International Network of Nuclear Structure and Decay Data Evaluators

ACTION 44, 20th NSDD Meeting, Kuwait, January 2013, IAEA report INDC(NDS)-0635: Proposed procedures/guidelines for ENSDF half-life evaluations (ground states and long-lived isomers).

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(1) Identify and accumulate ALL published measurements of the half-life of the specified nuclear level(s)

(2) Ensure that all of the above identified half-life data and origins (NSR key-numbers) are listed systematically (chronologically reverse) in the *Comments* area, or as a footnote in **Adopted Levels, Gammas data set.**

(3) Consider any other features of each specific measurement for either rejection or increased preference, based on your own experience and subjective judgements. Examples include the following:

- acceptance or rejection of *grey* references (publications that have not been fully peer reviewed: laboratory reports; conference proceedings; sometimes the journal issue of a set of conference papers),
- measurement technique (compared with others, the technique is judged/known to be more appropriate for the half-life being addressed),
- recognised difficulties and complications (e.g. impact of impurities, detector limitations, background subtraction, dead-time losses, relative to “standards”),
- known reliability or improvements in a particular measurement technique (improvements might make the date of the measurements important),
- regular and lengthy measurement programme of specific half-lives for important applications (normally a policy instigated by national standards laboratories, but also observed to be undertaken by others) can result in rejecting all but the most recently reported value; complications can also arise when the laboratory changes equipment/technique,
- if the same author(s) determine a particular half-life based on the same measurement technique/apparatus, only consider the most recent value in deducing the recommended value,

and various other imponderables.

An important issue of procedure is faced by any evaluator commissioned to derive a recommend half-life with an uncertainty (for example) at the 1σ level from a set of data varying widely with measurement techniques, data handling procedures by the measurers, and problems with the detail (or lack thereof) provided in a publication. Unrealistically low uncertainties are known to be reported in the field of half-life measurements (particularly obvious when systematic uncertainties are ignored by the experimenters), such that various subjective decisions may need to be taken by the evaluator:

- reject measurements that do not quantify the uncertainty (budgets) at all;

- reject or be cautious of measurements with uncertainties that are judged to be totally unrealistic and/or incorrect;
- reject or be cautious of half-life studies that suffer from insufficient measurement time when determining activity decay as a function of time in order to quantify the slope of such a plot, and which do not provide details of counting losses;
- increase the uncertainty in a particular measurement on the basis of known limitations in the measurement technique, hopefully described adequately in the paper;
- increase specific uncertainties during the course of the process of weighted-mean calculation, and subsequently recycle until the weighting of any particular half-life measurement does not exceed a prescribed level (one common practice is "no more than 50% weighting").

All actions of above type which involve some form of subjective judgement require full explanation of what was done and why, and should be included in the *Comments* area.

(4) Identify outliers, document and discard, based on the criteria adopted in least-squares analysis codes. Numerous averaging techniques have been proposed and developed (see VISUAL AVERAGING LIBRARY or AVETOOLS on NNDC webpage). Examples include:
   weighted mean (WM);
   limitation of the relative statistical weight (LRSW or LWM);
   normalised residuals (NR);
   Rajeval Technique (RT) (M.U. Rajput, T.D. MacMahon, Nucl. Instrum. Methods Phys. Res. **A312** (1992) 289-295);
   BootStrap (BS) (O. Helene, V.R. Vanin, Nucl. Instrum. Methods Phys. Res. **A481** (2002) 626-631); etc.

These disparate techniques use different methods to handle the uncertainties, identify outliers, and derive the mean value and uncertainty. LRSW, NR and RT use the uncertainties and occasionally inflate them to accommodate discrepant data; all three of these methods should be used simultaneously to identify outliers (i.e. defined as such if at least two of the methods identify a data point as an outlier). BS method ignores uncertainties, and therefore does not identify outliers. Software codes are available to run these methods of analysis simultaneously/together for direct and speedy comparison. There are eight different averaging methods in the Visual Averaging Library code (V-AVELIB) developed by Michael Birch at McMaster, and available through NNDC. This code also handles asymmetric uncertainties. Note that AVETOOLS does not handle asymmetric uncertainties.

