SAND2011-6525C

Fast Neutron Spectrometry for Nuclear Safeguards Measurements

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Presentation Outline

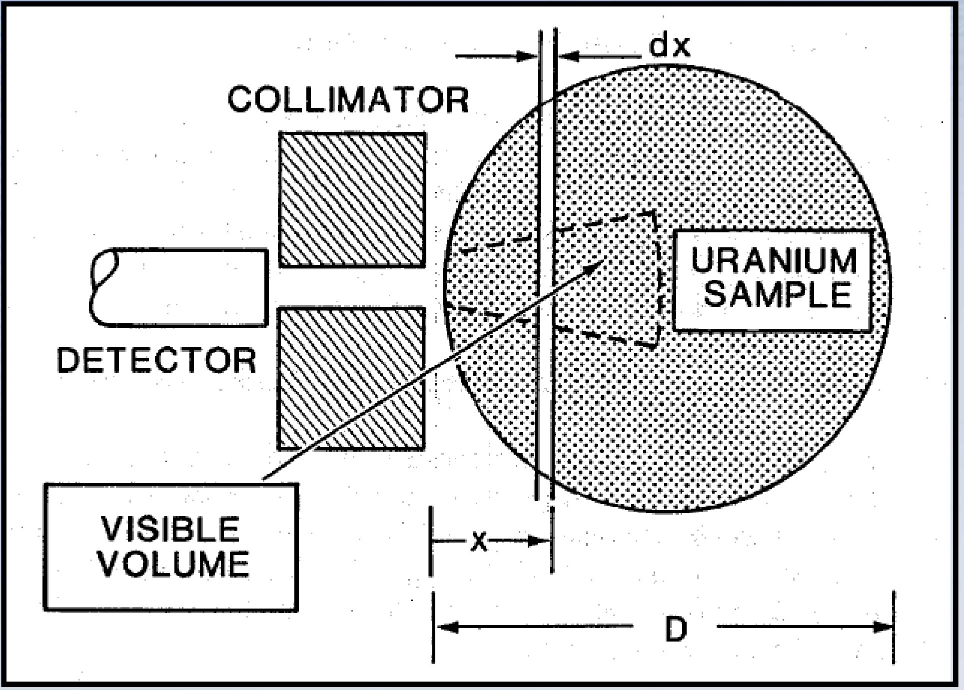
- Introduction to the problem
- Sandia's concept: imaging neutron spectrometers
- Pu vs. uranium
- Initial Pu calculations
- Discussion

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**

Current technology is good, but…

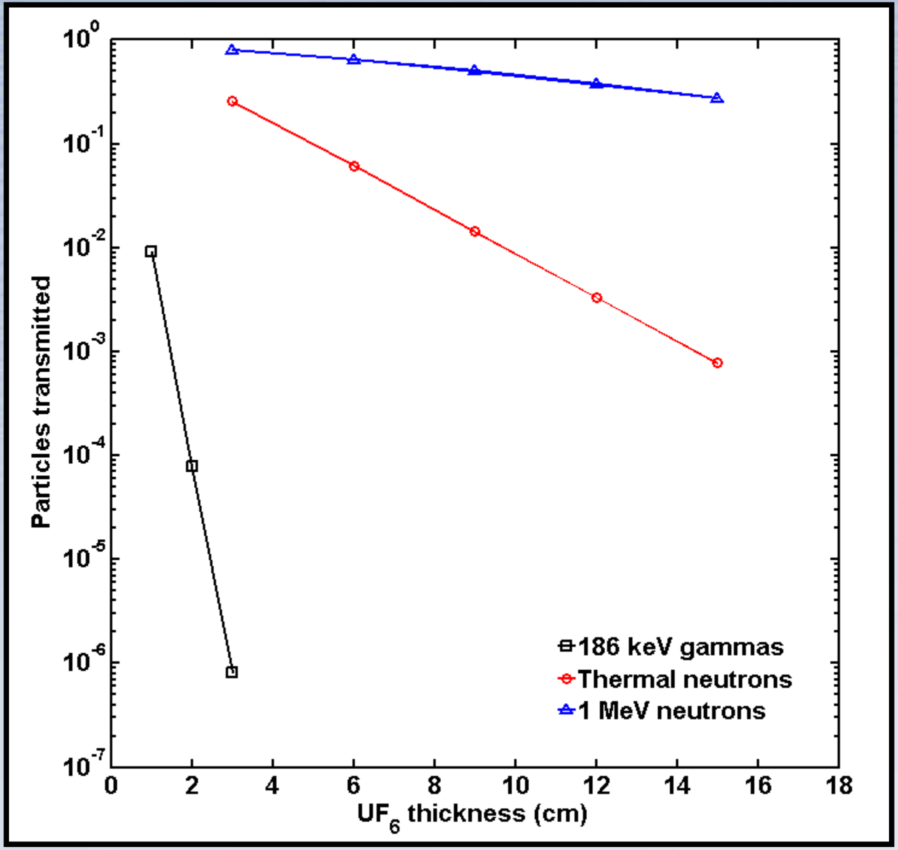
- **"Enrichment meter" measures gamma emissions from the uranium hexafluoride (UF_6)**
  - Gives local enrichment, not total mass
  - Sensitive to variations in container wall thickness
  - Not sensitive to material beyond outer skin of UF_6



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

Sandia's concept: directly measure fast neutron emissions

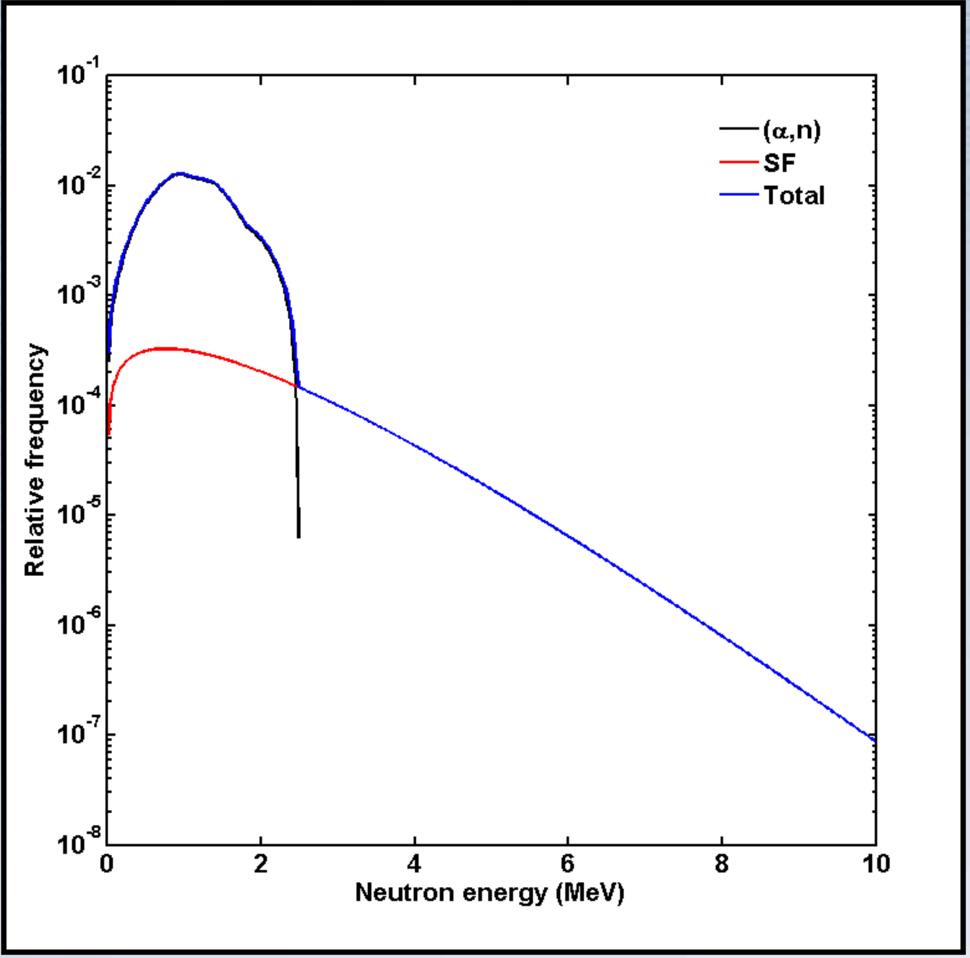
- Fast neutrons generated by independent processes within the UF_6 can provide an independent enrichment measurement that samples the entire UF_6 volume
- Neutron imaging of the UF_6 distribution detects unexpected UF_6 geometries and applies necessary corrections
- Sandia has developed expertise in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

