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I N D C INTERNATIONAL NUCLEAR DATA COMMITTEE

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

Summary Report of the Second Research Co-ordination Meeting

PTB, Braunschweig, Germany

10 - 12 May 2000

Prepared by

M. Herman and A.L. Nichols*

IAEA Nuclear Data Section
Vienna, Austria

*AEA Technology
Harwell, United Kingdom

September 2000

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Abstract

The Second Research Co-ordination Meeting to Update X- and γ-ray Decay Data Standards for Detector Calibration was held at PTB Braunschweig from 10 to 12 May 2000. A primary aim of this meeting was to review progress in the evaluation and recommendation of data under the auspices of the CRP. All CRP activities were reviewed, and actions agreed for the remaining 18 months of the programme.

September 2000

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Summary Report of the
Second Research Co-ordination Meeting on

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

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10 - 12 May 2000

SUMMARY

CRP members reviewed the status of their evaluations, as agreed originally at the previous meeting. Specific procedures were debated, and individual decay-data evaluations were discussed in some detail if difficulties had been experienced. Assessments were also presented on the coincidence method of detector calibration, high-energy gamma-ray emissions from suitable nuclear reactions, and covariance analyses of gamma energies and intensities; recommendations are in the process of being formulated in these particular areas for calibration purposes. The various presentations demonstrated good progress in all areas of the CRP, with an aim of completion by the end of 2001, and publication of an IAEA-TECDOC report in 2002.

BACKGROUND

An IAEA Consultants' Meeting in May 1998[1)] recommended the establishment of a Co-ordinated Research Programme to Update X- and Gamma-ray Decay Data for Detector Calibration (i.e., re-evaluate and update the recommended decay data in IAEA-TECDOC-619[2)]). Hence, an IAEA programme began in December 1998, with the first Co-ordinated Research Meeting of specialists to undertake this work to an appropriately high gamma-ray energy[3)]. The CRM at PTB Braunschweig represents the second of these meetings to monitor the progress in achieving the main objectives of the agreed programme.

A L Nichols was elected as Chair for the Braunschweig meeting, supported by M. Herman as Secretary (see Appendices 1 and 2 for agenda and attendees, respectively). The main application of X- and gamma-ray standards decay data is the calibration of detectors for the measurement of these emissions to high energies (~90 MeV). Other requirements were also considered, including nuclear medicine, dosimetry and safeguards. All progress was monitored, and some aspects of the work programme were re-assigned.

WORK STATUS

Radionuclidic decay data

The status of each individual evaluation for the radionuclides selected as X- and γ-ray standards is summarised in Table 1. Specific problems in these data were discussed in some detail (see below). Some radionuclides were also re-assigned to newly participating laboratories: CIEMAT/UNED to evaluate decay data for ^{66}Ga, ^{125}Sb and ^{243}Am. All half-life data are being evaluated for the CRP by Woods et al (NPL).

Table 1. Status of evaluations for radionuclides selected for inclusion in the recommended database

Nuclide	Evaluator	Completed	Comment
^{22}Na	INEEL	**x**	
^{24}Na	INEEL	**x**	
^{40}K	INEEL	**x**	
^{46}Sc	INEEL	**x**	
^{51}Cr	INEEL/PTB	**x**	
^{54}Mn	INEEL/PTB	**x**	
^{56}Mn	NPL/AEA		Evaluated, to be reviewed
^{55}Fe	LNHB	**x**	
^{59}Fe	LNHB		Evaluation underway
^{56}Co	NPL/AEA		To be re-evaluated: new measurements (eg., Raman and Molnar)
^{57}Co	KRI		Evaluated, to be reviewed
^{58}Co	LNHB	**x**	
^{60}Co	INEEL	**x**	
^{64}Cu	INEEL		To be evaluated
^{65}Zn	INEEL	**x**	
^{66}Ga	CIEMAT/UNED		New evaluator, to be evaluated; new measurements
^{67}Ga	KRI		Evaluated, to be reviewed
^{68}Ga	PTB	**x**	
^{75}Se	LBL/PTB	**x**	
^{85}Kr	NPL/AEA		Evaluated, to be reviewed
^{85}Sr	PTB		Evaluated, to be reviewed
^{88}Y	PTB		Evaluated, under review
^{93m}Nb	KRI		Evaluated, to be reviewed
^{94}Nb	NPL/AEA		Evaluated, to be reviewed

Nuclide	Evaluator	Completed	Comment
^{95}Nb	INEEL	**x**	
^{99}Mo	LNHB/ KRI		Evaluated, under review
^{99m}Tc	LNHB		Evaluated, under review
^{103}Ru	NPL/AEA		To be evaluated
^{106}Ru-^{106}Rh	NPL/AEA		To be evaluated
^{110m}Ag	INEEL		To be evaluated
^{109}Cd	PTB	**x**	
^{111}In	KRI	**x**	
^{113}Sn	INEEL	**x**	
^{125}Sb	CIEMAT/UNED		New evaluator, evaluation underway
^{123m}Te	LNHB	**x**	
^{123}I	LNHB		Evaluation underway
^{125}I	PTB	**x**	
^{129}I	KRI		Evaluation underway
^{131}I	LNHB		Evaluation underway
^{134}Cs	USP		To be evaluated
^{137}Cs	INEEL	**x**	
^{133}Ba	KRI		Evaluated and reviewed: to be revised
^{139}Ce	PTB	**x**	
^{141}Ce	PTB	**x**	
^{144}Ce	PTB		Evaluation underway
^{153}Sm	INEEL		Evaluated and reviewed: to be revised
^{152}Eu	USP		Evaluated, to be reviewed
^{154}Eu	KRI		Evaluated, to be reviewed
^{155}Eu	KRI		Evaluation underway
^{166m}Ho/^{166}Ho	PTB		Evaluated, to be reviewed

Nuclide	Evaluator	Completed	Comment
^{170}Tm	KRI	**x**	
^{169}Yb	PTB and LNHB		Evaluated, to be reviewed
^{192}Ir	LBL/INEEL/USP	**x**	
^{198}Au	PTB		Evaluated, to be reviewed
^{203}Hg	NPL/AEA		Evaluated, to be reviewed
^{201}Tl	PTB		Evaluated, to be reviewed
^{207}Bi	LNHB	**x**	
^{226}Ra (and daughters)	INEEL		To be evaluated; new measurements
^{228}Th (and daughters)	NPL/AEA		To be evaluated
^{234m}Pa	NPL/AEA		Evaluation underway
^{241}Am	KRI		Evaluated, to be reviewed
^{243}Am	CIEMAT/UNED		New evaluator, to be evaluated

INEEL - Idaho National Engineering and Environmental Laboratory (USA)

PTB - Physikalisch Technische Bundesanstalt (Germany)

NPL - National Physical Laboratory (UK)

LNHB - Laboratoire National Henri Becquerel (France)

AEA - AEA Technology (UK)

KRI - V.G. Khlopin Radium Institute (Russia)

LBL - Lawrence Berkeley Laboratory (USA)

USP - University of Sao Paulo (Brazil)

CIEMAT - Centro de Investigaciones Energeticas, Medioambientales y Tecnologicas (Spain)

UNED - Universidad Nacional de Educacion a Distancia (Spain)

Specific evaluation problems were discussed:

^{56}Mn atomic data – advised to ignore X_L data, and calculate average beta-energies via log(ft) program.

^{67}Ga – accuracy of electron capture decay to ^{67}Zn ground state may be overstated by measurement of Simpson and Ntsoane. A value of 3.6(20) per 100 disintegrations will probably be recommended.

^{93m}Nb – simple decay scheme with some uncertainty in α_K of 30.77 keV gamma-ray; also disagreement between measurements of this parameter. Theoretical value of 2.62(8) 10^4 will be adopted.

^{133}Ba – decay scheme balance gives Pγ(356 keV) of 62.1(11) %, with a ground state electron-capture decay of zero. The ICC data for the 81 keV gamma-ray transition does not need to be anomalous to generate a consistent decay scheme (no need to argue penetration effects for this M1+E2 transition).

^{155}Eu – relative gamma-ray emission probabilities are well known, but absolute emission probabilities are poorly characterised. Evaluator requested measurements of the absolute gamma-ray emission probabilities within the CRP.

^{203}Hg atomic data - X_L data required. Obtain X-ray energies form Browne and Firestone, and calculate conversion-electron data from gamma-ray emission probabilities.

Schönfeld had prepared a listing of X_K-ray energies and emission probabilities for all the radionuclides in Table 1 when possible. These data will be adopted in the evaluations. During the debate on X_L-rays, the team noted the poor quality of input data for the various subshells – interest in X_L-rays is for nuclei above A=200, and these parameters should only be quoted for ^{198}Au and above.

^{24}Na, ^{46}Sc, ^{60}Co, ^{94}Nb and ^{111}In internal conversion coefficients; Helene wished to know whether these parameters were derived for the CRP from measurements or extracted from theoretical tabulations: ^{24}Na from theory; ^{46}Sc from theory; ^{60}Co mixed (experiment and theory); ^{94}Nb from theory; ^{111}In mixed (from experiment and theory).

Some other points were noted:

(i) When relative emission probabilities have been measured, these parameters are evaluated as well as the normalisation factor; this combination of data is then used to generate absolute emission probabilities. Thus, both relative and absolute emission probabilities are included in the evaluation exercise, and both can be included in the IAEA-TECDOC.

(ii) Emphasis will be placed on the gamma rays most suited as detector calibrants, and only these emissions will be listed in the final CRP dataset (i.e., only a limited number of strong lines will be recommended).

(iii) Detailed comments and complete decay-data listings will not necessarily be included in the IAEA-TECDOC; however, the user will be referred to the parallel publications of the DDEP (published by CEA-PTB).

ACTIONS:

Helmer: combine LWEIGHT packages (limitation of statistical weight; Rajeval; normalised residual; Chechev procedure (see also 'Other issues')) – possible action on Browne (LBL).

Nichols: prepare comments on ^{56}Mn and ^{203}Hg evaluations for submission to Browne for review, and organise similar submissions for ^{85}Kr and ^{94}Nb (S. Woods (NPL)).

Los Arcos: agreed timetable of CRP evaluations – ^{125}Sb/^{125m}Te, ^{243}Am and ^{66}Ga.

Calibration by coincidence method (Hlaváč) – see Appendix 4

Fourteen radionuclides from the CRP list of X- and gamma-ray standards were judged to be of primary interest for detector calibration via the coincidence method: ^{24}Na, ^{46}Sc, ^{60}Co, ^{66}Ga, ^{75}Se, ^{88}Y, ^{94}Nb, ^{111}In, ^{123m}Te, ^{133}Ba, ^{134}Cs, ^{152}Eu, ^{154}Eu and ^{207}Bi, along with the 11.4 and 4.4 MeV cascade from the ^{11}B(p,γ)^{12}C* reaction. These nuclides cover the energy range from 80 to 2700 keV (and 80 to 11400 keV with the nuclear reaction included). Available data on angular correlations were assembled, and evaluated according to the recommended evaluation procedures. Theoretical calculations were performed for the second and fourth angular correlation coefficients (A_{22} and A_{44}), based on spin, transition multipolarities and mixing ratios. The evaluated data are in good agreement with the theoretical values, apart from ^{154}Eu.

ACTION:

Hlaváč – extend coincident approach for detector calibration to nuclear reactions so as to include higher energies.

Nuclear reactions (Marcinkowski) – see Appendix 4

Evaluations were made of the gamma-ray production cross sections from thermal neutron capture by ^{14}N, ^{35}Cl, ^{48}Ti and ^{52,53}Cr, and from resonance capture of protons by ^{14}N, ^{23}Na and ^{27}Al; all of these recommended data have been tabulated. Data uncertainties were also assessed. Evaluations were also undertaken for the intensity ratios for pairs of gamma-rays from multi-gamma cascades following proton capture by ^{45}Sc and ^{59}Co.

Various observations and recommendations can be made:

- uncertainties of cross sections for neutron capture by ^{14}N are of order 1-3%;
- cross sections of other reactions have higher uncertainties (factor of 2-3 greater);
- data re-measured for calibration purposes would achieve improved accuracy;
- could achieve higher energies (up to 18 MeV) by using the emissions from the ^{7}Li(p,γ) reaction;
- ^{11}B(p,γ_o) reactions could be used up to 90 MeV, with an uncertainty of 10% in the gamma-ray production cross sections;
- ^{14}N, ^{27}Al(p,γ_o) reactions should be evaluated in a search for better accuracy.

Covariances (Helene) – see Appendix 4

Covariances between gamma-ray emission probabilities were calculated for a few of the proposed primary standards in which the gamma-ray spectrum was dominated by two cascading transitions after beta-transition feeding. Two distinct groups were found that required different hypotheses:

(i) specific nuclides with very simple decay schemes (^{94}Nb and ^{46}Sc) have strongly correlated gamma-ray intensities;

(ii) other radionuclides (^{60}Co and ^{24}Na) require the inclusion of other levels and their beta feeding to give gamma-ray emission probabilities that are almost uncorrelated.

A HPGe detector was calibrated using radioactive sources that have simple decay schemes (one or two gamma-rays). ^{152}Eu was counted, and the branching ratios and beta feeding fractions were determined by fitting the appropriate expressions with the Least-Squares Method. Gamma-ray emission probabilities and their covariance matrix were calculated; a well-correlated data set is generated, and a similar exercise should be undertaken for the beta-decay data.

A covariance matrix was derived for the standard gamma-ray energies to be published by Helmer and van der Leun (Nucl Instrum Meth Phys Res (2000)), using the Least-Squares Method. The results show that the correlation coefficients for about 70 gamma-ray energies are greater than 0.9; while five gamma-ray energies have correlation coefficients smaller than -0.6. Gamma-ray emissions from ^{60}Co, ^{57}Co, ^{133}Ba, ^{192}Ir and ^{137}Cs sources were measured with a HPGe detector - these experimental results were used to improve the covariance matrix of the standard gamma-ray energies and their uncertainties as published by Helmer and van der Leun.

ACTION:

Helene - provide discussion report for the CRP on the correct use of the Helmer and van der Leun gamma-ray energies for Ge detector calibration, especially in terms of the covariances between the calibration values. This document should be incorporated into the resulting IAEA-TECDOC report.

