Beta decay studies with the Total Absorption Technique
a remedy against “Pandemonium”
ANL Colloquium

Berta Rubio
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Most unstable nuclei decay by either $\beta^+$ or $\beta^-$ decay

About 3000 out of 6000 synthesised in our laboratories.

Proton Drip Line

Neutron Drip Line
The beta decay process

\[ \beta^- : n \rightarrow p + e^- + \bar{v} \]
\[ \beta^+ : p \rightarrow n + e^+ + v \]

Fermi and Gamow-Teller

Single state, Fermi

Triplet state Gamow-Teller

The selection rules

\[ \Delta l = 0 \]
\[ \Delta s = 0,1 \]
\[ \Delta \pi = 0 \]

It happens real nuclei

\[ \beta^- : {}^A_z X_N \rightarrow {}^A_{z+1} X^*_{N-1} + e^- + \bar{v}_e \]
\[ \beta^+ : {}^A_z X_N \rightarrow {}^A_{z-1} X^*_{N+1} + e^+ + v_e \]
\[ \text{EC} : {}^A_z X_N + e^- \rightarrow {}^A_{z-1} X^*_{N+1} + v_e + X_{ray} \]
From the nuclear structure point of view the operator responsible for the process is very simple.

Or $\sigma \tau$ Gamow-Teller

$\tau$ Fermi

2p 1/2
1f 5/2
2p 3/2

$\sigma \tau$

1f 7/2

$\tau$

1f 5/2
2p 3/2
1f 7/2
The beta strength or transition probability governed by the spin-isospin operator

Theoretically

\[ B(GT) = \left| \langle \psi_f | \sum_k \sigma_k \tau_k^\pm | \psi_i \rangle \right|^2 \]

Experimentally

\[ B(GT) = k \frac{I_\beta(E)}{f(Q_\beta - E, Z)T_{1/2}} = k \frac{1}{ft} \]

From the experiment:

- \( I_\beta(E) \) - Beta feeding to states in the daughter nucleus
- \( T_{1/2} \) - Parent half life
- \( Q_\beta \) - Q-value
It is important if we want to describe one of the main processes that contribute to the origins and abundances of the heavy chemical elements in our World and in the part of the Cosmos accessible to our observation, the rapid neutron capture process (r-process).
Nuclear Astrophysics

Astrophysics
Defines stellar conditions
temperature, density, pressure
and chemical composition

Nuclear physics
Reaction cross sections,
n-capture probabilities, gamma
Absorption probabilities
decay probabilities, masses,
half lives
The pathways for the s- and r-processes

**s-process**: Neutron flux is low so beta decay occurs before a second neutron is captured. We slowly zigzag up in mass.

**r-process**: Neutron flux is enormous and many neutrons are captured before we get beta decays back to stability.
\[ B(GT) = \left| \langle \psi_f \left| \sum_k \sigma_k \tau_k^\pm \right| \psi_i \rangle \right|^2 = k \frac{1}{ft} \]

Calculations of T1/2 for waiting point nuclei

H. Grawe, K. Langanke and G. Martinez-Pinedo
Nuclear Structure and Astrophysics Reports
in Progress in Physics 70 (2007) 1525

Not so bad!!!
Larger differences (lack of experimental data)
Approximately 8% of the total energy generated during the fission process is related to the energy released in the natural decay of fission products, and is commonly called decay heat [1]. Once the reactor is shut down, the energy released in radioactive decay provides the main source of heating. Hence, coolant needs to be maintained after termination of the neutron-induced fission process in a reactor.

A. Algora et al PRL 105 105 (2010) 202501 Introduction....
Beta decay is a Weak process, and consequently slow

Typical constants
Pion-nucleon ("strong") 1
Electromagnetic $10^{-2}$
$\beta$ decay ("weak") $10^{-5}$
Gravitational $10^{-39}$
After the nucleus has been created there is some time available
Before it decays

Fusion evaporation projectile fragmentation, target spallation, fission...