(5) All acceptable half-life data to be analysed by means of these techniques
   - may need to define which method is the most appropriate – WM, LRSW, NRM, RT, BMR, BootStrap, others, and so adhere to consistency in the selection of the recommended half-life value and uncertainty,
   - role of reduced χ^2 in such analyses needs to be better defined, implemented and used to develop a more rigorous understanding of the data set adopted for full analysis.
   - when a new half-life measurement for a ground state or long-lived isomer comes to the evaluator's attention, the impact of that measurement on the currently

recommended ENSDF value should be assessed, and suitable adjustments made in ENSDF, if deemed necessary.

- as an overall guide:

  adopt WM value and uncertainty when measured half-life data are not discrepant;

  adopt value from LRSW or other procedures when measured half-life data exhibit discrepancies;

  the recommended uncertainty should generally be no lower than the lowest uncertainty to be found in sets of experimental half-life data that are not individually defined in terms of various types of separated component uncertainties (also see below);

  if the statistical and systematic components of the half-life uncertainty have been quantified as separate entities in the various measurements, the recommended overall uncertainty in the half-life should be the sum of the lowest systematic uncertainty to be found in the data set and the weighted mean of the statistical uncertainty;

  the final uncertainty should not be lower than 0.01%;

  the adopted analytical route should be clearly described in the *Comments* area (data accepted; data rejected; numerical method adopted/applied).

(6) **Literature coverage**: some half-life articles are published in non-nuclear physics or non-radioactivity journals, and can consequently be missed by NSR. Examples of such omissions can be found in journals that include Health Physics, Geochronology and Geochemistry, and Planetary and Earth Sciences. The DDEP group generally undertakes a more complete literature search than ENSDF for their selected set of nuclides, but they do not always register and request NSR key-numbers, when they make use of a reference not found in NSR. Examples of previously missing important articles on half-life measurements that were added to NSR about two months ago on request of one of the authors of this report: 1991Ma68 (*Health Physics* **61**, 511) for ^{214}Pb, ^{214}Bi; 1989Ma67 (*Health Physics* **57**, 121) for ^{218}Po; 2001Po32 (*Radiochemistry* **43**, 549) for ^{175}Hf, ^{181}Hf; and also several other references.

(7) **Useful article**: there is an interesting article by S. Pomme *et al.* from IRMM, Geel, published in *Journal of Radioanalytical and Nuclear Chemistry* **276** (2008) 335-339, which constitutes a useful document for evaluators of half-lives. Pomme and co-workers have also published significant articles on half-life measurements, mostly in *Applied Radiation and Isotopes*. A search of NSR can retrieve a list of some of these papers published during 2011-2014, where methodology and uncertainty budgets are discussed in good detail.

Examples (2010/11):

Co-62 half-life

Reference	Half-life (min)	Comments
1949Pa01	1.6 (2)	β-decay curves followed over six half-lives; decay curve shown
1960Pr05	1.9 (3)	β-decay curve not shown – only lists half-life
1962Va23	1.5 (1)*	β-decay curve followed over four half-lives; no discussion of impurities
1969Wa16	1.50 (4)#	γ-γ coincidence and high energy β; decay curves not shown – only lists half-life
1970Jo12	1.4 (2)	1129-keV γ decay followed for more than five half-lives; decay curves shown for several γ rays
	1.54(10)	Recommended value (LRSW – Limitation of Relative Statistical Weights)

* Uncertainty increased to ± 0.2 to reduce weighting to below 50%.

\# Uncertainty increased initially to ± 0.20 to reduce weighting to below 50%.