Experiments (Molnar)

A simple and accurate method of detector efficiency calibration has been developed by combining widely-accepted radionuclide standards with emissions from the ^{14}N(n,γ) reaction. This calibration can be used to determine new relative gamma-ray intensities for ^{56}Co, ^{66}Ga and the ^{35}Cl(n,γ) capture reaction, with up to 1% standard deviation for ^{56}Co and ^{66}Ga and 1.5% standard deviation for ^{35}Cl(n,γ) reaction. The Budapest data for ^{66}Ga agree with equivalent data from Lawrence Berkeley National Laboratory, and support the observed problem of adopting a linear extrapolation approach to the efficiency curve above 2.75 MeV.

ACTION:

Molnar - new data have been generated for the decay of ^{56}Co and ^{66}Ga that should be definitely communicated to the relevant CRP evaluators. Similarly, the resulting high-energy gamma-ray data for the ^{35}Cl(n,γ) reaction data should be communicated and considered for adoption by Marcinkowski.

Other issues: discrepant data

Participants acknowledged the usefulness of the bootstrap method of discrepant data analysis (as described by Helene), but deemed usage impractical in the current CRP.

Chechev noted his procedure for the handling of discrepant data. Under certain circumstances, there is a danger of overestimating the uncertainty if the unweighted average is chosen; similarly, too low an uncertainty could be recommended from the analysis of other discrepant data sets. Two rules were suggested to avoid these difficulties, and should be incorporated into LWEIGHT.

ACTIONS

Helmer: combine LWEIGHT packages (limitation of statistical weight; Rajeval; normalised residual; Chechev procedure) – possible action on Browne (LBL).

Nichols: prepare ^{56}Mn and ^{203}Hg comments on evaluation for submission to Browne to be reviewed, and to organise similar submissions for ^{85}Kr and ^{94}Nb (S. Woods (NPL)).

Los Arcos: agreed timetable of CRP evaluations – ^{125}Sb/^{125m}Te, ^{243}Am and ^{66}Ga.

Hlaváč: extend coincident approach for detector calibration to nuclear reactions so as to include higher energies.

Helene: provide discussion report for the CRP on the correct use of the Helmer and van der Leun gamma-ray energies for Ge detector calibration, especially in terms of the covariances between the calibration values. This document should be incorporated into the resulting IAEA-TECDOC report (as an Appendix).

Molnar: new data have been generated for the decay of ^{56}Co and ^{66}Ga that need to be definitely communicated to the relevant CRP evaluators. Similarly the resulting high-energy gamma-ray data for the ^{35}Cl(n,γ) reaction data should be communicated and considered for adoption by Marcinkowski.

REFERENCES

1. "Report on the Consultants' Meeting on the Preparation of the Proposal for a Co-ordinated Research Project to Update X- and γ-ray Decay Data Standards for Detector Calibaration", A Nichols and M Herman, INDC(NDS)-378, May 1998.
2. "X-ray and Gamma-ray Standards for Detector Calibration", IAEA-TECDOC-619, IAEA Vienna, 1991.
3. "Update of X- and γ-ray Decay Data Standards for Detector Calibaration and Other Applications", Summary Report of the First Research Co-ordination Meeting, 9-11 December 1998, M Herman and A Nichols, INDC(NDS)-403, July 1999.

APPENDIX 1

International Atomic Energy Agency

Second Research Co-ordination Meeting on

Update of X- and Gamma-ray Standards for Detector Calibration

PTB, Braunschweig, Germany

10 – 12 May 2000

AGENDA

Wednesday, 10 May

09:00-09:30 Opening Session
Opening address (E. Schönfeld, PTB)
(M. Herman, IAEA Nuclear Data Section)
Election of Chairman
Adoption of Agenda

09:30-09:45 Report on the Decay Data Evaluation Project meeting (Helmer)

09:45-10:00 Review of actions from 1st RCM

10:00-12:30 Status of radionuclide evaluations according to Table 1 of INDC(NDS)-403

12:30-14:00 Lunch Break

14:00-18:00 Status of evaluations
- Selection of gamma decay standards for detector calibration using coincidence method (Hlaváč)
- Compilation and evaluation of nuclear reactions relevant to detector calibration (Marcinkowski)
- Determination of covariances between gamma-ray intensities and energies (Helene)
- Summary report on the gamma-ray measurements and evaluation of decay data at LNHB (Bé)
- The final uncertainties of the evaluated values (Chechev)
- Consistency of neutron and proton capture intensity standards - new relative intensities for ^{56}Co, ^{66}Ga decay and ^{35}Cl(n,γ) reaction gamma-rays (Molnar)

Thursday, 11 May

09:00-12:30 Discussion of problems encountered in radionuclide evaluations
- ^{67}Ga - evaluation of the probability of the EC-transition to the ground state of ^{67}Zn
- ^{93m}Nb - ICC
- ^{133}Ba - ICC and gamma multipolarity admixtures
- ^{94}Nb, ^{111}In, ^{46}Sc, ^{60}Co and ^{24}Na – ICC
- ^{56}Mn and ^{203}Hg - X-ray and electron data

12:30-14:00 Lunch Break

14:00-18:00 Drafting of the meeting report

Friday, 12 May

09:00-12:30 Drafting of the meeting report
12:30-14:00 Lunch Break
Demonstrations
- BANDRRI: a decay data presentation tool (Los Arcos)
- Demonstration of the LNHB Web server (Bé)

14:00-16:00 Drafting of the meeting report

16:00-17:00 Adoption of the meeting report
Final discussion

APPENDIX 2

International Atomic Energy Agency

Second Research Co-ordination Meeting on

Update of X- and Gamma-ray Standards for Detector Calibration

Physikalisch Technische Bundesanstalt

Braunschweig, Germany

10 - 12 May 2000

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APPENDIX 3

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 (Schönfeld)

1.3. γ-ray standards for detector calibration/ordered by energy and radionuclide (all)

1.4. Covariances for selected γ-ray standards (Helene and 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 (Schönfeld)

3.3 Gamma-ray standards (Bé)

3.4. Covariances (Helene and 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 (Schönfeld)

5.3 (-ray standards (all evaluators)

5.4 Nuclear reactions (Marcinkowski)

6. LIST OF PARTICIPANTS (Herman)

APPENDIX 4

Technical input

Readers, please note:
the attachments were presented by CRP members
at the beginning of the meeting - subsequent discussions
have resulted in minor modifications to proposed work plans
that are NOT included in these texts.

IAEA Co-ordinated Research Programme to Update X- and Gamma-ray Decay Standards for Detector Calibration and Other Applications

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Progress Report

The overall objective of the IAEA-CRP to Update X- and Gamma-ray Decay Data Standards for Detector Calibration and Other Applications is to improve the quality of recommended decay data in such important non-energy applications as safeguards, material analysis, environmental monitoring, medicine, waste management, dosimetry and spectroscopy. ALN and Mike Herman (IAEA-NDS) prepared a summary report of the First CRP Meeting in Vienna (9-11 December 1998), that was issued as an IAEA-NDS document (INDC(NDS)-403).

DOS-based programs have been developed at Brookhaven National Lab, Lawrence Berkeley National Lab, CEA Saclay and PTB Braunschweig to assist in data processing. A 10-day workshop was organised at Lawrence Berkeley National Laboratory to develop understanding of the various decay-data parameters and outline the workings of the calculational tools:

GTOL, LOGFT, GABS, HISCC, ALPHAD, ENSDAT, EMISSION, ICC,

EC-CAPTURE, LWEIGHT and NSR.

ALN attended the second week of this programme. The codes of particular interest are EMISSION and ICC to calculate x-ray and theoretical internal conversion coefficients, respectively. Both were shown to contain errors, and have been corrected in the latest versions that were posted to CRP members in January 2000.

Work has focused on incorporating measured decay data for Mn-56, Sb-125 and Hg-203 into the evaluation exercises. Significant modifications have been made to the recommendations as a result of the various additions. Specific atomic data have also been derived for these radionuclides. The decay data for Sb-125 are also being evaluated by Amalia Williart Torres and co-workers at UNED, Madrid (with support from CIEMAT); these data will be compared for this radionuclide.

All of the evaluated data are subject to modification prior to adoption by the CRP. Radionuclides awaiting decay-data evaluation by ALN include Th-228 (and daughters: Ra-224, Rn-220, Po-216, Pb-212, Bi-212, Tl-208 and Po-212), and Pa-234m.

Report of Activities on Nuclear Decay Data Evaluation in Spain

J.M. Los Arcos[1], A. Williart[2] and M. Shaw[2]
[1] Unidad de Metrología de Radiaciones Ionizantes, CIEMAT, Madrid, Spain
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Nuclear Decay Data Evaluation activities in Spain began only very recently when a team of researchers, from CIEMAT (Unidad de Metrología de Radiaciones Ionizantes, declared National Standards Lab. for Ionizing Radiations, and Unidad de Informática) and from Universidad a Distancia (UNED) (Depto de Física de Materiales, Nuclear Physics Group), started developing the Spanish Reference Data Base for Ionizing Radiations (BANDRRI) and, simultaneously, joined the International Decay Data Evaluation Project (IDDEP).

BANDRRI is an Internet reachable, relational data base, which will offer to ionizing radiation users nation-wide the recommended (as endorsed by the Standards Laboratory) decay/interaction data values to be used in ionizing radiations applications, both in radioactivity and dosimetry fields. This data base has been designed and developed at CIEMAT and is now under implementation, with partial data stored for 21 radionuclides and 10 dosimetric materials, which wil soon be completed with atomic radiation data. User-friendly data search and presentation, its high level of interactivness and its ability for placing queries make it very easy to incorporate other data blocks of interest to "sectorial" applications. A short trial connection to BANDRRI will be shown during the Meeting.

Within the IDDEP, after a training period, the CIEMAT/UNED joint team has been assigned the evaluation of ^{125}Sb, ^{134}Cs, ^{94}Nb, ^{131}Cs, ^{59}Ni and ^{63}Ni. A preliminary evaluation of ^{125}Sb was done between November 1999 and February 2000, in order to test the computer codes and criteria adopted within IDDEP. The full scale evaluation, collecting and reviewing 40 papers, started in February and is now about to be finished. The evaluated values for the half-life, 41 gamma-ray energies and emission intensities, the internal conversion coefficients and the ENSDF data set will be presented at this meeting. Ongoing tasks are the estimation of secondary radiation emissions and the final balance of the decay scheme, which should be completed by June 15. Additional evaluation of ^{125m}Te will follow immediately in parallel to either that of ^{134}Cs or ^{94}Nb, to be completed by the end of September and the end of January 2001, respectively. Depending on CRP needs, other nuclides will also be evaluated.

Summary report on the gamma-ray measurements and evaluation of decay data at CEA-BNM/LNHB

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The Laboratoire National Henri Becquerel (LNHB) was created during the year 1999, it includes the former Laboratoire des Rayonnements Ionisants (LPRI) and the Laboratoire de Métrologie des Rayonnements Ionisants (LMRI). The LNHB is affiliated with the French National Bureau of Metrology (BNM) and it is a part of the Commissariat à l'Énergie Atomique (CEA).

Measurements of X and gamma emissions

- The measurements of the X and gamma-ray emissions of Yb-169 were carried out in the framework of an Euromet exercise. Eleven laboratories took part in this international comparison, all the results were studied and published in a report [1].
- The emission probabilities of the KX- rays following the decay of Np-237 in equilibrium with Pa-233 is on progress with other European laboratories.

Evaluation of decay data

- The evaluation of the Yb-169 decay data following the Euromet exercise was done in collaboration with the PTB. This evaluation takes into account all the available measurements before and including the Euromet exercise. The evaluation report was sent to the DDEP (Decay Data Evaluation Project) group.
- The evaluation of Mo-99 and Tc-99m is under review with the KRI.
- A complete set of documentation: recommended data and evaluation remarks is enclosed with this contribution for the Fe-55, Co-58, Te-123m and Bi-207 evaluations.
- A gamma-ray library, dedicated to gamma spectroscopy, is being updated and re-evaluated. This library contains 350 radionuclides and will be available with the version 2 of the NUCLÉIDE package.
- As mentioned in the previous CRP report, the following publications were made in the framework of the DDEP collaboration:
  - a CD-ROM with the NUCLÉIDE software [2]
  - a report [3] with the new evaluated nuclides. It includes:

    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.
  - a report [4] including all the comments on the evaluations of the above data and also including the same data sets presented in the ENSDF style.
- The program to calculate Internal Conversion Coefficients is now available [5].

Other related activities

- The NUCLÉIDE database was linked with the program ETNA (Efficiency Transfer for Nuclide Activity measurements), which provides both detector efficiency transfer for various types of sources for coaxial source-detector geometries and coincidence summing corrections, in order to automatically import the necessary radionuclide decay data [6].
- A special study, in the framework of a nuclear waste management program, about nuclides with long half-life (> 30 a) was carried out.

On progress and future work

- The evaluation of Fe-59 is on progress; the evaluation of I-131 and I-123 will follow.
- A new version of the NUCLÉIDE package which will run with Access 2000 is under development.
- A new version of the IN-NUC (previously Saisinuc) is under tests. Several improvements are writing, the main idea is to avoid mistakes in typing data and to reduce labor. It will include:
  - the gamma relative intensities and uncertainties and the automatic calculation of the absolute intensities and uncertainties;
  - the automatic input of atomic data and Q values from tables;
  - a direct link with the calculation tools: Lweight, EC-capture, ICC v3.99;
  - the calculation of the gamma transition intensities and of the conversion electron energies and intensities and their uncertainties ;
  - the making, from the data set, of the necessary input file to the Emission program and the transfer of the results to the NUCLÉIDE data set.

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Half-Life Evaluations

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The principal effort has concentrated on the identification and retrieval of published texts which contain relevant half-life data. To date, almost 700 potential references have been identified and retrieval is still underway although the majority of documents have now been received. Each article is read to determine its applicability and, where relevant, information is recorded on the method of measurement, whether or not the effects of impurities were considered, the length of the measurement period, the determined half-life value together with its declared uncertainty and confidence level. Evaluations are proceeding on the same basis as used for TECDOC-619, keeping the cut-off date as 1968, as before. The effects of impurities can be significant and retaining the 1968 date is appropriate given that the use of Ge detectors only started in about 1965. To date, complete sets of data have been retrieved and evaluated for some 19 radionuclides but additional checking and validation is still required.

Status of the work

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a) Covariances between gamma-ray energies

We attempted to determine covariances between gamma-ray energies using recent publication by Helmer and van der Leun of the most important gamma-ray energies used in detector calibration. The obtained results were used in a self-calibration procedure and as a consequence all gamma-ray energies, uncertainties and correlation coefficients were updated. The obtained results show that the correlation coefficients of about 70 gamma-ray energies are greater than 0.9. Some gamma-ray energies have correlation coefficients lesser than - 0.6.

b) Covariances between gamma-ray emission probabilities in very simple decay schemes.

