ATOMIC AND NUCLEAR DECAY DATA – A FEW RECENT OBSERVATIONS

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Decay Data Evaluation Project – training workshop, 21-25 October 2024, CEA Saclay, Gif-sur-Yvette, France

Half-life of ^{144}Ce

NSR reference	Half-life (d)	Selected detail, issues and uncertainties
1943Bo02	310	β^- activity monitored as a function of time; issues: approximation with no quantified estimate of uncertainty, and therefore not adopted in weighted-mean analyses
1944Jo01	297 ± 30	β^- activity monitored as a function of time; issues: lack of sufficient detail and defined as an outlier, and therefore not adopted in weighted-mean analyses
1951Sc56	282 ± 3	flow proportional counter over 1.5 years; superseded by laboratory measurements of 1956Sc87
1956Sc87	284.5 ± 1.0	end-window methane flow proportional counter over 6.5 years; issues: possible inadequate separation from other fission products, and no systematic uncertainties
1957Me47	285 ± 2	AECL 4πβ proportional counter over 4 years; issue: no sound provision of statistical + systematic uncertainties
1960Ea02	277 ± 4	vibrating-reed electrometer to monitor separate ^{226}Ra and ^{144}Ce sources in Ar-filled balanced ion chambers; issue: defined as an outlier, and therefore not adopted in weighted-mean analyses
1965Fl02	283.8 ± 0.6	2π internal gas-flow proportional counter; two sources counted over 3.6 and 3.4 years; issue: only statistical uncertainty quantified
1968La10	284.8 ± 1.0	^{144}Ce + ^{144}Pr separated from other fission products, and monitored by β^- and Ge(Li) γ–ray spectrometers; issue: uncertain purity of parent-daughter ^{144}Ce + ^{144}Pr
1968Re04	284.9 ± 0.8	end-window proportional counter over 2.4 years; issue: lacks any real detail, with no quantification of the systematic uncertainty
1976WaZH	285.08 ± 0.18	superseded by continued laboratory measurements published as 1983Wa26
1980Ho17	285.8 ± 0.1	4πγ pressurized ionization chamber over 2.9 years; issues: overall uncertainty stated to be a combination of statistical uncertainty (1σ) + ill-defined systematic uncertainties - lacks basic operational information to analyse and justify such a low uncertainty
1982HoZJ	284.9 ± 0.5	NBS well-type Ar-filled ionization chambers over 2.3 to 5.9 years; superseded by further NBS/NIST measurements of 1992Un01 and 2002Un02, and the study of 2012Fi12 after their discovery of problems in maintaining constant detector-source geometry within the NIST ionization chamber
1983Wa26	284.45 ± 0.14	4πγ pressurized ionization chamber over nearly 2 years, with full uncertainty analysis - judged as adequately quantified for weighted-mean analyses
1986Ol01	284.893 ± 0.008	4πβ gas-flow proportional counter and two NaI γ detectors (β–γ coincidence), with counting intervals "greater than one half-life of ^{144}Ce", implying less than two half-lives (i.e., over no more than 1.5 years); issues: systematic uncertainties not satisfactorily documented – items for correction listed, but not quantified
1992Un01	284.558 ± 0.038	NIST 4πγ pressurized ionization chamber over 2.7 years; superseded by further NBS/NIST measurements and the study of 2012Fi12 after their discovery of problems in maintaining constant detector-source geometry within the NIST ionization chamber
1997Ma75	286.14 ± 0.09	4πγ re-entrant-well ionization chamber over 5 years issue: potential difficulties correcting for ^{154}Eu impurity; also defined as an additional outlier at a late stage in the weighted-mean analysis
2002Un02	284.534 ± 0.032	NIST 4πγ pressurized ionization chamber over 3.0 years; superseded by further NBS/NIST measurements and the study of 2012Fi12 after their discovery of problems in maintaining constant detector-source geometry within the NIST ionization chamber
2012Fi12	284.35 ± 0.08	NIST 4πγ pressurized ionization chamber; 2012Fi12 replaced by 2014Un01 – uncertainty increased from ± 0.08 to 0.40 d; subsequently, 2014Un01 replaced by 2014Un01 corrigendum (2020) whereby uncertainty of ± 0.08 d was re-adopted
2014Un01	284.35 ± 0.40	
2020 re-adjustment of 2014Un01	284.35 ± 0.08	NIST 4πγ pressurized ionization chamber, re-corrected data with full uncertainty analysis - judged as adequately quantified for weighted-mean analyses

