Metrology for Nuclear Physics: Research and Applications

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g factor measurements of μs isomeric states in neutron-rich nuclei around ^{68}Ni produced in projectile-fragmentation reactions

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g-factors of isomeric states i

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RAPID COMMUNICATIONS

PHYSICAL REVIEW C **78**, 061302(R) (2008)

Single-particle behavior at $N = 126$: Isomeric decays in neutron-rich ^{204}Pt

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PHYSICAL REVIEW C **104**, 024321 (2021)

Magnetic moment of the $11/2^-$ isomeric state in ^{99}Mo and neutron spin g factor quenching in $A \approx 100$ nuclei

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Impact from nuclear science?

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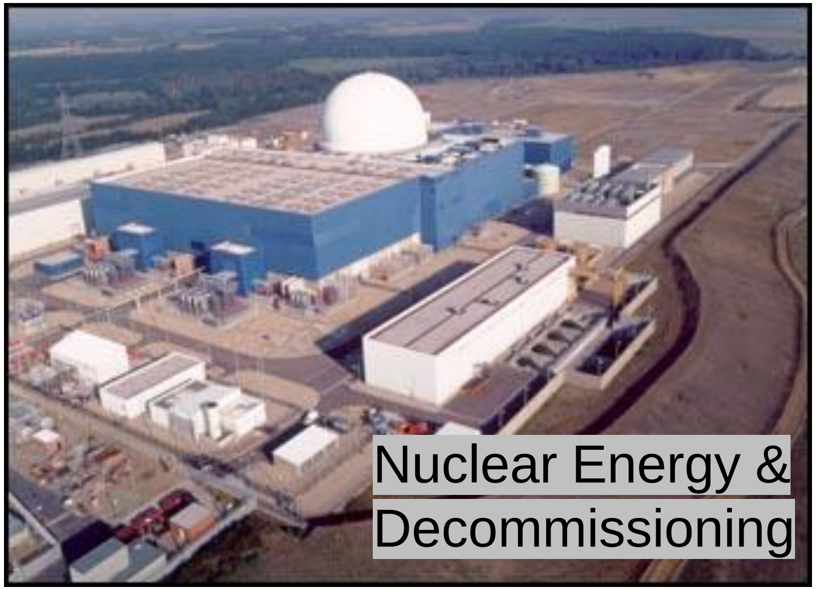
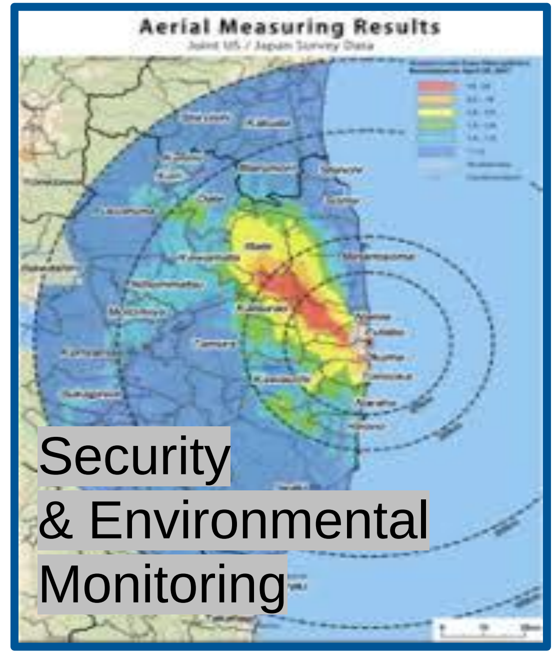
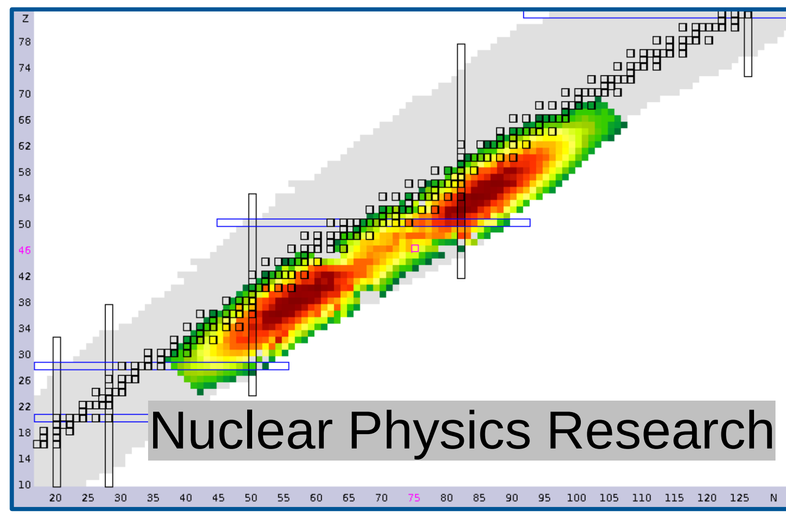






'*Keeping the lights on, fighting cancer, keeping us safe and keeping us curious*'

All rely on confidence in the measurement of radioactive materials.

Lots of applications of nuclear science measurement

- Nuclear power & waste management ($^{134,7}Cs$; ^{90}Sr; $^{239,240}Pu$, ^{241}Am…..)
- Medical thera(g)nostics (^{99m}Tc, ^{18}F, ^{82}Sr, ^{177}Lu $^{149,152,155,161}Tb$, ^{223}Ra…)
- NORMs (^{222}Rn; $^{226,228}Ra$, ^{40}K) & man-made environmental (^{90}Sr, ^{137}Cs)
- Chronology / long-term dating (^{14}C ; $^{238}U/^{206}Pb$; $^{40}K/^{40}Ar$)
- Nuclear forensics (e.g.,CTBTO weapons test), criticality monitoring
  - (Noble) gas radioactivity & isotopic ratios e.g. $^{133,135}Xe$ and $^{87,88}Kr$

All rely on **accurate Nuclear Metrology** via **traceable references standards**

which relate the activity (A) to the number of atoms (N) via

$$A = \lambda N = -(dN / dt)$$

What is a “primary standard”?

A measurement standard established using a **primary reference measurement procedure**”

A **primary measurement procedure** is:

“A reference measurement procedure used to obtain a measurement result **without relation to a measurement standard** for a quantity **of the same kind**”

Regan,, Judge,, Keightley & and Pearce (2018). Radionuclide metrology and standards in nuclear physics. Nuclear Physics News, **28(3**), pp.25-29.

Absolute primary standardisation of radioactive activity

- A 'primary' technique can measure, in parallel:
  - The Activity = $\lambda N = (N / \tau) = (0.693N) / T_{1/2}$.
  - The Absolute detection efficiency.

- 'No previous knowledge of nuclear data is required'…but:
  - Caveat 1: Some knowledge of the decay scheme is useful.
  - Caveat 2: The half-life ($T_{1/2}$) ~ inverse of the decay probability per unit time of the radionuclide (λ) is needed.

Primary Standards of Radioactivity

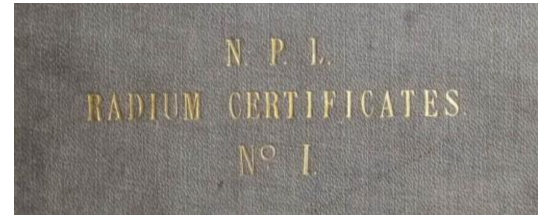
The first primary standard was a piece of radium ($T_{1/2}$=1600 ± 7) years, which was chemically purified and weighed

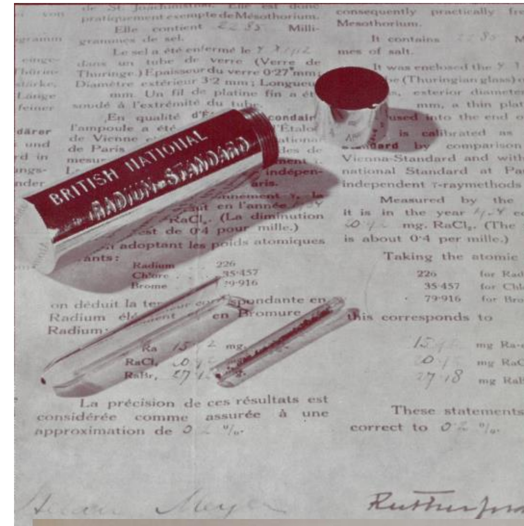
- 1 g of pure ^{226}Ra = activity of 1 Curie

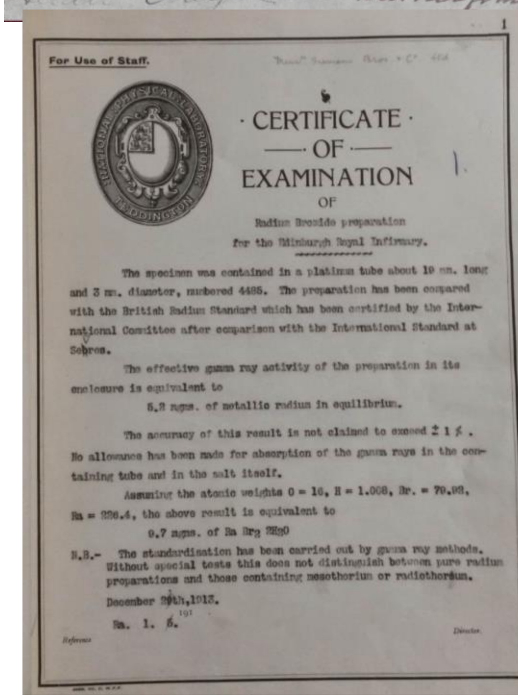
i.e. This was actually a **mass standard.**

There are many more radionuclides of interest which have shorter (or longer) half-lives than ^{226}Ra.

Radionuclide primary standards now are based on techniques rather than artefacts







For Use of Staff.

· CERTIFICATE ·
— OF —
EXAMINATION
OF

Radium Bromide preparation
for the Edinburgh Royal Infirmary.

The specimen was contained in a platinum tube about 10 mm. long and 3 mm. diameter, numbered 4485. The preparation has been compared with the British Radium Standard which has been certified by the International Committee after comparison with the International Standard at Sèvres.

The effective gamma ray activity of the preparation in its enclosure is equivalent to

5.8 mgms. of metallic radium in equilibrium.

The accuracy of this result is not claimed to exceed ± 1 %.

No allowance has been made for absorption of the gamma rays in the containing tube and in the salt itself.

Assuming the atomic weights O = 16, H = 1.008, Br. = 79.92, Ra = 226.4, the above result is equivalent to

9.7 mgms. of Ra Br2 2H2O

N.B.- The standardisation has been carried out by gamma ray methods. Without special tests this does not distinguish between pure radium preparations and those containing mesothorium or radiothorium.

December 29th, 1913.

Ra. 1. 5.

Reference

Director

JULY, 1931 REVIEWS OF MODERN PHYSICS VOLUME 3

THE RADIOACTIVE CONSTANTS AS OF 1930

REPORT OF THE INTERNATIONAL RADIUM-STANDARDS COMMISSION

BY M. CURIE, A. DEBIERNE, A. S. EVE, H. GEIGER, O. HAHN, S. C. LI
ST. MEYER, E. RUTHERFORD, AND E. SCHWEIDLER

I. INTRODUCTION

FOLLOWING the reorganization of the International Union of Chemi and of the International Atomic Weights Commission, the need arisen for the publication of special Tables of the Radioactive Constants.

This responsibility has been assumed by the International Rad Standards Commission chosen in Brussels in 1910, which has expressed willingness to cooperate with the International Union.

Besides the members, M. Curie, A. Debierne, A. S. Eve, H. Geiger Hahn, S. C. Lind, St. Meyer, E. Rutherford, E. Schweidler, the follow have taken part as experts: J. Chadwick, I. Joliot-Curie, K. W. F. Kohlrau A. F. Kovarik, L. W. McKeehan, L. Meitner and H. Schlundt, to whom desired to express especial obligations.

Recommended value.

Use of the value $3.7\cdot10^{10}$ is recommended in accord with reference 9.

1g ^{226}Ra equivalent to (SI) $3.7\text{x}10^{10}$ Bq

1 g of pure ^{226}Ra contains (N_A / 226) particles = **$2.7\text{x}10^{21}$ = N**

$T_{1/2}$(^{226}Ra) = 1600 yrs = $5.0\text{x}10^{10}$ s

λ (^{226}Ra) = ln2 /$T_{1/2}$ = **$1.37\text{x}10^{-11}$ s^{-1}**

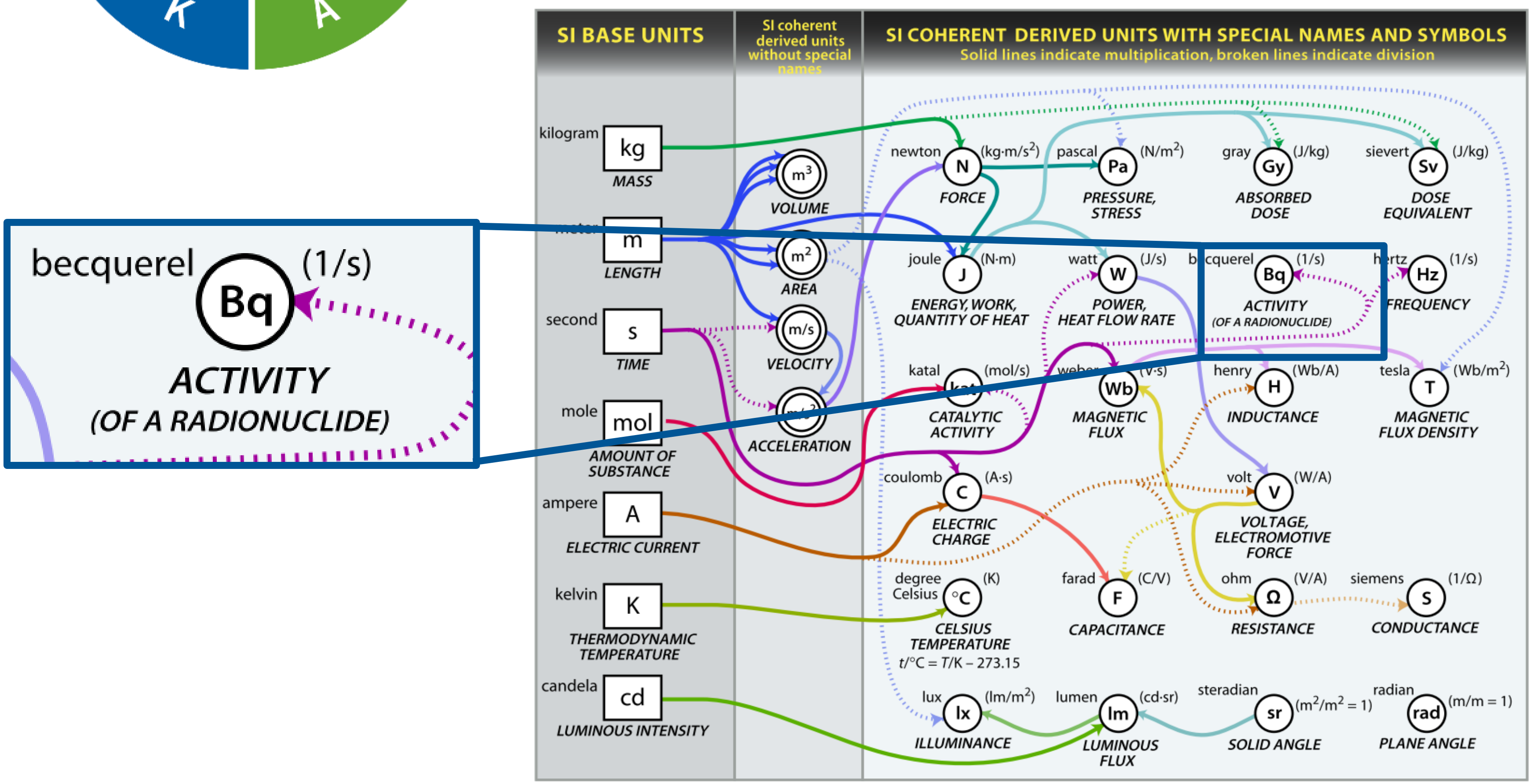
***A*(1g of ^{226}Ra) = λ . N = $3.7\text{x}10^{10}$ s^{-1}**

The Bq is a derived unit (s^{-1}) and the SI unit for radioactivity.

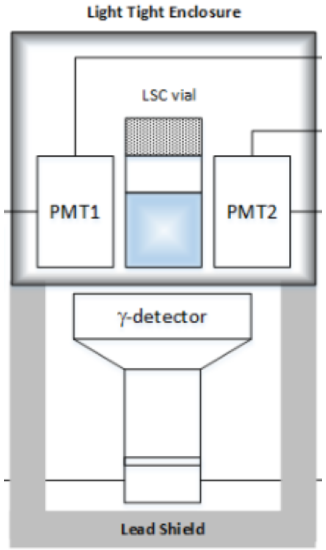
NPL National Physical Laboratory

7 independent base units.
All other 'derived units' are made from combinations of these 7. Examples:

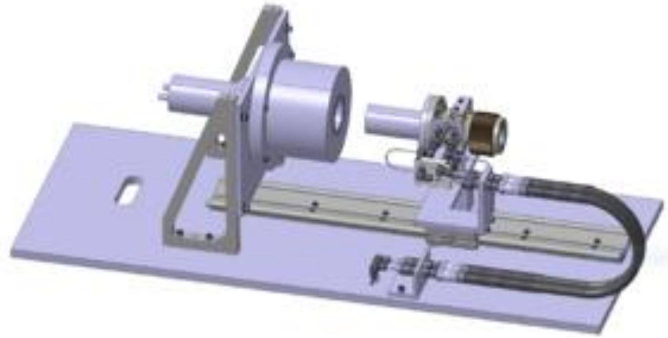
- $Bq = s^{-1}$
- $Pa = N.m^{2} = kg.m^{-1}s^{-2}$



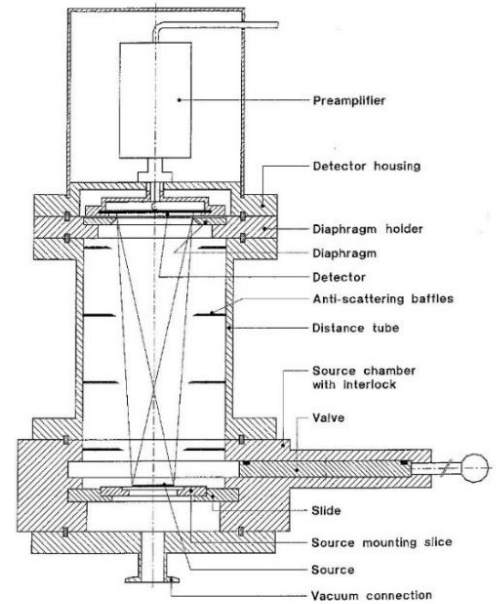
Primary standardisation techniques



4π(LS)-γ coincidence counting



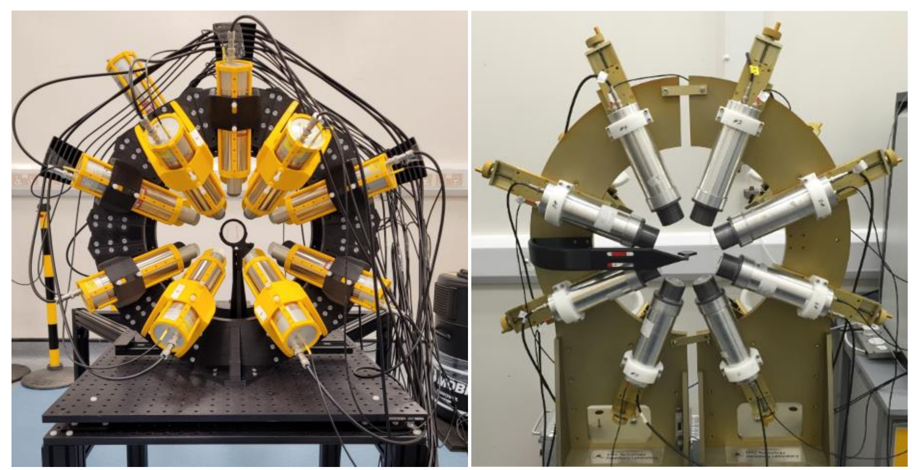
$4\pi\alpha/\beta$-γ coincidence counting



Defined by capability to measure simultaneously:
(1) The disintegration rate
(2) The detection efficiency

Defined Solid Angle α-counting

Liquid Scint. (LS) Triple-to-Double Coincidence Ratio counting



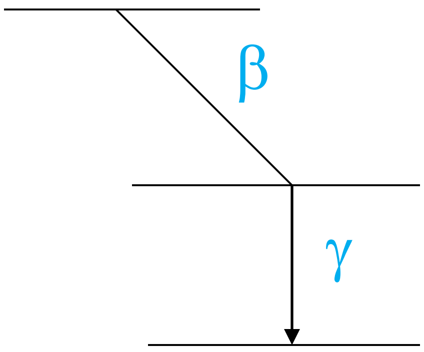
γ-γ coincidence counting

PHR et al., (2018). *Radionuclide metrology and standards in nuclear physics*. Nuclear Physics News, **28(3)**, pp.25-29.

