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Neptunium Subcritical Observation (NeSO) Experiment Design

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Outline

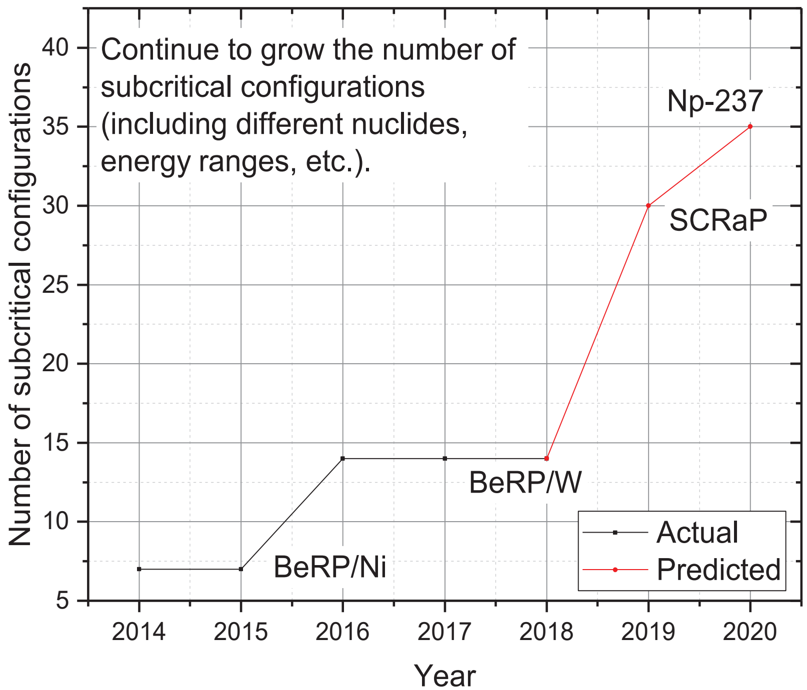
- **What is NeSO?**
- **Motivation**
- **The Sphere and Reflectors**
  – Material and configurations
- **Detectors and Analysis Method**
- **Monte Carlo Simulations**
  – Multiplication, Count Rates, Sensitivities, Uncertainties
- **Preliminary Measurements**
- **Composition Troubles**
- **Current and Future Work**

Overview of NeSO

- **Subcritical experiment with a 6kg sphere of Neptunium (“Np sphere”)**
- **Includes configurations with both the bare sphere and varying amounts of nickel reflection**
  - Nickel increases multiplication of system, leading to configurations spanning a variety of multiplication levels
- **To be performed at National Criticality Experiments Research Center (NCERC) at the Nevada National Security Site (NNSS)**
- **Goal is inclusion in International Criticality Safety Benchmark Evaluation Project (ICSBEP) Handbook**

Motivation

- **ICSBEP Benchmarks are crucial pieces of the nuclear data process**
  - Used in validation of new cross section sets
- **Help validate ^{237}Np nuclear data, and subcritical measurement methods**
  - Create a benchmark much more sensitive to ^{237}Np cross sections than any already in existence
- **^{237}Np is a byproduct of power reactors**
  - (n,γ) reactions of ^{235}U or (n,2n) reactions involving ^{238}U
  - ^{241}Am α-decay
- **Np sphere exists to better understand ^{237}Np critical mass**
  - Subject of previous critical benchmarks
- **Add to steadily growing group of NCERC subcritical benchmark measurements**



