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Nuclear Data Needs for γ–γ Coincidence Analysis

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PNNL is operated by Battelle for the US Dept. of Energy

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Outline

- PNNL and Our Shallow Underground Laboratory
- CASCADES Germanium Array
- Melusine and the Coincidence Lookup Library
- Nuclear Data
- Summary and Future Work

Installing 2nd CASCADES Cryostat

PNNL Locations in Washington State

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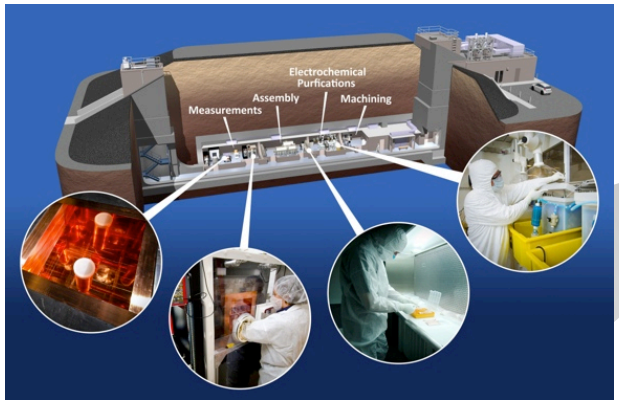
Pacific Northwest Center for
Global Security

Marine Research Operations
Sequim, Washington



PSF and the new shallow underground lab

Richland campus

PNNL Shallow Underground Laboratory

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Aalseth, C.E., et al. (2012). "A shallow underground laboratory for low-background radiation measurements and materials development." Rev. Sci. Inst. **83**(11): 113503.

http://tour.pnnl.gov/shallow-lab.html

CASCADES HPGe Array

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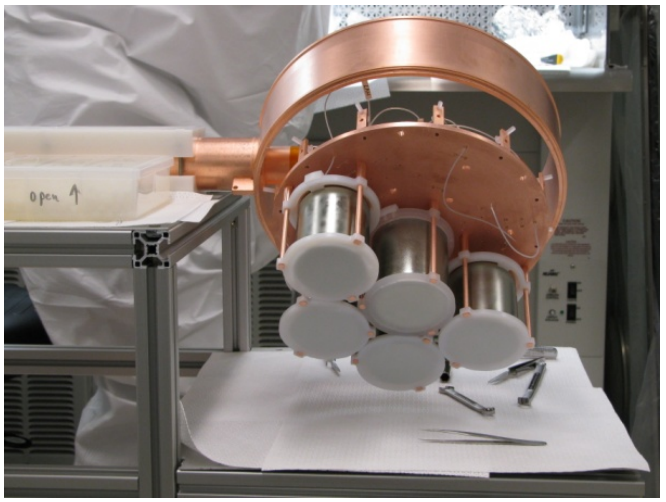
- 14 crystal HPGe array operating in shallow underground lab at PNNL
  - Two cryostats
  - 2 crystals in upper cryostat are not operating
- Low-background construction and techniques
  - PNNL electroformed copper cryostats
  - Low-background materials
  - Graded shielding
  - ~30 m.w.e. overburden
  - Active anti-cosmic veto
  - Radon exclusion system
- Development was funded by DOE Ground-Based Nuclear Explosion Monitoring (GNEM) Program (Leslie Casey)