Idealised example of the 4πβ–γ coincidence method

β–γ decay (100% fed single cascade)

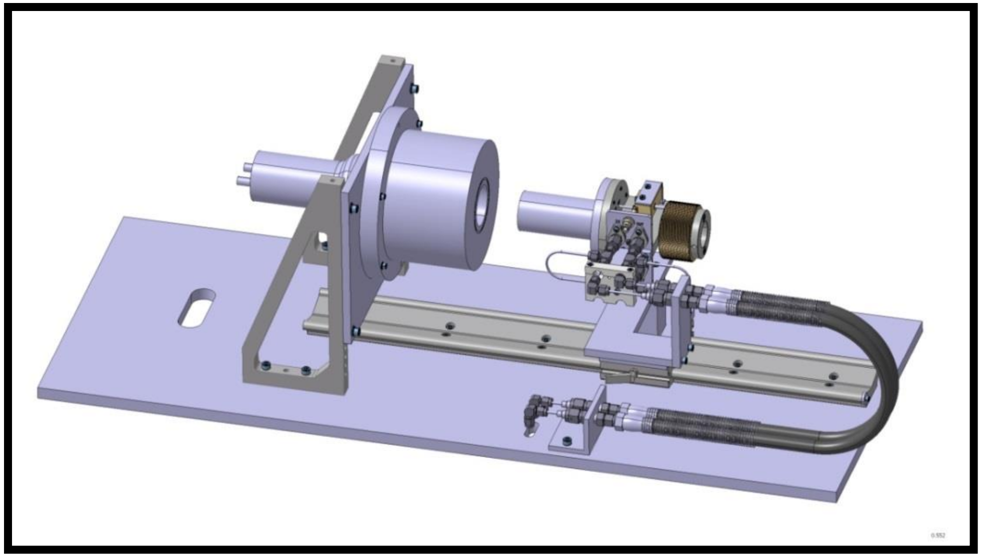
Count Rates



$N_\beta = N_o \varepsilon_\beta$

$N_\gamma = N_o \varepsilon_\gamma$

$N_c = N_o \varepsilon_\beta \varepsilon_\gamma$



Efficiencies:

$$\varepsilon_\beta = N_c/N_\gamma$$

$$\varepsilon_\gamma = N_c/N_\beta$$

$$Activity : N_o = \frac{N_\beta N_\gamma}{N_c}$$

Coincidence counting in nuclear physics is an ‘established’ idea.

MAY, 1940 R. S. I. VOLUME 11

The Application of the Method of Coincidence Counting to Experiments in Nuclear Physics

J. V. Dunworth
Denman Baynes Research Student, Clare College, Cambridge, England

(Received September 27, 1939)

The paper describes the principles involved in the application of coincidence counting to problems in nuclear physics, and gives details of the various methods of approach to the solution of nuclear level schemes, with some of the difficulties encountered. It is shown that the maximum source strength which can be used is inversely proportional to the coincidence resolving time and that the method of experiment has the great advantage over most other methods in that it can be used with very weak sources (sources having an activity of about 10^{-5} millicurie). An accurate knowledge of the absolute net efficiency of a Geiger counter for all types of radiation is required in the interpretation of the coincidence rates, and the method of achieving this at the same time as information is obtained about nuclear level schemes is explained. Further, it is pointed out that the method makes possible a simple and rapid determination of the total energy of disintegration of β-radioactive nuclei and therefore of the mass differences between parent and daughter nuclei.

times used* and demonstrating its constancy for all arrangements of Geiger counters used.

The errors shown are the theoretical statistical r.m.s. errors. The experimental weighted r.m.s. error of the resolving times in column 6 is 0.05×10^{-7} min. agreeing very closely with the statistical r.m.s. error of 0.03×10^{-7} min. Any lack of constancy of τ would have caused the former to be much greater than the latter. No genuine coincidence rates were operative due to the sources used in the above measurements (i.e., $G=0$).

Some idea of the order of magnitude of the various quantities involved in a nuclear coincidence experiment together with an indication of the method of experiment may be obtained

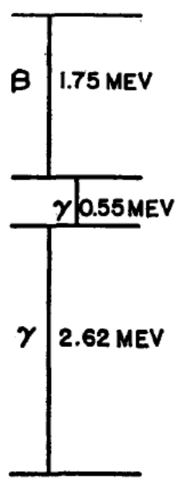
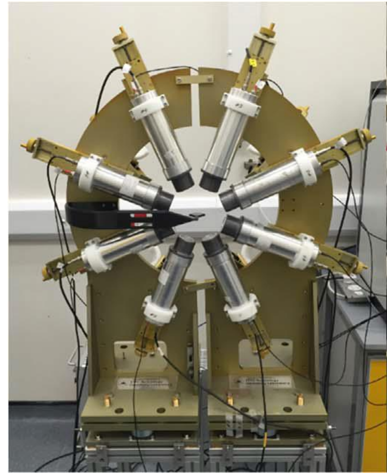


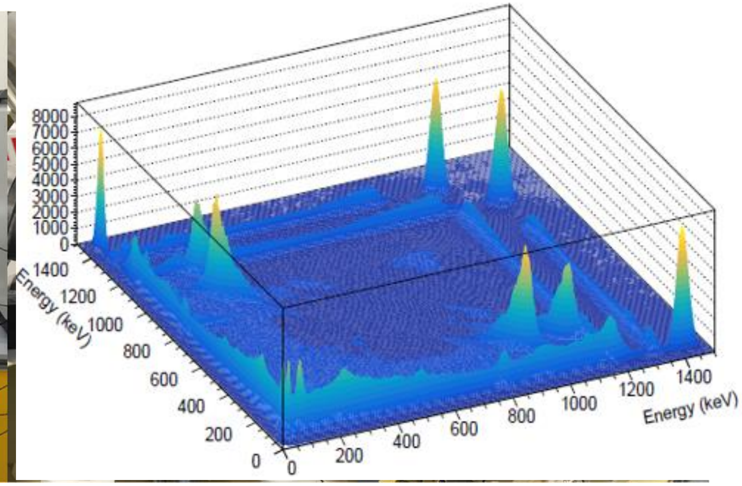
Fig. 1. Modified level scheme of ThC″.

John Vernon Dunworth,
Director, NPL 1964–1977
CIPM Vice-President 1968-1975
CIPM President 1975-1985

Standardisation of ^{60}Co: NANA (NAtional Nuclear Array))

- The multi-γ ray detector NANA used as a primary standard.
- Absolute activity of ^{60}Co determined using the γ-γ coincidence technique.
- Effect of angular correlations on the activity clearly observed





Contents lists available at ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

Investigation of γ-γ coincidence counting using the National Nuclear Array (NANA) as a primary standard

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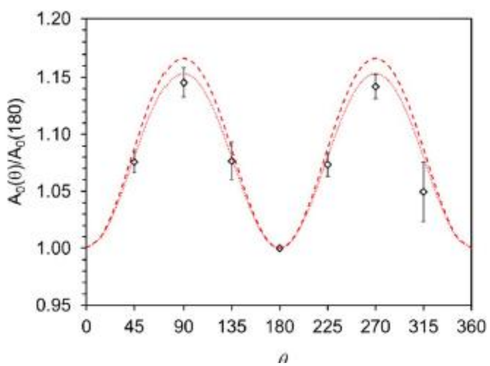
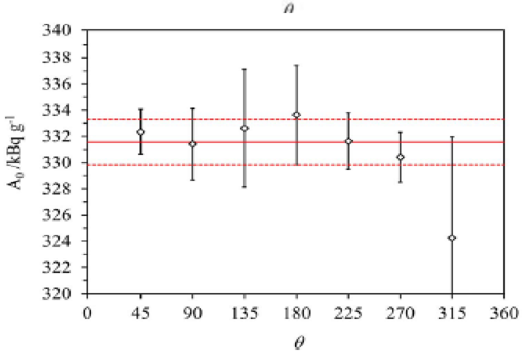




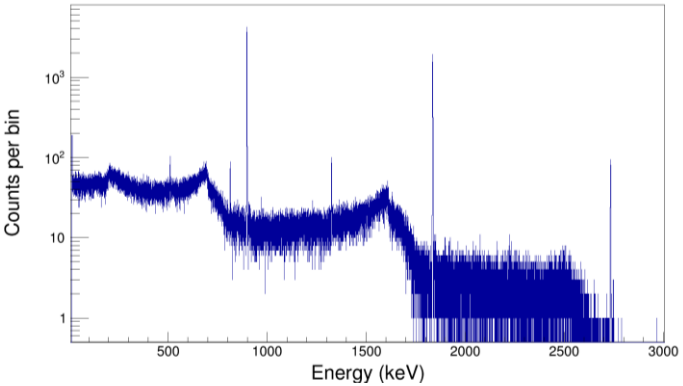
Standardisation technique	A_0 /kBq g^{-1}	$u(A_0)$ /kBq g^{-1}
NANA γ-γ coincidence counting	330.8	± 1.0
4π(LS)- γ DCC	330.92	± 0.86

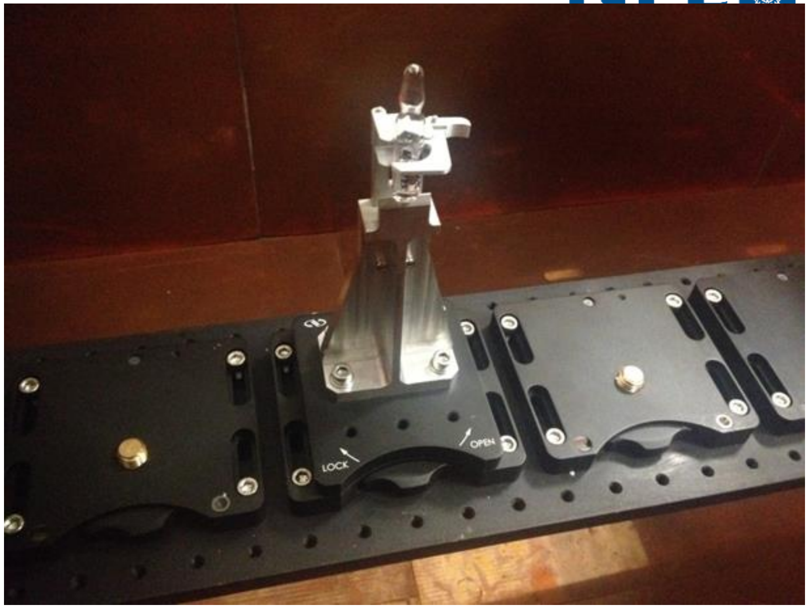
High resolution gamma spectrometry

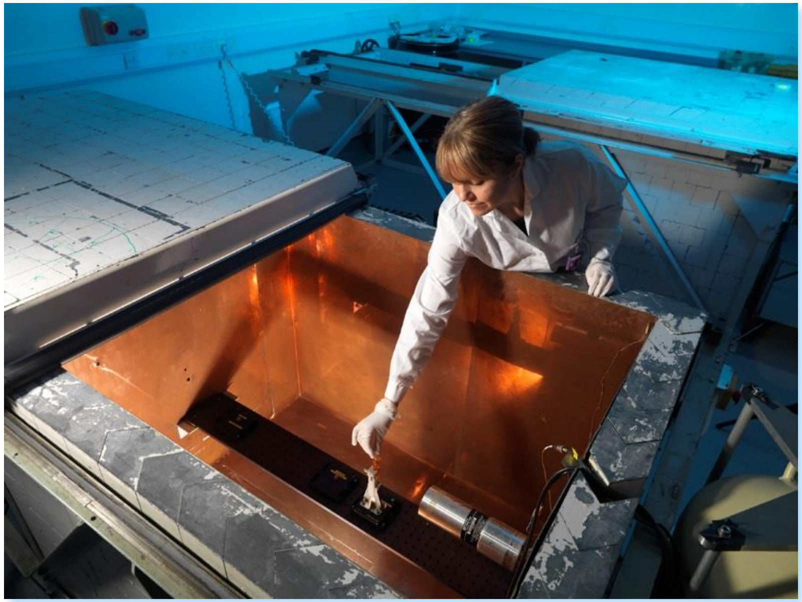
HPGe has excellent energy resolution allows for very clean peak identification.

HPGe measurements are not (usually) primary standards, but can be secondary..

1) **<u>Each geometry must have a precise calibration.</u>**
2) The nuclear data of the nuclides (i.e. $T_{1/2}$ and $P\gamma(\%)$ **must** be precise & accurate.

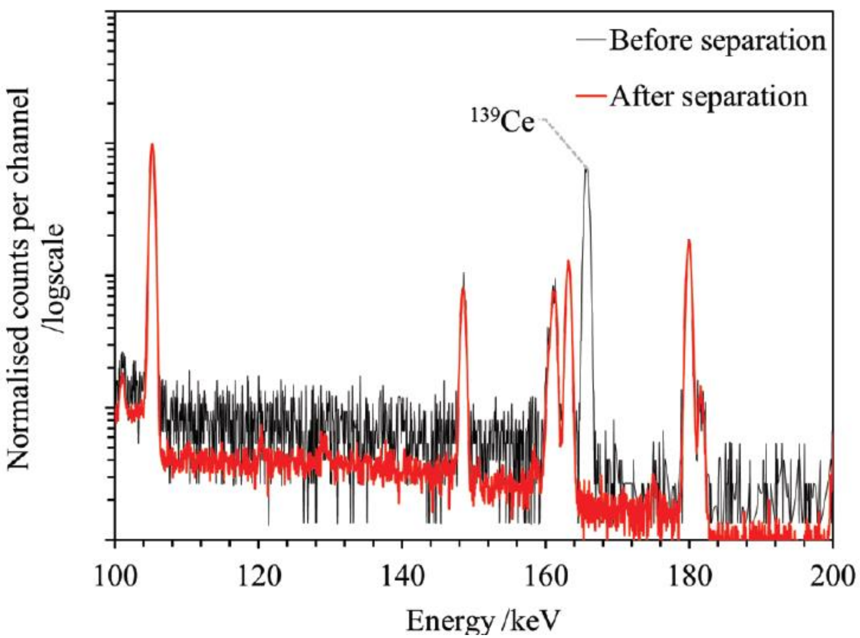




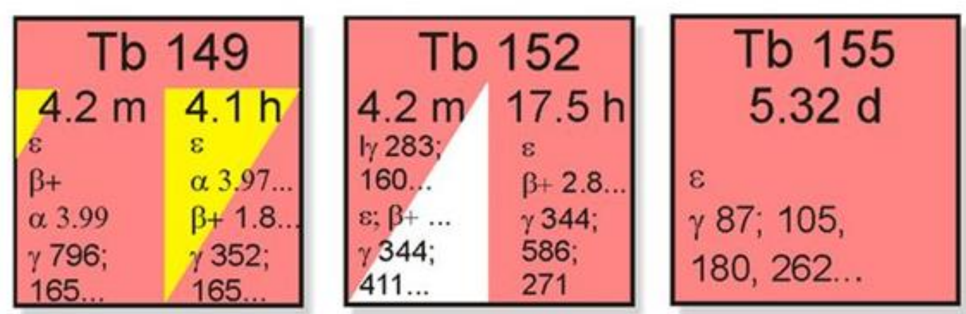


Need for highest precision nuclear decay data (e.g. $T_{1/2}$ values, $P_{\gamma}(\%)$) for standardisation….

- **<u>Chemical selectivity is really important</u>** for novel radionuclide standardisation - <u>See talk by Peter Ivanov</u>
- e.g. Separating A=155 into 'pure' ^{155}Tb from ^{139}Ce^{16}O



Isotope	$T_{1/2}$	Decay mode	Q-value	Main Gamma emissions (keV)	Application
^{149}Tb	4.118(25) h	α (16.7(14) %) ε+β⁺ (83.3(17) %)	4.0775(22) MeV 3.637(4) MeV	352 (29%) 165 (26%	α-therapy; PET
^{152}Tb	17.5(1) h	ε+β⁺ (100%)	3.990(40) MeV	344 (64%)	PET
^{155}Tb	5.32(6) d	ε (100 %)	820(10) keV	87 (32%) 105 (25%)	SPECT
^{161}Tb	6.89(2) d	β⁻ (100 %)	593.0(13) keV	49 (17%) 75 (10%)	β/auger-therapy; SPECT



SCIENTIFIC REPORTS

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Chemical Purification of Terbium-155 from Pseudo-Isobaric Impurities in a Mass Separated Source Produced at CERN

Received: 24 April 2019
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Ben Webster[1,2], Peter Ivanov[1], Ben Russell[1], Sean Collins[1], Thierry Stora[3], Joao Pedro Ramos[3,4], Ulli Köster[5], Andrew Paul Robinson[1,6,7] & David Read[1,2]

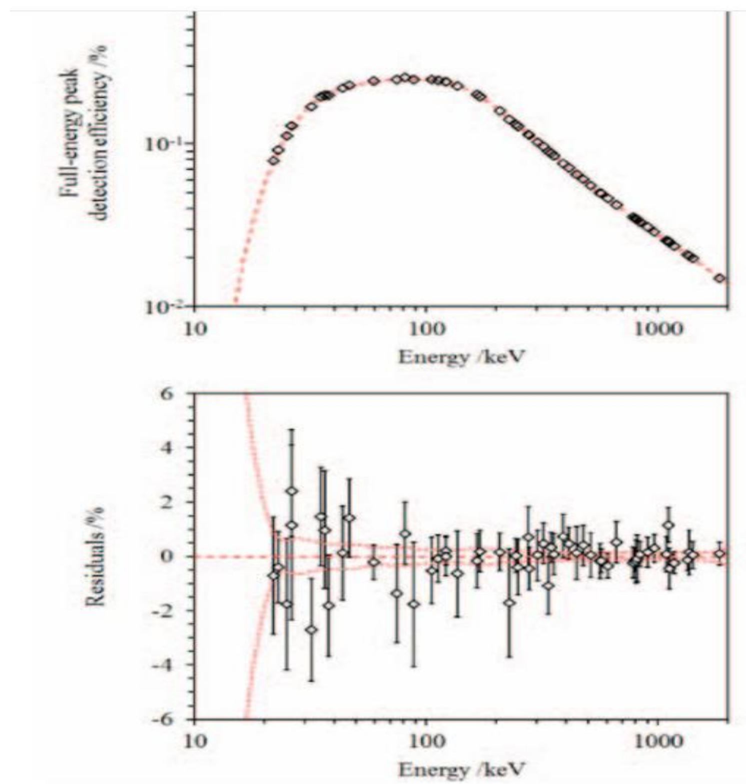
High resolution gamma spectrometry – in practice

The peak area N, gamma emission probability P_γ and full-energy peak detection efficiency ε_γ must all be precisely known.

$$A = \frac{N}{P_\gamma \, \varepsilon_\gamma \, t}$$

Considerations must also be given to;

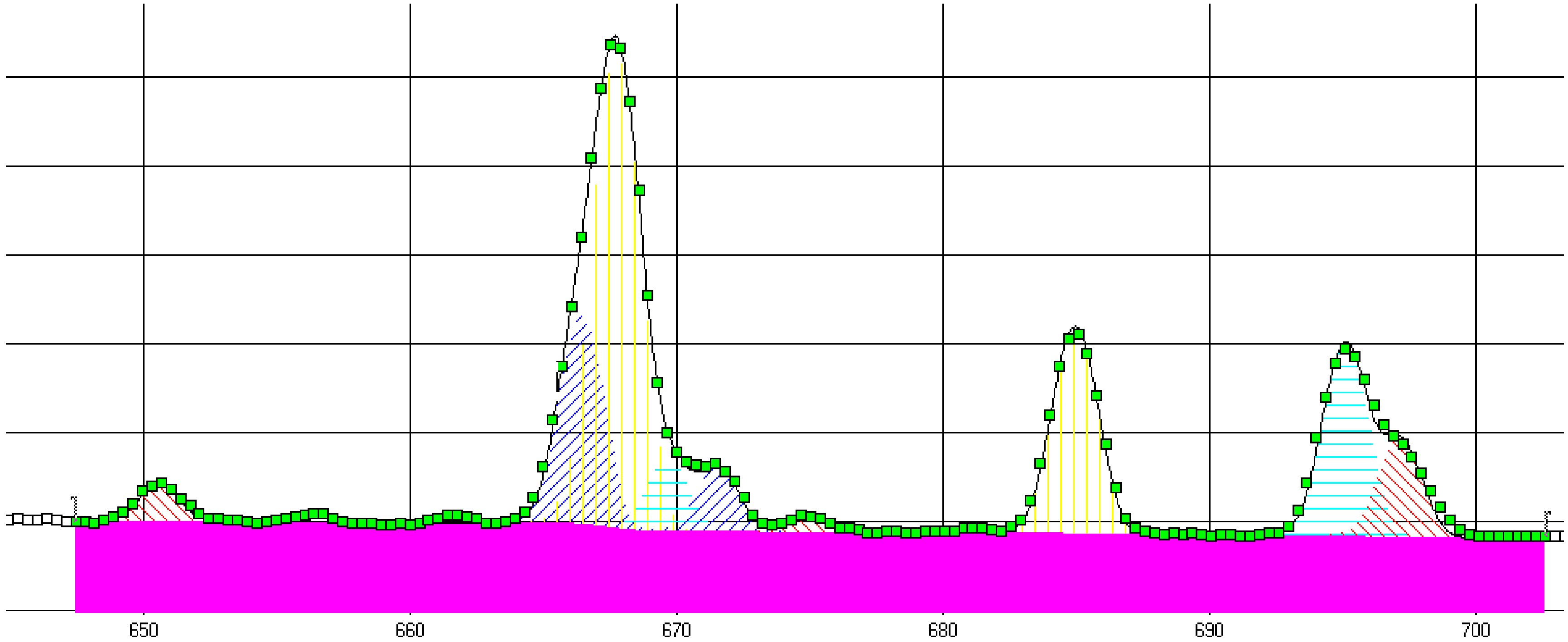
- sum corrections (in close geometry)
- dead times must be corrected
- in measurement decay of the nuclide.



