Daily Check-In:

Wednesday

Please take a moment to check in for today's session.

MIRION Connect AMERICAS 26
Annual Users' Conference

Advancing Radiation Safety.
Empowering Progress.

Quantification of Gamma Rays from Actinide Prompt Gamma Neutron Activation Analysis

C. Egozi

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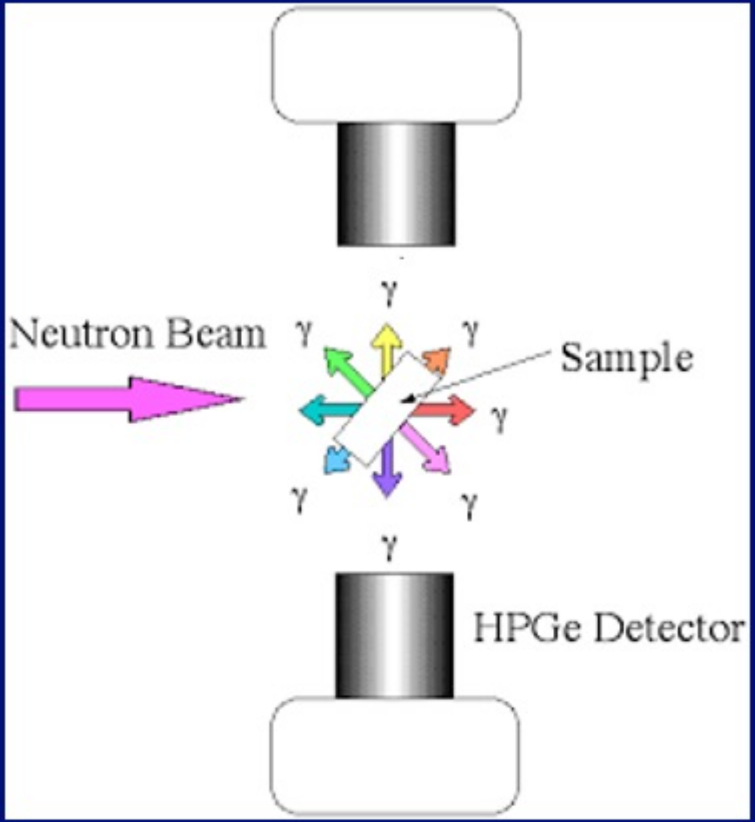
Agenda

1. Proposed experiment, what is PGNAA? ←
2. Prior experimental PGNAA
3. Prior MCNP PGNAA
4. Proposed measurement plan
5. Conclusions

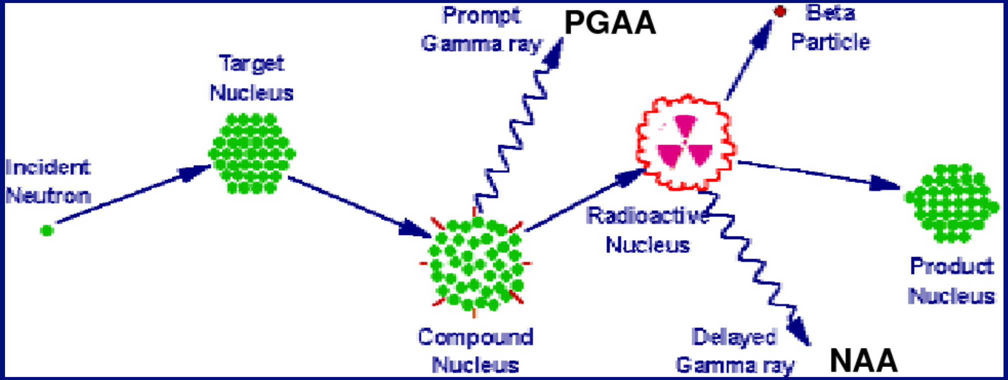
Proposed Experiment

- Irradiation of Pu and U isotopes
  - Measure gamma rays while irradiation is occurring
  - Known as Prompt Gamma Neutron Activation Analysis
- High energy gammas can be used for characterization of isotopes in highly shielded environments



Prompt Gamma Neutron Activation Analysis (PGNAA)

- A sample undergoes neutron radiative capture and emits a gamma in an (n,γ) reaction
- These gammas can have much higher energy than the decay gammas of a radionuclide
- The IAEA has a documented PGNAA compendium for 79 elements up to uranium



Elements Characterized by PGNAA

1 H																	2 He
3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne
11 Na	12 Mg											13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr
37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe
55 Cs	56 Ba	57* La	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn
87 Fr	88 Ra	89** Ac	104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt	110 *	111 *	112 *						
* Lanthanides				58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu
** Actinides				90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr

- Nearly all elements up to uranium, as well as ^{237}Np, ^{241}Am, and ^{242}Pu have been characterized
- IAEA database has information on PGNAA gamma rays for many isotopes (blue) but not for ^{239}Pu.

