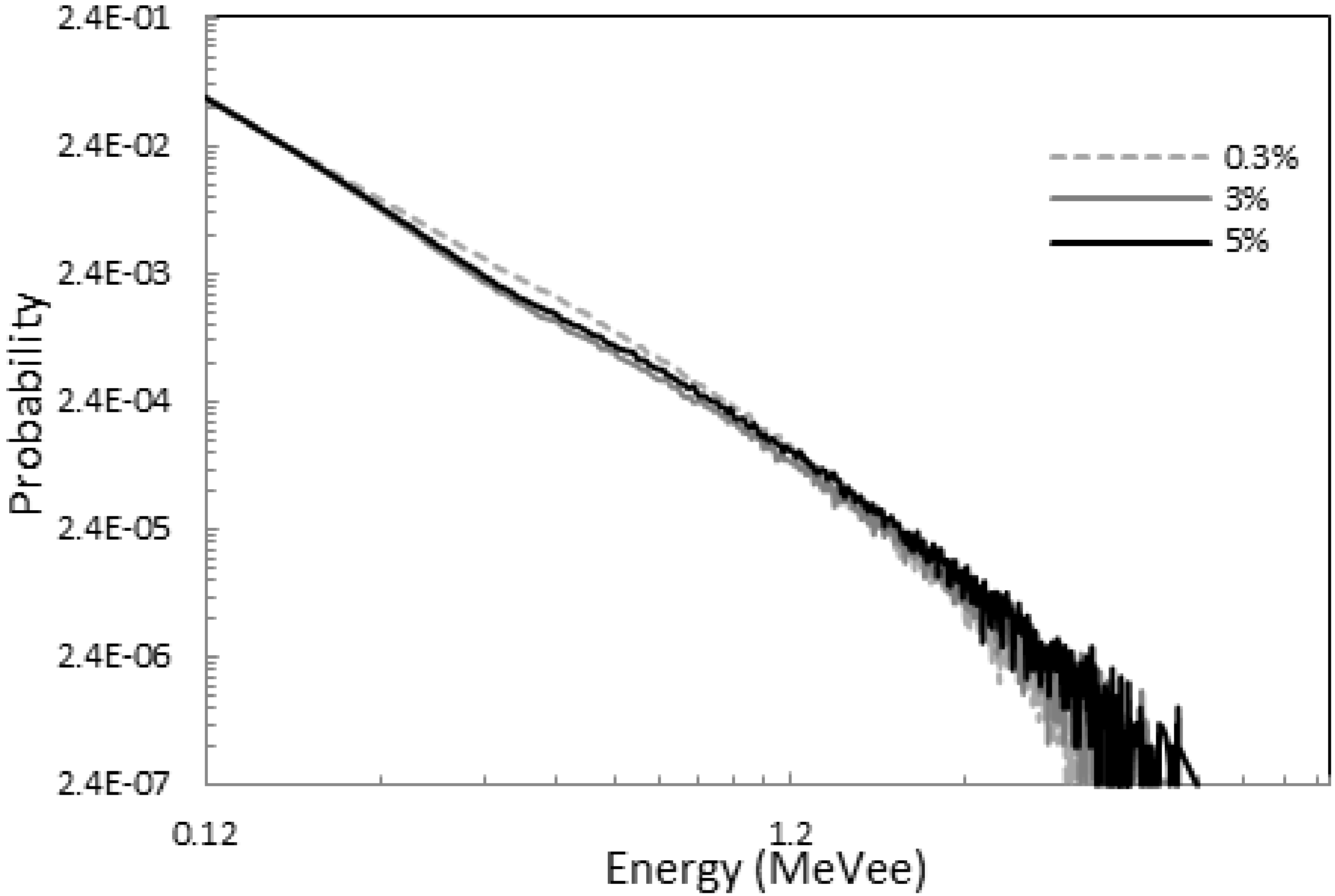


These spectra are normalized to more easily compare the distributions

The simulated spectra were analyzed using Principal Component Analysis (PCA)

