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Recommended standards for gamma ray intensities

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Abstract

Gamma ray data are used in more and more areas of application, and so over the years the demand for recommended gamma ray energies and intensities has increased. This paper proposes a list of gamma rays whose intensity is sufficiently well-known and they can be used for the calibration of gamma ray spectrometers and other applications; it is based on studies carried out by an international group of evaluators: the Decay Data Evaluation Project. One goal of this paper is to gather this set of data together in order to facilitate and generalize their use. In the first part, a brief description of the methodology followed throughout the evaluations is given, different methods of gamma ray intensity evaluation are presented, some typical examples of evaluations are shown; in the second part, the list of chosen nuclides is given along with their applications, and finally a list of recommended gamma ray intensities is presented.

Keywords

Applied radionuclides, Decay data, Gamma ray intensities, Evaluation techniques

1. Introduction

Over the years, there has been an increasing demand for recommended data such as gamma ray energies and intensities. Looking at various publications or databases related to decay data, the quantities of interest generally differ. Several reasons are at the origin of these Tapez une équation ici.differences, e.g. some results were not available at the time of the evaluation, different judgments brought to references could result in their rejection, different evaluation methodologies, etc. These data are used in many applications, for example, in the field of gamma ray spectrometry measurements, where the detectors used (such as Si(Li), HPGe) must be accurately calibrated. To meet the demand, a previous paper was published to recommend accurate gamma ray energies [1]. A parallel effort has been pursued by an international group (Decay Data Evaluation Project) to propose a list of gamma rays whose intensities are sufficiently well-known so as to make them appropriate for the calibration of gamma ray spectrometers and other applications. The Decay Data Evaluation Project (DDEP), was organized in 1995 by Helmer [2], [3]. It has included, throughout the years, various contributors from well-known scientific institutes (Table 1). The work has been constant and its recognition was achieved when the Bureau International des Poids et Mesures (BIPM— CCRI(II)) in 2004 decided to adopt these data as recommended values to be used in all the intercomparison exercises in the field of ionizing radiation metrology. Since then, the DDEP evaluations have been published in a series entitled "Monographie BIPM-5" [4] for which six volumes have already been issued. The objective of the DDEP is to provide carefully recommended data describing the nuclide decay, among them the γ ray intensities which are extensively used in many fields of application. The DDEP brings together scientists from several laboratories, most of which are the radionuclide standard laboratories in their respective country. The methodology followed in each evaluation is the same and the evaluator provides

written documentation of all data used, all decisions made and detailed calculations [5]. In conjunction with the IAEA Nuclear Data Section, two Coordinated Research Projects (CRP) have been organized and conducted with the participation of a number of DDEP evaluators using the DDEP methodology in both cases [6], [7], [8]. The aim of this paper, which is based on DDEP studies, is to provide a list of carefully evaluated gamma ray intensities for nuclides of interest in the field of detector calibration, nuclear waste management, medical applications, etc. In the first part, a brief description of the methodology followed throughout the evaluations is given, different methods of gamma ray intensity evaluation are presented, some typical examples of evaluations are shown; in the second part, the list of the chosen nuclides is given along with their applications, and finally a list of recommended gamma ray intensities is presented.