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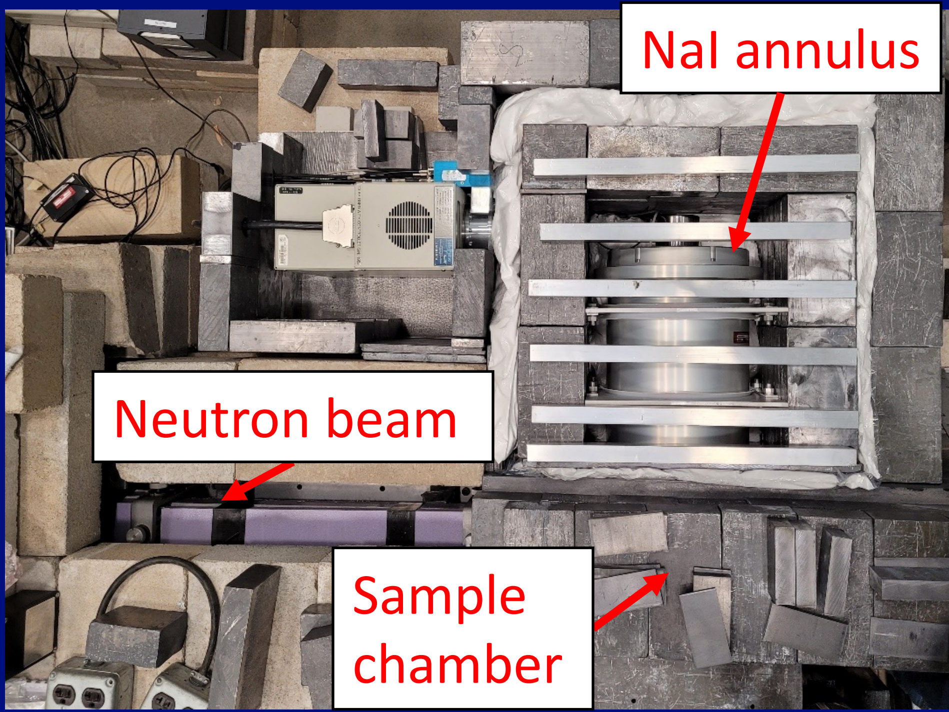
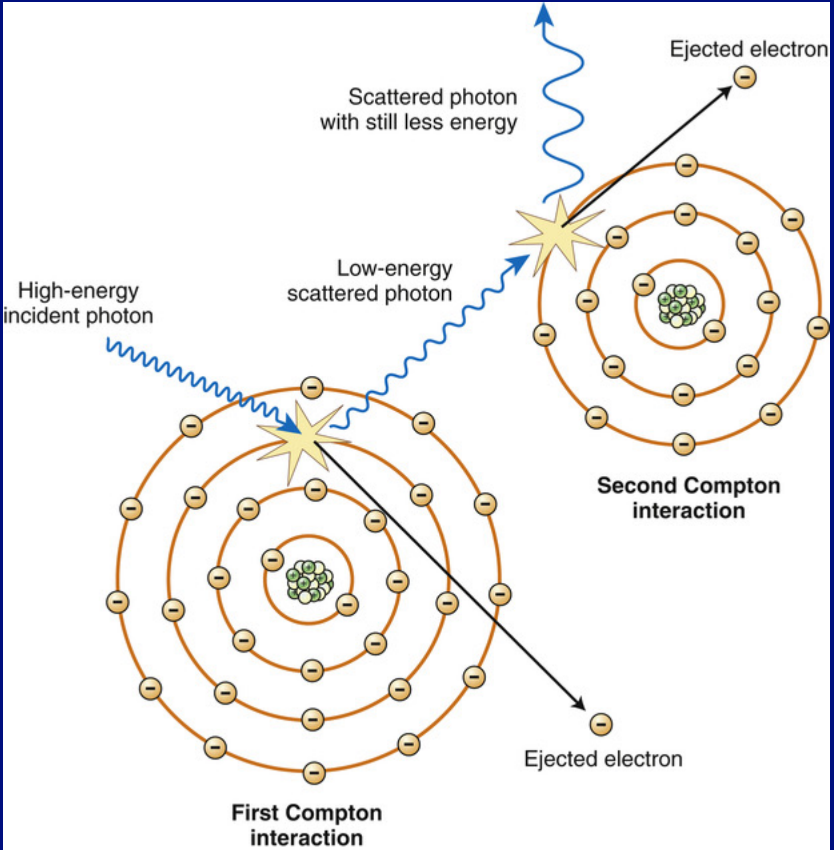
Prior Experimental Setup

- PGNAA at the University of Texas Nuclear Engineering Teaching Lab (NETL) has cold neutron beam line, lead lined sample holder, and Compton suppression (CS) with NaI annulus around HPGe detector