Particle evaporation
10^{-19} s

Gamma de-excitation
fs, ns, μs, ms

Beta-decay ms, s, min, days, years
This allows us to separate the nucleus of interest from the rest of the nuclei produced in the reaction.
Figure 4. The r-process path together with the yield expected from an ion source system based on a 1 Ci Californium fission source and the limit of known masses.
Today, for very unstable nuclei we perform beta decay studies at fragmentation facilities (GSI, MSU, GANIL, RIKEN...)

production

Separation in flight with the Fragment Separator (FRS@GSI)

selection

identification

implantation

spectroscopy

50Fe

Active stopper and Gamma array

15/4/11

B. Rubio@Caribu_Workshop
$$B(GT) = \left| \left\langle \psi_f \right| \sum_k \sigma_k \tau_{k}^{\pm} \left| \psi_i \right\rangle \right|^2 \equiv \langle \sigma \tau \rangle^2$$

- Beta-feeding for from stability
- Many states, daughter
- One state (g.s) parent

Further from stability, higher $Q_\beta$-values
How to measure beta intensity

\[ B(GT) = k \frac{I_\beta(E)}{f(Q_\beta - E, Z)T_{1/2}} \]
For high Q-values, Ge detectors fail to detect β-feeding at high excitation energy!!!
$ZA_N$
John Matin’s 1825 engraving "Pandemonium".

J. Milton, *Paradise Lost, Book I* (1667)
Total Absorption spectroscopy

Ideal case
Total absorption spectroscopy

Real case
The main problem is not the intrinsic efficiency but the material placed inside the detector or the holes needed to put the activity inside.

One has to solve the following equation:

\[ d_i = \sum_{j=1}^{j_{\text{max}}} R_{ij} f_j \]
**ISOLDE/OSIRIS TAS:**

Duke et al. NP A151 (1970) 609
Hornshoj et al. NP A239 (1975) 15

2x Ø15cm×10cm cryst. + Plastic scin. ring

**St. Petersburg TAS:**

Bykov et al. IAN SSSR 44 (1980) 918

Ø20cm×30cm (2 cryst.) + Si det

**INEL TAS:**


Ø25cm×30cm well + Si det.

Duke et al in the 70’s, Studswik and Isolde
Firestone 1974, 1975
Hardy and 1977
Russians in the 80’s
Greengood 90’s
GSI 1994
A big step forward came with the construction and exploitation of the Berkeley-GSI TAS 1997.

TAS at GSI

Detector “Plug”: cylinder of NaI (Ø4.7 cm x 15 cm)

Mainly for proton rich nuclei

Ge detector (Ø16 mm x 10 cm)

Si detectors (Ø22 mm x 1 mm)

Main Crystal: NaI cylinder (Ø35.6 cm x 35.6 cm)

Positron absorber: polyethylene (Ø51 mm x 21 mm)
Accurate response of the whole apparatus to the gamma rays and the betas: Geant Monte Carlo simulations

Solve the inverse problem:

\[ d_i = \sum_{j=1}^{j_{\text{max}}} R_{ij} f_j \]


**LR method:** polynomial smoothing

\[ f = \left( R^T \cdot V_d^{-1} \cdot R + \lambda B^T \cdot B \right)^{-1} \cdot R^T \cdot V_d^{-1} \cdot d \]

**ME method:** entropy maximization

\[ f_j^{(s+1)} = f_j^{(s)} \exp \left( \frac{2}{\lambda} \sum_i \frac{R_{ij}}{\sigma_i^2} \left( d_i - \sum_k R_{ik} f_k^{(s)} \right) \right) \]

**EM method:** Bayes Theorem

\[ f_j^{(s+1)} = \frac{1}{\sum_i R_{ij}} \sum_i R_{ij} f_j^{(s)} d_i \]
στ excitation viewed from the reaction perspective is a collective spin isospin excitation

Gaarde NPA369(1981)258
152Tm $2^-$

4.4 MeV

B$_{GT}$ ($g_A^2/4\pi$)

0.36

0.34

0.32

0.30

0.28

0.26

0.24

0.22

0.20

0.18

0.16

0.14

0.12

0.10

Energy (keV)

$\frac{g_A^2}{4\pi}$

1.40

1.75

4.6 MeV

$\frac{g_A^2}{4\pi}$

0.05

0.09

0.56

0.58

4.3 MeV

$\frac{g_A^2}{4\pi}$

nothing

B$_{GT}$ = 0.10

150Ho $2^-$

148Tb $2^-$

$\frac{g_A^2}{4\pi}$

0.20

0.14

150Ho $9^+$

148Tb $9^+$

B.Rubio “Frontiers of Collective Motions” CM2002 Japan, World Scientific
Are we sure our deconvolution method is correct?

