Developments in capture-γ libraries for nonprolieration applications

Aaron M. Hurst
amhurst@berkeley.edu
(c/o Richard B. Firestone)

Department of Nuclear Engineering
University of California, Berkeley

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Overview

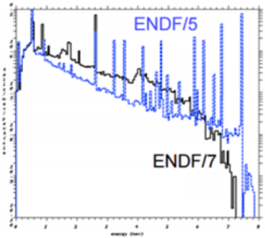
- Principal aim: Augment Evaluated Nuclear Data File (ENDF) libraries with high-resolution HPGe-quality γ-ray line data.
- Improved capture-γ data needed for several high-priority isotopes for accurate simulation of interrogation systems.

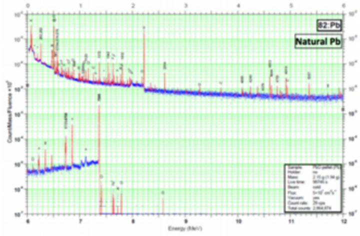
- Interrogation system: Evaluating munitions for presence of explosives, chemicals or nerve agents.
- Idaho National Laboratory - 80 units in use worldwide
- SF, DD, DT neutron sources (Gus Caffrey, Ed Seabury, INL)

Background and problem

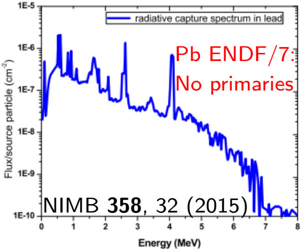
- Neutron-capture γ rays provide unambiguous isotopic fingerprint.
- Useful for NDA applications: passive interrogation (spontaneous-fission neutrons) and active interrogation (external neutron source).
- Bright high-energy *"primary"* γ rays ($E_\gamma \approx 4 - 12$ MeV) $\Rightarrow$ easily seen in spectra and clearly indicate presence of Special Nuclear Material (SNM, i.e. ^{235,238}U and ^{239}Pu), fission products and an array of materials associated with SNM.
- **NDA applications are predicated on accurate data!**
- Problem: There are well-known data gaps in capture-γ ENDF libraries; some isotopes (e.g., actinides) have no primaries at all.

Issues with Pb capture-γ data in ENDF

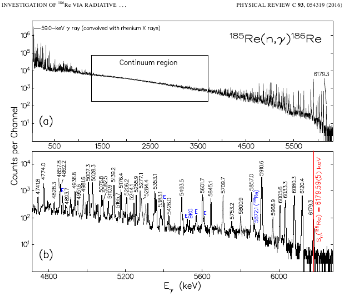


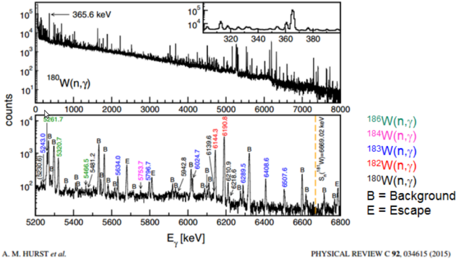


- "Real" (top; RHS) capture-γ spectrum for Pb reveals inconsistencies with MCNP simulation sourcing ENDF/7 (top; LHS).
- Migration from ENDF/5 to ENDF/7 capture-γ data becomes worse.
- *Published literature describing Pb simulations with ENDF/7!*



