Beta-strength, decay heat and anti-neutrino energy spectra from total absorption spectroscopy

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Complex *β*-decays

* β-transitions (mostly Gamow-Teller) are feeding highly excited states,

* these many, weak β -transitions are followed by the cascades of γ -transitions in the daughter nucleus,

* these weak γ-transitions are very difficult to detect with radiation detectors with low efficiency

Total absorption γ-spectroscopy

* to determine **true \beta-feeding** and resulting γ -decay patterns (nuclear structure),

* to determine **"decay heat"** released by radioactive nuclei produced during a nuclear fuel cycle

* to determine **anti-neutrino spectrum** associated with β -decay of fission products, $Y \rightarrow X^* + e^- + \overline{v}$

J. Hardy et al., Physics Letters 71 B, 307, 1977

A. Algora et al., PRL 105, 202501, 2010 K. P. Rykaczewski, Viewpoint in Physics 3, 94, 2010 K. P. Rykaczewski, 2013 McGraw-Hill Yearbook of Science and Technology, p.92



The true picture of the neutron-rich parent nucleus (Z,N), with many weak β-transitions and following low intensity γ-transitions.



N-RICH PARENT (Z,N)

Reactor antineutrino anomaly

PHYSICAL REVIEW D 83, 073006 (2011) Reactor antineutrino anomaly

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Deficit of detected anti-neutrinos vs expected counting rate at the level of 0.943(23) obtained after re-evaluation of 1980's integral β -energy spectra nicknamed "Schreckenbach data" for n + ²³⁵U, ^{239,241}Pu reactions

More general – the differences between the expected and measured \overline{v} -spectrum



Antineutrino energy spectra

Two basic pieces of information are needed to evaluate the energy spectrum of anti-neutrinos next to nuclear reactor:

1.Nuclear fuel content in a function of time (burn-up phase) 2.Knowledge of β -spectra (anti-v spectra) in fission products



from Ian Gauld, 2014 Nuclear Reactor Science (ORNL)

Ian Gauld is a keeper of Oak Ridge Isotope Generator code (ORIGEN) code, which is a part of SCALE package



β-spectra in fission products

Two methods:

- 1."Ab initio" (or summation calculations)
 - → use decay data base ENSDF and add up all beta-decays of fission products generating anti-neutrinos (fission yields are needed for reactor inventory nuclei)
- 2. Use integral experimental data on β -decays
- \rightarrow "Schreckenbach data" from 1980's (cumulative β -spectra for n+ ²³⁵U, ²³⁹Pu, ²⁴¹Pu)
 - e.g., K. Schreckenbach, G. Colvin, W. Gelletly, F. Von Feilitzsch, Phys. Lett. B 160, 325 (1985)

and recent n+ ²³⁸U cumulative β-spectra: Haag,.., Schreckenbach, PRL 112,122501, 2014

(and A. Hayes et al., PRL 112, 202501 (2014) for discussion of GT/FF in β-spectra de-convolution)



Our approach:

 \rightarrow use MTAS verified/improved (true) β -decay data for summation calculations

First step – let's check the effect of our MTAS results on fission products with respect to the number of interacting anti-neutrinos



Modular Total Absorption Spectrometer (MTAS)

*19 NaI(Tl) hexagonal shape blocks, each 21" long and 8" maximum diameter (SGC, Hiram, OH)



* the weight of a single detector is ~120 pounds, the total weight of the detector array is ~2 200 pounds



*MTAS is mounted at the adjustable cart to facilitate measurements at different beam line height (47" – 53")





MTAS shielding (background reduction)

The weight of (mostly Pb) shielding for this setup is **~12 000 pounds** *(with about 1" layer of solid lead and ~0.75" lead in lead wool blankets)*





Rate for MTAS (most exposed single module)

- without shielding ~16000 Hz (test stand near a lab with some activated materials)

- with four Pb blankets and paraffin ~600 Hz

- with "Pb house", Pb blankets and paraffin shielding ~160 Hz (now two SWX-227A layers instead of paraffin bricks)



AUXILIARY DETECTORS and MOVING TAPE DRIVE

* HPGe detector for sample composition monitoring,



- surrounding the tape-collected sample (tape by Ed Zganjar, LSU)
- the solid angle covered by two Si-detectors around 95% of 4π
- the low energy threshold for electrons < 50 keV
- the energy resolution for 976 keV electrons 2.5 % (~25 keV)





Beta energy loss spectra measured with segmented Si-detectors help to center radioactive samples in the middle of MTAS





MTAS γ -efficiency verified using calibrated ¹³⁷Cs, ⁵⁴Mn and ⁶⁵Zn single γ -line sources



Measurements vs response function simulations



black points – exp red line - simulations



Aleksandra Fijałkowska Charlie Rasco

Marek Karny







MTAS on-line to the HRIBF mass separator OLTF (On-line Test Facility = ISOL, $m/\Delta m \sim 600$)



Ge detector for monitoring – collection point

> Data acquisition based on digital pulse processing and using XIA Pixie16 modules (100 MHz)



MTAS at the HRIBF, January 2012 studies at mass A ~ 90 and 140 regions with OLTF and Tandem

The decays of fission products produced at the HRIBF on-line mass separator (OLTF) using Tandem beam and studied with new Modular Total Absorption Spectrometer (MTAS) are marked by yellow and red squares.