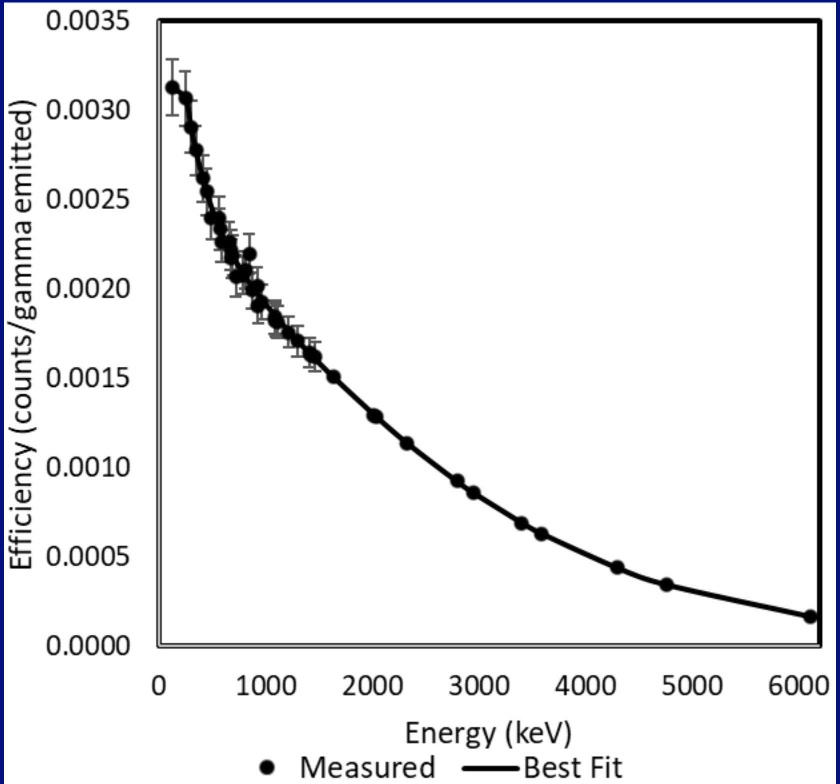
Compton Suppression

- In CS, HPGe surrounded by NaI annulus, and any photon reaching the two detectors simultaneously is not counted



Detector Efficiency

- Efficiency determined using a combination of a 65 hr count of a 65 kBq ^{152}Eu standard and a 6 hr PGNAA count of an iron foil
- Due to much higher energies of prompt gamma rays than decay gammas

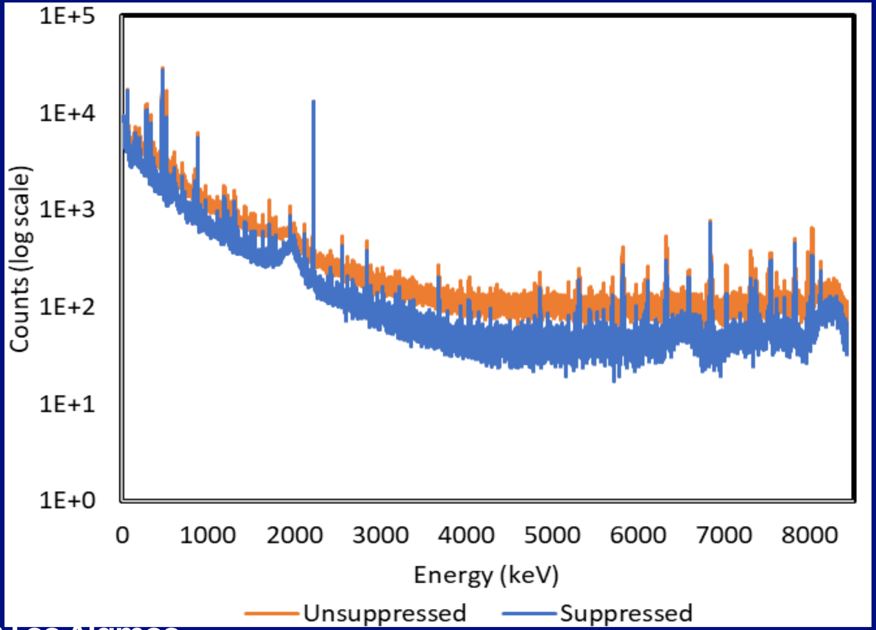


$$\varepsilon = e^{0.0526*\ln(E)^4-1.3192*\ln(E)^3+12.4060*\ln(E)^2-51.4877*\ln(E)+85.0532}$$