20 half-life publications (1943-2020): outliers; ill-defined uncertainties; restricted statistical uncertainties; inadequate quantification of systematic uncertainties; superseded by further measurements by same laboratory/equipment

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2014Un01	284.35 ± 0.40	
2020 re-adjustment of 2014Un01	**284.35 ± 0.08**	**NIST 4πγ pressurized ionization chamber, re-corrected data with full uncertainty analysis - judged as adequately quantified for weighted-mean analyses**
Recommended values	284.40 ± 0.05 d	UWM
	284.38 ± 0.08 d	LWM, $\chi^2/(N-1)$ of 0.38 and $\chi^2/(N-1)_{critical}$ at 99% confidence of 6.63; uncertainty adjusted from ± 0.07 to the accepted lowest-measured value of ± 0.08 days.

Decay data of ^{144}Ce

Specific future decay-data requirements for this fission product (2022):

1) **need a healthy number of fully acceptable half-life measurements with sound estimates of uncertainties – uncertainty budgets that stand up to critical inspection**

2) highly-resolved E_γ (keV) and absolute P_γ (%) measurements, particularly for the 33.57(3)- and 40.93(3)-keV γ rays that are in close proximity to XKL_2 of 36.965 keV, XKL_3 of 35.694 keV; and XKM_2 of 40.808 keV, XKM_3 of 40.906 keV, XKM_5 of 41.221 keV

3) consider further extensive conversion-electron measurements to assist in resolving the above (akin to J.S. Geiger, *et al*., 1960Ge05)

4) multipolarities/mixing ratios of 33.57- and 40.93-keV γ rays (M1+E2: both set currently at 100%M1)

5) IT decay of $^{144}Pr^m$ (single 59.03(3)-keV γ ray defined as highly-converted M3 transition) – would welcome accurate conversion-electron measurements

Decay data of $^{244}Am^{g,m}$

2009/2010 IAEA-DDEP evaluations → example evidence of inadequacies in measured decay data of the ground state

1) $^{244}Am^{g}$: only γ-ray studies of Hoff *et al*. provide uncertainty estimates in γ-ray probabilities that are expressed in terms of their relative intensity per 100 neutron captures in a high-flux reactor (1984Ho02)

2) no weighted-mean data could be derived – data of 1984Ho02 adopted wholesale (expressed in terms of the 743.977-keV gamma-ray emission probability (100)), and re-adjusted when deemed necessary

3) only the single beta-particle energy calculated from structural detail of proposed decay scheme, based on nuclear-level energy of 1040.188(12) keV from Akovali (2003Ak04) and Q_{β^-} of 1427.3(10) keV from Audi *et al*. (2003Au03) to determine energy and uncertainty of β-particle transition – this single β^- transition was assigned an emission probability of 100%

4) further measurements merited to determine the decay scheme of $^{244}Am^{g}$ with greater confidence, and quantify the γ-ray emission probabilities with improved clarity and certainty

Decay data of $^{244}Am^{g,m}$

2009/2010 IAEA-DDEP evaluations → also example evidence of inadequacies in measured decay data of the metastable state

1) $^{244}Am^{m}$: relatively simple decay scheme constructed from branching fraction measurements of Fields *et al*. and Gabeskipiya *et al*. (1955Fi36, 1976Ga31) and γ-ray studies of Hoff *et al*. (1984Ho02).