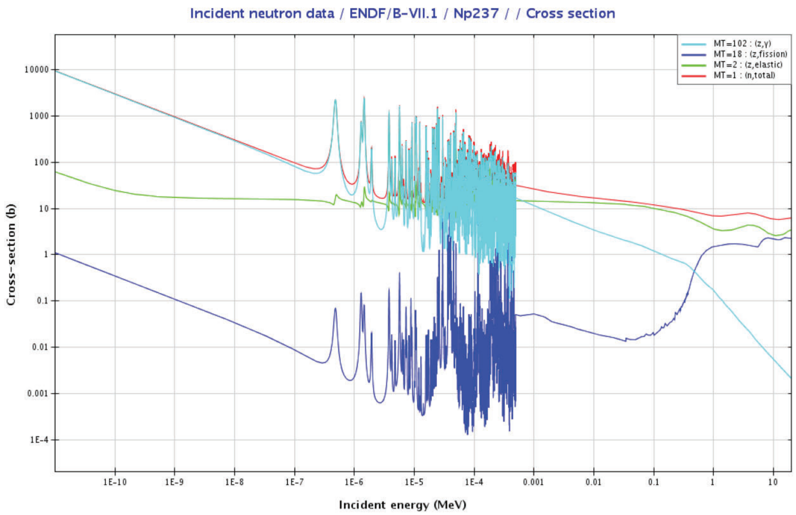
The Neptunium Sphere

- **Cast in 2001**
- **Total mass: 6070.4 grams**
  - ^{237}Np: 6060 grams
- **Radius: 4.149 centimeters**
- **Includes Tungsten and Nickel cladding**
  - Meant to decrease dose from ^{233}Pa γ-rays
  - Tungsten is 0.259 cm thick
  - Two layers of nickel, total 0.381 cm thick
- **Composition shown in table on right, from analysis of the surface**
  - Taken from previous critical benchmark
    - SPEC-MET-FAST-008, Np sphere surrounded by HEU
  - May not be representative of other parts of the sphere
  - Low emission rate
    - Spontaneous fission yield from PANDA Manual

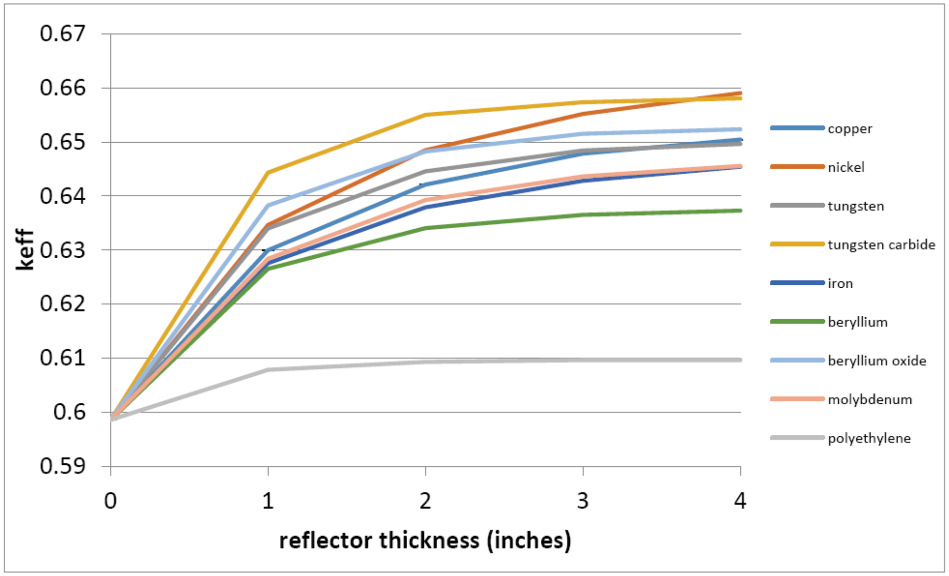
Nuclide	Mass (g)	S.F. yield (neutrons/s)
^{237}Np	6.06×10^{3}	6.90×10^{-1}
^{233}U	2.17×10^{-1}	1.87×10^{-4}
^{234}U	3.48×10^{-2}	1.75×10^{-4}
^{235}U	1.66	4.96×10^{-4}
^{236}U	9.28×10^{-3}	5.09×10^{-5}
^{238}U	1.87×10^{-1}	2.54×10^{-3}
^{238}Pu	9.83×10^{-2}	2.55×10^{2}
^{239}Pu	1.95	4.25×10^{-2}
^{240}Pu	1.40×10^{-1}	1.43×10^{2}
^{241}Pu	3.77×10^{-3}	1.88×10^{-4}
^{242}Pu	1.95×10^{-2}	3.35×10^{1}
^{241}Am	4.04×10^{-4}	4.76×10^{-4}
^{243}Am	1.12×10^{1}	-
Total	6.07×10^{3}	4.32×10^{2}