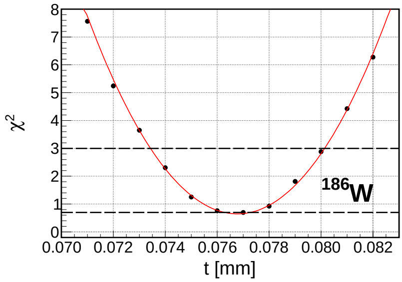
High-E_γ issues: lack of primary-γ data

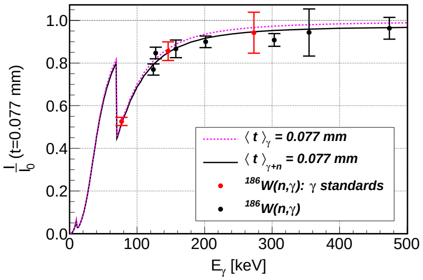




- Generally we can understand completely high-E_γ primary region $\Rightarrow$ very useful signatures for NDA.
- Often these data are missing from the ENDF libraries (e.g., actinides).
- Recent work has identified primary γ rays for the first time in ^{180}W(n,γ) and 50 new primaries have been measured in ^{185}Re(n,γ).

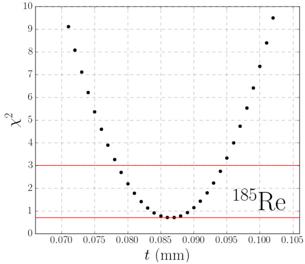
Low-E_γ issues: self-absorption capture-γ corrections I

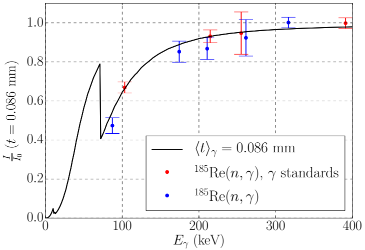




- High-ρ samples (e.g., W and Re) may require thickness correction to establish σ_γ
- Low-energy γ rays particularly susceptible to self absorption
- Adjust thickness until convergence is achieved for **all** γ lines: $\frac{\sigma_{\gamma_i}(E_\gamma)}{A_{\gamma_i}(E_\gamma)}$
- χ^2 method in Ref. [A. M. Hurst *et al.*, NIMB **362**, 38-44 (2015)]

Low-E_γ issues: self-absorption capture-γ corrections II

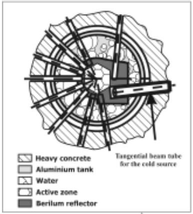


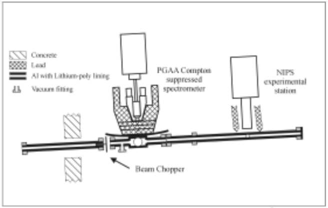


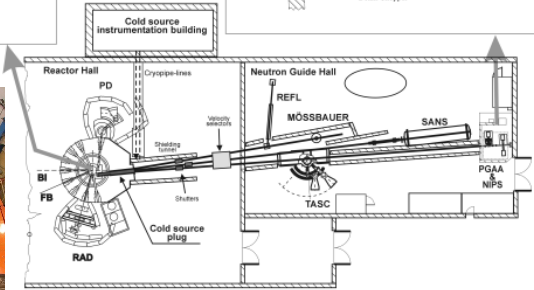
- "Ideal" σ_γ(Re) extracted relative to well-known standard σ_γ(Cl) from low-ρ $ReCl_3$ (hydrated) sample.
- Convergence achieved by comparison with lines in enriched ^{185}Re (pure) sample.

Analysis by D. A. Matters, AFIT [PRC 93, 054319 (2016)]

Experimental setup (1): Budapest Research Reactor



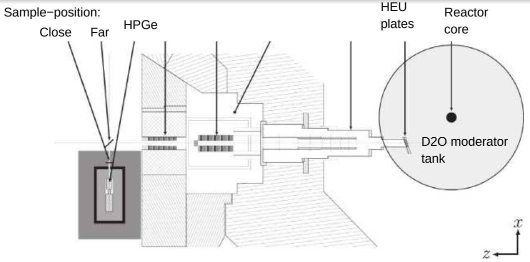




Thermal: $\sim 10^6/\mathbf{cm}^2/\mathbf{s}$

Cold: $\sim 10^8/\mathbf{cm}^2/\mathbf{s}$

Experimental setup (2): FRM-II Reactor Munich



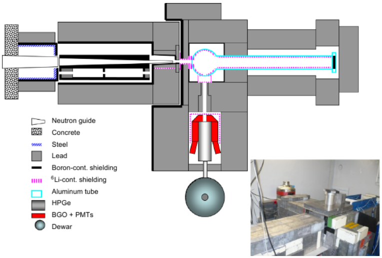
Fission neutrons:

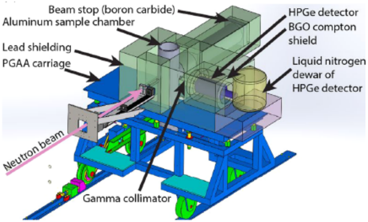
$\sim 1 \times 10^8/\text{cm}^2/\text{s}$

$\langle E_n \rangle \approx 2.1$ **MeV**

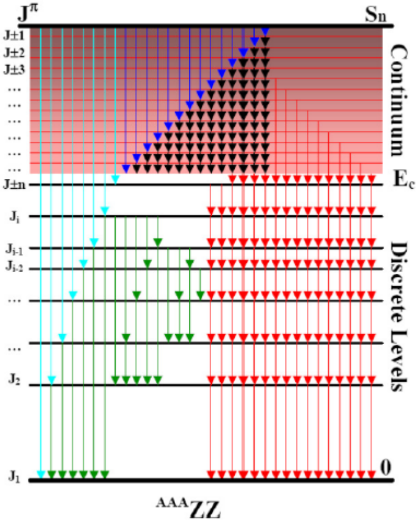
Thermal-equivalent:

$\sim 2.7 \times 10^{10}/\text{cm}^2/\text{s}$





Statistical modeling with DICEBOX



- DICEBOX: Monte Carlo code
- Generates (n,γ) decay scheme simulations originating at S_n
- All levels and γ rays below E_c are taken from experiment
- All levels and γ rays above E_c are randomly generated
- Primary γ rays from experiment when known
- Adopted photon strength function (PSF) and level density (LD) models to calculate partial transition widths and compare to experimental data:

$$\langle \Gamma_{if} \rangle = \frac{f(E_\gamma) \cdot E_\gamma^{2L+1}}{\rho(E_i, J_i, \pi_i)}$$

ENDF processing

- Validated E_γ and σ_γ data are processed for incorporation into ENDF and correlations with other parts of library verified.
- Integrity of library is checked against LANL and LLNL verification codes (PREPRO + NJOY + FUDGE).
- Upon successfully testing out and generating representative transport-simulation output (MCNP) $\Rightarrow$ libraries are sent to National Nuclear Data Center (NNDC) for further testing prior to dissemination in the next ENDF/B-VIII release.

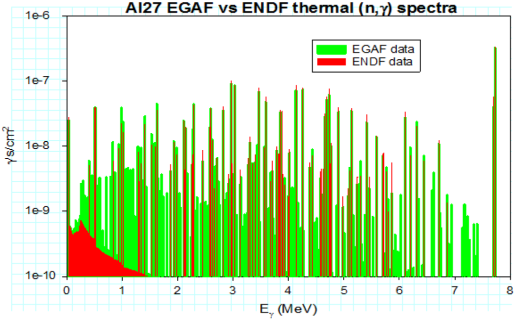
New capture-γ data in ENDF

ENDF/B-VIII.β2 libraries updated with new evaluated capture-γ lines

Z	**Element**	A	**Abundance [%]**	**No. (n,γ) lines**
3	Li	6	7.589	3
3	Li	7	92.411	3
5	B	11	80.18	13
9	F	19	100	166
11	Na	23	100	244
13	Al	27	100	290
14	Si	28	92.2297	56
17	Cl	35	75.771	382
17	Cl	37	24.229	76