2) $BF_{\beta-}$ of 0.99964(1) and BF_{EC} of 0.00036(1)

3) only γ-ray studies of Hoff *et al*. provide estimates of the γ-ray emission probabilities and their uncertainties per 100 neutron captures – therefore, no weighted-mean data could be derived, and the decay data of 1984Ho02 were adopted as published

4) energies of six β-particle emissions calculated from structural detail of proposed decay scheme from Akovali (2003Ak04) and $Q_{\beta-}$ of (1427.3(10) + 88.6(29) → 1516(3)) keV from Audi *et al*. (2003Au03) to determine energy and uncertainty of β^- emissions

5) further measurements merited to determine the decay scheme of $^{244}Am^{m}$ with greater confidence, and quantify all β^- and γ-ray emission probabilities with improved clarity and certainty

Decay data of $^{244}Am^{g,m}$

2009/2010 IAEA-DDEP evaluations → example evidence of inadequacies in measured decay data of both the ground and metastable states of ^{244}Am

- 2009/2010 evaluation: further measurements merited to determine the decay schemes of $^{244}Am^{g}$ and $^{244}Am^{m}$ with greater confidence, and quantify all β^- and γ-ray emission probabilities with improved clarity and certainty

- 2019 – highly relevant measurements carried out and published

  2019Na34: S. Nakamura, *et al.*, J. Nucl. Sci. Technol. **56**, 123-129 (2019).

  2019Tr05: R. Tripathi, *et al.*, Nucl. Instrum. Methods Phys. Res. **A922**, 143-149 (2019); corrigendum, Nucl. Instrum. Methods Phys. Res. **A935**, 239 (2019).

A good example of the healthy synergies that need to exist between decay-data evaluators and measurers

Decay data of $^{244}Am^{g,m}$

		$^{244}Am^{g}$			$^{244}Am^{m}$	
		2009/2010 IAEA-DDEP evaluation	2019Na34	2019Tr05	2009/2010 IAEA-DDEP evaluation	2019Tr05
Half-life		10.1(1) h	–	**10.01(3) h**	26(3) min	**26.13(43) min**
		absolute P_γ (%)			**relative P_γ (%)**	
E_γ (keV)	42.96	0.096(20)	–	–		
	99.38	5.0(11)	–	**4.73(15)**		
	153.86	19(4)	**17.1(12)**	**21.19(69)**		
	205.58	0.35(8)	–	**0.388(37)**		
	538.40	0.66(19)	**< 0.42**	**0.522(23)**		
	743.977	66(8)	**66.5(11)**	**67.7(36)**		
	897.840	28(8)	**28.0(10)**	**27.59(84)**		
E_γ (keV)	42.96				8.33(4)	–
	940.5				–	**17.1(14)**
	942.03				97(46)	**45.9(20)**
	977.80				–	**21.6(16)**
	983.46				–	**24.8(18)**
	1041.22				53(24)	**44.9(17)**
	1062.95				75(33)	**76.1(22)**
	1084.18				100	**100**
	1105.9				11(7)	–

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Auger-electron and X-ray decay data of ^{131}Cs

^{131}Cs ($t_{½}$ of 9.681(16) d): single 100% EC decay directly to ground state of daughter ^{131}Xe, with the emission of only Auger electrons and X-rays

- low-energy Auger electrons possess very short mean-free paths of the order of nanometers, and such high linear-energy transfer (LET) is an effective means of causing effective and efficient localised damage to cancer cells
- potential applications in brachytherapy and microdosimetry
- health warning: likelihood of achieving the above is dependent on identifying suitable chemical species to transport ^{131}Cs, achieve close contact and irradiate carcinoma precisely (chemistry constitutes the major problem that has to be addressed, rather than the atomic decay data)

Quantification of Auger-electron and X-ray shell and sub-shell emissions

BrIccEmis code, NS_RadList v1.0:

B.P.E. Tee, T. Kibédi, B.Q. Lee, A.E. Stuchbery, BrIccEmisDB Database and Computational Tool to Evaluate Atomic Relaxation Radiations , Department of Nuclear Physics and Accelerator Applications, Australian National University (ANU), Acton, Canberra, Australia, paper in preparation – to be submitted for publication in At. Data Nucl. Data Tables (2023Texx)

Auger-electron and X-ray decay data of ^{131}Cs

Quoted uncertainties include direct contributions from only electron capture and internal-conversion electrons (2023Texx) − these data do not include the significant uncertainties from the more complex but less well-defined atomic radiation probabilities as considered semi-quantitatively in EADL (1991PeZY)

– as specified by 1990PeZY, undeclared uncertainties in theoretical Auger-electron radiative rates are ~ 10% for the K and L shells (except for Coster-Kronig and super Coster-Kronig transitions), and can be of the order of 30% or more for the Coster-Kronig and super Coster-Kronig transitions and outer shells, while undeclared uncertainties in theoretical X-ray radiative rates are ~ 10% for the K and L shells, and can be of the order of 30% for the outer shells

1990PeZY: S.T. Perkins, *et al.*, "Tables and Graphs of Atomic Subshell and Relaxation Data Derived from the LLNL Evaluated Atomic Data Library (EADL), Z = 1 - 100", Lawrence Livermore National Laboratory report UCRL-50400, 30 (1991)

Code calculations in good agreement when benchmarked against Auger-electron experimental studies of A. Kovalík, *et al.*, "The Electron Spectrum from the Atomic De-excitation of $^{131}_{54}Xe$", J. Electron Spectrosc. Relat. Phenom. **95**, 231-254 (1998)

Auger-electron decay data of ^{131}Cs

Calculated Auger-electron spectrum for ^{131}Cs (BrIccEmis code, 2 March 2021 and NS_RadList v1.0, 8 April 2022): intensity cut-off of 0.05 per 100 decays for partial listings given below

Auger electrons		Mean energy (keV)	Energy (keV), 95% confidence range	Electrons per 100 decays (%) [a]
Auger total	(Xe)	0.623	0.003 – 3.879	**985.4 (20)**
Auger K_{tot}	(Xe)	25.878	23.516 - 32.437	**8.948 (17)**
Auger KLL	(Xe)	24.299	23.516 - 24.832	**5.947 (11)**
Auger KLX	(Xe)	28.606	27.908 – 29.608	**2.718 (5)**
Auger KXY	(Xe)	32.854	32.220 – 34.177	**0.2830 (5)**
Auger L_{tot}	(Xe)	2.997	0.130 – 4.362	**93.77 (+22, -21)**
Auger Coster-Kronig LLX	(Xe)	0.308	0.076 – 0.577	**13.92 (6)**
Auger LMM	(Xe)	3.282	2.659 – 4.009	**60.64 (+13, -12)**
Auger LMX	(Xe)	3.995	3.590 – 4.608	**17.89 (4)**
Auger LXY	(Xe)	4.715	4.399 – 5.297	**1.3282 (+28, -27)**
Auger M_{tot}	(Xe)	0.368	0.020 – 0.700	**229.5 (5)**
Auger Coster-Kronig MMX	(Xe)	0.101	0.007 – 0.280	**66.78 (15)**
Auger MXY	(Xe)	0.477	0.315 -0.720	**162.76 (32)**
Auger N_{tot}	(Xe)	0.026	0.003 – 0.075	**653.2 (+13, -14)**
Auger super Coster-Kronig NNN	(Xe)	0.010	0.002 – 0.035	**77.18 (16)**
Auger Coster-Kronig NNX	(Xe)	0.046	0.003 – 0.108	**86.79 (19)**
Auger NXY	(Xe)	0.025	0.003 – 0.037	**489.2 (10)**

[a] Quoted uncertainties include direct contributions from only electron capture and internal-conversion electrons (2023Texx) – these data do not include significant uncertainties from the more complex but less well-defined atomic radiation probabilities as considered semi-quantitatively in EADL, whereby uncertainties in the theoretical Auger-electron radiative rates are < 15% for the K and L shells (except for Coster-Kronig and super Coster-Kronig transitions), and can be of the order of 30% or more for Coster-Kronig and super Coster-Kronig transitions and the outer shells (1991PeZY).