The Reflectors

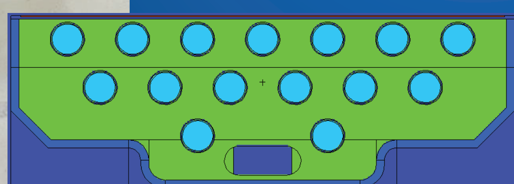
- ^{237}Np **is a threshold fissioner**
  - Reflecting materials such as polyethylene or graphite wouldn't increase multiplication as much
- **Simulations investigated a series of material choices**
  - Iron, tungsten, nickel, copper, beryllium, etc.
- **Nickel chosen due to larger range of multiplication factor values, and consistency with cladding**
  - Previous benchmark experience with Nickel 200, a high purity alloy



Graphic from JANIS



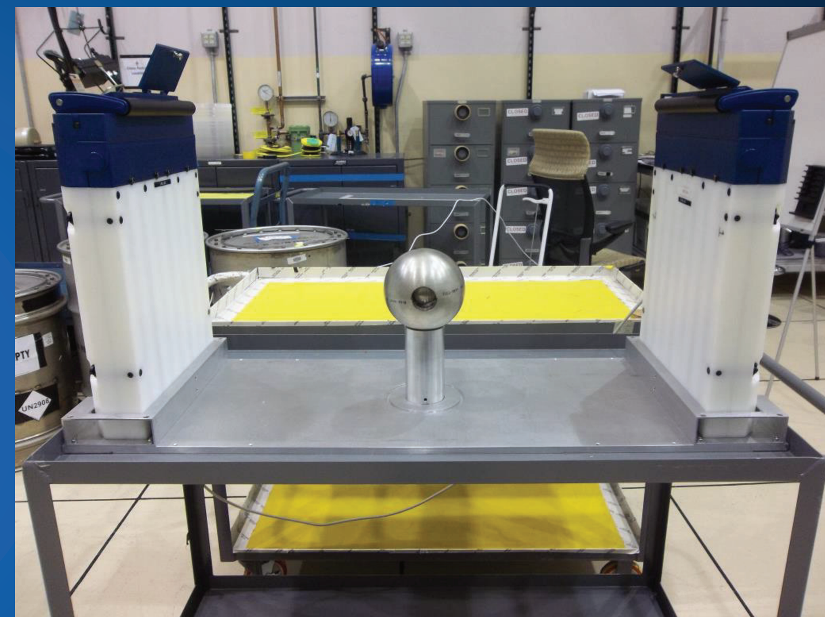
Detectors & Analysis Method



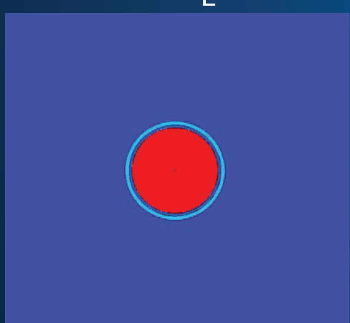
- **Neutron Multiplicity Array Detector (NoMAD)**
  - 15 ^{3}He tubes surrounded by polyethylene
  - Creates list-mode data
  - Two will be placed at 30 cm from the center of the sphere
- **Data will be analyzed with Hage-Cifarelli formalism of Feynman Variance-to-Mean technique**
  - Same as previous NCERC subcritical measurements
  - Allows to solve for leakage multiplication (M_L) from singles and doubles rates (R_1 and R_2)
    - M_L – number of neutrons that leave the system per starter neutron
    - R_1 – rate at which counts are recorded in the detector
    - R_2 – rate at which two neutrons from same fission chain are recorded in the detector

Final Configurations

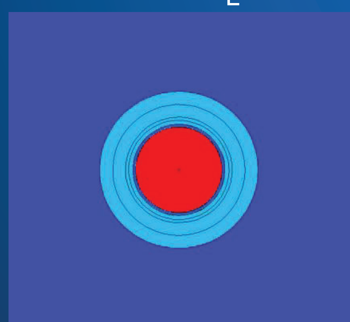
- **Bare (no added nickel), 0.6”, 1.1”, 2.1”, 3.6” Nickel**
  - A range of distinct M_L values
  - Smaller range than previous benchmarks, but still distinguishable
- **Nickel reflection from nesting spherical shells**
  - Similar in style to previous subcritical benchmarks



