



Current Decay Experiments

Libby McCutchan National Nuclear Data Center

WoNDRAM – June 20th, 2021



Experimental Campaigns

Funded through Nuclear Data Interagency Working Group Lab Call

- Improving the Nuclear Data on Fission Product Decays at CARIBU LLNL-ANL – 4 year project funded from FY2018
- Novel approach for Improving Nuclear Data for Antineutrino Spectra Predictions ANL - 4 year project funded from FY2018
- Beta-strength function, reactor decay heat and anti-neutrino properties from total absorption spectroscopy of fission fragments
 ORNL, 4 year project funded from FY2019

Any decay data on fission fragments helps !

- TAGS : Valencia/Jyvaskyla and MSU/SUN
- Discrete Spectroscopy
- Beta Shape Measurements



Required decay data: Beta intensities

One major ingredient is beta feeding intensities



S(E)=N W (W²-1)^{1/2}(W-W₀)² F(Z,W) C(Z,W)(1+ δ)



The problem for high Q values : Pandemonium





Required decay data : shape factor

 $I(E) = N W (W^{2}-1)^{1/2} (W-W_{0})^{2} F(Z,W) C(Z,W)_{screen} \delta_{WM} \delta_{rad}$ $C_{exp} = 1 + a_{1}W + a_{2}W^{2} + a_{3}W^{3} + b_{1}/W$

Shape factor : remaining correction accounting for nuclear structure C= 1 for allowed transitions – adopted in most calculations

Important in both

- Summation
- Conversion







MTAS measurements

Studies with ORNL's **Modular Total Absorption Spectrometer (MTAS**) at the ORNL's HRIBF and at the ANL's CARIBU facilities







Impressive efficiency, plotted here up to 6 MeV total 99%-98%, single γ-line 80%-70%





ENSDF: 240 y's, 63 levels

Brookhaven[•] Krzysztof Piotr Rykaczewski and Bertis Charlie Rasco Physics Division ORNL

Slide courtesy of K. Rykaczewski

MTAS measurements : light mass fission peak

	Nb 102	Nb 101	Nb 100	Nb 99	Nb 98	Nb 97	Nb 96	Nb 95	Nb 94	Nb 93	Nb 92	Nb 91	Nb 90	Nb 89
	1.3 s	7.1 s	1.5 s	15.0 s	2.86 s	72.1 m	23.35 h	34.991 d	20.3 ky	100.	34.7 My	680 y	14.60 h	2.03 h
	Zr 101	Zr 100	Zr 99	Zr 98	Zr 97	Zr 96	Zr 95	Zr 94	Zr 93	Zr 92	Zr 91	Zr 90	Zr 89	Zr 88
	2.3 s	7.1 s	2.1 s	30.7 s	16.90 h	2.80	64.032 d	17.38	1.53 My	17.15	11.22	51.45	78.41 h	83.4 d
	Y 100	Y 99	Y 98	Y 97	Y 96	Y 95	Y 94	Y 93	Y 92	Y 91	Y 90	Y 89	Y 88	Y 87
	735 ms	1.470 s	548 ms	3.75 s	5.34 s	10.3 m	18.7 m	10.18 h	3.54 h	58.51 d	64.00 h	100	106.65 d	79.8 h
	Sr 99	Sr 98	Sr 97	Sr 96	Sr 95	Sr 94	Sr 93	Sr 92	Sr 91	Sr 90	Sr 89	Sr 88	Sr 87	Sr 86
	269 ms	653 ms	429 ms	1.07 s	23.90 s	75.3 s	7.423 m	2.66 h	9.63 h	28.79 y	50.53 d	82.58	7.00	9.86
	Rb 98	Rb 97	Rb 96	Rb 95	Rb 94	Rb 93	Rb 92	Rb 91	Rb 90	Rb 89	Rb 88	Rb 87	Rb 86	Rb 85
	114 ms	169.9 ms	203 ms	377.5 ms	2.702 s	5.84 s	4.492 s	58.4 s	2.6 m	15.15 m	17.78 m	27.83	18.642 d	72.17
			Kr 95	Kr 94	Kr 93	Kr 92	Kr 91	Kr 90	Kr 89	Kr 88	Kr 87	Kr 86	Kr 85	Kr 84
fority 1 for decay heat	6 PI	1:	114 ms	210 ms	1.286 s	1.840 s	8.57 s	32.32 s	3.18 m	2.84 h	76.3 m	17.30	10.776 y	57.00
riority 2 for decay heat	4 pi	2:	Br 94	Br 93	Br 92	Br 91	Br 90	Br 89	Br 88	Br 87	Br 86	Br 85	Br 84	Br 83
priority for antineutrinos	13 r	·	70 ms	102 ms	343 ms	0.64 s	1.910 s	4.40 s	16.36 s	55.65 s	55.1 s	2.90 m	31.80 m	2.40 h
		V .	Se 93	Se 92	Se 91	Se 90	Se 89	Se 88	Se 87	Se 86	Se 85	Se 84	Se 83	Se 82
		20 ms	50 ms	100 ms	270 ms	>300 ns	410 ms	1.53 s	5.8 s	14.1 s	33 s	3.1 m	22.3 m	8.73