Crystal installation into 2nd cryostat

HPGe array in PNNL shallow lab

Technical Approach

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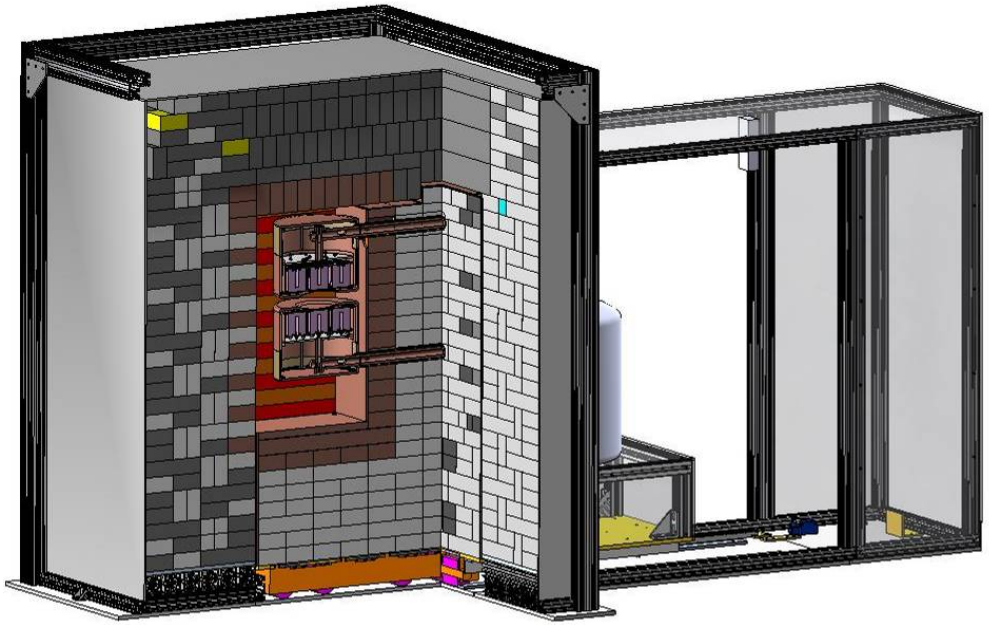
Performance Objectives

- High Efficiency
- High Selectivity
- Low Background



Sample Types

- Filter Paper
  - Whole sample
  - Archive portion
- Liquid sample after radiochemical dissolution
- Separated / group separated samples or solutions
- Swipes, etc
- International Monitoring System Samples?

Focus areas: nuclear forensics, material assay

“Melusine” Gamma Spectroscopy Analysis Code

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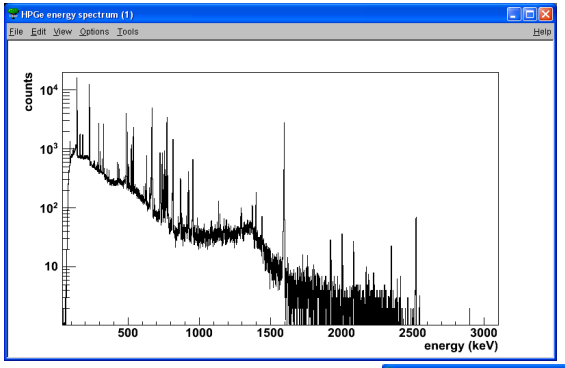
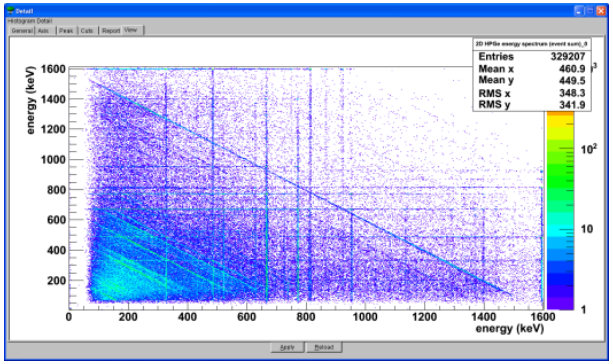
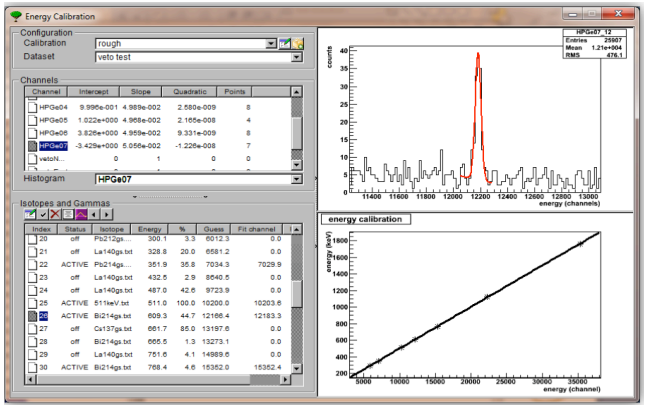
- Gamma spectroscopy code to support analysis of data from multi-coincident detector systems, e.g. CASCADES
- Provides typical gamma spectroscopy functionality
  - Energy and shape calibration
  - Detection efficiency calibration
  - Peak search
  - Isotope identification
  - Activity calculations
- Provides additional capabilities for coincidence systems
  - Flexible data reconstruction
  - 1- and 2-D histograms
  - User-defined coincidence, anti-coincidence, energy cuts, etc
- Relies on the Coincidence Lookup Library for coincidence signature intensities