Co-62m half-life

Reference	Half-life (min)	Comments
1949Pa01	13.9 (2)	β-decay curves followed over six half-lives; decay curve shown
1957Ga15	13.91 (5)*	γ decay measured in well-type scintillation detector; minor Cu-64 and Ni-65 impurities present; no decay curves shown – only lists half-life
1960Pr05	13.8 (2)	β-decay curve not shown – only lists half-life
1962Va23	13.9 (2)	β-decay curve followed over about two half-lives; no discussion of impurities
1969Wa16	14.00 (24)	High energy β and γ decay; decay curves not shown - only lists half-life
1969Mo04	13.8 (5)	1163-, 1172-, 2003- and 2103-keV γ decay followed for about six half-lives; decay curves shown for several γ rays
1970Jo12	13.5 (3)	1163- and 1173-keV γ decay followed for more than two half-lives; decay curves shown for several γ rays
	13.86 (9)	Recommended value (LRSW)

*Uncertainty increased to ± 0.20 to reduce weighting to below 50%.

Cu-62 half-life

Reference	Half-life (min)	Comments
1954Nu27	10.1 (2)[#]	Cu-62 milked from parent Zn-62
1965Eb01	9.76 (2)	Decay of positron annihilation radiation; Cu-64 impurity considered constant - no decay curves, only lists measured half-life
1965Li11	9.79 (6)	Decay of positron annihilation radiation corrected for Cu-64 activity, and fitting of excitation functions for Co-63(n,2n)Cu-62 reaction at En=12.6-19.6 MeV – lists half-life derived from these fittings
1969Bo11	9.7 (1)	Decay of positron annihilation radiation and fitting of excitation functions for Co-63(n,2n)Cu-62 reaction at En=13-18 MeV – lists half-life derived from these fittings
1969Jo07	9.73 (2)	Decay of positron annihilation radiation - no decay curves, only lists measured half-life
1975Ca40	9.80 (2)	γ-ray decay – no decay curves, only lists measured value
1997Zi06	9.68 (4)	4πβ liquid scintillation spectrometry, twelve independent measurements spanning two to four half-lives
	9.673 (26)	4πγ ionization chamber, two independent measurements spanning two to four half-lives
2002Un02	9.673 (8)*	Quote 1997Zi06, see above, but uncertainty is statistical only.
	9.74 (6)	LRSW: weighted average of the above with uncertainty expanded so that range includes the most precise value (9.673 min); data set exhibits significant inconsistencies that mitigate against LSWM approach
1997Zi06	9.68 (4)	4πβ liquid scintillation spectrometry, twelve independent measurements spanning two to four half-lives
	9.673 (26)	4πγ ionization chamber, two independent measurements spanning two to four half-lives
2002Un02	9.673 (8)*	Quote 1997Zi06, see above, but uncertainty is statistical only.
	9.675 (22)	Recommended value: from weighted average of two values in 1997Zi06. Uncertainty should be increased to 0.026.

[#] Rejected as outlier, and not included in the data sets for LRSW analyses.

[*] Not included in averaging.

Further comments:

2012Fi12 (NIST correction to 2002Un02 half-life data (see also footnote above for [*])) – adjusted value of 9.672(8) min has no impact on the analysis of the data published up to and including 2002. 2014Un01 report a half-life of 9.673(8) min, which is identical to their previous value and therefore has no impact on the analysis of the data published up to and including 2002.

<u>Half-life of Bi-207: review of ENSDF evaluation, 2010</u> (A.L. Nichols)

Each relevant paper considered in reasonable detail below. Comments are given in order of year of publication of each of the highly-relevant papers. Earlier half-life measurements are significantly less accurately characterised, and have not been assessed in this exercise.

<u>1978Ya04: Yanokura et al., Nucl. Phys. A229 (1978) 92-98</u>

Three different approaches were taken to measure the half-life of Bi-207.

(1). The absolute disintegration rate of At-211 in a purified sample was measured by means of a liquid scintillation counter, and a large volume of the same solution was used to study the gamma-ray decay of daughter Po-211 and Bi-207 with a heavily-shielded Ge(Li) detector, calibrated against IAEA standard γ-sources of Na-22, Mn-54, Co-57, Co-60, Ba-133 and Cs-137. The prominent 569.7-keV gamma ray was used to calculate the decay rate of Bi-207 (emission probability of 99.85% was used from Parsa and Markowitz, *J. Inorg. Nucl. Chem.* **36** (1974) 1429-1431), with a theoretical total internal conversion coefficient of 0.0221 adopted for this E2 transition). Thus, the half-life value for Bi-207 was "evaluated" to be 32.2 ± 1.3 years.

