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INTERNATIONAL DECAY DATA EVALUATION PROJECT

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Abstract

Basic concepts of, and information from, radionuclide decay are used in many applications. Many of these applications require a knowledge of half-lives and radiation energies and emission probabilities. For over 50 years, people have compiled and evaluated measured data with the goal of obtaining the best values of these quantities. This has resulted in numerous sets of recommended values, many of which still have scientific, historical, or national reasons for existing. These sets show varying degrees of agreement and disagreement in the quoted values and varying time lags in incorporating new and improved experimental results. A new informal international group has been formed to carry out evaluations for radionuclides of importance in applications; it is expected that the results will become an authoritative and widely accepted set of decay data.

I. Introduction

There are many fields that apply radionuclide decay data and the categories of data that are needed differ among these fields. I will limit this discussion to the data needed for applied γ-ray spectrometry; this includes applications such as nuclide identification and quantitative assay.

For almost any radionuclide one will find the related data in several compilations and the values of the quantities of interest will generally differ. Often these differences are insignificant for applied spectrometry and they can be ignored, but occasionally even small differences are important. Even if the differences are insignificant, they are a nuisance because one has to make a choice among them even though one may not have any basis for determining which is the best.

There are several reasons for these differences between evaluations, including

- different measurement results used as input data
  - done at various times so newer references available to later evaluator
  - some references were not available to an evaluator
  - different judgements as to which references to use
- different analytical techniques for combining data
  - select only single "best" set of values
  - average all values - weighted or unweighted
  - combine discrepant values in different ways
- different supporting data
  - selection of internal-conversion coefficient tables

2. What quantities

The quantities that are useful for applications involving γ-ray spectrometry can be divided into two groups, with the first including those quantities that are always needed, namely, the

- half-life,
- γ-ray energies (E_γ), and
- γ-ray intensities or emission probabilities (P_γ, in γ's per 100 decays or per decay).

This set of information is sufficient for most current γ-ray spectral-analysis codes. However, future analysis programs should be able to make the corrections for coincidence summing between sequentially emitted γ rays. This often becomes important for Ge semiconductor detector measurements on small samples placed close to the detector. For this correction, one also needs a second set of quantities, namely,

- for each nuclear level in the daughter nucleus
  - its energy
  - its α, β, or electron-capture feeding probability
- for each γ ray
  - its placement in the level scheme
  - the K-shell internal-conversion coefficient (α_K), and
  - the total internal-conversion coefficient (α).

In addition, it is suggested that any evaluation should include a set of comments stating which sets of experimental data were used and what decisions were made, so that other evaluators can determine the quality of the evaluation and accept it as is, or build on it without redoing the evaluation from the beginning.

3. Overview of files

3.1 File characteristics

The characteristics of several collections of decay data from the last 20 years [1-21] are summarized in Tables 1 and 2 including the approximate number of radionuclides, and of the above list of quantities those that are in the set of data. This is not a comprehensive list of the available files of decay data; especially, since it includes only one non-English file. One might reasonably assume that the older data sets have been completely replaced by the newer ones; however, this has not happened. My conversations have indicated that even the 1979 set of decay data of Erdtmann and Soyka [3] and the 1977 set of Kocher [1] are still used.

Comments are given here for the evaluations and compilations listed in Table 1; they are for simplicity, ordered by the country of origin.

International

Evaluated Nuclear Structure Data File (ENSDF), a computer file and the associated publication Nuclear Data Sheets (NDS) [19]; continuously up-dated on a cycle of ~ 6 years and has about 20,000 pages; it includes reaction data

International Atomic Energy Agency (IAEA) Coordinated Research Programmes (CRP); the first CRP was on transactinium nuclides published in 1986 [8] for 23 nuclides, the second CRP was on radionuclides used in the calibration of γ-ray detectors [11] included 36 radionuclides. A unique aspect of these projects was that they included programs of precise measurements in addition to the evaluations and thereby produced a significant improvement in the quality of the data available.