Measure **an ampoule of primary standardized solution** (i.e. in Bq/ml).

Radionuclide Metrology and Standards in Nuclear Physics. *Nuclear Physics News*, *28*(3), (2018) 25–29

Peak fitting in gamma spectrometry



Determining the peak area N precisely can also be an issue – there are many complicating factors:

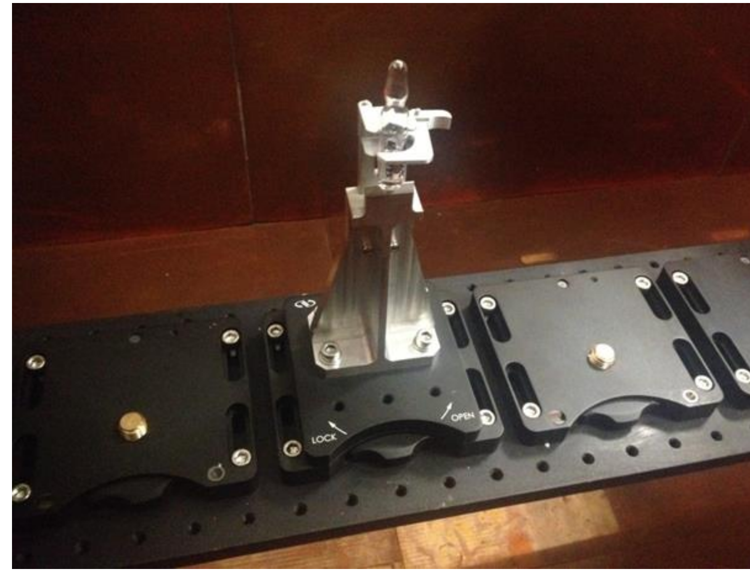
Convolution – two peaks close together;

Interference – two or more peaks at the same energy;

Background structures – such as backscatter peaks and Compton edges

Absolute γ-ray emission probabilities

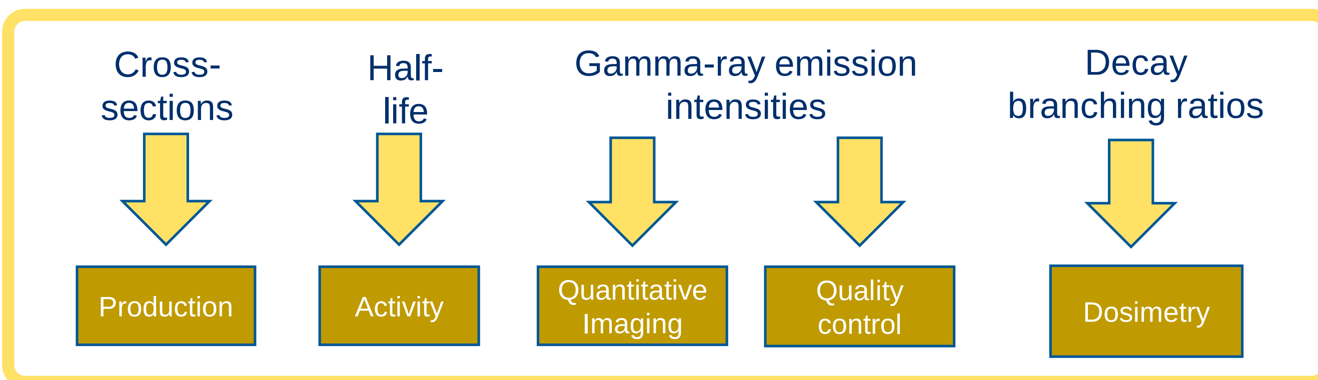
$$I_\gamma(E) = \frac{N(E)}{A_0 \cdot \varepsilon(E) \cdot t \cdot m} \cdot k_1 \cdot k_2 \cdot k_3 \cdot k_4 \cdot k_5$$



E	Energy of the γ-ray emission
I_γ	Emission probability of γ-ray
N	Net peak area of full-energy peak
A_0	Activity at the reference time
ε	Full-energy peak efficiency
t	Live time of measurement
m	Mass of solution
k_1	Correction for radioactive decay
k_2	Correction for pulse pile-up (aka Random summing)
k_3	Correction for true coincidence summing (TCS)
k_4	Correction for self-absorption in source
k_5	Correction for transient equilibrium (^{211}Pb onwards

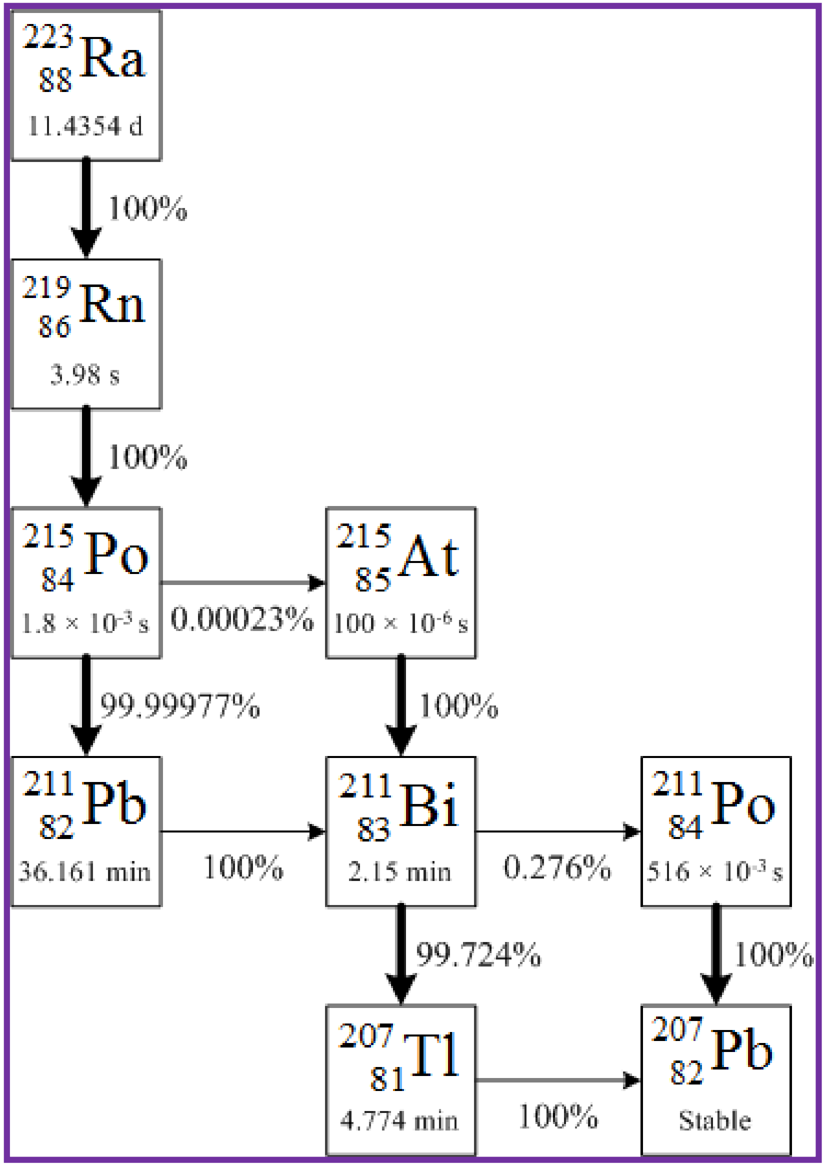
Measure an ampoule of primary standardized solution (i.e. in Bq/ml).
Measure gammas using a VERY well calibrated HPGe detector at different distances, and very well-defined geometries.

Importance of nuclear data in Nuclear Medicine



A ‘high impact’ example:

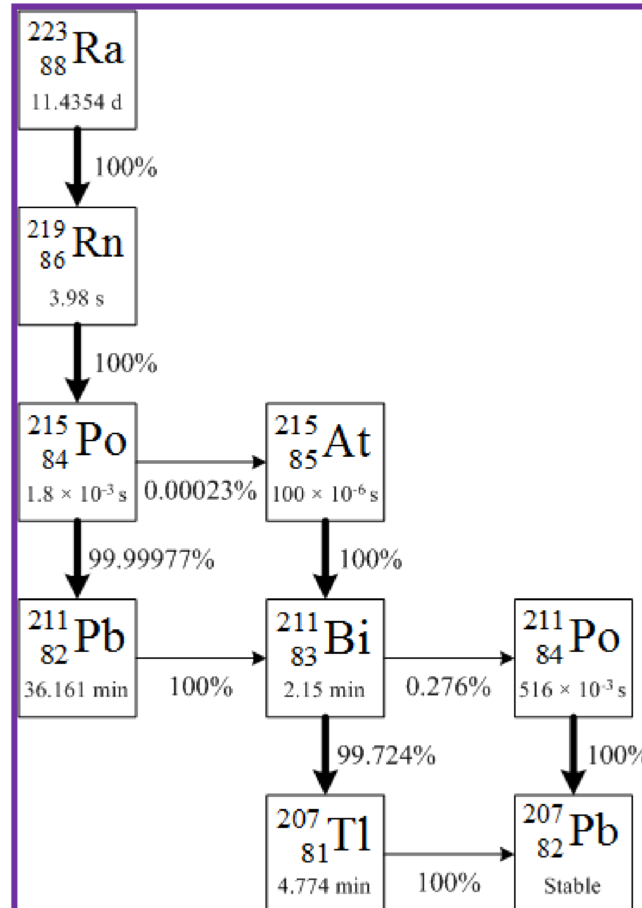
The standardisation of ^{223}Ra.



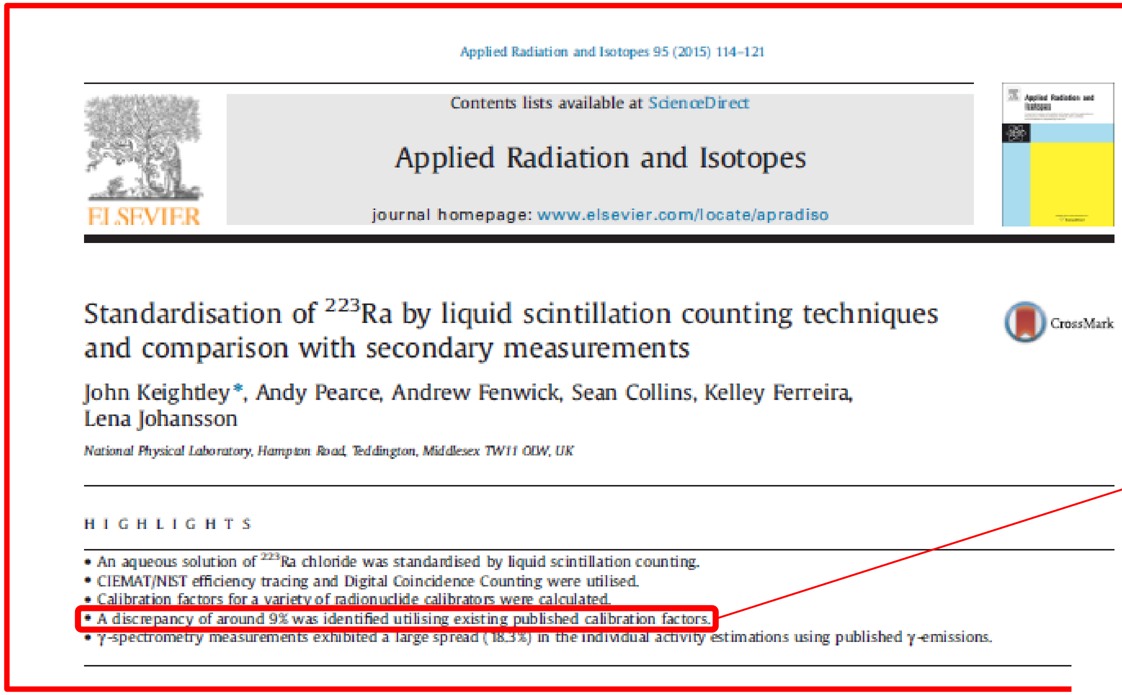
Primary Standards of α-Emitting Radiopharmaceuticals.

Alpha-emitting radionuclides have potential for treating tumours. $^{223}RaCl_2$ (Xofigo) was the first a-emitting drug approved by the US FDA.

Used in >3,000 clinics worldwide.



A problem…absolute ^{223}Ra standardisation at NPL



Applied Radiation and Isotopes 95 (2015) 114–121

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

Standardisation of ^{223}Ra by liquid scintillation counting techniques and comparison with secondary measurements

John Keightley*, Andy Pearce, Andrew Fenwick, Sean Collins, Kelley Ferreira, Lena Johansson

National Physical Laboratory, Hampton Road, Teddington, Middlesex TW11 0LW, UK

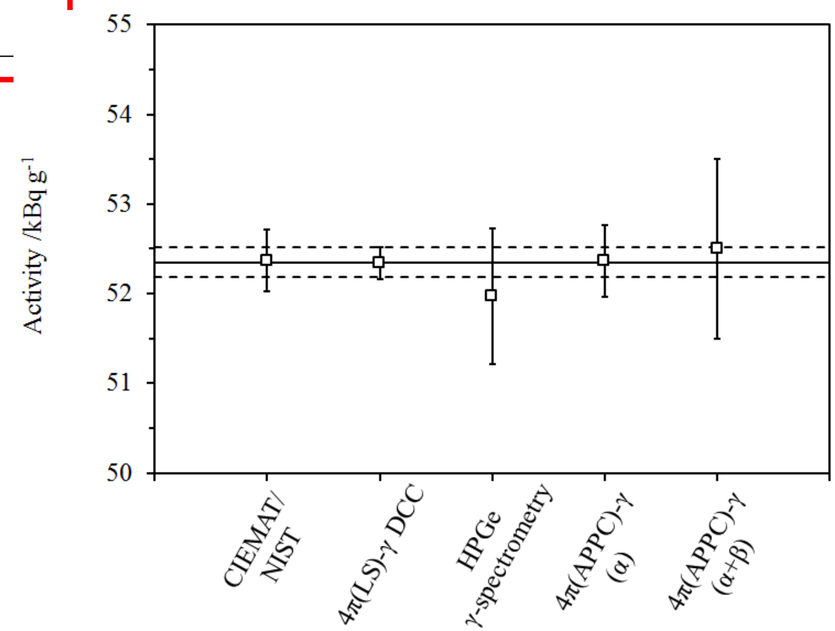
HIGHLIGHTS

- An aqueous solution of ^{223}Ra chloride was standardised by liquid scintillation counting.
- CIEMAT/NIST efficiency tracing and Digital Coincidence Counting were utilised.
- Calibration factors for a variety of radionuclide calibrators were calculated.
- A discrepancy of around 9% was identified utilising existing published calibration factors.
- γ-spectrometry measurements exhibited a large spread (18.3%) in the individual activity estimations using published γ-emissions.

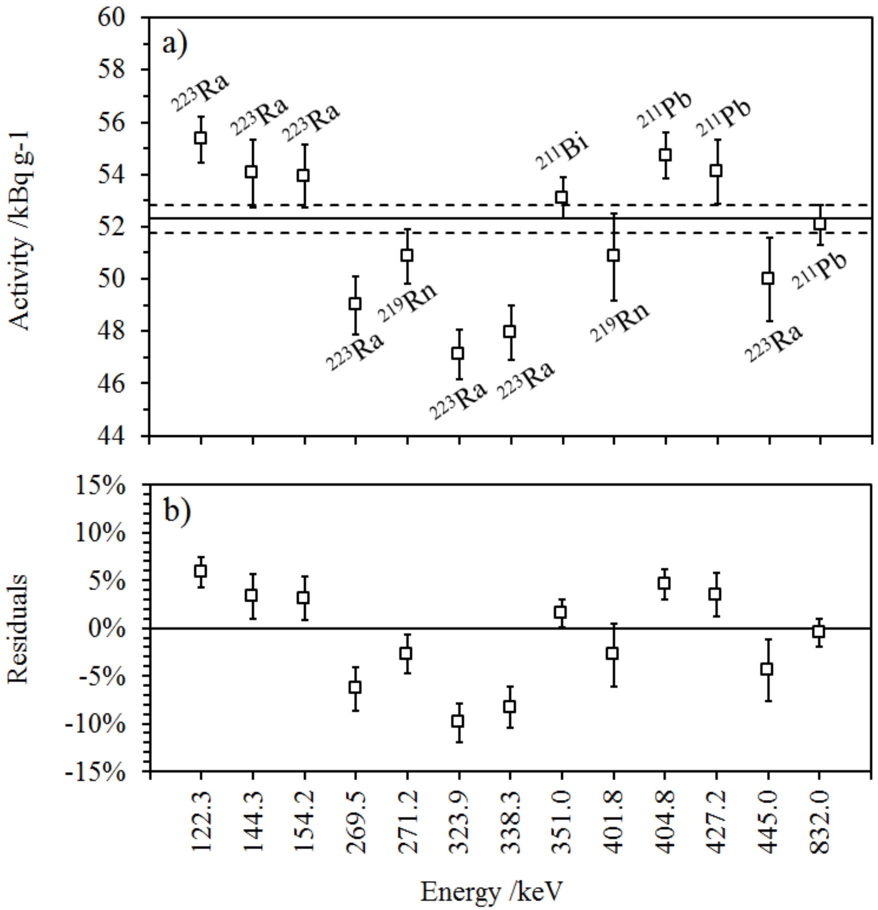
9% discrepancy from NIST

- Different techniques showed excellent agreement….
  - Including the γ-spectrometry.

- Do need new measurements of the γ-ray emission probabilities?



Large discrepancies in measurements with published γ-ray emission probabilities of ^{223}Ra.



- Main γ-ray emissions from ^{223}Ra using nucl data taken from DDEP.
- 18% range in deduced activity using different transitions. **This is not good enough.**
- However, the weighted mean gives the 'right' result.
- Previous results 'suspicious' due to the large spread.

Applied Radiation and Isotopes 99 (2015) 46–53

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NPL

Direct measurement of the half-life of ^{223}Ra

CrossMark

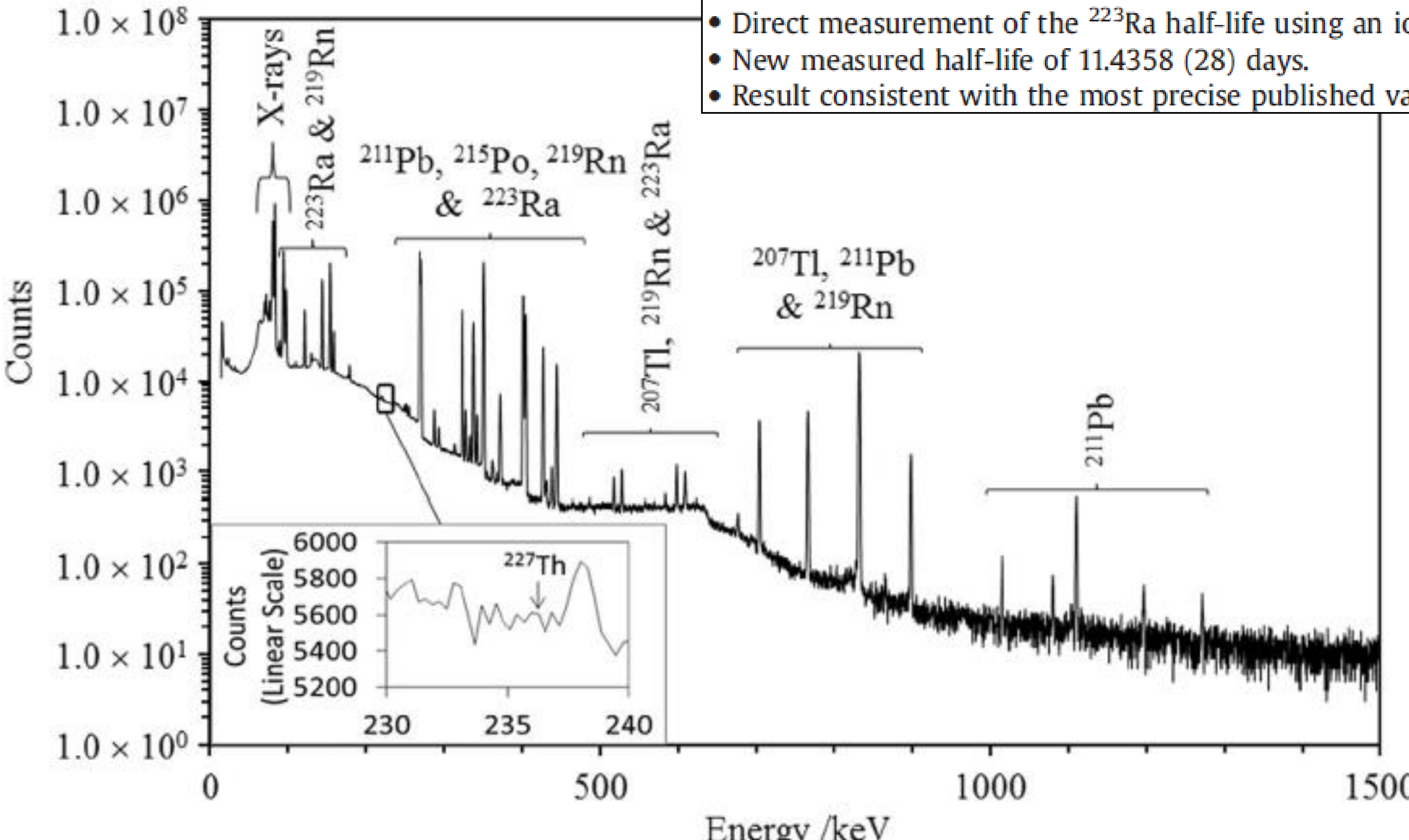
S.M. Collins [a,*], A.K. Pearce [a], K.M. Ferreira [a], A.J. Fenwick [a], P.H. Regan [a,b], J.D. Keightley [a]

[a] National Physical Laboratory, Hampton Road, Teddington, Middlesex TW11 0LW, United Kingdom

[b] Department of Physics, University of Surrey, Guildford, Surrey GU2 7XH, United Kingdom

HIGHLIGHTS

- Direct measurement of the ^{223}Ra half-life using an ionisation chamber.
- New measured half-life of 11.4358 (28) days.
- Result consistent with the most precise published value.



