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INDC INTERNATIONAL NUCLEAR DATA COMMITTEE

UPDATE OF X- AND γ-RAY DECAY DATA STANDARDS FOR DETECTOR CALIBRATION AND OTHER APPLICATIONS

Summary Report of the First Research Co-ordination Meeting

IAEA Headquarters, Vienna, Austria
9 - 11 December 1998

Prepared by

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July 1999

IAEA NUCLEAR DATA SECTION, WAGRAMERSTRASSE 5, A-1400 VIENNA

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Abstract

The discussions and conclusions of the First Research Co-ordination Meeting to Update X- and γ-ray Decay Data Standards for Detector Calibration are described in this summary report. The agreed list of radionuclides to be evaluated is given, along with the evaluation procedures and assignment of tasks among participants of the CRP.

July 1999

TABLE OF CONTENTS

Summary Report of the First Research Co-ordination Meeting on UPDATE OF X- AND γ-RAY DECAY DATA STANDARDS FOR DETECTOR CALIBRATION AND OTHER APPLICATIONS

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SUMMARY

CRP members reviewed the suggested radionuclides and decay data required for detector calibration and other applications, based on the findings of an IAEA Consultants' Meeting in November 1997. Agreed work programmes were formulated during the CRP Meeting and tasks defined clearly to achieve an updating of X- and γ-ray standards for detector calibration and other specific applications. The work programme is scheduled to be completed by 2001, with a mid-term review in early 2000.

BACKGROUND

The 1997 meeting of the International Nuclear Data Committee (INDC) strongly recommended that the Nuclear Data Section (NDS) place more emphasis on the development of improved nuclear structure data for "standards" applications. The main use of such standards data is in the calibration of X- and γ-ray detectors for the determination of both the energies and emission probabilities of X- and γ-rays of importance in nuclear technology. Other nuclear techniques (for example radiotherapy) suffer from a lack of high-energy calibration standards, and there is a need to provide such data for the calibration of γ-ray detectors up to 25 MeV. Following the INDC recommendation, and taking into account the results of a Consultants' Meeting held at IAEA Headquarters on 24 - 25 November 1997[1], the NDS has initiated a Co-ordinated Research Project (CRP) on the "Update of X- and γ-ray Decay Data Standards for Detector Calibration".

A.L. Nichols was elected to chair the CRP Meeting, supported by M. Herman as Secretary (see Appendices 1 and 2 for agenda and attendees, respectively). After appropriate debate, it was agreed that the new CRP should improve and extend the standards for decay data recommended by the previous CRP on the same subject (IAEA-TECDOC-619)[2]. This highly-respected database needs to be updated because of the new experimental data that became available after the earlier CRP had been completed. Members of the new CRP will review and modify the list of radionuclides most suited for detector calibration and consider the needs of such applications as:

- safeguards,
- materials analysis,
- environmental monitoring,
- medicine,
- waste management,
- dosimetry.

All of the recommended data must meet stringent quality requirements as by their very nature these parameters will propagate any inaccuracy directly into new measurements. An acceptable evaluation must be based on a well-defined set of precise procedures, supplemented with the judicious use of well-established theories.

OVERALL OBJECTIVE

The overall objective of the CRP is to improve detector calibration procedures in the most important nuclear applications including safeguards, materials analysis, environmental monitoring, medicine, waste management, dosimetry and spectroscopy.

SPECIFIC RESEARCH OBJECTIVES

The specific research objective of the CRP is to provide an extended and updated set of high-quality and internationally-accepted standards data (particularly radionuclide half-lives, X- and γ-ray energies and emission probabilities) for a number of radionuclides and specific nuclear reactions which are relevant for X- and γ-ray detector calibration. The CRP will:

1. revise the decay data of 35 radionuclides contained in the TECDOC-619 and supplement the database with 27 additional radionuclides, including some parent-daughter decay chains (see Table 1 for a detailed list). Decay data will be compiled, evaluated, and recommended (half-lives and X-, γ-ray and alpha-particle emission probabilities),
2. compile and evaluate γ-ray emission probabilities from several nuclear reactions appropriate for detector calibration in the range from 10 to 25 MeV,
3. analyze low-energy α-induced reactions, typically used in neutron sources of the Po-Be type, to obtain calibrated intensities of the 4.439 MeV γ line from ^{12}C,
4. provide a detailed description and recommended data for the coincidence calibration method,
5. investigate the feasibility and usefulness of including uncertainty correlations in the evaluation procedure.

Selection of radionuclides

Members of the CRP agreed that the basic database will contain decay data (radionuclide half-lives, X-, γ-ray and alpha-particle energies and emission probabilities) for the nuclei listed in Table 1. This selection is also motivated by the use of the listed nuclides in specific applications (see Table 2).

The original list of radionuclides suggested by the IAEA Consultants' Meeting was reviewed in detail[1]. Some nuclides were deleted (^{7}Be, ^{44}Ti, ^{124}I, ^{161}Tb, ^{195}Au and ^{239}Np) because of their lack of availability as standards, half-life considerations and/or inherent complexity, while others were added as more appropriate standards (^{123m}Te and ^{166}Ho (medical needs)).

Table 1. Radionuclides selected for inclusion in the recommended database

NUCLIDE	TECDOC-619	TO BE EVALUATED BY:
^{22}Na	X	INEEL
^{24}Na	X	INEEL
^{40}K		INEEL
^{46}Sc	X	INEEL
^{51}Cr	X	INEEL/PTB
^{54}Mn	X	INEEL/PTB
^{56}Mn		NPL/AEA
^{55}Fe	X	LPRI
^{59}Fe		LPRI
^{56}Co	X	NPL/AEA
^{57}Co	X	KRI
^{58}Co	X	LPRI
^{60}Co	X	INEEL
^{64}Cu		INEEL
^{65}Zn	X	INEEL
^{66}Ga		PTB
^{67}Ga		KRI
^{68}Ga		PTB
^{75}Se	X	LBL/PTB
^{85}Kr		NPL/AEA
^{85}Sr	X	PTB
^{88}Y	X	PTB
^{93m}Nb	X	KRI
^{94}Nb	X	NPL/AEA
^{95}Nb	X	INEEL

NUCLIDE	TECDOC-619	TO BE EVALUATED BY:
^{99}Mo		LPRI/ KRI
^{99m}Tc		LPRI
^{103}Ru		NPL/AEA
^{106}Ru-^{106}Rh		NPL/AEA
^{110m}Ag		INEEL
^{109}Cd	X	PTB
^{111}In	X	KRI
^{113}Sn	X	INEEL
^{125}Sb	X	NPL/AEA
^{123m}Te		LPRI
^{123}I		LPRI
^{125}I	X	PTB
^{129}I		KRI
^{131}I		LPRI
^{134}Cs	X	USP
^{137}Cs	X	INEEL
^{133}Ba	X	KRI
^{139}Ce	X	PTB
^{141}Ce		PTB
^{144}Ce		PTB
^{153}Sm		INEEL
^{152}Eu	X	USP
^{154}Eu	X	KRI
^{155}Eu	X	KRI
$^{166m}Ho/^{166}Ho$		PTB

NUCLIDE	TECDOC-619	TO BE EVALUATED BY:
^{170}Tm		KRI
^{169}Yb		PTB and LPRI
^{192}Ir		LBL/INEEL/USP
^{198}Au	X	PTB
^{203}Hg	X	NPL/AEA
^{201}Tl		PTB
^{207}Bi	X	LPRI
^{226}Ra (and daughters)		INEEL
^{228}Th (and daughters)	X	NPL/AEA
^{234m}Pa		NPL/AEA
^{241}Am	X	KRI
^{243}Am	X	NPL/AEA

INEEL - Idaho National Engineering and Environmental Laboratory (USA)
PTB - Physikalisch Technische Bundesanstalt (Germany)
NPL - National Physical Laboratory (UK)
LPRI - Laboratoire Primaire des Rayonnements Ionisants (France)
AEA - AEA Technology (UK)
KRI - V.G. Khlopin Radium Institute (Russia)
LBL - Lawrence Berkeley Laboratory (USA)
USP - University of Sao Paulo (Brazil)

Table 2. Justification for the selection of radionuclides

NUCLIDE	X- AND/OR γ-RAY STANDARD	DOSIMETRY STANDARD	MEDICAL APPLICATIONS	ENVIRONMENTAL MONITORING	WASTE MANAGEMENT	SAFEGUARDS
^{22}Na	P		x			
^{24}Na	P					
^{40}K				x		
^{46}Sc	P					
^{51}Cr	P		x			
^{54}Mn	P			x	x	
^{56}Mn			x			
^{55}Fe	P		x		x	
^{59}Fe			x			
^{56}Co	S					
^{57}Co	P		x			x
^{58}Co	P			x		
^{60}Co	P		x	x	x	x
^{64}Cu			x			
^{65}Zn	P			x	x	
^{66}Ga	S		x			
^{67}Ga			x			
^{68}Ga			x			
^{75}Se	S		x			
^{85}Kr				x		
^{85}Sr	P		x	x		
^{88}Y	P					
^{93m}Nb		x				
^{94}Nb	P					
^{95}Nb	P			x		
^{99}Mo			x			
^{99m}Tc			x			
^{103}Ru			x	x		

NUCLIDE	X- AND/OR γ-RAY STANDARD	DOSIMETRY STANDARD	MEDICAL APPLICATIONS	ENVIRONMENTAL MONITORING	WASTE MANAGEMENT	SAFEGUARDS
^{106}Ru - ^{106}Rh			x	x		
^{110m}Ag				x	x	
^{109}Cd	P			x		
^{111}In	P		x			
^{113}Sn	P					
^{125}Sb				x		
^{123m}Te	P					
^{123}I			x			
^{125}I	P	x	x			
^{129}I				x	x	
^{131}I		x	x	x		
^{134}Cs				x		
^{137}Cs	P	x		x	x	
^{133}Ba	S		x			
^{139}Ce	P			x		
^{141}Ce	P			x		
^{144}Ce	P		x	x		
^{153}Sm			x			
^{152}Eu	S			x	x	x
^{154}Eu				x	x	x
^{155}Eu	P			x	x	
^{166m}Ho			x(^{166}Ho)			x
^{170}Tm	P					
^{169}Yb			x			
^{192}Ir		x	x			
^{198}Au	P					
^{203}Hg	P					
^{201}Tl			x			
^{207}Bi			x			
^{226}Ra		x		x	x	

NUCLIDE	X- AND/OR γ-RAY STANDARD	DOSIMETRY STANDARD	MEDICAL APPLICATIONS	ENVIRONMENTAL MONITORING	WASTE MANAGEMENT	SAFEGUARDS
^{228}Th	P			x		
^{234m}Pa				x	x	
^{241}Am	P			x	x	x
^{243}Am					x	

P Primary standard for detector efficiency calibration

S Secondary standard for detector efficiency calibration

These radionuclides will be evaluated on the basis of the rules and procedures given in Appendix 3.

Selection of nuclear reactions

The following nuclear reactions were adopted as γ-ray calibration standards for high-energy applications:

1. $^{19}F(p,\alpha\gamma)^{16}O^*$
2. $^{12}C(p,p')^{12}C^*$
3. $^{11}B(p,\gamma)^{12}C^*$
4. $^{12}C(\alpha,\alpha')\ ^{12}C^*$
5. $^{16}O(p,\alpha p)^{12}C^*$

Their cross sections, and the energies and transition probabilities of the most prominent high-energy γ-lines will be evaluated and recommended. Other possible nuclear reactions need to be considered by Marcinkowski and co-workers (IPJ).

Coincidence Calibration

The method of coincidence calibration (see Appendix 5, contribution 10) can be used with sources which emit two cascading γs for the absolute determination of detector efficiency by comparing singles and coincidence intensities of the full energy peak measured simultaneously in an additional detector. The ratio of these two intensities, corrected for correlation effects, gives the absolute total efficiency of the detector directly. Absolute source activity cancels out in this ratio and does not need to be known, thus eliminating one of the major sources of uncertainty and allowing absolute calibration without absolutely calibrated sources. The following radionuclides have been used in the coincidence calibration method: ^{24}Na, ^{60}Co, ^{88}Y, ^{152}Eu and ^{207}Bi. Since all of these nuclides will be considered by the CRP as traditional calibration standards (see Table 1), application of the coincidence method will only require evaluations of γ–γ correlations. The coincidence method can also make use of high-energy photons from proton capture reactions such as $^{11}B(p,\gamma)^{12}C$, which will also be evaluated within the CRP. Thus, particle-γ–γ correlations will be evaluated so that the $^{11}B(p,\gamma)^{12}C$ reaction can be used in the coincidence calibration method.

Covariances

"Determining decay data correlations and calibrating the detector efficiency with multigamma-ray sources" (Appendix 5, contribution 13) describes the approach that will be adopted in a study of the ^{134}Cs gamma-ray emission probabilities. If possible, similar data analyses will be performed for ^{192}Ir and ^{152}Eu.

More technical details are given in Appendix 5, contributions 11-14, and an adequate format will be developed to publish the resulting data.

RECOMMENDED DATA OUTPUTS

The recommended decay data will be published as an IAEA-TECDOC containing the database in the form of tables, along with detailed descriptions of the status of these data and comments associated with the evaluation procedure. The electronic version of the database will be made available for on-line retrieval from the IAEA-NDS Web server. A preliminary format for the IAEA TECDOC is given in Appendix 4 (as discussed by CRP members).

RELEVANT MEASUREMENTS

Physikalisch Technische Bundesanstalt (PTB)

Measurements of half-lives and γ-ray emission probabilities carried out at the Physikalisch-Technische Bundesanstalt (PTB) since 1991.

