Cross Section Measurement and Evaluation for Nuclear Applications

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Historical Background

The US Nuclear Data Program (USNDP) was founded 70 years ago by an act of Congress to provide evaluated data for nuclear energy.

Today the USNDP is divided into two nuclear data evaluation efforts

- **ENDF** Neutron cross section database for nuclear engineering calculations.
- **ENSDF** Nuclear structure database primarily used in basic nuclear physics research.

Both efforts have been highly successful but are facing significant challenges to remain relevant into the future.

The two efforts have drifted apart and lost contact with the nuclear physics and applications communities. The primary goal of this ANS&T project seeks to develop an new direction for the LBNL nuclear data effort.



Goals of this ANS&T Project

Primary Goals

- Measure thermal neutron cross sections with guided neutron beams at the Budapest and Munich Reactors.
- Measure surrogate cross sections with STARS/LiBeRACE at the LBNL 88" cyclotron.
- Study the physics of statistical level densities and γ -ray strengths.

Secondary Goals

- Bridge the gap between the reaction and structure data communities.
- Develop a broad collaborations with the greater international nuclear physics and application communities.
- Establish the LBNL Isotopes Project as the lead applied experimental physics group at the 88" cyclotron.

Challenges Facing Nuclear Data

1. Loss of expertise

- a. Retirement of key personnel
- b. Lack of interest/investment by foreign collaborators

2. Outdated methods

- a. "Card image" data formats
- b. Fortran 1 software

3. Disconnect from current research

- a. Insufficient participation of evaluators in basic research
- b. Lack of quality research directed towards nuclear applications
- c. Concentration on local institutional concerns of the moment
- d. Poor communication between the ENDF and ENSDF evaluators
- e. Lack of experimental facilities for nuclear data in the US

4. Failure to address emerging research interests

- a. Nuclear astrophysics
- b. Homeland security
- c. Energy and environment

Disappearing ENSDF Evaluators

Active ENSDF evaluators are near or beyond retirement (red) or temporary commitments (green).

- No evaluators from Western Europe
- Weakening commitment from Asia
- Excessive reliance on one person

In less than 5 years it may be impossible to evaluate ENSDF

Latest Mass Chain Evaluation Pipeline

A Evaluator(s)

- 27 Basunia
- 31 Ouellet, Singh
- 32 Ouellet, Singh
- 34 Nica, Singh
- 35 Chen, Cameron, Singh
- 36 Nica, Cameron, Singh
- 37 Cameron, Chen, Singh, Nica
- 44 Chen, Singh, Cameron
- 61 Zuber, Singh
- 75 Negret, Singh
- 77 Nica, Singh
- 85 Singh

A Evaluator(s)

- 89 Singh
- 92 Baglin
- 114 Blachot
- 129 Timar, Elekes, Singh
- 142 Johnson, Symochko, Fadil, Tuli
- 150 Basu, Sonzogni
- 152 Martin
- 159 Reich
- 161 Reich
- 192 Baglin
- 222 Singh, Jain, Tuli
- 249 Abusaleem

The EGAF Opportunity

- In the 1990's a guided neutron beam was constructed at the Budapest Reactor. Precise γ -ray cross sections (σ_{γ}) were measured on all stable elemental targets. This led to the
 - First σ_{γ} database this data didn't exist in ENDF or ENSDF
 - Evaluated Gamma-ray Activation File (EGAF) IAEA CRP http://www-pub.iaea.org/MTCD/publications/PubDetails.asp?publd=7030
 - Measurement of total thermal neutron cross sections (σ_0)
 - Development of the Reaction Input Parameter Library (RIPL)
 - New applications to chemistry, geology, energy, environment
 - New theoretical methods for ENSDF evaluation
 - New γ -ray and cross section data for ENDF
 - International collaboration to study statistical model photon strengths and level densities
 - Interest in the surrogate reaction method
 - Applications of Nuclear Science and Technology proposal

Budapest Reactor Measurement of Prompt γ-rays with Neutron Beams



Reactor and neutron guide hall. The PGAA (capture gamma) station located ≈ 30 m from the reactor wall.