“Singles” and coincidence spectra from thermal irradiation of ^{235}U





Energy calibration panel

“Melusine” Gamma Spectroscopy Analysis Code

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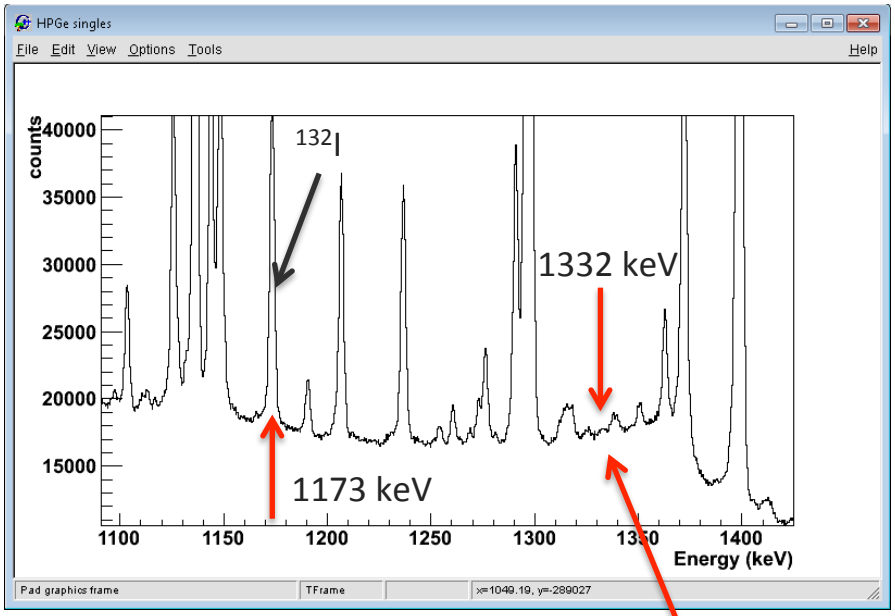
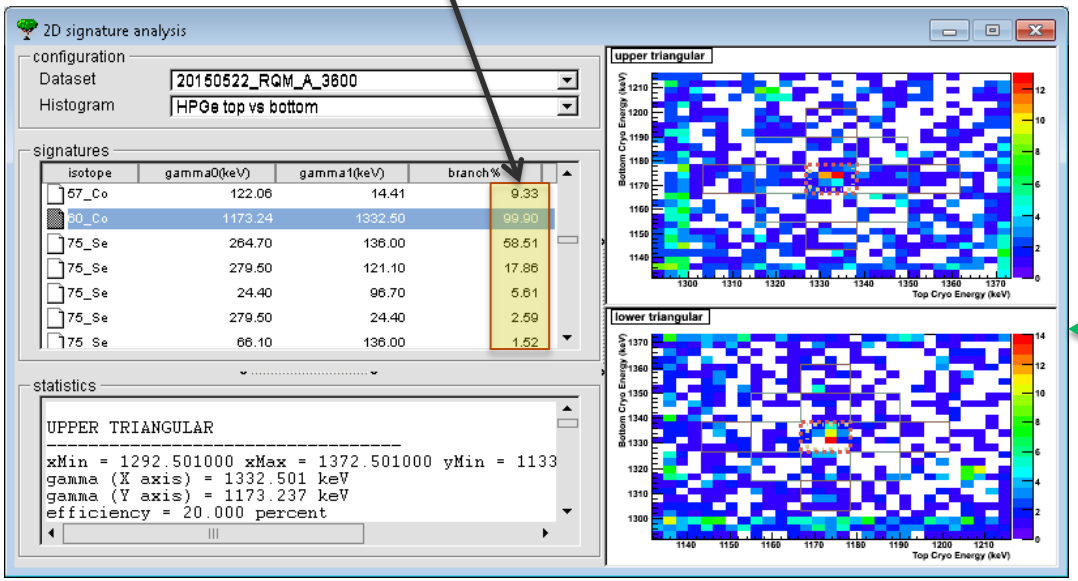
Nuclear data from the CLL drives