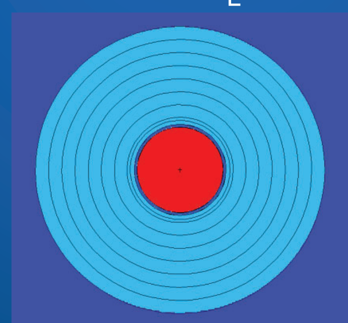
Bare – M_L 1.94



1.1” Ni – M_L 2.10



3.6” Ni – M_L 2.21



Statistical uncertainties ≤ 0.0005

Monte Carlo Simulations

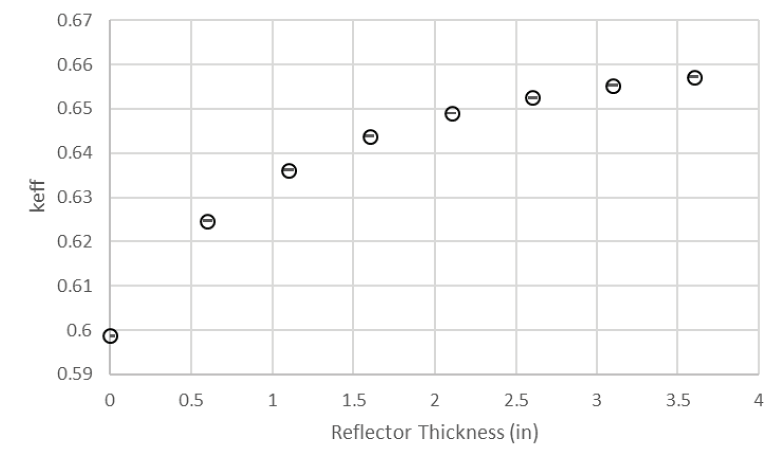
- **MCNP® version 6.2 KCODE criticality source computations, with ENDF/B-VII.1 cross sections**
  - Used to determine the effective multiplication factor k_{eff} for each experimental configuration
  - 5,000 active cycles, 10,000 neutrons per cycle
- **Can estimate total multiplication and leakage multiplication from k_{eff}**

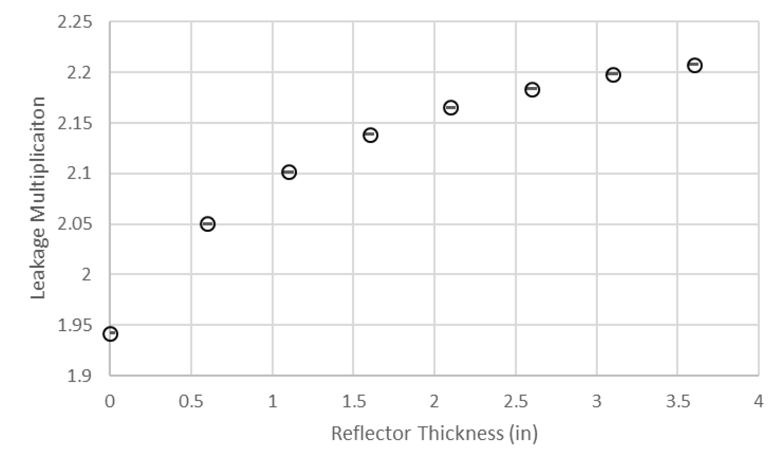
$$k_{eff} = \frac{k_p}{1-\beta_{eff}} \qquad M_T = \frac{1}{1-k_p}$$

$$M_L = \frac{1}{\bar{\nu}}\left[(\bar{\nu} - 1 - \alpha)M_T + 1 + \alpha\right]$$

$$\alpha = \frac{\Sigma_c}{\Sigma_f}$$

- **β_{eff} - effective delayed neutron fraction**
- **k_p - prompt multiplication factor**
- **M_T – total multiplication, the number of neutrons produced per starter neutron**
- **$\bar{\nu}$ - average number of neutrons produced in fission**