The label"1" indicates the highest priority for decay heat measurement established by the Nuclear Energy Agency evaluation in 2007.

Y88	Y89	Y90	Y91	Y92	Y93	Y94	Y95
106.65 d		64.00 h	58.51 d	3.54 h	10.18 h	18.7 m	10.3 m
Sr87	Sr88	Sr89	Sr90	Sr91	Sr92	Sr93	Sr94
		50.53 d	28.79 y	9.63 h	2.66 h	7.42 m	75.3 s
Rb86	Rb87	Rb88	Rb89	Rb90	Rb91	Rb92	Rb93
18.64 d		17.78 m	15.15 m	2 158 s	58.4 s	4 .49 s	5.84 s
Kr85	Kr86	Kr87	Kr88	Kr89	Kr90	Kr91	Kr92
10.77 y		76.3 m	2.84 h	3.15 m	32.32 s	8.57 s	1.84 s
Br84	Br85	Br86	Br87	Br88	Br89	Br90	Br91
31.80 m	2.90 m	55.1 s	55.65 s	16.36 s	4.40 s	1.91 s	541 ms
Se83	Se84	Se85	Se86	Se87	Se88	Se89	Se90
22.3 m	3.1 m	31.7 s	15.3 s	5.50 s	1.53 s	410 ms	300 ms
As82	As83	As84	As85	As86	As87	As88	As89
19.1 s	13.4 s	4.02 s	2.02 s	945 ms	610 ms	300 ms	200 ms

La 137	La 138	La 139	L a 140	L a 141	L a 142	La 143	
^{60 ky}	^{0.09}	^{99.91}	1.68 d	3.92 h	92.6 m	14.3 m	
Ba 136	Ba 137	Ba 138	Ba 139	Ba 140	Ba 141	Ba 142	
_{7.85}	11.23	71.7	83.06 m	12.75 d	18.27 m	10.7 m	
Cs 135	Cs 136	Cs 137	Cs 138	Cs 139	Cs 140	Cs 141	
2.3 My	13.16 d	_{30.17 y}	32.2 m	9.27 m	63.7 s	24.94 s	
Xe 134	Xe 135	Xe 136	Xe 137	Xe 138	Xe 139	Xe 140	
10.44	9.10 h	8.87	3.83 m	14.08 m	^{39.68 s}	13.6 s	
I 133	I 134	I 135	I 136	l 137	I 138	I 139	
20.8 h	^{52 m}	6.61 h	^{84 s}	24.2 s	6.4 s	2.29 s	
Te 132	Te 133	Te 134	Te 135	Te 136	Te 137	Te 138	
3.2 d	12.5 m	41.8 m	18.6 s	17.5 s	2.49 s	1.4 s	
Nuclear Science NEA/WPEC-25 INTERNATIONAL Evaluation Co-operation Volume 25 ASSESSMENT OF FISSION PRODUCT DECAY DATA FOR DECAY HEAT CALCULATIONS A report by the Working Party on International Evaluation Co-operation of the NEA Nuclear Science Committee							
CO-ORDINATOR T. Yachida Musashi Instinte of Technology JAPAN Musashi Instinte of Technology International Atomic Energy Agency AUSTRIA							

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National Laboratory

Priority "1" $^{139}Xe \rightarrow ^{139}Cs$ decay (T $_{1/2}$ = 39.7 s) cumulative yield of ^{139}Xe in n_{th} + ^{235}U fission is about 5%



MTAS result:

average **gamma** energy release per ¹³⁹Xe decay increased from 935 keV to 1370 keV **46.5 % increase**

average **beta** energy release per ¹³⁹Xe decay decreased from1774 keV to 1573 keV **13% decrease**



Aleksandra Fijałkowska et al., Nuclear Data Sheets 120, 26 (2014)

Measured and **modified** ¹³⁹Xe decay in central and inner rings of MTAS









 10^{1}



Energy [keV]

Electron and anti-neutrino energy spectra in ¹³⁹Xe decay



In ¹³⁹Xe decay, the average anti-neutrino energy is shifted down from 2348 keV (ENSDF) to 2114 keV (MTAS), i.e., by 234 keV



Anti-neutrino interactions with protons



A. Strumia and F. Vissani, PL B 564, 42, 2003

Interacting anti-neutrinos emitted in ⁸⁹Kr and ¹³⁹Xe decay

Number of interactions of ⁸⁹Kr and ¹³⁹Xe anti-neutrinos per 10 keV energy bin (in % x 10⁻⁴³ cm² units).



reactor antineutrino anomaly 94.3(23) % → ~ 6 % missing

MTAS results for ⁸⁹Kr and ¹³⁹Xe decays account for **2.9 of the** *"missing* **6%**" difference in the reactor anti-neutrino anomaly for **a fresh nuclear fuel load**, and for **1.8** and **1.5 out of the missing 6%** for the burn-up phases corresponding to **2% FIMA and 3% FIMA**, respectively, if applied to the anti-neutrino anomaly as calculated from all data contained in ENSDF.