39 decays measured between 2012 and 2016



Slide courtesy of K. Rykaczewski

MTAS measurements : heavy mass fission peak

Ce 139	Ce 140	Ce 141	Ce 142	Ce 143	Ce 144	Ce 145	Ce 146	Ce 147	Ce 148	Ce 149
137.641 d	88.450	32.508 d	11.114	33.039 h	284.8 d	2.98 m	13.52 m	56.4 s	56 s	5.3 s
La 138	La 139	La 140	La 141	La 142	La 143	La 144	La 145	La 146	La 147	La 148
0.090	99.910	1.6781 d	3.92 h	92.6 m	14.3 m	40.9 s	24.8 s	6.27 s	4.015 s	1.26 s
Ba 137	Ba 138	Ba 139	Ba 140	Ba 141	Ba 142	Ba 143	Ba 144	Ba 145	Ba 146	Ba 147
11.232	71.698	83.06 m	12.752 d	18.27 m	10.7 m	14.5 s	11.5 s	4.31 s	2.22 s	893 ms
Cs 136	Cs 137	Cs 138	Cs 139	Cs 140	Cs 141	Cs 142	Cs 143	Cs 144	Cs 145	Cs 146
13.16 d	30.1671 y	32.2 m	9.27 m	63.7 s	24.94 s	1.689 s	1.791 s	994 ms	582 ms	323 ms
Xe 135	Xe 136	Xe 137	Xe 138	Xe 139	Xe 140	Xe 141	Xe 142	Xe 143	Xe 144	Xe 145
9.10 h	8.87	3.83 m	14.08 m	39.68 s	13.60 s	1.73 s	1.22 s	511 ms	388 ms	188 ms
I 134	I 135	I 136	I 137	I 138	I 139	I 140	I 141	I 142	I 143	I 144
52.0 m	6.61 h	45s 84 s	24.2 s	6.4 s	2.29 s	860 ms	430 ms	~200 ms	100 ms	50 ms
Te 133	Te 134	Te 135	Te 136	Te 137	Te 138	Te 139	Te 140	Te 141	Te 142	
12.5 m	41.8 m	2 18.6 s	17.5 s	2.49 s	1.4 s	>300 ns	300 ms	100 ms	50 ms	
Sb 132	Sb 133	Sb 134	Sb 135	Sb 136	Sb 137	Sb 138	Sb 139			_
2.79 m	2.5 m	780 ms	1.68 s	923 ms	450 ms	500 ms	300 ms			
Sn 131	Sn 132	Sn 133	Sn 134	Sn 135	Sn 136	Sn 137				
56.0 s	39.7 s	1.45 s	1.12 s	530 ms	250 ms	190 ms				

7 priority 1 for decay heat
 4 priority 2 for decay heat
 8 priority for antineutrinos

38 decays measured between 2012 and 2016



MTAS current campaign at CARIBU at ANL





Decays of 14 isotopes marked by black ovals were studied during the experiments with MTAS at CARIBU.

Slide courtesy of K. Rykaczewski

And more TAGS measurements

Valencia/Jyvaskyla collaboration





Nuclear Data on Fission Product Decays at CARIBU

• Fission product selection by compact CARIBU isobar separator

LLNL-ANL teams performs various decay experiments:

- CARIBU delivers mass-separated beams of any fission product with $t_{\rm 1/2}$ > 25 ms



Kay Kolos Nick Scielzo







Beta-delayed γ-ray branching ratios

Nuclear data for fission yields - impacts nuclear forensics



Fission-product isomer-to-ground state ratios

Understanding of fission dynamics and angular momentum



Guy Savard Filip Kondev







Slide courtesy of K. Kolos

Fission product decays



Precision branching ratio measurements

Radiopure sample harvested at CARIBU

- Ions deposited on thin carbon foil
- Precision gamma-ray measurements at TAMU



 Isotopes measured so far: ⁹⁵Zr, ¹⁴⁷Nd, ¹⁴⁴Ce and ¹⁵⁶Eu

Figures: Most recent results for ¹⁵⁶Eu

→ Comparison of current (NNDC) evaluated data with our results for γ-ray branching ratios

K. Siegl, K. Kolos, N. D. Scielzo et al. "Beta-decay half-lives of ^{134,134m}Sb and their isomeric yield ratio produced by the spontaneous fission of ²⁵²Cf" PRC 98, 054307 (2018)