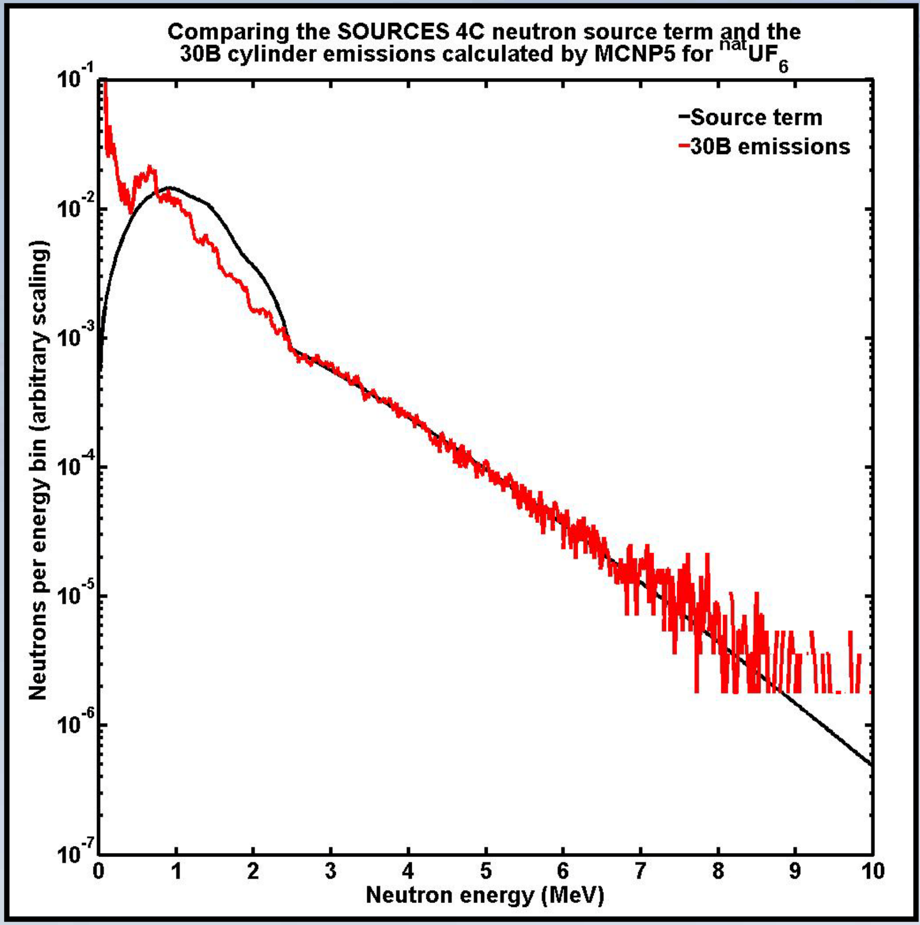


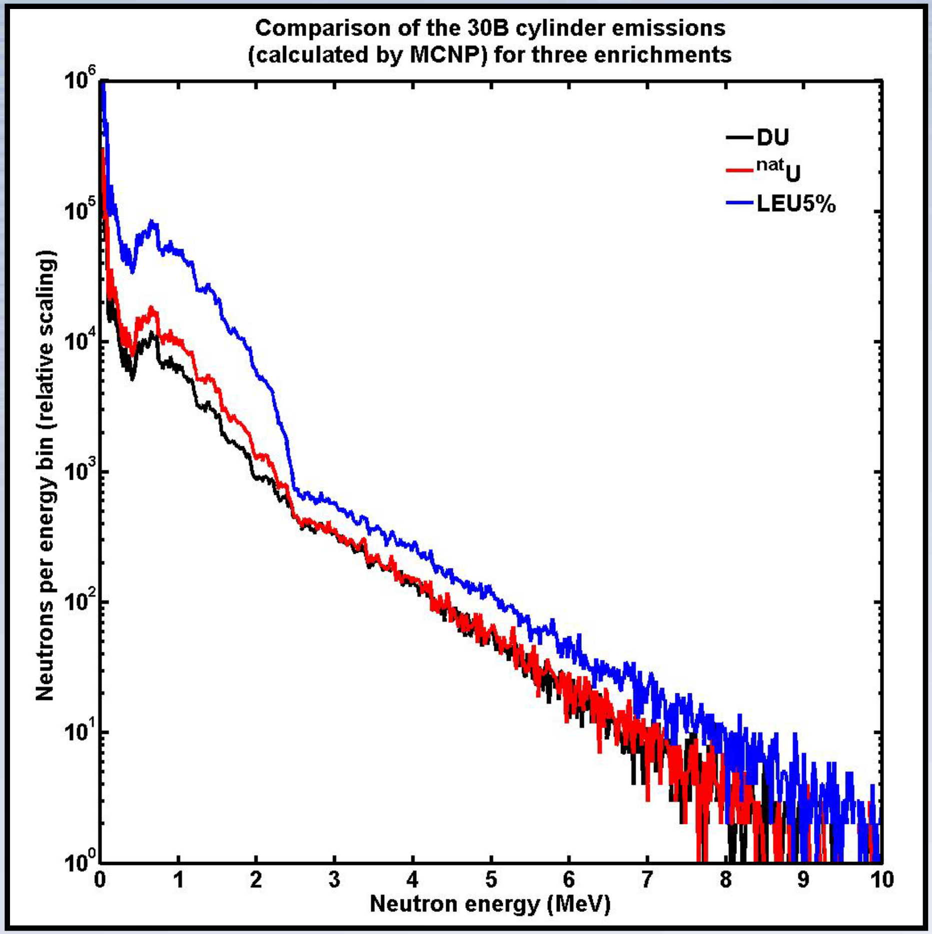
SOURCES4C calculation of neutron spectrum for 5% enriched UF_6

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





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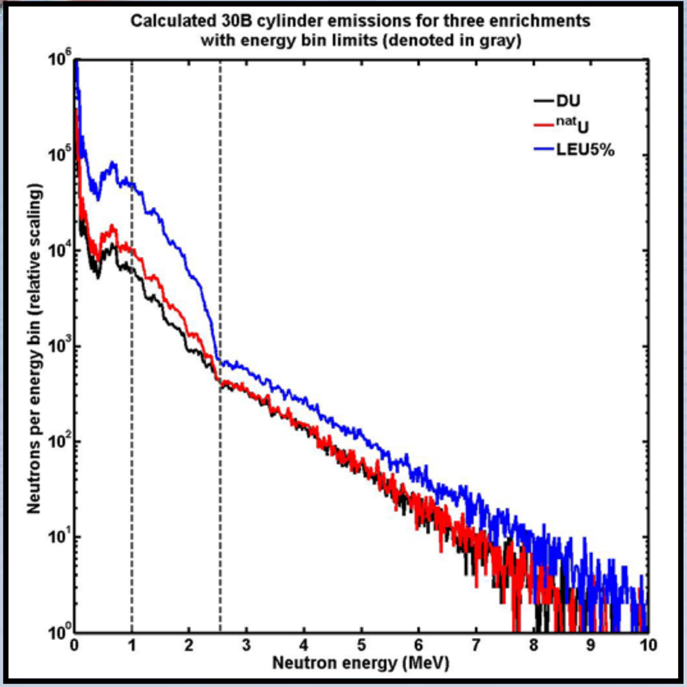
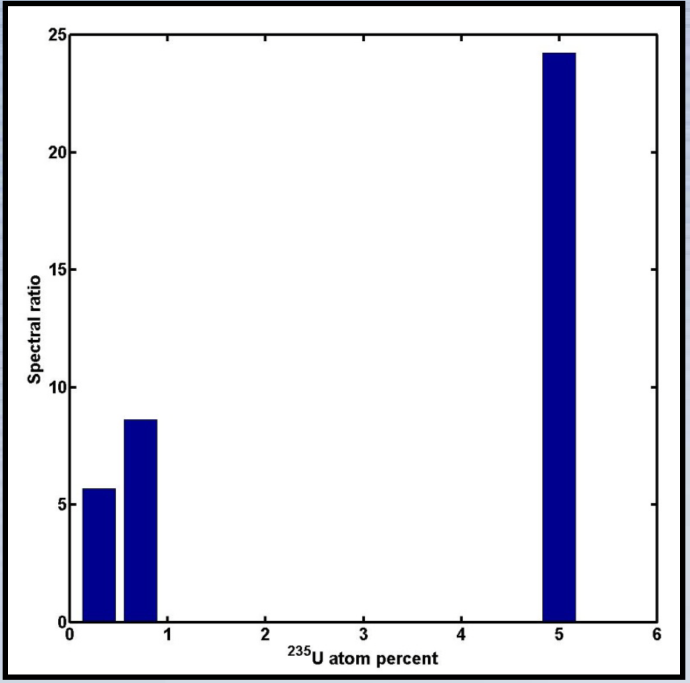
The enrichment can be inferred from the neutron energy spectrum