- PCA is a technique that allows representation of data having many variables with a small set of significant variables
- Data dimensionality reduction is key aspect without losing characteristic data features
- The UF_6 fast neutron spectrum is a function of filling profile, ^{235}U enrichment and mass, and scattering environment
- Filling profile may vary from cylinder to cylinder
- PCA will characterize parameters including: declared ^{235}U enrichment, mass, and filling profile
- Cylinders with unexpected ^{235}U enrichment or very unusual material distribution may be detected as an anomaly using PCA

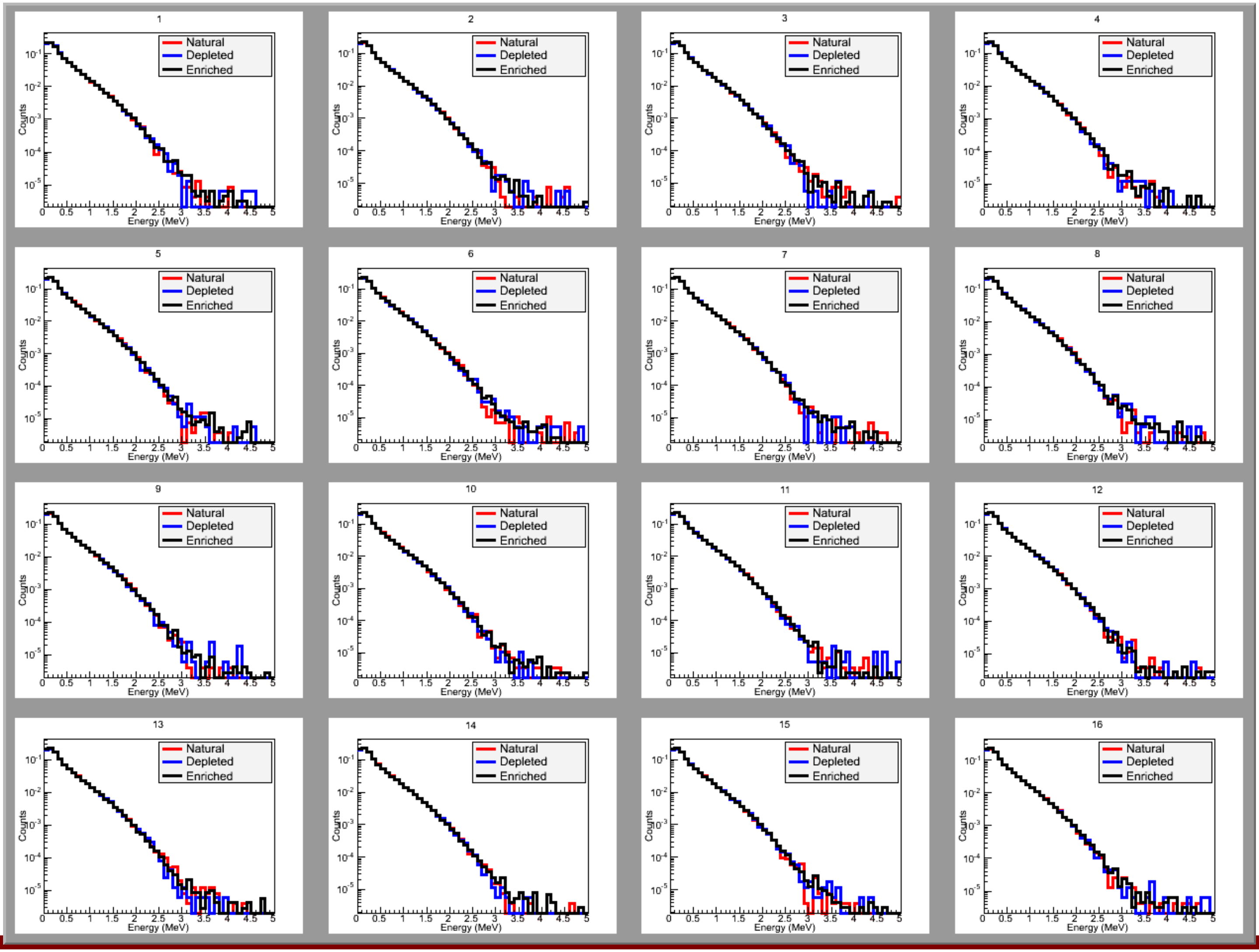
PCA Technique

- PCA requires the transformation of data into orthogonal feature space
- Transformed data is a linear combination of original data
- Transformed data is uncorrelated
- Reveal as much of the original variance in the transformed data space
- The new data are ordered according to the degree of variance and are called principal components (PC)
- The first few PC's are carry the most variance in the characterized data