X-ray decay data of ^{131}Cs

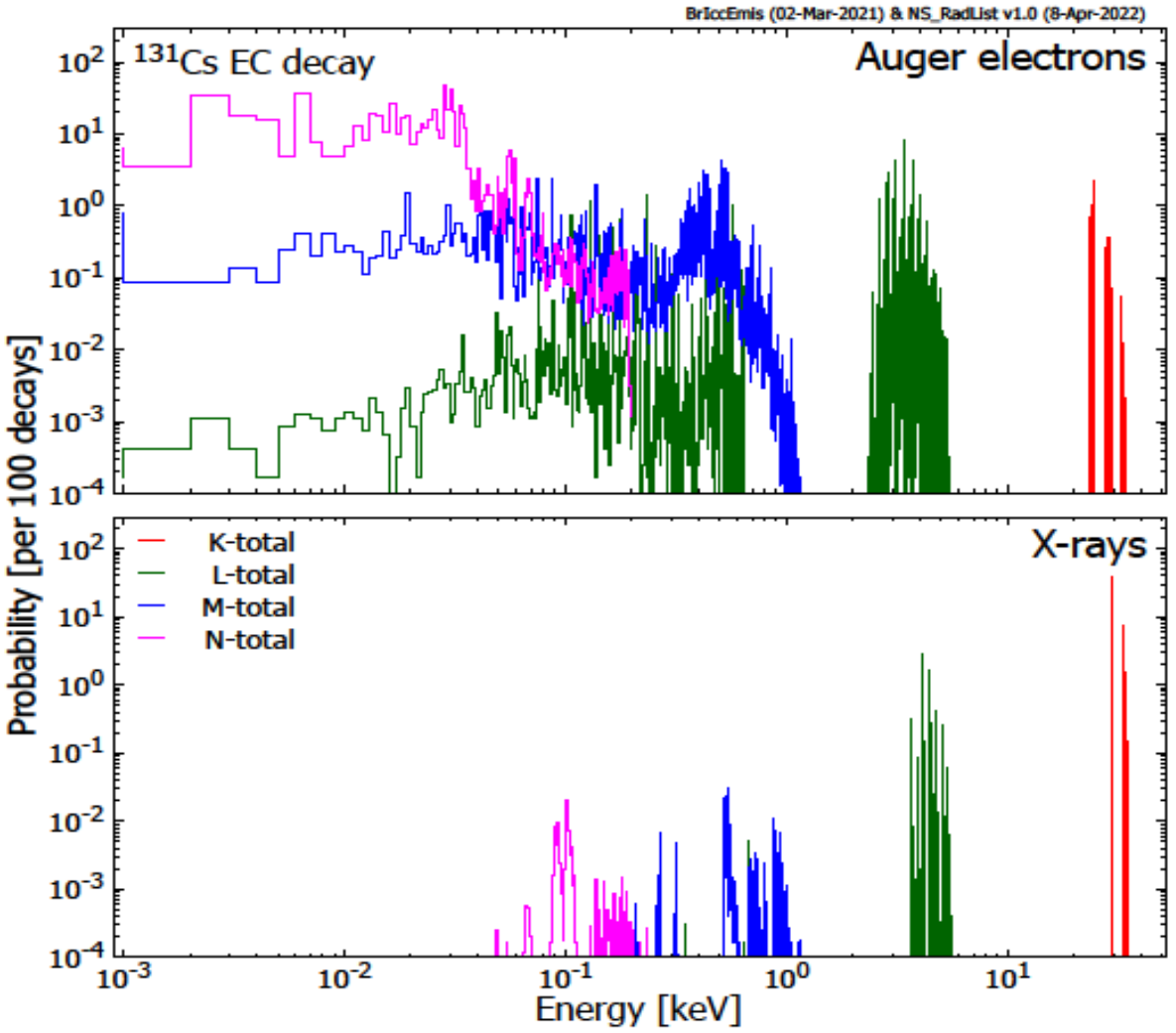
Calculated X-ray spectrum for ^{131}Cs (BrIccEmis code, 2 March 2021 and NS_RadList v1.0, 8 April 2022): intensity cut-off of 0.05 per 100 decays for partial listings given below