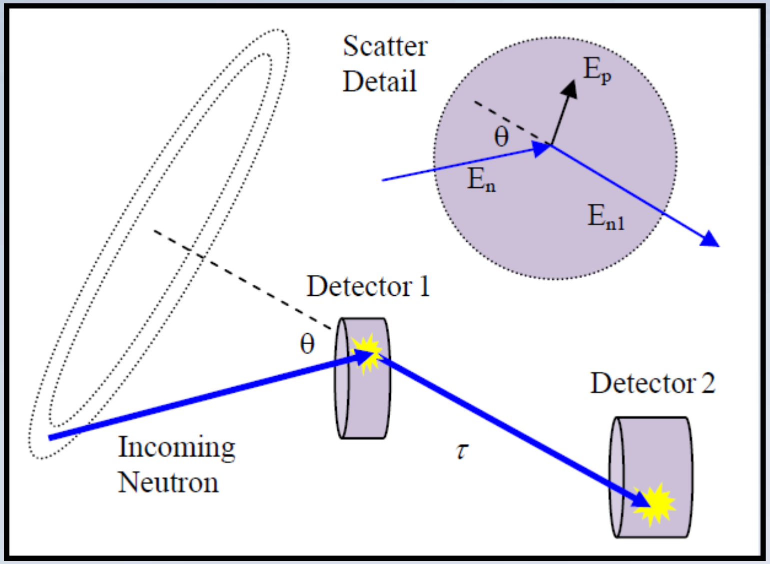


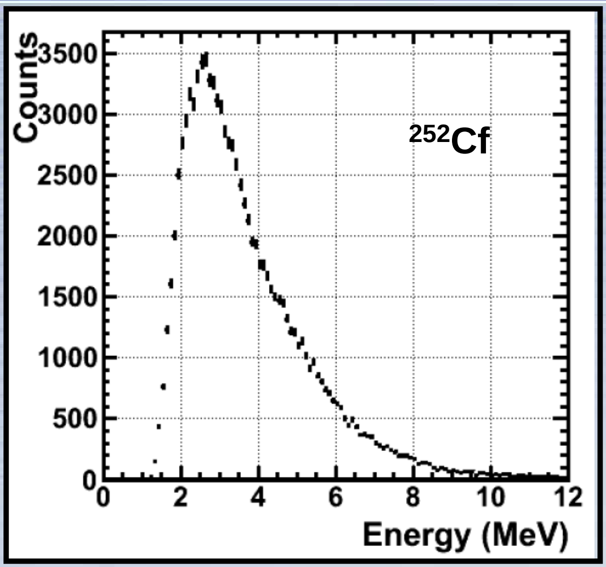


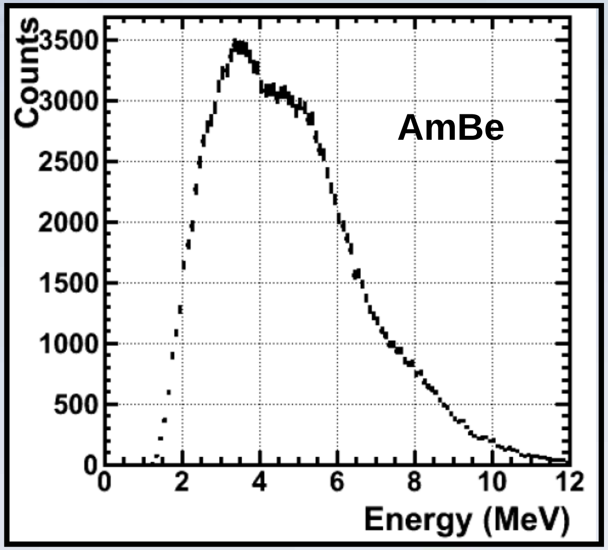
- **The ratio of neutrons in the (α,n) and S.F. regions is a function of enrichment**
  - Cut data at the end of the (α,n) spectrum (~2.54 MeV)
  - A realistic detector will have a detection threshold (choose 1 MeV)
- **For the simulated data, the ratio is a monotonic function of enrichment**

Neutron spectrometry measurements will 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)







Considering PuO_2 holdup accountancy

- **Consider PuO_2 holdup in a reprocessing facility**
  - Gloveboxes
  - Hulls
- **A combination of imaging (to locate hotspots) and spectrometry (to measure total activity) may be useful**
  - Spectrometry technique: follows directly from UF_6 enrichment measurement concept
  - Imaging: could potentially couple with Hausladen's imager, or may find another imaging technique is better

La Hague reprocessing facility

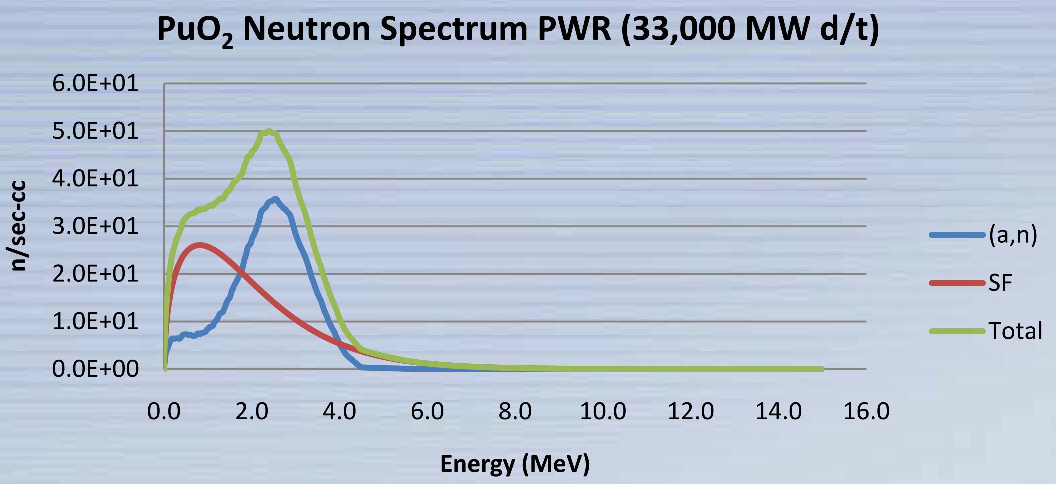
Plutonium vs. uranium measurements

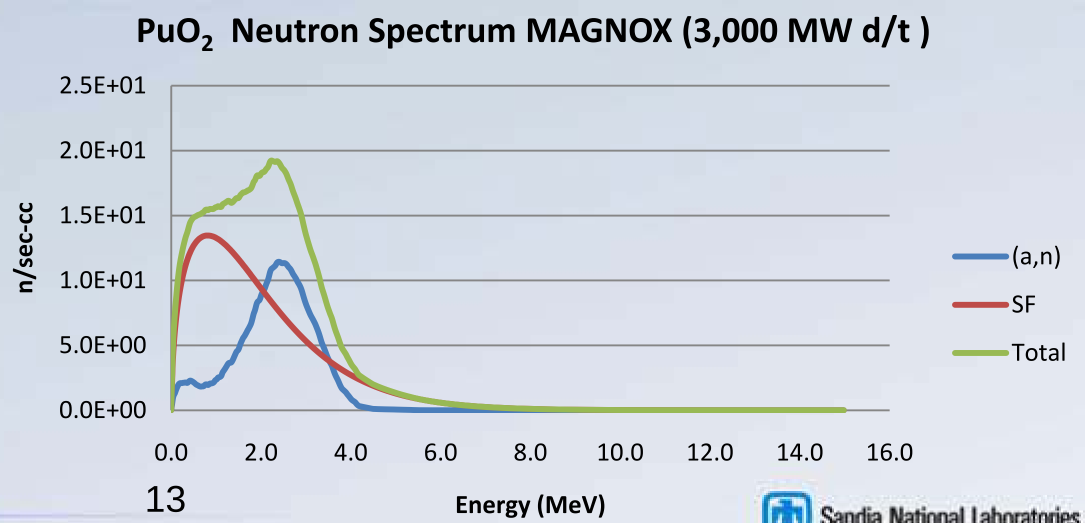
- **Consider PuO_2. How do the proposed measurements compare to UF_6?**

- **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





Working an example

- **Data from various online sources accumulated**
  - Varying isotopic content for PuO_2.