Summary

Modular Total Absorption Spectrometer with its auxiliary Si strip detectors, HPGe detector, shielding structure and tape collector system was constructed, characterized and applied to the decay studies of **over twenty** fission products, seven of highest NEA priority, at the HRIBF at Oak Ridge.

MTAS is the **largest and the most efficient** Total Absorption Spectrometer ever built (factor 7 in volume). Its **modular construction** helps to verify the analysis of complex decay patterns (now similar construction - DTAS from Valencia).

Test decay chain ${}^{142}Ba -> {}^{142}La -> {}^{142}Ce$, where earlier measured high resolution and total absorption data were used to generate the β -strength distribution, demonstrated a reasonable agreement between MTAS experimental spectra and simulated MTAS response.

Deduced true beta-strength patterns offer the guidance and verification for the theoretical calculations of beta-decay process in neutron-rich nuclei (to be done, mostly deformed nuclei).



Summary (results)

New data on decays of abundant ^{235,238}U fission products, like ⁸⁹Kr and ¹³⁹Xe, demonstrated an increase in average γ-energy emitted per decay (decay heat) by 28 % and 47 %, respectively.

New beta-decay schemes based on MTAS data, with an increased beta feeding to the high energy states (hence lower beta transition energies) affect also anti-neutrino spectra. Lower energies of anti-neutrino largely reduce the number of interactions with protons (matter).

 89 Kr – 41% and 139 Xe - 20%

These two decays alone reduce the number of reactor antineutrinos interacting with protons by about 1.5 % (up to 3 %)

Single detector experiments detecting anti-neutrinos next to the power reactors should take into account the correct(ed) antineutrino energy spectra, when deducing the magnitude of "reactor anti-neutrino anomaly"



Anti-neutrino interactions with protons deduced from MTAS measurements for ⁸⁶Br

MTAS vs ENDSF



In average, in ⁸⁶Br decay we have about **7% less** of anti-neutrino interactions with protons.

It is NOT a typical case, since the beta feeding to high energy levels (low end-point energies) was partially compensated by the increased feeding to the ground state.



Total β -gated MTAS spectrum for ¹⁴²La decay

Earlier measurements for ¹⁴²La decay included total absorption spectroscopy [see Greenwood et al. NIM A390 (1997)]



TAGS based simulations are close to MTAS data

Marzena Wolinska-Cichocka et al., Nuclear Data Sheets 120, 22 (2014)





Maybe it is not the end ...

New proposal submitted to DOE Nuclear Data Program is asking for a funding of two-weeks of ORNL Tandem operation for the OLTF-MTAS

Parent	Half-life	Ion rate at 50 nA (pps)	MTAS Q _β (Q _β -Emax level) [MeV]	²³⁸ U yield [%]	Shifts required
136I ^{gs,m}	83s/47s	6.8E5	6.9 (0.3/2.8)	1.3	1
138I	6.2 s	1.3E4	7.8 (2.5)	3.1	1
139I	2.3 s	1.5E3	6.8 (5.1)	2.1	2
140I	0.86 s	1.0E2	8.7 (6.9)	0.9	3
140Xe	14 s	1.0E2	4.1 (1.5)	4.9	3

MTAS "discovery window": Qβ - Emax_level



new beginning (of experiments at the Tandem)

Parent	Half-life	Ion rate at 50 nA	MTAS Q _β (Q _β -Emax level) [MeV]	²³⁸ U vield	Shifts required
		(pps)		[%]	requireu
85Br	2.9 m	5.7E5	2.9 (0.8)	0.74	1
88Br	16 s	1.3E5	9.0 (2.0)	1.5	1
88Kr	2.8 h		2.92 (0.15)	2.0	1
89Br	4.4 s	1.5E4	8.3 (3.5)	1.9	2
90Br	1.9 s	2.8E3	10.4 (4.6)	1.4	2
91Br	0.54 s	1.8E2	9.8 (5.4)	0.8	3





MTAS result:

average **gamma** energy release per ⁸⁹Kr decay increased from 1801 keV to 2304 keV

28 % increase

the average energy of **beta** particles per 89Kr decay decreased from 1462 keV to 1222 keV **20% decrease**