1. Measurement of half-lives with ionisation chambers

1.1. Long-lived radionuclides

	Measurement	Last evaluation	reference
Kr-85	1978	1995	[1]
Ag-108m	1984	1995	[1]
Ba-133	1974	1995	[1]
Eu-152	1974	1996	[2]
Eu-154	1978	1996	[2]

1.2. Short-lived radionuclides

Ge-68	1992	1993	[3]
Rb-81	1997	1997	[4]
Sm-153	1996	1998	[5]
Re-186	1994	1994	[6]
Re-188	1997	1997	[7]
Rn-222	1995	1995	[4]

2. Measurement of γ-ray and X-ray emission probabilities

Se-75	[11]
Ga-68	[3]
Te-123m	[8]
Ce-141	[9]
Re-186	[6]
Yb-169	[10]
Sm-153	[5]
Np-237	[4]

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Khlopin Radium Institute (KRI)

Measurements of ^{226}Ra will be performed in 1999.

University of Sao Paulo

Measurements of single spectra for ^{152}Eu and ^{192}Ir and coincidence spectra for ^{134}Cs will be performed.

National Physical Laboratory (NPL)

Measurements of ^{129}I are planned for 1998-2001.

New measurements are needed for the following radionuclides due to significant discrepancies.

Nuclide	Quantity	Uncertainty needed
^{22}Na	half-life	0.01%
^{24}Na	half-life	0.02%
	relative γ-ray intensity (only for weak lines of 0.0004% to 0.00002%)	
^{46}Sc	half-life	0.01%
^{51}Cr	half-life	0.004%
	γ-ray emission probability	0.5%
^{75}Se	half-life	0.003%

Nuclide	Quantity	Uncertainty needed
^{65}Zn	half-life	0.008%
^{95}Nb	half-life	0.01%
	relative γ-ray intensity	5% for 234 keV and 0.5% for 724 keV, relative to 756 keV
^{137}Cs	half-life	0.06%, but need several measurements
	internal-conversion coefficient α(661)	0.5%

ACTIONS

1. Schönfeld: report to Herman on any recent and on-going PTB measurements of direct interest to the CRP (by 10 January 1999) – (see above).
2. All evaluators who have completed specific CRP evaluations to report on any measurement discrepancies and needs to Herman (by 10 January 1999).
3. All evaluators with reports etc. of relevance to CRP half-life evaluations to provide Woods with information for re-assessment/debate (by 1 March 1999).
4. Marcinkowski: review TECDOC-619 for other suitable candidates for high-energy γ-rays.
5. PTB/INEEL/LPRI ensure (via NDS) transfer of calculational routines / PC programs to all decay data evaluators (31 January 1999).
6. Bé: redraft "Rules for Evaluation" and resubmit to NDS by 15 January 1999 (see Appendix 3).
7. All evaluators: provide to Reher by 1 March 99 relevant information on X-ray emission probabilities :
   - completed evaluations,
   - data and references,
   - tools and software for generating X-ray data (fluorescence yields, etc).
8. Hlaváč: expand on the description of the coincidence method, giving more details on technical aspects and goals to be achieved before the next meeting (by 15 January 1999).
9. Vanin: provide draft of covariance chapter and submit to Herman (by 15 January 1999).
10. Herman: explore with CRP members an appropriate route for the generation of the evaluated and recommended data for TECDOC and CD-ROM issue (by March 2000).

INDIVIDUAL TASKS

CRP members provided brief descriptions of their various relevant programmes (see attachments) during the meeting. However, some of the proposals made in these attachments have been significantly modified as a consequence of subsequent discussions.

The final assignment of the tasks is given below:

M.-M. Bé (LPRI, Gif-sur-Yvette) work plan

1. evaluate decay schemes and transition probabilities for ^{55}Fe, ^{59}Fe, ^{58}Co, ^{99m}Tc, ^{123m}Te, ^{123}I, ^{131}I and ^{207}Bi;
2. evaluate together with PTB decay scheme and transition probabilities for ^{169}Yb;
3. evaluate together with Khlopin Institute decay scheme and transition probabilities for ^{99}Mo.

V. Chechev (KRI, St. Petersburg) work plan

1. evaluate decay schemes and transition probabilities for ^{57}Co, ^{67}Ga, ^{93m}Nb , ^{111}In, ^{129}I, ^{133}Ba, ^{154}Eu, ^{155}Eu, ^{170}Tm and ^{241}Am;
2. evaluate together with LRPI decay scheme and transition probabilities for ^{99}Mo.

R. Helmer (INEEL, Idaho Falls) work plan

1. evaluate decay schemes and transition probabilities for ^{22}Na, ^{24}Na, ^{40}K, ^{46}Sc, ^{60}Co, ^{64}Cu, ^{65}Zn, ^{95}Nb, ^{110m}Ag, ^{113}Sn, ^{137}Cs, ^{153}Sm and ^{226}Ra (and daughters);
2. evaluate together with PTB decay schemes and transition probabilities for ^{51}Cr and ^{54}Mn;
3. evaluate together with LBL and Sao Paulo decay scheme and transition probabilities for ^{192}Ir.

S. Hlaváč (SAS, Bratislava) work plan

1. select suitable sources for application in the coincidence calibration method;
2. examine and select statistical methods needed for evaluation of angular correlations in decay data;
3. perform evaluation of angular correlations for selected sources;
4. prepare detailed description of the coincidence method.

A. Marcinkowski (IPJ, Warsaw) work plan

1. evaluate cross sections for the production of γ-rays with energies of 6129.39 and 7117.0 keV in the ^{19}F(p,α γ)^{16}O* reaction;
2. evaluate cross sections for the production of γ-rays with energies of 4.44 and 15.11 MeV from ^{12}C*;
3. prepare a list of reactions suitable for the production of ^{12}C* and to compile and evaluate cross sections for these reactions including inelastic proton scattering on ^{12}C and radiative proton capture on ^{11}B (to evaluate emission probabilities of γ-rays with energies of 4.44 and 15.11 MeV from ^{12}C*).

O. Helene (University of Sao Paulo) work plan

1. develop procedures for the determination of covariances between decay data as well as between decay data and fundamental constants;
2. perform high quality measurements of the single spectra for ^{152}Eu and ^{192}Ir, and coincidence spectra for ^{134}Cs;

3. evaluate together with INEEL/LBL decay data for ^{134}Cs, ^{152}Eu and ^{192}Ir (taking into account all available experimental information) along with related covariances.

D. Reher (IRMM, Geel) work plan

1. based on the available data from CRP members (e.g. PTB, LPRI, INEEL), perform complementary literature search and collect relevant X-ray emission probabilities for all nuclides in Table 1 where these data are relevant;
2. with the support of PTB and LPRI, establish a set of tools for the calculation of auxiliary data, such as EC probabilities, fluorescence yields, etc;
3. in co-operation with PTB, LPRI and others, evaluate X-ray emission probabilities for the nuclides in Table 1 where relevant for specific applications.

E. Schönfeld (PTB, Braunschweig) work plan

1. evaluate decay schemes and transition probabilities for ^{66}Ga, ^{68}Ga, ^{85}Sr, ^{88}Y, ^{109}Cd, ^{125}I, ^{139}Ce, ^{141}Ce, ^{166m}Ho, ^{166}Ho, ^{198}Au and ^{201}Tl;
2. evaluate together with INEEL decay schemes and transition probabilities for ^{51}Cr and ^{54}Mn;
3. evaluate together with LPRI decay scheme and transition probabilities for ^{169}Yb;
4. evaluate together with LBL decay scheme and transition probabilities for ^{75}Se;
5. evaluate atomic shell data and K- and L-X-ray emission probabilities (calculated using the adopted atomic shell data and nuclide specific data).

M. Woods (NPL, Teddington) work plan

1. collect the published half-life data for all radionuclides in Table 1;
2. revisit statistical data analysis package and evaluate half-lives for all nuclides in Table1;
3. evaluate decay schemes and emission probabilities for ^{56}Mn, ^{56}Co, ^{85}Kr, ^{94}Nb, ^{103}Ru, ^{106}Ru-^{106}Rh, ^{125}Sb, ^{203}Hg, ^{228}Th (and daughters), ^{234m}Pa and ^{243}Am.

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2. "X-ray and Gamma-ray Standards for Detector Calibration", IAEA-TECDOC-619, IAEA, Vienna, 1991.

APPENDIX 1

First Research Co-ordination Meeting on the

Update of X- and Gamma-Ray Decay Data Standards for Detector Calibration

IAEA Headquarters, Vienna, Austria
9 - 11 December 1998

AGENDA

Wednesday, 9 December

09:30 – 10:00 Opening Session
- Opening address (D.W. Muir, Head, IAEA Nuclear Data Section, M. Herman, IAEA Nuclear Data Section)
- Election of Chairman
- Adoption of Agenda

10:00-10:15 Scope of the CRP – M. Herman

10:15-12:30 Presentations by the participants (related activities, current status of evaluations, needs)

12:30-14:00 Lunch Break

14:00-15:00 Presentations by the participants (cont.)

15:00-17:00 Selection of radionuclides and calibration reactions

17:00-18:00 Assignment of tasks

Thursday, 10 December

09:00-11:30 Discussion and adoption of evaluation procedures

11:30-12:30 Discussion of the needs for new experiments

12:30-14:00 Lunch Break

14:00-16:00 Discussion and adoption of the layout and format of the final document

Friday, 11 December

09:00-12:30 Discussion of the scope of the planned CRP on decay data for actinides

12:30-14:00 Lunch Break

14:00-17:00 Drafting and adoption of the Meeting report
Final discussion

APPENDIX 2

First Research Co-ordination Meeting on the

Update of X- and Gamma-Ray Decay Data Standards for Detector Calibration

IAEA Headquarters, Vienna, Austria
9 - 11 December 1998

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APPENDIX 3

Evaluation rules and guidance

(excerpt from: CEA/LPRI - Table of Radionuclides, ISBN 2 7272 0202 6)

Two main groups of data sources are used to obtain the recommended data :

- specific values evaluated from all available original publications (for example : half-life),
- compiled data already evaluated by specialists (for example : Q-values), if a new experimental value exists, it may be taken into account. In this case, the corresponding reference is mentioned in the reference list of this radionuclide.

1 Rules for evaluation

For simplification, all intermediate stages in compilation and evaluation are not presented. These stages essentially comprise the following :

- critical analysis of published results and, if necessary, correction of these results to account for more recent values hitherto unavailable to the experimentalists. Results without associated uncertainties are, as a rule, not used. The rejection of some values is always discussed ;
- for private communications, the only ones used are those for which we have all the necessary information directly from the scientist carrying out the measurements ;
- estimation of new uncertainties affecting individual results ;
- determination of the best value and of the standard uncertainty.

1.1 Evaluation of uncertainties

Definitions by the "Guide to the expression of uncertainty in measurement" [1]:

Uncertainty (of measurement) : a parameter, associated with the result of a measurement, that characterizes the dispersion of the values that could reasonably be attributed to the measurand.

Standard uncertainty : uncertainty of the result of a measurement expressed as a standard deviation.

Type A evaluation (of uncertainty) : method of evaluation of uncertainty by the statistical analysis of a series of observations.

Type B evaluation (of uncertainty) : method of evaluation of uncertainty by means other than the statistical analysis of a series of observations.

The uncertainties given by authors are re-evaluated by combining the standard uncertainties σ_A and σ_B by using the general law of variance propagation:

$$u_c = \sqrt{\sigma_A^2 + \sigma_B^2} \tag{1}$$

where: u_c : combined standard uncertainty
σ_A : type A standard deviation
σ_B : type B standard uncertainty

When the authors give insufficient information about their uncertainty calculations, the combined uncertainty u_c, may be estimated by the evaluator, based on his knowledge of the measurement method(s).

1.2 Determination of the best value and of the associated uncertainty.

1.2.1 Case of results obtained by one author using one method:

In this case the procedure is normally carried out by the author himself. Sometimes only the final result, for the mean value and the combined standard uncertainty, is given in the original publication. If details are known, the procedure is the following:

If there are n individual values a_i $(i = 1 \cdots n)$, the best value is the arithmetical mean:

$$\bar{a} = \sum_{i=1}^{n} \frac{a_i}{n} \tag{2}$$

with the type A standard deviation:

$$\sigma_A(\bar{a}) = \left[\frac{\sum_i (a_i - \bar{a})^2}{n(n-1)}\right]^{1/2} \tag{3}$$

If there are m contributions σ_{Bj} (j $= 1 \cdots$ k) to the type B standard uncertainty, independent of each other:

$$\sigma_B(\bar{a}) = \left[\sum_{j=1}^{m} \sigma_{Bj}^2\right]^{1/2} \tag{4}$$

The combined standard uncertainty is:

$$u_c(\bar{a}) = \sqrt{\sigma_A^2(\bar{a}) + \sigma_B^2(\bar{a})} \tag{5}$$

The recommended value is:

$$a = \bar{a} \pm u_c(\bar{a}) \tag{6}$$

1.2.2 Case of results obtained by several authors employing the same method:

If there are n individual values $\bar{a}_i$ $(i = 1 \cdots n)$, each having a standard deviation σ_{Ai} and a type B uncertainty σ_{Bi} the best value is obtained by taking the mean weighted by the inverse of the variances.

$$\bar{a} = \frac{\sum_j (\bar{a}_j / \sigma_{Ai}^2)}{\sum_j (1/\sigma_{Ai}^2)} \tag{7}$$

The associated values u_c, σ_A, σ_B are :

$$\sigma_A(\bar{a}) = \left[\sum_i (1/\sigma_{Ai}^2)\right]^{-1/2} \tag{8}$$

$\sigma_B(\bar{a}) = \sum_i (\sigma_{Bi})_{min}$ *or* $\sigma_B(\bar{a}) = \sqrt{\sum_i (\sigma_{Bi})_{min}^2}$ or $\sigma_B(\bar{a}) = (\sigma_B)_{min}$
depending on the individual case. However, $\sigma_B(\bar{a})$ cannot be less than the smallest σ_{Bi}.