Thermal neutron cross γ -ray sections measured with Compton suppressed HPGe detector for all elemental targets.

- Low background
- Internal calibration
- Z=1,83,90,92 except for He, Pm.



Thermal (n,γ) Cross Sections – Light Isotopes

Target	σ_0 (Budap.)	σ ₀ (Atlas*)	Target	σ_0 (Budap.)	σ₀(Atlas*)
⁶ Li	52.6(22) mb	44.8(3) mb	²⁶ Mg	38.8(14) mb	38.4(6) mb
⁷ Li	46.3(13) mb	45.2(14) mb	²⁷ AI	232.2(17) mb	231(3) mb
⁹ Be	8.8(6) mb	8.5(3) mb	²⁸ Si	186(2) mb	171(3) mb
¹⁰ B	3.90(11) mb	3.05(16) mb	²⁹ Si	128(4) mb	119(3) mb
¹¹ B	9.06(20) mb	5.5(33) mb	³⁰ Si	112(6) mb	107(2) mb
¹² C	3.86(6) mb	3.53(7) mb	³¹ P	169(5) mb	165(3) mb
¹³ C	1.51(3) mb	1.37(4) mb	³² S	542(7) mb	518(14) mb
¹⁴ N	78.5(7) mb	80.1(6) mb	³³ S	449(7) mb	454(20) mb
¹⁵ N	39(3) μb	24(8) μb	³⁴ S	285(8) mb	256(9) mb
¹⁶ O	197(7) μb	190(20) μb	³⁵ Cl	44.00(20) b	43.6(4) b
¹⁹ F	9.36(12) mb	9.51(9) mb	³⁷ Cl	50.0(8) mb	43.3(6) mb
²³ Na	541(3) mb	517(4) mb	³⁹ K	2.28(4) b	2.1(2) b
²⁴ Mg	535(20) mb	538(13) mb	⁴⁰ K	86(7) b	30(8) b
²⁵ Mg	196(8) mb	199(3) mb	⁴¹ K	1.62(3) b	1.46(3) b

More than 50% of cross sections disagree within their uncertainties

Thermal (n,γ) Cross Sections– Heavy Isotopes



DICEBOX* Statistical Model Calculations

When the (n,γ) continuum feeding is important it can be calculated if

E_c 1. $σ_{\gamma}$ deexciting levels below a cutoff energy E_{crit} is complete. \downarrow

2. Primary σ_{γ} populating the levels below E_{crit} from the capture state is complete.

3. J^{π} of levels below E_{crit} is well known.

4. Level density $\rho(E > E_{crit}, J)$ is known.

5.Photon strength $f(E_{\gamma})$ deexciting or populating levels above E_{crit} is known. $\downarrow \downarrow \downarrow \downarrow$

* M. Krticka, F. Becvar, Charles University, Prague

Statistical Model Ingredients

- A. Structure data from the Reaction Input Parameter Library (RIPL)
- Level density models Β.

Constant temperature (1937)

 $\rho(E,J) = \frac{f(J)}{T} \exp\left(\frac{E - E_0}{T}\right)$

- Back-shifted Fermi Gas (1937) $\rho(E,J) = f(J) \frac{\exp\left(2\sqrt{a(E-E_1)}\right)}{12\sqrt{2}\sigma_1 a^{1/4} (E-E_1)^{5/4}}$
- C. The spin distribution of the level density $f(J)(1937) = \frac{2J+1}{2\sigma^2} \exp\left[-\frac{(J+1/2)^2}{2\sigma^2}\right]$
- D. Photon Strength

spin flip models, 1958

E1 (Brink-Axel, 1955) $f_{BA}^{(E1)}(E_{\gamma}) = \frac{1}{3(\pi\hbar c)^2} \frac{\sigma_G E_{\gamma} \Gamma_G^2}{(E_{\gamma}^2 - E_G^2)^2 + E_{\gamma}^2 \Gamma_G^2}$ <u>M1 – Single particle,</u> spin flip models 1958 $\frac{f^{(E1)}}{f^{(M1)}} = 5-7 \text{ or } f(M1) = 1.2 \times 10^{-8} MeV^{-3}$