Marek Karny et al., in presentation at ND 2013 Aleksandra Fijałkowska et al., Nuclear Data Sheets 120, 26 (2014)





Electron and anti-neutrino spectra in ⁸⁹Kr decay



In ⁸⁹Kr decay, the average anti-neutrino energy is shifted from 1914 keV (ENSDF) to 1650 keV (MTAS), i.e., by 264 keV



Anti-neutrino energy spectrum deduced from MTAS



A. Fijałkowska et al., Acta Phys. Pol. B 45, 545, 2014

MTAS-corrected β-decay scheme of ⁸⁶Br (+14 % in decay heat) points to a larger fraction of low energy anti-neutrinos than can be expected from ENDSF.
Note that the energy threshold for the v+p+n+e⁺ reaction is 1.8 MeV, exactly at the energy region where the change occurred.









Beta decay of very neutron-rich nuclei is indeed interesting and complex



Part of beta-strength "under S_n " is easier (not easy) to study and compare to models (it is *easier* to investigate, if you have right tools like **MTAS** array)

MTAS amplitude stability



MTAS Temperatures at OLTF - December 28-29, 2011

Amplitude stability is not a problem when room temperature is stable (gain matching/testing with 1115 keV γ-line from ⁶⁵Zn) Work on an active amplification control in progress: K.C. Goetz, R. Grzywacz (UTK) (laser light pulser split over reference detector and MTAS modules)



Holifield Radioactive Ion Beam Facility at Oak Ridge



January 2012



Anti-neutrino energy spectra from beta-decay of fission products

$$z \mathbf{X}_{N} \xrightarrow{\beta^{-}} z_{+1} \mathbf{Y}_{N-1} + \mathbf{e}^{-} + \overline{\mathbf{V}}$$



A=142 test measurement



Time scale of A=142 experiment and analysis time gates





Anti-neutrino energy spectrum deduced from MTAS



MTAS-corrected β -decay scheme of ⁸⁶Br (+14 % in decay heat) points to a larger fraction of low energy anti-neutrinos than can be expected from ENDSF. Note that the energy threshold for the ∇ + p \rightarrow n + e⁺ reaction is 1.8 MeV, exactly at the energy region where the change occurred.

Anti-neutrino interactions with protons deduced from MTAS measurements for ¹³⁹Xe decay

¹³⁹Xe makes 3.3% of n_{th} +²³⁸U direct fission products and 4.3% for n_{th} + ²³⁵U



MTAS: number of ¹³⁹Xe anti-neutrino interactions with protons reduced by 21%

Neutrino probability in % per 10 keV

In ⁸⁹Kr decay, this effect is as large as 41 % !



Impact on the reactor anti-neutrino measurements with a single detector !





Earlier measurements for ¹⁴²Ba decay: * high resolution Ge detectors data [see ENSDF / NDS 112, 8 (2011)] * total absorption spectroscopy [see Greenwood et al. NIM A390 (1997)]

high resolution data are similar to TAGS data MTAS data agree with previously adopted β -decay pattern

Marzena Wolinska-Cichocka et al., Nuclear Data Sheets 120, 22 (2014)



Sterile anti-neutrinos rule ! (see DNP APS-JPS, Oct. 2014)

- T. Maruyama \rightarrow dark matter candidate
- Warren et al (Notre Dame) \rightarrow enhancing of SN explosion energy
- Maricic 10 MCi activities 51 Cr (Q_{EC}=0.75 MeV) as a source of mono-energetic neutrinos 10⁴ Ci 144 Ce- 144 Pr 3 MeV β —endpoint (see PRL107, 2011).
- Padgett (former UT, now LLNL), Orrell (PNNL) better cumulative β-spectra
- Mumm (NIST), Classen (LLNL) \rightarrow great prospects of PROSPECT (at HFIR)
- Lasserre (Saclay) \rightarrow Nucifer detector 7m from Osiris reactor (Saclay)
- Learned, Vogelar NuLAT 3D anti-neutrino detection at 1500 MW Navy Reactor ..

Warnings:

No new cosmological concordance with massive sterile neutrino, PRL113,041301, 2014 Daya Bay data interpretation does not require an extra neutrino, PRL 113,141802, 2014



Efforts by the Neutrino Community

Numerous Very Short Baseline Experiments ranging from operating to planning stages





Neutrino-4, Russia

Operational, D=7m

Nearly Operational, D=6-13m

Experimental Concept reactor core detector 1 -4m detector 1 -4m detector 2 reactors under consideration: NIST, ATR, HFIR

PROSPECT, USA

Coming soon at HFIR ?

CORMORAD – Italy, PANDA – Japan, SOLiD - France

from Libby Mc Cutchan (Nuclear Data BNL) Caribu decay spectroscopy workshop, May 2014.