Finally σ_A and σ_B are combined quadratically:

$$u_c(\bar{a}) = \sqrt{\sigma_A^2(\bar{a}) + \sigma_B^2(\bar{a})} \tag{9}$$

The recommended value is:

$$a = \bar{a} \pm u_c(\bar{a}) \tag{10}$$

1.2.3 Case of results obtained by different methods:

When different methods have been applied, a weighted average is calculated using the combined uncertainties of the individual values as weights.

If there are n independent values a_i, each having its own combined standard uncertainty u_{c_i}, then to each value, a weight p_i proportional to the inverse of the square of its individual u_{c_i} can be assigned.

$$a_w = \frac{\sum_{i=1}^n p_i \, a_i}{\sum_{i=1}^n p_i} \tag{11}$$

where the weights are $p_i = 1/u_{c_i}^2$.

An internal and an external uncertainty can be assigned to the mean value [2][3]:

$$\sigma_{int}(a_w) = \left[\sum_i (1/u_{c_i}^2)\right]^{-1/2} \tag{12}$$

The internal variance $\sigma_{int}^2(a_w)$ is the expected uncertainty of the mean, based on the individual *a priori* variances $u_{c_i}^2$ (by uncertainty propagation).

The external uncertainty is also calculated :

$$\sigma_{ext}(a_w) = \left[\frac{\sum (a_i - a_w)^2 / u_{c_i}^2}{(n-1) \sum (1/u_{c_i}^2)} \right]^{1/2} \tag{13}$$

The external variance $\sigma^2_{ext}(a_w)$ includes the scatter of the data and is based on the amount by which each a_i deviates from the mean, measured as a fraction of its given uncertainty u_{c_i}.

The ratio :

$$\sigma_{ext}/\sigma_{int} = \sqrt{\chi^2/(n-1)} \tag{14}$$

is a measure of the consistency of the data [2][3]. If this ratio is significantly greater than unity, this may be taken as a hint that at least one of the input data has an underestimated u_{c_i} which, probably, should be enlarged.

The method of limitation of the relative statistical weight [3][4] is recommended. When there are three or more values, the uncertainty of a value contributing more than 50 % to the total weight is increased to give a contribution less than 50 %. The weighted average is recalculated and used as final value if the $\chi^2/(n-1)$ value for this data set is < 2. If $\chi^2/(n-1) \leq 1$ the recommended value is :

$$a = a_w \pm \sigma_{int}(a_w) \tag{15}$$

If $1 < \chi^2/(n-1) \leq 2$,

$$a = a_w \pm (\text{the larger of } : \sigma_{int}(a_w) \text{ and } \sigma_{ext}(a_w)) \tag{16}$$

If the $\chi^2/(n-1)$ value is > 2, the weighted or unweighted mean is chosen, depending on whether or not the uncertainties of the average values make them overlap with each other. In either case, the uncertainty can be enlarged to cover the most accurate value.

The parameters evaluated according to these rules are half-lives, energies of emitted radiations, number of emitted particles, and some internal conversion coefficients.

The remaining values given in these tables are generally taken from compilations.

1.3 Balancing of decay schemes

All the probabilities for transitions and emitted radiations correspond to balanced schemes. This procedure offers the advantage of having a consistent set of values.

This balance implies obvious relationships such as the following:

- in each horizontal plan of a decay scheme the sum of the transition probabilities for all the cut transitions (α, $\geq \gamma$, EC) is equal to 1 (or 100 %). This is also valid for the highest cut where only (γ or α, EC) transitions are cut, and for the lowest cut, where sometimes only γ transitions are cut;

- for an excited level, the sum of the transition probabilities for transitions feeding this level (γ, β, ...) is equal to the sum of transition probabilities (including conversion electrons) of those transitions starting from this level;

- in the general case, where the relative emitted photon numbers $P(rel)_{\gamma_i}$ of the γ emission are known with respect to one of them, such as γ_1, and if we assume no

feeding to the ground state from α, β, and EC transitions, all the absolute emitted photon numbers for the γ emissions P(abs)$_{\gamma_i}$ can be calculated by :

$$P(abs)_{\gamma_i} = P(rel)_{\gamma_i} \times \frac{1}{k} \tag{17}$$

where k, called the "normalization factor", is deduced from :

$$k \sum_i P(rel)_{\gamma_i}(1 + \alpha_{t_i}) = 1 \tag{18}$$

where the sum is only over the γ-transitions feeding the ground state.

2 Compilations

2.1 β and electron capture transitions

The β-transition energies are, depending on the individual case, evaluated on the basis of experimental data (maximum β energies), or derived from disintegration energies supplied by the AUDI and WAPSTRA Tables [5], and the γ-transition energies. The average β energies are generally computed [6], and the values of lg ft are calculated from the tables of GOVE and MARTIN [7], as well as ϵ/β^+ if possible.

The energies of electron capture transitions are derived from the disintegration and γ energies. In the absence of experimental values, the probabilities for capture $P_K, P_L \ldots$, are calculated using ratios of the radial components of electron wave functions [8],[9],[10], and corrective terms for the exchange $X^{L/K}$ [11],[12],[13],[14],[15] taken from tables.

2.2 γ transitions

The internal conversion coefficients of pure transitions are evaluated and compared with theoretical values [16],[17] which are sometimes preferred, if the experimental values are too uncertain. The theoretical values are deduced from the RÖSEL et al. tables interpolated with a cubic spline method for $30 \leq Z \leq 104$ and from BAND et al. for $Z < 30$. The uncertainties taken for theoretical values are estimated to be 3%.

For some M1 and E2 transitions the internal conversion coefficients are calculated according to [18] in order to account for penetration effects.

The internal conversion coefficients of multipole transitions (e.g.: M1 + E2) are sometimes derived from tables, with the use of experimental mixing ratios:

$$\alpha_i(M1 + E2) = (1 - \delta^2)\ \alpha_i\ (M1) + \delta^2 \alpha_i\ (E2) \tag{19}$$

i = K, L, ..., t.

2.3 Level spins and parities

Level spins and parities are generally extracted from the Nuclear Data Sheets [19].

2.4 Constants of the atomic shell

The K-shell fluorescence yields ω_K and their uncertainties are taken from the evaluation of BAMBYNEK et al. [20],[21],[22] with uncertainties going from 1 % ($Z > 35$) to 10 % ($Z = 5$), and from subsequent experimental results.

The mean L-shell fluorescence yields $\bar{\omega}_L$ are taken from the evaluation of SCHÖNFELD et al. [23]. This evaluation use experimental values [24],[25],[26] and theoretical values [27]. The relative uncertainties are ≤ 4 % (for $Z > 29$).

The mean M-shell fluorescence yields $\bar{\omega}_M$ are taken from the fit made by HUBBELL [26],[28], based on experimental values.

The relative X-ray emission rates $K\beta/K\alpha$ are taken from SCHÖNFELD et al. [23], and the $K\alpha_1/K\alpha_2$ from the theoretical values of SCOFIELD [29]. The uncertainties are assumed to be of the order of 1 %.

The X-ray radiation energies are taken from the tables of BEARDEN [30].

The relative emission probabilities of K-Auger electron groups are deduced from the X-ray ratio [23] with an uncertainty the order of 3 %.

The energies of the K and L-Auger electrons are taken from the table of LARKINS [31].

The mean number of vacancies created in the L shell (with K hole) n_{KL} and in the M shell (with L hole) $\bar{n}_{LM}$ are estimated from the preceding values.

2.5 m_0c^2 energy

The m_0c^2 energy is taken as 510,999 06 (15) keV given by the CODATA Group (1986) [32].

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APPENDIX 4

Proposed Layout of TECDOC

TECDOC report will be assembled at NDS from the contributions provided by the participants. This document will evolve throughout the three years, and CRP participants should aim towards the following structure (name of the person responsible for the section is given between brackets):

1. **RECOMMENDED DATA**
   - 1.1 Half-lives (Woods)
   - 1.2 X-ray standards for detector calibration/ordered by energy and radionuclide (Reher/Schönfeld)
   - 1.3 γ-ray standards for detector calibration/ordered by energy and radionuclide (all)
   - 1.4 Covariances for selected γ-ray standards (Vanin)
   - 1.5 Nuclear reactions for detector calibration (Marcinkowski)
   - 1.6 Coincidence calibration (Hlaváč)

2. **INTRODUCTION/SUMMARY**
   - 2.1. Objectives of the CRP
   - 2.2. Achievements and conclusions

3. **EVALUATION PROCEDURES**
   - 3.1 Half-lives (Woods)
   - 3.2 X-ray standards (Reher/Schönfeld)
   - 3.3 Gamma-ray standards (Bé)
   - 3.4. Covariances (Vanin)
   - 3.5. Coincidence calibration (Hlaváč)

4. **EVALUATION SHEETS**
   - 4.1. Radionuclides (all evaluators)
   - 4.2. Nuclear Reactions (Marcinkowski)
   - 4.3. Coincidence calibration data (Hlaváč)

5. **REMAINING DISCREPANCIES**
   - 5.1. Half-lives (Woods)
   - 5.2. X-ray standards (Reher/Schönfeld)
   - 5.3. γ-ray standards (all evaluators)
   - 5.4. Nuclear reactions (Marcinkowski)

6. **LIST OF PARTICIPANTS** (Herman)

APPENDIX 5

Technical input

Readers, please note:

the attachments were presented by CRP members

at the beginning of the meeting - subsequent discussions

have resulted in modifications to proposed work plans

that are NOT included in these texts.

Presentation of the Decay Data Group Activities at LPRI. (Links with International Evaluation Works)

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The Primary Ionizing Radiation Laboratory (LPRI) is affiliated with the French National Bureau of Metrology (BNM) which is responsible for organizing metrology in France. The LPRI is in charge of the establishment, preservation and improvement of national standards for the units used in ionizing radiations measurements. The LPRI is also a laboratory of the Commisariat à l'Energie Atomique (CEA) which is in charge of all the activities relating to atomic energy.

The LPRI working program comprises: evaluation of decay data, absolute activity measurements, X- and gamma-ray spectrometry, development of new measuring techniques, etc.

In the field of decay data evaluation, LPRI is primarily working on the following three subjects:

1) NUCLEIDE software

The evaluation of decay data for the "Table de Radionucléides" by BNM-CEA/LPRI began in 1974, continued to 1987 and four volumes were published.

NUCLEIDE is the computerized form of this "Table de Radionucléides".

The NUCLEIDE software was entirely developed by LPRI with the objectives of making it easier to update and add data and, obviously, to offer easy access to the nuclear and atomic decay data to the user by "click on the button" facilities.

The aim of this Table is to provide recommended data for nuclides of special interest for metrology or practical applications like nuclear medicine, monitoring and reactor shielding, etc.

Primary recommended data comprise half-lives, decay modes, X-rays, gamma-rays, electron emissions, alpha- and beta-particle transitions and emissions, and their uncertainties. All the references used for the evaluations are given.

In order to update the data of the nuclides already present and to add new evaluations, the Laboratoire Primaire des Rayonnements Ionisants (LPRI, France) and the Physikalisch-Technische Bundesanstalt (PTB, Germany) established a cooperative agreement; they were then joined by the Idaho National Engineering and Environmental Laboratory (INEEL, USA), the Lawrence Berkeley National Laboratory (LBNL, USA) and the Khlopin Radium Institute (KRI, Russia). This international collaboration is based on an informal agreement, the initial work of this group was to discuss and to agree on a methodology to be used in these evaluations. The data and associated uncertainties were evaluated from all available experiments and taking into account theoretical considerations.

The main steps for the evaluation of the data and their uncertainties are:

- a critical analysis of all available original publications in order to accept or not each value and its uncertainty reduced to the combined standard uncertainty ;
- the determination of the best value which is either the weighted or the unweighted average of the retained values, this is decided after examination of the reduced χ^2 value. With a weighted average, each weight is limited to 50%. The uncertainty, designated uc, is the greatest of the internal or external uncertainty values. For a discrepant set of data, it may be expanded to cover the most precise input value.

Following this works:

- a CD-ROM with the NUCLEIDE software and the complete database will be distributed at the beginning of 1999;
- a report with the new evaluated nuclides will be published in February 1999. The layout of this report is the same as those of the previous Table (see Bi-207 as example). It will include:

Na-22, K-40, Ce-139, Co-60, Zn-65, Nb-95, Zr-95, Sn-113, Cs-137, Al-26, Se-75, Re-188, Ir-192, Ir-194, Ge-68, Ga-68, I-125, Ce-141, Fe-55, Bi-207, Co-58, Te-123m, C-14, Cl-36, H-3, In-111, S-35.

2) Internal Conversion Coefficients

Up to now LPRI has used the Rösel et al. tables and an interpolation program (working under Windows PC) to calculate the ICC values.

Work to create a database with all the experimental values has begun with the purpose of comparing these values with those deduced from Rösel et al., Band et al. and Hager et al. tables.

Recently, a group which includes Dr. Band and Dr. Raman has developed a new program based on the Dirac-Fock atomic model. This program tries to resolve the cases where the theoretical values given by the previous models and the experimental values differ by several percents, for example in the case of low gamma energy and high L transitions.

In order to check this new program and use it to calculate easily the ICC, contacts with Dr. Band have been established.

3) Nuclide evaluations

The status is:

- Finished evaluations: Fe-55, Bi-207;
- On progress: Mo-99, Tc-99m, Tc-99
- Planned for 1999: Yb-169 (this evaluation will be done with PTB and after the end of the Euromet exercise);
- Later: Fe-59, Kr-85, I-129, I-131.

Report on the activity of the Radionuclide Data Center 1996 - 1998

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1. Measurements of Decay Data

Experimental determination of KX-ray and soft gamma-ray emission probabilities in decays of ^{153}Gd, ^{155}Eu and ^{169}Yb (finished).