		Atom Fractions					
		Magnox	SO1	SO2	PWR	PWR	PWR
BURNUP		3000	8167	6808	33000	43000	53000
Pu	238	0.001	0.00557	0.00462	0.013	0.02	0.027
Pu	239	0.8	0.71886	0.73218	0.566	0.525	0.504
Pu	240	0.169	0.1905	0.18812	0.232	0.241	0.241
Pu	241	0.027	0.0721	0.06384	0.139	0.147	0.152
Pu	242	0.003	0.01295	0.01124	0.047	0.062	0.071

Use "effective" isotope masses

- **PWR reactor with 43,000 MW d/t burn up**

Element	Isotope	Rho [g/cc]	Atom Fraction	Stoichiometry	Isotope Mass/ Molecule	Atom Dens. [#/cc]	For 1 cm^3 true Pu mass (g)	Eff 240 mass (g)	Eff 239 mass (g)
Pu	238	11.5	0.02	1	4.76	5.12E+20	**10.1255**	**95.04192**	**2015.646**
Pu	239	11.5	0.525	1	125.475	1.34E+22			
Pu	240	11.5	0.241	1	57.84	6.17E+21			
Pu	241	11.5	0.147	1	35.427	3.76E+21			
Pu	242	11.5	0.062	1	15.004	1.59E+21			
O	16	11.5	0.99757	2	31.92224	2.55E+22			
O	17	11.5	0.00038	2	0.01292	9.73E+18			
O	18	11.5	0.00205	2	0.0738	5.25E+19			
				SUMS:	**238.506**	**2.55E+22**			

$$m_{240}^{eff} = 2.52 * m_{238} + m_{240} + 1.68 * m_{242}$$

(Pu-240 eff mass proportional to SF contribution)
(Pu-239 eff mass proportional to (α, n) contribution)

$$m_{239}^{eff} = \frac{13400}{38.1} * m_{238} + m_{239} + \frac{14.1}{38.1} * m_{240} + \frac{1.3}{38.1} * m_{241} + \frac{2}{38.1} * m_{242}$$

Pu-239 mass coefficients calculated using data from: http://www.fas.org/sgp/othergov/doe/lanl/lib-www/la-pubs/00326406.pdf
Pu-240 effective mass formula : "Nuclear Safeguards, Security, and Nonproliferation" Doyle, James E.

SOURCES4C Outputs

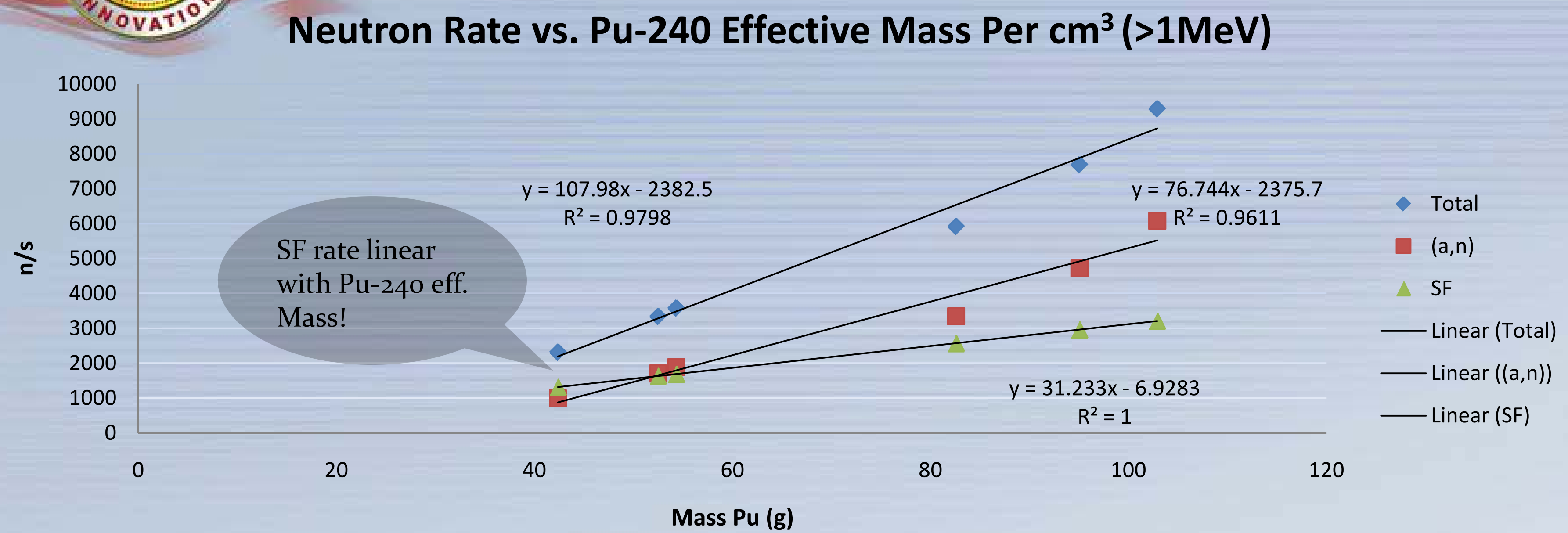
- **Sources outputs data for:**
  - (alpha, n)
  - Spontaneous Fission
  - Delayed neutrons (ignored in this case ~ 0)
  - Total neutron contribution
- **Want to compare "Total" spectra to (a, n) and SF individually.**

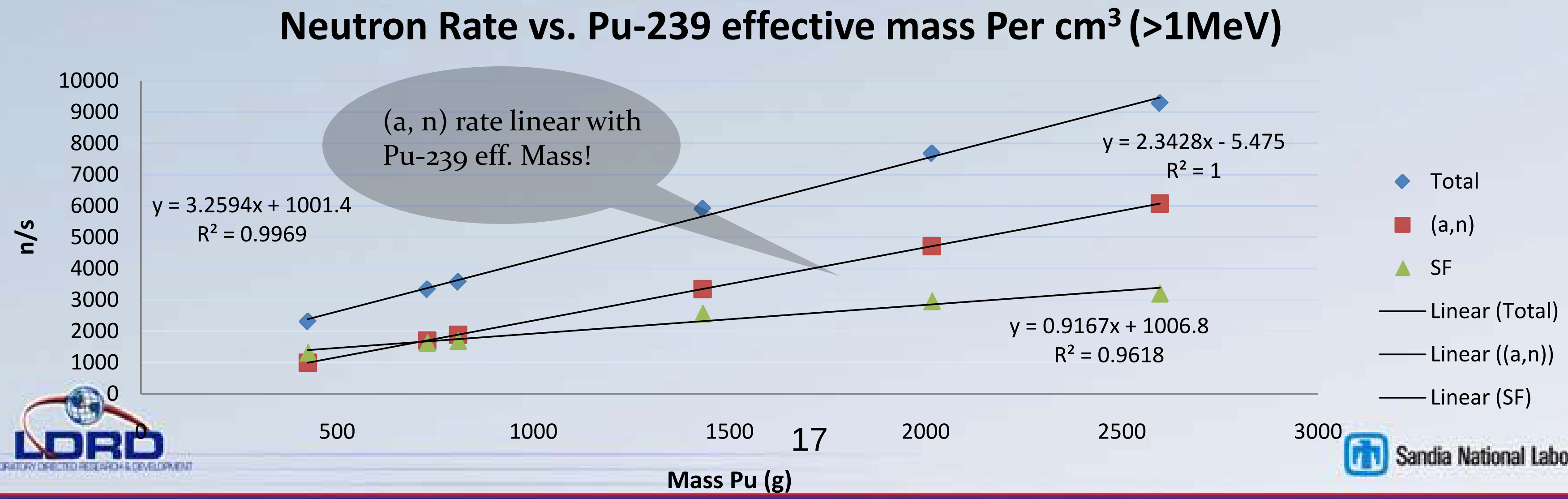
Absolute Totals (neutrons/sec-basis)

E (MeV)	(a,n)	(sf)	(dn)	Total
0.000E+00	0.000E+00	0.000E+00	0.000E+00	0.000E+00
2.000E-02	4.571E-01	2.202E+00	0.000E+00	2.659E+00
4.000E-02	9.447E-01	3.990E+00	0.000E+00	4.934E+00
6.000E-02	1.171E+00	5.117E+00	0.000E+00	6.288E+00
8.000E-02	1.293E+00	6.000E+00	0.000E+00	7.293E+00
1.000E-01	1.485E+00	6.739E+00	0.000E+00	8.224E+00
1.200E-01	1.664E+00	7.378E+00	0.000E+00	9.042E+00
1.400E-01	1.825E+00	7.941E+00	0.000E+00	9.766E+00
1.600E-01	1.965E+00	8.444E+00	0.000E+00	1.041E+01
1.800E-01	2.048E+00	8.898E+00	0.000E+00	1.095E+01
2.000E-01	2.086E+00	9.310E+00	0.000E+00	1.140E+01
2.200E-01	2.093E+00	9.687E+00	0.000E+00	1.178E+01
2.400E-01	2.092E+00	1.003E+01	0.000E+00	1.212E+01
2.600E-01	2.113E+00	1.035E+01	0.000E+00	1.246E+01
2.800E-01	2.118E+00	1.064E+01	0.000E+00	1.276E+01
3.000E-01	2.132E+00	1.091E+01	0.000E+00	1.304E+01
3.200E-01	2.139E+00	1.116E+01	0.000E+00	1.330E+01
3.400E-01	2.117E+00	1.139E+01	0.000E+00	1.351E+01
3.600E-01	2.106E+00	1.160E+01	0.000E+00	1.371E+01

Energy bins (0 to 14.98 MeV)

Plotted Results of Ideal (alpha, n) and Spontaneous Fission neutron contributions calculated by SOURCES4C with 1MeV threshold.