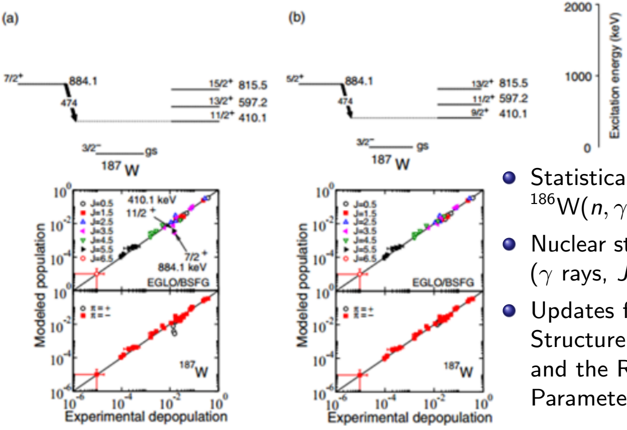
A comparison: Before and after

New $\sigma_\gamma(E_\gamma)$ data (green) reveal marked improvement in quality and quantity over previous capture-γ data in ENDF (red) for ^{27}Al(n,γ)



High-resolution data needed for accurate simulations of interrogation systems

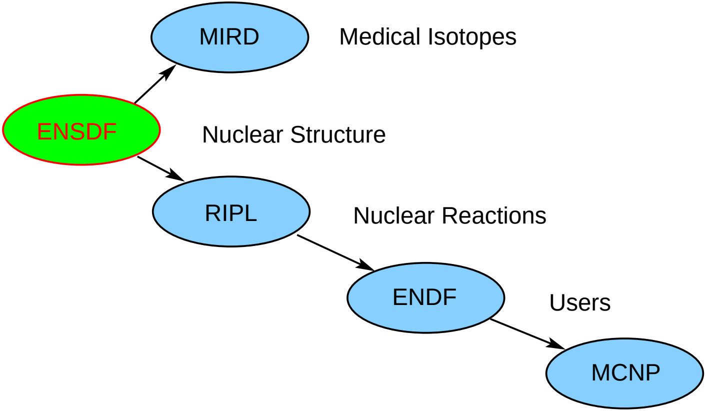
Structure data improvements



A. M. HURST *et al.* PHYSICAL REVIEW C **89**, 014606 (2014)

- Statistical-model analysis ^{186}W(n,γ)
- Nuclear structure improvements (γ rays, J^{π})
- Updates for Evaluated Nuclear Structure Data File (ENSDF) and the Reference Input Parameter Library (RIPL)

Nuclear data library development



Summary and outlook

- New improved neutron-capture γ-ray line data were incorporated into and used to upgrade 9 ENDF libraries: ^{6}Li, ^{7}Li, ^{11}B, ^{19}F, ^{23}Na, ^{27}Al, ^{28}Si, ^{35}Cl, and ^{37}Cl.
- All libraries successfully passed testing and generated representative output for MCNP transport applications.
- Now available in ENDF/B-VIII.β2 via the NNDC at Brookhaven National Laboratory.
- Many more isotopes on *"priority list"* need improved capture-γ data.
- Widespread problems and data gaps (including missing primaries) augmented with new evaluated capture-γ data will provide enhanced threat-detection signatures for interrogation systems.
- **More than 10** peer-reviewed publications (Physical Review, Nuclear Science and Engineering, Nuclear Data Sheets, Nuclear Instruments and Methods) in the **past 2 years** alone.
- Thesis fodder for graduate students (so far: 4 PhDs, 1 MS).