Applied Radiation and Isotopes 99 (2015) 46–53

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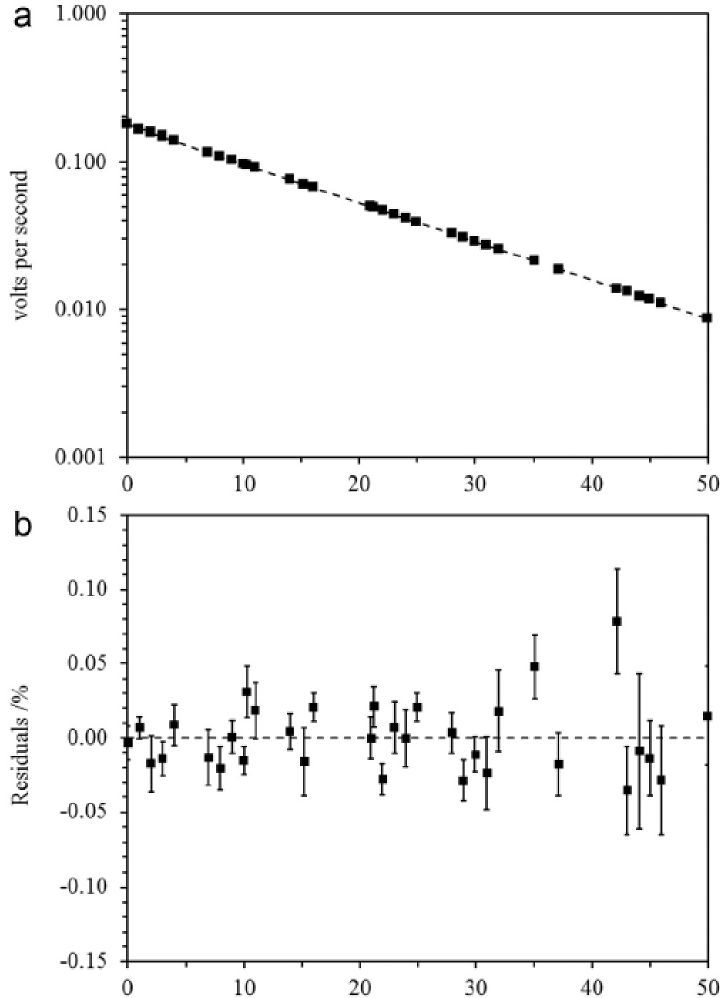
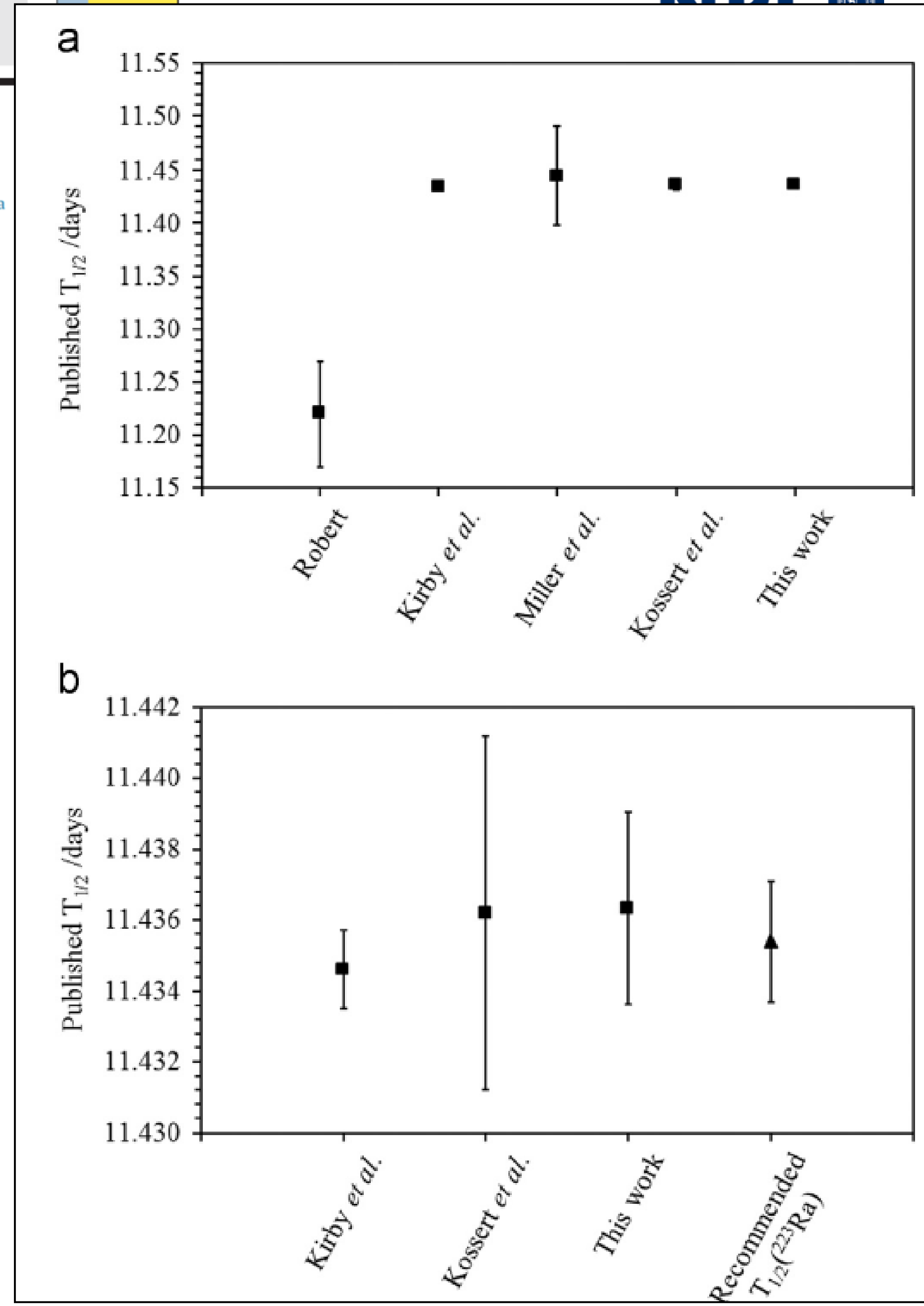
Direct measurement of the half-life of ^{223}Ra

S.M. Collins [a,*], A.K. Pearce [a], K.M. Ferreira [a], A.J. Fenwick [a], P.H. Regan [a,b], J.D. Keightley [a]

[a] National Physical Laboratory, Hampton Road, Teddington, Middlesex TW11 0LW, United Kingdom
[b] Department of Physics, University of Surrey, Guildford, Surrey GU2 7XH, United Kingdom





Applied Radiation and Isotopes 102 (2015) 15–28

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

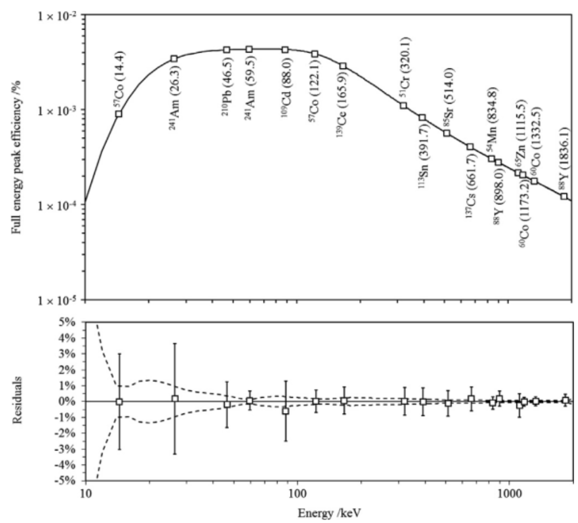
Precise measurements of the absolute γ-ray emission probabilities of ^{223}Ra and decay progeny in equilibrium

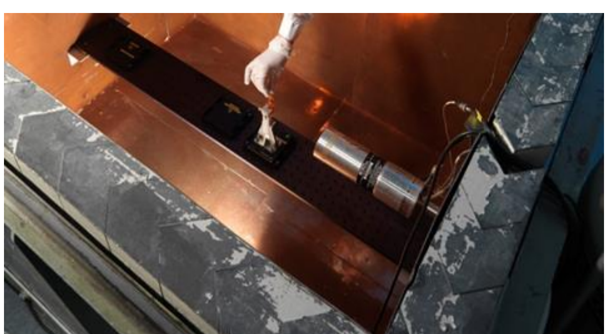
S.M. Collins [a,*], A.K. Pearce [a], P.H. Regan [a,b], J.D. Keightley [a]

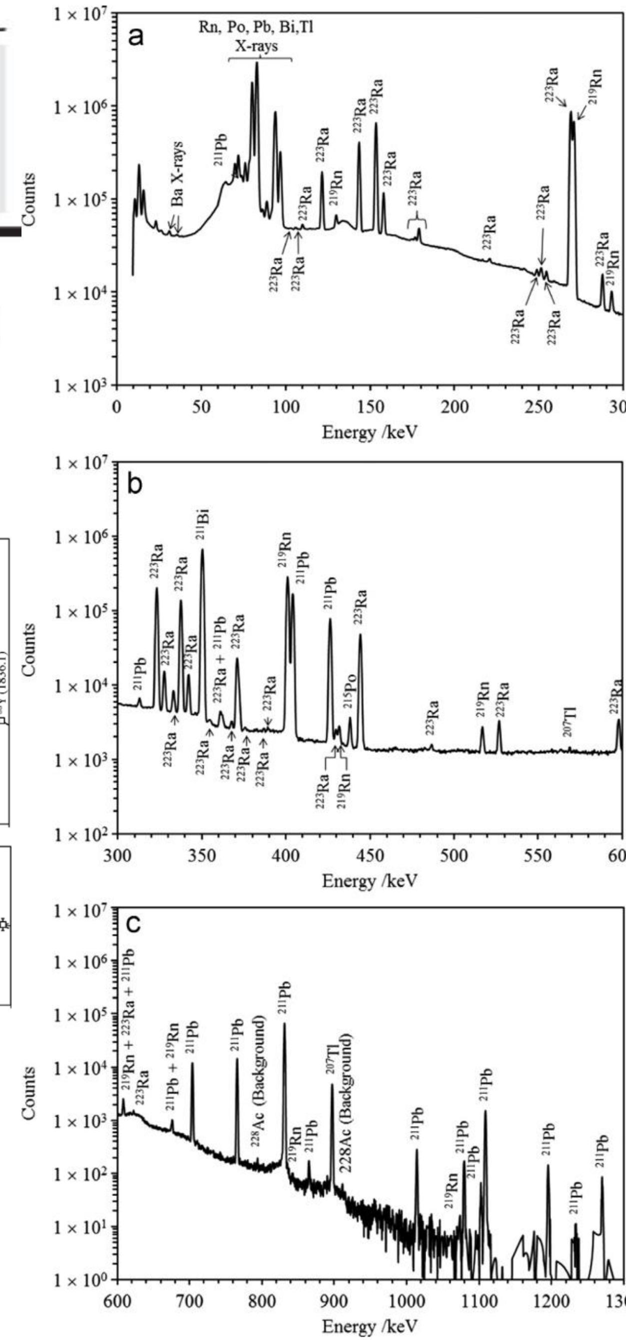
[a] National Physical Laboratory, Hampton Road, Teddington, Middlesex TW11 0LW, United Kingdom
[b] Department of Physics, University of Surrey, Guildford, Surrey GU2 7XH, United Kingdom

Table 3
Full-energy peak efficiency calibration points for the HPGe γ-ray spectrometers BART and LORAX.

Energy/keV	Radionuclide	Full-energy peak efficiency /%	
		BART	LORAX
14.41	^{57}Co	$2.96(10) \times 10^{-2}$	$9.0(3) \times 10^{-2}$
26.24	^{241}Am	$1.29(5) \times 10^{-1}$	$3.44(12) \times 10^{-1}$
35.49	^{125}I	$1.52(4) \times 10^{-1}$	–
46.54	^{210}Pb	$1.647(23) \times 10^{-1}$	$4.26(6) \times 10^{-1}$
59.54	^{241}Am	$1.674(10) \times 10^{-1}$	$4.33(3) \times 10^{-1}$
88.03	^{109}Cd	$1.67(4) \times 10^{-1}$	$4.29(8) \times 10^{-1}$
122.06	^{57}Co	$1.601(11) \times 10^{-1}$	$3.88(3) \times 10^{-1}$
136.47	^{57}Co	$1.537(24) \times 10^{-1}$	$3.54(6) \times 10^{-1}$
165.86	^{139}Ce	$1.416(12) \times 10^{-1}$	$2.893(25) \times 10^{-1}$
320.08	^{51}Cr	$8.54(6) \times 10^{-2}$	$1.107(10) \times 10^{-1}$
391.70	^{113}Sn	$7.16(6) \times 10^{-2}$	$8.23(7) \times 10^{-2}$
514.00	^{85}Sr	$5.61(5) \times 10^{-2}$	$5.63(5) \times 10^{-2}$
661.66	^{137}Cs	$4.53(4) \times 10^{-2}$	$4.06(3) \times 10^{-2}$
834.84	^{54}Mn	$3.716(14) \times 10^{-2}$	$3.038(12) \times 10^{-2}$
898.04	^{88}Y	$3.499(20) \times 10^{-2}$	$2.793(13) \times 10^{-2}$
1115.54	^{65}Zn	$2.929(22) \times 10^{-2}$	$2.162(16) \times 10^{-2}$
1173.23	^{60}Co	$2.822(8) \times 10^{-2}$	$2.045(6) \times 10^{-2}$
1332.49	^{60}Co	$2.534(14) \times 10^{-2}$	$1.770(5) \times 10^{-2}$
1836.05	^{88}Y	$1.93(9) \times 10^{-2}$	$1.218(5) \times 10^{-2}$







Most up to date, accurate data on ^{223}Ra decay.

NPL

Applied Radiation and Isotopes 102 (2015) 15–28

Precise measurements of the absolute γ-ray emission probabilities of ^{223}Ra and decay progeny in equilibrium

CrossMark

S.M. Collins [a,*], A.K. Pearce [a], P.H. Regan [a,b], J.D. Keightley [a]

[a] National Physical Laboratory, Hampton Road, Teddington, Middlesex TW11 0LW, United Kingdom
[b] Department of Physics, University of Surrey, Guildford, Surrey GU2 7XH, United Kingdom

Table 4
Absolute γ-ray emission probabilities per 100 decays of ^{223}Ra and decay progeny in equilibrium.

Energy (keV)	Source	I_γ (%)	Energy (keV)	Source	I_γ (%)	Energy (keV)	Source	I_γ (%)	Energy (keV)	Source	I_γ (%)
103.9(5)	^{223}Ra	0.0119(6)	323.9(6)	^{223}Ra	3.655(18)	438.8(6)	^{215}Po	0.0533(7)	675.4(6)	^{211}Pb	0.0058(6)
106.7(4)	^{223}Ra	0.0213(11)	328.4(6)	^{223}Ra, ^{211}Po, ^{207}Tl	0.2021(16)	445.0(6)	^{223}Ra	1.218(6)	676.9(6)	^{219}Rn	0.0184(5)
110.8(5)	^{223}Ra	0.0512(10)	333.9(6)	^{223}Ra	0.0756(6)	462.8(6)	^{219}Rn	0.0011(5)	704.6(7)	^{211}Pb	0.498(3)
122.3(5)	^{223}Ra	1.312(6)	338.3(6)	^{223}Ra	2.605(13)	487.3(5)	^{223}Ra	0.0083(3)	707.8(7)	^{219}Rn	0.0034(4)
130.6(5)	^{219}Rn	0.1478(10)	342.9(6)	^{223}Ra, ^{211}Pb	0.1958(21)	500.2(6)	^{223}Ra	0.0013(5)	711.4(7)	^{223}Ra	0.0037(3)
144.3(5)	^{223}Ra	3.481(16)	351.1(6)	^{211}Bi	13.17(7)	504.1(6)	^{211}Pb	0.0022(4)	727.4(7)	^{223}Ra	0.0024(7)
154.2(5)	^{223}Ra	6.02(3)	355.5(6)	^{223}Ra	0.0124(15)	517.6(6)	^{219}Rn	0.0453(5)	766.4(7)	^{211}Pb	0.685(4)
158.7(5)	^{223}Ra	0.749(4)	361.7(6)	^{211}Pb	0.0341(7)	522.6(6)	^{223}Ra	0.0021(6)	831.9(7)	^{211}Pb	3.448(16)
175.6(5)	^{223}Ra	0.01578(10)	363.0(6)	^{223}Ra	0.0192(9)	527.6(6)	^{223}Ra	0.0659(8)	835.6(7)	^{219}Rn	0.00364(19)
177.4(5)	^{223}Ra	0.0426(8)	368.4(6)	^{223}Ra	0.0134(4)	531.4(6)	^{223}Ra	0.0028(9)	865.8(6)	^{211}Pb	0.00540(21)
179.7(5)	^{223}Ra	0.1613(10)	371.7(6)	^{223}Ra	0.435(3)	537.5(6)	^{223}Ra	0.0033(6)	891.3(7)	^{219}Rn	0.00107(20)
221.4(5)	^{223}Ra	0.0304(10)	372.9(6)	^{223}Ra	0.1133(13)	542.1(6)	^{223}Ra	0.0026(6)	897.8(7)	^{211}Po, ^{207}Tl	0.2725(15)
224.0(5)	^{219}Rn	0.0056(14)	376.2(6)	^{223}Ra	0.0056(4)	545.9(6)	^{223}Ra	0.0028(6)	1014.7(7)	^{211}Pb	0.0171(4)
249.4(5)	^{223}Ra	0.0375(9)	383.3(5)	^{223}Ra	0.0023(6)	555.9(5)	^{219}Rn	0.0026(7)	1074.5(7)	^{219}Rn	0.00044(12)
251.9(5)	^{223}Ra	0.0640(11)	386.3(5)	^{223}Ra, ^{219}Rn	0.0052(7)	564.4(5)	^{219}Rn	0.0035(4)	1080.1(7)	^{211}Pb	0.01228(21)
255.1(5)	^{223}Ra	0.0499(13)	390.1(5)	^{223}Ra	0.0053(7)	569.6(7)	^{211}Po, ^{207}Tl	0.0043(5)	1103.3(8)	^{211}Pb	0.00380(12)
269.5(6)	^{223}Ra	13.37(7)	401.8(6)	^{219}Rn	6.57(3)	573.7(7)	^{223}Ra	0.0029(13)	1109.5(8)	^{211}Pb	0.1113(7)
271.3(6)	^{219}Rn	10.75(6)	404.8(6)	^{211}Pb, ^{215}At	4.011(19)	598.6(7)	^{223}Ra	0.0867(12)	1196.2(8)	^{211}Pb	0.01052(17)
288.2(6)	^{223}Ra	0.1498(16)	427.1(6)	^{211}Pb	1.890(9)	609.3(7)	^{223}Ra, ^{219}Rn, ^{211}Pb	0.0543(7)	1234.3(8)	^{211}Pb	0.00092(8)
293.6(5)	^{219}Rn	0.0688(7)	430.4(6)	^{223}Ra, ^{211}Pb	0.0206(19)	619.8(6)	^{219}Rn	0.0056(12)	1270.7(8)	^{211}Pb	0.00624(19)
313.7(6)	^{211}Pb	0.0276(5)	432.4(6)	^{223}Ra	0.0297(14)	623.4(5)	^{223}Ra	0.0082(8)			

Comparison of NPL & (new) NIST and PTB Data.