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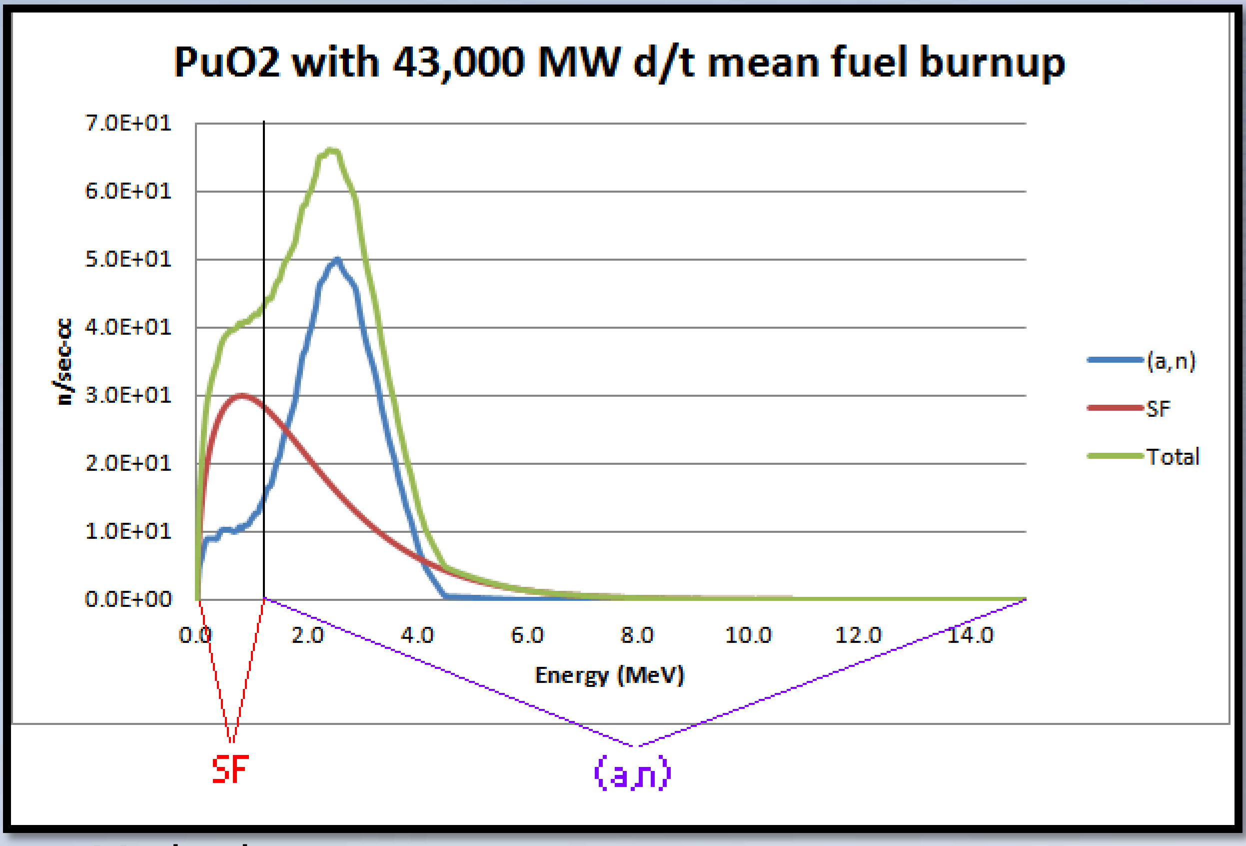
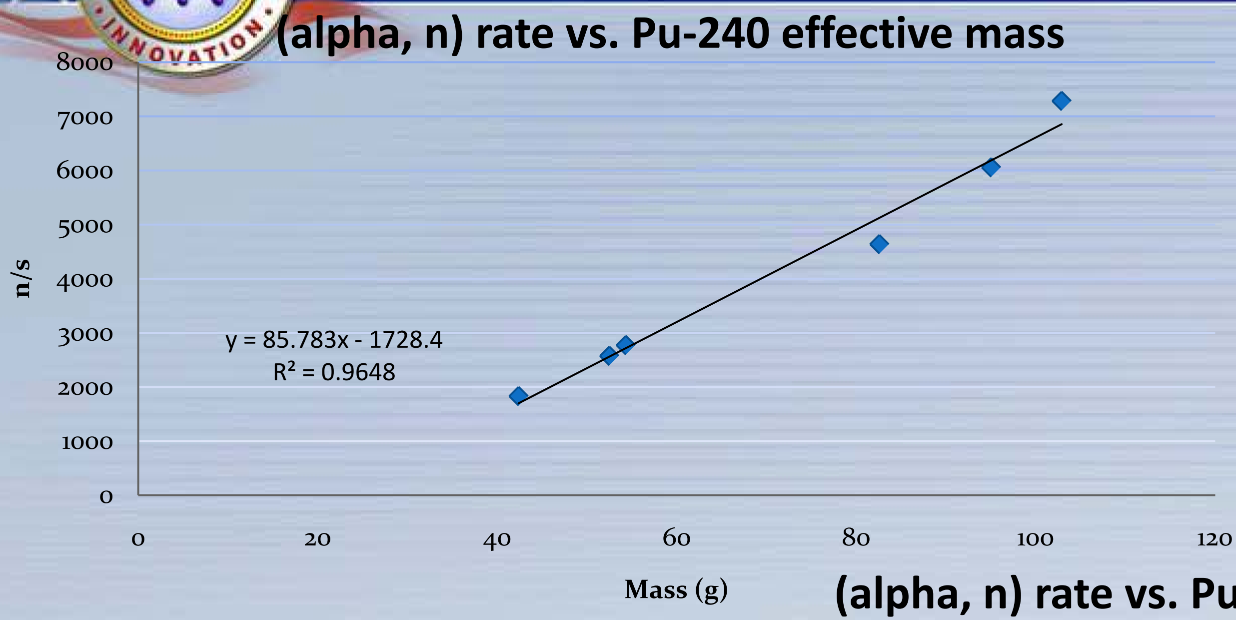
Assigning SF, (α,n) contributions



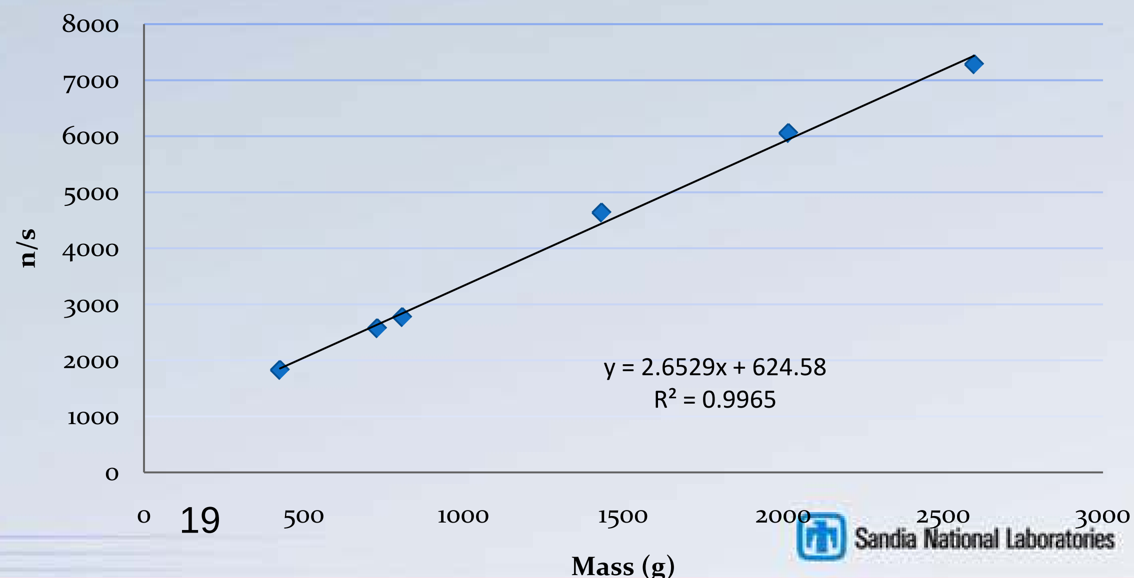
- Look for a point to split graph in two
  - SF and (α, n)
- Integrate using bin boundaries
- Why?
  - Detector sees total only (Green Curve)
  - Need to use simulations to tell SF from (α, n) contributions