Acknowledgments

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- **Charles University Prague** – F. Bečvář, M. Krtička
- **Budapest Research Reactor** – T. Belgya, L. Szentmiklósi
- **FRM-II Munich** – Zs. Révay, **C. Genreith**
- **AFIT** – **D. A. Matters**, **A. G. Lerch**[*], J. W. McClory, S. R. McHale
- **US ARL** – J. J. Carroll
- **OSU** – **D. Turkoglu**

[*]DTRA
[&]Walmart

Student training

Student	Degree	Project	Institute	Current Position
A. Ureche	Ph. D.	$^{139}La(n,\gamma)$	UC Berkeley [NSSC]	Year 2
D. A. Matters	Ph. D.	$^{185,187}Re(n,\gamma)$	Air Force Institute of Technology	Final Year
A. G. Lerch	M.S. 02/2014	$^{185}Re(n,\gamma)$	Air Force Institute of Technology	Group Leader, DTRA
C. Genreith	Ph. D. 10/2014	$^{242}Pu(n,\gamma)$	Jülich	Postdoc, FRM-II
D. Turkoglu	Ph. D. 11/2014	$^{93}Nb(n,\gamma)$	Ohio State University	Postdoc, NIST

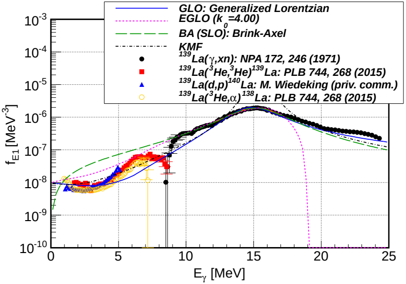
^{140}La: an abundant fission product

IAEA cumulative fission yields [% per fission]

Fission Nucleus	Fission/(n,γ) Product	Thermal Yield [%]	Fast Yield [%]	14-MeV Yield [%]
^{232}Th	^{140}**La**	-	7.7	5.7
^{233}U	^{140}**La**	6.5	6.2	4.7
^{235}U	^{140}**La**	6.3	6	4.5
^{238}U	^{140}**La**	-	6.0	4.6
^{239}Pu	^{140}**La**	5.3	5.3	-
^{241}Pu	^{140}**La**	5.8	5.4	-

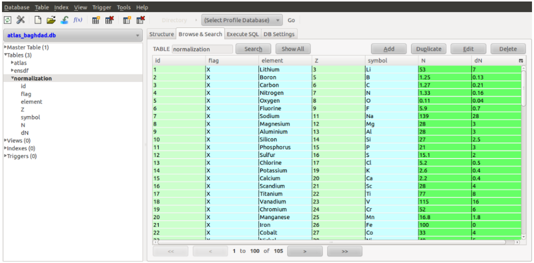
"*$A \approx 143$ Isotopes: New data for high-yield fission fragments are needed for accurate prediction of inventory in used nuclear fuel assemblies, development of better physics models for calculation-based nuclear forensic tools ...*", White paper on NDNCA: http://bang.berkeley.edu

Low-energy PSF in ^{140}La



- Provide students with an interesting thesis topic.
- Nature of photon strength function (PSF) at low energy is a hotly-contested issue.
- PSF has fundamental application in statistical model for γ decay provides structural insights
- Reproduce known data $\Rightarrow$ validate regions without data $\Rightarrow$ **improve capture-γ library!**

Baghdad $(n, n'\gamma)$ Atlas: SQL relational database



id	flag	element	Z	symbol	N	dN
1	X	Lithium	3	Li	53	7
2	X	Boron	5	B	1.25	0.13
3	X	Carbon	6	C	1.27	0.21
4	X	Nitrogen	7	N	1.33	0.16
5	X	Oxygen	8	O	0.11	0.04
6	X	Fluorine	9	F	5.9	0.7
7	X	Sodium	11	Na	139	28
8	X	Magnesium	12	Mg	28	3
9	X	Aluminium	13	Al	28	3
10	X	Silicon	14	Si	27	2.5
11	X	Phosphorus	15	P	21	3
12	X	Sulfur	16	S	15.1	2
13	X	Chlorine	17	Cl	5.2	0.5
14	X	Potassium	19	K	2.6	0.4
15	X	Calcium	20	Ca	2.2	0.4
16	X	Scandium	21	Sc	28	4
17	X	Titanium	22	Ti	77	8
18	X	Vanadium	23	V	115	16
19	X	Chromium	24	Cr	52	6
20	X	Manganese	25	Mn	16.8	1.8
21	X	Iron	26	Fe	100	0
22	X	Cobalt	27	Co	33	4