Covariance between gamma-ray probabilities were calculated in a few cases of primary standard sources with gamma-ray spectrum dominated by two cascading transitions following a beta transition which feeds the most excited level. We found two distinct groups of nuclides that due to their decay schemes require different sets of hypotheses: (i) nuclides with very simple decay schemes such as ^{111}In, ^{94}Nb and ^{46}Sc (the latter two with gamma intensities strongly correlated), (ii) nuclides that require inclusion of additional levels and beta feedings in the calculations (such as ^{24}Na and ^{60}Co). The latter group furnished gamma-ray emission probabilities almost uncorrelated.

c) ^{152}Eu

A HPGe detector was calibrated using radioactive sources with simple decay schemes (one or two gamma-rays). ^{152}Eu source was counted and the branching ratios and beta feeding fractions were determined by fitting the appropriate expressions with the Least-Squares Method. From the data, the gamma-ray emission probabilities and their covariance matrix were calculated.

d) Discrepant data: bootstrap procedure

The recommended value for a physical quantity must include its standard deviation. Besides its role on estimating confidence intervals, the standard deviation is the required quantity for combining present data with new ones. However, reducing discrepant data is a cumbersome task, since their probability density functions are unknown: in this case many difficulties arise when one intends to determine both a point and interval estimates. In order to overcome this difficulty, we propose using the median to estimate the measured quantity and using a bootstrap procedure to estimate the median standard deviation. A test of the method was performed.

The correlations between the emission probabilities of the more intense gamma rays in ^{152}Gd following ^{152}Eu decay

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The variance matrix between the emission probabilities of the strong gamma-rays following β^- decay of ^{152}Eu were determined in a specially designed experiment. The emission probabilities are deduced from the decay scheme, whose branching-ratios and beta feeding fractions were fitted to the observed peak areas.

1. Introduction

In a previous work [1], we presented the steps needed to find out the correlations between gamma-ray emission probabilities. Here, we show the results obtained by the application of the method to the intense gamma-rays of the β^- decay of ^{152}Eu.

We start calibrating both the total and the total-energy absorption detector efficiencies. Then we proceed to determine the decay parameters: branching-ratios and beta feeding fractions, with their full variance matrix (i.e., including the covariances), by fitting the observed peak areas with appropriate formulas deduced from the decay scheme, taking into account the secondary detection effects, particularly summing. Finally, from these data and the decay scheme, we calculate the emission probabilities along with the variance matrix.

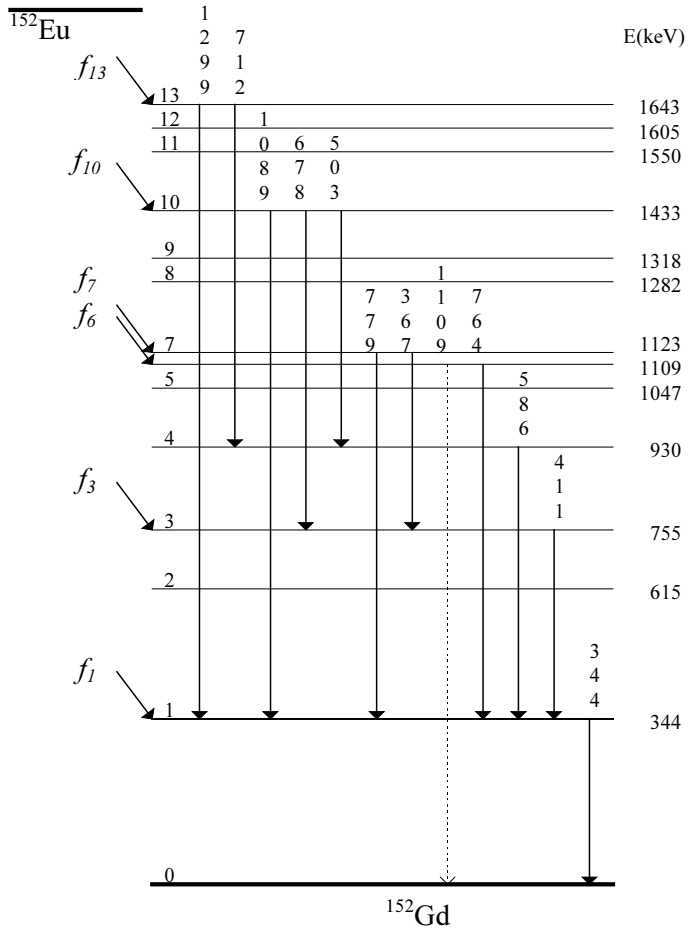

Figure. Part of ^{152}Eu decay scheme, showing the gamma-ray transitions that furnished data for the fit and the beta feeding fractions taken as free parameters.

2. The relation between peak-areas and ^{152}Eu β^- decay parameters

The number of counts in a 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 formulas are well described in the literature [2-6]. In this first experiment with ^{152}Eu, only the intense gamma-ray transitions from the β^- decay were taken into account, which are shown in Figure. In this summary, the symbols for the fraction of β^- decay to level i, f_i, and the branching ratios of electromagnetic transition from level i to level j, κ_{ij}, will be required. The transitions that are not used in the fit must be taken into account, leading to the separation of

the feeding fractions in two parts, $f_i = f_i' + f_{unaccounted}$, were only the f_i' are fitted. The constraints also must be modified, that on the beta feeding fractions is $\sum f_i = 0.2792$ and that on the branching-ratios sum are $\sum_{j,acconunted} \kappa_{ij} = 1 - \sum_{j,unsccounted} \kappa_{ij}$. Therefore, from all the ^{152}Gd levels taken into account, only four branching-ratios are free parameters to fit: $\kappa_{13\text{-}1}$; $\kappa_{10\text{-}1}$; $\kappa_{10\text{-}2}$; $\kappa_{7\text{-}1}$.

3. The detection system and the efficiency calibration

The system consists in a 160 cm^3 HPGe detector, with the source placed 20 cm away the detector capsule. Calibrated sources of ^{88}Y, ^{60}Co, ^{54}Mn with activities between 2 and 4 kBq calibrated in 4πβ–γ detector were used, along with ~30 kBq sources of ^{137}Cs, ^{152}Eu (only the low energy gamma-rays), and ^{133}Ba. The intention is to replace the data of Eu and Ba with data coming from sources with simple decay schemes: ^{57}Co, ^{51}Cr, etc., but we did not succeed in getting the material for preparing the corresponding calibrated sources yet. We do understand that using strong sources, in particular of Eu and Ba, warrant bad results. However, this is the prototype experiment, not intended to go beyond a rough estimate of the correlations (we are not giving emission probabilities).

4. Determining the quantities related to the decay scheme

All the gamma-ray transitions with emission probabilities greater than 0.1% were selected for observation, except the 1109 keV ($\gamma_{7\text{-}0}$) that constitutes a doublet with the 1112 keV. We were left, therefore, with 11 equations for the peak areas of the transitions plotted as full lines in the Figure, and 10 parameters: f_1 ; f_3 ; f_7 ; f_{10} ; f_{11} ; $\kappa_{13\text{-}1}$; $\kappa_{10\text{-}1}$; $\kappa_{10\text{-}2}$; $\kappa_{7\text{-}1}$; and the number of disintegrations. The constraints: the total feeding by the β^- channel, and the sums of the branching-ratios for the levels 13, 10 and 7, were used to eliminate the parameters f_6 , and $\kappa_{13\text{-}4}$, $\kappa_{10\text{-}4}$, $\kappa_{7\text{-}3}$, respectively. Uncertainties in the conversion coefficients were neglected throughout the calculations, but they can be included analogously to the uncertainties due to other scheme dependent quantities.

The disintegration data was obtained from a 10 kBq ^{152}Eu source, with the detector heavily shielded. The peaks were fitted with a gaussian plus exponential tails over a step and a 2nd degree polynomial. The least-squares fit [7,8] was performed, resulting in a chi-square value equal to 2.23 with one degree of freedom, with 15% probability of being exceeded. In Table I, we list the decay parameters obtained, including the number of disintegrations (Ω), standard deviations and the correlation matrix.

Table 1. ^{152}Eu β^- fitted decay parameters. The first column presents the names of the parameters, the second the values and the third the standard deviations. The rest of the table is the correlation matrix.

parm.	value	uncert.	correlation matrix									
			f_1	f_3	f_7	f_{10}	f_{13}	$\kappa_{13\text{-}1}$	$\kappa_{10\text{-}1}$	$\kappa_{10\text{-}3}$	$\kappa_{7\text{-}1}$	Ω
f_1	0.0732	0.0016	1.0	-.06	-.98	-.77	-.79	.17	-.20	.16	-.54	.86
f_3	0.00911	0.00013	-.06	1.0	-.01	-.04	.01	-.04	.19	-.25	.65	-.01
f_7	0.1436	0.0012	-.98	-.01	1.0	.67	.71	-.19	.09	-.06	.48	-.81
f_{10}	0.0254	0.0003	-.77	-.04	.67	1.0	.64	-.06	.48	-.40	.37	-.71
f_{13}	0.0188	0.0002	-.79	.01	.71	.64	1.0	-.04	.24	-.18	.39	-.87
$\kappa_{13\text{-}1}$	0.8943	0.0013	.17	-.04	-.19	-.06	-.04	1.0	-.01	-.09	-.09	.05
$\kappa_{10\text{-}1}$	0.714	0.003	-.20	.19	.09	.48	.24	-.01	1.0	-.94	.06	-.23
$\kappa_{10\text{-}3}$	0.1936	0.0022	.16	-.25	-.06	-.40	-.18	-.09	-.94	1.0	-.04	.18
$\kappa_{7\text{-}1}$	0.9403	0.0008	-.54	.65	.48	.37	.39	-.09	.06	-.04	1.0	-.44
Ω	123.3E8	1.1E8	.86	-.01	-.81	-.71	-.87	.05	-.23	.18	-.44	1.0

The overall precision is not good and also we do not have a reliable efficiency calibration. However, the results obtained are compatible with published data.

5. Determining the emission probabilities

With the decay scheme parameters of ^{152}Eu, all the gamma-ray transition probabilities along with their variance matrix can be deduced. The result is shown in Table 2, only for the gamma-ray used for calibration purposes. Note that all the correlations are moderately important, which follows from properties of the decay scheme, reflected in the set of equations.

Table 2. Gamma-ray emission probabilities and correlation for the intense gamma-rays.

energy	intensity	standard	correlation matrix				
		deviation	1299	1089	779	411	344
1299	0,01684	0,00018	1,00	0,59	0,68	0,36	-0,28
1089	0,01816	0,00025	0,59	1,00	0,59	0,34	-0,25
779	0,1352	0,0012	0,68	0,59	1,00	0,58	-0,37
411	0,02244	0,00009	0,36	0,34	0,58	1,00	-0,20
344	0,26692	0,00009	-0,28	-0,25	-0,37	-0,20	1,00

6. Conclusion

We presented the correlation matrix of the gamma-ray emission probabilities of the strong gamma-rays following the β^- decay of ^{152}Eu which forms a moderately correlated data set. However, this is not the simplest data set to be used when performing a precise efficiency calibration, when summing effects corrections are inevitable. Sum depends on the level feeding fractions from the parent nucleus and the branching ratios, which in turn form the easiest data set to determine when looking for the gamma-ray emission probabilities. We conclude, therefore, that it would be useful to publish the beta feeding fractions and the branching-ratios, with the respective variance matrix.

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Covariances between gamma-ray energies

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Covariance between experimental data are as significant as variance both in the evaluation of uncertainties, and to perform statistical tests. When standard data are used in calibrations, their covariances must be taken into account. Covariances are also necessary in order to update values every time new data are obtained.

We attempted to determine covariances between gamma-ray energies using recent publication by Helmer and van der Leun [1] of the most important gamma-ray energies for use in detector calibration. The obtained results were used in a self-calibration procedure and in consequence gamma-ray energies, uncertainties and correlation coefficients were updated.

1 - Energy calibration: input data and least square procedure

In ref. [1] Helmer and van der Leun used four different types of input data in order to update gamma-ray energies. The four groups of data are described below.

(a) Measured wavelengths of 22 gamma-ray transition energies using double-flat Si crystal. The Si lattice parameter adopted in ref. [1] is *d=0.192015540(40)nm*. Since those gamma-ray energies depend on the common factor *d*, they are correlated. Therefore, these data were transformed back to the ratio between *d* and gamma-ray wavelengths, and uncertainties were "unpropagated". In this case the data considered are $R_i = \lambda_i / d$, and the gamma-ray energies are related to R_i by $R_i = f / E_i$, where *f=hc/ed*. The data R_i were supposed to be non correlated.

(b) The group of gamma-ray energies relative to the 412 keV transition of ^{198}Au. (Data with superscript *h* in table 4 of ref. [1] were not taken into account.) The uncertainty of the 412 keV transition was not included.

(c) Gamma-ray energy differences were taken into account without transformation.

(d) The set of gamma-ray energies measured with HPGe detectors, included without transformation.

The fundamental constant *f* was also considered an experimental datum as well as a parameter to be fitted. As expected, *f* remains unchanged in the fit. The other parameters to be fitted are the gamma-ray energies.

Since some relations between gamma-ray energies and the experimental data are not linear, the Least-Squares Method was applied interactively.

Apart some details, the above procedure gives the same results quoted by Helmer and van der Leun in ref. [1]. The only relevant difference is the covariance matrix, not calculated in [1]. Table I shows the most important correlation coefficients ($\rho>0.95$ and $\rho<-0.5$).

Table I - The fifth column shows the correlation coefficients between some recommended gamma-ray energies (second and fourth columns, in keV).