Radionuclide	Energy /keV	I_γ(NPL) %	I_γ(NIST) %	I_γ(PTB) %	$\chi^2/(n-1)$
^{223}Ra	122.3	1.312 (6)	1.30 (1)	1.304 (12)	0.3
^{223}Ra	144.3	3.481 (16)	3.51 (3)	3.469 (20)	0.3
^{223}Ra	154.2	6.02 (3)	6.08 (6)	6.03 (5)	0.2
^{223}Ra	269.5	13.37 (7)	13.24 (12)	13.16 (15)	0.5
^{223}Ra	323.9	3.655 (18)	3.63 (2)	3.661 (21)	0.3
^{223}Ra	338.3	2.605 (13)	2.59 (2)	2.614 (13)	0.3
^{223}Ra	445.0	1.218 (6)	1.217 (8)	1.222 (6)	0.1
^{219}Rn	271.2	10.75 (6)	10.69 (10)	10.87 (12)	0.3
^{219}Rn	401.8	6.57 (3)	6.56 (4)	6.62 (4)	0.3
^{211}Pb	404.8	4.011 (19)	4.01 (3)	4.05 (5)	0.1
^{211}Pb	427.2	1.890 (9)	1.89 (1)	1.912 (10)	0.8
^{211}Pb	832.0	3.448 (16)	3.48 (3)	3.430 (17)	0.5
^{211}Bi	351.0	13.17 (7)	13.11 (9)	13.24 (6)	0.4
No. of γ-rays reported		83	15	43	

Now, good agreement between the three main NMIs ☺.

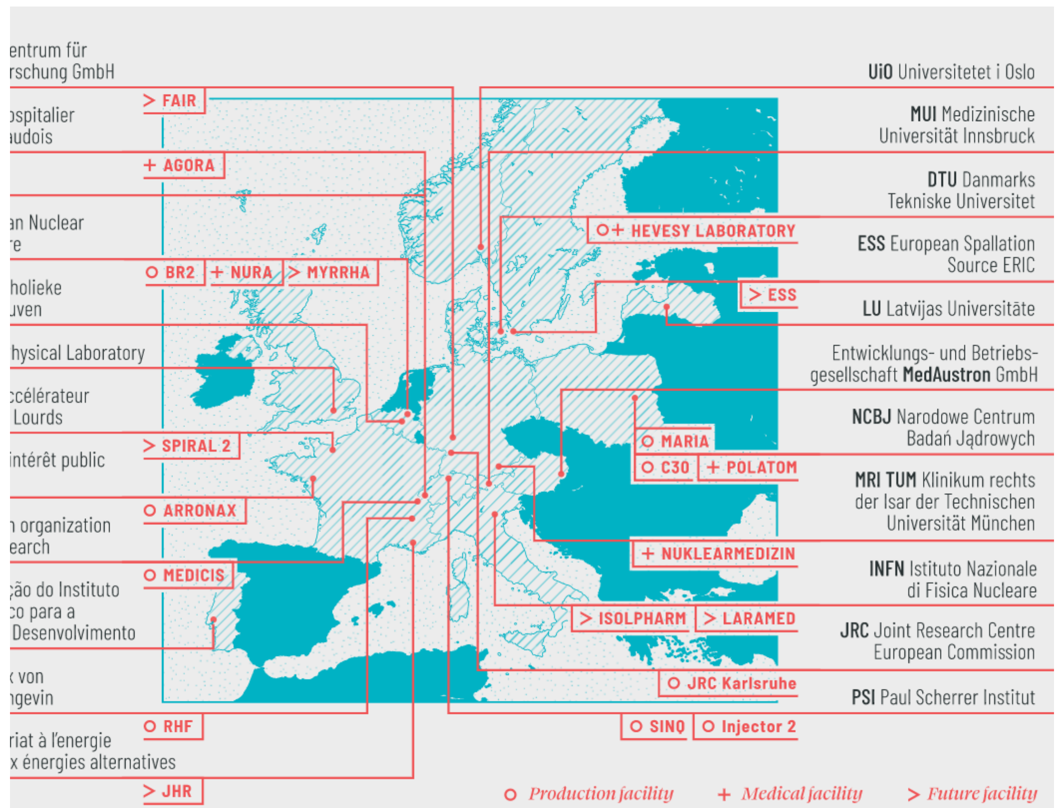
PRoduction of high purity Isotopes by mass Separation for Medical Applications

- Suite of medically relevant nuclides produced and being studied (often with sparse Nuclear Data)

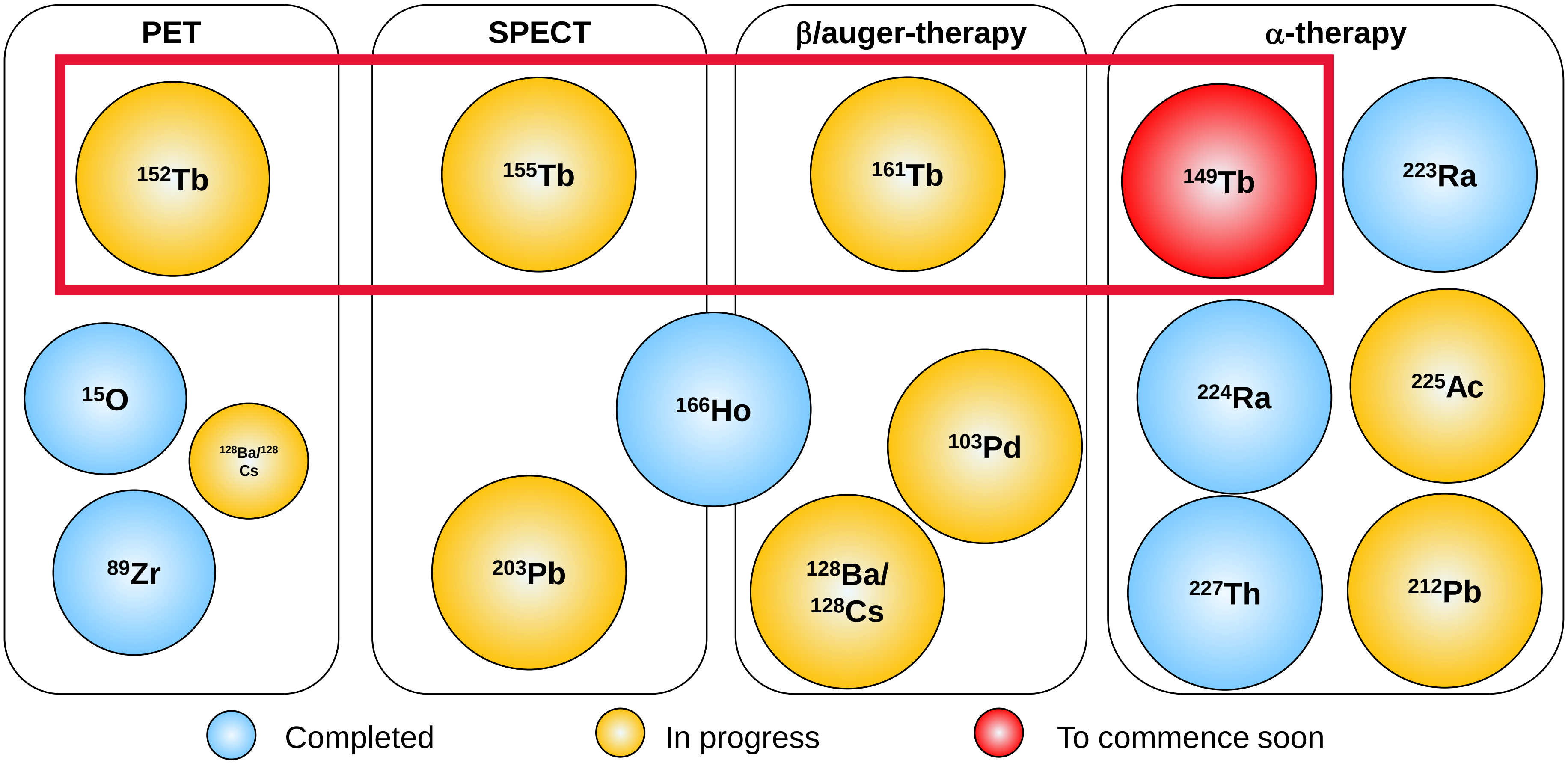
- 23 Partners from 13 countries
  - 2 National measurement Institutes

Our objectives are:

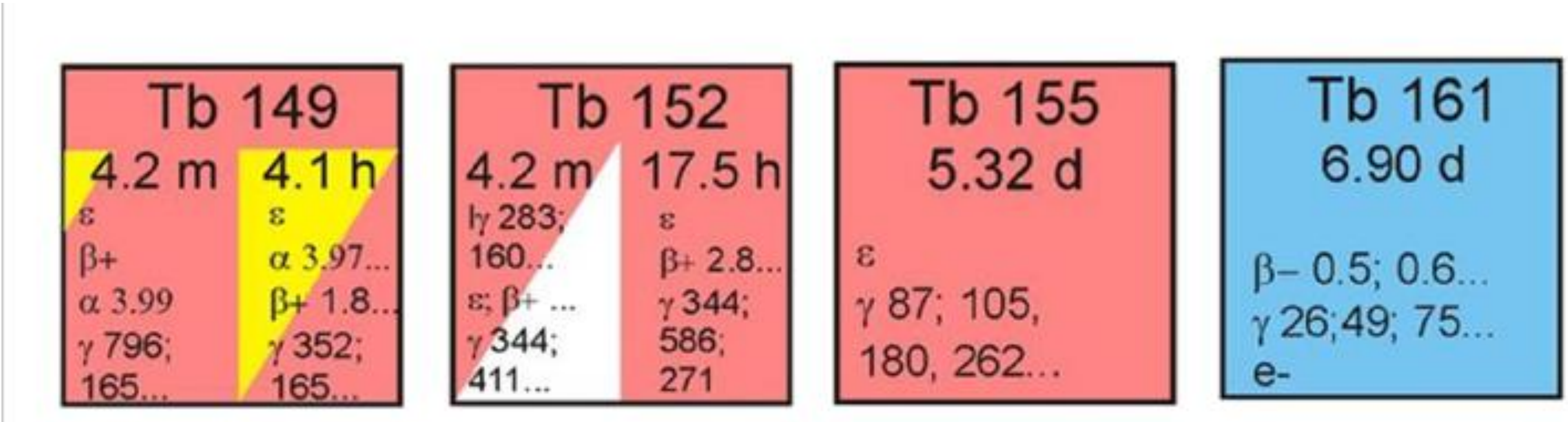
- Provide access to new radionuclides and new purity grades for the medical research
- Create a common entry port and web interface to the starting research community
- Enhance clarity and regulatory procedures to enhance research with radiopharmaceuticals
- Improve the delivered radionuclide data and regulation, along with biomedical research capacity
- Ensure sustainability of PRISMAP on the long term



Medical radionuclides primary standardisations at NPL since 2015



The Terbium Toolbox

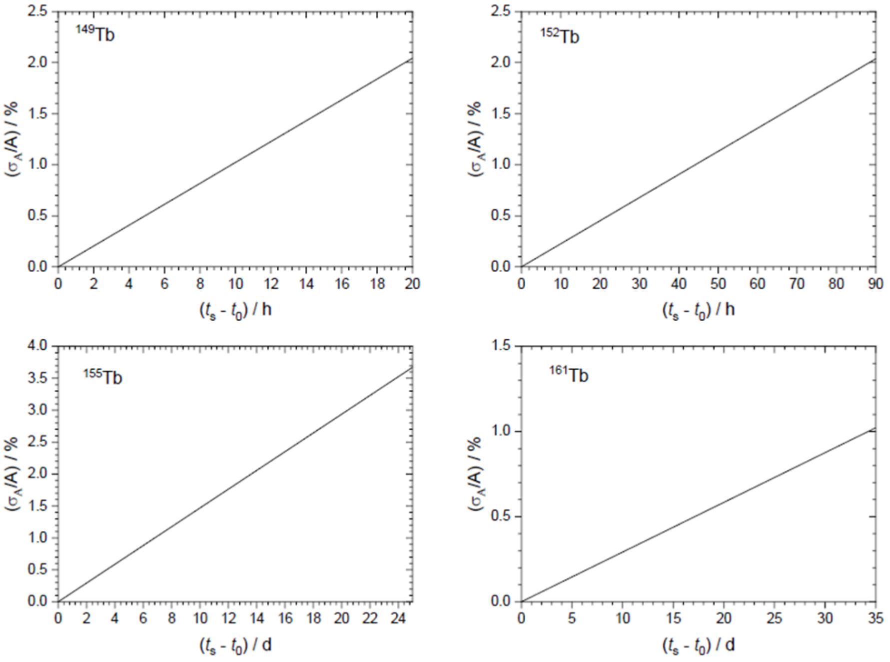


Isotope	$T_{1/2}$	Decay mode	Q-value	Main Gamma emissions (keV)	Application
^{149}Tb	4.118(25) h	α (16.7(14) %) ε+β$^+$ (83.3(17) %)	4.0775(22) MeV 3.637(4) MeV	352 (29%) 165 (26%	α-therapy; PET
^{152}Tb	17.5(1) h	ε+β$^+$ (100%)	3.990(40) MeV	344 (64%)	PET
^{155}Tb	5.32(6) d	ε (100 %)	820(10) keV	87 (32%) 105 (25%)	SPECT
^{161}Tb	6.89(2) d	β$^-$ (100 %)	593.0(13) keV	49 (17%) 75 (10%)	β/auger-therapy; SPECT

The half-lives of the Tb quartet before our work…

Radionuclide	$T_{1/2}$	$u(T_{1/2})/T_{1/2}$
^{149}Tb	(4.118 ± 0.025) h	0.61 %
^{152}Tb	(17.5 ± 0.1) h	0.57 %
^{155}Tb	(5.32 ± 0.06) d	1.1 %
^{161}Tb	(6.89 ± 0.02) d	0.29 %

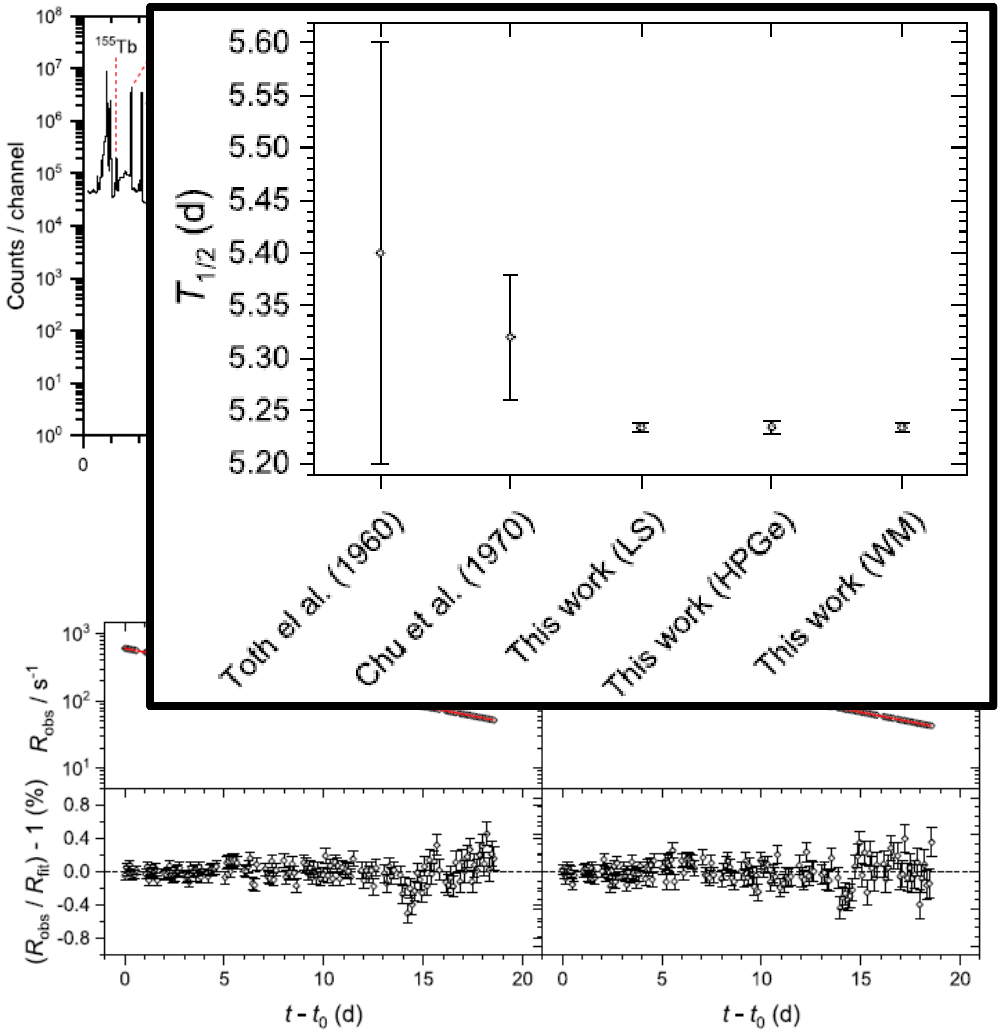
$$\left(\frac{\sigma_A}{A}\right)^2 = \left(\lambda t_d + \lambda t_m + \frac{1 - \left(\frac{1 - e^{-\lambda t_m}}{\lambda t_m}\right)}{\frac{1 - e^{-\lambda t_m}}{\lambda t_m}}\right)^2 \left(\frac{\sigma_{T_{1/2}}}{T_{1/2}}\right)^2$$

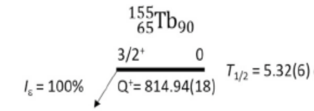


Sean Collins, Rob Shearman et al., (NPL)

Chemical selectivity is really important for novel radionuclides e.g. Separating ‘pure’ ^{155}Tb from ^{139}Ce^{16}O

$$R_t(R_0,\lambda) = R_0 \cdot e^{-\lambda t} \cdot \frac{(1-e^{-\lambda \Delta t})}{\lambda \Delta t}$$

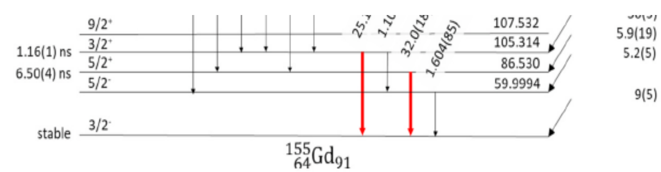




$T_{1/2}(^{155}Tb)$=5.2346(36) days

1.6% relative difference to evaluated Value of 5.32(6) days.

Order of magnitude improvement in the precision.