(2). A Bi-207 half-life of 31.7 ± 3.7 years was determined from a source prepared for liquid scintillation counting, but after complete decay of At-211, whereby the large uncertainty was attributed to the poor detection efficiency when gamma counting this particular liquid sample (?).

(3). And finally, the half-life of Bi-207 was also determined from the EC/α branching ratio, the emission probability of the 6868-keV α transition from Po-211 to the 569.7-keV nuclear level in Pb-207, the half-life of At-211, and the decay probability of Bi-207 feeding the 5769.7-keV nuclear level in Pb-207. A half-life value of 33.4 ± 0.8 years was calculated via this method. The authors assigned the small uncertainty to the counting statistics involving the 569.7-keV gamma ray – this value was adopted as the definitive recommended half-life through rather nebulous reasoning (simply because the value was deemed to be the most accurate?).

Systematic uncertainties are ignored in this set of studies, and are difficult to extract from the contents of the paper. Furthermore, such issues as the data sources for the direct 569.7-keV gamma-ray study need to be re-assessed (emission probability and ICC(total)) to derive a new half-life value, rather than simply adopt the original value of 32.2 ± 1.3 years. The half-life derived from the liquid sample should simply be discarded as seriously inaccurate. Finally, the half-life calculated from the EC/α branching ratio and other derived nuclear data needs to be re-assessed (and discard if deemed inappropriate).

<u>1990Al11: Alburger and Harbottle, *Phys Rev.* C41 (1990) 2320-2324</u>

An end-window gas-flow proportional counter was used to determine the decay of β^- radiation from two samples of Ti-44 and one sample of Bi-207. Consideration of the detailed and overall performance of this system can be found in Alburger *et al*. *Earth Planetary Sci. Letts.* **78** (1986) 168-176. Long-term drift in counter voltage was deemed to be of the order of less than 0.5 V (c.f. 25 V to achieve the equivalent of 1σ statistical uncertainty); box pressure would have to vary by 0.15″ compared with monitored changes of better than 0.03″. Changes in temperature of 2°F would result in 1σ standard deviation change in activity ratios, while a variation from 30% to 80% in the relative humidity would also cause a variance of 1σ standard deviation. These latter parameters were only monitored close to the end of the earlier studies on Si-32/Cl-36 with the following observations: temperature fluctuated from 72.4 to 74.7°F, and average relative humidity varied between 35% to 76% - judged as unfortunate and important variations in any attempt to define SYSTEMATIC uncertainties. Fluctuations of the data points from a smooth exponential decay were observed that are approximately THREE times the statistical uncertainty, and the authors assigned this unusual behaviour to variations in the temperature and relative humidity. Uncertainties were also identified with the operating pressure for the system – judged by the authors as operational under somewhat lower conditions than optimum. Other considerations involved studies of restoration of operational stability (system required a week to re-stabilize of any power shut-down), and change to a new gas supply (no observable effect). One might judge an overall SYSTEMATIC uncertainty of the order of ± 1.5 for a value of 34.9 years, without consideration of source preparation, radionuclidic purity and stability.

Clearly, the uncertainties quantified in this paper are only the STATISTICAL uncertainties from the relative activity measurements for Cl-36, Ti-44 and Bi-207 (Figs. 1, 2, 3, 4). A recommended value of 34.9(4) years is derived by the authors for the half-life of Bi-207.

Consideration of a combination of systematic and statistical uncertainties could result in a significant adjustment to 34.9 ± 2.0 years. However, there are a number of imponderables in this analysis that can be seen to justify the rejection of the half-life value from this particular study by the original 207 mass chain evaluators.