Authors	Laboratory/list of evaluated nuclides
V. Bé, M.M. X. Chisté, Mougeot	Laboratoire National Henri Becquerel, France/ C-11, N-13, O-15, F-18, K-40, Ar-41, Ca-47, Sc-47, Fe-52, Mn-52, Co-58, Fe-59, Cu-64, Zn-65, Ga-67, Ga-68, Se-75, Kr-85, Rb-86, Rb-88, Y-90m, Sr-92, Y-92, Tc-99m, Ag-108, Ag-108m, I-123, Te-123m, Sb-124, I-125, Sb-126, Sn-126, Xe-127, I-131, Cs-134, Ce-139, Ce-144, Pr-144, Nd-147, Sm-153, Yb-169, Lu-176, Ta-178, Ta-182, Au-195, Pb-203, Bi-207, Pb-210, Bi-214, Pb-214, Ra-226, Am-243
Ε. Browne, Baglin	C. Lawrence Berkeley National Laboratory, USA/Al-26, Sc-44, Ti-44, Ni-57, Ga- 66, Eu-152, Re-188, Ir-192, Ir-194
V.P. Chechev, N.K. Kuzmenko	V.G. Khlopin Radium Institute, Russia/ Co-57, Mo-99, In-111, I-129, Ba-133, Cs-137, Ce-141, Eu-154, Eu-155, Tm-170, Ra-223, Pa-233, Th-233, Np-236, Np-236m, Np-237, Np-238, Np-239, U-239, Am-241, Cm-243, Cm-245
R.G. Helmer	Idaho National Engineering and Environmental Laboratory, USA/Be-7, Na-24, Sc-46, Co-60, Nb-95, Nb-95m, Zr-95, Ag-110, Ag-110m, Sn-113, In-113m, Ba- 140, La-140, Gd-153, Gd-159
F.G. Kondev	Argonne National Laboratory, USA/Lu-177, Hg-206, Tl-209, Tl-210, Pb-211
T.D. MacMahon, A. Arinc, A. M.J. Pearce, Woods	National Physical Laboratory, UK/Co-56, Ru-106, Ac-228, Pa-231
E. Schönfeld, Dersch	R. Physikalisch-Technische Bundesanstalt, Germany/ Cr-51, Mn-54, Sr-85, Y-88, Cd-109, Ho-166, Ho-166m, Re-186, Au-198, Tl-201
A.L. Nichols	University of Surrey, UK/Mn-56, Pd-109, Sb-127, Te-132, Hg-203, Tl-208, Bi- 212, Pb-212, Bi-215, Rn-219, Ra-224, Th-228, Am-244
M. Galán	Laboratorio de Metrología de RadiacionesIonizantes, Spain/Na-22, I-133, Xe- 133, Xe-133m, Xe-135m
A. Luca	IFIN-HH/Radionuclide Metrology Laboratory, Romania/ Bi-211, Ra-228, Pa- 234, Th-234

Table 1. List of the DDEP evaluators and contributors from 1995 to 2012.

2. Evaluation methodology

2.1. General evaluation approach and rules

The first step of an evaluation is to compile all the available existing data then an analysis, critical and statistical, is made for each quantity of the decay scheme. All evaluations are based on available experimental data, supplemented with theoretical calculations or considerations when necessary. Only the main stages in the compilation and evaluation are presented here, more details can be found in Ref. [9]. These stages comprise the following.•critical analysis of published results and, if necessary, correcion of these results to account for more recent values hitherto unavailable to the original experimentalists; as a rule, results without associated uncertainties are discarded; some results can be rejected and their rejection is documented.•data obtained through private communications are only used when there is no published article available and when all of the necessary information has been provided directly by the scientist carrying out the measurements.•adjustments may be made to the reported uncertainties.•recommended values are derived from an analysis of all the retained measurement results (or theoretical considerations), along with the standard deviations corresponding to the 1σ confidence level.

2.2. Evaluation of uncertainties and statistical process

The following definitions of uncertainties have been extracted from the "Guide to the expression of uncertainty in measurement" [10].

Uncertainty (*of measurement*): parameter associated with the result of a measurement that characterizes the dispersion of the values that could reasonably be attributed to the measurand. *Standard uncertainty*: uncertainty of the result of a measurement expressed as a standard deviation.

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

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

 When necessary and when sufficient details are known, the uncertainties given by authors can be re-evaluated by combining the standard uncertainties σ_A and σ_B through the general law of variance propagation:

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

Where u_c is the combined standard uncertainty, σ_A is the type A standard deviation, and σ_B is the type B standard uncertainty.

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

In the case of gamma ray intensities, the published results have generally been obtained over a number of years and by a variety of scientists from various laboratories. When several results have been published by the same laboratory only the last one is retained in the evaluation process. Hence, the data in the set can be considered as independent and a weighted average is calculated using the combined uncertainties of the individual values as weights. For *n* independent values a_i , each with a combined standard uncertainty u_{ci} , a weight p_i proportional to the inverse of the square of the individual u_{ci} can be assigned to each value.

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

where the weights are $/u_{c_i}^2$

An internal and an external uncertainty can be assigned to the mean value [11]:

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

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

The external uncertainty is given by the equation:

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

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

A measure of the consistency of the data is given by the ratio [2], [3]:

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

If this ratio is significantly greater than unity, at least one of the input data most probably has an underestimated uci which should be increased. A critical value of $\chi^2/(n-1)$ at 1%

confidence level is used as a practical test for discrepant data. The following table lists critical values of $\chi^2/(n-1)$ for an increasing degree of freedom $\nu=n-1$:

If $\chi^2/(n-1) \le$ critical $\chi^2/(n-1)$, the recommended value is given by $a = a_w \pm \sigma_{int}(a_w)$