K. Kolos, A. M. Hennessy, N. D. Scielzo et al. "New approach to precisely measure γ -ray intensities for long-lived fission products, with results for the decay of 95 Zr" NIM A 1000 165240 (2020)



Demonstrated resolution of >10⁷ allowing isomers to be separated

- lons are trapped
- Excited to a particular radius in trap
- Allowed to rotate freely in trap along circle
- Ejected after a period of time to measure location



• More measurements to come! Slide courtesy of K. Kolos

Isomer-to-g.s. measurement on ^{102,102m}Nb

Decay Spectroscopy with Gammasphere

Funded under : Novel approach for Improving Nuclear Data for Antineutrino Spectra Predictions

Two experimental campaigns

- ¹⁴⁴La, ^{146g,m}La, ¹⁴⁴Ba, ¹⁴⁶Ce
- ^{102g,m}Nb, ^{104g,m}Nb, ¹⁰²Zr, ¹⁰⁴Zr, ¹⁰²Mo, ¹⁰⁴Mo



Gammasphere for Beta Decay New moving tape system + 6 plastic scintillators



F.G. Kondev et al., EPJ Web of Conferences 223, 01028 (2019)



Gammasphere vs. TAGS

Where does Pandemonium take over?

141Ce 32.511 D	142Ce >5E+16 Y	143Ce 33.039 H	144Ce 284.91 D
β-: 100.00%	11.114% 2β-	β-: 100.00%	β-: 100.00%
582.7	-745.7	1461.6	318.6
140La 1.67855 D	141La 3.92 H	142La 91.1 M	143La 14.2 M
β-: 100.00 %	β 10.00%	β-: 100.00 %	β-: 100.00 %
3760.2	25	4509	3435
139Ba 83.06 M	140Ba 12.7527 D	141Ba 18.27 M	142Ba 10.6 M
β-: 100.00 %	β-: 100.00%	β-: 100.	β-: 100.00 %
2312.5	1047	3199	2182
138Cs 33.41 M	139Cs 9.27 M	140Cs 63.7 S	141Cs 184 S
β-: 100.00 %	β-: 100.00 %	β-: 100.00 %	β-: 10. 10%
5375	4213	6219	p-n: 0.64% 5255

¹⁴¹Ba:

3.2 MeV Q value High fission yield Discrete measurement: 1980's Greenwood TAGS : 1990's









Excellent agreement with TAGS for strong transitions

All the pieces are coming together



A.A. Sonzogni et al., Phys. Rev. C 91, 011301(R) (2015).





TAGS measurements

PRL 115, 102503 (2015)

PHYSICAL REVIEW LETTERS

week ending 4 SEPTEMBER 2015

Total Absorption Spectroscopy Study of ⁹²Rb Decay: A Major Contributor to Reactor Antineutrino Spectrum Shape

A.-A. Zakari-Issoufou,¹ M. Fallot,¹ A. Porta,^{1,*} A. Algora,^{2,3} J. L. Tain,² E. Valencia,² S. Rice,⁴ V. M Bui,¹ S. Cormon,¹ M. Estienne,¹ J. Agramunt,² J. Äystö,⁵ M. Bowry,⁴ J. A. Briz,¹ R. Caballero-Folch,⁶ D. Cano-Ott,⁷ A. Cucoanes,¹ V.-V. Elomaa,⁸ T. Eronen,⁸ E. Estévez,² G. F. Farrelly,⁴ A. R. Garcia,⁷ W. Gelletly,^{2,4} M. B. Gomez-Hornillos,⁶ V. Gorlychev,⁶ J. Hakala,⁸ A. Jokinen,⁸ M. D. Jordan,² A. Kankainen,⁸ P. Karvonen,⁸ V. S. Kolhinen,⁸ F. G. Kondev,⁹ T. Martinez,⁷ E. Mendoza,⁷ F. Molina,^{2,4} I. Moore,⁸ A. B. Perez-Cerdán,² Zs. Podolyák,⁴ H. Pentilä,⁸ P. H. Regan,^{4,10} M. Reponen,^{8,4} J. Rissanen,⁸ B. Rubio,² T. Shiba,¹ A. A. Sonzogni,¹¹ C. Weber,^{8,8} and IGISOL collaboration⁸



PRL 117, 092501 (2016) PHYSICAL REVIEW LE

TERS

week ending 26 AUGUST 2016

Decays of the Three Top Contributors to the Reactor $\bar{\nu}_e$ High-Energy Spectrum, ⁹²Rb, ⁹⁶gsY, and ¹⁴²Cs, Studied with Total Absorption Spectroscopy