1991Li10: Lin and Harbottle, *J. Radioanal. Nucl. Chem.* 153 (1991) 51-55
Note same common author for 1990 and 1991 publications (Harbottle).

An inadequate paper, with insufficient detail and lack of clear traceability. Used gamma-ray spectroscopy to monitor the disintegration rates of individual gamma rays, and calculated half-life data from a combination of these disintegrations rates, "known" gamma abundances and the detector efficiency curve. Measured gamma-ray abundances are compared with equivalent data from the NBS certification of the Bi-207 source, and recommendations to be found in *Nucl. Data Sheets* **43** (1984) 383.

Interestingly, three half-live values are quoted in this paper:

(1). 31.6 ± 0.7 years from "only" the major 569-keV gamma line;
(2). 32.7 ± 0.7 years from the 569- and 1063-keV gamma lines;
(3). 32.7 ± 0.8 years from the 569-, 1063- and 1770-keV gamma lines.

There is an argument to be made for just adopting the half-life value of 31.6 ± 0.7 years, although a reasonable understanding of the recommended uncertainty is required (and is judged to be unrealizable).

1992Un01: Unterweger *et al., Nucl. Instrum. Methods Phys. Res.* A312 (1992) 349-352

2002Un02: Unterweger, *Appl. Radiat. Isot.* 56 (2002) 125-130

Represent a small part of a long-term NBS/NIST exercise to monitor, characterise and revise the decay half-lives of an extensive list of radionuclides maintained and stored within NIST. These studies have been ongoing for approximately five decades, based on measurements by means of 4πγ pressurized ionization chambers and (more recently) high-resolution HPGe detectors.

Both of these papers lack sufficient detail, but refer to detailed descriptions and equipment and techniques to be found in NBS Special Publication 626 (1982) 85 and NBS Special Publication 250-10 (1987). However, specific systematic uncertainties are noted, such as the lower response of the ionization chambers that was believed to arise from instabilities in the old battery pack, and improvements noted after the vibrating reed electrometer and capacitor bank were replaced with a multi-range electrometer. Other unexplained changes also occurred periodically in the response of the ionization chamber to radium references sources prior to 1973.

The 1992 publication contains a recommended half-life for Bi-207 of 11523 ± 19 days which is equivalent to 31.55 ± 0.05 years (1 year (mean tropical year) ≡ 365.2422 days), which had only been followed for 0.6 half-lives (~ 19 years). Uncertainties are quantified in terms of Statistical Uncertainty (10.0) and Other Uncertainty (16.0), although I am uncertain as to what these numbers really mean.

The 2002 publication contains a recommended half-life for Bi-207 of 11523.0 ± 15.0 days which is equivalent to 31.55 ± 0.04 years (1 year ≡ 365.2422 days), which had been followed for 0.9 half-lives (~ 28 years). Uncertainties are also quantified in terms of Statistical Uncertainty (9) and Other Uncertainty (12), although I remain uncertain as to what these numbers mean.

Concluding Remarks

I would recommend discarding:
- half-life (2) from Yanokura et al;
- half-life of Alburger and Harbottle;
- half-lives (2) and (3) of Lin and Harbottle;
- ignore 1992 half-life of Unterweger *et al.* (replaced by recommended 2002 value).

Rework and accept half-lives (1) and (3) from Yanokura et al (however, may still discard re-worked half-life (3));
accept half-life (1) of Lin and Harbottle;
accept 2002 half-life of Unterweger.

Bi-207 half-life: 2011Ko04 – F.G. Kondev, S. Lalkovski, NDS 112 (2011) 707-853
Recommended $T_{1/2}$: 31.55 y *4*

$T_{1/2}$: From 2002Un02, using $4\pi\gamma$ pressurized ionization chamber at NIST; statistical uncertainty 0.025 y and systematic uncertainty 0.033 y. No impurities in the sources were observed using HPGe; decay has been followed over a period of t ≈ 28 y. The value agrees with that of 31.55 y 5 reported by the same group (1992Un01), when decay was followed over a period of t ≈ 19 y. **Value superior to others described below.**