United States of America

Table of Isotopes [2, 20, 24-29], first edition published in 1940 [24], through the 7th edition in 1978 [2], and the new 8th edition [20]; includes reaction data.

Table of Radioactive Isotopes [7]; a decay-data file from Table of Isotopes group

Evaluated Nuclear Data File /B (ENDF/B) [12]; this file contains cross sections and other information for use in nuclear reactor technology.

Others include "Radioactive Atoms" [22], "Nuclear Decay Data for Selected Radionuclides" [23], "Nuclear Decay Data for Radionuclides Occurring in Routine Releases from Nuclear Fuel Cycle Facilities" [1]; and "Table of Nuclear-Decay Data" [6]. Others: [9] and [30].

United Kingdom

file related to the nuclear reactor industry; part for heavy elements [4] and activation products [15].

Germany

a list [13] prepared at the national standards laboratory Physikalisch-Technische Bundesanstalt (PTB).

"The Gamma Rays of the Radionuclides, Tables for Applied Gamma Ray Spectrometry" [3]

"Catalog of Gamma Rays from Radioactive Decay" [5].

France

Table de Radionucléides [18] from Laboratoire Primaire des Rayonnements Ionisants (LPRI)

Sweden

a conveniently formatted set of decay data from ENSDF [14]

a fission-product data file [17].

A significant limitation of most of these sets of data is that, as shown in Table 2, there are no comments indicating the origin of the data for a particular radionuclide and especially what processing was done by the authors. This limitation is understandable since such documentation would require a great deal of effort and would take up a great deal of space. However, this makes it impossible for others to judge the quality of the evaluations and for subsequent evaluators to make good use of the results.

It is also clear that if one is interested in, for example the placement of γ rays, most of these sets of decay data do not meet the need.

3.2 Relationships among files

Some of the sets of decay data in Table 1 involve major efforts in evaluation of the original measurements, others start with an existing evaluated decay-data set and add new measured results, and others involve only the selection of information from other decay-data files. It is reasonable to expect that each evaluator has made use of many of the previously published decay-data sets, so there are complex interrelations among the data sets. Since the ENSDF file has a long history and is widely available, it has often been used as a starting point for other evaluations.

My understanding of the relationships of some of these files is as follows.

ENSDF based files:

Nuclear Data Sheets (NDS) [19], each NDS publication is from a recent, complete update of that portion ENSDF. Between NDS publications for an A-chain, the contents of ENSDF may be partially updated, so ENSDF may be more current than the NDS.

NCRP-58 [6], from the ENSDF file with other information added

Table of Radioactive Isotopes [7] from ENSDF file with other quantities added and some different data handling methods

Table of Isotopes, 8th edition [20] based on the ENSDF file as it existed in about 1994.

ENDF/B Decay-Data File [12] from ENSDF with editing and additional quantities computed.

Ekström [14] is a direct extraction from ENSDF.

Mixed origin:

Physikalisch-Technische Bundesanstalt (PTB) [16]
Debertin and Helmer [10]

Independent evaluations:

Table of Isotopes, 7th edition
Table de Radionucléides [18]
AEA-1 and AEA-2 [4, 15]
Fission Products [17]
Erdtmann and Soyka [3]
Reus and Westmeier [3, 5]
IAEA CRP's [8, 11]
Chechev and Chukreev [13]

4. Comparison of Files

4.1 Agreement among data

For a large number of radionuclides the values in the different files are be quite similar; this is illustrated in Table 3 where the quoted half-lives, and γ-ray energies and emission probabilities are listed for the well-studied decay scheme of ^{137}Cs. All of the half-lives and all of the P_γ values after 1980 agree very well. Since 1986, the uncertainties quoted for the P_γ values are either 0.07 or 0.2, but these uncertainties are small enough that the difference is not significant for the applied user, even for the precise detector efficiency calibrations. However, the fact that only these two values occur suggests that the evaluators have used different methods of analysis, and this fact is of interest.