Laurentzian resonance ~8.5 MeV, Γ_{SF} ~4 MeV

E. Problems

- No consideration of parity
- No consideration of nuclear structure
- Little progress for over 50 years

Reference Input Parameter Library (RIPL-3)



International Atomic Energy Agency Nuclear Data Services Provided by the Nuclear Data Section

R. Capote, M. Herman, P. Oblozinsky, P.G. Young, S. Goriely, T. Belgya, A.V. Ignatyuk, A.J. Koning, S. Hilaire, V.A. Plujko, M. Avrigeanu, O. Bersillon, M.B. Chadwick, T. Fukahori, Zhigang Ge, Yinlu Han, S. Kailas, J. Kopecky, V.M. Maslov, G. Reffo, M. Sin, E.Sh. Soukhovitskii and P. Talou *Nuclear Data Sheets - Volume 110, Issue 12, December 2009, Pages 3107-3214*

Nuclear structure data from ENSDF.

Initial nuclear structure data for RIPL provided to T. Belgya by LBNL Isotope Project.

No one has formal responsibility for evaluating the RIPL library.

RIPL is the primary source of nuclear structure data for statistical model calculations with **DICEBOX**, **EMPIRE**, and other programs.

- Incomplete ENSDF level schemes
- Unclear ENSDF spin/parity assignments
- ENSDF was not evaluated for this purpose
- Lack of coordination between the reaction and structure communities

A goal of this ANS&T project is to improve the RIPL database

New Physics Resulting from this ANS&T Proposal



Analysis of primary γ -ray reduced transition strengths indicates strong nuclear structure dependence for M1 and E1 transitions.

¹⁶⁸Er K-Dependence of the Statistical Model



Analysis of population/depopulations ratios in ¹⁶⁸Er indicate K-dependence and problems with spin distribution

New Physics Resulting from this ANS&T Proposal



DICEBOX Population/Depopulation Plot*



DICEBOX can determine the admixture of spins for the capture state Discrepancies from the straight line indicate problems with the nuclear structure data.

* M. Krticka, R.B. Firestone, D.P. McNabb, B. Sleaford, U. Agvaanluvsan, T. Belgya, and Z.S. Revay, Phys. Rev. C 77, 054615 (2008).

J^{π} Assignments using DICEBOX



Palladium Isotopes Results*

Isotope	σ_0 (literature) (b)	σ_0 (this work) (b)
¹⁰² Pd(n,γ) ¹⁰³ Pd	1.6±0.2	1.1±0.4
¹⁰⁴ Pd(n,γ) ¹⁰⁵ Pd	0.65±0.30	0.75±0.26
¹⁰⁵ Pd(n,γ) ¹⁰⁶ Pd	21.0±1.5	21.7±0.5
¹⁰⁶ Pd(n,γ) ¹⁰⁷ Pd	0.30±0.03	0.36±0.10
¹⁰⁸ Pd(n,γ) ¹⁰⁹ Pd ^g	7.6±0.5	8.6±0.6
¹⁰⁸ Pd(n,γ) ¹⁰⁹ Pd ^m	0.185±0.011	0.185±0.010
¹¹⁰ Pd(n,γ) ¹¹¹ Pd	0.70±0.17	0.34±0.10

* M. Krticka, R.B. Firestone, D.P. McNabb, B. Sleaford, U. Agvaanluvsan, T. Belgya, and Z.S. Revay, Phys. Rev. C 77, 054615 (2008).

Surrogate Reaction Cross Sections

First results for this ANS&T project



Spin Determination with STARS/LiBeRACE



Analysis courtesy of Mathis Wiedeking, iThemba LABS, South Africa

Improved Level Schemes for DICEBOX with STARS/LiBeRACE



28 new states, 99 new transitions, 2 states confirmed, 4 transitions confirmed, 3 spins confirmed/corrected, 2 low energy decay properties significantly different.