- Method:
  - Calculated double derivatives to look for the 2 peaks
  - Averaged the integral between the two peaks to find the energy at which to split the graph

Results From Splitting the "Total" Spectra



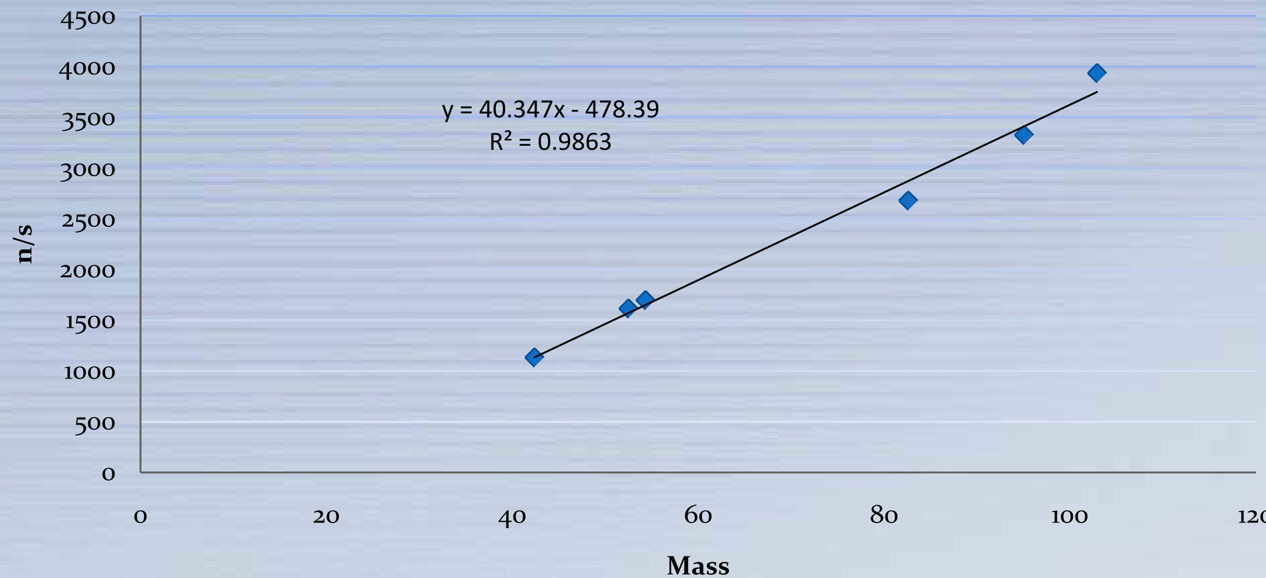
SOURCES4C "Total" output was "split" to derive these values.



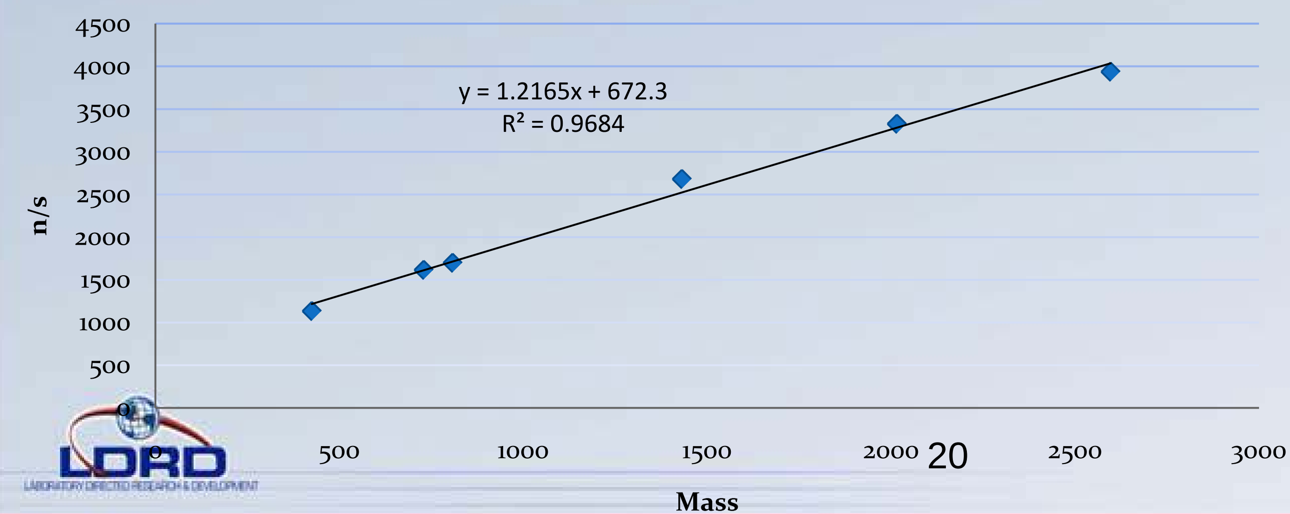
Data shown here confirms our expected result that there is a higher correlation between the (alpha, n) rate and Pu-239 effective mass compared to the Pu-240 effective mass.
(No threshold set)

Spontaneous Fission rates showed a higher correlation with the Pu-240 effective mass compared to the Pu-239 effective mass. This confirmed our expectations. (No threshold set)

Spontaneous Fission Rate vs. Pu-240 effective mass



Spontaneous Fission Rate vs. Pu-239 effective mass

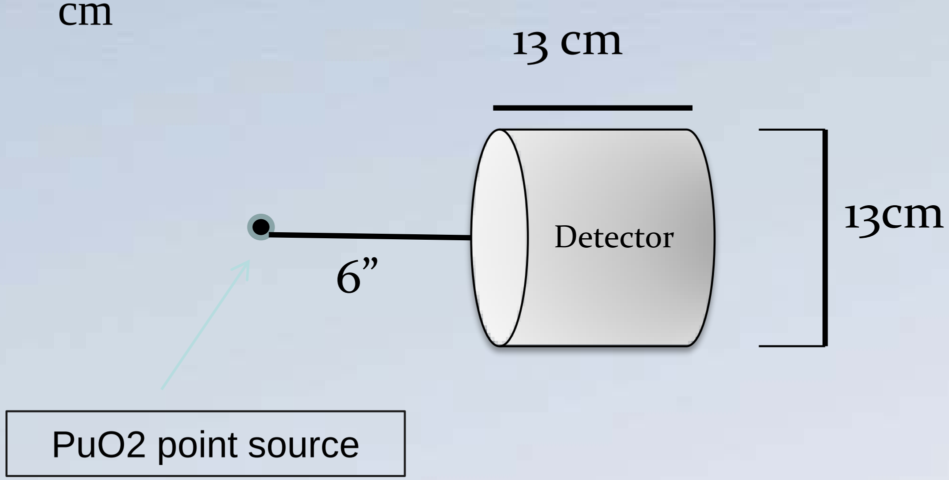


<u>Purpose of measuring rates as a function of mass:</u>

To find a relationship concerning all 5 isotopes where we can back-calculate amount of Pu-240 and Pu-239.

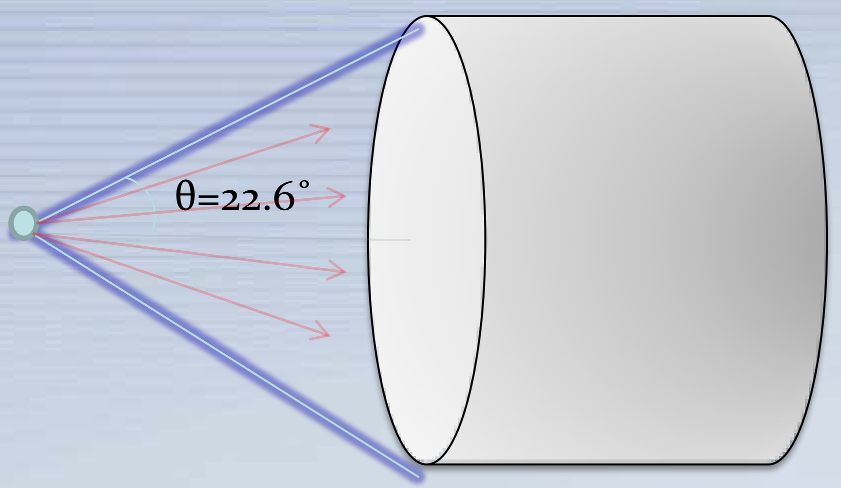
Next Step: Apply a detector response in order to confirm and verify values obtained from SOURCES4C and to simulate what we would see experimentally.