B. C. Rasco,^{1,2,3,4,*} M. Wolińska-Cichocka,^{5,2,1} A. Fijałkowska,^{6,3} K. P. Rykaczewski,² M. Karny,^{6,2,1} R. K. Grzywacz,^{3,2,1}
 K. C. Goetz,^{7,3} C. J. Gross,² D. W. Stracener,² E. F. Zganjar,⁴ J. C. Batchelder,^{8,1} J. C. Blackmon,⁴ N. T. Brewer,^{1,2,3}
 S. Go,³ B. Heffron,^{3,2} T. King,³ J. T. Matta,² K. Miernik,^{6,1} C. D. Nesaraja,² S. V. Paulauskas,³ M. M. Rajabali,⁹
 E. H. Wang,¹⁰ J. A. Winger,¹¹ Y. Xiao,³ and C. J. Zachary¹⁰

G.S. Branch 91 (3) %





Discrete Spectroscopy





5 HPGe Clover Detectors Large plastic scintillator



Beta spectra for short-lived fission products

Isotopes that emit many high energy vs contribute the most to the signal:

- Large (cumulative) fission yields
- Large Q values
- Large BRs to low-lying states
- → short-lived isotopes at the mass peak such as ⁹²Rb, ⁹⁶Y, ¹⁴²Cs, etc.

Most of the important transitions are first forbidden... but reactor calculations assume a nearly allowed shape

Recent calculations indicate different firstforbidden operators lead to large distortions from allowed shape

We are making precision investigations of the spectral shape!



Journal of Physics G: Nuclear and P

Self-consistent calculation of the reactor antineutrino spectra including forbidden transitions

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We started with ⁹²Rb – highestimpact decay



Preliminary results indicate $0^- \rightarrow 0^+$ transition has allowed shape

Slide courtesy of K. Kolos

LLNL-PRES-816784

Nuclear Structure repurposed

PHYSICAL REVIEW C 101, 044311 (2020)

Editors' Suggestion

Shape evolution of neutron-rich ^{106,108,110}Mo isotopes in the triaxial degree of freedom

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$106_{MO} \qquad 60 \qquad \gamma (deg) \qquad 4.8 \\ 3.6 \\ 2.4 \\ 20 \\ 0.2 \\ \beta \\ 0.4 \\ 0.0 \\ 1.2 \\ 0.0 \\ 0.0 \\ 1.2 \\ 0.0 \\$

PHYSICAL REVIEW C 103, 024323 (2021)

Ground-state and decay properties of neutron-rich ¹⁰⁶Nb

A. J. Mitchell[©],^{1,*} R. Orford,^{2,3,†} G. J. Lane[©],¹ C. J. Lister[©],⁴ P. Copp,^{4,‡} J. A. Clark,³ G. Savard,^{3,5} J. M. Allmond,⁶ A. D. Ayangeakaa,^{7,8} S. Bottoni[©],^{3,§} M. P. Carpenter[©],³ P. Chowdhury,⁴ D. A. Gorelov[©],^{3,9} R. V. F. Janssens[©],^{7,8} F. G. Kondev,³ U. Patel,^{1,||} D. Sewervniak,³ M. L. Smith[©],^{1,¶} Y. Y. Zhong[©],¹ and S. Zhu^{3,#}





High Priority List for Reactor Antineutrinos P. Dimitriou, A.L. Nichols, IAEA report INDC(NDS)-0676, IAEA (Austria, Vienna, 2015)

Kr-91 🔆	Sr-97 🔆	Nb-98 🔆	I-138 🔆
Rb-88 🔆	Y-94 🔆	Nb-100	Xe-139 🔆
Rb-90 🔆	Y-95 🔆	Nb-101	Xe-141
Rb-92 🔆 🌔	Y-96 🔆 🄇	Nb-102 【 🔘	Cs-139 🔆
Rb-93 🔆 🌔	Y-97/97m 🔆	Nb-104m 🅄 🌔 🔘	Cs-140 🔆 🌔
Rb-94 🔆	Y-98m 🔆 🌔	Te-135 🔆	Cs-141 🔆
Sr-95 🔆	Y-99	I-136/136m 🔆	Cs-142 🔆 【
Sr-96 🔆	Zr-101 🌂	I-137 🔆 🌔	La-146 🔆 🔘



Valencia/Jyvaskyla TAGS

ANL - Gammasphere

Summary and Conclusions

- 100's of experiments on data decay relevant to reactor antineutrinos have been performed in the last 10 years.
- For beta intensities we are approaching multiple measurements from multiple groups for nearly all "high-priority" isotopes.
- Next, hopefully, will come similar campaigns of beta shape measurements.
- And even more data from future RIB facilities fully documented so can be utilized for reactor antineutrino calculations