Contents lists available at ScienceDirect

Applied Radiation and Isotopes 190 (2022) 110480

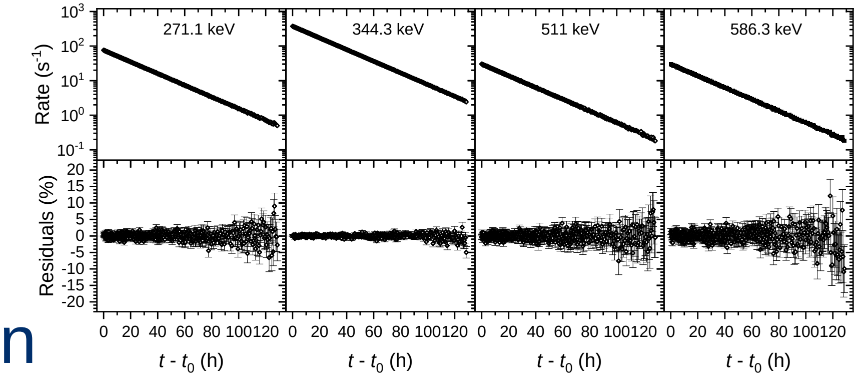
Half-life determination of ^{155}Tb from mass-separated samples produced at CERN-MEDICIS

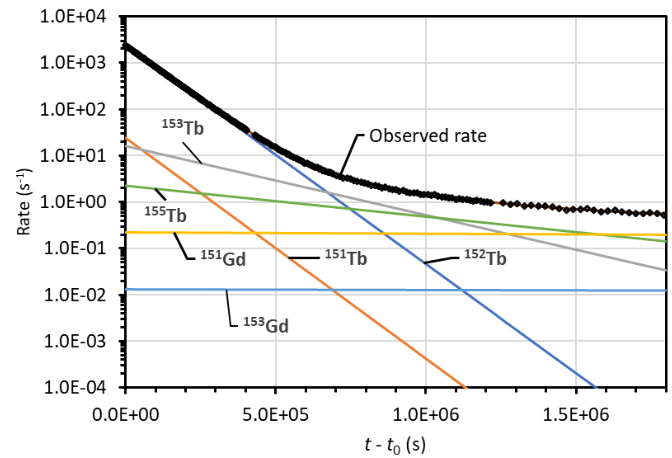
S.M. Collins [a,b,*], A.P. Robinson [a,c,d], P. Ivanov [a], U. Köster [e], T.E. Cocolios [f], B. Russell [a], B. Webster [a,b], A.J. Fenwick [a], C. Duchemin [f,g], J.P. Ramos [f,g,h], E. Chevallay [g], U. Jakobsson [i,j], S. Stegemann [f], P.H. Regan [a,b], T. Stora [g]

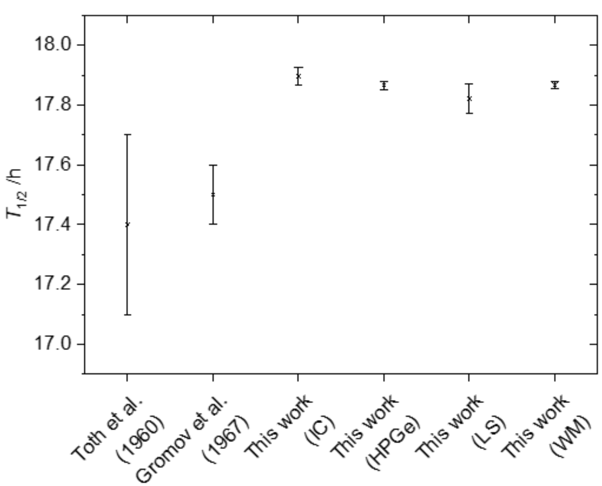
Half-life of ^{152}Tb

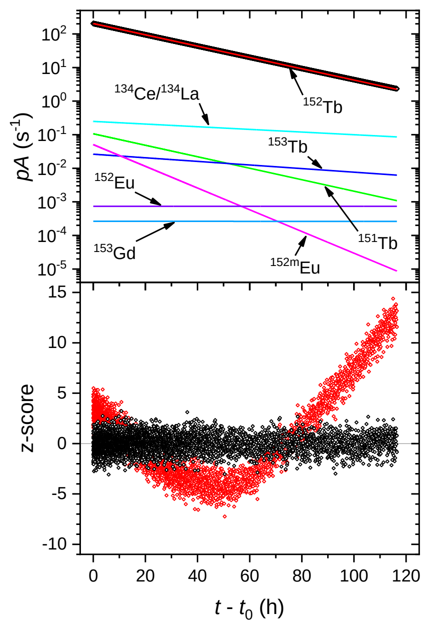
$T_{1/2}$= 17.867(11) h

- 2.1 % relative difference to evaluated half-life of 17.5(1) d
- x10 improvement in the precision
- IC & LS affected by contaminants – results in poor precision vs HPGe

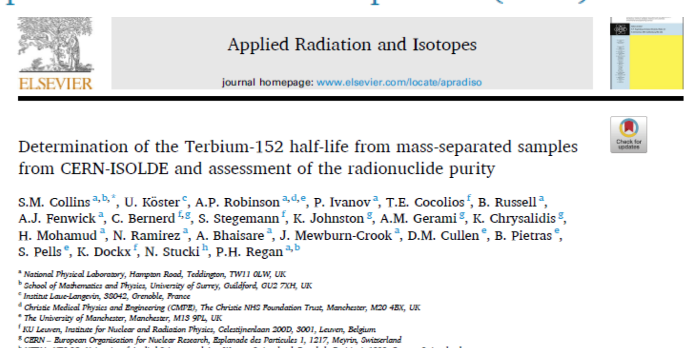








Applied Radiation and Isotopes 202 (2023) 111044



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Applied Radiation and Isotopes
journal homepage: www.elsevier.com/locate/apradiso

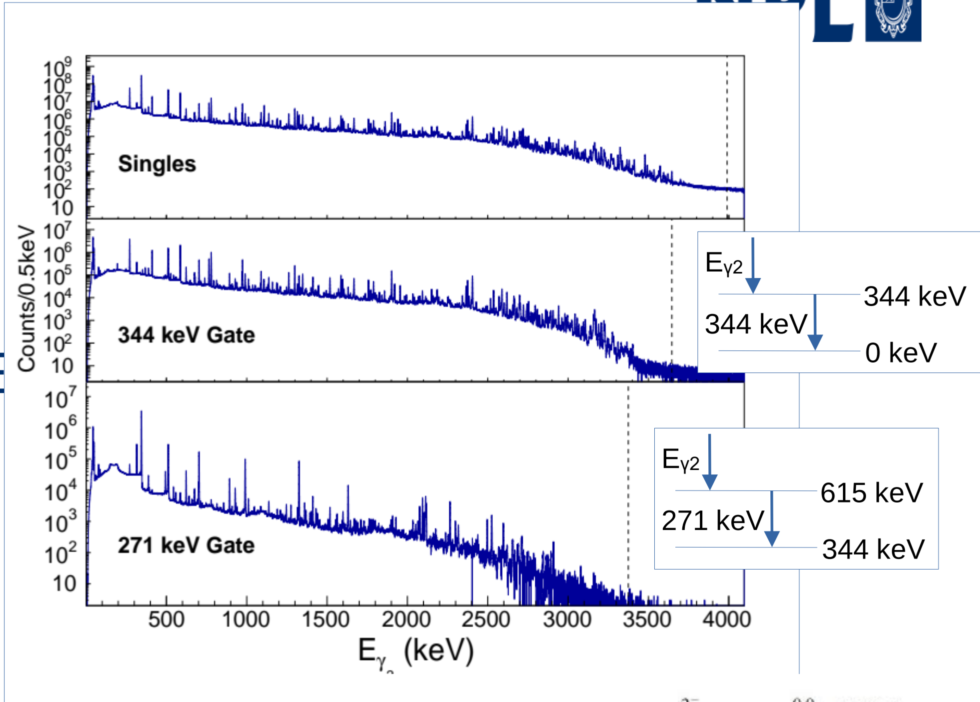
Determination of the Terbium-152 half-life from mass-separated samples from CERN-ISOLDE and assessment of the radionuclide purity

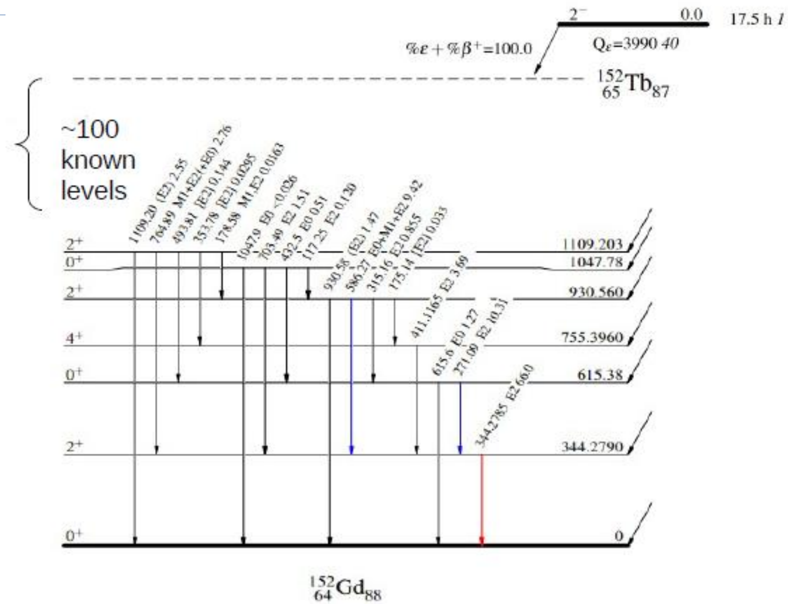
S.M. Collins [a,b,*], U. Köster [c], A.P. Robinson [a,d,e], P. Ivanov [a], T.E. Cocolios [f], B. Russell [a], A.J. Fenwick [a], C. Bernerd [f,g], S. Stegemann [f], K. Johnston [g], A.M. Gerami [g], K. Chrysalidis [g], H. Mohamud [a], N. Ramirez [a], A. Bhaisare [a], J. Mewburn-Crook [a], D.M. Cullen [e], B. Pietras [e], S. Pells [e], K. Dockx [f], N. Stucki [h], P.H. Regan [a,b]

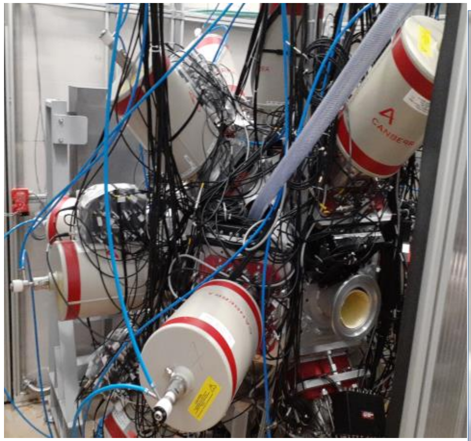
[a] National Physical Laboratory, Hampton Road, Teddington, TW11 0LW, UK
[b] School of Mathematics and Physics, University of Surrey, Guildford, GU2 7XH, UK
[c] Institut Laue-Langevin, 38042, Grenoble, France
[d] Christie Medical Physics and Engineering (CMPE), The Christie NHS Foundation Trust, Manchester, M20 4BX, UK
[e] The University of Manchester, Manchester, M13 9PL, UK
[f] KU Leuven, Institute for Nuclear and Radiation Physics, Celestijnenlaan 200D, 3001, Leuven, Belgium
[g] CERN – European Organisation for Nuclear Research, Esplanade des Particules 1, 1217, Meyrin, Switzerland
[h] HEPIA, HES-SO, University of Applied Sciences and Arts Western Switzerland, Rue de la Prairie 4, 1202, Geneva, Switzerland

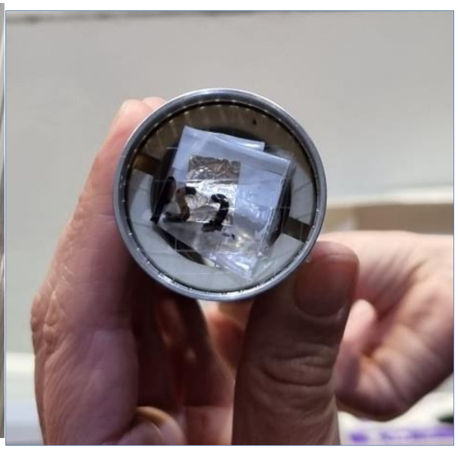
$^{152}Tb \rightarrow ^{152}Gd$ Decay spectroscopy: talk by Ed O'Sullivan.

- Poorly understood, beta feeding of high excitation energy states close to Q-value window ~3.9 MeV not known (Pandemonium)
- Sources made at CERN-ISOLDE (proton on Tantalum target)
- Measured decay gammas at ILL using the FIPS HPGe array
  - New transitions, new levels
  - Angular correlations.

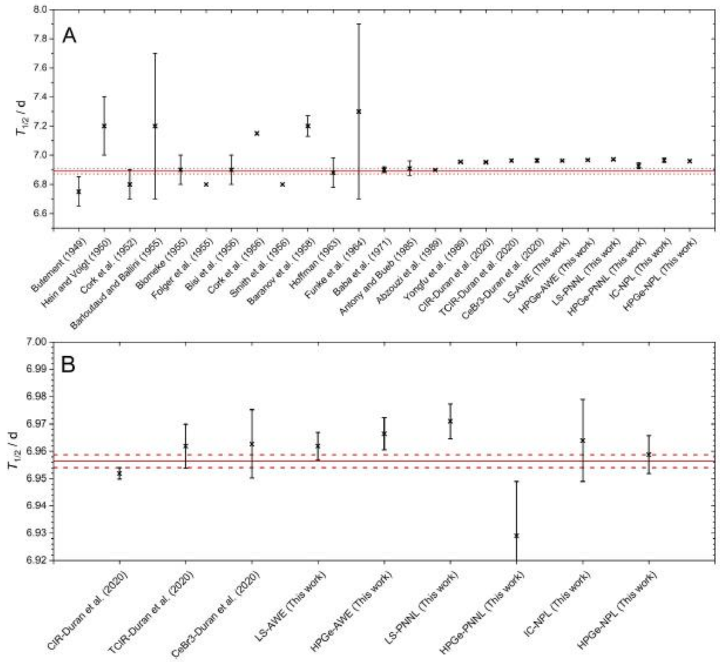


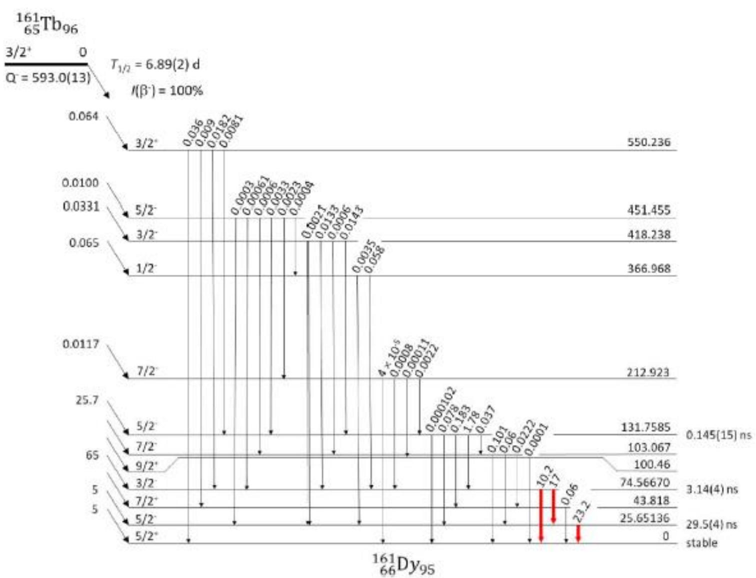






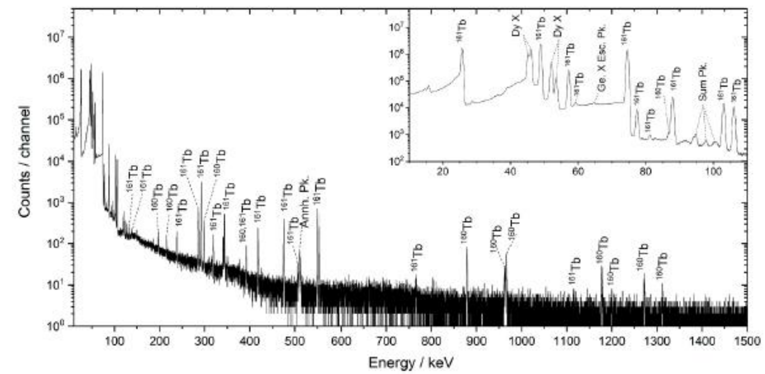
Half-life of ^{161}Tb





$T_{1/2}$ = 6.9637(29) d

- 6 measurements from 2 laboratories
- 1 % diff. to evaluated $T_{1/2}$ of 6.89(2) d
- x7 improvement in the precision.



Applied Radiation and Isotopes 182 (2022) 110140

Applied Radiation and Isotopes
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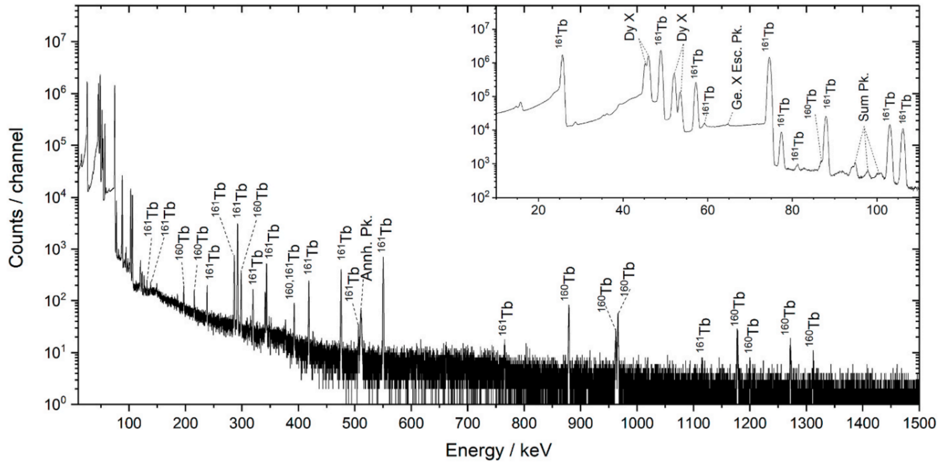
journal homepage: www.elsevier.com/locate/apradiso

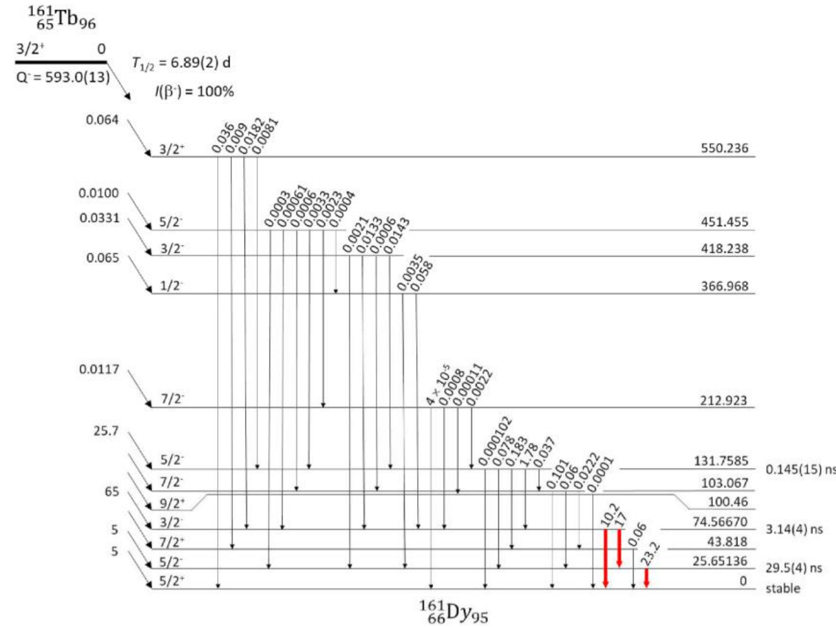
Determination of the ^{161}Tb half-life

S.M. Collins [a,b,*], C. Gilligan [c], B. Pierson [d], N. Ramirez [a], M. Goodwin [b,c], A.K. Pearce [a], B. C. Archambault [d], M.M. Haney [d], P.H. Regan [a,b]

[a] National Physical Laboratory, Hampton Road, Teddington, Middlesex, TW11 0LW, United Kingdom
[b] Department of Physics, University of Surrey, Stag Hill, Guildford, GU2 7XH, United Kingdom
[c] Atomic Weapons Establishment, Aldermaston, Reading, Berkshire, RG7 4PR, United Kingdom
[d] Pacific Northwest National Laboratory, 902 Battelle Blvd, Richland, WA, 99354, United States

^{161}Tb decay spectroscopy

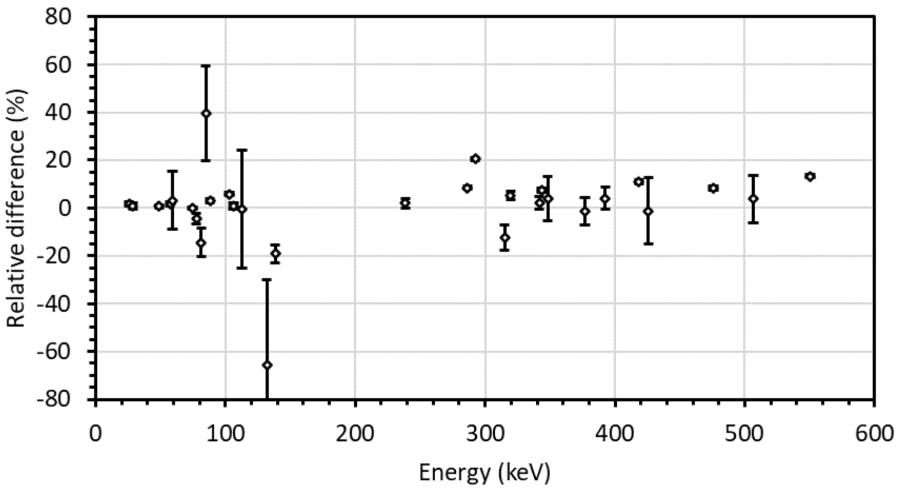




ENSDF: $I\gamma$(74.6 keV) = 10.20(54) %

This work: $I\gamma$(74.6 keV) = 10.183(57) %

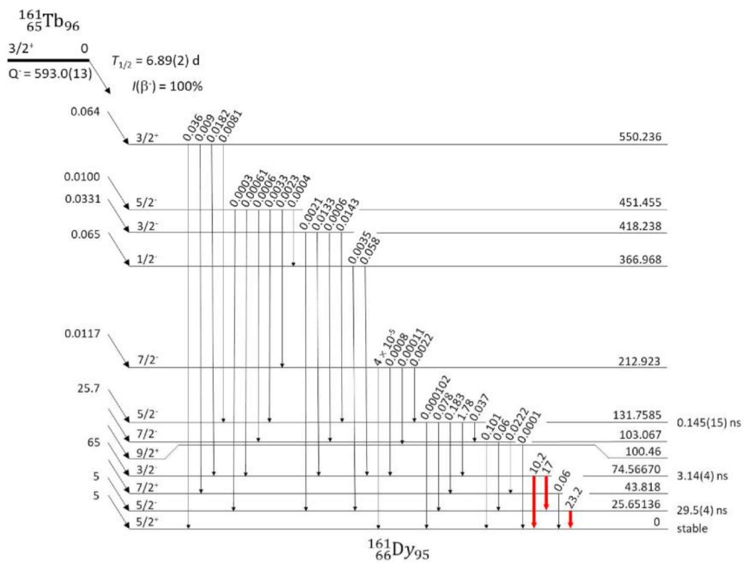
Comparison of normalised emission intensities of this work to ENSDF