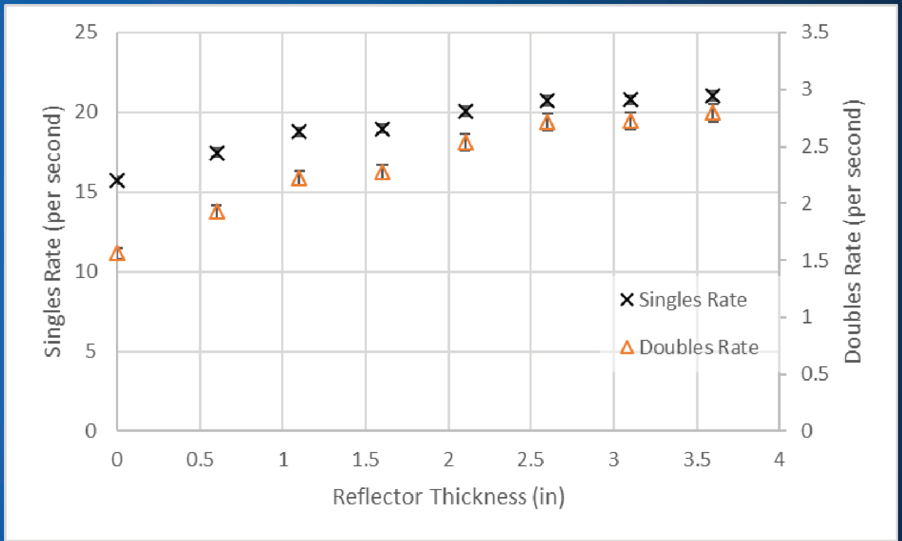
Estimation of Count Rates

- **Can further estimate singles and doubles rates from leakage multiplication**

$$R_1 = \varepsilon b_{11} F_S \qquad R_2 = \varepsilon^2 b_{21} F_S$$

$$b_{11} = M_L \overline{\nu_{S1}}$$

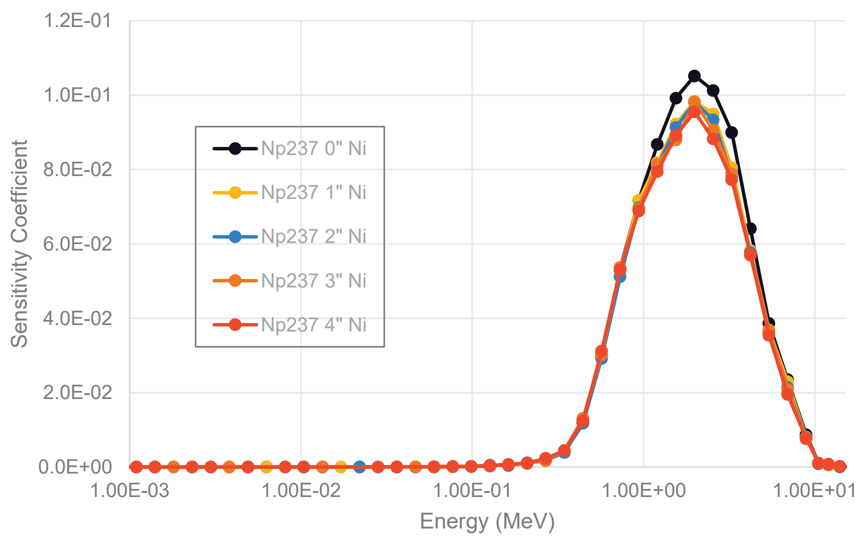
$$b_{21} = M_L^2 \left[\overline{\nu_{S2}} + \frac{M_L - 1}{\overline{\nu_{I1}} - 1} \overline{\nu_{S1}\nu_{I2}} \right]$$



- **ε – detector efficiency**
- **F_S - spontaneous fission rate**
- **$\overline{\nu_{In}}$ – nth reduced factorial moment of the induced fission neutron multiplicity distribution**
- **$\overline{\nu_{Sn}}$ – nth reduced factorial moment of the spontaneous fission neutron multiplicity distribution**

Cross Section Sensitivities

- **Integral and continuous energy cross section sensitivity coefficients were also calculated for various thicknesses of nickel**
- **Much more sensitive to ^{237}Np than previous benchmarks**
  - SPEC-MET-FAST-014, a critical benchmark that had the Np sphere with HEU and iron had total coefficient of 0.1401
- **Fast system, fast sensitivities**
- **Sensitivity coefficients to plutonium isotopes are very small (< 4.8E-4)**