If $\chi^2/(n-1)$ > critical $\chi^2/(n-1)$, the method of limitation of the relative statistical weight [11] is recommended when there are three or more values; uncertainty of a value contributing more than 50% to the total weight is increased to give a contribution less than 50% (weighting factor < 0.50). The weighted and unweighted average and critical $\chi^2/(n-1)$ are then recalculated: if $\chi^2/(n-1) \le$ critical $\chi^2/(n-1)$, the recommended value is given by

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

If $\chi^2/(n-1)$ > critical $\chi^2/(n-1)$, the weighted or unweighted mean is chosen, depending on whether or not the uncertainties of the average values result in overlap. If overlap occurs, the weighted average is recommended; otherwise the unweighted average is chosen. In either case, the uncertainty can be increased to cover the most accurate value.

2.3. Determination of the best value and associated uncertainty for the γ ray intensities

From the results of the statistical process, as described above, the evaluated γ ray intensities are derived. However, to obtain confident γ ray intensity values, it is necessary to know not only the intensity values themselves but also the rest of the nuclide decay scheme. The overall consistency of the decay scheme being the best check of the quality of γ ray intensities. The goal of the decay data evaluation is to construct the decay scheme of the studied nuclide, i.e. to determine the energy and intensity of all emissions occurring in its decay: α , β^- , β^+ , conversion and Auger electrons, γ rays and X rays, as well as the electron capture (EC) probabilities, branching ratios, etc.

When the decay scheme is built, its overall consistency is checked by controlling:

- − at each nuclear level in the daughter nucleus, the sum of the probabilities of the transitions populating it is equal to the sum of those depopulating it;
- − the total energy carried away by various emissions is equal to the available decay energy;
- − the X-ray intensities deduced from the decay scheme data are consistent with the measured ones if available.

In most cases, the determination of the γ -ray energies and intensities is the first step, but to achieve the decay scheme construction other quantities are required:

- − for each nuclear level, its energy and the probabilities of the α, β or EC transitions feeding it;
- − for each γ transition, its placement in the decay scheme and its related internal conversion coefficients which are measured or theoretically calculated.

However, from time to time, the intensities of the γ rays are derived also from other measurements or physical quantities, for example:

- $\overline{}$ Sometimes the α emission intensities have been extensively measured, then using the information listed above the same process is conducted but using the α intensities as the starting point.
- − The decay scheme characteristics, such as the probability balance, are determined from the knowledge of the internal conversion coefficients, measured or theoretically calculated.

Below the different methods used to obtain the best values for the γ ray intensities and examples illustrating them are given.

a) Evaluation based on relative γ ray intensities and internal conversion coefficients

Most often the γ ray intensities have been measured as *relative* values, i.e. one of the γ rays, generally the most intense, is chosen as a reference line and its intensity is arbitrarily fixed to a certain value (e.g. 100) and the intensity of the other rays are given relative to this reference value.

However, users require *absolute* values, i.e. intensities in percent of nuclide decays. The transformation from relative to absolute values assumes the knowledge of a normalization factor N which is deduced from examination of the decay scheme. One verification of the decay scheme consistency is that for 100% of nuclide decays, 100% of the transitions (α, β, γ, and EC) must populate the ground state of the daughter(s). That is

$$
\sum_{i} I_{\gamma i} [1 + \alpha_{Ti}] + (P_{\beta} + P_{\alpha} + P_{EC}) = \frac{100\%}{N}
$$

Where the sum $\sum_i I_{\gamma i} [1 + \alpha_{\gamma i}]$ is over the γ transitions feeding the ground state(s); $I_{\gamma i}$ is the relative emission intensity of the *i*th gamma-ray, α*Ti* is its total internal conversion coefficient and $P_{\beta} + P_{\alpha} + P_{EC}$, is the sum of β , α , EC transitions populating the ground state. *N* is the normalization factor between the relative and absolute scales.

In the case where there are no α , β or EC transitions to the ground state, N, the normalization factor, is then deduced from the measured $I_{\gamma i}$ values and the related $\alpha_{\gamma iS}$ and, the associated uncertainty, *dN*, is calculated following the law of uncertainty propagation:

$$
N = \frac{100\%}{\sum_{i} I_{\gamma i} [1 + \alpha_{Ti}]} \quad \text{and} \quad dN^2 = +\sum_{i} \left(\frac{\partial N}{\partial I_{\gamma i}} dI_{\gamma i} \right)^2 + \sum_{i} \left(\frac{\partial N}{\partial \alpha_{Ti}} d\alpha_{Ti} \right)^2
$$

The α_T coefficients are obtained from theoretical calculations or measured values depending on the nuclide. When theoretical coefficients were preferred, in most cases they were interpolated from the tables of Band et al. [12] by using the computer code BrIcc [13] with the so called "Frozen orbital" approximation. In some earlier evaluations, they were interpolated from Rösel et al. tables [14]. For each evaluation, details on this point are refined in Ref. [5].