Beta Energy (keV)	Level (keV)	Intensity (%) ENSDF	Intensity (%) This work
42.8	550.236	0.064 ± 0.004	0.07123 ± 0.00054
141.5	451.455	0.0100 ± 0.0011	0.008924 ± 0.00074
174.8	418.238	0.0331 ± 0.0021	0.03556 ± 0.00026
226	366.968	0.065 ± 0.005	0.0761 ± 0.0024
380.1	212.923	0.0117 ± 0.0016	0.00953 ± 0.00068
460	131.759	25.7 ± 1.6	26.24 ± 0.41
	103.067		
	100.46		
522	74.5667	65 ± 4	63.9 ± 1.1
	43.818		
567.3	25.6514	5 ± 5	5.7 ± 1.6
589	0	5 ± 5	3.9 ± 1.2

Sean Collins, Rob Shearman et al., (NPL)

^{161}Tb beta decay branching ratios from absolute emission intensities



Beta Energy	Level	Intensity (%)	
(keV)	(keV)	ENSDF	This work
42.8	550.236	0.064 ± 0.004	0.07123 ± 0.00054
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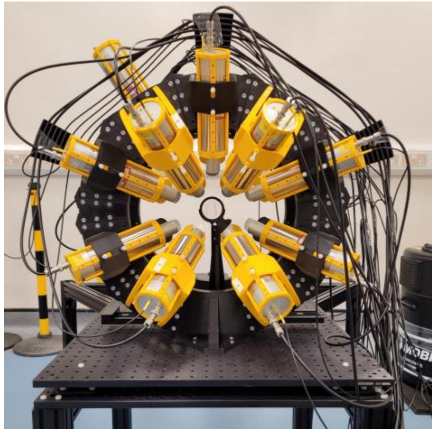
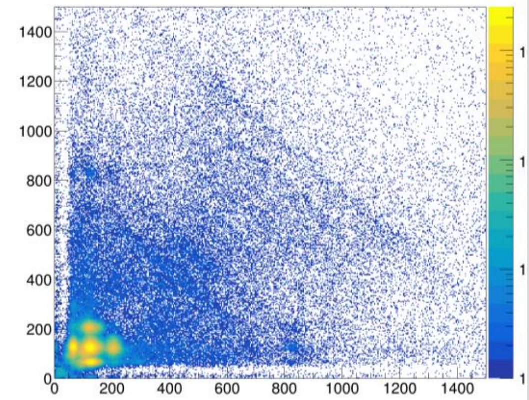
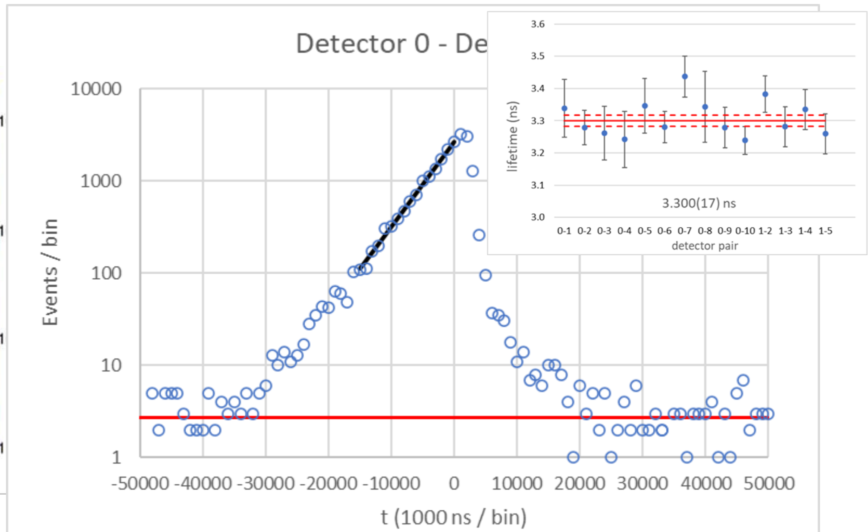






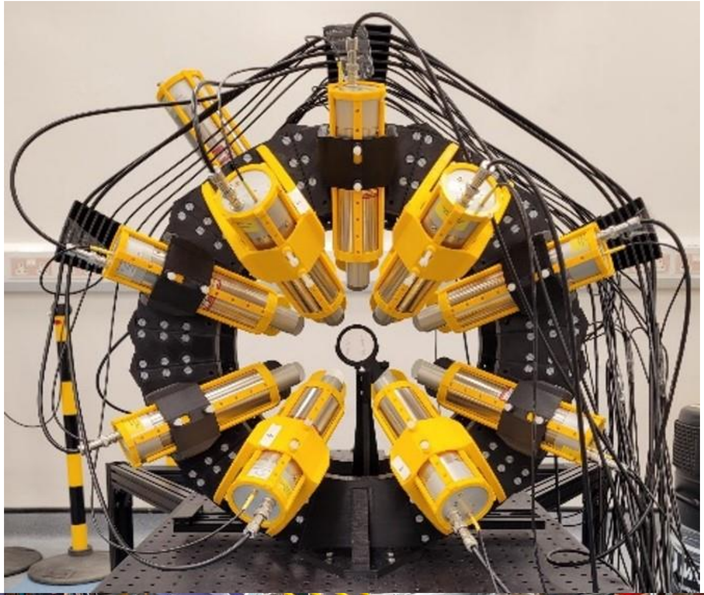
Discrepant from ENSDF lifetime of 3.14(4) ns.

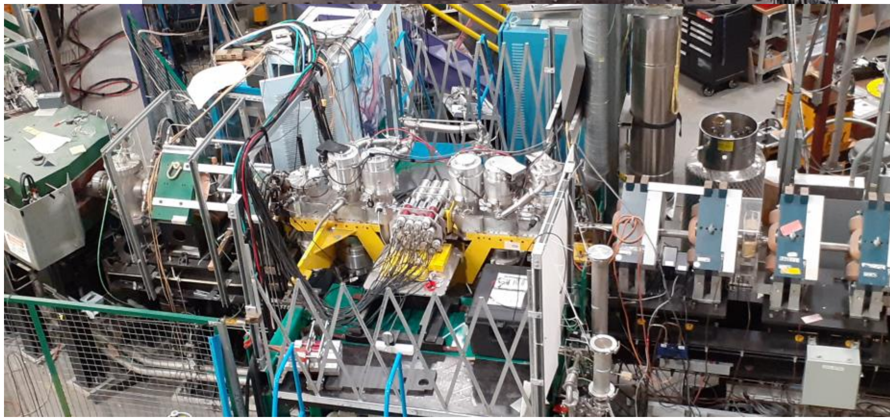
S.M.Collins, R. Shearman et al.,

NANA for External Experiments

- National Nuclear Array (NANA) developed for Nuclear data and Primary standardisation work,

- Also allows collaboration with international experiments in nuclear physics ‘curiosity driven’ research.





Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment
Volume 1056, November 2023, 168597

Full Length Article

Response of the FAst TIMing Array (FATIMA) for DESPEC at FAIR Phase-0

M.M.R. Chishti [a b], S. Jazrawi [a c], R. Shearman [c], P.H. Regan [a c], Zs. Podolyák [a], S.M. Collins [a c], M. Górska [g], B. Cederwall [i], A. Yaneva [g o], G.X. Zhang [d e], J. Cederkall [b], A. Goasduff [f], H.M. Albers [g], S. Alhomaidhi [g], A. Banerjee [g], A.M. Bruce [k], G. Benzoni [h t], B. Das [i], T. Davinson [l], L.M. Fraile [m]... V. Werner [n]

Applied Radiation and Isotopes
Volume 134, April 2018, Pages 290-296

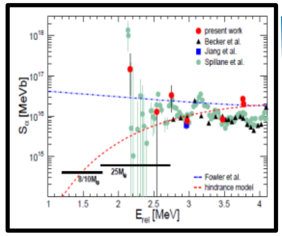
Investigation of γ-γ coincidence counting using the National Nuclear Array (NANA) as a primary standard

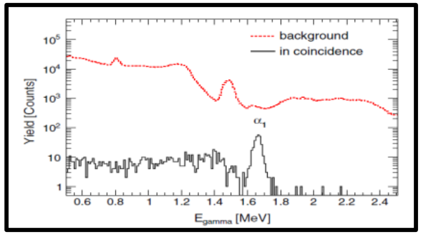
S.M. Collins [a b], R. Shearman [a b], J.D. Keightley [a], P.H. Regan [a b]

NANA collaborating in nuclear structure & astrophysics

NPL

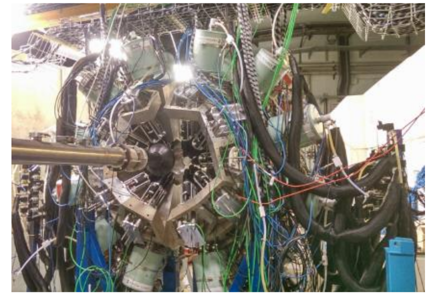
STELLA @ IPN-ORSAY
$^{12}C+^{12}C$ nucleosynthesis

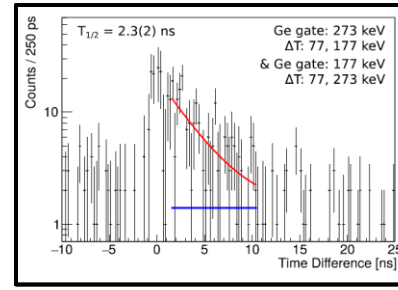




M.Heine et al., NIM **A903** (2018) p1-77
G.Fruet et al., PRL **124** (2020) 192701

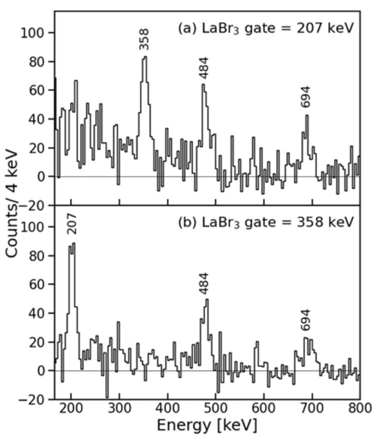
NuBALL at IJC Lab
^{166}Dy, ^{178}W, ^{238}U(n,f)

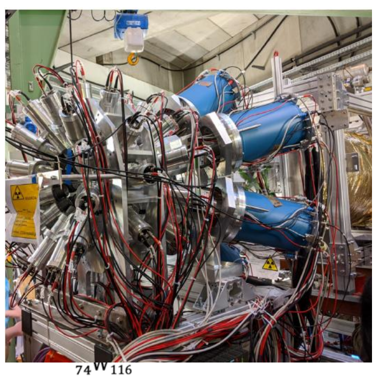




^{166}Dy: R.L.Canavan et al., PR **C101** (2020) 024313
^{178}W M.Rudigier et al., PL **B301** (2021) 135140
$^{134-8}$Te: G.Hjaeffner et al., PR **C103** (2021) 034317

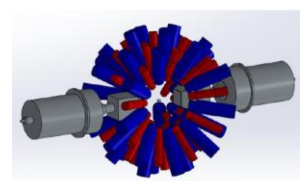
FATIMA @DESPEC, FAIR Phase-0
^{94}Pd, ^{190}W, ^{170}Er beam, …..

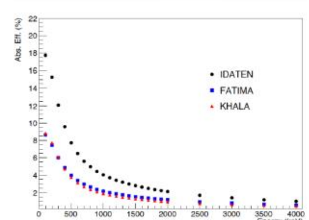


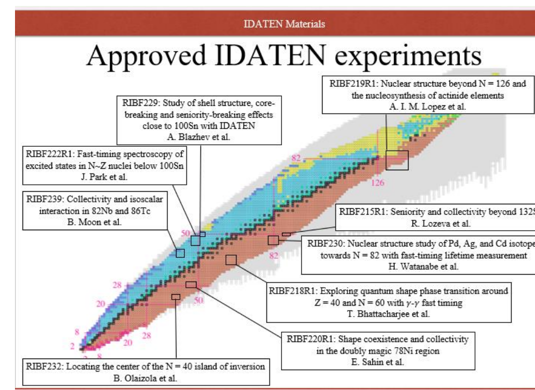


^{94}Pd: A.Yanena et al., PL B 138805 (2024)
^{190}W: E.Sahin et al., PL B 138976 (2024)

IDATEN at RIBF-RIKEN
^{94}Pd (test), ^{82}Nb, ~^{78}Ni, ~^{100}Zr, ^{128}Pd..



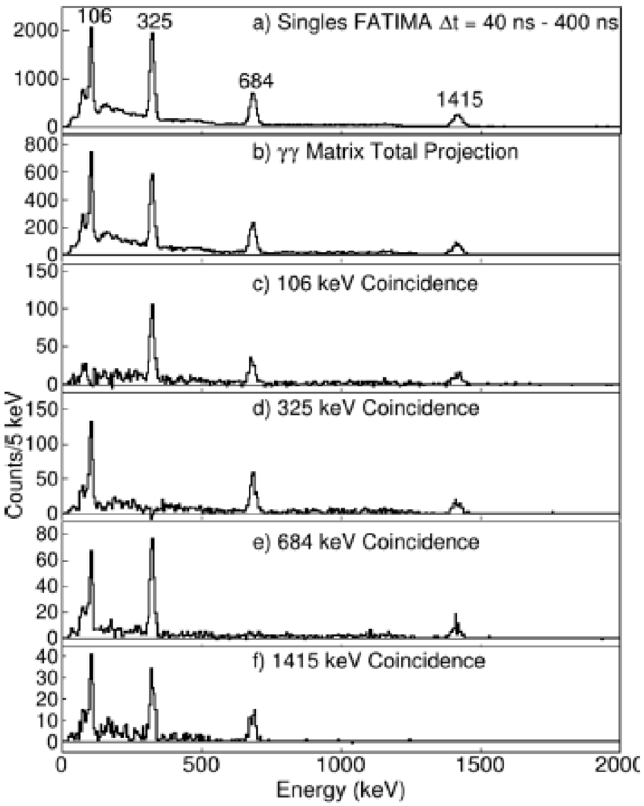


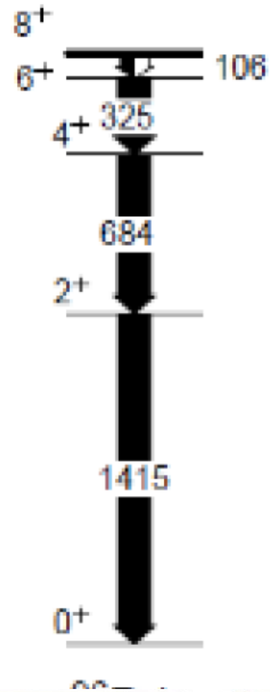


NP2112-RIBF212: Fast-timing γ-ray spectroscopy of exotic nuclei at RIBFH. Watanabe, P. H. Regan, and B. Moon

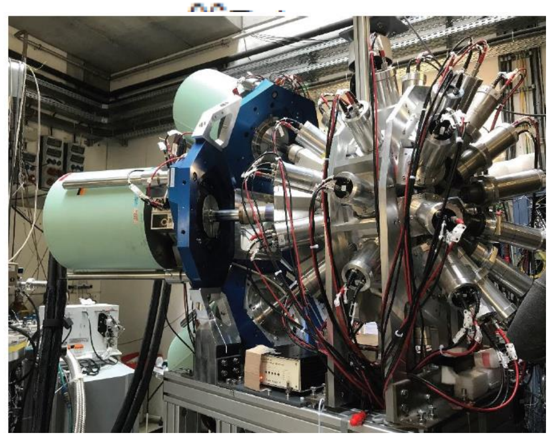
Application of $\gamma\gamma$-singles for ε_γ of FATIMA

Nuclear Inst. and Methods in Physics Research, A 1056 (2023) 168597





$$\epsilon_{\gamma_1} = \frac{I_{\gamma_1\gamma_2}(1+\alpha_2)}{I_{\gamma_2}}$$



Nuclear Inst. and Methods in Physics Research, A 1056 (2023) 168597

Contents lists available at ScienceDirect

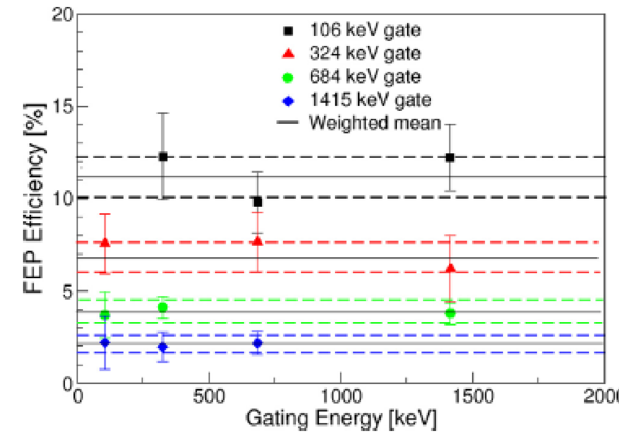
Nuclear Inst. and Methods in Physics Research, A

journal homepage: www.elsevier.com/locate/nima

Full Length Article

Response of the FAst TIMing Array (FATIMA) for DESPEC at FAIR Phase-0

M.M.R. Chishti, S. Jazrawi, R. Shearman et al.



Nuclear Inst. and Methods in Physics Research, A 1056 (2023) 168597

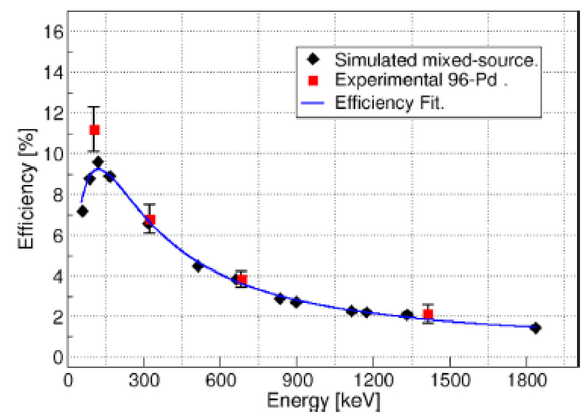
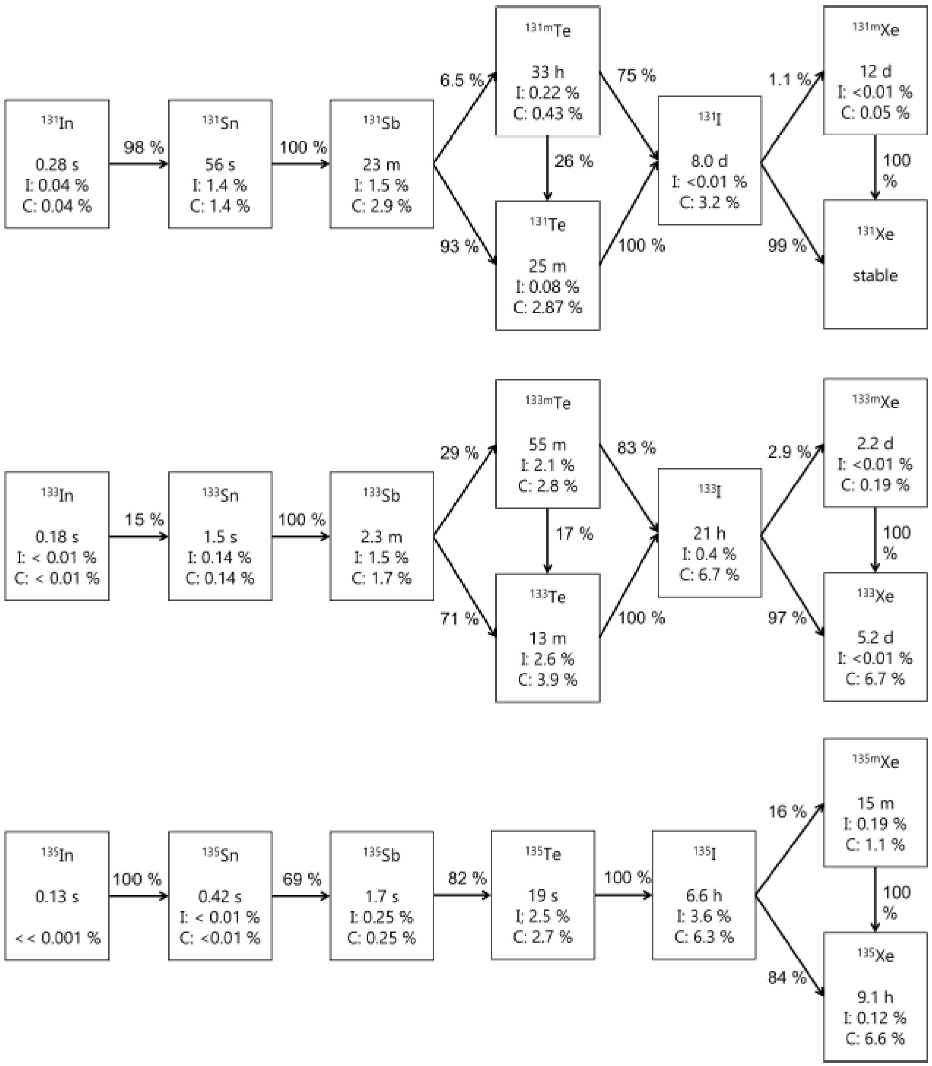


Fig. 5. Full-energy peak (FEP) efficiency response for the FATIMA array determined from the GEANT4 simulations and compared with the experimental, in-situ values calculated for the $I^\pi = 8^+$ isomeric decay cascade in ^{96}Pd.