target	sample	compound	E [keV]	dE [keV]	BR	dBR	cross section [mb]	error cs [mb]
Pd	E	110Pd	356.9	0.2	0.88	0.04	0.944019648	0.172460592992611
Pd	E	110Pd	373.8	0.08	100.0	0.0	107.27496	18.9814874907528
Pd	E	110Pd	398.8	0.2	5.2	0.5	5.57829792	1.12336132006618
Pd	E	110Pd	401.0	0.7	0.7	0.3	0.75092472	0.348174955859454
Pd	E	110Pd	439.76	0.08	23.6	0.3	25.31689056	4.49117641358686
Pd	E	110Pd	463.9	0.4	0.18	0.02	0.193094928	0.040344498162347
Pd	E	110Pd	477.5	0.3	1.02	0.15	1.094204592	0.251750073340714
Pd	E	110Pd	547.04	0.1	9.2	0.5	9.86929632	1.82681433414117
Pd	E	110Pd	572.89	0.1	5.4	0.3	5.79284784	1.07433557076616
Pd	E	110Pd	584.48	0.1	1.65	0.1	1.77003684	0.331057002919737
Pd	E	110Pd	641.0	1.1	0.04	0.015	0.042909984	0.017792572389775
Pd	E	110Pd	648.51	0.16	0.51	0.04	0.547102296	0.105889509617127
Pd	E	110Pd	653.1	0.2	0.52	0.05	0.557829792	0.112336132006618
Pd	E	110Pd	656.42	0.15	0.93	0.06	0.997657128	0.18789605156465
Pd	E	110Pd	672.4	1.1	0.039	0.015	0.0418372344	0.017712404890167
Pd	E	110Pd	687.7	0.3	0.16	0.02	0.171639936	0.037184360451213
Pd	E	110Pd	722.5	0.4	0.11	0.015	0.118002456	0.026360715904069
Pd	E	110Pd	729.9	1.0	0.07	0.02	0.075092472	0.025236127807713
Pd	E	110Pd	762.2	0.4	0.13	0.02	0.139457448	0.032698904990246
Pd	E	110Pd	770.3	0.2	0.61	0.05	0.654377256	0.127607310508747
Pd	E	110Pd	773.0	0.8	0.11	0.03	0.118002456	0.038362374058153
Pd	E	110Pd	796.83	0.1	1.84	0.12	1.973859264	0.372227763547894
Pd	E	110Pd	813.52	0.1	4.2	0.3	4.50554832	0.859729566441032
Pd	E	110Pd	838.5	0.3	3.0	0.5	3.2182488	0.782281986690531
Pd	E	110Pd	840.9	0.7	1.6	0.4	1.71639936	0.525702074118599
Pd	E	110Pd	905.2	0.2	0.82	0.06	0.879654672	0.168431622615454
Pd	E	110Pd	941.5	1.2	0.031	0.014	0.0332552376	0.016130086887437
Pd	E	110Pd	978.8	0.5	0.078	0.016	0.0836744688	0.022667317671123

- A. Demidov *et al.*, *Atlas of Gamma-Ray Spectra from Inelastic Scattering of Reactor Fast Neutrons*, (Moscow, Atomizdat 1978).
- 105 data sets: 76 natural; 29 enriched
- Over 7000 $(n, n'\gamma)$ lines
- Visualization of data using relational tables
- Use SQLite GUI to manipulate data or execute SQL code
- Absolute γ-ray cross sections normalized to $2_1^+ \rightarrow 0_{gs}^+$ in ^{56}Fe (847 keV, 468 mb)
- *Query: retrieve σ_γ for enriched $^{110}Pd(n, n'\gamma)$ where $300 \leq E_\gamma \leq 1000$ keV*