Example: ¹³⁴Cs decays by β⁻-particle emissions to excited levels in ¹³⁴Ba, none of which populate the ground state, and with a negligible electron capture branch. Several papers have been published, all of which report γ ray intensity values measured relative to the 604 keV γ ray. Measured values of the K conversion coefficients, α_K , have also been reported, and a good agreement was found compared with the theoretical values. Hence by application of the above reasoning, an absolute intensity value of 97.63 (8) % was calculated for the 604 keV γ ray, and all the other absolute intensities were then derived.

b) Evaluation based on measured absolute γ ray intensities

From time to time, *absolute* measurements of γ emission intensities are reported. Such measurements suppose that the mass activity of the nuclide under study was measured by using a method which is independent, as far as possible, of the decay scheme, such as the coincidence counting method.

Example: ¹⁵³Sm decays by β⁻-particle emissions to various levels in ¹⁵³Eu, including one to the ground state, hence the normalization can be tricky. Fortunately, several absolute measurements of the main γ emission intensities have been published, especially for the strongest γ ray of 103 keV. The value of 29.19 (16) % was adopted and used to convert all of the other relative γ ray intensities to absolute ones.

c) Evaluation, *from the decay scheme data*, *based on relative γ ray intensities and theoretical considerations*

In some cases, only the relative γ ray intensities are available but, fortunately, theoretical considerations can help in building the decay scheme.

Example: ¹²⁴Sb decays by β⁻-particle emissions to excited levels in ¹²⁴Te. In 2009 an international exercise was conducted to improve its decay scheme [15], [16], with relative and absolute γ ray intensities determined by several laboratories. Looking at the decay scheme, it was apparent that it would be best to use the relative results and calculated internal conversion coefficients, because the energies of the transitions involved are relatively high and their respective multipolarities are E2, hence the conversion coefficients can be considered very reliable. This process led to the adoption of a value of 97.775 (20) % for the 602 keV γ ray, which compares very favorably with 97.77 (26) % as derived from the measured results.

d) Evaluation based on relative γ ray intensities and other parameters

Special cases may arrive, especially when there is a direct transition from the parent nuclide to the ground state of the daughter nuclide. Ideally, an experimental determination of the transition probability would have been made, but such measurements prove to be particularly difficult.

Example: ⁶⁷Ga decays by electron capture to excited levels in ⁶⁷Zn, with an additional transition direct to the ground state. The evaluated relative emission intensities were used, with the absolute emission intensity of the 93 keV γ ray $I(\gamma_{93})$ to calculate the absolute gamma-ray emission intensities. Initially, $I(\gamma_{93})$ was determined from the total internal conversion coefficient $\alpha_T(\gamma_{93})$ theoretically calculated and the evaluated values of the conversion electron intensities $I(ce_{93})$ deduced from measurement results.

e) Evaluation based on α particle emission intensities and balancing the decay scheme

For nuclides decaying by α emission, often several measurements of the α particle emission intensities are available; when, *a contrario*, the γ emission intensities have not been measured or have large uncertainties. In such cases, the construction of the decay scheme starts with the critical examination of the α emission intensity measurement results and the γ transitions are deduced from the decay scheme balance.

Example: ²¹¹Bi decays mainly by α particle emission to a single excited level in ²⁰⁷Tl. The available experimental data on the unique γ ray occurring in the ²¹¹Bi α decay are discrepant. Therefore, the γ emission intensity was derived from the decay scheme balance using the much more precise values of the α particle intensity.

2.4. Comments on X-ray intensities and 511 keV gamma photons

Electron capture and internal conversion processes give rise to the creation of vacancies in the electronic shells and sub-shells of the daughter atom. The filling of a vacancy is followed by the emission of X-rays or Auger electrons and the creation of new vacancies in less bound shells. The intensities of the various K and L X-rays emitted following the re-arrangement process can be calculated according to parameters of the decay scheme (electron capture and γ transition probabilities, conversion coefficients) and the related atomic constants (fluorescence yields, Coster-Krönig transitions, etc.). They can then be compared with any measured values. Unfortunately, there are typically very few experimental results available. For this reason we do not consider X-rays in this paper.