4.2 Differences Among Data

Differences and Time Lags

There are cases, some well known, where there have been significant problems; two examples are given in Tables 4 and 5.

The case of ^{234}Pa (1.1 min) has become quite well known for many years. This case is especially interesting because the γ ray from this nuclide is often used to determine the amount of ^{238}U present; often a very important question. The P_γ(1001) values in the evaluations before 1993 were all based on a single 1963 measurement. Unfortunately, the 1971 measured value was published in a laboratory report, and it was not used by most evaluators. Researchers who were involved in the assay of ^{238}U by means of γ-ray spectrometry became aware of significant discrepancies between these results and those from other measurement methods and this provided the impetus for the newer measurements given in Table 4 which give a change in this P_γ of 30%. The earliest of these newer measurements was published in 1986 and it took a long time for it to get into the widely circulated evaluations.

The case of the P_γ ratio for ^{144}Ce is interesting because it is a problem that was identified by a user of the decay data. The assay of ^{144}Ce is usually based on the 133-keV γ ray because it is the most intense line; the 80-keV line can then be used to verify the radionuclide assignment. But, if the $P_\gamma(80)/P_\gamma(133)$ ratio used in the assay is too small, the analysis procedure may assign the residual area of the 80-keV peak to another radionuclide with a γ ray at this energy (e.g., ^{133}Xe). The evaluations in Table 5 show a wide range of ratios, from 0.103(10) to 0.137(4) for the last five values. It is clear that the uniquely low value of ref. [46] was adopted in four of the evaluations and the next lowest value of ref. [48] was adopted by, or strongly influenced, several others. In the course of investigating this problem, the author found that two metrology laboratories had unpublished measurements on this nuclide; these values are given as the 1992 entries. The best value of this ratio is clearly about 0.139, and not the 0.103 - 0.123 previously quoted; of the compilations of the last ten years only the PTB list has a value this large.

Differences in Evaluation Methods

In spite of 50 years of experience in compiling and evaluating decay data, there are areas in which the methodology is not agreed upon. Over the past few years there have been discussions within the Non-Neutron Nuclear Data Working Group of the ICRM on evaluation methods. And, various authors such as Zijp [50], Gray et al. [51], Woods [52], James et al. [53], and Rajput [54] have described alternative methods to deal with discrepant sets of data. Results from these methods are compared in ref. [54].

The data in Table 6 show a comparison of the half-lives for three radionuclides from five different methods of "averaging" the measured values. The first two cases, ^{90}Sr and ^{137}Cs, illustrate the variation when the measured values are not consistent, while the last case, ^{154}Eu, illustrates the results when the measured values are consistent. The first two methods make use of the original uncertainties assigned to the measured values, while the last three methods adjust these uncertainties, if it is found that the measured values are not consistent.

The question is how to treat the discrepancies between the measured values. It would, of course, be desirable to be able to find errors or limitations in the original measurements so that one could knowingly modify the original uncertainties or even reject some of the measurements; however, most evaluators do not have sufficient knowledge about any particular set of measurements to do this. Therefore, one is left to use some ad hoc data selection or some analytical method to adjust the weights of the individual measurements and/or the uncertainty in the evaluated value. The last three methods adjust the weights in different ways.

The values of the half-lives deduced by the various methods are in good agreement; it is the uncertainties that are different. The Limitation of Relative Statistical Weight (LRSW) method [50] and the Rajeval method [54] give uncertainties that differ by factors of about 6 and 10 for the first two nuclides. This is a serious difference. For ^{90}Sr, the external uncertainty of the weighted mean is 0.14, which agrees fairly well with the large uncertainty from the LRSW method, while for ^{137}Cs, the external uncertainty for the weighted mean of 0.03 agrees with the small value from the Rajeval method. Whether the external uncertainty of the weighted mean agrees with the LRSW or Rajeval method is related to the relationship of the most precise measured value with the weighted mean.