- **MCNP – Gaussian Energy Broadening function applied**
  - Geometry: point source of PuO_2 placed 6 inches away from midpoint of scintillation detector (EJ309).
    - Detector: 5 inch (12.7 cm) diameter, 5 inch depth.
      - Including aluminum covering of 0.15 thickness, overall dimensions = 13 cm

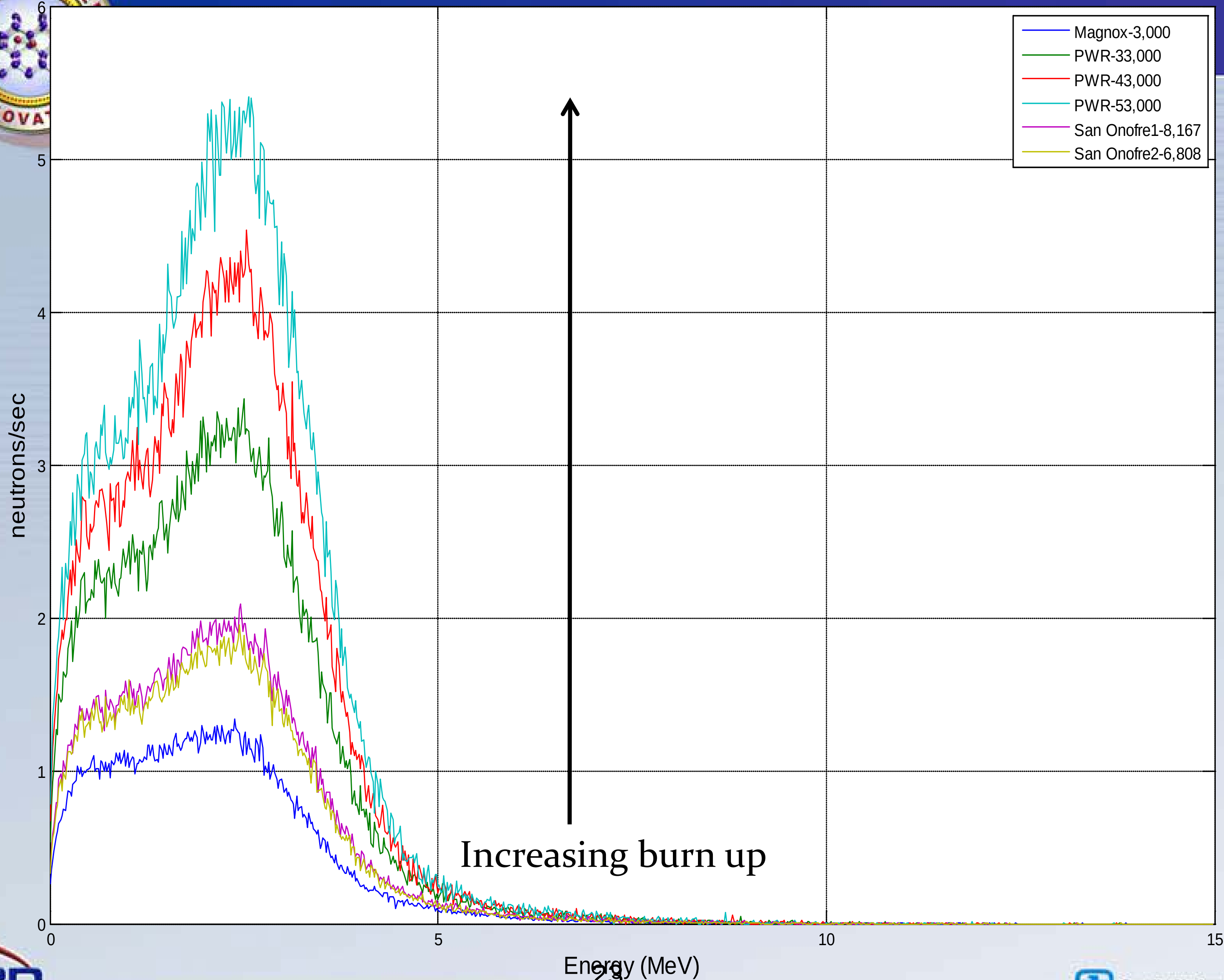


MCNP Simulations

- **Ran MCNP simulations for these spent fuel compositions:**
  - Magnox (Burn up=3,000)
  - PWR1 (Burn up=33,000)
  - PWR2 (Burn up=43,000)
  - PWR3 (Burn up=53,000)
  - San Onofre 1 (Burn up=8,167)
  - San Onofre 2 (Burn up=6,808)



Neutron Energy spectrum with Gauss Energy Broadening



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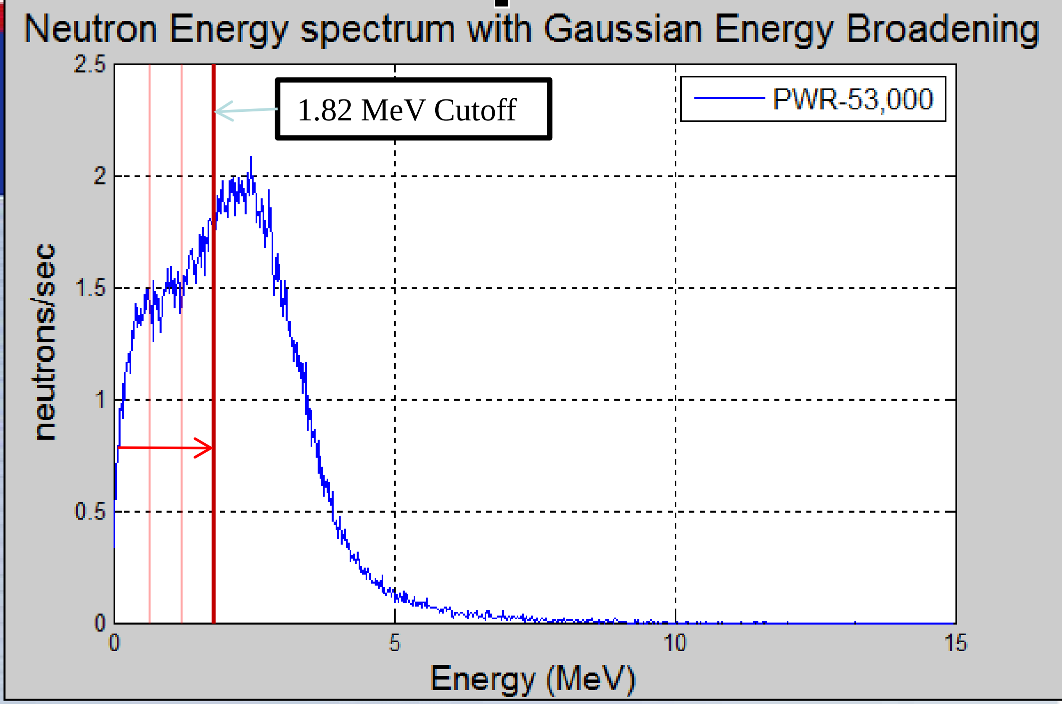
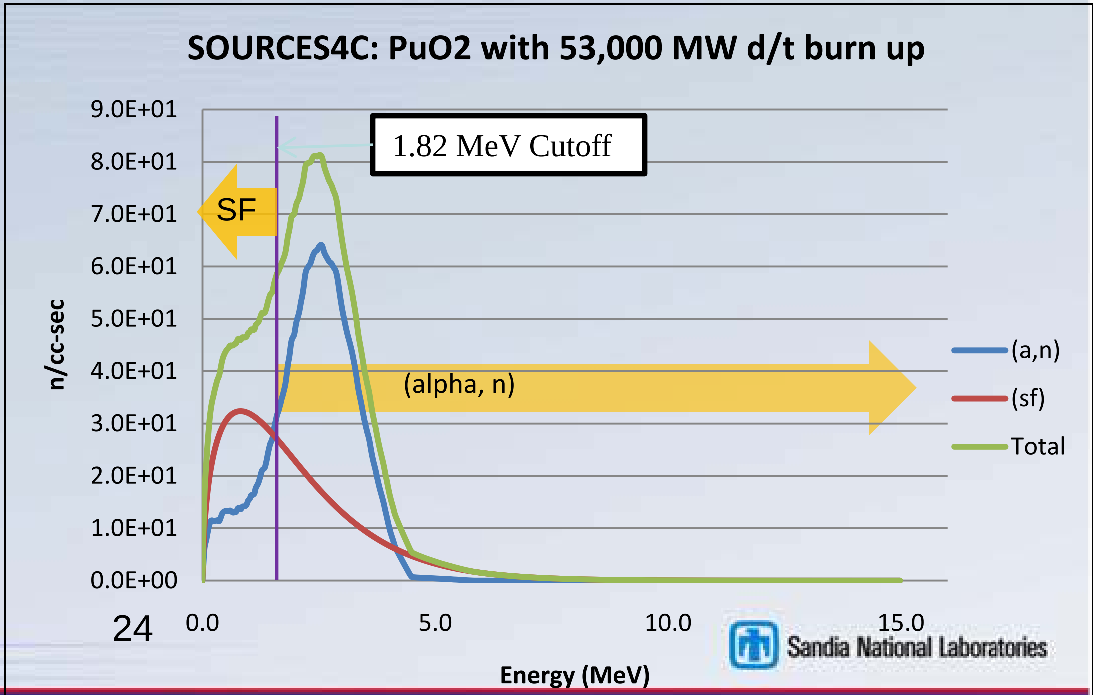
MATLAB