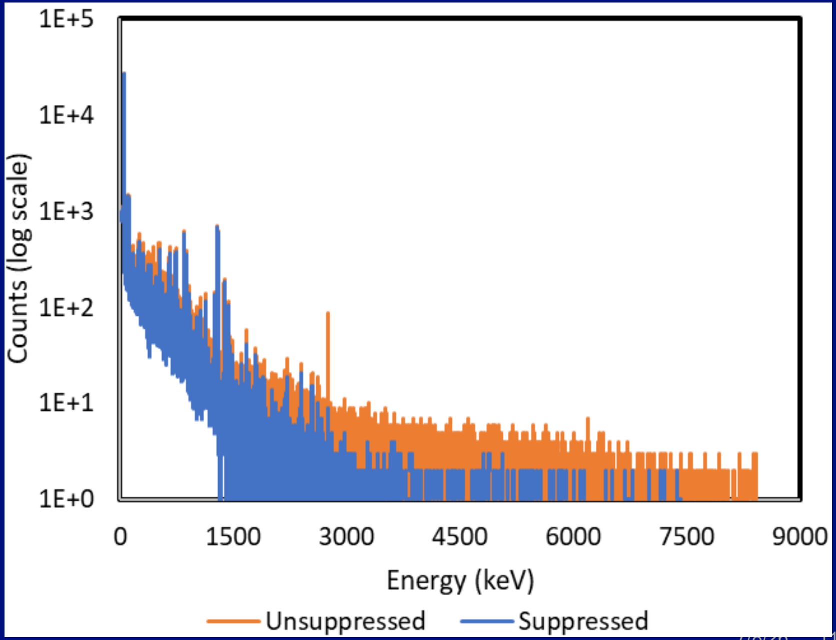
PGNAA for ^{239}Pu with CS

- ^{239}Pu foil counted on system (in CS and unsuppressed modes) for 4 hrs with PGNAA, then for 8 hrs after activation to eliminate fission product gammas

4 hr count



8 hr count



PGNAA Yield for ^{239}Pu with CS

- 12 gamma rays identified from CS spectrum as ^{239}Pu PGNAA gammas
- Efficiencies calculated with efficiency curve equation
- The yields were calculated by:

$$Y_{^{239}Pu} = Y_{^{58}Ni} * \frac{\varepsilon(E)_{^{58}Ni}}{\varepsilon(E)_{^{239}Pu}} * \frac{m_{^{58}Ni}}{m_{^{239}Pu}} * \frac{C_{^{239}Pu}}{C_{^{58}Ni}}$$

Energy (keV)	Net Peak Area (counts)	Efficiency (counts/g)	Yield (barns)	% Uncertainty
1427.53	4819 ± 149	1.63E-3 ± 7.64E-8	4.98 ± 0.55	11.09
1633.03	4179 ± 146	1.51E-3 ± 7.06E-8	4.67 ± 0.52	11.22
2015.2	1996 ± 147	1.29E-3 ± 6.07E-8	2.6 ± 0.34	13.05
2032.31	706 ± 151	1.28E-3 ± 6.03E-8	0.93 ± 0.23	24.49
2321.65	413 ± 90	1.14E-3 ± 5.33E-8	0.61 ± 0.16	25.59
2790.08	298 ± 69	9.22E-4 ± 4.33E-8	0.54 ± 0.15	27.07
2944.12	325 ± 100	8.58E-4 ± 4.03E-8	0.64 ± 0.21	33.43
3401.32	204 ± 69	6.89E-4 ± 3.23E-8	0.5 ± 0.18	36.89
3579.6	668 ± 112	6.31E-4 ± 2.96E-8	1.78 ± 0.36	20.02
4294.47	244 ± 63	4.37E-4 ± 2.05E-8	0.94 ± 0.27	28.42
4754.15	1048 ± 103	3.43E-4 ± 1.61E-8	5.14 ± 0.75	14.51
6102	3975 ± 100	1.64E-4 ± 7.70E-9	40.76 ± 4.44	10.9

New ^{239}Pu Source Characterization

- 2 new sources were obtained, with ^{239}Pu electrodeposited on stainless steel and certified activities of 17.8 ± 0.89 kBq and 13.9 ± 0.70 kBq
- Their activities were determined experimentally with passive gamma spectrometry relative to the 1.81E6 ^{239}Pu standard, using the 129 keV gamma.

	Net counts	Count Time (s)	Measured Activity (kBq)
1.81E3 ± 12.1 kBq standard	11019 ± 106	504.86	n/a
17.8 ± 0.9 kBq source	5586 ± 140	25107.7	18.5 ± 0.5
13.9 ± 0.7 kBq source	3137 ± 121	18335.7	14.2 ± 0.6

^{239}Pu Yield Normalization

- The (n,γ) reaction rate was determined with the 884 keV ^{134}Xe fission product gamma and calculated from the fission product reaction rate equation:

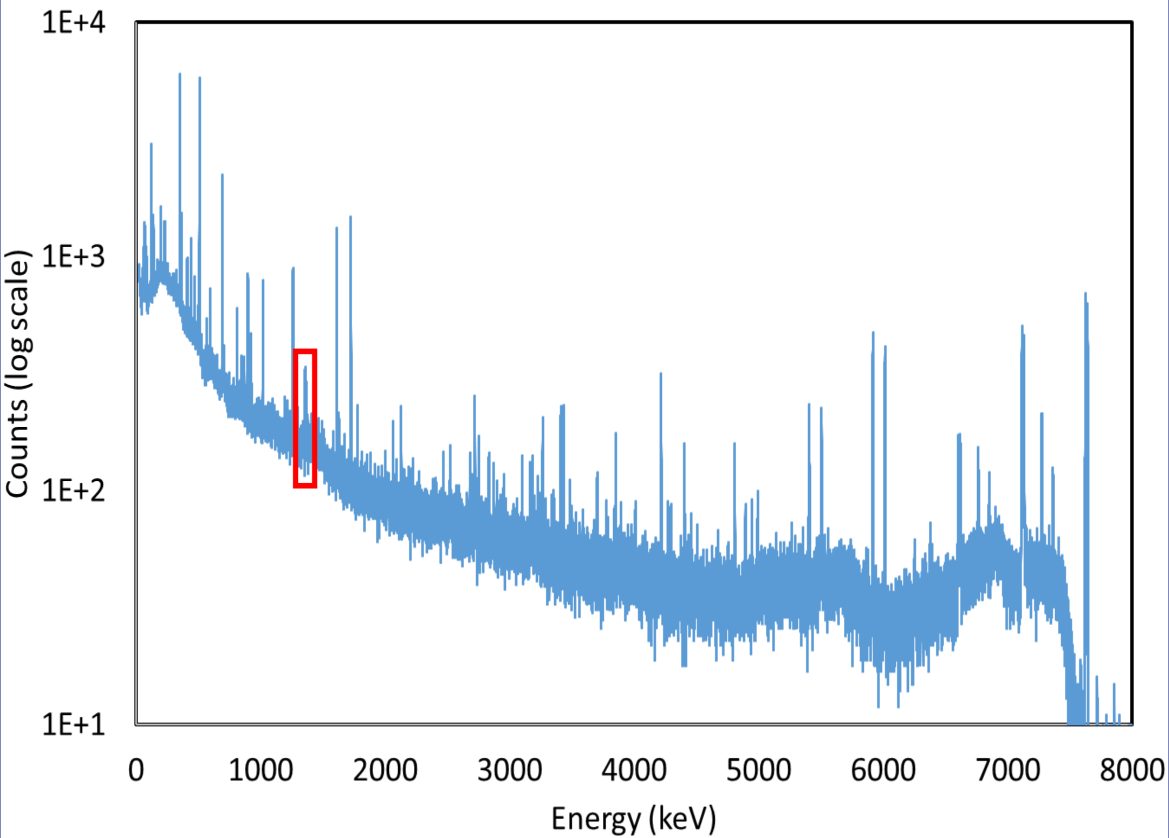
$$RR_f = \frac{\frac{N*\ln(2)}{t_{1/2}}}{\left(1-e^{-t_c*\frac{\ln(2)}{t_{1/2}}}\right)*\left(1-e^{-t_i*\frac{\ln(2)}{t_{1/2}}}\right)}$$

- The capture reaction rate, gamma net areas, active radius, and efficiencies were used to calculate the normalized prompt gamma yields (Y_n): $Y_n = \frac{N}{\varepsilon*RR_\gamma*\pi*r^2}$

Energy (keV)		Net area	Yield Normalization
2015	0.001294 ± 0.000081	2329 ± 150	941.38
6102	0.000164 ± 0.000048	3975 ± 99	12658.9

^{239}Pu Mass Determination

- The ^{239}Pu mass was determined with the normalized yields and relative to the ^{57}Fe 1358 keV prompt gamma from a 4-hour iron foil PGNAA count



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New ^{239}Pu Mass

- The mass was calculated by: $m_{239} = \frac{Y_{57} * N_{239} * \varepsilon_{57} * m_{57}}{Y_{239} * N_{57} * \varepsilon_{239}}$
- And uncertainty propagated by:

$$\Delta m_{{}^{239}_{17.8}Pu} = m_{{}^{239}_{17.8}Pu} * \sqrt{\left(\frac{\Delta N_{{}^{239}_{17.8}Pu}}{N_{{}^{239}_{17.8}Pu}}\right)^2 + \left(\frac{\Delta Y_{{}^{57}Fe}}{Y_{{}^{57}Fe}}\right)^2 + \left(\frac{\Delta \varepsilon_{{}^{239}_{E}Pu}}{\varepsilon_{{}^{239}_{E}Pu}}\right)^2 + \left(\frac{\Delta N_{{}^{134}Xe}}{N_{{}^{134}Xe}}\right)^2 + \left(\frac{\Delta N_{{}^{57}Fe}}{N_{{}^{57}Fe}}\right)^2 + \left(\frac{\Delta N_{{}^{239}_{1.81}Pu}}{N_{{}^{239}_{1.81}Pu}}\right)^2 + 0.05^2}$$