2. Horizontal Evaluations of Decay Data

1996 half-life evaluations for 42 radionuclides used for X-ray and gamma-ray detector calibration (by Chechev, V.P.).

Evaluations of decay data within the framework of DDEP cooperation: ^{3}H, ^{14}C, ^{35}S, ^{36}Cl, ^{111}In (finished), ^{57}Co, ^{99}Mo, ^{99m}Tc, ^{170}Tm (in progress).

Re-evaluating of half-lives, X- and gamma-ray energies and emission probabilities for ^{44}Ti, ^{57}Co, ^{67}Ga, ^{85}Kr, ^{99}Mo, ^{99m}Tc, ^{111}In, ^{129}I, ^{133}Ba, ^{154}Eu, ^{155}Eu, ^{170}Tm, ^{203}Hg (within the framework of CRP task, in progress)

3. Other Related Activities on Nuclide Data

Development of Database for NUCLIDE GUIDE and NUCLIDE CHART, PC system, containing short information of all known nuclides. For radioactive nuclides it includes the evaluated half-lives, mass excesses, decay energies (with uncertainties) and some other characteristics (radiation energies and emission probabilities) without uncertainties.

Making (together with Atominform, Moscow) the International Chart of Nuclides (wall-type) on the basis of the above Database and nuclear data reported by CNDC (China), M.S. Antony (France) and NDC JAERI (Japan) (the first version is finished).

ISTC Project No.1227 "TRANSURANIUM RADIONUCLIDES: PRODUCING HIGHLY ENRICHED ISOTOPE SAMPLES, MEASURING EMISSION PROBABILITIES OF RADIATIONS AND DECAY DATA EVALUATION" is submitted for consideration to the International Science and Technology Centre from three participating institutions (VNIIEF, Sarov, and KRI and VNIIM, St.Petersburg). It includes measurements of the alpha- and gamma-ray emission probabilities for ^{238}Np, ^{241}Am, ^{242m}Am, $^{243-248}$Cm and re-evaluations of decay data for the 20 transuranium radionuclides.

Translating to English the Russian reference book "Evaluated Values of Nuclear Characteristics of Transuranium Nuclides" by V.P.Chechev et al., Energoatomizdat, Moscow, 1988 (in progress).

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Search for optimum approach to evaluating data with different consistency (A Proposal for Producing Recommended Averages and Uncertainties)

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The derivation of a recommended value and uncertainty from a discrepant set of data is an important problem for evaluators. In recent years a number of data evaluation procedures have been proposed. A review and testing of these methods has been made in /1,2/.

At present it is recognized that one of the very useful methods for evaluating data is the procedure of a limitation of relative statistical weights (LWM) /3/. On the basis of this method a computer program LWEIGHT has been developed by E. Browne /4/ and T.D. MacMahon. It uses also the Chauvenet's criterion /5/ for rejecting outliers, and works successfully in evaluating discrepant data when $1<\chi^2/(n-1)<2$. However for a number discrepant data sets with $\chi^2/(n-1)>2$ the LWEIGHT choice of an unweighted mean gives rise to doubt (see my "Remarks on the LWEIGHT program").

Now there is a possibility to make corrections in programs and to unite the best findings in searches for an optimum approach to evaluating data with different consistency. This can be achieved by testing different statistical procedures, and proposing the use of tS /6/ or a modified Bayesian procedure (MBAYS) /2/ to give the most reliable uncertainties for recommended values, as well as using the LWM method of CRP in 1991 and the LWEIGHT program.

I have examined the data sets with different consistency which cover measurement results for half-lives of about 50 radionuclides /6,7/. There is not a single universal statistical procedure for producing recommended averages for all sets of data that are both consistent and discrepant. Therefore we have developed, in collaboration with A. Egorov, the computer program EV1NEW that uses several procedures /1,2/ of the program LWEIGHT and includes the LWM method as one of the component steps.

The principles of our program consist in a successive motion from the initial collection of all available experimental results to the final data set which is used for calculating the recommended value. The final uncertainty depends on a degree of the data inconsistency /7/. For rejecting some results this program gives only RECOMMENDATION: the decision to adopt or reject data is made by the EVALUATOR ("Yes" or "No").

The first step from the initial data set ("0") to the data set "1" consists, if necessary, in omitting unreliable or revised later measurement results. The next step ("1" $\rightarrow$ "2") is connected with the estimation of contributions of the different experimental results to the total χ^2 value. In this step the evaluator can reject one or two statistical outliers (see the example for ^{155}Eu, Table 1). In forming the data set ("3") the possible adjustment of the uncertainty of one of the results occurs due to applying the above-mentioned LWM method.

Table 1

Experimental values of ^{155}Eu half-life (in days)

Reference	Data set "1"	Data set "2"	Data set "3"
	χ^2 =334.9 $(\chi^2)_6{}^{0.05}$ =14.1	χ^2 =6.14 $(\chi^2)_5{}^{0.05}$ = 12.6	χ^2 =5.68 $(\chi^2)_5{}^{0.05}$ =12.6
98Si**	1739(8)	1739(8)	1739(8)
93Th04	1735(22)	1735(22)	1735(22)
92Un01	1739.0(5)	1739.0(5)	1739(7)[b]
83Wa26	1737(23)	1737(23)	1737(23)
74Da24	1708(18)	1708(18)	1708(18)
72Em01	1812(4)	Omitted [a]	-
72Su09	1653(51)	1653(51)	1653(51)
70Mo23	1698(74)	1698(74)	1698(74)

[a] The value from 72Em01 has been omitted on the basis of statistical considerations.

[b] The rule "of 50%" weight leads to a significant increase of the 92Un01 uncertainty.

It can be noticed that the rule of "50 % weight" has not been used in /6,7/ to evaluate some radionuclide half-lives (in particular, ^{155}Eu). This possible deviation from the LWM rule is foreseen in the program for those radionuclidic measurements associated with a considerable improvement of experimental technique.

The final phase of our program is directed to obtaining the "best" value through the resulting "3" set of selected data. The program compares results obtained with the different statistical procedures /1,2/ but chooses the weighted mean as an evaluated value expanding the final uncertainty in dependence of the χ^2 value. A modification of this program includes the five ways of adjusting the final uncertainty with increasing χ^2 and the three modes of classification of discrepant data sets by means of comparing χ^2 to the tabulated value $(\chi^2)_{n-1}{}^{0.05}$ for the significance level 0.05 (Table 2).

Table 2

Distribution of 48 half-life data sets: degree of discrepancy determined by χ^2 value and recommended uncertainty of the evaluated value.

Mode of set	Degree of data discrepancy (on χ^2 value)	Number of sets	Recommended uncertainty
	$\chi^2 \leq (n-1)$	12	σ internal uncertainty
	$(n-1)<\chi^2\leq(\chi^2)_{n-1}^{0.05}$	7	$S=\sigma\times[\chi^2/(n-1)]^{1/2}$ external uncertainty
	$(\chi^2)_{n-1}^{0.05}<\chi^2\leq10(\chi^2)_{n-1}^{0.05}$	25	$t_{n-1}^{0,68}\times S$ expanding S for low n or $\sigma\times[\chi^2/(n-2)]^{1/2}$ MBAYS uncertainty
	$\chi^2>10(\chi^2)_{n-1}^{0.05}$	4	$\sigma\times[\chi^2/(n-3)]^{1/2}$ BAYS uncertainty

This approach is based on the computational study of Kafala et al. /2/ in which the MBAYS procedure was determined to be most reliable for evaluating discrepant data and on the assumption that the discrepancy of data is connected with partial or total incorrectness of the experimental uncertainties. The two extreme modes of expressing the final uncertainty of the weighted mean correspond to consistent data sets (σ) and greatly discrepant data sets when the Bayesian procedure is used, implying a random distribution of the experimental uncertainties. The intermediate cases (largest number) correspond to the use of an external uncertainty or the MBAYS procedure. It should be noted that the uncertainty values for the intermediate mode (3) (tS and MBAYS) almost coincide for $n\geq4$ and therefore have been united. t is the Student's coefficient for (n-1) degrees of freedom and the confidence level 0.68 (for details see /6,7/).

Half-lives of radionuclides corresponding to data sets with $\chi^2>10(\chi^2)_{n-1}^{0.05}$ can be recommended for additional measurements.

Also this approach avoids an unweighted mean which is not acceptable for averaging.

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Remarks on the LWEIGHT program

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The LWEIGHT program is successful in evaluating discrepant data when $1< \chi^2/(n-1)<2$. However in case of discrepant data sets with $\chi^2/(n-1)>2$ the LWEIGHT choice of an unweighted mean (UWM) rises doubts.

In many cases, of early measurement with large uncertainties, a choice of UWM leads, in fact, to the assignment of EQUAL WEIGHTS to both old results with small accuracy and the best experimental data.

As a consequence the recommended average does not agree with the best experimental results. Increasing the uncertainty of the recommended UWM to embrace latter results is not desireable as the obtained large error of the recommended value does not correspond the modern experimental accuracy.

Rejecting old experimental results leads to a weighted mean (WM) and improves the situation but this procedure brings in some amount of subjectivity.

Uniformity of evaluations is violated by using both WM and UWM for different evaluations. Increasing the final uncertainty for greatly discrepant data can be achieved with other methods (MBAYS and BAYS) or by the 1991 CRP method including the lowest uncertainty value for WM (but not for UWM).

Conclusion: UWM should not be used for averaging the values with different accuracy.

Evaluation of Decay Data: On-going Activities

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1 Introduction

A number of UK decay-data evaluation programmes are supported by the UK Industrial Management Committee (primarily through British Nuclear Fuels Limited) and the UK Atomic Energy Authority. Some of these evaluations have proved to be particularly complex (short-lived fission products), while others require the assessment of equivalent decay data for adjacent radionuclides because of a dearth of direct measurements. The decay-data defined below are split between requests for fission-product data (BNFL) and fusion activation products (UKAEA). While the fission-product studies are close to completion (within next 6 months), the work for the UKAEA will continue into 2000.

2 Fission Product Decay Data

BNFL staff requested improved decay-data evaluations for specific fission products on the basis of OECD-NEA discussions and agreed collaboration to prepare JEFF-3 (Joint Evaluated Fission and Fusion file for the NEA Data Bank). A set of 27 thermal fission products (plus short-lived daughters and related metastable/ground states) were identified as important from the point of view of radiotoxicity, fuel reprocessing, monitoring standards and delayed-neutron emissions (Table 1), and also because of a lack of adequate decay-data files. Hence, a two-year evaluation exercise has been underway since mid-1996 to produce a comprehensive set of recommended decay data for these nuclides based on published measurements and a well-defined evaluation procedure. Theoretical data for a further 35 short-lived fission products have also be considered for adoption from other sources (Table 2).

A list of 37 radionuclides evolved for discrete decay-data evaluation after a review of the requirements of the nuclear industry with respect to decay heat, recycling, reprocessing and delayed-neutron emissions; a detailed assessment was subsequently carried out, and these evaluation needs were reduced to 27 radionuclides (plus short-lived daughters and related metastable/ground states). The decay data for a number of these radionuclides have been evaluated from 1996 to 1998 and data files assembled (see Table 3 for the consistency of the recommended data), along with the assessment and evolution of recommended decay data for the other 35 short-lived fission products (see Table 2).

3 Activation Product Decay Data

The decay data files of approximately 50 radionuclides within the EAF library have been identified by Robin Forrest (UKAEA) as being problematic or incomplete when used for fusion reactor applications. These problematic radionuclides were extended, with the addition of related metastable states and daughter nuclides. Some of the data files do not contain any gamma-ray emissions, while others exhibit inconsistencies between the mean gamma energies and component radiations listed in the files. The aim of the work programme has been to undertake decay-data evaluations for these radionuclides, as specified by the UKAEA.

The recommended decay data exhibit good to excellent consistency, as indicated in Table 4, when Q-values and branching fraction data are compared with the discrete emission data (i.e. gamma rays, x-rays, Auger and conversion electrons, EC transitions and beta particles). These decay-data files are being assembled at regular intervals in ENDF-6 format for the construction of the Joint Evaluated Fission and Fusion library (JEFF-3) - Nuclear Energy Agency, OECD.

4 Concluding Remarks

Evaluation efforts continue within the UK to improve the recommended decay data for specific radionuclides of importance in both the power-based fission programme and R & D fusion studies. All resulting decay-data files are generated in ENDF-6 format as requested by the UK nuclear power industry, and checks are made to quantify the consistency and completeness of the recommended decay schemes (1). Suitable procedures have been developed and implemented over a period of approximately 20 years to achieve the desired credibility and quality of data (2).

An assessment has been made of the proposed decay-data evaluations for the IAEA-CRP to Update X- and Gamma-ray Decay Data Standards for Detector Calibration (3) in order to generate an appropriate work programme as the UK contribution. Decay-data re-assessments and evaluations (and gamma-ray emission probabilities in particular) could be undertaken for the following radionuclides:

> Fe-59, Co-56, Kr-85, Sr-85, Y-88, Nb-94, Ru-106/Rh-106, I-129, Eu-155 and Hg-203, with the work load shared between AEA Technology (A L Nichols) and NPL (S A Woods).

References

1 J.S. Backhouse and A.L. Nichols, Assessment and Evaluation of Decay Data for Nuclear Reactor Applications, Appl. Radiat. Isot., **49** (1998) 1393-1396.

2 A.L. Nichols, Evaluation of Activation Product Decay Data, Nucl. Instrum. Meth. Phys. Res., **A369** (1996) 516-522.

3 A. Nichols and M. Herman, Report on the Consultants' Meeting on Preparation of the Proposal for a Coordinated Research Project to Update X- and γ-ray Decay Data Standards for Detector Calibration, INDC(NDS)-378, May 1998.