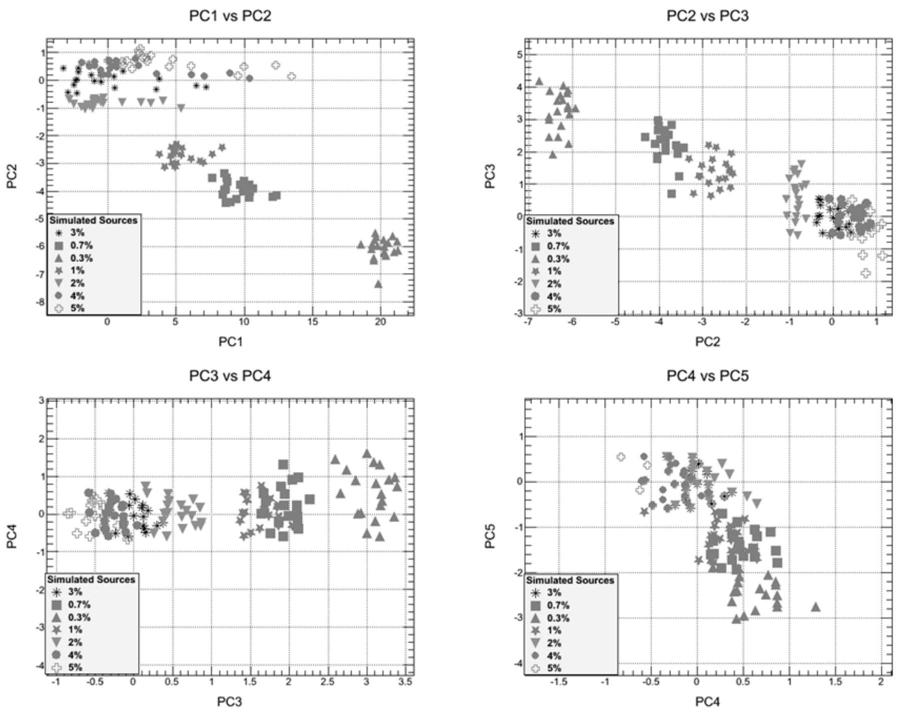
Preliminary data

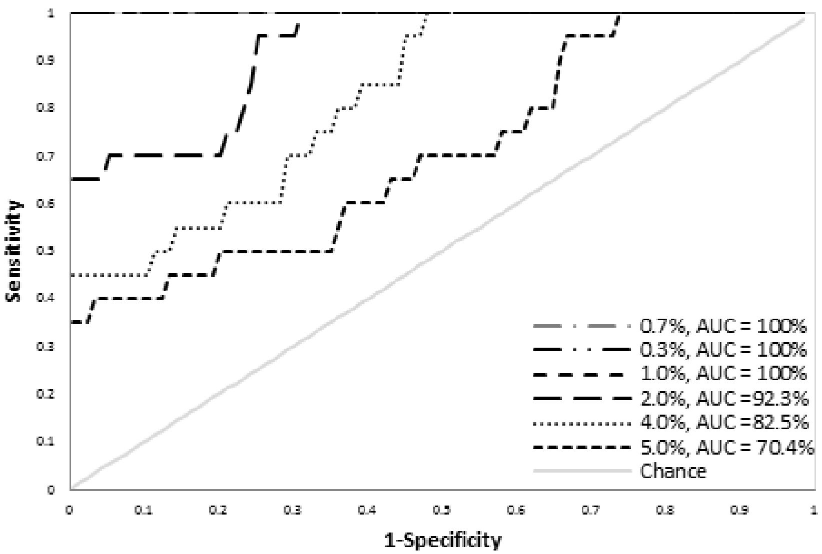
- Full transport simulations of neutrons within 30B cylinders and out to the Neutron Scatter Camera
- The detector response function was folded on to the detected neutron energy spectrum
- The PCA algorithm was trained using 3% ^{235}U
- Other enrichments ranging from depleted to 5% were clearly discriminated using the PCA approach

The simulated detector response spectra for 16 NSC cells



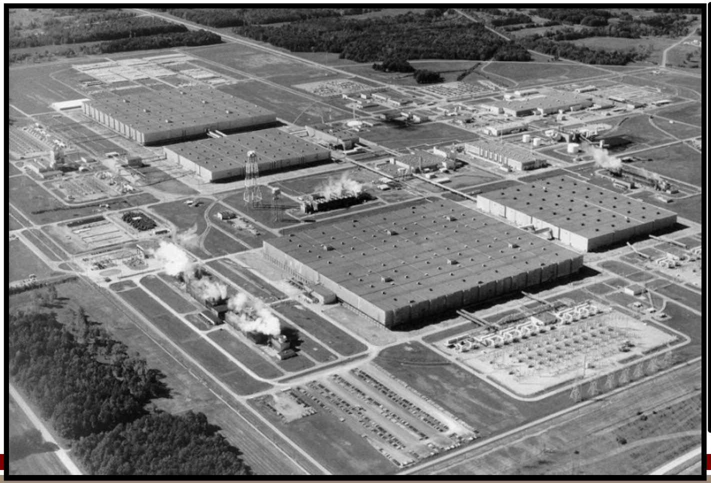
PCA Technique





Field measurements were performed to validate our results

- It is important to measure filled 30B cylinders with to collect data with
  - The appropriate (α,n) source term
  - A complex source with multiplication, scattering
  - Appropriate rates (neutrons and gammas)
  - Realistic backgrounds
- The Paducah Gaseous Diffusion Plant hosted our measurements