Are we sure that we cannot measure the $I_\beta$ if we use a very powerful Ge gamma array
Six EUROBALL CLUSTER detectors in close geometry
Underneath the resonance, there is a clear fine structure
A. Algora, B. Rubio et al PRC 50 (2002)

150 Ho β-decay

β-strength
TAS vs CLUSTERCUBE

$S^\beta_{\gamma}$ (10^{-6}s^{-1}keV^{-1})

0.4
0.3
0.2
0.1

0 2 4 6 8

Ex[MeV]

0 2 4 6 8

1064 γ-rays
295 levels

$\frac{S^HR_{\beta}}{S^{TAS}_{\beta}} = 0.4$
Encouraged by the success of these experiments and similar experiments carries out by the Warsaw group (M. Karny, K. Rykaczewski et al) We decided to construct a new TAS and put it at ISOLDE (CERN), But this time optimised for short half lives (less than 1 s)
The Lucrecia TAS at Isolde is a NaI single-crystal $\Phi 38\text{cm} \times 38\text{cm}$ (+ ancillary detectors )Ge for X-rays and plastic for $\beta$ particles
What can we learn from beta decay about the deformation of these nuclei?
Clarly prolate

Mixture of prolate and oblate

76Sr

oblate

prolate

154/11
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Ph.D thesis Valencia

Ph. D thesis Starsbourg
Beta (and alpha) decay represents 8% of the heat produced in a reactor when it is operation and 100% when the reactor stops.

A knowledge of how the decay heat varies with the time is needed because the cooling has to be maintained after the reactor has been switched off.

It is also important when the fuel is changed (waiting time) or when it is finally removed from the reactor and must be properly stored and shielded.
Decay heat: definition

\[ f(t) = \sum_i \left( \overline{E_{\gamma,i}} + \overline{E_{\beta,i}} + \overline{E_{\alpha,i}} \right) \lambda_i N_i(t) \]

- \( E_{\gamma,i} \), \( E_{\beta,i} \), \( E_{\alpha,i} \) Mean Decay energy of the nucleus \( i \) (gamma, beta or alpha)
- \( \lambda_i \) Decay constant of the nucleus \( i \)
- \( N_i \) Number of nuclei \( i \) at the cooling time \( t \)

Requirements for the calculations: large databases that contain all the required information (inventory of nuclides, half-lives, mean \( \gamma \)- and \( \beta \)-energies released in the decay, \( n \)-capture cross sections, fission yields, etc, etc …)
A long standing problem: “The γ-ray discrepancy between decay heat summation calculations and experimental data

Yoshida and co-workers (Journ. of Nucl. Sc. and Tech. 36 (1999) 135) and the work of the
Two main difficulties:
a) Refractory elements are difficult to extract in normal isol facilities
Second difficulty:
b) Pandemonium effect affects decay energy calculations

\[ \bar{E}_\beta = \sum_i I_\beta (E_i) \langle E_{\beta,i} \rangle \]
\[ \bar{E}_\gamma = \sum_i I_\beta (E_i) E_i \]

\( \bar{E}_\beta \) overestimation
\( \bar{E}_\gamma \) underestimation

Parent \((Z,N)\)
\[ \begin{array}{c}
\alpha_{10} \\
\beta\text{-transition} \\
E_\beta^i \\
\gamma\text{-transition} \\
\end{array} \]

Daughter \((Z+1,N-1)\)

Parent \((Z,N)\)

Missing levels
(Loss of \(\beta\)-strength)

\[ \alpha_1 \]

Daughter \((Z+1,N-1)\)
WPEC-25 (\(^{102,104,105,106,107}\text{Tc}, \^{105}\text{Mo}, \^{101}\text{Nb})

The “polar” experiences (motivations):
to study of the beta decay of nuclei relevant to the decay heat problem by means of the total absorption technique,
spokespersons
A. Algara and J.L. Taín (Valencia)
The ion gas guide technique mass separation and isobar separation

Use of the JYFLTRAP as a high resolution separator (first time that this kind of setup was used combined with a TAS)

50 MeV p on 15 mg/cm² nat U target

The process is fast enough for the ions to survive as single charged ions. The system is chemically insensitive and very fast (sub-ms).
Experimental setup at Jyväskylä

St. Petersburg TAS

Si det.