^{108}Ag	433.9	^{125}Sb	463.4	0.96
^{124}Sb	1368.2	^{124}Sb	2090.9	0.96
^{152}Eu	678.6	^{152}Eu	1089.7	0.96
^{152}Eu	688.7	^{152}Eu	810.5	1.00
^{152}Eu	867.4	^{152}Eu	1112.1	0.99
^{152}Eu	1212.9	^{152}Eu	1457.6	1.00
^{154}Eu	1246.1	^{154}Eu	1494.0	0.99
^{160}Tb	879.4	^{160}Tb	966.2	0.97
^{161}Tb	25.7	^{161}Tb	74.6	0.97
^{161}Tb	48.9	^{161}Tb	74.6	0.99
^{169}Yb	177.2	^{169}Yb	307.7	0.96
^{169}Yb	198.0	^{169}Yb	261.1	0.96
^{182}Ta	84.7	^{182}Ta	152.4	0.96
^{182}Ta	1121.3	^{182}Ta	1189.0	1.00
^{182}Ta	1121.3	^{182}Ta	1221.4	1.00
^{182}Ta	1121.3	^{182}Ta	1231.0	0.98
^{182}Ta	1121.3	^{182}Ta	1257.4	0.97
^{182}Ta	1121.3	^{182}Ta	1273.7	0.99
^{182}Ta	1121.3	^{182}Ta	1289.1	1.00
^{182}Ta	1121.3	^{182}Ta	1373.8	0.99
^{182}Ta	1121.3	^{182}Ta	1387.4	0.99
^{182}Ta	1189.0	^{182}Ta	1221.4	1.00
^{182}Ta	1189.0	^{182}Ta	1231.0	0.98
^{182}Ta	1189.0	^{182}Ta	1257.4	0.97
^{182}Ta	1189.0	^{182}Ta	1273.7	0.99
^{182}Ta	1189.0	^{182}Ta	1289.1	1.00
^{182}Ta	1189.0	^{182}Ta	1373.8	0.99
^{182}Ta	1189.0	^{182}Ta	1387.4	0.99
^{182}Ta	1221.4	^{182}Ta	1231.0	0.98
^{182}Ta	1221.4	^{182}Ta	1257.4	0.97
^{182}Ta	1221.4	^{182}Ta	1273.7	0.99
^{182}Ta	1221.4	^{182}Ta	1289.1	1.00
^{182}Ta	1221.4	^{182}Ta	1373.8	0.99
^{182}Ta	1221.4	^{182}Ta	1387.4	0.99
^{182}Ta	1231.0	^{182}Ta	1273.7	0.99
^{182}Ta	1231.0	^{182}Ta	1289.1	0.98
^{182}Ta	1231.0	^{182}Ta	1373.8	0.99
^{182}Ta	1231.0	^{182}Ta	1387.4	0.99
^{182}Ta	1257.4	^{182}Ta	1273.7	0.96
^{182}Ta	1257.4	^{182}Ta	1289.1	0.97
^{182}Ta	1257.4	^{182}Ta	1373.8	0.96
^{182}Ta	1257.4	^{182}Ta	1387.4	0.96
^{182}Ta	1273.7	^{182}Ta	1289.1	0.99
^{182}Ta	1273.7	^{182}Ta	1373.8	1.00
^{182}Ta	1273.7	^{182}Ta	1387.4	0.99
^{182}Ta	1289.1	^{182}Ta	1373.8	0.99
^{182}Ta	1289.1	^{182}Ta	1387.4	0.99
^{182}Ta	1373.8	^{182}Ta	1387.4	0.99
^{192}Ir	588.6	^{192}Ir	884.5	0.97
^{198}Au	675.9	^{198}Au	1087.7	0.98
^{75}Se	66.1	^{75}Se	198.6	-0.66
^{75}Se	96.7	^{75}Se	303.9	-0.68
^{75}Se	121.1	^{75}Se	279.5	-0.75
^{75}Se	136.0	^{75}Se	264.7	-0.51
^{110}Ag	446.8	^{110}Ag	937.5	-0.65
^{133}Ba	160.6	^{133}Ba	223.2	-0.76
^{133}Ba	160.6	^{133}Ba	276.4	-0.69

2 - Updating energy data

Below, we show how some results obtained from the measurement of gamma-ray energies change adopted values and uncertainties of other non measured gamma-ray transition energies.

Gamma-ray energies from the ^{133}Ba, ^{192}Ir, ^{137}Cs, ^{60}Co, and ^{57}Co decay were measured in a HPGe detector. The calibration was made using gamma-ray energies from ref. [1] and the covariance matrix determined in this work. A self-calibration procedure [2] was applied.

Since the whole covariance matrix was used in all steps of the calibration, energies from the measured transitions as well as the energies of all transitions correlated with the measured ones were changed as a consequence of the measurement. Also, the covariance matrix of all gamma-ray energies were changed.

Table 2 shows energies and uncertainties of the measured transitions quoted in [1] and the final results obtained in the calibration. Table 3 shows some of the non-measured transitions and uncertainties that were changed as a consequence both of the new results of the measured quantities, and the procedure that included the complete covariance matrices.

Table 2 - Gamma-ray energies from ^{133}Ba, ^{192}Ir, ^{137}Cs, ^{60}Co, and ^{57}Co decay: energies and uncertainties from ref. [1] and from this work.

Energy (keV) from ref. [1]	Energy (keV) this work
53.1622(6)	53.6124(5)
79.6144(12)	79.6142(11)
80.9967(9)	80.9962(8)
160.6120(16)	160.6104(13)
223.2368	223.2371
276.3989(12)	276.3994(11)
302.8508	302.8512
356.0129(7)	356.0134(4)
383.8485(12)	383.8472(8)
661.657(3)	661.6577(6)
1173.228(3)	1173.2227(16)
1332.492(4)	1332.4962(21)
205.79430(9)	205.79430(8)
295.95650(15)	295.95650(14)
308.45507(17)	308.45513(15)
316.50618(17)	316.50602(15)
468.06885(26)	468.06877(25)
484.5751(4)	484.5751(4)
588.5810(7)	588.5819(5)
604.41105(25)	604.41116(25)
612.46215(26)	612.46203(26)
884.5365(7)	884.53745(55)
122.06065(12)	122.06063(13)

Table 3 - Some not measured gamma-ray energies were changed as a consequence of the measurement of correlated energies. This table shows some energies and uncertainties of unmeasured transitions before and after the experiment

Values before the experiment (ref. [1])		Values after the experiment	
Energy (keV)	Uncertainty (eV)	Energy (keV)	Uncertainty (eV)
^{94}Nb 702.6446	1.2	702.6452	0.9
^{110}Ag 657.7599	1.0	657.7604	0.9
^{124}Sb 645.8504	1.3	645.8511	1.0
^{125}Sb 635.9511	1.2	635.9518	1.0
^{132}Cs 667.7159	1.2	667.7165	1.0
^{144}Ce 696.5097	1.2	696.5103	0.9

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Covariances between gamma-ray emission probabilities in very simple decay schemes

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The covariances between the gamma-ray emission probabilities of the two strongest electromagnetic transitions following the decays of ^{94}Nb, ^{111}In, ^{46}Sc, ^{60}Co, and ^{24}Na were determined from their decay schemes and available data, resulting in correlation coefficients equal to 0.96, 1, 0, 0.015, and 0.04, respectively. The required assumptions and experimental and theoretical values have yet to be checked by the reviewers of these nuclides.

Introduction

When the gamma-ray spectrum is dominated by two cascading electromagnetic transitions following a beta transition which feeds only the most excited level, it is possible to calculate the covariance between the two gamma-ray emission probabilities. From all the nuclides called primary calibration standards in the 1999 Summary Report, the decay schemes of five nuclides fit well this simple picture, forming two groups, according to their detailed decay schemes. The nuclides: ^{94}Nb, ^{111}In, and ^{46}Sc, have truly simple decay schemes while ^{60}Co and ^{24}Na required the consideration of a few other levels and beta feedings in the calculations. For all other nuclides, it was impossible to retrieve the covariances from the existing data, being required specially designed experiments to determine them. In the following, we will present the calculations for the two groups of nuclides separately.

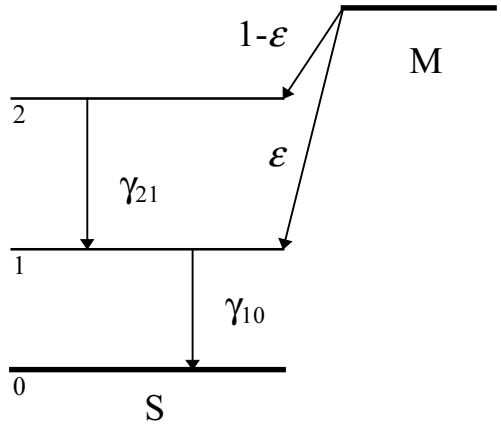

Figure 1. Decay schemes of ^{94}Nb, ^{111}In and ^{46}Sc for calculation purposes.

The cases of ^{94}Nb, ^{111}In and ^{46}Sc

The decay of these nuclides can be very well represented by the simple scheme of Fig. 1, were ε represents the beta feeding fraction to the intermediate level. Tables 1 and 2 show the values of the quantities needed to evaluate the covariances and the assumptions about each nuclide.

Since the internal conversion coefficients (ICC) are the main source of error in these cases, the resulting emission probabilities are correlated almost to the extent that the ICC are correlated. For the spherical nuclei ^{94}Mo and ^{46}Ti, with stretched E2 transitions of about the same energies, the correlation coefficient between the theoretical values of the ICC should approach 1. With respect to ^{111}In, we assumed that the quoted values are experimental and uncorrelated, due to the very different precision quoted for the two ICCs of about the same value, signing that the main source of error is not common to both values.

Table I. Beta feeding fractions outside the gamma cascade.

Nuclide	ε(%)	comments
^{46}Sc	0.0036(7)	neglecting γ_{20} cross-over $I_{\gamma 20}$~10^{-5} %
^{94}Nb	0	4th forbidden β transition
^{111}In	0.005(5)	feed through 151 keV γ-ray

Table II. Internal conversion coefficients. The fourth column presents the assumed correlation coefficient for the two ICCs at left, represented by ρ_α. The last row shows ρ_α between the theoretical values for the ICCs of ^{111}Cd.

Nuclide	Energy (keV)	ICC	ρ_α	Comments
^{46}Sc→^{46}Ti	1120.545 889.277	$9.2\cdot10^{-5}$ $1.57\cdot10^{-4}$	1	theoretical values for stretched E2 cascade, similar energies
^{94}Nb→^{94}Mo	702.622 871.091	$1.85\cdot10^{-3}$ $1.08\cdot10^{-3}$	1	theoretical values for stretched E2 cascade, similar energies
^{111}In→^{111}Cd	171.28 245.4	0.103(3) 0.0628(7)	0	experimental values, precision from counting statistics
			(0.5)	theoretical values for different multipolarities; experimental mixing-ratio for γ_{21}.

In the case of figure 1, the gamma-ray emission probabilities I_γ and respective standard deviations σ_γ are given by

$$I_{\gamma 21} = \frac{1-\varepsilon}{1+\alpha_{21}} \text{ with } \frac{\sigma_{\gamma 21}}{I_{\gamma 21}} = \sqrt{\frac{\sigma_{\alpha 21}^{\ 2}}{\left(1+\alpha_{21}\right)^2} + \frac{\sigma_\varepsilon^{\ 2}}{\left(1-\varepsilon\right)^2}} \tag{1}$$

and

$$I_{\gamma 10} = \frac{1}{1+\alpha_{10}} \text{ with } \frac{\sigma_{\gamma 10}}{I_{\gamma 10}} = \frac{\sigma_{\alpha 10}}{1+\alpha_{10}} \tag{2}$$

where the ICCs are symbolized by α_{ij} . The relative covariance is

$$\frac{\operatorname{cov}\left(I_{\gamma 21}, I_{\gamma 10}\right)}{I_{\gamma 21} I_{\gamma 10}} = \frac{\operatorname{cov}\left(\alpha_{21}, \alpha_{10}\right)}{\left(1+\alpha_{21}\right)\left(1+\alpha_{10}\right)} = \frac{\rho_\alpha \sigma_{\alpha 21} \sigma_{\alpha 10}}{\left(1+\alpha_{21}\right)\left(1+\alpha_{10}\right)} \quad . \tag{3}$$

A straightforward calculation leads to the correlation coefficient between the gamma-ray emission probabilities,

$$\rho_\gamma = \frac{\mathrm{cov}\left(I_{\gamma 21}, I_{\gamma 10}\right)}{\sigma_{\gamma 21}\sigma_{\gamma 10}} = \frac{\rho_\alpha}{\sqrt{\dfrac{1}{\left(1+\alpha_{21}\right)^2} + \dfrac{1}{\left(1-\varepsilon\right)^2}\left(\dfrac{\sigma_\varepsilon}{\sigma_{\alpha 21}}\right)^2}} \qquad (4)$$

which, for small conversion coefficients and beta feeding outside the gamma cascade, reduces to the cleaner formula

$$\rho_\gamma \cong \frac{\rho_\alpha}{\sqrt{1+\left(\dfrac{\sigma_\varepsilon}{\sigma_{\alpha 21}}\right)^2}} \quad .$$

In table III, we quote the values calculated by formula (4) above with the data from tables I and II, where it was assumed 3% of precision in the theoretical ICCs. Note that the results rely on the values of the uncertainties, being almost independent of the exact values of the decay quantities, reflecting the fact that these correlations are mostly a decay scheme property.

Table III. Correlation ρ_γ between the emission probabilities of the two strongest gamma-rays of a few nuclides.

Nuclide	**gamma energies**	**ρ_γ**	**comments**
^{46}Sc	1121;889	0.96	assuming $\sigma_{\alpha 21}/\alpha_{21} \approx 3\%$
^{94}Nb	703;871	1	
^{111}In	171;245	0	assuming ICCs are experimental (ρ_α=0)

The cases of ^{60}Co and ^{24}Na

The situation gets somewhat more complicated for these nuclides because there are other levels feed in the beta decay, with both strong gamma-rays loosing a bit of intensity due to a cross-over and γ_{10} getting some intensity that does not come through γ_{21}. The decay scheme is better represented in Fig. 2 than in Fig 1. The data required for the calculations can be found on tables IV and V, with the appropriate comments.

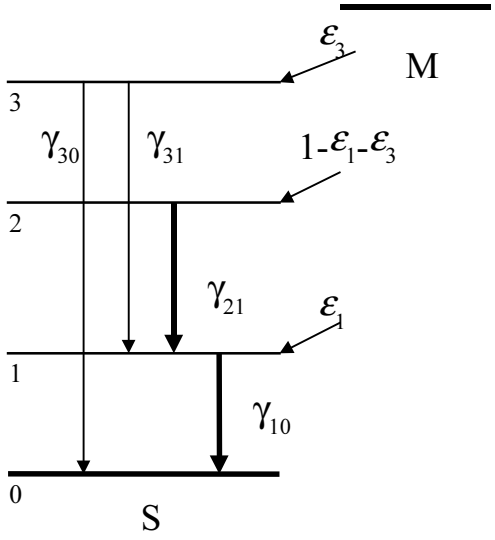

Figure 2. Decay schemes of ^{24}Na and ^{60}Co for calculational purposes.