Paducah Deployment

Paducah Deployment

The Paducah DAQ was chosen to be representative of the NSC

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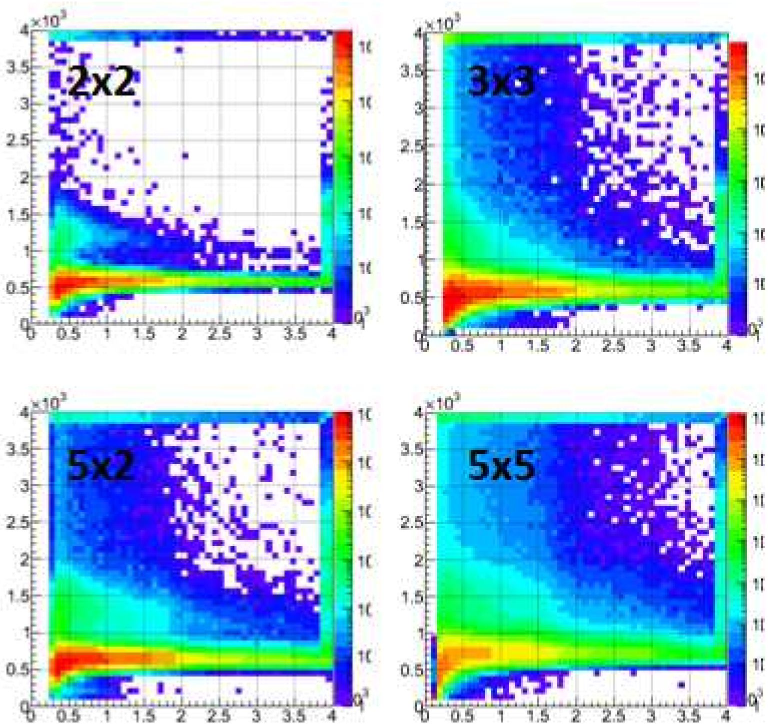
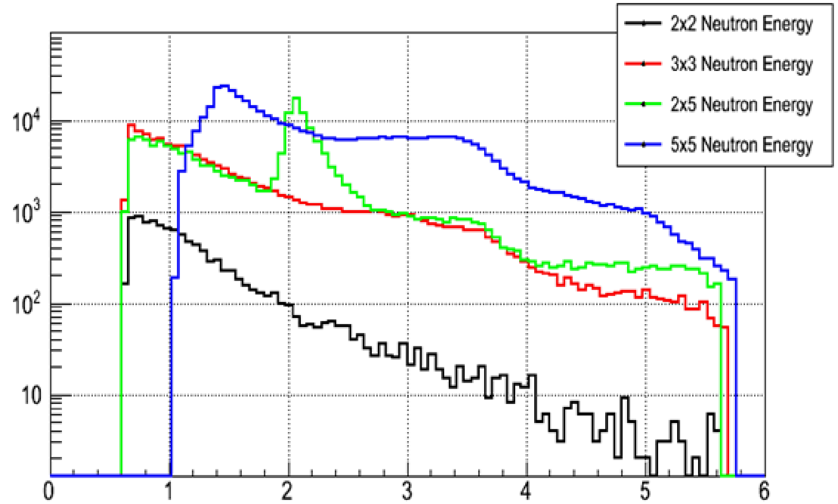
- Wanted a "NSC-like" data acquisition system that afforded more flexibility and had a larger assortment of detector sizes
  - 4 EJ-309 liquid scintillator cells: 2"x2", 3"x3", 5"x2", 5"x5"
  - 5"x5" NaI(Tl) crystal
  - ^{3}He tube
  - Analog electronics were similar to the NSC electronics at that time
    - PSD, ADC, scaler rates, trigger gate for coincident measurements
- Measurements of several enrichments
  - 0.711%, 2.00%, 3.60%, 4.00%, 4.95%
  - 4.00%, 4.95% (downblended Russian material)
- Goals
  - Measurement of neutron spectra
  - Discover and work through difficulties associated with field measurements

Pulse pileup from high gamma rates created PSD challenges

PSD plots (log intensity scale)