Nonetheless, X-rays are commonly used, particularly to calibrate γ ray detectors in the 5– 20 keV range (e.g. ${}^{55}Fe$) or to control the amount of some actinides (e.g. ${}^{241}Am$) in radioactive waste. Recommended data on X-ray intensities for a number of nuclides can be found in BIPM Monographie-5 [4], [8] and the associated comment files and also in the IAEA report [6], [7].

When a β ⁺ transition occurs, the positron annihilates with an electron of the medium being traversed, which leads to the creation of two γ photons of energy 511 keV. This 511 keV emission is extensively used in nuclear medicine imaging (PET) and is also employed in γ spectrometry techniques to determine the activity of a β^+ emitter in a solution [17].

It is noteworthy that a positron can annihilate before completely slowing down, then the two emitted photons have energies greater than m_0c^2 , as would be obtained from annihilation at rest. In γ ray spectrometry, this phenomenon has the effect of removing, from the 511 keV peak, a fraction of the annihilation photons, producing a continuous photon spectrum. To take this effect into account the energy of this line is given as 511 keV *without uncertainty* since the energy is not exactly 511 keV. The magnitude of this effect depends on the material within which the positrons are stopped, such that the intensity of the 511 keV ray does not have a unique value but must be determined taking into account the experimental set-up.

As a rule, the adopted 511 keV ray emission intensity is two times the positron emission intensity (evaluation method "f" in Table 3).

3. Recommended gamma ray intensities

In Table 2 a list of chosen radionuclides is given. It corresponds to the list of nuclides for which decay data were evaluated by the DDEP collaboration up to the end of 2012 [5], [15].

Table 2. Nuclides and applications.

Applications of the isotopes listed are shown in the second and third columns of this table.

In Table 3 the recommended gamma ray intensities, greater than 1%, are presented (fourth column). In addition, the half-lives, the gamma-ray energies and the total internal conversion coefficients (ICC T) are also given, as recommended and published by the DDEP.

Table 3. Recommended γ ray intensities (>1%). Uncertainty (in parenthesis) given is the combined uncertainty at one standard deviation.

a From the decay scheme using measured relative γ ray intensities and theoretical or measured ICCs $(α_r)$.

b From the measured absolute intensities or using the measured absolute intensity of the strongest γ ray and relative intensities of the other γ-rays.

c From the decay scheme using theoretical considerations and relative γray intensity values.

d Evaluation based on relative γ-ray intensities and other parameters.

e Evaluation based on absolute α-particle intensities and decay scheme balance.

f Deduced from β ⁻-decay intensity, see Section 2.4 for explanations.

⁎ The evaluation method is given according to Section 2.3.

** The published DDEP intensity value was updated taking into account/based on new experimental data for: ⁵¹Cr [19], ⁸⁸Rb [20], ¹⁰⁹Cd [21], ¹¹³Sn [22], ¹²³I [23], ¹⁷⁶Lu [24], ¹⁷⁷Lu [25], [26], and 203Hg [27].

⁎⁎⁎ From recent accurate measurement of the Ho-166m half-life [28].

When DDEP evaluations are not available, e.g. for several nuclides of practical interest which were present in previous evaluations of some DDEP members [18], the above characteristics were updated by the authors. In addition, some published DDEP intensity values were updated in order to take into account new experimental data, by following the same evaluation methodology (see footnotes in Table 3).

4. Summary and comments

This article proposes a list of γ rays which are commonly used in various fields of application. The decay data characteristics of 150 nuclides have been studied and carefully evaluated, paying attention to the overall consistency of the decay scheme. A list of 656 γ -ray intensities is presented, as extracted from the DDEP database. These data appear across the seven volumes of Monographie-5 [4] published under the auspices of the Bureau International des Poids et Mesures (BIPM). As such, these data have been adopted for all international comparison exercises in the field of ionizing radiation metrology. One goal of this paper is to gather this set

of data together in order to facilitate and generalize their use, for example, in the more and more numerous simulation studies of detector response. This evaluation work has been conducted by members of the Decay Data Evaluation Project, who have carried out and reviewed all the evaluations and, to whom, the authors of this article are greatly thankful. As in many areas of research, funding is continually being reduced and manpower is becoming more and more difficult to find, hence we would like to use this opportunity to encourage other scientists to collaborate with us and help us expand the Decay Data Evaluation Project.

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