TAS det (NaI(tl))
(Det 1 & det 2).

Det 1: 20 cm diam., 20 cm length, 5 cm hole
Det 2: 20 cm diam, 10 cm length
LNPI design (St. Petersburg)
New feature: trap-assisted spectroscopy
Results of the analysis for $^{104}$Tc

$T_{1/2} = 1098(18)$ s; $Q_\beta = 5516(6)$ keV

$E_\beta$(TAGS) = 931 (10) keV
$E_\beta$(JEFF-3.1) = 1595 (75) keV \quad \Delta E_\beta = -664$ keV

$E_\gamma$(TAGS) = 3229 (24) keV
$E_\gamma$(JEFF-3.1) = 1890 (31) keV \quad \Delta E_\gamma = 1339$ keV

Results of the analysis for $^{102}$Tc

$T_{1/2} = 5.28(15)$ s; $Q_{\beta} = 4532(9)$ keV

$E_{\beta}(\text{TAGS}) = 1935 (11)$ keV
$E_{\beta}(\text{JEFF-3.1}) = 1945 (16)$ keV \quad \Delta E_{\beta} = -10$ keV

$E_{\gamma}(\text{TAGS}) = 106 (23)$ keV
$E_{\gamma}(\text{JEFF-3.1}) = 81 (5)$ keV \quad \Delta E_{\gamma} = 25$ keV
Reactor Decay Heat in $^{239}$Pu: Solving the $\gamma$ Discrepancy in the 4–3000-s Cooling Period

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$^{101}$Nb, $^{107}$Tc, $^{105}$Mo, $^{106}$Tc

$^{104}$Tc, $^{105}$Tc, $^{102}$Tc

Courtesy A. Sonzogni
Test of nuclear models:
Preliminary QRPA calculations (K.L. Kratz, priv. Com)
$T_{1/2}(\text{exp}) = 35.6 \text{ s}$

$[1-4.5] \text{ MeV}$
$\sum_{\text{TAGS}} = 87.99\%$
$\sum_{\text{Theo}} = 92.05\%$

$[0-0.5] \text{ MeV}$
$\sum_{\text{TAGS}} = 11.51\%$
$\sum_{\text{Theo}} = 7.94\%$

$[0-0.5] \text{ MeV}$
$\sum_{\text{TAGS}} = 11.51\%$
$\sum_{\text{Theo}} = 67.84\%$
Impact of the results for $^{235}$U

![Graph showing the time x decay heat (MeV/fission) over time after fission (S).]
- TAS measurements can play an important role in decay heat calculations.

- Our results show that approximately two thirds of the discrepancy is solved in the range 300-3000 s for $^{239}\text{Pu}$, and this is mainly related to the impact of the decay of $^{104,105}\text{Tc}$.

- From the available information (databases) it is clear that there is still a large amount of work to be done ($^{235}\text{U}/^{239}\text{Pu}$, $^{232}\text{Th}/^{233}\text{U}$ cycle). It requires close collaboration with the experts of the field in order to determine priorities.
This work has stimulated activities at HRIBF and ANL
ARRA Project: **Decay Studies of Fission Products w/ a new Modular Total Absorption Spectrometer (MTAS)**

PI’s: K. P. Rykaczewski (ORNL) and R.K. Grzywacz (UTK/ORNL)

A **Modular Total Absorption Spectrometer (MTAS)** has been constructed from 19 NaI(Tl) scintillator segments. MTAS is designed to perform decay studies with pure beams of neutron-rich nuclei produced in the $^{238}$U fission at HRIBF. The total absorption gamma spectra measured with MTAS will be used to derive a true beta-feeding pattern and resulting beta strength function. The studies are important for the verification and development of the microscopic description of neutron-rich matter will be performed as well as applied studies of decay heat released by radioactive nuclei produced in nuclear fuels at power reactors.

**Status:** the MTAS has been manufactured at the SGC (Hiram, OH) and delivered to the HRIBF. The tests done using digital electronics show the energy resolution superior to requested specs. Two PhDs were hired full time, one PhD part time.