- Emission probabilities for coincidence signatures are required (along with detection efficiency) to convert measured signal to activity
- Coincidence summing corrections





Non-detect for ^{60}Co in singles mode

^{60}Co detected in coincidence

Coincidence Lookup Library

Smith et al. IEEE Transactions on Nuclear Science, V51, N3, June 2004

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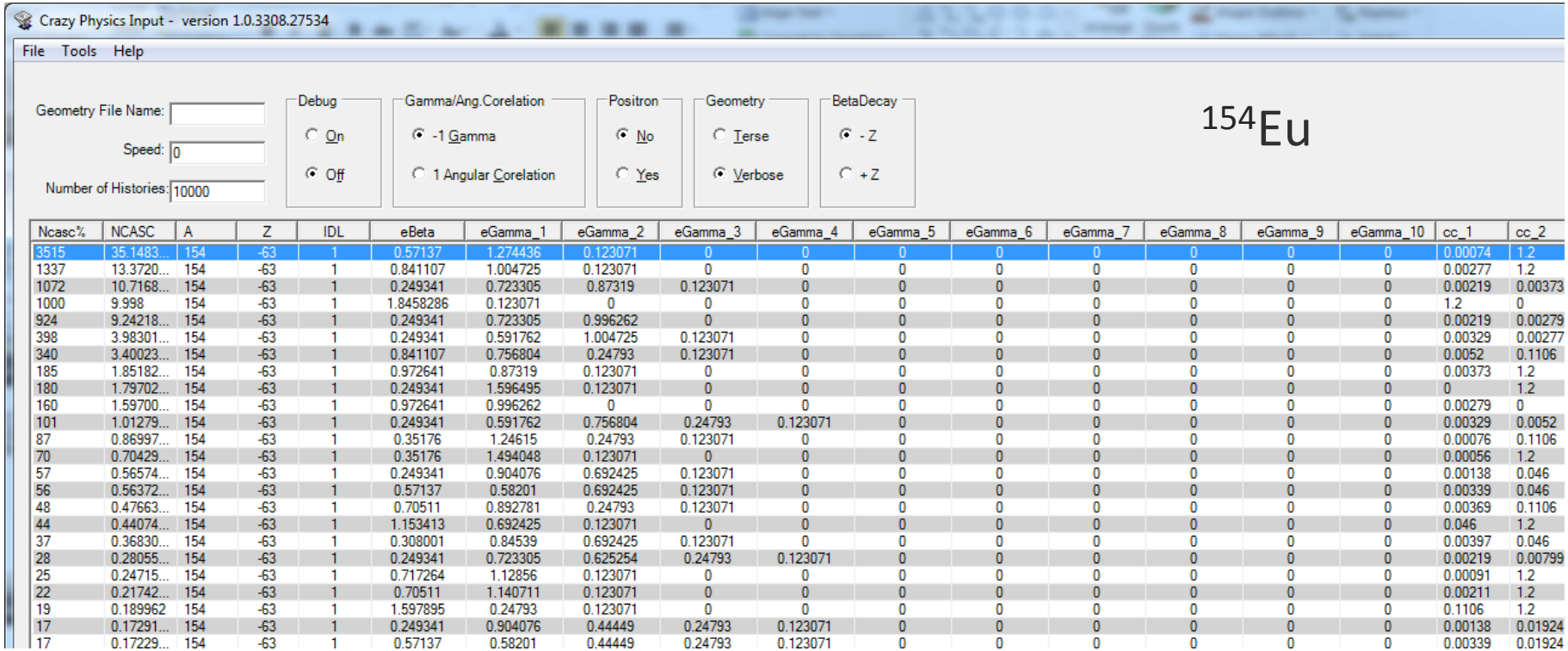
- The Coincidence Lookup Library (CLL) was developed at PNNL ~ 2004-2006
  - Calculates path probabilities based on published decay structures
  - Provides conversion coefficients for gammas (from published nuclear data tables)
  - Provides associated β-max (from published nuclear data tables)
  - Provides limited x-ray data