- Detection limits calculated by Currie equation:

$$L_D = 2.71 + 4.65\sqrt{B}$$

New ^{239}Pu Mass

- Activities agree within 11%

	17.8 ± 0.89 kBq	13.9 ± 0.70 kBq
^{57}Fe net area	7252 ± 173	6956 ± 165
^{57}Fe efficiency	0.001671 ± 0.000061	0.001671 ± 0.000061
2015 keV ^{239}Pu net area	1097 ± 127	932 ± 118
6102 keV ^{239}Pu net area	1651 ± 115	1475 ± 112
2015 keV ^{239}Pu mass	7.90E-06 ± 1.17E-06	7.00E-06 ± 1.03E-06
6102 keV ^{239}Pu mass	6.96E-06 ± 6.86E-07	6.49E-06 ± 6.39E-07
2015 keV ^{239}Pu activity (kBq)	18.14 ± 2.68	16.07 ± 2.37
6102 keV ^{239}Pu activity (kBq)	16.00 ± 1.58	14.90 ± 1.47
Average activity (kBq)	17.07 ± 3.03	15.48 ± 2.75
2015 keV DL (kBq)	7.62	8.48
6102 keV DL (kBq)	4.39	4.50

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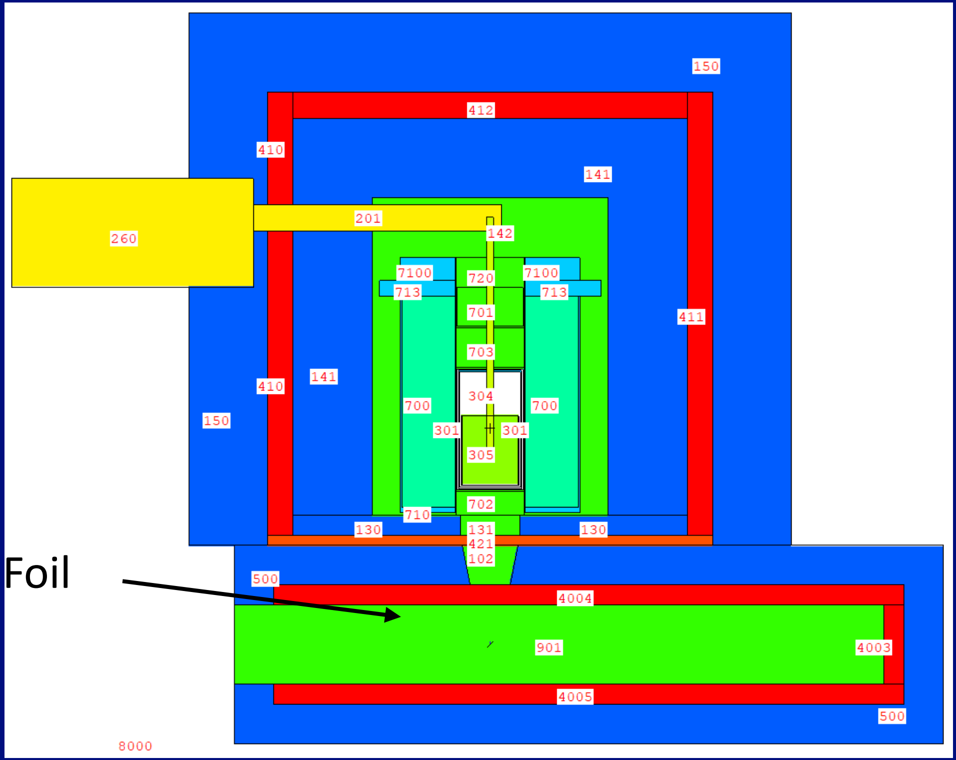
MCNP of PGNAA System

- An MCNP simulation was generated to reflect the experimental setup as accurately as possible

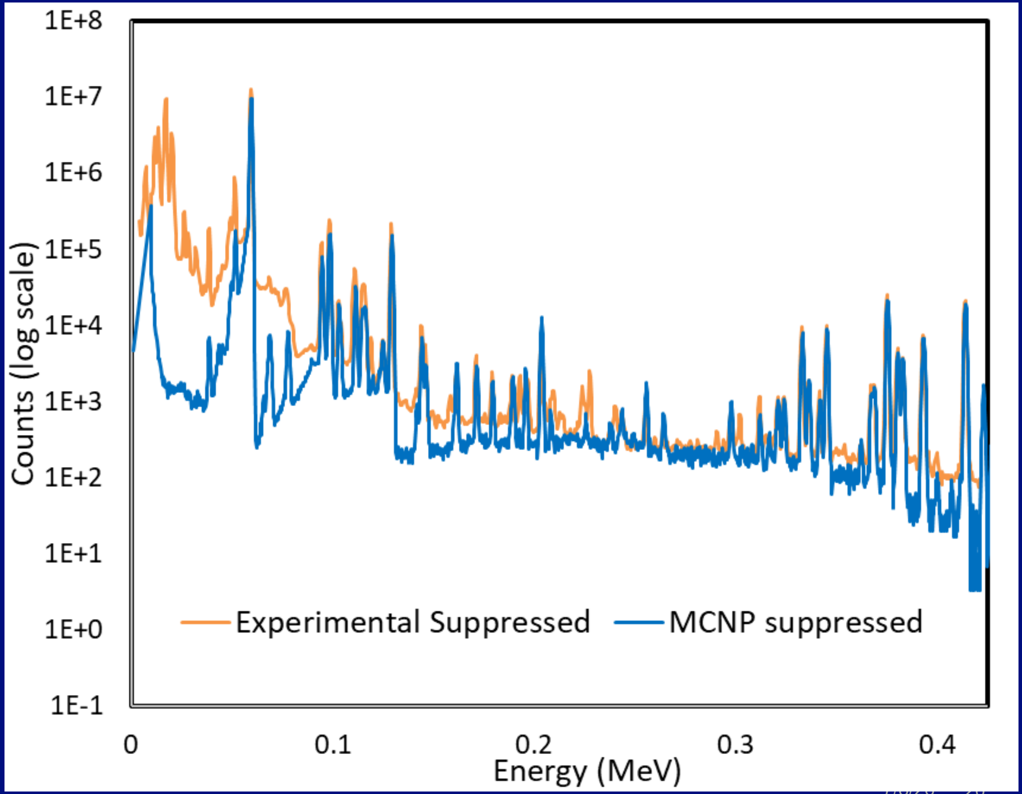
Materials Legend:

- 301, 303, 710, 7100, 711, 713, 714, : Al
- 302, 304, 8000: Vacuum
- 305, 307: HPGe
- 306, 202: Cu
- 700: NaI
- 701, 702, 703, 131, 142, 151, 720, 901, 102: Air
- 200: ^{239}Pu foil
- 130, 140, 141, 150, 160, 500: Pb
- 201, 260: Cryocooler
- 410, 411, 412, 4003, 4004, 4005: Li_2CO_3 (Natural)
- 421: Li_2CO_3 (Enriched to 95% Li_6)
- 800, 801, 802, 803: Concrete

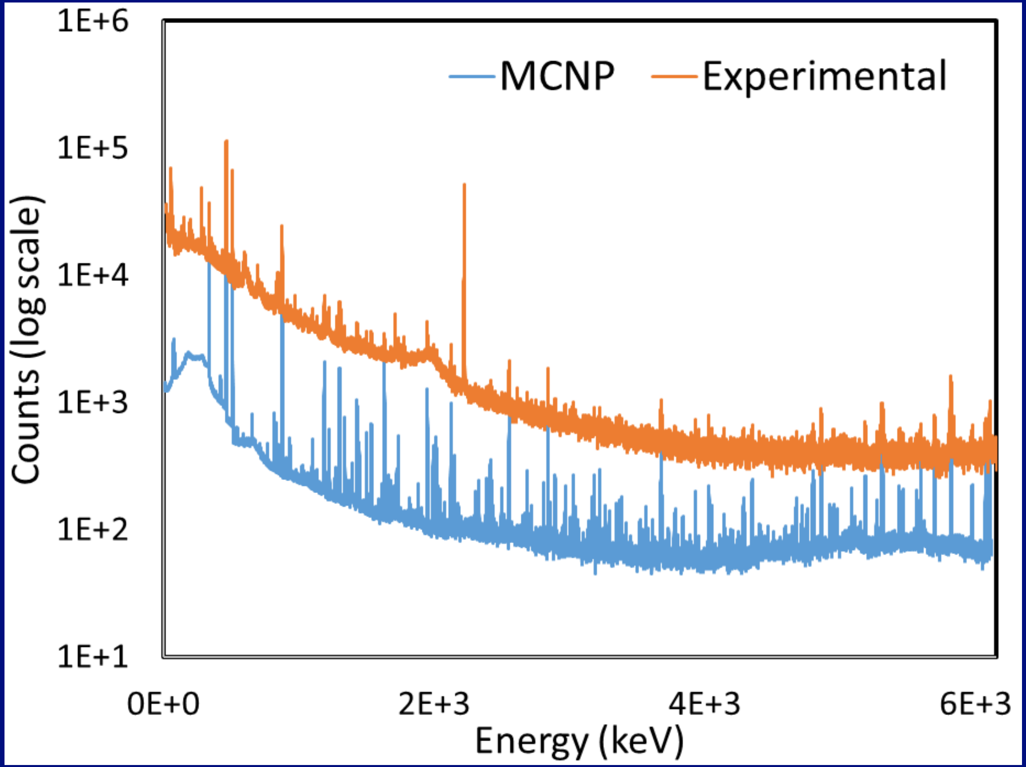


MCNP vs Experimental CS

- The MCNP model was used with an F8 PHL tally for Compton suppression and the spectral results were compared to experiment
- Above 100 keV, the spectra are in agreement



MCNP vs Experimental PGNAA Spectra



- The MCNP model was split into two inputs
  - One for ^{239}Pu with gamma rays given in SI card
  - One for nickel with neutron source to induce radiative capture
- Spectra were combined and compared to experimental results

MCNP vs Experimental PGNAA Net Areas

- Net areas of ^{239}Pu and ^{58}Ni prompt gammas were within 11% of experimental values above 1000 keV
- Significant amount of PFA experimentally (but not in the model), emits 1633 keV fluorine prompt gamma, resulting in little confidence in the ^{239}Pu 1633 keV gamma.