X-rays		Mean energy (keV)	Energy (keV), 95% confidence range	Photons per 100 decays (%) [a]
X_{tot}	(Xe)	27.478	4.120 – 34.408	**83.75 (16)**
XK_{tot}	(Xe)	30.410	29.458 – 34.408	**74.43 (14)**
XKL_2	(Xe)	29.458	29.458	**21.38 (4)**
XKL_3	(Xe)	29.778	29.778	**39.52 (+7, -8)**
XKM	(Xe)	33.606	33.563 – 33.625	**10.980 (21)**
XKM_2	(Xe)	33.563	33.563	**3.702 (7)**
XKM_3	(Xe)	33.625	33.625	**7.193 (14)**
XKN	(Xe)	34.417	34.408 – 34.420	**2.319 (4)**
XKN_2	(Xe)	34.408	34.408	**0.7773 (15)**
XKN_3	(Xe)	34.420	34.420	**1.526 (3)**
XL_{tot}	(Xe)	4.310	3.643 – 5.059	**8.683 (+18, -17)**
XM_{tot}	(Xe)	0.635	0.268 – 0.981	**0.5127 (10)**
XN_{tot}	(Xe)	0.110	0.086 – 0.193	**0.12145 (+23, -22)**

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Auger-electron and X-ray decay data of ^{131}Cs

Calculated Auger-electron and X-ray spectra for ^{131}Cs
BrIccEmis code, 2 March 2021 and NS_RadList v1.0, 8 April 2022



Applications

Reactor decay-heat calculations

Distant non-invasive monitoring of reactor operations – antineutrinos

Nuclear medicine

Medical applications

A.L. Nichols, "Status of the Decay Data for Medical Radionuclides: Existing and Potential Diagnostic γ Emitters, Diagnostic β^+ Emitters and Therapeutic Radioisotopes", Radiochim. Acta **110**, 609-644 (2022)

Diagnostic γ emitters and single-photon emission computed tomography (SPECT):

- further measurements required rather than immediate decay-data evaluations

β^+ emitters for positron emission tomography (PET):

- ^{61}Cu 100% EC/β^+ decay – although identified as a reasonably well-defined decay scheme, recent extensive γ-ray studies have revealed inconsistencies which need to be confirmed and fully quantified along with additional β^+ experimental studies, **prior to a re-evaluation of the decay scheme**

Non-standard β^+ emitters:

- ^{38}K and ^{45}Ti 100% EC/β^+ decay possess low-intensity EC/β^+ branches that **merit further investigation to improve their clarity**

Palliative and therapeutic radionuclides:

- further X-ray, internal-conversion and Auger-electron studies of ^{125}I 100% EC decay merited **prior to re-evaluation**
- further X-ray and internal-conversion electron studies of both ^{161}Tb and ^{169}Er 100% β^- decay merited **prior to re-evaluation**

Potential therapeutic radionuclides:

- ^{67}Cu 100% β^- decay – **full re-evaluation of decay scheme**
- ^{114}In^m, ^{117}Sn^m, ^{193}Pt^m and ^{195}Au^m IT decay – re-assessment of internal-conversion electron probabilities for radiotherapy, **along with re-evaluations of decay schemes**

ALN: previous and on-going decay-data evaluations

DDEP evaluations: "Date" applies to completion of evaluation and preparation of detailed comments; (2) means second attempt at a decay scheme evaluation for DDEP.