Ncasc%	NCASC	A	Z	IDL	eBeta	eGamma_1	eGamma_2	eGamma_3	eGamma_4	eGamma_5	eGamma_6	eGamma_7	eGamma_8	eGamma_9	eGamma_10	cc_1	cc_2
3515	35.1483...	154	-63	1	0.57137	1.274436	0.123071	0	0	0	0	0	0	0	0	0.00074	1.2
1337	13.3720...	154	-63	1	0.841107	1.004725	0.123071	0	0	0	0	0	0	0	0	0.00277	1.2
1072	10.7168...	154	-63	1	0.249341	0.723305	0.87319	0.123071	0	0	0	0	0	0	0	0.00219	0.00373
1000	9.998	154	-63	1	1.8458286	0.123071	0	0	0	0	0	0	0	0	0	1.2	0
924	9.24218...	154	-63	1	0.249341	0.723305	0.996262	0	0	0	0	0	0	0	0	0.00219	0.00279
398	3.98301...	154	-63	1	0.249341	0.591762	1.004725	0.123071	0	0	0	0	0	0	0	0.00329	0.00277
340	3.40023...	154	-63	1	0.841107	0.756804	0.24793	0.123071	0	0	0	0	0	0	0	0.0052	0.1106
185	1.85182...	154	-63	1	0.972641	0.87319	0.123071	0	0	0	0	0	0	0	0	0.00373	1.2
180	1.79702...	154	-63	1	0.249341	1.596495	0.123071	0	0	0	0	0	0	0	0	0	1.2
160	1.59700...	154	-63	1	0.972641	0.996262	0	0	0	0	0	0	0	0	0	0.00279	0
101	1.01279...	154	-63	1	0.249341	0.591762	0.756804	0.24793	0.123071	0	0	0	0	0	0	0.00329	0.0052
87	0.86997...	154	-63	1	0.35176	1.24615	0.24793	0.123071	0	0	0	0	0	0	0	0.00076	0.1106
70	0.70429...	154	-63	1	0.35176	1.494048	0.123071	0	0	0	0	0	0	0	0	0.00056	1.2
57	0.56574...	154	-63	1	0.249341	0.904076	0.692425	0.123071	0	0	0	0	0	0	0	0.00138	0.046
56	0.56372...	154	-63	1	0.57137	0.58201	0.692425	0.123071	0	0	0	0	0	0	0	0.00339	0.046
48	0.47663...	154	-63	1	0.70511	0.892781	0.24793	0.123071	0	0	0	0	0	0	0	0.00369	0.1106
44	0.44074...	154	-63	1	1.153413	0.692425	0.123071	0	0	0	0	0	0	0	0	0.046	1.2
37	0.36830...	154	-63	1	0.308001	0.84539	0.692425	0.123071	0	0	0	0	0	0	0	0.00397	0.046
28	0.28055...	154	-63	1	0.249341	0.723305	0.625254	0.24793	0.123071	0	0	0	0	0	0	0.00219	0.00799
25	0.24715...	154	-63	1	0.717264	1.12856	0.123071	0	0	0	0	0	0	0	0	0.00091	1.2
22	0.21742...	154	-63	1	0.70511	1.140711	0.123071	0	0	0	0	0	0	0	0	0.00211	1.2
19	0.189962	154	-63	1	1.597895	0.24793	0.123071	0	0	0	0	0	0	0	0	0.1106	1.2
17	0.17291...	154	-63	1	0.249341	0.904076	0.44449	0.24793	0.123071	0	0	0	0	0	0	0.00138	0.01924
17	0.17229...	154	-63	1	0.57137	0.58201	0.44449	0.24793	0.123071	0	0	0	0	0	0	0.00339	0.01924