Table 1: Fission Product Nuclides - Requirements for Evaluated Decay Data

Radionuclide	Priority	Importance
45-Rh-106*	high	Instrumentation for recycling
57-La-140	high	Fission product standard
62-Sm-147	high	Instrumentation for recycling
34-Se-79*	high	Radiotoxicity
40-Zr-93*	high	Radiotoxicity
50-Sn-126*	high	Radiotoxicity
51-Sb-127*	medium	Reprocessing
53-I-132*	medium	Reprocessing
52-Te-132	medium	Reprocessing
53-I-138	medium	Reprocessing/Delayed neutron emission
59-Pr-143	medium	Reprocessing
59-Pr-144*	medium	Reprocessing
65-Tb-161	medium	Reprocessing
35-Br-88	medium	Delayed neutron emission
35-Br-89	medium	Delayed neutron emission
35-Br-90	medium	Delayed neutron emission
37-Rb-94	medium	Delayed neutron emission
39-Y-98m*	medium	Delayed neutron emission
53-I-137	medium	Delayed neutron emission
39-Y-99	low	Delayed neutron emission
51-Sb-135	low	Delayed neutron emission
53-I-139	low	Delayed neutron emission
35-Br-87	low	Delayed neutron emission
35-Br-91	low	Delayed neutron emission
37-Rb-95	low	Delayed neutron emission
37-Rb-93	low	Delayed neutron emission
33-As-85	low	Delayed neutron emission

* Additional short-lived daughters and related metastable/ground state radionuclides were also evaluated.

Table 2: Other Important Short-lived Fission Products - Adoption of USENDF/B-VI Decay Data Unless Stated Otherwise

Radionuclide	Quoted Half-life (sec) - USENDF/B-VI	Continuum Spectra - Energy Range (keV)*		
		Gamma	Beta	Neutron
39-Y-104	0.12825	0(500) - 12730	0 - 12690	0 - 5510
39-Y-105	0.14688	0(500) - 10820	0 - 10790	0 - 6840
40-Zr-105	0.49263	0(500) - 8290	0 - 8260	0 - 2260
40-Zr-106	0.90709	0(500) - 6380	0 - 6350	0 - 2570
40-Zr-107	0.24295	0(500) - 9230	0 - 9200	0 - 3950
41-Nb-109	0.31537	0(500) - 8760	0 - 8730	0 - 5300
42-Mo-109	1.4085	0(500) - 6700	0 - 6670	0 - 1200
42-Mo-111	0.46637	0(500) - 8020	0 - 7990	0 - 2210
42-Mo-112	0.97537	0(500) - 6020	0 - 5990	0 - 2720
43-Tc-113	0.65238	0(500) - 7540	0 - 7510	0 - 4080
43-Tc-114	0 20226	0(500) - 10610	0 - 10580	0 - 4790
43-Tc-115	0.27044	0(500) - 8870	0 - 8840	0 - 5910
43-Tc-116	0.11549	0(500) - 11860	0 - 11830	0 - 6650
44-Ru-115	0.87844	0(500) - 7250	0 - 7220	0 - 1400
44-Ru-116	1.7004	0(500) - 5510	0 - 5480	0 - 2150
44-Ru-117	0.34277	0(500) -8500	0 - 8470	0 - 3180
44-Ru-118	0.66235	0(500) - 6530	0 - 6500	0 - 3680
44-Ru-119	0.19495	0(500) - 9290	0 - 9260	0 - 4440
45-Rh-118	0.31565	0(500) - 9970	0 - 9940	0 - 3410
45-Rh-120	0.17246	0(500) - 10770	0 - 10730	0 - 4830
45-Rh-121	0.24956	0(500) - 8790	0 - 8760	0 - 5990
46-Pd-121	0.64367	0(500) - 7560	0 - 7530	0 - 1520
51-Sb-141	No entry in ENDF/B-VI; theoretical and JENDL data			
57-La-152	0.28495	0(500) - 8810	0 - 8770	0 - 3980
58-Ce-153	1.4688	0(500) - 5820	0 - 5790	0 - 1620
58-Ce-154	2.0161	0(500) - 5010	0 - 4970	0 - 1640
58-Ce-158	No entry in ENDF/B-VI; theoretical and JENDL data			
59-Pr-156	0.37926	0(500) - 8690	0 - 8660	0 - 2790
59-Pr-157	0.38001	0(500) - 8130	0 - 8100	0 - 3590
60-Nd-157	2.4833	0(500) - 5560	0 - 5520	None
60-Nd-158	2.6949	0(500) - 5000	0 - 4970	0 - 320
60-Nd-159	0.64159	0(500) - 7150	0 - 7120	0 - 1230
60-Nd-160	0.78856	0(500) - 6350	0 - 6320	0 - 1830
61-Pm-159	3.0005	0(500) - 5650	0 - 5620	0 - 410
61-Pm-160	0.72892	0(500) - 7800	0 - 7770	0 - 1130

* Expressed in terms of incremental units of 10 keV starting from zero (first incremental energy step of continuum gamma spectra is from zero to 500 keV).

Table 3: Comprehensive Evaluations - Consistency of Fission-Product Data Sets (1996-98)

Radionuclide	Consistency (% Deviation)	Radionuclide	Consistency (% Deviation)
33-As-85	0.0988*	(51-Sb-126)	-0.0653
34-Se-79	0.0000	(51-Sb-126m)	-0.1714
(34-Se-79m)	-0.0962	(51-Sb-126n)	-0.3560
35-Br-87‡	*	51-Sb-127	-0.0431
35-Br-88	0.2554*	51-Sb-135	-0.0198*
35-Br-89	0.0534*	(52-Te-127)	-0.0037
35-Br-90	0.1331*	(52-Te-127m)	-0.0908
35-Br-91	0.0274*	52-Te-132	0.1077
37-Rb-93	-0.0182*	53-I-132	-0.0832
37-Rb-94+	*	(53-I-132m)	-0.3723
37-Rb-95+	*	53-I-137	0.1276*
(39-Y-98)	-0.0432*	53-I-138	-0.1955*
39-Y-98m	-0.2944*	53-I-139	0.0552*
39-Y-99	-0.0741*	57-La-140	-0.0108
40-Zr-93	1.2384	59-Pr-143	0.0000
(41-Nb-93m)	-0.3678	59-Pr-144	0.0382
45-Rh-106	-0.0243	(59-Pr-144m)	-0.0860
(45-Rh-106m)	-0.0487	62-Sm-147	-0.0023
50-Sn-126	0.0293	65-Tb-161	-0.0324

Additional short-lived daughter and related metastable/ground state radionuclides are in parenthesis, and were also evaluated.

* Beta-decay mode only.

‡ Evaluation underway.

+ Evaluation completed - awaiting data processing.

Table 4: Evaluated Decay Data - Fusion Activation Products (1996-2000)

Radionuclide	Half-life	Consistency (% Deviation)
7-N-17	4.17(4) sec	0.0724†
(25-Mn-58)	65.2(5) sec	-0.3450
25-Mn-58m*	2.7(6) sec	0.1037
31-Ga-77	13 sec	
33-As-82	14 sec(?)	
(33-As-82m)	19 sec(?)	
34-Se-79*	6.0(5) x 10^5 y	0.0000
34-Se-79m	3.90(2) min	-0.0962
38-Sr-87m	2.808(6) h	-0.0154
39-Y-96*	5.37(7) sec	-0.0151
(39-Y-96m)	9.62(15) sec	0.0079
(39-Y-96n)	(?)	N/A
41-Nb-100	1.4(1) sec	0.0733
(41-Nb-100m)	2.9(2) sec	-0.0167
43-Tc-97*	2.6(4) x 10^6 y	-0.0047
43-Tc-97m	90.2(11) d	0.0621
46-Pd-109	13.46(1) h	0.0090
(46-Pd-109m)	4.71(3) min	0.0367
46-Pd-112	20.3(2) h	-0.0306
(47-Ag-107m)	44.1(4) sec	-0.0525
(47-Ag-109m)	39.8(2) sec	-0.1869
(47-Ag-114)	4.7(1) sec	‡
47-Ag-114m*	0.00150(5) sec	‡
(47-Ag-115)	20.5(4) min	0.0434
47-Ag-115m*	18.6(8) sec	0.1918
48-Cd-107	6.52(2) h	-0.0289
49-In-112	14.7(7) min	0.1052
(49-In-112m)	20.7(1) min	-0.1202
56-Ba-129	2.38(11) h	-0.0730
56-Ba-129m*	2.14(5) h	0.0550
58-Ce-147	57(2) sec	0.0269

Radionuclide	Half-life	Consistency (% Deviation)
59-Pr-143	13.56(1) d	0.0000
59-Pr-144	17.28(2) min	0.0382
(59-Pr-144m)	6.9(7) min	-0.0860
59-Pr-150	6.1(4) sec	-0.6261
(61-Pm-152)	4.12(9) min	-0.3799
(61-Pm-152m)	7.5(1) min	-0.7796
61-Pm-152n*	14.4(7) min	-0.0401
(65-Tb-156)	5.17(12) d	-0.3867
65-Tb-156m*	24.4(10) h	0.5785
65-Tb-156n*	5.1(3) h	-0.0964
67-Ho-160**	25.3(7) min	-0.2337
67-Ho-160m	5.0(1) h	-0.5027+
67-Ho-160n**	2.9(2) sec	0.2220
67-Ho-161	2.48(12) h	0.0565
(67-Ho-161m)	6.77(6) sec	0.1297
(67-Ho-170)	43 sec(?)	
67-Ho-170m	2.8 min(?)	
72-Hf-178m	4.0(3) sec	‡
72-Hf-178n	31(1) y	‡
72-Hf-180m	5.5(1) h	‡
75-Re-191*	9.7(4) min	0.0000
75-Re-192*	6.2(8) sec	0.0566
76-Os-185	93.8(9) d	‡
(76-Os-190m)	9.9(4) min	‡
(76-Os-191m)	13.1(1) h	0.0520
76-Os-195*	6.5(6) min	-0.0396
77-Ir-187	10.5 h	
(77-Ir-190)	12.0(2) d	‡
(77-Ir-190m)	1.120(3) h	‡
77-Ir-190n	3.087(12) h	‡
(77-Ir-191m)	4.9 sec	
77-Ir-191n**	5.5 sec(?)	
(77-Ir-192)	74.2 d	

Radionuclide	Half-life	Consistency (% Deviation)
77-Ir-192m	1.5 min	
(77-Ir-192n)	241 y	
77-Ir-197	5.8 min(?)	
77-Ir-197m**	8.9 min(?)	
78-Pt-193*	50(9) y	2.0682
(78-Pt-193m)	4.34(3) d	-0.3390
(79-Au-192)	5.0 h	
79-Au-192m	0.029 sec(?)	
80-Hg-199m	42.1(9) min	‡
(82-Pb-201)	9.4 (1) h	‡
82-Pb-201m*	61(3) sec	‡
83-Bi-208*	3.68(4) x 10^5 y	0.0635
84-Po-208*	2.93(4) y	-0.0380

† Beta-decay mode only.

* No gamma lines in EAF/JEF library.

** No EAF/JEF data file.

\+ Datum for decay scheme with 124 gamma-ray transitions; with addition of a further 42 gamma rays not placed in the decay scheme, Consistency is -6.4818%.

‡ Evaluation completed - awaiting data processing.

Nuclides in parenthesis have not been requested, but were included for completeness.

N/A, not applicable (judged to be insufficient evidence for existence of nuclide).

Nuclides without Consistency values are awaiting evaluation.

Contribution to the IAEA Nuclear Data Section CRP Meeting on X-ray Standards, IAEA Vienna, 9 – 11 December 1998

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Before starting to evaluate X-ray emission data from literature, some tools needed to be prepared, e.g. the way to calculate EC probabilities, P_i, or internal conversion coefficients, α_i. Authors rather calculate than measure EC probabilities or they are taken from other sources. Other auxiliary data are fluorescence yields, ω_i, Coster-Kronig transition probabilities, f_{ij}, and n_{KL}, the average number of primary L_i sub-shell vacancies produced by transitions to the K-shell.

Except for the EC probabilities and the internal conversion coefficients, most of these auxiliary data can be found in Bambynek-1972, Bambynek-1977, Bambynek-1984 and Schönfeld-1996. It is expected (Nichols-1997, p.8), that the tools for generating internal conversion coefficients (interpolations from the tables of Rösel, ...) would be made available to the CRP.

A beginning was made by generating an algorithm for calculating EC probabilities under the form of an EXCEL template. Input are the relative occupation numbers of the shells and sub-shells, the binding energies, the amplitudes of the bound electron wave functions, the exchange and overlap correction factors, B(x), from Vatai and from Bahcall, the neutrino energy, the decay energy Q_{EC}, and in case of a non-ground state decay, the isomeric transition energy E_γ. The EC probabilities are calculated for both exchange and overlap corrections of Vatai and Bahcall, in order to have a source for the allocation of uncertainties on the P_i.

The exchange and overlap correction tables in Bambynek-1977 are incomplete, especially above Z = 54. Inter- and extrapolations are necessary for the higher shell electron capture probabilities. As these higher shell electron capture probabilities are rather small, the large uncertainties, due to the extrapolations will not affect significantly the lower shell electron capture probabilities. But they need to be calculated in order to get the correct balance between these probabilities.

In the range of $40 < Z < 50$ it can be shown, that a quadratic fit of B(K), B(L1), B(M1) values and extrapolation to B(N1) gives consistant values which are (2.46+0.13)% lower than the B(N1) from the tables. This encourages a quadratic fit-extrapolation method for the missing values of B(N1) and correcting for 2.46% "undershoot".

For the O-shell there are no values - but as O-capture is very unprobable, the exchange and overlap corrections for O-shells are set to 1.

The algorithms and the EXCEL template are ready now. The next step is to compare calculations of the P_i with evaluated experimental results.