Table IV. Emission probabilities required to calculate the correlation between gamma-ray intensities.

Nuclide	ε_1(%)	$I_{\gamma 30}$(%)	$I_{\gamma 31}$(%)	comments
^{60}Co	0.12(3)	0.0012(2)	0.0076(8)	"level 3" is indeed between levels 2 and 1 and is feed mainly by the 347 keV γ-ray
^{24}Na	0.003(3)	0.0011(2)	0.052(4)	"level 3" represents both the 3525 and 4238 keV levels

Table V. Correlation coefficients ρ_α for the conversion coefficients: internal (ICC) and pair (PCC).

Nuclide	energy (keV)	ICC(+PCC)	ρ_α	comments
^{60}Co →^{60}Ni	1173 1332	$1.74(4)\cdot10^{-4}$ $1.62(7)\cdot10^{-4}$	1	include PCC; theoretical values for stretched E2 cascade, similar energies
^{24}Na→^{24}Mg	2754 1369	$2.7\cdot10^{-6}$ $9.8\cdot10^{-6}$	1	do not include PCC; theoretical values for stretched E2 cascade, similar energies

The analogous of formulas (1-3) of section 2 are

$$I_{\gamma 21} = \frac{1-\varepsilon_1 - I_{t30} - I_{t31}}{1+\alpha_{21}} \;,\; \frac{\sigma_{\gamma 21}}{I_{\gamma 21}} = \sqrt{\frac{\sigma_{\alpha 21}{}^2}{\left(1+\alpha_{21}\right)^2} + \frac{\sigma_{\varepsilon 1}{}^2 + \sigma_{t30}{}^2 + \sigma_{t31}{}^2}{\left(1-\varepsilon_1 - I_{t30} - I_{t31}\right)^2}} \qquad (5)$$

$$I_{\gamma 10} = \frac{1-I_{t30}}{1+\alpha_{10}}, \frac{\sigma_{\gamma 10}}{I_{\gamma 10}} = \sqrt{\frac{\sigma_{\alpha 10}{}^2}{\left(1+\alpha_{10}\right)^2} + \frac{\sigma_{t30}{}^2}{\left(1-I_{t30}\right)^2}} \qquad (6)$$

$$\operatorname{cov}\left(I_{\gamma 21}, I_{\gamma 10}\right) = I_{\gamma 21} I_{\gamma 10} \frac{\operatorname{cov}\left(\alpha_{21}, \alpha_{10}\right)}{\left(1+\alpha_{21}\right)\left(1+\alpha_{10}\right)} + \frac{\sigma_{t30}{}^2}{\left(1+\alpha_{21}\right)\left(1+\alpha_{10}\right)} \quad \text{or} \qquad (7)$$

$$\frac{\operatorname{cov}\left(I_{\gamma 21}, I_{\gamma 10}\right)}{I_{\gamma 21} I_{\gamma 10}} = \frac{\operatorname{cov}\left(\alpha_{21}, \alpha_{10}\right)}{\left(1+\alpha_{21}\right)\left(1+\alpha_{10}\right)} + \frac{\sigma_{t30}{}^2}{\left(1-\varepsilon_1 - I_{t30} - I_{t31}\right)\left(1-I_{t30}\right)} \;, \qquad (7')$$

where the subscript *t* means transition, for instance, I_{t30} is the total (gamma+electron) transition intensity between levels 3 and 0. We prefer formula (7') to (7), because adimensional values are easier to deal with, even if it is not clearer.

There is no simple analogous for the case of figure 2 of the expression (4), the correlation between the gamma-ray emission probabilities. Therefore, table VI lists the relative standard deviations of the gamma-ray intensities given by (5) and (6), the relative covariance given by (7'), and the correlation calculated by the definition

$$\rho_\gamma = \frac{\mathrm{cov}\left(I_{\gamma 21}, I_{\gamma 10}\right)}{\sigma_{\gamma 21}\sigma_{\gamma 10}}.$$

In the calculations, the transition intensities between levels 3→0 and 3→1 were taken as the respective gamma-ray intensities, neglecting the other weak electron transitions. In the case of ^{24}Na, it was also neglected the contribution of 2869.5 keV γ-ray to $I_{\gamma 10}$, and, in the case of ^{60}Co, the 2506 keV cross-over transition.

Table VI. Correlation and relative covariance between the emission probabilities of the two strong gamma-rays, along with other values required for the calculations.

Nuclide	$\sigma_{\gamma 21}/I_{\gamma 21}$	$\sigma_{\gamma 10}/I_{\gamma 10}$	$\mathrm{cov}(I_{\gamma 21}, I_{\gamma 20}) / (I_{\gamma 21}\ I_{\gamma 10})$	ρ_γ
^{60}Co	$3.0\cdot10^{-4}$	$7.3\cdot10^{-6}$	$3.2\cdot10^{-11}$	0.015
^{24}Na	$5.0\cdot10^{-5}$	$2.0\cdot10^{-6}$	$4.0\cdot10^{-12}$	0.04

Conclusion

When the gamma-ray emission probabilities are deduced from simple decay schemes, it is possible to calculate their statistical correlation coefficient if the correlation between the internal conversion coefficients is known.

In the cases studied here, it turned out that the correlation coefficient is either negligible or very high, reflecting the existence or not of concurrent transitions in the decay scheme.

References

Data for ^{60}Co and ^{111}In are from *Table de Radionuclides*, BNM (1999); for the other nuclides, from ENSDF and the program ICC98. These correlation values can integrate the standard, provided all the data presented here is checked by the reviewers of these nuclides, which should also fill in the appropriate values and references.

Evaluation of Decay Schemes and Transition Probabilities for ^{57}Co, ^{67}Ga, ^{93m}Nb, ^{99}Mo, ^{111}In, ^{129}I, ^{133}Ba, ^{154}Eu, ^{155}Eu, ^{170}Tm and ^{241}Am

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Summary

An analysis of decay schemes and associated experimental data has been fulfilled for 9 of 11 radionuclides to be evaluated on the CRP work plan for V.Chechev (KRI, St. Petersburg). The ^{111}In, ^{133}Ba and ^{170}Tm decay data evaluations have been reviewed by the participants of the CRP and DDEP. The reviews of the ^{67}Ga and ^{241}Am evaluations are in progress. The evaluations of the ^{57}Co, ^{93m}Nb, ^{99}Mo, ^{154}Eu decay data have been prepared as drafts and an analysis for ^{129}I and ^{155}Eu is next.

Remarks on the final uncertainties of evaluated values

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There is not a single universal statistical procedure for producing recommended averages for sets of data, especially discrepant ones and those that constitute the small numbers of values (n). Therefore, if a detailed analysis results in the rejection of some values or the increase of osome uncertainties on certain physical grounds is impossible or difficult, the intuition of the evaluator becomes very important in the choice of a statistical procedure to give the "best value" [1, 2]. As seen for many evaluations when $1<\chi^2/(n-1)<2$, the LWEIGHT [3] or the EV1NEW [4] computer programs lead to good agreement between the two sets of results. Both programs use the Limitation of Relative Statistical Weights (LRSW) method and increase the final uncertainty of the weighted average of the discrepant data. Some problems arise for data sets with $\chi^2>2(n-1)$ and $\chi^2<(n-1)$.

In the first case (χ^2 greater than 2 (n-1), there is a danger of overestimating the uncertainty especially if an unweighted average is chosen. Examples of such overestimations were given earlier in my evaluations of the ^{90}Sr, ^{99}Mo and ^{111}In half-live [5]. For highly discrepant data, the Bayesian procedure can be used as well as the Normalised Residuals (NORM) and Rajeval (RAJ) techniques. These procedures adjust input uncertainties and can delete discrepant data without rejecting other values. However, I believe that these methods (especially RAJ) lead to unjustifiably low uncertainties in comparison with the Bayesian procedure (see Annex below, in which the data set from [4,5] are used).

In the second case (small χ^2), there is a risk of assigning too low an uncertainty to the evaluated value. This possibility can be avoided by adopting the two following rules:

(1) use the smallest of the input measurement uncertainties (σ_{min}) as a final uncertainty of the evaluated value, if σ_{min} is more than the uncertainty obtained from a statistical procedure; this rule is justified by the fact that almost any measurement is indirect and the measurements uncertainty includes the systematic error of the measurement method;

(2) use tS (t is the Student's coefficient for (n-1) degrees of freedom and a confidence level 0.68) as a final uncertainty of the evaluated value instead of an external uncertainty S; this rule is justified by the desirability to expand the final uncertainty as the number of data decrease and a primordial normal distribution transforms to Student's form.

I will give only one example of the evaluation of the experimental K internal conversion coefficient for the 84 keV gamma transition in ^{170}Yb. The weighted mean (WM) of eight measurements for α_K(84 keV) [1.48(5), 1.41(4), 1.37(4), 1.41(5), 1.46(7), 1.39(3), 1.41(3) and 1.43(4)] is 1.414 with an internal uncertainty of 0.014, a reduced χ^2 of 0.6 and an external uncertainty of 0.011. If we take formally WM=1.414(14) we will discard a systematic uncertainty of the measurement method connected, for example, with detection efficiency and use of K fluorescence yield (ω_K) which contributes significently to the total uncertainty of the best experimental values of α_K. Taking this into account, the smallest of the input uncertainties should be chosen as a final uncertainty of the WM: α_K(exp)=1,41(3). It should be noted that the third digit in WM cannot be given as no experimentalist can declare such an accuracy.

Annex

Half-life of ^{99}Mo measurements (n=15) in hours: 66.0(1), 66.0(15), 66.96(9), 67.2(2), 65.6(2), 66.7(1), 65.93(24), 65.95(4), 66.69(6), 66.5(2), 66.02(1), 66.16(30), 65.945(5), 65.924(6), 65.942(12).

The results of statistical processing with the different techniques: UWM=66.24(12), WM=65.952(3), CHV=66.24(12), UINF=65.952(19), PINF=65.952(19), BAYS=65.952(20), MBAYS=65.952(19), LWM=66.24(19), IEXW=66.25(12), NORM=65.941(7), RAJ=65.942(5). Recommended value is 65.952(20) [BAYS]. As seen NORM and RAJ give the smallest uncertainties (except WM).

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4. V.P. Chechev and A.G. Egorov. Appl. Radiat. Isot. 52(2000)601-608.
5. V.P. Chechev. INDC(NDS)-399, IAEA, Vienna, March 1999, pp. 116-122.

Recommended cross sections for thermal neutron capture by ^{14}N, ^{35}Cl, ^{48}Ti and ^{52,53}Cr, for resonance capture of protons by ^{14}N, ^{23}Na and ^{27}Al, and the ratios of intensities from multi-γ cascades following proton capture

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1. Thermal neutron capture reactions

Neutron capture reactions at thermal energies have proved to be a useful tool in efficiency calibration of high energy γ-ray detectors. The γ-ray emission probabilities P_γ are usually known to an accuracy suitable for that purpose (see e.g. IAEA-TECDOC-619 [1]). The emission probability is related to the total capture cross section σ_C ,

$$\sigma_\gamma = \sigma_C \frac{\Gamma_\gamma}{\Gamma} \text{ , with } \Gamma = \Gamma_\gamma = \sum_i \Gamma_{\gamma i} \text{ ,} \quad (1)$$

and the cross section $\sigma_{\gamma i}$ for emission of specific γ_i-rays of given energy $E_{\gamma i}$ is given by,

$$\sigma_{\gamma_i} = \sigma_C \frac{\Gamma_{\gamma i}}{\Gamma} = \sigma_C P_{\gamma i} \text{ .} \quad (2)$$

In Table 1 the total neutron capture cross sections for the target nuclei of interest are gathered. These cross sections are determined with an accuracy of 1% to 3%.

Table 1. Total neutron capture cross section at thermal energy.

Reaction	σ_C(b)	Ref.
^{14}N(n,γ)^{15}N	0.0798(14)	[2]
^{35}Cl(n,γ)^{36}Cl	43.60(46)	[3]
^{48}Ti(n,γ)^{49}Ti	8.5(2)	[4]
^{52}Cr(n,γ)^{53}Cr	0.86(3)	[5]
^{53}Cr(n,γ)^{54}Cr	18.6(6)	[6]

In Tables 2 to 5 the γ_i-ray-emission cross sections $\sigma_{\gamma i}$ calculated according to Eq. (2), using the emission probabilities $P_{\gamma i}$ per neutron capture from refs [7-11]for the thermal neutron capture reactions considered are listed.

Table 2. The evaluated thermal neutron capture cross section $\sigma_{\gamma i}$ for selected γ_i-rays from the $^{14}N(n,\gamma)^{15}N$ reaction and the corresponding γ_i-ray emission probabilities per neutron capture $P_{\gamma i}$ from ref. [7].

$E_{\gamma i}$(keV)	$\sigma_{\gamma i}$(mb)	$P_{\gamma i}$
1678.174(55)	5.77(18)	0.0723(18)
1884.879(21)	14.89(32)	0.1866(25)
2520.418(15)	4.62(1)	0.0579(7)
3532.013(13)	7.37(15)	0.0924(9)
3677.772(12)	11.88(24)	0.1489(15)
4508.783(14)	13.20(27)	0.1654(17)
5269.169(12)	23.96(45)	0.3003(20)
5297.817(15)	17.01(33)	0.2131(18)
5533.379(13)	15.79(32)	0.1975(21)
5562.062(17)	8.50(10)	0.1065(12)
6322.337(14)	14.90(28)	0.1867(14)
7298.914(33)	7.76(15)	0.0973(9)
8310.143(29)	3.36(8)	0.0422(5)
9149.222(47)	1.30(3)	0.0162(2)
10829.087(46)	10.89(25)	0.1365(21)

The uncertainty of $P_{\gamma i}$ in ref. [8] consists of a rather small statistical error supplemented by an 8% systematic error. This systematic error [8], however does not affect the ratios of P/P and therefore both uncertainties are given in the brackets in Table 3.

Table 3. The evaluated thermal neutron capture cross section $\sigma_{\gamma i}$ for selected γ_i-rays from the $^{35}Cl(n,\gamma)^{36}Cl$ reaction and the corresponding γ_i-ray emission probabilities per neutron capture $P_{\gamma i}$ from ref. [8].