5. International Decay Data Evaluation Project

In the above discussion, the problems illustrated are:

- many sets of decay data to choose from,
- long time lag to get new measurements into files, and
- little communication with users who have identified problems.

Over the past two years, a new international collaboration has been formed to address these problems; it is called the Decay Data Evaluation Project (DDEP). This group is based on an informal agreement and consists of M.-M. Bé, Laboratoire Primaire des Rayonnments Ionisants (LPRI) in France, E. Schönfeld, Physikalisch-Technische Bundesanstalt (PTB) in Germany, T. D. MacMahon, Centre for Analytical Research in the Environment (CARE), Imperial College in the United Kingdom; and in the United States E. Browne, Lawrence Berkeley National Laboratory (LBNL); J. K. Tuli, Brookhaven National Laboratory (BNL); and this author, who is the coordinator of the DDEP. The latter three are members of the group that provides evaluations for the ENSDF system. LPRI and PTB also have established a formal agreement to cooperatively evaluate and publish decay data. Their new publication, Table of Radionuclides, will be an extension of the existing LPRI Table de Radionucléides.

One strength of the DDEP is that it brings together the expertise of several laboratories that are experienced in the precise measurement of the quantities involved in radioactive decay. This is illustrated by the fact that LPRI and PTB are the radionuclide standards laboratories of their respective countries and they have extensive experience in the precise measurement of half-lives and γ-ray emission probabilities.

The initial effort of the this group was to establish a list of ~250 radionuclides that are of importance in the various applications and should be evaluated. Then this group discussed, and agreed on, the methodology to be used in these evaluations. This include:

Account for (i.e., use or explicitly exclude) all measurements of a quantity.

Generally use the Limitation of Relative Statistical Weight Method of computing the average of a set of values. This provides a procedure for treating a discrepant set of data.

Use the Rösel et al. internal-conversion coefficients [55] if theoretical values are used.

Use the Schönfeld data [56] for the electron-capture probabilities for various atomic shells and the Schönfeld and Janssen evaluation of the fluorescence yields [57].

Provide written documentation of all data used and all decisions and calculations.

All evaluations will be approved by all evaluators before they are final.

An example of the level of thoroughness that we are trying to establish is that in the evaluation of

the decay of ^{75}Se, the evaluator considered relative γ-ray intensity data from 25 measurements.

The evaluations that have been prepared and are in various stages of the review process include: ^{24}Na, ^{65}Zn, ^{68}Ge, ^{68}Ga, ^{75}Se, ^{95}Zr, ^{95}Nb, ^{109}Cd, ^{113}Sn, ^{139}Ce, ^{140}Ba, ^{140}La, ^{141}Ce, ^{143}Pr, ^{153}Sm, ^{153}Gd, ^{188}Re, and ^{194}Ir. Other evaluations that are have been prepared in a draft form include: ^{7}Be, ^{22}Na, ^{40}K, ^{41}Ar, ^{46}Sc, ^{51}Cr, ^{54}Mn, ^{60}Co, ^{137}Cs, and ^{137m}Ba.

The preparation of high-quality evaluations does not, in itself, address any of the three problems listed above. In the future, a goal of this project will be to address the first problem by advertizing the project and convincing other people who are preparing sets of decay data to use these results. If this is successful, this should reduce the number of different values that are found in various sets of decay data. The fact that this is possible is illustrated by the γ-ray energies in Table 3. Since the 1979 publication of ref. [58], most evaluators have used the values from this paper and the consistency shown in the table has resulted.

In the past and at present, most of the decay-data files present the data on a printed page which prevents up-dating values as new data becomes available. The ENSDF data are in a computer file from which the data for any desired decay scheme can be extracted at any time. Therefore, revised evaluations could be made available in a short time. In the future, other files should become available over Internet and the World Wide Web. Although these new methods of data delivery will not necessarily solve this problem, it is hoped that the electronic decay-data files will develop plans to, and methods of, up-dating the data in a more timely manner. For example, in the ENSDF system the decay data may be up-dated without the necessity of up-dating a whole A-chain. If this is implemented, it may be possible to avoid the long delays that have occurred in the past in up-dating individual decay schemes.