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Bambynek-1984	W. Bambynek in X-84, X-84, Inner shell Processes..., Leipzig 1984.
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Future:

Before the evaluation starts, several questions have to be answered:

- Is it necessary to re-evaluate nuclides, that are currently being evaluated elsewhere – partly even by members of this CRP?
- What is the reason for all these parallel evaluations?
- As probably all of us have to economise our activities – wouldn't it be possible to base our efforts on existing evaluations by using existing data-sets and just add new data?
- If not – wouldn't it be useful to have a central system in collaboration with other evaluators where all relevant basic information and evaluation procedures plus computer programs are stored, in order to easily and continuously update evaluations with a minimum of effort?
- Another subject to decide on is – whether evaluations are to be based on experimental data only – and where to fill gaps with theoretical data. As theoretical and experimental data are not always coherent (atomic data, internal conversion) a decision on what to propose to the user of such data is necessary.
- Radionuclide decay data have been measured for at least 50 years now. Do we consider all these data as eligible for evaluation, or do we say that 'the value measured in 1953 is not reliable because of the use of antique equipment'? Experiences with modern electronics in data measurements suggest that older measurements might be wrong,
- Last remark: From my experience with users of radioactive decay data I know that they are practically never aware of the latest and best evaluations. When the new edition of the CRP results are available, an advertising campaign is proposed to stop users from employing the "wrong" tables.

Proposal:

I therefore propose to generate a central pool of experimental data from literature containing the remarks and the conclusions of all evaluators. Such data pools exist in some laboratories – but just as a sort of private data sources – and with a high probability of being lost after retirement of the concerned scientist. Additionally, these volatile data pools are not consistent with those of other evaluators. Furthermore, all auxiliary data sets and tables, and computer programs and used algorithms should be stored centrally and be available to CRP members.

As soon as a publication of concern to the CRP is available, it should be circulated to all evaluators including comments. Then a final assessment should be made at the IAEA ND-section, after consulting CRP members, and the data be entered into the data pool, available to all CRP members. This would enable the IAEA ND-Section to upgrade evaluations at regular instances and without big efforts. Between these regular upgrades, evaluations should be frozen for at least 3 to 5 years.

There will be changes in the composition of the CRP in the future – even if this CRP is composed for the next 3 – 4 years. With the above mentioned system, a high degree of continuity is guaranteed. A new-comer has access to an existing pool of data and procedures; he/she does not need to start from zero, and can concentrate only on new developments and data.

Final remark:

When looking at publications of radioactive decay data it is clear that few authors fulfil the requirements of CRP evaluators. It is time to publish in relevant journals, a vade-mecum for scientists doing data measurements, containing a check-list for the description of experiments, uncertainties, covariances, etc. – which assist greatly in international radioactive decay data evaluation programmes.

How can the radionuclide data evaluation work carried out in the Physikalisch-Technische Bundesanstalt (PTB) during the last years be coordinated with the present CRP?

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I have been engaged in the field of absolute activity measurements of radioactive sources since 1964. A growing part of my work over recent years has been the evaluation of radionuclide data. In this respect two collaborations have to be mentioned:

Since 1993 there has been a collaboration between the LPRI (Laboratoire Primaire des Rayonnements Ionisants at Saclay / France) and the PTB. Both institutions agreed to jointly issue a "Table of radionuclides" to continue the "Table de Radionucleides" formerly issued by the LPRI alone [1]. This includes the reevaluation of formerly evaluated data as well as the addition of new radionuclides to the list of approximately 200 radionuclides which are of metrological interest and which are often applied in different fields. In the tables evaluated values of half-lives, transition probabilities and transition energies of β^-, β^+, EC and gamma transitions, internal conversion coefficients of gamma transitions, emission probabilities of X-ray and gamma quanta, Auger and conversion electrons and annihilation radiation and nuclear shell data are compiled.

An "Introduction" has been prepared [2] which includes the agreed evaluation rules, the collection and evaluation of atomic shell data have been carried out, and a computer version of the table was developed (at LPRI). Data for the following radionuclides have been evaluated in the PTB since 1996: Ge-68/Ga-68, Sr-85, Y-88, Cd-109, I-125, Ce-141, Yb-169, Re-186, Au-198, Tl-201.

Since 1996 the PTB has also contributed to the International Decay Data Evaluation Project (IDDEP) co-ordinated by R. G. Helmer, Idaho Falls USA [3]. This project is closely related to the ENSDF (Evaluated Nuclear Structure data File). Within the framework of this project it is planned to evaluate very carefully the nuclear decay data of about 250 radionuclides. The PTB has contributed to the evaluation of decay data of the following radionuclides: Be-7, Na-22, Na-24, Cr-51, Mn-54, Se-75, Cs-137/Ba-137m, Ce-139.

The work of these two projects is closely connected in order to avoid duplication of work. I urge the avoidance of duplication in the present research project. It makes no sense to recommend at the same time different values for the same quantity. This means: what already has been done within the framework of the above-mentioned projects should be absorbed into the present research project.

Concerning the present research project, I can imagine two ways of coming to the desired data. One way is characterised by some sort of specialisation. This means data for half-lives are evaluated by a specialist for half-lives, gamma-ray energies are evaluated by a specialist

for gamma ray energies, emission probabilities of gamma-rays are evaluated by a specialist for gamma ray emission, emission probabilities of X-rays are evaluated by a specialist for X-ray emission and so on. This system has partially been followed in the IAEA-TECDOC-619 (1991). As few scientists are experienced in all of these fields, I see some advantage in this approach. The other alternative would be for one person to be formally responsible for all the data of one radionuclide. Of course, this does not exclude, that he contacts other specialists in order to improve his data. We might discuss here these two versions. Perhaps a compromise between both types is the best way to produce the "best figures".

At the end of my contribution I would like to say some words concerning atomic shell data – because this is my special field – and programs developed at the PTB for the calculation of emission probabilities of K- and L-X rays and of K-Auger electrons. When evaluating atomic shell data like fluorescence yields and relative X-ray emission probabilities, one can adopt one of two methods. The one way is to collect data for the individual case and to evaluate a best value from these figures (which are very rare for some elements). The other way is to collect the data for all elements and to make a least squares fit, which is possible because there is a monotonically dependence of these data from the atomic number Z. At the PTB we have adopted the last approach. The results of these efforts are compiled in a number of publications [4 – 7].

A computer program has been developed at PTB [8] which enables the emission probabilities of K-X rays and K-Auger electrons to be calculated on the basis of implemented data files (K-shell fluorescence yields, relative K-X ray emission probabilities) and nuclide-specific data (such as transition probabilities, fractional capture probabilities and conversion coefficients). As these values are based uniformly on evaluated data, they can be regarded to be the most probable values which should be recommended for general use in X-ray spectrometry. On the other hand one can compare these calculated values with measured values (which often do not exist in the desired quality). From the agreement (or disagreement) between calculated and measured values one can draw conclusions concerning the internal consistency of all the data which are used as input for the calculated data.

The development of a program is planned at the PTB which allows one to calculate the components of the total X-ray emission probability. For this purpose we need three L-subshell fluorescence yields instead of one mean L-shell fluorescence yield, three fractional L-subshell capture probabilities (instead of one), the three components of the L-conversion coefficient and the vacancy transfer coefficients to the three L-subshells instead of one integral figure n_{KL}. Moreover we need the relative L-X ray emission probabilities for those radiation which are emitted when there are vacancies in the three L subshells (Campbell and Wang). In principle all the data are available in the literature, although the accuracy does not often correspond to the desired level. On the other hand, measured individual L-X ray emission probabilities have uncertainties of several percents. Taking this in mind, calculated values may be of interest in many cases, even if their uncertainties are rather large.

The PTB is prepared to contribute their data and programs to the present co-ordinated research project. As already mentioned, duplicate work should be avoided in order to work efficiently.

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Decay data related activities at NPL

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1. Introduction

Within the Radioactivity Group at NPL, the majority of the work is funded by the UK Department of Trade and Industry in support of the requirements of the UK National Measurement System. The ultimate customer is the UK base as a whole and encompasses interests in the medical, nuclear environmental and other relevant sectors. The funding mechanism is based on three year programmes which are endorsed by a cross-industry advisory committee which assesses the relative importance of programme proposals and prioritizes the available funding accordingly. Any work undertaken therefore in the nuclear decay data area is that required to support particular shortcomings that have been identified at the programme formulation stage as well as that required to underpin other related activities, such as radionuclide standardisations.

2. Work completed since last CRP

In many cases, the absolute values for gamma emission probabilities rely on the quality and availability of absolute standardisations of particular radionuclides. Since the first CRP, a number of radionuclides have been re-standardized or standardized for the first time. The results of these standardisations are being fed into the BIPM-based international database on equivalence and will allow better, and hopefully lower, estimates to be made of the uncertainties attached to particular decay scheme measurements. In the half-life area, better and new standardisations of very long-lived radionuclides will allow stochiometric measurements to be achieved.

Since the beginning of the decade, as well as re-standardisations, new standards have been produced for:

^{32}P, ^{89}Sr, ^{90}Y, ^{106}Ru, ^{153}Sm, ^{186}Re, ^{188}Re, ^{192}Ir, ^{210}Bi, ^{210}Pb, ^{233}Pa, ^{237}Np, ^{238}Pu, ^{239}Np, ^{239}Pu, ^{243}Am, ^{244}Cm.

New decay data measurements have been published for:

^{56}Co, ^{90}Sr, ^{125}Sb, ^{153}Sm, ^{154}Eu, ^{192}Ir, ^{239}Np, ^{243}Am.

In 1995, NPL published a report RSA(EXT)53 which recommended, for the UK user community, additional decay data for 53 radionuclides, supplementing TECDOC-619. These data were extracted from currently available evaluated databases, using JEF2.2 and UKPADD2 where possible and occasionally ENSDF.

3. Ongoing and Future Work

From the 1995-1998 programme, work is now being completed on a number of radionuclides and results of standardisations and measurements of nuclear decay data are to be published in the near future. This will include the radionuclides:

^{210}Bi, ^{210}Pb, ^{210}Po, ^{233}Pa, ^{237}Np, ^{238}Pu, ^{239}Pu, ^{244}Cm.

Investigations are also planned in the 1998-2001 programme and include the radionuclides:

^{95m}Tc, ^{99}Tc, ^{129}I, ^{209}Po, ^{237}Np.

4. Dissemination of Recommended Decay Data

In respect of recommendations to the user community on preferred nuclear decay data, NPL recommends that the IAEA TECDOC-619 data takes precedence. NPL regularly conducts intercomparison exercises covering all the relevant sectors (medical, nuclear, environmental). In these, participants are recommended to use the data in the IAEA TECDOC-619 and NPL Report RSA(EXT)53. In the reporting sheets for these exercises, participants are also asked to quote the data source that they use and this subject is always on the agenda at the follow-up workshops. Over the past decade, this approach has produced a dramatic improvement in the quality of the data used by the user community as well as its awareness of up-to-date data sources. It has also resulted in pressure from the users on to manufacturers and there is an increased willingness for suppliers of software to either update their nuclear decay data libraries or to allow users to access and modify the libraries themselves.

Compilation and evaluation of high-energy γ-ray standards from nuclear reactions.

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Abstract

The strongest deexcitation line in $^{12}C^*$ at 4.438 MeV results from the decay of its first excited state at 4.439 MeV. Excited states above the 4.439 MeV level decay primarily by breakup into α-particles, and hence they are not important sources of γ-ray lines. An exception is the 15.11 MeV level, which, because of conservation of isotopic spin, cannot decay by emitting α-particles and hence deexcites only by γ-ray emission. The cross sections for exciting this level were compiled and it was found that the intensity of the 15.11 MeV line is at most about 2% of the 4.438 MeV line intensity. At proton energies lower than 23 MeV the overall contribution of the higher excitet states to the 4.44 MeV γ-rays is less than 0.5 mb.

The various excitation modes of the 4.439 MeV and the 15.11 MeV levels involving proton and α-particle projectiles are following:

- $^{11}B(p,\gamma)^{12}C^*$,
- $^{12}C(p,p')^{12}C^*$,
- $^{12}C(\alpha,\alpha')^{12}C^*$,
- $^{14}N(p,2pn)^{12}C^*$,
- $^{14}N(\alpha,\alpha pn)^{12}C^*$,
- $^{16}O(p,\alpha p)^{12}C^*$,
- $^{16}O(\alpha,2\alpha)^{12}C^*$,

while the reactions,

- $^{12}C(p,2p)^{11}B^*$,
- $^{12}C(\alpha,\alpha p)^{11}B^*$,

excite the 4.444 MeV level in ^{11}B, which has an energy threshold of 22 MeV. The resulting γ-line of energy 4.443 MeV cannot be resolved from the 4.439 MeV one because the kinematical Doppler broadning blends the two into a single feature.

The $^{11}B(p,\gamma^{12}C^*$ reaction

Proton capture by ^{11}B at incident resonance energy 163 keV populates the level at excitation energy of 16.1058 MeV in ^{12}C, which decays to the first excitet level at 4.439 MeV producing

the cascading γ-rays of energies 4.44 MeV and 11.7 MeV with intensity of 92 quanta per 100 decays. The other resonances in the proton capture yield curve are of less practical importance,

proton energy	γ-ray energies	cross section	width
163 keV	16.11, 11.68, 4.43	0.157 mb	7 keV
675 keV	12.15, 4.43	0.050 mb	322 keV
1388 keV	17.23, 12.80	0.053 mb	1270 keV
2630 keV	13.94, 4.43, 2.14		300 keV.

However, in applications of the proton capture reaction as a source of calibration γ-rays common use is made of the primary high-energy γ-transitions from the capturing states (within the energy spread of the proton projectiles) to the ground state and the subsequent excited states of ^{12}C. The available data on the primary γ-rays following proton capture with energies from 4 MeV to 90 MeV are gathered in [Mar98].