Nuclear Data

- The CLL uses static nuclear data, e.g. Table of Isotopes, 8th edition (1999)
- The CLL does not provide uncertainty in the signature intensity, or in the conversion coefficient
- For some isotopes, the CLL does not successfully parse the nuclear data
  - A number of isotopes are not successfully calculated
  - e.g. ^{55}Co, ^{56}Co, ….
- We are not aware of published data tables that provide the emission probability for coincident $\gamma-\gamma$, γ-x, and $\beta-\gamma-\gamma$ signatures
- Monte-Carlo decay simulators are available which also utilize available nuclear data to provide particle generator capabilities
  - SCH2FOR (Laedermann and Decombaz, Appl Radiat Isot, V52, N3, Mar 2000)
  - G4RadioactiveDecay (Agostinelli *et al*, NIMA 506, 2003)
- Nuclear Data Gaps in the CLL (and elsewhere?)
  - Angular correlation data
  - Conversion coefficients often calculated (often not measured)
  - Missing data

Coincidence Data in NuDat Decay Radiation Search

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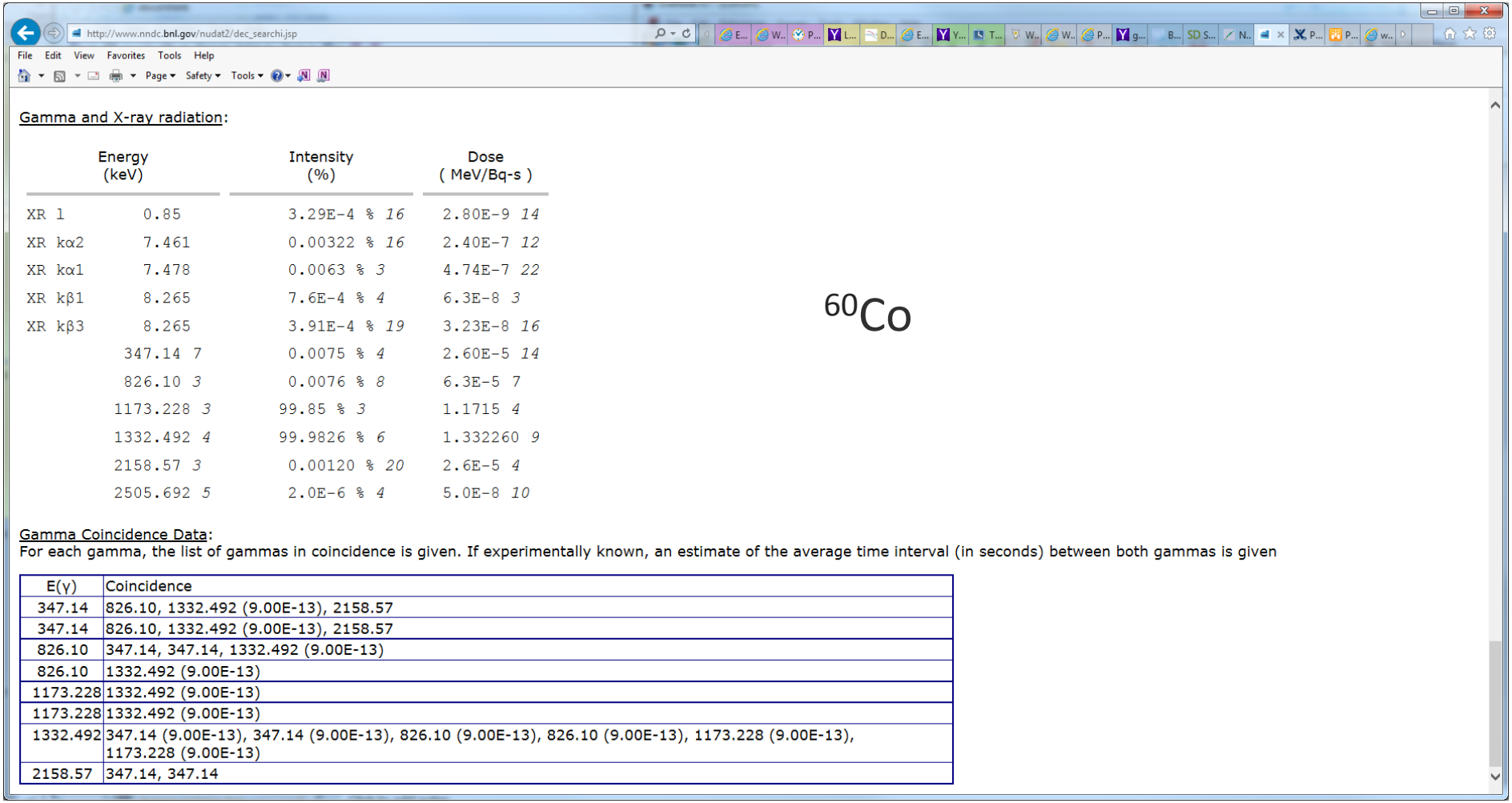