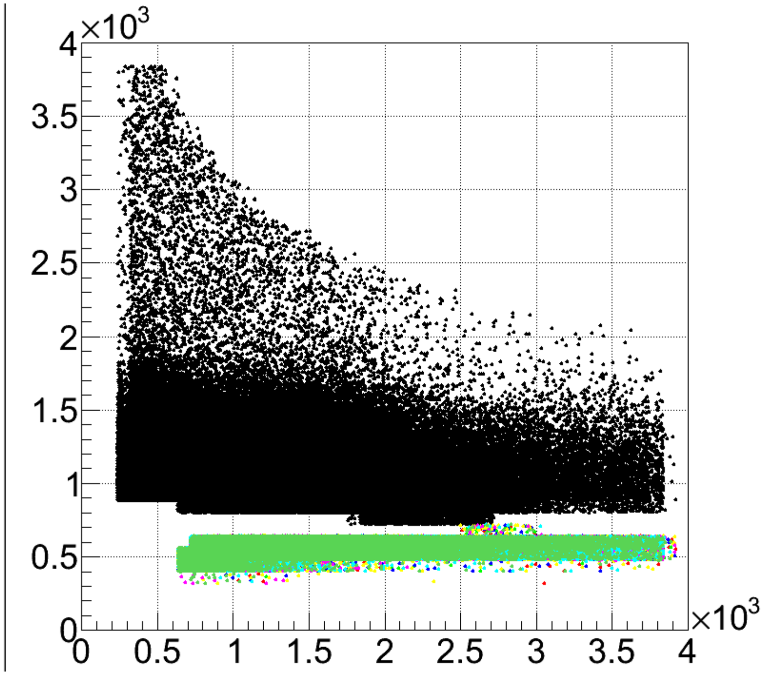


Neutron pulse height spectrum

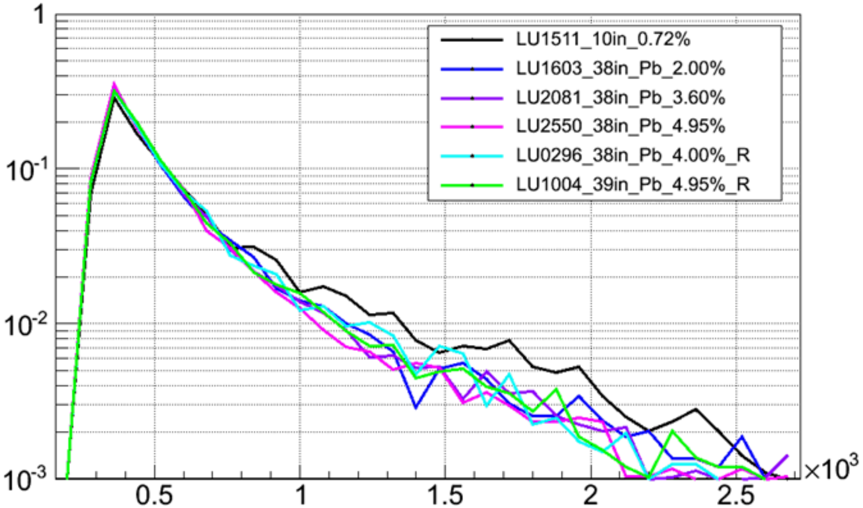


Focus on the smallest EJ-309 cell (manageable pileup)

PSD plot (black=neutrons, green=gammas)