The ^{12}C(p,pγ)^{12}C* reaction and comparison with the ^{12}C(p,p')^{12}C* reaction

Due to the exceptionally weak γ-branchings of excited states in ^{12}C other than the 4.439 and 15.11 MeV states, the cross sections for inelastic scattering determined via detection of either the γ-rays or by detection of the scattered inelastic groups of nucleons do not differ within typical experimental errors. The practical equality of these cross sections was verified experimentally, e.g. the inelastic cross sections to the 4.44 MeV level at 12 MeV incident energy determined by integrating the angular distribution of scattered protons was found to be 267±14 mb, in good agreement with the cross section for γ-ray production, which had been found to be 262±26 mb.

The differential cross sections for excitation of the 4.439 MeV state in the (p,p') reaction on ^{12}C were measured in the proton energy range from 6.65 to 65 MeV and in the (n,n') reaction at neutron energies 22.0, 24.0, 26.0 and 28.2 MeV. Cross sections for excitation of the 15.11 MeV state were measured via proton scattering of incident energies from 20.5 to 45.0 MeV. These inelastic cross sections have no immediate application in calibration procedures. However, by fitting with Legendre polynomial series they enable extraction of the integral inelastic cross sections that characterize the strength of the reaction as a γ-ray source. In conjunction with the angular distributions of the emitted γ-rays they can be already immediately used for intensity calibration of a γ-ray detector.

The 4.44 MeV γ-rays

The threshold for excitation of the 4.439 MeV level in ^{12}C is 4.81 MeV and 5.92 MeV for incident protons and α-particles, respectively. The cross sections for production of the 4.44 MeV γ-rays in the (p,p'), (p,p') + (p,2p), (α,α') and (α,α') + (α,αp) reactions were evaluated by [Ram79]. The proton data cover the incident energy range from threshold to about 1000 MeV/nucleon and the α-particle data cover the energy range from threshold to about 40 MeV/nucleon. In the light of what was said above the difference between the γ-ray production data and the (p,p') data is entirely due to the excitation of the 4.444 MeV level in ^{11}B by the (p,p2p) reaction relevant above the 22 MeV threshold. The γ-ray production via α-particle scattering includes the unseparable contribution from the (α,αp) reaction too. The information on the angular distributions of emitted γ-rays is available but sparce. These data are not self-consistent since some of them are not symmetric about 90°. On the other hand, we know that the γ-ray angular distributions when averaged over the emission angles of the inelastic

protons must be symmetric about 90°. Only the angular distributions measured by [Dye81] fulfil this requirement.

Cross sections for proton induced spallation of ^{16}O are spread over the energy range from 20 MeV/nucleon to 1000 MeV/nucleon. For proton spallation of ^{14}N the cross section measured at 120 MeV was used to estimate that the 4.438 MeV γ-ray production cross section from ^{14}N is larger than from ^{16}O by about a factor 2.4.

The 15.11 MeV γ-rays

The 90° cross sections for production of the 15.11 MeV γ-rays by bombarding ^{12}C with protons were measured from the threshold energy at 16.37 MeV to 24.4 MeV. The uncertainty in absolute normalization is ±20%. These results duplicate quite well the results of earlier measurements. The earlier data cover the incident proton energy range from threshold to 48.5 MeV. The same cross sections were remeasured from threshold to 24 MeV by [Mea73]. All these data sets are consistent in magnitude but differ in the energy resolution of the proton projectiles and the projectile energy grid. The Legendre polynomial expansion coefficients for the angular distributions of the 15.11 MeV γ-rays are also available. Only even Legendre polynomials contribute resulting in distributions symmetric with respect to 90°. The latter data have been extended in framework of the present evaluation by fitting with Legendre polynomials of the angular distributions of scattered protons measured by [Ger75] at the incident proton energies 22.5, 30, 32, 34, 36, 38, 40, 42.5 and 45 MeV.

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Coincidence method for semiconductor detector calibration

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The coincidence method has been used successfully for decades in nuclear spectroscopy and various applications. It is considered to be the only feasible method to study complex decay and level schemes of atomic nuclei. Furthermore, a widely accepted application of the coincidence method is the determination of the absolute activity of standards for detector calibration. However, the coincidence method can be applied more generally and allows the determination of the absolute detector efficiency .

At present the absolute calibration of photon detectors proceeds in two steps. In the first step determination of the absolute source intensity is performed using $\beta - \gamma$ coincidence method. This results in an absolutely calibrated standard, which is used in the second step to determine the absolute efficiency of the photon detector.

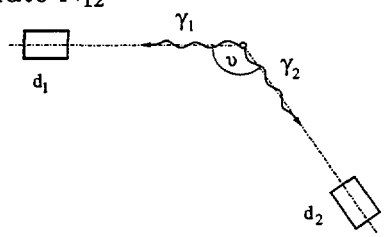
Use of the coincidence method can potentially reduce the number of steps in detector calibration procedure to a single step, thus reducing the error of the calibration. This approach may be especially useful for several high energy photon sources (e.g. ^{11}B + p $\rightarrow^{12} C^*$), which are difficult to calibrate absolutely.

The coincidence method is rather simple and can be used if the source nucleus decays by two cascading photons γ_1 and γ_2. The calibration setup consist of two detectors and is shown schematically below. Detectors d_1 and d_2 are used for measurement of gamma ray γ_1 and γ_2, respectively. The following relations hold for detection rates N_1 in detector d_1, N_2 in detector d_2 and the coincidence rate N_{12}

$$N_1 = \Omega_1 A$$

$$N_2 = \Omega_2 A$$

$$N_{12} = \Omega_1 \Omega_2 A W(\theta),$$



where A is the unknown decay rate of the calibration source, Ω_i is the efficiency of detector i for gamma ray γ_i and $W(\theta)$ is the angular correlation function, i.e. angular distribution of γ_2 with respect to the gamma ray γ_1 detected in d_1. The angle θ is defined by the position of both detectors. These three equations can be solved for three unknown variables Ω_1, Ω_2 and A

$$\Omega_1 = \frac{N_{12}}{N_2} \frac{1}{W(\theta)}$$

$$\Omega_2 = \frac{N_{12}}{N_1}\frac{1}{W(\theta)}$$

$$A = \frac{N_1 N_2}{N_{12}} W(\theta).$$

From these relations the efficiency of detector d_1 for gamma ray γ_1 can be determined form the ratio of coincidence to single counts of gamma ray γ_2 in detector d_2. The same holds for gamma ray γ_2 detected in d_1 and γ_1 detected in d_2. Thus we do not need to know efficiency of detector d_2 to determine the efficiency of detector d_1.

These simple relations can be further refined to describe the realistic experimental situation with respect to the full energy peak efficiency of the calibrated detector, calibration sources with more complicated decay schemes and finite detector volumes. A two-parametric data acquisition system is necessary in order to perform precise efficiency measurement. Further reduction in measurement errors can be achieved by measuring the time distribution between coincident γ rays.

For precise detector calibration using coincidence method slightly different needs in decay data of calibration sources arise. A crucial parameter is the number of gamma rays γ_2 per single photon γ_1, thus population of individual levels, branching ratios of electromagnetic transitions as well as the internal conversion coefficients need to be known. However, there is no need to know absolutely the number of nuclei that decay during the efficiency measurement; thus the half-life of the source is irrelevant and does not need to be known precisely.

On the other hand, spatial and time correlations of both photons are important. However, since the half-lives of the majority of nuclear levels in nuclei used for calibration are much shorter than the time resolution of commonly used photon detectors, we can assume that both photons are emitted at the same time.

The spatial correlations between two successive γ transitions are at present well understood. Correlation depends on the spins and parities of all three nuclear levels involved as well as on the multi-polarities of both γ transitions. General theory of correlations is rather complex, but in principle the correlation function can always be reduced to the sum of Legendre polynomials

$$W(\theta) = \sum_{k_{even}} A_{kk} P_k(\cos\theta).$$

Since the spins of the nuclear levels of most nuclei used for detector calibration are rather low, directional correlations are weak and the first two or three members of the above sum describe satisfactorily the observed correlations. The influence of the correlations can be further reduced by measurement at an angle of 125^o, where the second Legendre polynomial equals to zero. Nevertheless, the influence of angular correlations should be taken into account in the coincidence method.

The least squares procedure in energy gamma-ray spectroscopy

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Covariances are important if the whole statistical properties of experimental data are to be preserved and statistical tests are to be reliable.

If the necessary details of experimental procedure are given, it is possible to recover information from published data to derive covariances. However, in some cases, it can be impossible to recover the experimental details needed to determine the covariances. In these cases, a detector can be calibrated with sources whose gamma-ray energies covariance matrix is known then interpolating the energies so that the required secondary standards can be calculated, along with the corresponding covariance matrix.

We discuss below how (a) to calibrate a detector system and (b) how to interpolate data taking into account variances and covariances of the input data, the adjusted parameters, and the interpolated data.

a) Calibration

Consider the data set (E_i,C_i), i=1, 2, ...n, where the energy data E_i have a covariance matrix $\mathbf{V_0}$ and the independent data C_i were supposed statistically independent, $(\mathrm{cov}(C_i,C_j)=0$ for $i\neq j)$ with variances $(\mathbf{V_c})_{ii}=\sigma_{ci}^2.\delta_{ij}$. If a function $E=a_1+a_2C+a_3C^2$ is to be fitted to the data, the (column) vector of the adjusted parameters $\tilde{\mathbf{A}}$ is given by

$$\tilde{\mathbf{A}} = (\mathbf{X}^t\mathbf{V}^{-1}\mathbf{X})^{-1}\mathbf{X}^t\mathbf{V}^{-1}\mathbf{E} \quad , \quad (1)$$

where $\mathbf{E}$ is the (column) vector with elements E_i, and

$$\mathbf{X} = \begin{bmatrix} 1 & C_1 & C_1^2 \\ 1 & C_2 & C_2^2 \\ \vdots & & \vdots \\ 1 & C_n & C_n^2 \end{bmatrix} \quad (2)$$

is the design matrix. The covariance matrix of $\tilde{\mathbf{A}}$ is

$$V_{\tilde{A}} = (\mathbf{X}^t\mathbf{V}^{-1}\mathbf{X})^{-1} \quad . \quad (3)$$

The covariance matrix $\mathbf{V}$ is given approximately by

$$\mathbf{V} = \mathbf{V_0} + \tilde{a}_2^2\mathbf{V_C} \quad . \quad (4)$$

It is assumed that the relation $E(C)=a_1+a_2C+a_3C^2$ is approximately linear, $a_3C<<a_2$. Since Eq. (4) needs the value $\tilde{a}_2$, and to calculate $\tilde{a}_2$ we need $\mathbf{V}$, the fit procedure is iterative.

If the data obey Gaussian distribution, then

$$\chi^2 = (\mathbf{E} - \mathbf{X}\tilde{\mathbf{A}})^t \mathbf{V}^{-1} (\mathbf{E} - \mathbf{X}\tilde{\mathbf{A}}) \quad (5)$$

obeys a chi-square distribution and can be used as a quality-of-fit test.

b) Interpolation

Energies corresponding to measured values C'_i calculated from the adjusted calibration function, are given by

$$\mathbf{E}' = \mathbf{X}'\tilde{\mathbf{A}} \qquad (6)$$

with covariance matrix

$$\mathbf{V}' = \mathbf{X}'\mathbf{V}_{\tilde{\mathbf{A}}}\mathbf{X}'^t + \tilde{\mathbf{a}}_2^2 \mathbf{V}_{C'} \quad . (7)$$

In this last equation $\mathbf{V}_{C'}$ is the covariance matrix of the data C' and the matrix X' is given by an equation analogous to eq. (2).

With this canonical procedure variances and covariances can be taken into account from the beginning without introducing different procedures for each specific particular energy value being sought.

Ref [1] shows some additional details that must be used in the calibration procedure for gamma-ray spectroscopy in order to keep, in all steps, the entire covariance matrices.

[1] V. R. Vanin, G. Kenchian, M. Moralles, O. Helene and P. R. Pascholati, Nucl. Instr. and Meth. A391(1997)338.

Determining covariances between gamma-ray emission probabilities in very simple decay schemes.

Vito R. Vanin and Otaviano A.M. Helene

Assume that the decay scheme represented in figure 1 applies to nucleus M decaying to S. Representing the gamma-ray emission probabilities by P_i and the internal conversion coefficients by α_i, it is obtained :

$P_i = \frac{1}{1+\alpha_i}$ for i=1,2 , with standard deviations $\sigma_{P_i} = P_i \frac{\sigma_{\alpha_i}}{1+\alpha_i}$ and covariance

$$\mathrm{cov}(P_1,P_2) = P_1 P_2 \frac{\mathrm{cov}(\alpha_1,\alpha_2)}{(1+\alpha_1)(1+\alpha_2)}.$$

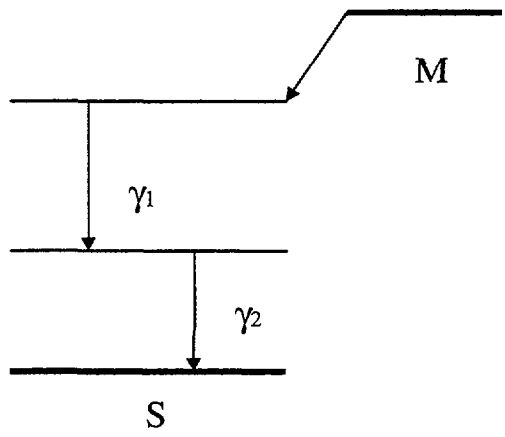

Figure 1. Decay scheme with two γ-rays and a single beta branch.