The last problem listed above involves feedback from the users of the decay data when they identify potential problems. This will be considered at a later time, but with the Internet and WWW communications system, it may now be practical to solicit responses from users.

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Table 1. Decay Data Files from 1977 to 1996

Publication Year	Data File	Reference	Label	From	Number of Nuclides
1977	Kocher	1	Koc	USA-ORNL	240
1978	Table of Isotopes	2	T of I-78	USA-LBL	All
1979	Erdtmann & Soyka	3	E & S	Germany	All
1981	Nichols & James	4	AEA-1	United Kingdom	125
1983	Reus & Westmeier	5	R & W	Germany	All
1985	NCRP-58	6	NCRP	USA	200
1986	Table of Radioactive Isot.	7	TRI	USA-LBL	All
1986	IAEA-CRP	8	IAEA	International	23
1988	Identification Catalog	9	IC	USA-ORAU	82
1988	Debertin & Helmer	10	D&H	Germany & USA-INEL	77
1990	ENDF/B-V1	12	ENDF	USA-INEL	776
1991	IAEA-CRP	11	IAEA	International	36
1991	Chechev & Chukreev	13	C&C	USSR	34
1992	Ekström	14	EK	Sweden	All
1993	Nichols	15	AEA-2	United Kingdom	230
1993	PTB	16	PTB	Germany	200
1993	Fission Products	17	FP	Sweden	400
1983-1988[a]	Table de Radionucléides	18	T de R LARA	France	150/200
1983-1996[b]	ENSDF	19	NDS	International	All
1996	Table of Isotopes	20	T of I-96	USA-LBL	All
1997 - [c]	Table of Radionuclides	21	T of R	France & Germany	??

[a] Evaluation effort was continuous, but each radionuclide was only done once.

[b] Evaluations and publication is continuous. This range of years includes all of the evaluations that have not been replaced in later issues.

[c] Continuation of the evaluations done in ref. 18, but will be in a revised form.

Table 2. Quantities in Decay Data Files

File	Year	Half-life	E_γ, P_γ	All γ's	P_α, P_β	γ place-ment	α^a	Comments
Koc	1977	Yes	Yes	No	Yes	No	No	None
T of I-78	1978	Yes	Yes	Yes	Yes	Yes	No	References
E&S	1979	Yes	Yes	Yes	No	No	No	None
AEA-1	1981	Yes	Yes	Yes	Yes	No	Yes	References
R&W	1983	Yes	Yes	Yes	No	No	No	None
NCRP	1985	Yes	Yes	No	Yes	No	Few	None
TRI	1986	Yes	Yes	Yes	No[b]	No	No	None
IAEA	1986, 1991	Yes	Yes	No	Yes	No	Some	Some
ENDF/B-VI	1990	Yes	Yes	Yes	Yes	Yes	Yes	Detailed
C&C	1991	Yes	Yes	Yes	Yes	No	No	None
AEA-2	1993	Yes	Yes	No	No	No	No	None
PTB	1993	Yes	Yes	No	No	No	No	Brief
FP	1993	Yes	Yes	No	No	No	No	Some
T de R	1983-88	Yes	Yes	Yes	Yes	Yes	Yes	References
ENSDF & NDS	1983-96	Yes	Yes	Yes	Yes	Yes	Yes	Detailed
T of I-96	1996	Yes	Yes	Yes	Yes	Yes	No	None

[a] Internal-conversion coefficients

[b] The total β^- and β^+ spectra are given in binned form, but the decays to individual final levels are not listed separately.