Nickel Thickness	^{237}Np Total Cross Section Sensitivity
0.0	7.94E-01 $\pm$ 1.91E-3
1.0	7.52E-01 $\pm$ 2.03E-3
2.0	7.37E-01 $\pm$ 2.06E-3
3.0	7.37E-01 $\pm$ 2.14E-3
4.0	7.26E-01 $\pm$ 2.11E-3

As a function of energy

Over all energies

Sensitivity Analysis and Uncertainty Quantification

- **Perturb certain parameters by multiple times their uncertainty in each direction, treat difference in results as a derivative**
  - This is the sensitivity
- **To obtain uncertainty due to a parameter, multiply this derivative by the uncertainty in the parameter**
- **From table below, can see that count rates are very sensitive to plutonium content**
  - Leakage multiplication is less sensitive

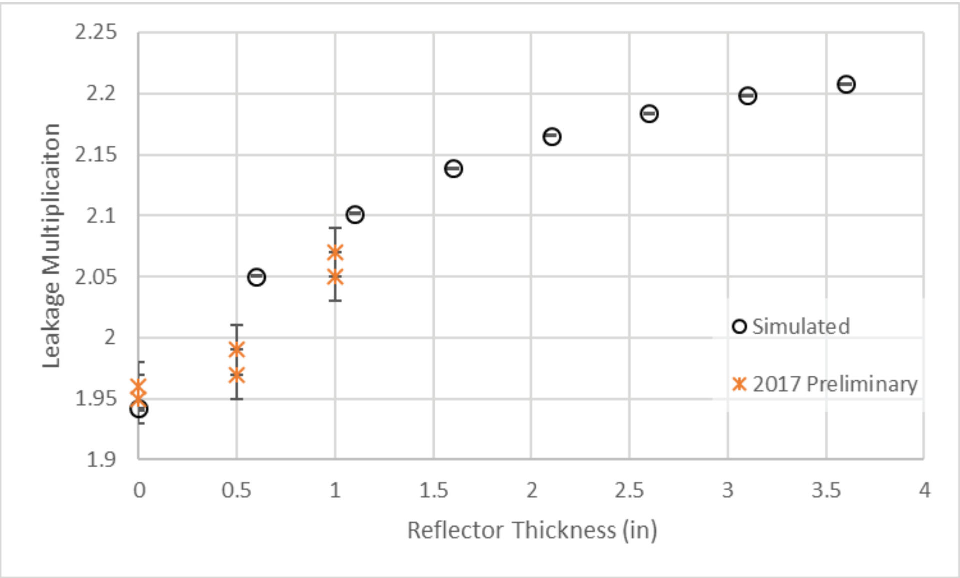
$$S_{k,x} = \frac{k_P - k_R}{P_x}$$

$$\delta k_x = u_x S_{k,x}$$

- **$S_{k,x}$ - sensitivity of benchmark parameter k to experimental parameter perturbation x**
- **P – Perturbation**
- **R – Reference**
- **δk_x - Uncertainty in k due to uncertainty in x**
- **u_x - uncertainty in x**

	M_L Sensitivity	M_L Uncertainty	R_1 Sensitivity	R_1 Uncertainty	R_2 Sensitivity	R_2 Uncertainty
Np Radius ± 2.74 mils	-0.6462	0.004497	0	0	0	0
Ni Cladding Thickness ± 2 mils	0.1029	0.001046	0	0	0	0
Ni Mass ± 0.5%	0.0008057	0.0008057	0	0	0	0
Pu Content +61g/–2g	0.0002269	0.014298	7.141	449.9	0.7139	44.97

Preliminary Measurements



Case	R_1	R_2	M_L
Bare	174.95 ± 0.36	17.753 ± 0.351	1.95 ± 0.02

- **Performed in Feb 2017 with 2 NoMAD systems at 47cm from center of sphere**
  - 30 minute measurements, much shorter than benchmark
- **Measured M_L matches with simulated data for non-reflected case**
  - Reflected shows right shape, but not exactly representative of benchmark shells
    - Current shells not made to fit Np sphere, some gaps present
    - Leftover from previous subcritical benchmark
- **Count rates do not agree**
  - Over an order of magnitude higher