Deliverables of the ANS&T Proposal

Funding:

- FY2011 \$380K
- FY2012 \$350K
- Additional effort is supported by Isotopes Project baseline and ARRA funding

Participants:

- R.B. Firestone (PI), Aaron Hurst (staff)*, Shamsu Basunia (staff)[†], Postdoc (TBD)
- Guests: H. Choi (Seoul University), Zs. Revay (Budapest), T. Belgya (Budapest)
- L. Bernstein (LLNL), M. Krticka (Charles U., Prague), K. Abusaleem (U. Jordan)

Deliverables:

- Thermal neutron cross sections Z<20, Fe, ⁷⁴Ge, Y, Eu, Gd, W (expt. Complete)
- Surrogate reaction cross sections for ²³⁷Np and ²³⁸Pu (published)
- ¹⁶⁷Er(n,g),(d,p) reactions, statistical model study (expt. completed)
- Analyze LBNL Total Absorption Spectrometer (TAS) data (paper presented)
- Publication format for EGAF in ENSDF presented and accepted by USNDP
- Develop collaboration between the research and data communities (done)
- Publish experimental results in peer reviewed journals (in progress).

* Newly hired through the ANS&T grant, † Position preserved though ARRA

Collaborators

LBNL Isotopes Project - R.B. Firestone, A. Hurst, S. Basunia, E. Browne LLNL Nuclear Data Library – B. Sleaford, N. Summers LLNL STARS/LiBeRACE – J. Ressler, J. Burke, N. Scielzo, I. Thomson, J. Escher, R. Casperson, R. Austin, C. Mattoon, N. Patel LLNL NIF - L. Bernstein, D. Bleuel, J.A. Caggiano, D. Schneider, W. Stoeffl **University of Richmond** – C. Beausang **Youngstown State U. (Ohio)** – J. Carroll Budapest/Munich Reactors – Zs. Revay, T. Belgya, L. Szentmiklosi Charles University, Prague - M. Krticka, F. Becvar Seuol University, Korea – H.D. Choi **University of Jordan** – K. Abusaleem **IAEA Nuclear Data Section** –D. Abriola, R. Capote, M. Kellett, V. Zerkin **iThemba Labs, South Africa** - M. Wiedeking University Oslo - S. Siem, A. Goergen, M. Guttormsen, A.C. Larsen More people are welcome to collaborate.

Publications and Papers

- 1. J. M. Allmond, C. W. Beausang, J. O. Rasmussen, T. J. Ross, M. S. Basunia, et al, *Particle-* γ spectroscopy of the (p, d- γ)155Gd reaction: Neutron single-quasiparticle states at N = 91; Physical Review C, 81, 064316, 2010.
- D. Scielzo, J. E. Escher, J. M. Allmond, M. S. Basunia, et al, *Measurement* of *γ*-emission branching ratios for 154,156,158Gd compound nuclei: Tests of surrogate nuclear reaction approximations for (n,γ) cross sections; Physical Review C 81, 034608, 2010.
- Surrogate measurement of the 238Pu(n, f) cross section; J. J. Ressler, J. T. Burke, J. E. Escher, C. T. Angell, M. S. Basunia, et al, *Physical Review* C, 83, 054610, 2011.
- 4. J. Gibelin, M. Wiedeking, L. Phair, P. Fallon, S. Basunia, et al, Channel selection of neutron-rich nuclei following fusion-evaporation reactions of light systems; *Nuclear Instruments and Methods in Physics Research A*, 648, 109-113, 2011.
- 5. R.B. Firestone and Zs. Revay, *Comparison of IUPAC k₀ values and neutron cross sections to determine a self-consistent set of data for neutron activation analysis*, Radiochimica Acta (in press).