CTBTO: Gaseous Radionuclides.

- Co-located with UN and IAEA in Vienna
- Organisation created to support implementation and verification of international Comprehensive Test Ban Treaty
- Aim to detect any nuclear explosion conducted on Earth – in the underground, underwater or in the atmosphere
- International Monitoring System (IMS) has been established to coordinate monitoring and sharing of data
- IMS comprised of 321 stations and 16 laboratories across the globe, with sensors for:
    - Seismic (underground), hydroacoustic (undersea), infrasound (atmosphere) and radionuclide (particulates and radioxenon in the atmosphere)



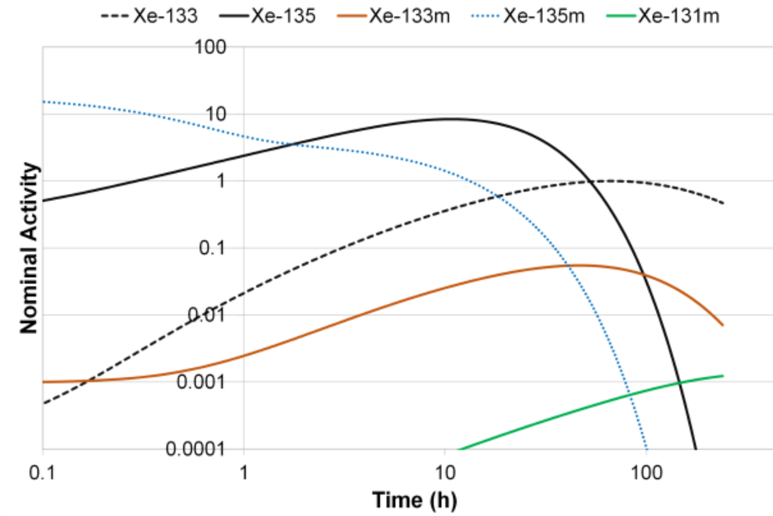


FIGURE 2.12: Simulated activity of five radioxenon isotopes relative to ^{133}Xe maximum activity, using ^{235}U thermal neutron-induced fission yields for 10 days. ^{133}Xe: black dashed; ^{135}Xe: black solid; ^{133m}Xe: orange; ^{131m}Xe: green; ^{135m}Xe: blue dotted. The activities here are calculated based on no fractionation from the parent nuclei. ^{135m}Xe

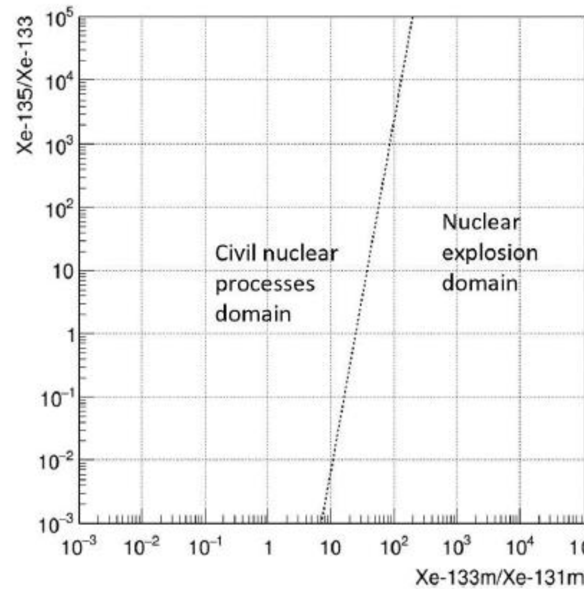
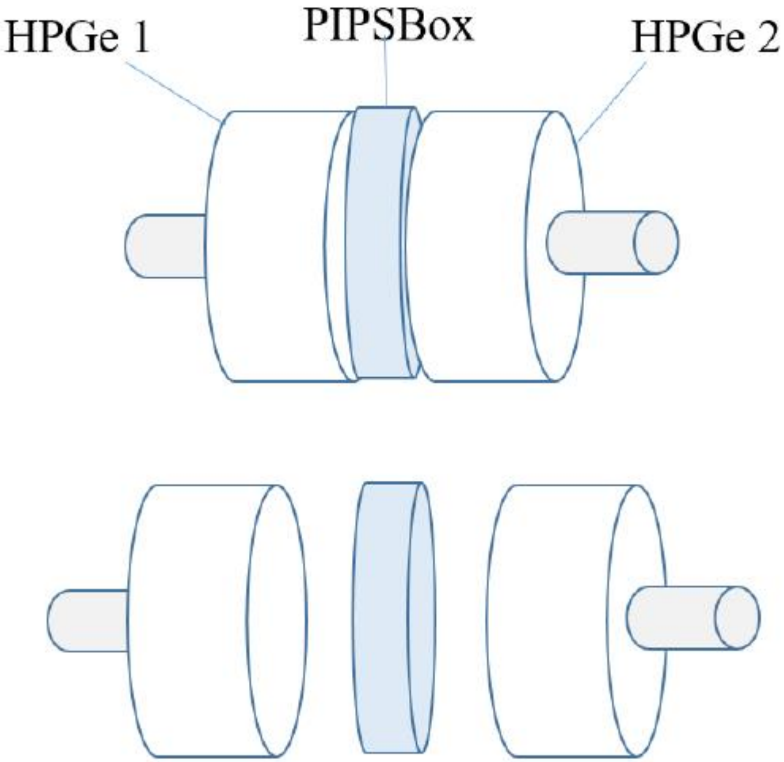
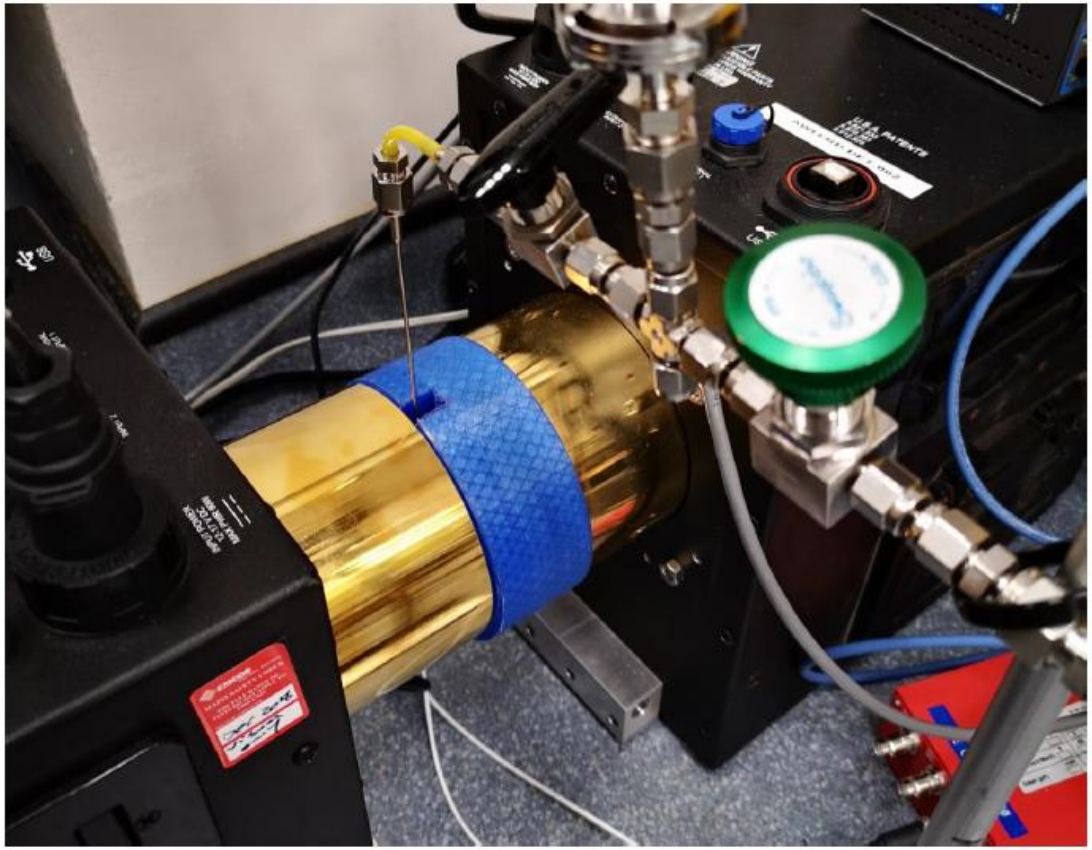


FIGURE 2.14: Radioxenon isotopic ratios plot showing the ratios that are consistent with civil nuclear facilities (left) and ratios that are consistent with a nuclear explosion (right). Figure from Goodwin *et al.* [40]

β-β-γ-γ





Goodwin et al, 2019. A high-resolution β-γ coincidence spectrometry system for radioxenon measurements. NIMA, 978, 164452. https://doi.org/10.1016/j.nima.2020.164452

Irradiate ^{235}U targets to produce noble gaseous radioactive (Kr, Xe) sources

Journal of Environmental Radioactivity 238–239 (2021) 106733

ELSEVIER

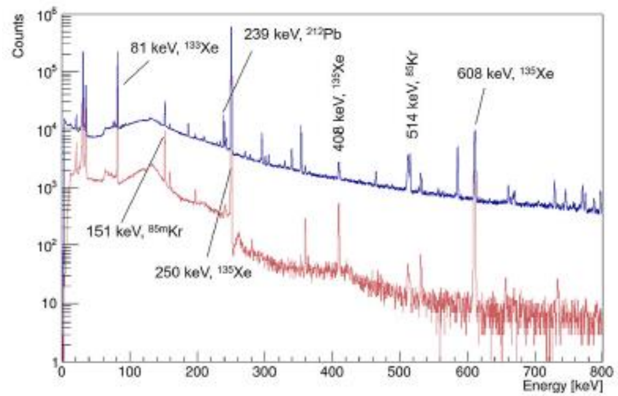
Journal of Environmental Radioactivity

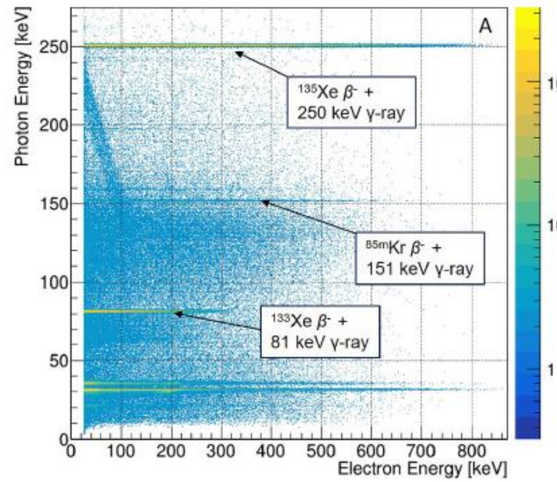
journal homepage: www.elsevier.com/locate/jenvrad

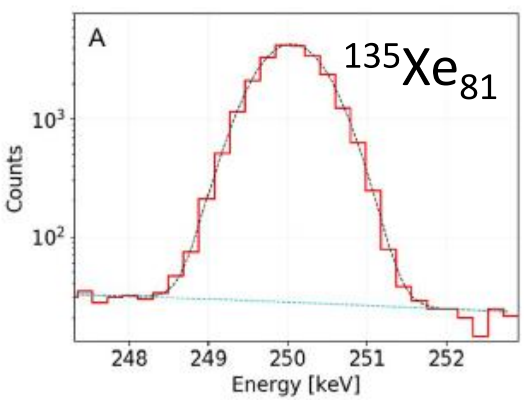
Production and measurement of fission product noble gases

Matthew A. Goodwin [a,b,*], Steven J. Bell [c], Richard Britton [d], Ashley V. Davies [a], Marc Abilama [c], Sean M. Collins [b,c], Robert Shearman [c], Patrick H. Regan [b,c]

[a] AWE Aldermaston, Reading, Berkshire, RG7 4PR, UK
[b] Department of Physics, University of Surrey, Guildford, GU2 7XH, UK
[c] National Physical Laboratory, Teddington, Middlesex, TW11 0LW, UK
[d] Provisional Technical Secretariat, CTBTO, Vienna, Austria







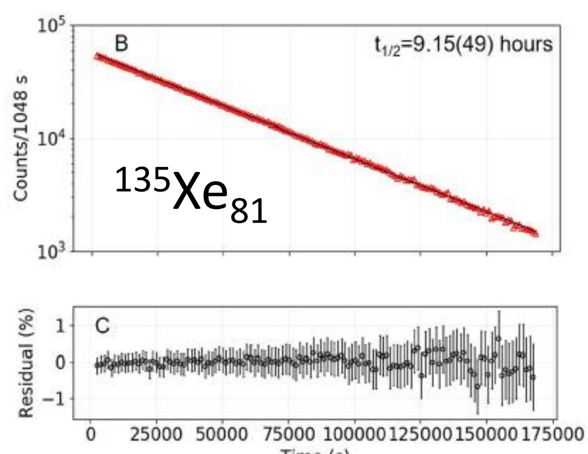


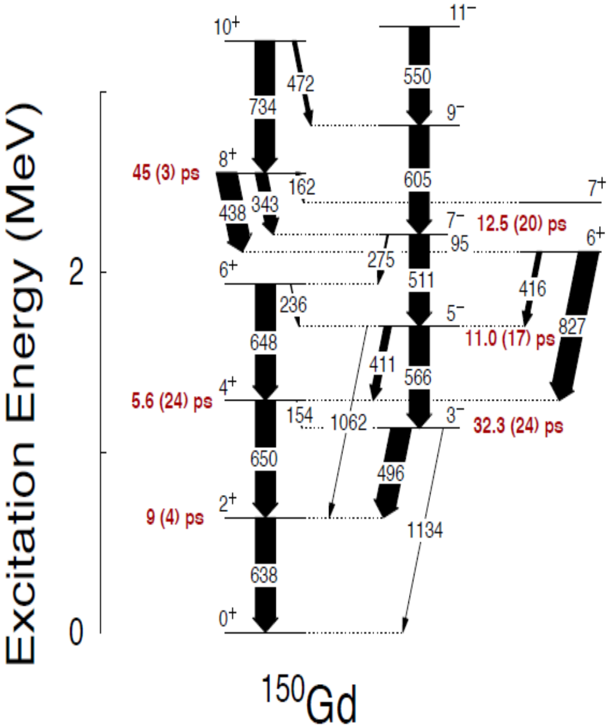
Table 3

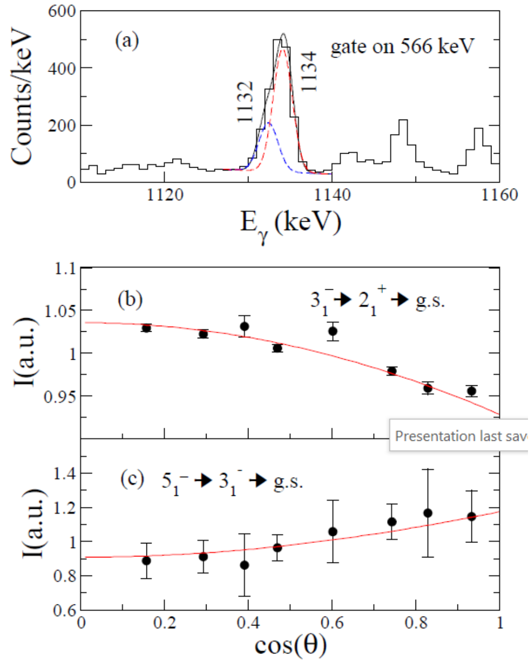
Nuclide identification from a peak search of the γ-ray spectrum from Extraction 1 using the full acquisition (t = 170,731 s).† X-rays from Xe/Cs minus Ge fluoresence (escape peak). The relative γ-ray intensity is the full energy peak integral divided by the simulated γ-ray detection efficiency, decay-corrected to the start of the acquisition relative to the 250 keV ^{135}Xe peak (see Eq. (1)).

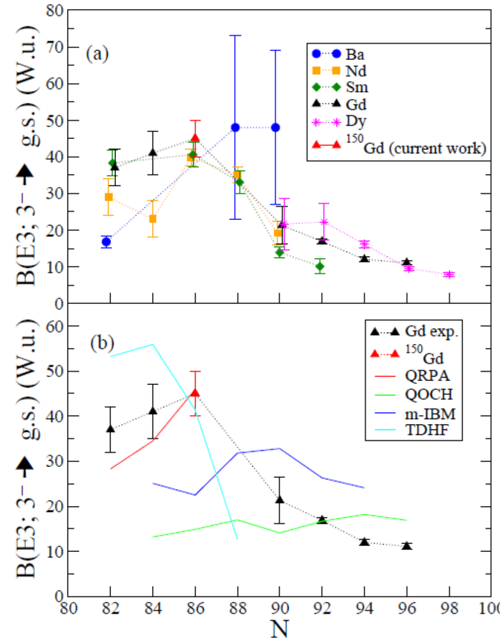
Fitted Energy (keV)	Parent Nucleus	Signature Type	γ-ray emission probability (%)	Rel. γ-ray Intensity (RGI) (× 1000)	Comment
20.4	Xe/Cs†	e^--X	–	–	Multiplet
30.7	Xe/Cs X K_α	e^--X	–	–	Multiplet
35.3	Xe/Cs X K_β	e^--X	–	–	Multiplet
80.9	^{133}Xe	$\beta-\gamma$	37.3(4)	47.4(7.1)	81.0 + 79.6 keV
122.8	^{88}Kr	$\beta-\gamma$	0.20(1)	0.807(12)	
129.1	^{85m}Kr	$\beta-\gamma$	0.30(8)	4.73(71)	
151.4	^{85m}Kr	$\beta-\gamma$	75.2(8)	38.1(5.7)	
158.6	^{135}Xe	$\beta-\gamma$	0.29(1)	2.40(36)	
196.6	^{88}Kr	$\beta-\gamma$	26(1)	4.42(67)	
233.4	^{133m}Xe	γ	10.1(2)	0.65(98)	
240.6	^{88}Kr	$\beta-\gamma$	0.25(1)	0.78(12)	
250.2	^{135}Xe	$\beta-\gamma$	90.0(3)	1000(7)	
305.1	^{85m}Kr	$\beta-\gamma$	14.0(4)	5.32(80)	^{85m}Kr > ^{85}Kr
358.5	^{135}Xe	$\beta-\gamma$	0.22(1)	1.94(29)	
390.0	^{88}Kr	$\beta-\gamma$	0.64(5)	0.87(14)	
407.7	^{135}Xe	$\beta-\gamma$	0.36(2)	5.83(88)	
438.9	^{88}Rb	$\beta-\gamma$	0.015(4)	1.35(21)	
451.1	^{85m}Kr	$\beta-\gamma$	0.011(4)	0.88(14)	
454.4	^{135}Xe	$\beta-\gamma$	0.004(1)	0.55(9)	
514.3	^{85}Kr	γ	0.43(1)	4.76(72)	^{85}Kr > ^{85}Rb
526.4	^{135m}Xe	γ	80.6(6)	35.5(5.4)	
530.3	^{133}I	$\beta-\gamma$	87(2)	2.81(42)	
608.3	^{135}Xe	$\beta-\gamma$	2.9(1)	81.4(1.2)	^{214}Bi Interference
731.9	^{135}Xe	$\beta-\gamma$	0.055(4)	0.39(6)	
834.9	^{88}Kr	$\beta-\gamma$	13(2)	6.75(10)	
898.2	^{88}Rb	$\beta-\gamma$	14.4(2)	1.73(27)	
1836.5	^{88}Rb	$\beta-\gamma$	22.8(1)	5.07(78)	

Precision metrology important in nuclear structure: Direct measurement of B(E3:$3^- \to 0^+$) in ^{150}Gd

NPL National Physical Laboratory







E_i [keV]	τ [ps]	J_i^π	J_f^π	E_f [keV]	Mult.	I_γ exp.	$\alpha(\times 10^{-3})$	Exp.
1134.30(2)	32.3(24)	3_1^-	2_1^+	638.05(2)	E1	100(1)	4.79	$8.7(7)\times 10^{-5}$
			0_1^+	0.0	E3	0.27(2)	4.39	45(5)
1699.91(3)	11.0(17)	5_1^-	4_1^+	1288.42(3)	E1	46(1)	7.38	$1.4^{+3}_{-2}\times 10^{-4}$
			3_1^-	1134.30(2)	E2	100(1)	10.15	18(3)
			2_1^+	638.05(2)	[E3]	0.10(2)	5.13	53^{+15}_{-12}

^{150}Gd studied @IFIN-HH with ROSPHERE by

i) RDM ^{140}Ce(^{13}C,3n)^{150}Gd

Ii) ^{147}Sm(^{6}Li,3n)^{150}Tb EC $\to$ ^{150}Gd decay

Maximal octupole collectivity across the Z = 64 isotopic chain: B(E3) values in ^{150}Gd, S.Pascu et al., submitted to PRL July 2024

Summary

- ‘Realising the becquerel’ is scientifically challenging..

- Primary & secondar) standard methods vary for each species...

- High precision data is needed for medical radioisotopes.

- Public confidence requires real-time radioactive gas metrology.

- There are strong and direct links between ‘curiosity driven’ nuclear physics and nuclear metrology for societal benefit.