$E_{\gamma i}$(keV)	$\sigma_{\gamma i}$(b)	$P_{\gamma i}$
516.73(8)	9.9(9)	0.227(9,20)
786.26(5)	4.20(40)	0.096(5,9)
788.40(5)	6.54(58)	0.150(3,12)
1164.72(5)	11.21(97)	0.257(8,22)
1600.82(6)	1.50(15)	0.0343(17,32)
1950.93(6)	8.15(67)	0.187(4,15)
1959.13(6)	5.28(40)	0.121(4,10)
2863.04(16)	2.62(23)	0.060(2,5)
3061.71(16)	1.53(13)	0.035(2,3)
5715.20(22)	2.24(19)	0.0514(6,42)
6110.82(22)	8.59(70)	0.197(2,16)
6619.42(23)	3.539(29)	0.0810(10,66)
6627.50(23)	2.02(17)	0.0464(10,38)
6977.56(24)	0.97(9)	0.0223(9,20)
7413.7(2)	4.36(36)	0.1000(10,81)
7790.05(25)	3.75(30)	0.0861(8,69)
8578.21(26)	1.28(11)	0.0294(6,24)

Table 4. The evaluated thermal neutron capture cross section $\sigma_{\gamma i}$ for selected γ_i from the $^{48}Ti(n,\gamma)^{49}Ti$ reaction. and the corresponding γ_i-ray emission probabilities per neutron capture $P_{\gamma i}$ from ref. [9]

$E_{\gamma i}$(keV)	$\sigma_{\gamma i}$(b)	$P_{\gamma i}$
341.707(4)	2.20(12)	0.2480(21)
1381.766(4)	7.49(44)	0.855(21)
1498.687(6)	0.416(26)	0.0489(13)
4881.645(21)	0.383(21)	0.0451(5)
6419.003(19)	2.59(15)	0.305(3)
6760.634(20)	3.94(22)	0.463(4)

The uncertainties of the emission probabilities in Table 4 contain a statistical error which amounts from 1% to 3% for the strong γ-transitions of interest and a systematic error due to the shapes of the efficiency calibration curves that was estimated to be 3%. An additional systematic error originates from the calculation of the absolute intensities. In this calculation the main uncertainty is caused by the capture cross section for ^{48}Ti of Table 1, which is known with an error of 2.4%. The total standard deviation of the intensities amounts to about 5% [9] and is included in the uncertainty of $\sigma_{\gamma i}$. However, only the first two uncertainties contribute to the errors of the ratios $P_{\gamma i}/P_{\gamma j}$.

Table 5. The evaluated thermal neutron capture cross section $\sigma_{\gamma i}$ for selected γ_i-rays from the ^{52}Cr(n,γ)^{53}Cr reaction. and the corresponding γ_i-ray emission probabilities per neutron capture $P_{\gamma i}$ from ref. [10].

$E_{\gamma i}$(keV)	$\sigma_{\gamma i}$(b)	$P_{\gamma i}$
2320.9(2)	0.128(16)	0.15(3)
5619.3(6)	0.129(26)	0.149(10)

The data for ^{52}Cr in Table 5 are rather uncertain as they were obtained in the early seventies or late sixties (see the references in [10]). However also the newer data of ref. [8] did not improve the experimental errors.

Table 6. The evaluated thermal neutron capture cross section $\sigma\gamma i$ for selected γi-rays from the 53Cr(n,γ)54Cr reaction and the corresponding γi-ray emission probabilities per neutron capture $P\gamma i$ from ref. [11].

$E_{\gamma i}$(keV)	$\sigma_{\gamma i}$(b)	$P_{\gamma i}$
834.861(52)	18.6(6)	1.0
1784.66(100	1.77(25)	0.095(13)
2239.07(100	2.42(29)	0.130(15)
6645.34(65)	2.31(25)	0.124(13)
7099.71(65)	1.90(29)	0.102(15)
8884.12(65)	11.22(153)	0.603(80)

2. Resonance proton capture reaction

Proton capture in the resonance region is also often used as a source of γ-rays for calibration of germanium detectors. To specify eqs. (1) and (2) for a resonance one has to replace the thermal neutron capture cross section σ_C by the proton capture cross section σ_R at the resonance energy E_R,

$$\sigma_R = 4\pi\lambda\!\!\!^{-}_{p}{}^{2}\frac{(2J+1)}{(2I_1+1)(2I_0+1)}\frac{\Gamma_p}{\Gamma} \quad . \qquad (3)$$

The resulting radiative capture cross section at the resonance energy is:

$$\sigma_\gamma = 4\pi\lambda\!\!\!^{-}_{p}{}^{2}\frac{(2J+1)}{(2I_1+1)(2I_0+1)}\frac{\Gamma_p\Gamma_\gamma}{\Gamma^2}, \qquad (4)$$

and the cross section for emission of a specific γ_i-ray is expressed via the emission probability $P_{\gamma i}$,

$$\sigma_{\gamma i} = \sigma_\gamma P_{\gamma i}. \qquad (5)$$

Using the measured resonance strenghts $S_{p\gamma}=(2J+1)\Gamma_p\Gamma_\gamma/\Gamma$ and the resonance widths Γ from refs. [12,13] the proton capture cross sections σ_γ were obtained from Eq. (4).

Table 7. Calculated resonance proton capture cross sections.

Reaction	E_p(keV)	Γ(eV)	$S_{p\gamma}$(eV)	σ_γ(mb)
$^{14}N(p,\gamma)^{15}O$	278	1700(500)	0.084	0.0886(261)
$^{23}Na(p,\gamma)^{24}Mg$	1416	90(30)	27(6)	75(30)
$^{27}Al(p,\gamma)^{28}Si$	992	70(14)	24(2)	81(17)

Taking the experimental emission probabilities $P_{\gamma i}$ for selected γ_i-rays [12-14] the corresponding emission cross sections $\sigma_{\gamma i}$ were calculated from eq. (5). The results are tabulated below.

Table 8. The evaluated proton capture cross sections $\sigma_{\gamma i}$ for selected γ_i-rays from the 14N(p,γ)15O reaction at the resonance energy ER=278 keV and the corresponding emission probabilities per proton capture $P_{\gamma i}$ from ref. [12].

$E_{\gamma i}$(keV)	$\sigma_{\gamma i}$(mb)	$P_{\gamma i}$
764.0	0.021(6)	0.232(6)
1381.0	0.051(15)	0.575(4)
2374.0	0.014(4)	0.158(6)
5183.0	0.014(4)	0.158(6)
6176.0	0.051(15)	0.575(4)
6793.0	0.021(6)	0.232(6)

Table 9. The evaluated proton capture cross sections σγi for selected γi-rays from the 23Na(p,γ)24Mg reaction at the resonance energy ER=1416 keV and the corresponding emission probabilities per proton capture Pγi from ref. [13].

$E_{\gamma i}$(keV)	$\sigma_{\gamma i}$(mb)	$P_{\gamma i}$
2754.0	70(28)	0.94(1)
8929.0	71(28)	0.93(1)

Table 10. The evaluated proton capture cross sections σγi for selected γi-rays from the 27Al(p,γ)28Si reaction at the resonance energy ER=992 keV and the corresponding emission probabilities per proton capture Pγi from ref. [14].

E_{γ}(keV)	$\sigma_{\gamma i}$(mb)	$P_{\gamma i}$
1778.9	76.6(166)	0.948(15)
10760.4	61.9(135)	0.766(15)

It should also be noted that the emission probabilities in Tables 2-6 and 8-10 should be applied for calibration purposes to spectra taken at an angle $\theta=55^{o}$ with respect to the beam direction under the assumption that the $\theta=55^{o}$ intensities are proportional to the 4π-integrated γ-ray yields since at this angle $P_2(\cos\theta)=0$ and this minimizes the influence of the angular distributions.

It has been already emphasized that the ratios of the emission probabilities $P_{\gamma i}/P_{\gamma j}$ are usually determined with better accuracy than $P_{\gamma i}$ itself. Therefore to determine the relative detector efficiency for high energies the "two-line method" is used in termal-neutron-capture and proton capture reactions. The calibration will be completely independent of previously measured efficiencies for high-energy γ-rays, when a high-energy and a low-energy γ-transition from the initial capturing state and via an intermediate state, both have 100% γ-ray branching to one level. This results in an intensity ratio $P_{\gamma 1}/P_{\gamma 2}=1$. Since the efficiency for the low-energy γ-rays is known from radioactive sources, the calculation of the efficiency for the high-energy γ-rays is straightforward. However, such ideal situations are extremely rare and therefore the (p,γ) reactions on light nuclei, with their broad choice of many sharp resonances, have certain advantages over the thermal-neutron capture reactions. So far eight such cascades were reported in [15] with uncertainties of $P_{\gamma 1}/P_{\gamma 2}$ estimated at about 2%. Later the intensity ratio of strong cascades in the more complicated multi-γ decay of six $^{23}Na(p,\gamma)^{24}Mg$ and $^{27}Al(p,\gamma)^{28}Si$ resonances has been measured with an accuracy of 0.5% and of three $^{11}B(p,\gamma)^{12}C$ resonances at a level of precision of 1-2% [16]. The latter extend the calibration energy range up to 13.92 MeV [16]. These data are included in the IAEA-TECDOC-619 [1].

For the more complicated multi-γ decays the final uncertainty in $P_{\gamma i}/P_{\gamma j}$ is essentially determined by our knowledge of the competing transitions that feed or deexcite the intermediate level or by the accuracy of efficiency calibration in the experiment that provided the intensity ratio. Bearing this in mind the content of ref. [1] has been extended to cover the intermediate calibration energies ranging from 8.36 MeV to 11.10 MeV by including cascades from five $^{45}Sc(p,\gamma)^{46}Ti$ and $^{59}Co(p,\gamma)^{60}Ni$ resonances or groups of resonances reported in [17-19]. The two targets are easily available. In case

of the thick-target measurements of [17] interference between resonances of opposite parity may destroy the symmetry of the angular distributions of the primaries around 90^o and therefore the calibration should take place at an angle of 90^o with respect to the proton beam in order to reproduce the geometry used in [17]. The above mentioned calibration error in intensity of the high-energy γ-rays with respect to the intensity of the low-energy γ-s amounts up to 9% and determines the overall uncertainty of the ratios $P_{\gamma 1}/P_{\gamma 2}$ and $P_{\gamma 1}/P_{\gamma 3}$ measured in [17] but is less in [18]. This accuracy is typical for routine (p,γ) experiments involving complicated multi-γ cascades from a high-excited capturing state to an intermediate state, even when the latter decays with a 100% γ-ray branching to one level.

In Table 11 we attach the energies and intensities of pairs of cascading γ-rays at1370 keV, 1377 keV and 1660 keV proton bombarding energies in the ^{45}Sc(p,γ)^{46}Ti energies [17,18] and at 1479 keV, 1540 keV and 2101 keV proton resonance energies in the ^{59}Co(p,γ)^{60}Ni experiment [19].

Table 11. Proton capture reactions with subsequent emission of γ-rays in cascade at energies Eγ1, Eγ2 and/or Eγ3 with emission probabilities P1, P2 and/or P3. Proton resonance energy is Ep.

Reaction	E_p(MeV)	$E_{\gamma 1}$(MeV)	$E_{\gamma 2}$(MeV)	$E_{\gamma 3}$(MeV)	P_1/P_2	P_1/P_3	Ref.
^{45}Sc(p,γ)^{46}Ti	1.377	9.688	1.121	0.889	0.947(67)	0.054(4)	[18]
	1.370-1.377	10.807	0.889		0.338(31)		[17]
		9.686	1.121	0.899	0.309(28)	0.185(17)	[17]
	1.653-1.660	11.084	0.889		0.217(20)		[17]
		9.963	1.121	0.889	0.439(40)	0.265(24)	[17]
^{59}Co(p,γ)^{60}Ni	1.479	8.479	1.333		0.194(14)		[19]
		8.359	1.333		0.167(12)		[19]
	1.540	8.419	1.333		0.136(10)		[19]
	2.101	8.971	1.333		0.086(6)		[19]

Errors are given in round brackets.

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Consistency of neutron and proton capture intensity standards New relative intensities for ^{56}Co, ^{66}Ga decay and ^{35}Cl(n,γ) reaction gamma rays

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Introduction

Efficiency calibration of gamma-ray detectors above 3 MeV is problematic because no primary radioactive standards are available at such high energies. For this reason, the multi-gamma emitters ^{56}Co ($T_{1/2}$=71 d) and ^{66}Ga ($T_{1/2}$=9.5 h) have long been considered attractive candidates for secondary standards providing γ rays up to 3.5 MeV and 4.8 MeV, respectively [1,2]. Unfortunately, early measurements of emission probabilities have relied on linear extrapolation of the full energy peak efficiency on a log-log scale [3,4], leading to biased results above 2 MeV γ-ray energy [5,6]. Unfortunately, many authors used those data for calibration thereafter. Hence the previous evaluation of standards has not come up with a firm recommendation, at least for ^{66}Ga [2]. Therefore, re-measuring the relative intensities of ^{56}Co and ^{66}Ga decay γ rays is a task of great importance.

Above 4.8 MeV only reaction γ rays can be used for detector calibration. In Refs. [1,2] suitable thermal neutron and proton resonance capture reactions are listed, both types provide γ rays up to about 11 MeV. So far only the ^{14}N(n,γ)^{15}N reaction has been accepted for a primary intensity standard [2] because its decay scheme is sufficiently simple to allow a simultaneous fit of emission probabilities and efficiencies, constrained by a model function. Relative accuracies of about 1% have been achieved at McMaster [7], forming the basis of the 1991 recommendation [2]. These data have been confirmed recently [8], hence the less accurate Argonne results [9], which also systematically disagree, have to be abandoned. It would be instructive to see, however, if efficiency calibrations based on the nitrogen capture lines on one hand, and on a combination of two-step cascades with 1:1 intensity ratios from various proton resonances (point pair method) on the other hand, provide consistent results.

Equivalence of calibrations with neutron and proton capture standards has been tested by us indirectly, in a precision measurement of relative intensities of ^{56}Co using a detector calibrated with nitrogen capture γ rays. Besides, relative intensities of comparable accuracy have also been obtained for the ^{35}Cl(n,γ)^{36}Cl reaction, a popular secondary standard thanks to its much higher cross-section. The 1991 recommendation [2] carries an 8% systematic uncertainty due to inaccuracies of efficiency calibration [10], making this important energy standard [11] less suitable for efficiency calibration. The new secondary standard has in turn been used to determine new, accurate relative intensities for the ^{66}Ga decay γ rays.