Table 3. Comparison of Values in Evaluations for ^{137}Cs

Evaluation (year)	Half-life (years)	E_γ (keV)	P_γ (%)
Koc (1977)	30.17(3)	661.645(9)	85.1(3)
E & S (1979)	30.1	661.62	84.62
R & W (1983)	30.14	661.6(1)	85.1
T de R (1984)	30.15(6)	661.660(3)	85.2(2)
NCRP (1985)	30.0(2)	661.660(3)	85.1(3)
TRI (1986)	30.0(2)	661.660(3)	85.21(7)
D&H (1988)	30.25(11)	661.660(3)	85.20(20)
NDS-90 (1990)	30.1(2)	661.660(3)	85.21(7)
IAEA (1991)	30.18(15)	661.660(3)	85.1(2)
C&C (1991)	30.21(11)	661.660(2)	85.22(7)
AEA-2 (1993)	30.17	661.66	
PTB (1993)	30.17(16)	661.66	85.1(2)
NDS-94 (1994)	30.07(3)	661.660(3)	85.1(2)
T of I-96	30.07(3)	661.660(3)	85.1(2)

Table 4. Comparison of Values for ^{234}Pa (1.1 min) in Evaluations and Measured P_γ Values

Evaluation (year)	E_γ (keV)	P_γ (%)
Koc (1977)	1001.025(22)	0.589
T of I-78 (1978)	1001.2(2)	0.59
E & S (1979)	1001.03	0.59
AEA-1 (1981)	1001.00(2)	0.59(10)
R & W (1983)	1001.0(1)	0.59
NDS-83 (1983)	1001.03(3)	0.59(8)
TRI (1986)	1001.00(3)	0.65(9)
LARA (1987)	1001.03	0.59
PTB (1993)	1001.03	0.839(12)
NDS-94 (1994)	1001.03(3)	0.837(10)
T of I-96 (1996)	1001.03(3)	0.837(10)

Measurements after 1965

Year	P_γ(%)	Reference
1971	0.83	[35]
1986	0.834(7)	[36]
1990	0.839(5)	[37]
1992	0.79(4)	[38]
1992	0.845(21)	[39]
1992	0.82(3)	[40]

Table 5. Comparison of Values for ^{144}Ce in Evaluations and Measured P_γ Ratio

Evaluation (year)	$P_\gamma(80)/P_\gamma(133)$[a]
Koc (1977)	0.152(16)
NDS-79 (1979)	0.102(10)
E & S (1979)	0.148
R & W (1983)	0.102
T de R (1983)	0.102
NCRP (1985)	0.135(7)
TRI (1986)	0.103(10)
NDS-89 (1989)	0.123(5)
C&C (1991)	0.121(7)
PTB (1993)	0.137(4)
T of I-96 (1996)	0.123(5)

Measured values

Year	$P_\gamma(80)/P_\gamma(133)$[a]	Reference
1969	0.22(2)	[41]
1970	0.148(12)	[42]
1970	0.143(14)	[43]
1970	0.16(1)	[44]
1976	0.150(4)	[45]
1976	0.102(10)	[46]
1977	0.134(8)	[47]
1984	0.123(5)	[48]
1992	0.140(4)	priv. comm.
1992	0.1379(7)[b]	priv. comm.

[a] For the evaluations and some measurements, the author computed the ratio from the individual P_γ values.
[b] This is not the entire uncertainty.

Table 6. Half-life Values from Difference Averaging Methods
All values are in years and are from MacMahon [49].

Method	^{90}Sr	^{137}Cs	^{154}Eu
Weighted mean[a]	28.56(2)	30.10(1)	8.593(4)
Bayesian[b] [51]	28.56(14)	30.10(3)	8.593(1)
LRSW [50]	28.60(17)	29.93(21)	8.593(4)
Normalized residual [53]	28.83(5)	30.06(3)	8.593(4)
Rajeval [54]	28.80(3)	30.10(2)	8.593(4)
Number of measurements	9	18	5

[a] Internal uncertainty is given.
[b] This is equivalent to the weighted mean with the external uncertainty.