Publications and Papers

- 6. A.M. Hurst, N.C. Summers, B.W. Sleaford, R.B. Firestone, *Gamma spectrum from neutron capture on tungsten isotopes,* Proceedings of the International Conference on Nuclear Data and Technology 2010, Jeju Island, Korea.
- A.M. Hurst, R.B. Firestone, B.W. Sleaford, Zs. Revay, M. Krticka, T. Belgya, M.S. Basunia, R. Capote, H. Choi, D. Dashdorj, J. Escher, A. Nichols, L. Zsentmiklosi, *Data evaluation methods and improvements to the neutron-capture g-ray spectrum,* Proceedings of the Second International Ulaanbaatar Conference on Nuclear Physics and Applications, July 26-30 2010, Ulaanbaatar Mongolia.
- B.W. Sleaford, R.B. Firestone, N. Summers, J. Escher, A. Hurst, M. Krticka, S. Basunia, G. Molnar, T. Belgya, Z. Revay, and H.D. Choi, *Capture Gamma-Ray Libraries for Nuclear Applications,* Proceedings of the International Conference on Nuclear Data for Science and Technology, April 26-30, 2010, Jeju Island, Korea (in press).
- 9. A.M. Hurst, N.C. Summers, B.W. Sleaford, and R.B. Firestone, *Gamma Spectrum from Neutron Capture on Tungsten Isotopes*, Proceedings of the International Conference on Nuclear Data for Science and Technology, April 26-30, 2010, Jeju Island, Korea (in press).

Publications and Papers

- 10. R.B. Firestone, *Determination of γ-ray transition probabilities and decay branching intensities from neutron capture data, invited* paper presented the Decay Data Evaluation Project (DDEP) Workshop, 9-11 June, 2010, Madrid.
- 11. R.B. Firestone, *Deviations from the Statistical Model in Capture* γ *-ray and* EC/β +*-decay Data,* invited paper presented at the 3rd Workshop on Level Density and Gamma Strength, May 23-27, 2011, Oslo.
- 12. R.B. Firestone, *Elemental Analysis with Prompt and Delayed Gamma-rays using Guided Neutron beams, Neutron Generators, and Cosmic Rays,* invited paper presented at the 21st International Conference on the Applicationof Accelerators in Research and Industry, August 8-13, 2010, Fort Worth TX.
- 13. R.B. Firestone, ¹⁰⁰*Mo*(²⁸*Si*,*xnyp*) *with the LBNL Total Absorption Spectrometer (TAS),* invited paper presented at the Workshop on Decay Spectroscopy at CARIBU: Advanced Fuel Cycle Applications, Nuclear Structure and Astrophysics, April 14-16, 2011, Argonne National Laboratory.

Future Developments

1. Nuclear Data

- Complete EGAF database with publication in the literature and inclusion in the ENSDF, ENDF, and DDEP databases.
- Measure new surrogate reaction and thermal neutron cross sections.
- Develop new statistical models of M1 and beta decay strengths.
- Update the on-line *Table of Isotopes* (Collaboration with UC Berkeley Nuclear Engineering Department).

2. Research

- Take on the STARS/LiBeRACE leadership role at the 88" cyclotron.
- Continue guided neutron beam measurements at the Budapest and Munich reactors.
- TAS measurements at CARIBOU
- Systematic studies of level densities and M1/E2 photon strengths
- Applications of nuclear data to energy and environment

Importance to the Nation

Nuclear data is critical to the success of many scientific disciplines

- Reactor Design poor data leads to costly overdesign of reactors
- Homeland Security nuclear data will be critical in dealing with a dirty bomb attack
- National Defense nuclear data is beginning to move to China
- Nuclear Chemistry expertise in NAA/PGAA is no longer in the US
- Nuclear Medicine the cost of radiation therapies depends on nuclear data
- Education thousands of students and Universities use the *Table of Isotopes* website.
- Environment radiation is a part of our environment that can be used to study the questions currently most interesting to society

It is important that nuclear data scientists not only provide the best possible data but also participate in real world applications of the data