If the parent nucleus also feeds the ground state of the daughter nucleus as shown in figure 2, there is an additional contribution to the covariance. Representing the probability of feeding the ground state by ε_g: $P_i = \frac{1-\varepsilon_g}{1+\alpha_i}$. Assuming ε_g statistically independent of the α_i, then $\mathrm{cov}(\varepsilon_g,\alpha_i) = 0$ and

$$\sigma_{P_i} = P_i \sqrt{\left(\frac{\sigma_{\alpha_i}}{1+\alpha_i}\right)^2 + \left(\frac{\sigma_{\varepsilon_g}}{1-\varepsilon_g}\right)^2},$$ and

$$\mathrm{cov}(P_1,P_2) = P_1 P_2 \left(\frac{\mathrm{cov}(\alpha_1,\alpha_2)}{(1+\alpha_1)(1+\alpha_2)} + \left(\frac{\sigma_{\varepsilon_g}}{1-\varepsilon_g}\right)^2 \right)$$

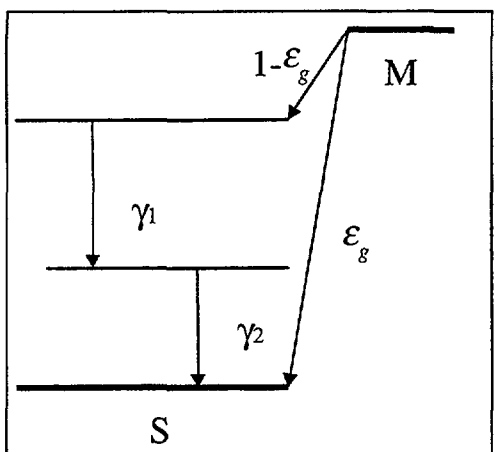

Figure 2. Decay scheme with 2 γ-rays and two beta branches.

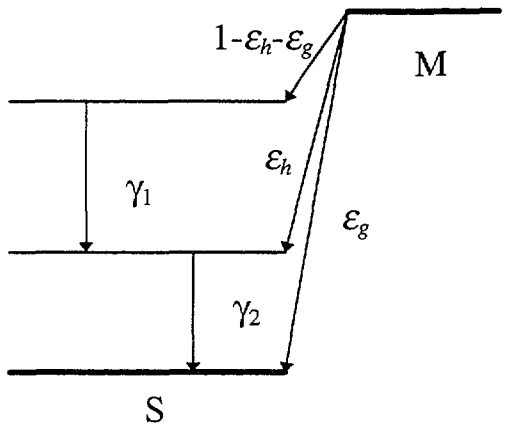

Figure 3. Decay scheme with 2 γ-rays and 3 beta branches.

The formula increase in complexity when there is an additional beta branch to the intermediate level, as implied by the scheme shown in figure 3. The emission probabilities are given by $P_1 = \frac{1-\varepsilon_h-\varepsilon_g}{1+\alpha_1}$ and $P_2 = \frac{1-\varepsilon_g}{1+\alpha_2}$ with standard deviations

$$\sigma_{P_2} = P_2\sqrt{\left(\frac{\sigma_{\alpha_2}}{1+\alpha_2}\right)^2+\left(\frac{\sigma_{\varepsilon_g}}{1-\varepsilon_g}\right)^2}$$ and

$$\sigma_{P_1} = P_1\sqrt{\left(\frac{\sigma_{\alpha_1}}{1+\alpha_1}\right)^2+\frac{\sigma_{\varepsilon_h}^2+\sigma_{\varepsilon_g}^2+2\,\mathrm{cov}\left(\varepsilon_h,\varepsilon_g\right)}{\left(1-\varepsilon_h-\varepsilon_g\right)^2}}$$

and covariance $\mathrm{cov}(P_1,P_2) = P_1P_2\left(\frac{\mathrm{cov}(\alpha_1,\alpha_2)}{(1+\alpha_1)(1+\alpha_2)}+\frac{\sigma_{\varepsilon_g}^2+\mathrm{cov}\left(\varepsilon_h,\varepsilon_g\right)}{\left(1-\varepsilon_g\right)\left(1-\varepsilon_h-\varepsilon_g\right)}\right)$,

assuming that ε_g and ε_h are statistically independent of the α_i so $\mathrm{cov}\left(\varepsilon_g,\alpha_i\right)=0$ and $\mathrm{cov}\left(\varepsilon_h,\alpha_i\right)=0$.

When $\mathrm{cov}(\alpha_1,\alpha_2)=\mathrm{cov}\left(\varepsilon_h,\varepsilon_g\right)=0$, $\varepsilon_h \ll 1$, and $\varepsilon_g \ll 1$, the covariance between the gamma-ray emission probabilities as described by figures 2 and 3 reduces to $\mathrm{cov}(P_1,P_2)\cong P_1P_2\sigma_{\varepsilon_g}^2$. In the same limit conditions, the covariance is null in the case shown in figure 1.

Determining decay data correlations and calibrating the detector efficiency with multigamma-ray sources.

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The aim of this work is to determine the steps needed to define the correlations between decay data. This problem is linked to the inverse problem, that is, the detector efficiency calibration with multigamma-ray sources taking into account the correlations between the decay data. The calibration procedure must give sound results even in unfavorable geometry, for instance, when the source is placed near the detector. We describe first the calibration method, followed by the procedure to determine the decay data and variance matrix.

The number of counts in the full-energy peak observed with HPGe detectors depends on a number of quantities related to the decay scheme and also on many secondary detection effects. The formula are well described in the literature [1-5].

The idea is to fit the efficiency calibration curve or the decay scheme quantities directly to the peak-area data in the most straightforward calculation, avoiding corrections that would introduce uncertainties and correlations that are difficult or tedious to evaluate.

In almost any geometry, the summing of coincident gamma-rays must be corrected, which leads to choosing the parent's feeding ratios and daughter's branching-ratios as preferred decay data. Also a calibration of the total-to-peak detection ratio is required. The use of analytical calibration functions makes the calculations easier.

The least-squares method (LSM) gives unbiased estimates of the parameters of the calibration function and, under the assumption of linearity near the estimated values of the parameters, also gives *the* estimates of minimum variance. In complex calculations like this one, it is better to formulate the LSM with matrices. Both the LSM properties and the appropriate formalism are well described in the literature [6,7].

The calibration function can be fitted by the LSM to the experimental data. Besides the counting statistics, there are contributions to the data variances from the uncertainties in the activity, the quantities related to the decay scheme, and the total-to-peak detection ratio calibration. The fitting procedure is iterative, since the equations are not linear in the efficiency calibration parameters due to the correction of sum effects, quadratically dependent on the efficiency.

Therefore, from the point of view of the calculations presented here, we find that it is easier and more useful to standardize the feeding factors and the branching ratios than the gamma-ray emission probabilities.

The same formula used to model the peak areas in the calibration experiment can be used to determine the decay data with a previously calibrated detector, by using the appropriate design matrix. The variance matrix has four components: counting statistics, efficiency calibration, total-to-peak gamma-ray detection ratio calibration, and other decay data quantities such as conversion coefficients. Also in this case the fitting procedure is iterative because the formula are generally not linear.

Pile-up and sum with bremsstrahlung, both internal and external, must be always considered, at least to check whether they are negligible effects. Also the assumption that the directional correlation between gamma-rays is negligible when correcting sum effects may be inadequate. Some standard bremsstrahlung and directional correlation data could be provided in order to stimulate the experimentalists to use the required corrections routinely.

Since the correlations between the scheme-dependent factors are usually not known, secondary standards with known correlations should be developed against detectors calibrated with monochromatic sources or sources with simple decay schemes that enable precise corrections, like ^{60}Co, which should be kept as robust primary standards. Also, monochromatic sources are always needed to calibrate the total-to-peak ratio.

For testing the procedure, the aim was to calibrate a 30% HPGe detector using sources of ^{88}Y (5000 Bq) , ^{60}Co (2000 Bq) and ^{51}Cr(2000 Bq), and then determine the ^{207}Bi decay data; however, we did not succeed in preparing the ^{51}Cr source in time for presentation at the meeting. The ^{88}Y and ^{60}Co sources were calibrated in a 4π β-γ detector, and we found that the activity values are uncorrelated. The accuracy of the geometrical dimensions of frames and encapsulation is 0.1 mm, which is enough to assure a 0.1% relative precision in the efficiency for the source-detector distance adopted, 20 cm. We substituted a ^{137}Cs (25000 Bq) Amersham source for the ^{51}Cr source and compensated the difference in encapsulation.

After the efficiency and total-to-peak ratio calibrations, a ^{207}Bi Amersham source was monitored and the decay data were determined. In this case, we obtained very small correlation coefficients, due to the characteristics of the ^{207}Bi decay scheme: each quantity derives mainly from one of the lines, and the efficiencies at the gamma-ray energies are almost uncorrelated because they are well separated in energy. Two quantities would be more correlated if they depended mainly on the same line, or if they were determined from different combinations (with significant weights) of the same set of lines.

From the set of decay data fitted, all the gamma-ray transition probabilities and their variance matrices can be determined by appropriate equations.

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Determination of covariances between gamma-ray energies from ^{161}Tb, ^{172}Hf, ^{241}Am and ^{198}Au decays

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Gamma-ray energies from ^{161}Tb, ^{172}Hf, ^{241}Am and ^{198}Au were estimated by using published data, taking into account covariances between the data. Data from ref. [1] and [2] were used for the input.

The experimental ratio between the 84 keV gamma-ray energy from ^{170}Tm decay and the 49 keV from ^{161}Tb decay as well as ratios between the 26 and 75 keV gamma-ray transitions and the 49 keV transition from ^{161}Tb decay were taken from ref. [1]. The ratio between the 84 keV and the standard 411 keV from ^{198}Au was taken from ref. [2]. From these data a set of experimental values were determined for the 26, 49 and 75 keV gamma-rays from ^{161}Tb decay and the 411 keV transition from ^{198}Au. The data obtained are y(1)=25.65135(4) keV, y(2)=48.91533(7) keV, y(3)=74.56668(8) keV and y(9)=411.80205(17) keV. The covariance matrix of these four values was determined from the general covariance propagation formula.

The differences between gamma-ray energies were taken from Table 4 in ref. [2]:

$$\begin{aligned} y(4) &= E1(Tb) - E1(Am) = -0.6933(2)\ \text{keV} \\ y(5) &= E1(Tb) - E(Hf) = 1.7184(2)\ \text{keV} \\ y(6) &= E1(Hf) - E1(Am) = -2.4118(9)\ \text{keV} \\ y(7) &= E2(Tb) - E2(Am) = -10.6256(1)\ \text{keV} \\ y(8) &= E3(Tb) - E2(Am) = 15.0260(2)\ \text{keV} \end{aligned} \qquad (1)$$

The covariance matrix of these energy differences was supposed to be diagonal.

The parameters to be fitted are the gamma-ray energies a_1=E1(Tb), a_2=E2(Tb), a_3=E3(Tb), a_4=E(Hf), a_5=E1(Am), a_6=E2(Am) and a_7=E(^{198}Au). The fitted parameters are given by $\tilde{\mathbf{A}}=(\mathbf{X}^t\mathbf{V}^{-1}\mathbf{X})^{-1}\mathbf{X}^t\mathbf{V}^{-1}\mathbf{Y}$ and their variance matrix is given by $\mathbf{V}_{\tilde{a}}=(\mathbf{X}^t\mathbf{V}^{-1}\mathbf{X})^{-1}$, where $\mathbf{X}$ is the design matrix relating the parameters to the experimental data.

The results are a_1=25.65135 (4) keV, a_2=48.91534 (7) keV, a_3=74.56669 (8) keV, a_4=23.93294 (20) keV, a_6=26.34465 (20) keV, a_6=59.54089 (11) keV and a_7=411.80206 (17) keV, with the correlation matrix given by

$$\mathbf{C}_{\tilde{a}} = \begin{bmatrix} 1.00 & 0.94 & 0.78 & 0.18 & 0.18 & 0.56 & 0.29 \\ 0.94 & 1.00 & 0.83 & 0.17 & 0.17 & 0.59 & 0.31 \\ 0.78 & 0.83 & 1.00 & 0.14 & 0.14 & 0.54 & 0.37 \\ 0.18 & 0.17 & 0.14 & 1.00 & 0.08 & 0.10 & 0.05 \\ 0.18 & 0.17 & 0.14 & 0.08 & 1.00 & 0.10 & 0.05 \\ 0.56 & 0.59 & 0.54 & 0.10 & 0.10 & 1.00 & 0.20 \\ 0.29 & 0.31 & 0.37 & 0.05 & 0.05 & 0.20 & 1.00 \end{bmatrix} \qquad (6)$$

The chi-square value obtained was 1.31 with 2 degrees of freedom.

Table 1 compares these results with the results from ref. [2].

Table 1

This note		Ref 2	
Value (keV)	std. dev. (keV)	Value (keV)	std. dev. (keV)
25.65135	0.00004	25.65135	0.00003
48.91534	0.00007	48.91533	0.00005
74.56669	0.00008	74.56669	0.00006
23.93294	0.00020	23.9330	0.0002
26.34465	0.00020	26.3446	0.0002
59.54089	0.00011	59.5409	0.0002
411.80206	0.00017	411.80205	0.00017

As can be seen from Table 1, differences between the energy-values determined in this note and in Ref. [2] are not very large. However, some aspects must be considered. First, the relevant quantity in statistical tests is the variance (and covariances) which, in some cases, is different by a factor 2. Consequently, the qualitative conclusion of a statistical test can be changed significantly due to changes in the standard deviations. The second point to be noted is the change of non-measured quantities, such as the 411 keV energy in this example. Estimates of the uncertainties of quantities which are correlated with other quantities must be revised every time those quantities are reevaluated, and this can be accomplished only if the entire covariance matrix is known [3].

If the necessary details of the experimental procedure are given, it is possible to recover information from published data in order to obtain the covariances. In some cases, however, it can be impossible to recover the experimental information needed to determine the covariances. In these cases, a detector can be calibrated with sources whose gamma-ray energies variance matrix is known and then the energies and variances for the required secondary standards can be calculated by an interpolation procedure.

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