Efficiency calibration and ^{56}Co decay

The measurements have been carried out on a thermal neutron guide at the Budapest Research Reactor, using a Compton-suppressed HPGe spectrometer with a source-to-detector

distance of 23.5 cm. A simple yet accurate method of detector efficiency calibration has been invented by combining the widely accepted multi-gamma secondary standards ^{133}Ba and ^{152}Eu with the primary standards ^{24}Na and the ^{14}N(n,γ) reaction at high energies. For the two radioisotopes the 1991 recommendation [2] was used between 81–1408 keV, while for nitrogen the recent data by Jurney *et al.* [8] were preferred because of their better accuracy near the low-energy end of 1678 keV. The data points were least-squares fitted with a seventh-order polynomial on a log-log scale, using orthogonal polynomials and including a normalising factor for each separately measured data set but ^{152}Eu. The latter has been a calibrated source, hence absolute efficiencies could be obtained for full-energy (FE) and single-escape (SE) peaks, respectively, as shown in Figure 1. As only relative intensities had to be determined, the uncertainty of the calibrated standard was ignored, hence accuracies better than 0.5% between 100–3500 keV and 1% up to 6000 keV could be achieved for relative FEP efficiency.

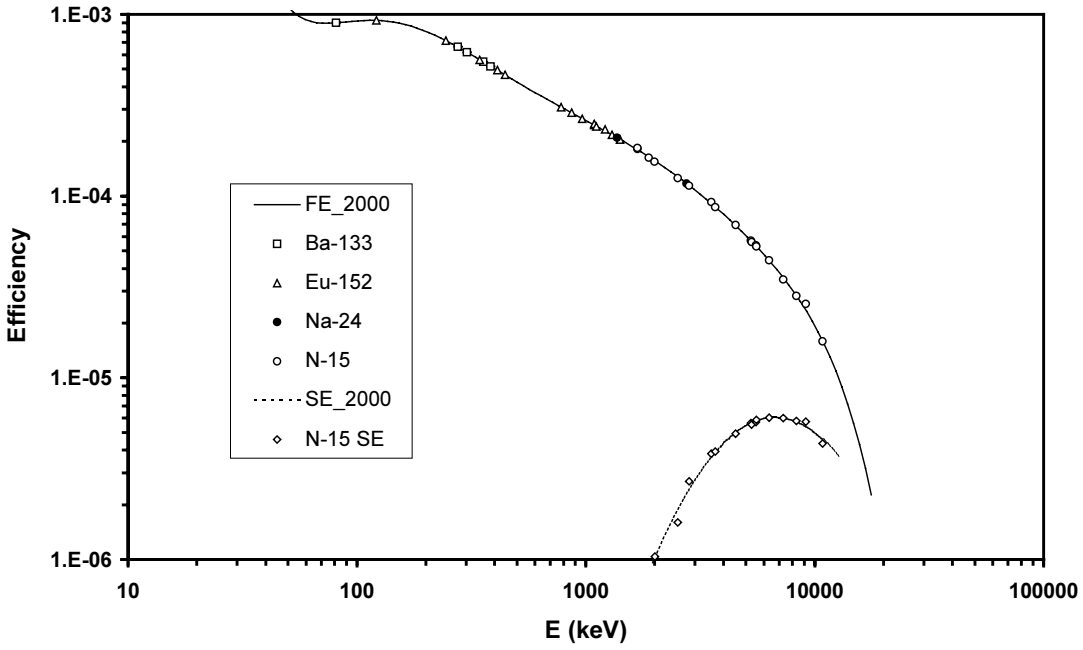

Figure 1. Absolute full energy (FE) and single escape (SE) efficiencies of the Budapest gamma-ray spectrometer in Compton suppression mode

Using the above calibration curve, new relative intensities have been determined for 20 gamma rays of ^{56}Co with an accuracy of 1% or better. Above 2.5 MeV the new intensities are systematically higher than those in the 1991 recommendation [2] (also adopted by the latest ENSDF evaluation [12]) for the aforementioned reasons, although the large uncertainties of the recommended data tend to mask this effect. Therefore, a new evaluation has been made including only those data for which the high-energy efficiencies were directly measured, rather than extrapolated or calculated by Monte Carlo methods. The methods varied from neutron capture [13,18] to proton resonances [14,15] and high-energy decay lines [16,17]. For the rest of data sources [4,19,20] only lines up to the 2598 keV transition were retained. The resulting weighted averages [21] are compared with the new data in Table 1. The agreement is excellent, as indicated by the reduced chi-square value of 0.96, demonstrating the equivalence of the various calibration procedures. Besides, the standard

uncertainties have become more uniform, typically smaller than 1%, with an unweighted average of 0.7%.

Table 1. New relative intensities for 20 strong ^{56}Co gamma-ray transitions, compared to the corresponding weighted averages of literature data [21]

E_γ (keV)	I_γ Wt. avg.	Unc.	% Unc.	I_γ Present	Unc.	% Unc.	Z-score/ Chi-sq.
847	100.000	0.200	*0.2*	100.000	0.300	*0.3*	
977	1.425	0.009	*0.6*	1.432	0.006	*0.4*	+0.6
1038	14.090	0.050	*0.4*	14.186	0.043	*0.3*	+1.5
1175	2.255	0.014	*0.6*	2.264	0.009	*0.4*	+0.5
1238	66.300	0.300	*0.5*	66.356	0.199	*0.3*	+0.2
1360	4.275	0.015	*0.4*	4.244	0.017	*0.4*	–1.4
1771	15.520	0.050	*0.3*	15.284	0.076	*0.5*	–2.6
1811	0.643	0.004	*0.6*	0.644	0.007	*1.1*	+0.2
1964	0.715	0.006	*0.8*	0.704	0.005	*0.7*	–1.4
2015	3.041	0.016	*0.5*	2.999	0.018	*0.6*	–1.7
2035	7.790	0.040	*0.5*	7.766	0.039	*0.5*	–0.4
2113	0.378	0.009	*2.4*	0.373	0.004	*1.0*	–0.5
2213	0.389	0.004	*1.0*	0.390	0.004	*1.1*	+0.2
2598	17.020	0.060	*0.4*	16.960	0.085	*0.5*	–0.6
3010	1.030	0.030	*2.9*	1.019	0.013	*1.3*	–0.3
3202	3.220	0.030	*0.9*	3.216	0.019	*0.6*	–0.1
3253	7.900	0.060	*0.8*	7.898	0.047	*0.6*	+0.0
3273	1.864	0.015	*0.8*	1.861	0.013	*0.7*	–0.2
3451	0.945	0.011	*1.2*	0.936	0.008	*0.9*	–0.7
3548	0.195	0.003	*1.5*	0.198	0.002	*1.2*	+0.8
Sum/Avg.	248.995	0.046	*0.9*	248.73	0.046	*0.7*	0.96

The ^{35}Cl(n,γ) reaction as secondary standard

In the same series of experiments, the capture reaction ^{35}Cl(n,γ) was also re-measured. For 35 intense gamma rays, spanning the energy range of 292–8579 keV, relative intensities could be determined with at least 2% accuracy, with 28 cases falling in the 0.5-1.5% accuracy range. According to a recent evaluation [22], the previously recommended data by Spits and Kopecky [2,10] do not agree with the latest high-quality measurements by Coceva *et al.* [23] which, in turn, agree well with earlier measurements by Krusche *et al.* [11]. The probable reason for the discrepancy is that in those works [11,23] only the nitrogen standard has been used, whereas in Ref. [10] various capture reactions have been utilised for calibration. Therefore, in Table 2 the present relative intensities were compared only with the new evaluation. The agreement is excellent again, and this is especially gratifying when the γ rays accurately measured before [23] are considered. These data and the precisely known capture cross-section and γ-ray energy values [11] render the ^{35}Cl(n,γ) reaction an excellent secondary standard for high-energy spectroscopy.

Table 2. New relative intensities for 35 strong gamma-rays from the $^{35}Cl(n,\gamma)$ reaction, compared to newly evaluated literature data [22]

E_γ (keV)	I_γ Wt. avg.	Unc.	% Unc.	I_γ Present	Unc.	% Unc.	Z-score/ Chi-sq.
292	0.967[a]	0.147	*15.2*	1.003	0.011	*1.1*	–0.2
436	3.860[a]	0.588	*15.2*	3.437	0.022	*0.6*	+0.7
517	89.338[b]	5.147	*5.8*	86.296	0.505	*0.6*	+0.6
632	1.173[a]	0.176	*15.0*	1.235	0.020	*1.6*	–0.3
786	38.676[b]	1.287	*3.3*	38.314	0.173	*0.5*	+0.3
788	60.000[b]	1.324	*2.2*	59.612	0.232	*0.4*	+0.3
937	2.165[a]	0.324	*14.9*	1.950	0.023	*1.2*	+0.7
1131	7.026[b]	0.206	*2.9*	7.054	0.047	*0.7*	–0.1
1165	100.000[c]	2.647	*2.6*	100.000	0.374	*0.4*	
1327	4.669[a]	0.699	*15.0*	4.567	0.037	*0.8*	+0.1
1601	12.809[b]	0.327	*2.6*	13.749	0.072	*0.5*	–2.8
1951	71.287[b]	2.096	*2.9*	71.844	0.369	*0.5*	–0.3
1959	46.176[b]	1.029	*2.2*	46.586	0.247	*0.5*	–0.4
2035	2.750[a]	0.276	*10.0*	2.651	0.042	*1.6*	+0.4
2676	5.779[b]	0.140	*2.4*	5.995	0.064	*1.1*	–1.4
2845	4.669[a]	0.478	*10.2*	3.931	0.045	*1.1*	+1.5
2864	21.213[b]	0.404	*1.9*	20.712	0.149	*0.7*	+1.2
3016	4.158[a]	0.213	*5.1*	3.663	0.049	*1.4*	+2.3
3062	12.945[b]	0.243	*1.9*	12.661	0.102	*0.8*	+1.1
3116	3.654[a]	0.202	*5.5*	3.323	0.045	*1.3*	+1.6
3429	3.290[a]	0.169	*5.1*	3.052	0.048	*1.6*	+1.4
3981	3.779[a]	0.202	*5.4*	3.718	0.054	*1.5*	+0.3
4083	2.886[a]	0.151	*5.2*	2.853	0.048	*1.7*	+0.2
4440	3.849[b]	0.085	*2.2*	4.126	0.073	*1.8*	–2.5
4980	13.294[b]	0.353	*2.7*	13.601	0.140	*1.0*	–0.8
5517	6.210[b]	0.154	*2.5*	6.278	0.076	*1.2*	–0.4
5715	19.522[b]	0.551	*2.8*	20.432	0.217	*1.1*	–1.5
5903	4.059[b]	0.114	*2.8*	4.167	0.082	*2.0*	–0.8
6111	75.662[b]	2.390	*3.2*	77.407	0.760	*1.0*	–0.7
6620	28.787[b]	0.588	*2.0*	28.349	0.344	*1.2*	+0.6
6628	17.243[b]	0.404	*2.3*	16.395	0.218	*1.3*	+1.8
6978	8.419[b]	0.235	*2.8*	8.196	0.122	*1.5*	+0.8
7414	38.676[b]	0.882	*2.3*	36.931	0.479	*1.3*	+1.7
7790	30.551[b]	0.699	*2.3*	29.705	0.423	*1.4*	+1.0
8579	10.070[b]	0.210	*2.1*	9.884	0.185	*1.9*	+0.7
Sum/Avg	312.938	0.436	*3.5*	309.38	0.183	*1.3*	1.35

[a] From Krusche *et al.* [11]
[b] From Coceva *et al.* [23]
[c] Normalising transition

The ^{66}Ga decay

Using the new ^{35}Cl(n,γ) data as calibration standards, new relative intensities have been determined for 21 gamma rays of ^{66}Ga, with an accuracy of 1% or better. For ^{66}Ga, the new Budapest data agree well with parallel data [26] from Lawrence Berkeley National Laboratory, obtained by calibrating their HPGe detector with absolute sources such as ^{228}Th (primary standard) and ^{238}Pu/^{13}C, providing the 6.13 MeV ^{16}O line, as well as with ^{56}Co. With this method of calibration, however, somewhat lower accuracy could be obtained.

Table 3. New relative intensities for 21 strong ^{66}Ga gamma-ray transitions, compared to the corresponding ENSDF data [25]

E_γ (keV)	I_γ ENSDF	Unc.	I_γ Present	Unc.	% Unc.	Z-score/ Chi-sq.	Contaminant
686	0.680	0.020	0.696	0.006	*0.9*	+0.8	
833	15.950	0.160	15.862	0.044	*0.3*	–0.5	
1039	100.000		100.000	0.166	*0.2*		
1332	3.260	0.030	3.171	0.009	*0.3*	–2.8	
1418	1.680	0.020	1.660	0.008	*0.5*	–0.9	
1507	1.520	0.025	1.498	0.007	*0.5*	–0.8	
1898	1.130	0.025	1.046	0.008	*0.8*	–3.2	
1918	5.650	0.020	5.364	0.026	*0.5*	–8.6	^{57}Ni
2189	15.120	0.150	14.405	0.054	*0.4*	–4.5	
2422	5.210	0.050	5.058	0.029	*0.6*	–2.6	
2751	63.100	0.500	61.264	0.260	*0.4*	–3.3	
3228	4.190	0.030	4.063	0.027	*0.7*	–3.1	
3380	4.040	0.030	3.960	0.027	*0.7*	–2.0	
3422	2.360	0.040	2.325	0.018	*0.8*	–0.8	
3432	0.805	0.011	0.784	0.009	*1.1*	–1.5	
3766	0.410	0.011	0.423	0.007	*1.7*	+1.0	
3790	3.010	0.030	3.028	0.024	*0.8*	+0.5	
4086	3.520	0.050	3.454	0.026	*0.8*	–1.2	
4295	10.950	0.120	10.598	0.069	*0.6*	–2.5	4608 SE
4461	2.278	0.024	2.280	0.020	*0.9*	+0.1	
4806	4.920	0.050	5.030	0.038	*0.8*	+1.8	
Sum/Avg	249.783	0.066	245.970	0.042	*0.7*	8.08	

Since the 1991 recommendation new data with high statistical precision have become available [24]. Unfortunately, the efficiency calibration has been tied up with the erroneous data of Refs. [3,4]. Hence the intensities had to be corrected above 2 MeV by the ENSDF evaluator [25] according to the recipe of McCallum and Coote [5], *i.e.* multiplied by the function

$$F(E_\gamma) = 1.053 - 0.079E_\gamma + 0.026(E_\gamma)^2$$

to account for the deviation of efficiency from a linear function. This function has been determined from a comparison of a linear extrapolation with the actual efficiency measured with proton resonances [5]. It is these corrected values [25] which have been compared with the new Budapest data in Table 3. In view of the fact that the ENSDF uncertainties do not include the contribution of the correction (estimated to at least 5% [5]), the agreement is

reasonable. Hence the negative bias, amounting to 30% at the highest energy, is verified for the old data [3,4], as well as for the more recent data [24] calibrated with them.