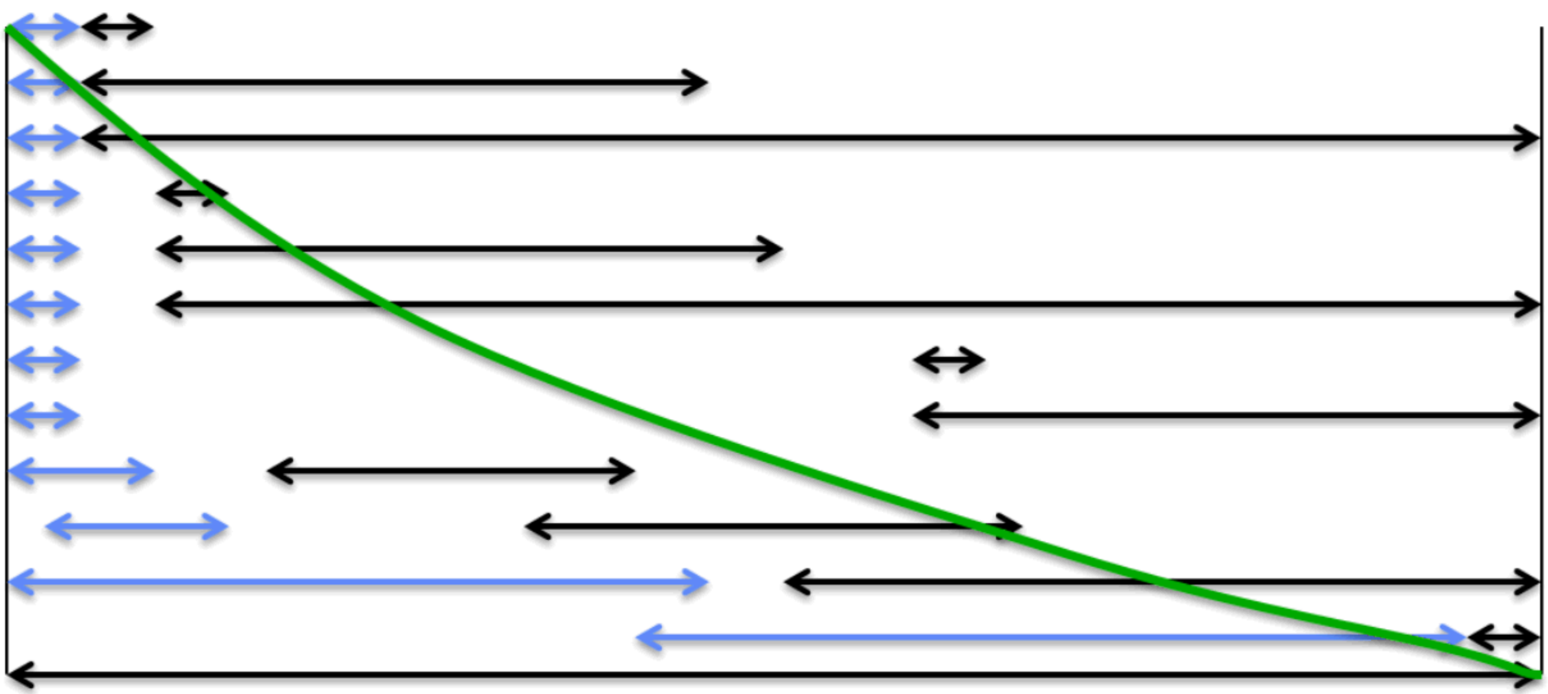


Neutron energy spectra for each 30B cylinder

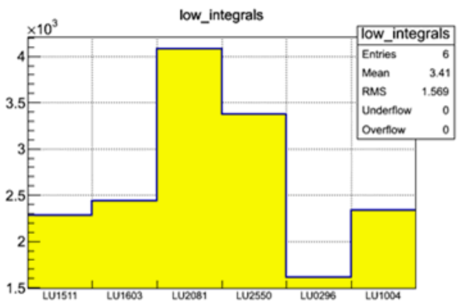


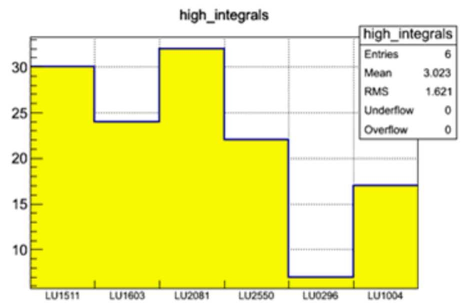
A simple two-bin analysis

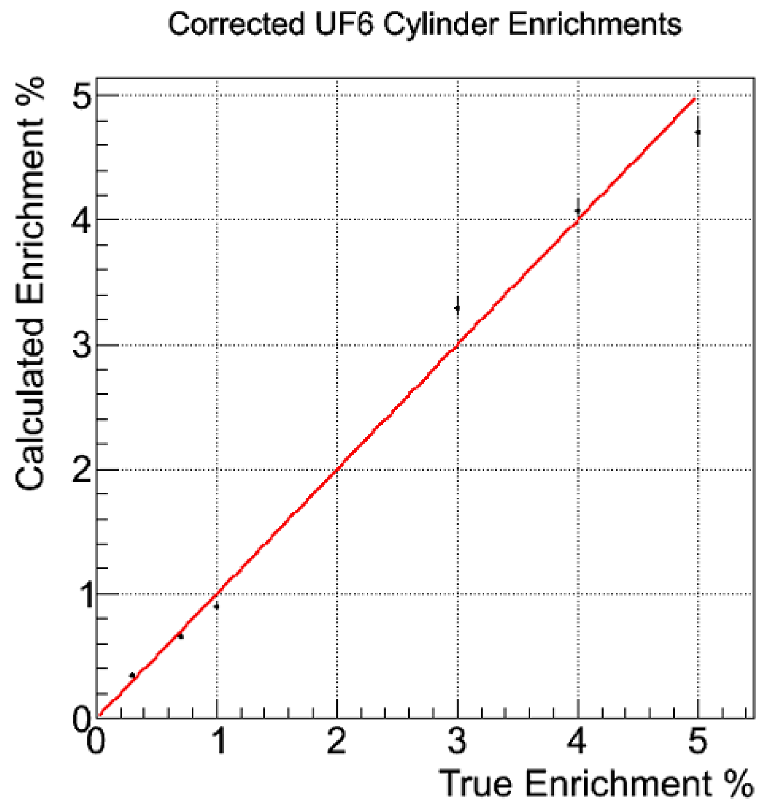
- Initialize 2 windows, low and high
- Find the total number of bins in the spectrum
- Loop over all non-overlapping window ranges.



Analysis results indicate spectroscopy works!







Roughly 0.25 MeV to 1 MeV low window, and 1 MeV to 6 MeV high window

Extrapolating detector rates to count time/counting uncertainty

Counting uncertainty vs. time for a 2” EJ-309 cells

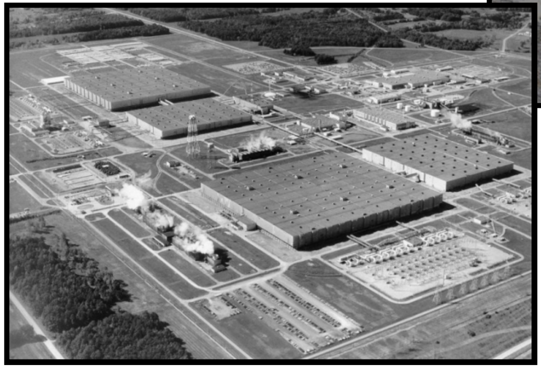
Time (hrs)	High-energy counts	Uncertainty
1	6.5	0.39
2	13	0.28
5	32.5	0.18
10	65	0.12
15	97.5	0.10
20	130	0.09
30	195	0.07
40	260	0.06