Gamma Ray (keV)	Isotope	Experimental net area	MCNP net area	% Difference
339	^{58}Ni	106918 ± 574	39688 ± 304	62.9
465	^{58}Ni	490126 ± 825	123883 ± 403	74.7
1427	^{239}Pu	9446 ± 250	10067 ± 159	6.57
1435	^{239}Pu	7998 ± 255	8670 ± 156	8.40
1633	^{239}Pu	54417 ± 316	25557 ± 197	53.0
1949	^{58}Ni	15040 ± 229	12708 ± 152	15.5
1992	^{58}Ni	3956 ± 200	3582 ± 116	9.45
2015	^{239}Pu	5039 ± 198	5279 ± 123	4.76
2032	^{239}Pu	1034 ± 180	1152 ± 108	11.4
2554	^{58}Ni	10845 ± 190	11182 ± 137	3.10
2842	^{58}Ni	10685 ± 183	11891 ± 134	11.3
3025	^{58}Ni	3128 ± 146	3070 ± 96	1.85
4754	^{239}Pu	2371 ± 149	2116 ± 94	10.8
6102	^{239}Pu	9676 ± 162	10738 ± 123	11.0

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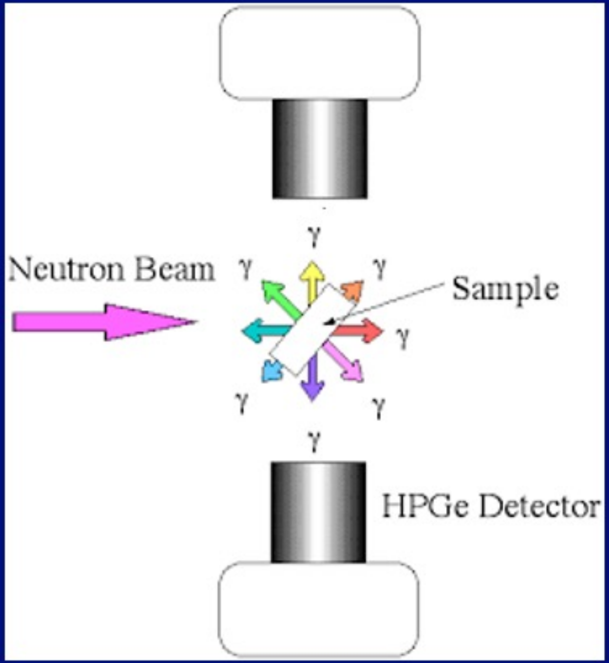
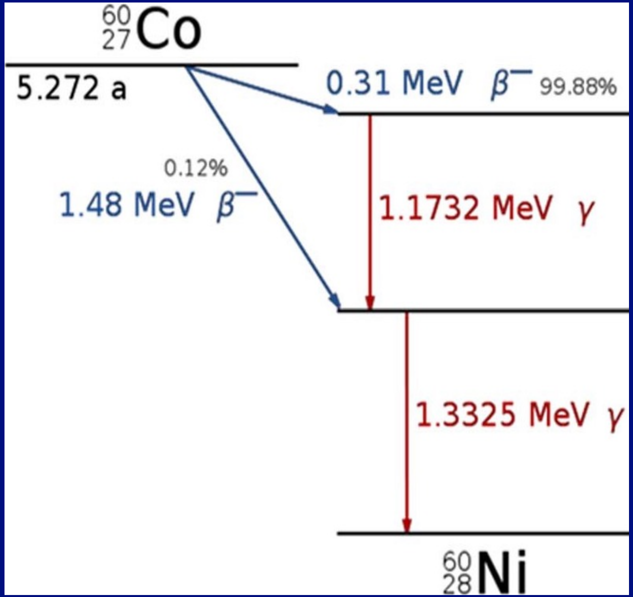
Proposed Measurements

- Experimental measurements to be performed at the Los Alamos Neutron Science Center (LANSCE)
- MCNP simulations to optimize shielding and geometry
- In collaboration with the MCNP team to potentially incorporate isotopic PGNAA data into MCNP database
- Isotopes of interest include ^{239}Pu, ^{240}Pu, and ^{235}U encased in Kapton
- To be counted with two HPGe detectors in coincidence to determine decay scheme



Gamma-Gamma Coincidence

- Two HPGe or BGO detectors facing each other with source between them
- Utilized to generate decay scheme of radionuclides
- Coincidence counting with energy gating can be performed at LANSCE to create decay scheme of prompt gammas of isotopes of interest



Conclusions

- PGNAA produces much higher gamma rays than radioactive decay does, and therefore the gammas are far less likely to become attenuated
- This could be significant for ^{239}Pu detection within nuclear material storage containers, where it is not always possible to remove the source for gamma spectrometry measurements.
- Also potentially for interrogation of highly shielded ^{239}Pu within criticality safety limits
- Necessary to perform measurements with pure samples due to prior interferences from metal foils

Questions?

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Research & Technology Forum

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