- **Wrote a code to use the ratio between (alpha, n) and SF rates to find cutoff line.**
  - Compared (alpha, n) to SF ratio rates across all energies in MCNP to the corresponding ratios in SOURCES4C
  - Found energy where difference in ratios is minimum and uses that point as cutoff to split the MCNP spectrum.
    - Left of line = SF
    - Right of line = (a,n)
  - Example: PWR with 53,000 burn up
    - Minimum difference in ratios of (a,n):SF between SOURCES4C and MCNP occurred at 1.82 MeV.





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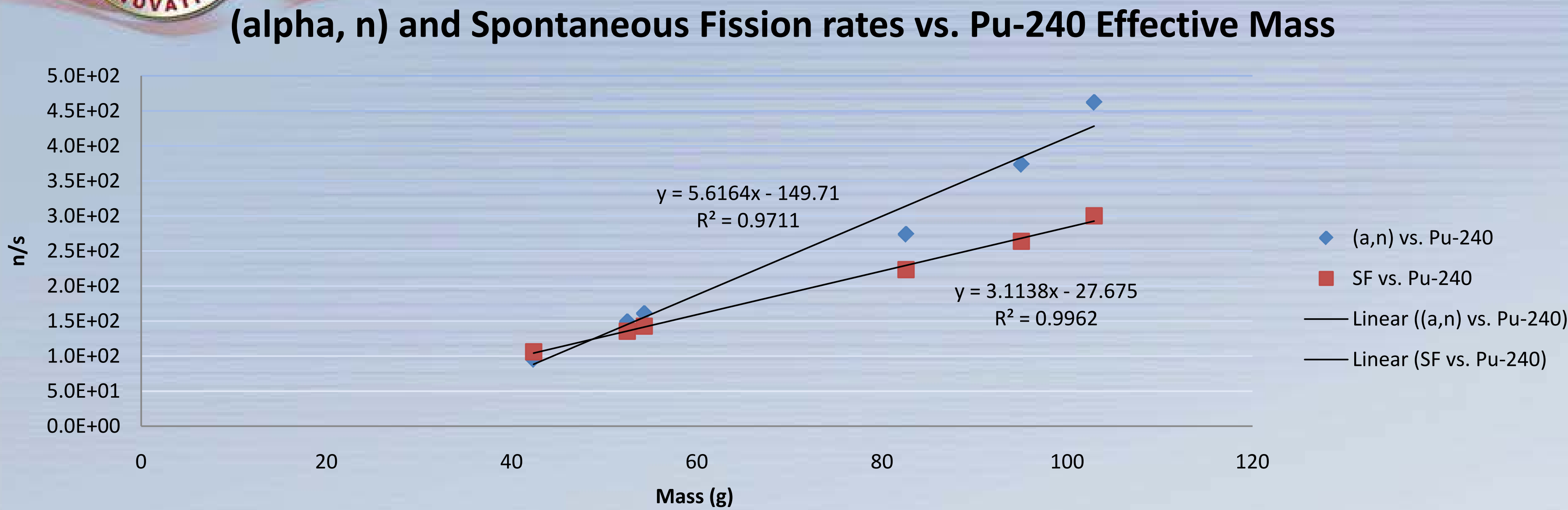
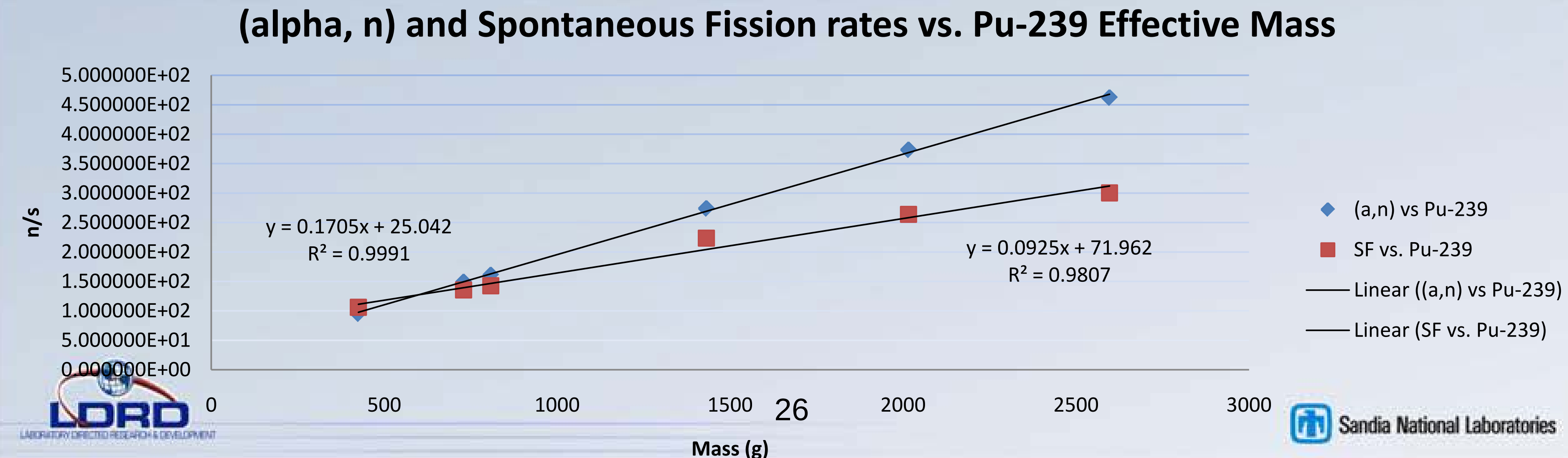
Compiled Results

Classification	True Pu mass for 1 cm^3	Effective Mass Pu-239	Effective Mass Pu-240	Integration of (a,n) spectrum (no threshold)	Integration of SF spectrum (no threshold)
PWR 33000	10.14134	1431.2524	82.5852	2.740000E+02	2.235000E+02
PWR 43000	10.13925	2015.6457	95.04192	3.736000E+02	2.639000E+02
PWR 53000	10.1394	2596.7244	102.89928	4.622000E+02	3.003000E+02
Magnox 3000	10.14289	425.27011	42.37944	9.52300E+01	1.061000E+02
SO1-8167	10.180538	808.00756	54.3256152	1.610000E+02	1.427000E+02
SO2-6808	10.18065	729.46677	52.4894256	1.492000E+02	1.355000E+02

MCNP Simulated Neutron Emission rates vs. Effective Masses





Discussion and Summary

- **We propose direct use of neutron signatures for nuclear safeguards material accountancy**
  - Two physical processes create neutrons with different energy spectra
  - For UF_6, simulations indicate enrichment can be extracted from emitted neutrons, even after full transport
  - For PuO_2, problem is more difficult due to varying isotopics, but initial analysis hints that it may be possible to estimate Pu mass
- **Advantages:**
  - Spectrum shape/magnitude gives UF_6 enrichment/mass
  - Spectrum shape/magnitude gives PuO_2 isotopic ratio/mass
  - Use of an imaging system (Neutron Scatter Camera or coded aperture system) suppresses backgrounds, allows mapping of material distribution

Future work

- **Develop a function of effective Pu-239 and Pu-240 masses to predict the “true” Pu sample mass.**
  - Estimate systematic error using this method.
  - Compare to existing technologies.
- **Consider realistic scenarios in reprocessing facilities**
  - Provide estimates of backgrounds, signal magnitudes.
  - Material distribution (imaging needs; neutron attenuation) and chemical forms (affects (alpha,n) distribution) in real plant process streams
- **Perform experiments/modeling for different detector concepts to determine optimal instrument**