50	325	0.06
60	390	0.05

Time to 5% counting uncertainty vs. cell size,

2" Cells	Hours	3" Cells	Hours	5“ Cells	Hours
1	50	1	14.8	1	3.2
2	25	2	7.4	2	1.6
5	10	5	3.0	5	0.64
10	5	10	1.5	10	0.32
15	3.3	15	1.0	15	0.21
20	2.5	20	0.74	20	0.16

Some challenges were revealed during this investigation

- Assaying cylinders using reasonable dwell times
- High gamma emissions of cylinders (technology choices help)
- High backgrounds from neighboring cylinders (separation is important—imaging could help, if practical)
- Isotopic challenges
  - $^{234}U/^{235}U$ ratio assumption
  - Downblended Russian material



Summary

- Direct measurement of neutron signatures for UF6 material accountancy appears to be a possible safeguards technique
  - Two physical processes create neutrons with different energy spectra
  - Simulations and experiments indicate enrichment can be extracted from emitted neutrons, even after full transport
- Advantages:
  - Sensitive to entire cylinder volume
  - Imaging (if possible) suppresses backgrounds from nearby cylinders, allows one to map material distribution within a cylinder
- Possible future work:
  - Detector optimization (detection medium, segmentation)
  - Verify insensitivity to internal geometry
  - Active interrogation?

Special thanks to…

- Financial support
  - Sandia LDRD office
  - NA-24 Human Capital Development
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  - Erik Brubaker, Laura Kogler, Pete Marleau, Stan Mrowka (Sandia staff)
  - Michael Streicher and Edward Cazalas (summer interns)
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