Radionuclide	Date	Radionuclide	Date	Radionuclide	Date
25-Mn-56	01/2004(2)	52-Te-127m	05/2012	83-Bi-212	02/2011(2)
30-Zn-63	03/2013	52-Te-132	08/2009	83-Bi-215	07/2011
34-Se-73	03/2014	**53-I-120**	√	84-Po-212	02/2011(2)
35-Br-76	07/2016	53-I-132		84-Po-216	01/2011(2)
40-Zr-89	09/2014	**55-Cs-131**	**09/2021**	85-At-211	02/2011
42-Mo-99/43-Tc-99m	√	58-Ce-144	06/2014	85-At-219	09/2010
43-Tc-94m	07/2014	59-Pr-142	02/2017	86-Rn-219	02/2011
43-Tc-97m		**59-Pr-142m**	√	86-Rn-220	01/2011(2)
43-Tc-99m	√	59-Pr-144	07/2014	88-Ra-224	01/2011(2)
45-Rh-103m	**07/2024**	59-Pr-144m	07/2014	90-Th-228	04/2010(2)
46-Pd-103/45-Rh-103m	**08/2024**	**73-Ta-178 (7^-)**	√	95-Am-242	12/2010
46-Pd-109	04/2009	**73-Ta-178 ((1+))**		95-Am-242m	04/2010
49-In-111/48-Cd-111m	√	80-Hg-203	01/2004(2)	95-Am-244	08/2009
51-Sb-126		81-Tl-208	07/2010(2)	95-Am-244m	02/2009
51-Sb-127	03/2012	**82-Pb-201**			
52-Te-127	05/2012	82-Pb-212	02/2011(2)		

"…… standing on the shoulders of giants" (in letter dated 1675 written by Isaac Newton)

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IAEA Research Coordination Meeting on "Gamma-ray standards for detector calibration", Rome, 11-13 June 1987
Back: Peter Christmas (NPL),
Dick Helmer (INEL), Nelcy Coursol (LMRI),
Klaus Debertin (PTB)
Front: Alan Nichols (UKAEA, Winfrith),
Walter Bambynek (CEC-JRC, Geel),
R. Jedlovsky (OMH), Frank Schima (NIST),
Y. Yoshizawa-san (Hiroshima University)
Alex Lorenz (IAEA, Nuclear Data Section)



IAEA Consultants' Meeting on "Gamma-ray standards for detector calibration", Grenoble, 30, 31 May 1985
Left to right:
Walter Bambynek (CEC-JRC, Geel),
Peter Christmas (NPL), Klaus Debertin (PTB),
Alan Nichols (UKAEA, Winfrith),
Alex Lorenz (IAEA, Nuclear Data Section),
Rene Vaninbroukx (CEC-JRC, Geel),
Alan Fudge (UKAEA, Harwell),
Dick Helmer (INEL)

elsewhere: T. Barta (OMH), Nelcy Coursol (LMRI), T. Yoshizawa-san (Hiroshima University, photographer)



Second IAEA Research Coordination Meeting on “Update of X- and gamma-ray decay data standards for detector calibration and other applications”, Braunschweig, 10-12 May 2000

Back row:

Alan Nichols (AEA Technology, Culham/Harwell, UK), Gabor Molnar (Institute of Isotope and Surface Chemistry, Budapest, Hungary), Stanislav Hlaváč (Slovak Academy of Sciences, Bratislava, Slovakia), Eckart Schönfeld (PTB, Braunschweig, Germany), Mike Herman, (IAEA-NDS, Vienna, Austria), José Maria Los Arcos (CIEMAT, Madrid, Spain)

Front row:

Andrzej Marcinkowski (Andrzej Soltan Institute for Nuclear Sciences, Warsaw, Poland), Otaviano Helene (Universidade de São Paulo, Brazil), Valery Chechev (V.G. Khlopin Radium Institute, St. Petersburg, Russia), Marie-Martine Bé (DAMRI/LNHB, Saclay, France), Dick Helmer (INEL, Idaho Falls, USA), Mike Woods (NPL, Teddington, UK)