http://www.nndc.bnl.gov/nudat2/dec_searchi.jsp

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Gamma and X-ray radiation:

	Energy (keV)	Intensity (%)	Dose (MeV/Bq-s)
XR l	0.85	3.29E-4 % *16*	2.80E-9 *14*
XR kα2	7.461	0.00322 % *16*	2.40E-7 *12*
XR kα1	7.478	0.0063 % *3*	4.74E-7 *22*
XR kβ1	8.265	7.6E-4 % *4*	6.3E-8 *3*
XR kβ3	8.265	3.91E-4 % *19*	3.23E-8 *16*
	347.14 *7*	0.0075 % *4*	2.60E-5 *14*
	826.10 *3*	0.0076 % *8*	6.3E-5 *7*
	1173.228 *3*	99.85 % *3*	1.1715 *4*
	1332.492 *4*	99.9826 % *6*	1.332260 *9*
	2158.57 *3*	0.00120 % *20*	2.6E-5 *4*
	2505.692 *5*	2.0E-6 % *4*	5.0E-8 *10*

^{60}Co

Gamma Coincidence Data:
For each gamma, the list of gammas in coincidence is given. If experimentally known, an estimate of the average time interval (in seconds) between both gammas is given

E(γ)	Coincidence
347.14	826.10, 1332.492 (9.00E-13), 2158.57
347.14	826.10, 1332.492 (9.00E-13), 2158.57
826.10	347.14, 347.14, 1332.492 (9.00E-13)
826.10	1332.492 (9.00E-13)
1173.228	1332.492 (9.00E-13)
1173.228	1332.492 (9.00E-13)
1332.492	347.14 (9.00E-13), 347.14 (9.00E-13), 826.10 (9.00E-13), 826.10 (9.00E-13), 1173.228 (9.00E-13), 1173.228 (9.00E-13)
2158.57	347.14, 347.14

http://www.nndc.bnl.gov/nudat2/dec_searchi.jsp

Summary and Path Forward

- Laboratory use of multi-crystal γ–γ coincidence systems is increasing
  - CASCADES, Dual Clover, etc
  - Analysis capabilities are typically system-specific, do not use consistent algorithms and nuclear data
- Future validated γ–γ methods will need
  - Consistent nuclear data
  - Validated algorithms
  - Interlaboratory comparisons
- Near-term focus should be on nuclear data and problem-specific algorithms
  - PNNL is pursuing a collaboration with Dave Brown (BNL) to develop new γ–γ coincidence analysis algorithms and associated nuclear data tables