Isotopes Project Analysis of Deep Sea Vents



Black smoker at the East Pacific Rise

J. Anal. At. Spectrom., 2001, 16, 1–7

	ALVIN 917-R4	ALVIN 1457-1R-C	ALVIN 1461-2R
0	45.9 [*]	41(6), 44.9*	45.1*
s	20.0 (0.2)	0.151 (0.005)	0.16 (0.01)
Ca	11.3 (0.2)	7.22 (0.11)	7.25 (0.13)
Fe	9.28 (0.11)	9.65 (0.08)	9.37 (0.09)
Cu	7.67 (0.07)		
Al		7.10 (0.07)	7.06 (0.12)
Mg	1.8 (0.2)	3.98 (0.11)	3.6 (0.2)
Zn	1.36 (0.05)		
Р		0.85 (0.18)	1.6 (0.2)
Ni	1.17 (0.003)	0.022 (0.002)	
Ti		1.097 (0.008)	1.060 (0.010)
Si	0.55 (0.05)	22.6 (0.3)	22.3 (0.3)
Н	0.368 (0.004)	0.0290 (0.0005)	0.027 (0.001)
K	0.27 (0.06)	0.138 (0.004)	0.16 (0.01)
Cl	0.194 (0.002)	0.0566 (0.0005)	0.0188 (0.0005)
Mn		0.154 (0.002)	0.161 (0.004)
Na	0.140 (0.014)	1.97 (0.04)	1.96 (0.05)
V		0.042 (0.002)	0.046 (0.003)
Co	0.0066 (0.0011)	0.0045 (0.0003)	0.0058 (0.0009)
Sc		0.0039 (0.0002)	0.0058 (0.0005)
Cd	0.00352 (0.00005)		0.00024 (0.00003)
В	0.00220 (0.00002)	0.000659 (0.000007)	0.000658 (0.000008)
Dy		0.00099 (0.00008)	0.00111 (0.00014)
\mathbf{Gd}	0.000050 (0.000006)	0.000524 (0.000007)	0.000556 (0.000010)
Sm	0.00033 (0.00003)	0.000330 (0.000005)	0.000340 (0.000007)

Concentrations (Wt.%) measured by PGAA

Isotope Project discovered of the Younger Dryas impact*





Impact markers found in a narrow sediment layer dated to 12.9 kyr BP at >25 sites in North America and Belgium show that a large comet exploded over the Laurentide Ice Sheet possibly creating the Great Lakes and causing

Onset of >1000 years of Younger Dryas cooling

• Sudden extinction of mammoths and other megafauna PGAA/NAA/XRF analysis shows that the comet was enriched in Ir and Fe/Ti with a *similar composition to lunar Procellarum KREEP Terrane*.

*R.B. Firestone, et al, Proc. Nat. Acad. Sci. 104, 16016–16021 (2007).



Fe/Ti microspherule

Nanodiamonds



Glass-like carbon

Isotopes Project discovers meteorites in 35 kyr BP Mammoth tusks*





Mammoth tusks and bison, musk ox, and rhinocerous skulls dated to \approx 34,000 BP contain micrometeorites. Tusk micrometeorite with characteristic burn ring.

Beringian extinctions occurred at this time. Probable cause is meteor impact into Sithylemenkat Lake, AK.

* *Micrometeorite Impacts in Beringian Mammoth Tusks and a Bison Skull*, R.B. Firestone, A. West, Zs. Stefanka, Zs. Revay, J.T. Hagstrum, 2007 AGU Fall Meeting, San Francisco.

PGAA Analysis of Bison Particle				
Z	EI	Wt(oxide)	±(%)	
1	Н	9.0%	3.2	
5	В	7.4 ppm	6	
6	С	36%	5	
7	Ν	8.4%	4.0	
9	F	10.4%	5	
11	Na	0.34%	5	
14	Si	0.35%	13	
15	Р	14.5%	3.3	
16	S	0.33%	9	
17	CI	82 ppm	5	
19	Κ	250 ppm	20	
20	Ca	19.2%	3.0	
22	Ti	120 ppm	26	
23	V	150 ppm	16	
24	Cr	0.08%	14	
25	Mn	0.109%	3.9	
26	Fe	1.01%	4	
28	Ni	400 ppm	18	
Mass of Fe=360 μg				
Radius of micrometeorite ≈1 mm				

Isotopes Project discovers four prehistoric near-Earth Supernovae*





Cosmic rays from Near-Earth SN correlate with changes in global temperature.

Renormalized comparison indicates SN are standard candles.

Date	Distance*	
(kyr BP)	(PC)	
44	≈110	
37	≈180	
32	≈160	
22 (Vela SN)	=250±30	

Evidence of 4 prehistoric supernovae <250 pc from Earth during the past 50,000 years. 18 more were found over past 300 kyr in the ¹⁰Be ocean sediment record

* R.B. Firestone, paper PP31D-1386, American Geophysical Union Fall Meeting, December 2009.

Thank you for your attention