Summary

The equivalence of efficiency determination procedures based on neutron and proton capture lines has been verified and the deviation of high-energy efficiency from linearity confirmed. The new, accurate relative intensities for ^{56}Co and ^{66}Ga extend the range of secondary radioactive standards up to 4.8 MeV. Extreme care has to be taken with any high-energy intensity value obtained in the past with the help of ^{56}Co or ^{66}Ga calibration sources, and corrections have to be made using the present data of high accuracy. Relative intensities have also been improved for the ^{35}Cl(n,γ) reaction, a useful secondary standard in a wide energy range, between 0.3–8.5 MeV. The new data are supported by other most recent measurements of a slightly lower precision [26,27].

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Selection and evaluation of gamma decay standards for detector calibration using coincidence method

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Abstract

Coincidence method for calibration of gamma detectors using suitable calibration standards with two cascading gamma rays is analyzed. From the list of recommended gamma ray standards currently under reevaluation by the CRP, 14 radionuclides were selected as the potential source candidates for the coincidence method. The following sources were selected ^{24}Na, ^{46}Sc, ^{60}Co, ^{66}Ga, ^{75}Se, ^{88}Y, Nb^{94}, ^{111}In, ^{123m}Te, ^{133}Ba, ^{134}Cs, ^{152}Eu, ^{154}Eu and ^{207}Bi. Reaction ^{11}B (p,γ) ^{12}C* was also selected as a source of high energy gamma rays. Experimental data on angular correlation coefficients for selected sources were collected from the literature and evaluated according to the recommended procedure. Theoretical angular correlation coefficients were calculated and compared to the evaluated data.

1. Introduction

Coincidence method is being 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. Second very important and widely accepted application of coincidence method is determination of the absolute activity of standards for detector calibration. However, coincidence method is more general and allows to determine the absolute detector efficiency too.

At present the absolute calibration of photon detectors proceeds in two steps. In the first step determination of the absolute source intensity is performed usually by beta-gamma coincidence method. Result of this step is an absolutely calibrated standard, which is used in the second step for determination of 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, reducing thus the uncertainty of the calibration. This possibility may be especially useful for several high energy photon sources (e.g. ^{24}Na, ^{11}B + p → ^{12}C*), which are difficult to calibrate absolutely.

2. Principle of the gamma-gamma coincidence method

The coincidence method is rather simple and can be used if the source nucleus decays by two cascading photons $\gamma_1$and γ_2. Simplified decay scheme of a nucleus with quantum numbers of decaying levels and gamma transitions is given in fig. 1. A general case with beta decay branches populating all levels of daughter nucleus is shown, where ei denotes feeding of level i in the daughter nucleus, b_i is a branching ratio of gamma ray γ_i, E_i is the level energy of the daughter nucleus, J^{π}_i, m_i are spin and magnetic substate of level i and $\lambda_i\nu_i$ is the multipolarity of γ_i radiation. Ideal source for coincidence method is a source with beta decay branch ε_2=1, populating only the second excited level with energy E_2 and both gamma ray branching ratios $b_{1,2}$=1.

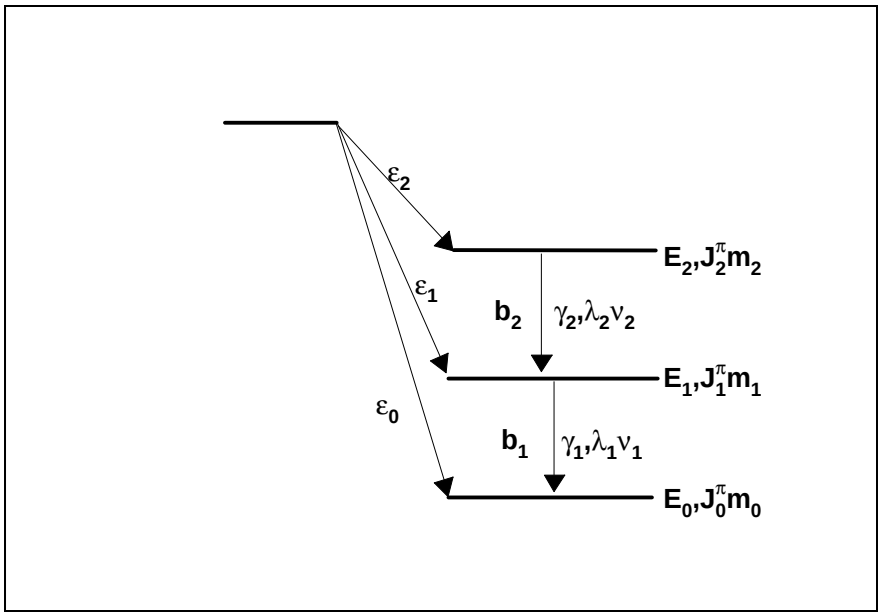

Fig. 1. Cascade of two gamma rays with mutlipolarities of both γ rays and characteristics of all levels involved. Symbols are explained in the text.

Numbers of photons γ_1 detected in detector d_1 (N_1), number of photons γ_2 registered in d_2 (N_2) and number of events (N_{12}) where both photons are registered in the respective detector can be written as follows

$$N_2 = A\omega_2 \frac{\varepsilon_2 b_2}{1+\alpha_2}$$

$$N_{12} = A\omega_1\omega_2 \frac{\varepsilon_2 b_1 b_2}{(1+\alpha_1)(1+\alpha_2)} W(\vartheta) F(\omega_1\omega_2\vartheta)$$

$$N_1 = A\,\omega_1 \frac{(\varepsilon_2 b_2 + \varepsilon_1) b_1}{1+\alpha_1}$$

where ω_1 and ω_2 are full energy peak efficiences, $W(\theta)$ is the angular correlation function and $F(\omega_1\omega_2\theta)$ is a correction for finite solid angle of both detectors.

Sources suitable for detector calibration using coincidence method were selected from the set of sources, recommended as calibration sources in the present CRP. The ideal source should have only two cascading γ-rays with β-decay feeding only the second excited level of the daughter nucleus. Therefore the main selection criteria were following:

- Two cascading γ rays $E_{\gamma 2}$ and $E_{\gamma 1}$ are emitted in the decay of parent nucleus
- β-decay branching to the second level E_2 in daughter nucleus ε_2 should be close to 100%
- β-decay branching to the first level E_1 in the in the daughter nucleus ε_1 should be minimal
- both γ-ray branching ratios B_1 and B_2 should be close to 100 %.

Sources selected from the whole set are given in the Tab. 1, all relevant data were taken from ref. 1. Together 14 radioactive sources covering energy region from 81 keV to 2754 keV were selected. In order to increase further the highest energy, we selected also a reaction $^{11}B(p,\gamma)^{12*}C$, which has a resonance at incident proton energy of 153 keV The excited final nucleus ^{12}C deexcite by emission of two high enegy γ rays with energies of 11670 keV and 4430 keV.

Parent	ε_2(%)	E_2(keV)	$E_{\gamma 2}$(keV)	B_2(%)	ε_1(%)	E_1(keV)	$E_{\gamma 1}$(keV)	B_1(%)
^{24}Na	99,94	4.122,90	2.754,03	99,94	0,00	1.368,70	1.368,63	100,00
^{46}Sc	100,00	2.009,80	1.120,55	99,99	0,00	889,30	889,28	100,00
^{60}Co	99,93	2.505,80	1.173,24	100,00	0,06	1.332,50	1.332,50	100,00
^{66}Ga	27,70	3.791,20	2.751,85	84,44	0,00	1.039,39	1.039,30	100,00
^{75}Se	95,80	400,70	136,00	64,62	<0,9	264,70	264,66	98,10
^{88}Y	94,90	2.734,10	898,04	100,00	5,50	1.836,10	1.836,06	100,00
^{94}Nb	98,10	1.573,70	702,62	100,00	0,00	871,10	871,09	100,00
^{111}In	90,00	416,70	171,28	100,00	0,00	245,40	245,40	100,00
^{123m}Te	--	247,60	88,46	100,00	--	159,10	158,97	100,00
^{133}Ba	86,00	437,00	356,02	86,93	<3,0	81,00	81,00	100,00
^{134}Cs	70,11	1.400,60	795,86	100,00	0,01	604,72	604,70	100,00
^{152}Eu	13,80	1.123,20	778,90	100,00	8,20	344,30	344,28	100,00
^{154}Eu	36,30	1.397,50	1.274,44	96,71	10,00	123,10	123,07	100,00
^{207}Bi	84,18	1.633,40	1.063,66	100,00	8,79	569,70	569,70	100,00
^{12*}C	100,00	16.105,80	11.670,00	92,00	0,00	4.438,90	4.430,00	100,00

Number of coincidenced events depends on angular correlation function W(θ), where θ is the angle between two γ-ray detectors. Angular correlation function depends on quantum characteristics of all involved levels i.e. on quantum numbers J^{π}_2, J^{π}_1 , J^{π}_0 and multipolarities $\lambda_i\nu_I$ of both γ-rays. Angular correlation function W(θ) can be calculated theoretically according to the following formulas (ref. 2)

$$W(\theta)=\Sigma A_{kk}\ P_k(\cos\theta)$$

$$A(J_2,\lambda_2,J_1,\lambda_1,J_0)=F_k(J_2,\lambda_2,J_1)\ F_k(J_0\lambda_1 J_1)$$

$$F_k(J_f,\lambda,J_i)=(-1)^{1-J_i-J_f}\ (2J_i+1)^{1/2}\langle\lambda 1\lambda -1|k0\rangle W(J_iJ_i\lambda\lambda;kJ_f),$$

where $P_k(\cos\theta)$ are Legendre polynomials, $\langle\lambda 1\lambda -1|k0\rangle$ are Clebsh-Gordon coefficients and $W(J_iJ_i\lambda\lambda;kJ_f)$ are Racah coefficients. The last formula holds for pure electromagnetic

transitions, which can be generalized in the case of mixed multipole transitions

$$A_{kk}(J_i\lambda\lambda'J_f)=1/(1+\delta^2)[F_k(J_f\lambda\lambda J_i)+2\delta F_k(J_f\lambda\lambda'J_i)+\delta^2 F_k(J_f\lambda'\lambda'J_i)]$$

$$F_k(J_f\lambda\lambda'J_i)=(-1)^{1-J_i-J_f}\ [(2J_i+1)(2\lambda+1)(2\lambda'+1)]^{1/2}\langle\lambda 1\lambda'-1|k0\rangle W(J_iJ_i\lambda\lambda';kJ_f).$$

The angular correlation coefficients for all radionuclei given in Tab.1. were retrieved from literature and evaluated using statistical code lweigh. In two instances (^{24}Na, ^{46}Sc) no experimental data were found in the literature, for two another nuclei (^{111}In, ^{123m}Te) only single data on angular correlations were found. For all other nuclei several sources describing angular correlation measurements were found.

Data necessary for calculation of angular correlations coefficients for all nuclei are given in Tab. 2. In the last four columns of Tab.2 the angular correlation coefficients are given. Coefficients A_{22}^{theory} , A_{44}^{theory} are calculated according to formulas given above, using CERNLIB library (ref. 3) for Clebsh-Gordon and Racah coefficients. Coefficients A_{22}^{WM}, A_{22}^{WM} are weighted means calculated with the code lweight (ref. 4). The agreement between experimental and theoretical values is reasonable in all instances except for ^{154}Eu, where theoretical coefficients predict higher anisotropy.

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Parent	Daughter	J^{π}_2	$\lambda_2\nu_2$	δ_2	J^{π}_1	$\lambda_1\nu_1$	δ_1	J^{π}_0	A_{22}^{theory}	A_{44}^{theory}	A_{22}^{WM}	A_{44}^{WM}
^{24}Na	^{24}Mg	4^+	?	-	2^+	E2	-	0^+	0.1020	0.0091	-	-
^{46}Sc	^{46}Ti	4^+	E2	-	2^+	E2	-	0^+	0.1020	0.0091	-	-
^{60}Co	^{60}Ni	4^+	E2(+M3)	-0.0025	2^+	E2	-	0^+	0.1020	0.0091	0.1012(22)	0.0658(24)
^{66}Ga	^{66}Zn	1^+	M1+E2	?	2^+	E2	-	0^+	-0.2455	0.0000	-	-
^{75}Se	^{75}As	$5/2^+$	E1	-	$3/2^-$	M1+E2	-0.044(6)	$3/2^-$	-0.0331	0.0000	-0.028(4)	0.0015(18)
^{88}Y~	^{88}Sr	3^-	E1	-	2^+	E2	-	0^+	-0.0714	0.0000	-0.0692(32)	0.0009(19)
^{94}Nb	^{94}Mo	4^+	E2	-	2^+	E2	-	0^+	0.1020	0.0091	0.0968(34)	0.0141(38)
^{111}In	^{111}Cd	$7/2^+$	M1+E2	-0.144(3)	$5/2^+$	E2	-	$1/2^+$	0.0312	-0.0014	-	-
^{123m}Te	^{123}Te	$11/2^-$	M4	-	$3/2^+$	M1+E2	0.062(6)	$7/2^-$	-0.1203	0.0000	-	-
^{133}Ba	^{133}Cs	$1/2^+$	E2	-	$5/2^+$	M1+E2	-0.151(2)	$7/2^+$	0.0359	-0.0016	0.0369(17)	0.0036(11)
^{134}Cs	^{134}Ba	4^+	E2	-	2^+	E2	-	0^+	0.1020	0.0091	0.0993(90)	0.0050(17)
^{152}Eu	^{152}Gd	3^-	E1(+M2)	0.002	2^+	E2	-	0^+	-0.0730	0.0000	-0.0730(19)	0.002(7)
^{154}Eu	^{154}Gd	2^-	E1+M2	0.032(15)	2^+	E2	-	0^+	0.2731	0.0003	0.123(13)	0.005(2)
^{207}Bi	^{207}Pb	$13/2^+$	M4	-	$5/2^-$	E2	-	$1/2^-$	0.2208	-0.0180	0.224(29)	-0.023(17)
^{11}B	^{12*}C	2^+	M1	-	2^+	E2	-	0^+				

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