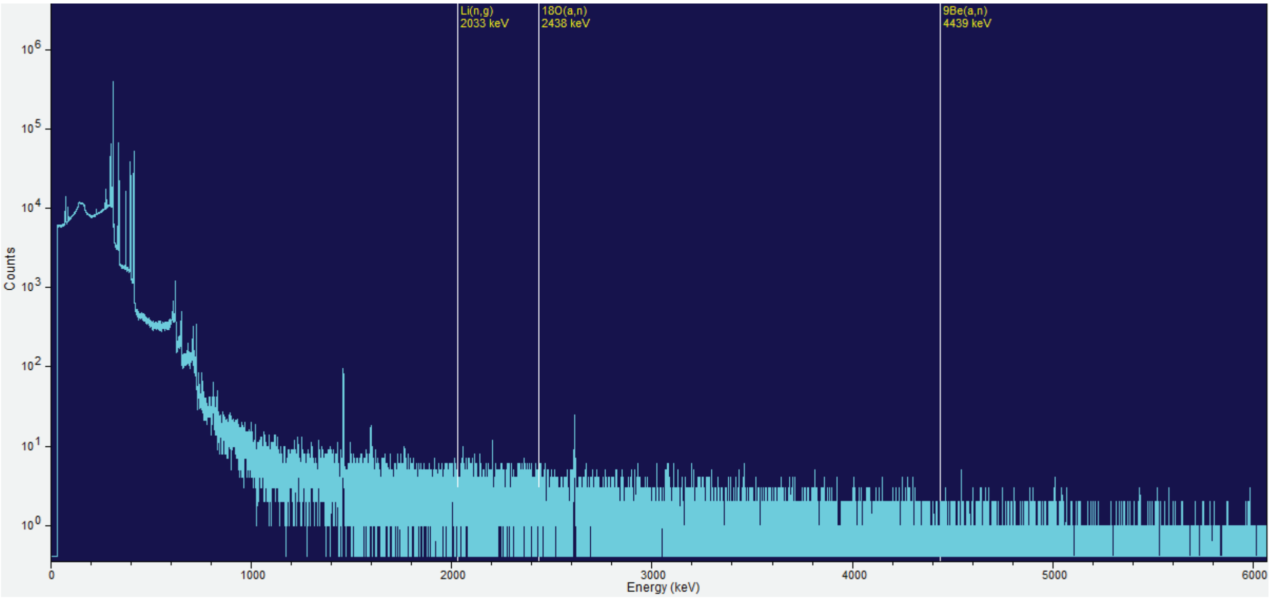
Composition Uncertainty

- **SPEC-MET-FAST-008, a previous critical benchmark with the sphere, says it contains 2 grams**
  - 2002 LANL report estimated 63 grams of Pu based on emission rate
- **Neutron emission rate appears to be changing over time**
- **Unsure if extra neutrons are from plutonium, curium, or something else entirely**
- **Also appears to be a hot spot, as detectors place on opposite sides of the sphere reported a approximately 15% difference in counts**

Date	Estimated Emission Rate (n/s)
2002	12,000
February 2017	8,700
February 2018	8,400
June 2018	8,300
August 2018	8,150

Composition Uncertainty, cont.

- **Gamma spectroscopy difficult due to shielding**
  - Lower energy peaks are suppressed
- **Looking at spectrum, (α, n) emission unlikely due to lack of high energy characteristic peaks**
  - Even though peaks are suppressed, complete lack of counts in some peaks points to lack of (α, n) emission



Conclusion

- **While disagreement in count rates is discouraging, the agreement between the measured and simulated leakage multiplication is still promising for a potential criticality safety benchmark**
  - Benchmark would just focus on leakage multiplication, similar to many critical benchmarks only focusing on k_{eff}
  - Leakage multiplication is relatively insensitive to uncertainty in plutonium content
- **This proposed experiment is much more sensitive to 237Np than any previous benchmark**
  - Will be very important to validating nuclear data related to this nuclide
- **Execution of the experiment is expected to take place in the near future**
  - All parts are been received
- **After execution, benchmark will be written for submission to ICSBEP**

Thank you!

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