**Funding:** $698 K capital + $882 K operations (includes $815 K salaries) = $1580 K

Funds committed/spent: $658 K capital and $512 K operations = $1270 K
Workshop on "Decay Spectroscopy at CARIBU: Advanced Fuel Cycle Applications, Nuclear Structure and Astrophysics"
April 14-16, 2011 at Argonne National Laboratory

A workshop on "Decay Spectroscopy at CARIBU: Advanced Fuel Cycle Applications, Nuclear Structure and Astrophysics" will be held at Argonne National Laboratory on April 14-16, 2011.

The aim of the workshop is to discuss opportunities for decay studies at the Californium Rare Isotope Breeder Upgrade (CARIBU) of the ATLAS facility with emphasis on advanced fuel cycle (AFC) applications, nuclear structure and astrophysics research. The workshop will consist of review and contributed talks. Presentations by members of the local groups, outlining the status of relevant in-house projects and available equipment, will also be organized. Time will also be set aside to discuss and develop working collaborations for future decay studies at CARIBU.

Topics of interest include:
- Decay data of relevance to AFC applications with emphasis on reactor decay heat
- Discrete high-resolution gamma-ray spectroscopy following radioactive decay and related topics
- Calorimetric studies of neutron-rich fission fragments using Total Absorption Gamma-ray Spectrometry (TAGS) technique
- Beta-delayed neutron emissions and related topics
- Decay data needs for nuclear astrophysics

Workshop Organizers
Dr. Michael Carpenter, Argonne National Laboratory
Prof. Partha Chowdhury, University of Massachusetts Lowell
Dr. Jason Clark, Argonne National Laboratory
Dr. Filip Kondej, Argonne National Laboratory
Dr. Kim Lister, Argonne National Laboratory
Dr. Dariusz Seweryniak, Argonne National Laboratory

Please visit the Workshop website for additional information about registration, program, lodging and transportation to Argonne.
http://www.anl.gov/capabilities/nd/AFC-Apr11/

Figure 4. The r-process path together with the yield expected from an ion source system based on a 1 Ci californium fission source and the limit of known masses.
Future
Three LoI presented in Feb 2011 (Tain et al, Algora et al, Rubio et al)

**Figure 2:** Layout of the DESIR hall with the permanent setups and general purpose places as foreseen today.

Valencia-Surrey TAS
Present: GSI
Z = 1 – 92
(from p to U)
Up to 2 GeV/nucleon
Some cooling

Future
Facility for Antiproton and Ion Research (FAIR)

Beams at FAIR:
Intensity: factor 100 (prim. beams)
10 000 fold (second. beams)
Z = -1 – 92
(anti-protons to uranium)
Up to 35 - 45 GeV/u
„full beam cooling“

Darmstadt, Germany, Hessen
Super-FRS will deliver beams at three different branches.
IN a Fragment Separator the ions are identified in Mass and Z, but a cocktail of ions arrive to the focal plane.
Implantation Detector
Sensitive to betas

Waiting time according with the half-life
Emission of β particle (or proton)
Correlation with gamma radiation

NUSTAR collaboration:
DESPEC experiment

High energy ions (fragmentation, fission, ...
) separated and identified with the
Super-FRS and implanted in an active
stopper

AIDA:
stack of DSSSD

132 modules:
LaBr₃:Ce

17 modules:
NaI(Tl)

Valencia, Madrid, Gatchina, Darmstadt,
Debrecen, Jyvaskyla, Surrey
Summary

Total Absorption Spectroscopy is a fundamental tool for beta decay studies of nuclei far from the stability.

These studies are important to test our models far from the stability, and in particular to guide these models when they are applied to r-process nuclei.

They are very important as input for decay heat calculations. This has a strong social impact as it has been (sadly) put in evidence in Fukushima. They are needed for the design of advanced Fuel Cycle reactors.

There are the working TAS spectrometers in Europe (plus some Russians).
Two other ones will be built and installed at the two large scale facilities (as defined in the ESFRI road map) in Europe.

Two other ones are being installed in the USA.

The analysis of these experiments is not trivial, but we think it is today under control.

Total Absorption Spectroscopy is an important part of the present and future facility at a number of laboratories including CARIBu@ANL.
Thank you for your attention