Others (not used in the NDS evaluation):
32.7 y *8* (1991Li10) by measuring the activity of a calibrated ^{207}Bi source (t ≈ 17 y after the source was calibrated) with a HPGe detector; value determined by averaging activities for 569γ, (Iγ = 97.75%), 1063γ (Iγ = 76.0% 14) and 1770γ (Iγ = 6.95% 13); $T_{1/2}$ = 31.6 y *7*, when the activity was deduced using 569γ only. The quoted uncertainty is statistical only. A sizable systematic uncertainty can be expected, given the uncertainties in the nuclear data parameters used in the calibration of the source.

34.9 y *4* (1990Al11) using a gas-flow proportional counter system; the uncertainty is statistical only and quoted at 2σ level; the source was produced by bombarding a Pb target with 22-MeV deuterons following chemical separation; the measurements were followed over a period of t = 3.4 y. A break in the singles rates were observed around t = 1.7 y after the beginning of the measurements. So the data were analyzed in two separate parts yielding $T_{1/2}$ = 34.88 y 21 from the first 27 points (up to t = 1.7 y) and 35.2 y *9* from the next eight points; the quoted $T_{1/2}$ is higher than the adopted one. The quoted uncertainty is statistical only, although a large systematic uncertainty should be expected owing to sensitivity of the measurements to temperature and humidity changes. It is worth noting that $T_{1/2}$ = 66.6 y *16* was reported by this group (1990Al11) for ^{44}Ti, which is higher than other precise measurements of 58.9 y *3* (2006Ah10) and 60.7 y *13* (1999Wi01).

33.4 y *8* (1978Ya04) deduced indirectly using the decay of a ^{211}At source and knowledge of the ε/α branching ratio of ^{211}At (0.583/0.417), the emission probability of 6568-MeV α to the 569.7-keV level of ^{207}Pb (0.58% 1), the half-life of ^{211}At (7.23 h *2*) and the total emission probability of 569.7γ fed in ^{207}Bi ε decay (99.85%). The quoted uncertainty is statistical only, but a large systematic uncertainty can be expected. The authors also quote a value of 32.2 y 13 using the disintegration rate of ^{211}At in a purified sample measured by the means of a liquid scintillation counter and by adopting the 569.7γ to determine the decay rate of ^{207}Bi. A measurement performed after a complete decay of ^{211}At yielded $T_{1/2}$ = 32.2 y *37*, whereby the large uncertainty was attributed to the poor detection efficiency when gamma counting this particular sample.

38 y *4* (1972Ru10) using a ^{207}Bi source by counting the 569.7-keV gamma ray, using a NaI(Tl) scintillation spectrometer over a period of t = 0.5 y.

38 y *3* (1961Ap01) deduced indirectly using the decay of ^{211}At source and knowledge of the α branching ratio of ^{211}At (40.9%), the half-life of ^{211}At (7.214 h *35*) and the

total emission probability of 569.7γ that is fed in ^{207}Bi ε decay (assumed 100% gamma-ray emission probability and 2.2% total α).

28 y *3* (1959So12) using the parent-daughter activity of ^{207}Po and ^{207}Bi.

Concluding Remarks

On balance, we sympathise with the rejection of much of the existing half-life data (2010/11), with the emphasis placed solely on the NIST measurements of 2002Un02 to the exclusion of all other studies.

Further comments, February 2014:
Amongst other publications since 2010, 2012Fi12 and 2014Un01 from NIST provides strong evidence that some of their reported half-life measurements over many years are systematically incorrect because of previously undetected physical movements of the source holder within the ionization chamber used to perform the work. The impact on the measured half-life of ^{207}Bi shows a change from (11523 ± 15) d to (11403 ± 61) d. which represents a decrease in the half-life of ≈ 1%. An adjusted half-life value of 31.22 y *17* constitutes a significant correction to the originally recommended half-life and uncertainty of 31.55 y *4* reported by 2002Un02 and adopted in ENSDF – the uncertainty at the 1σ confidence level has increased by a factor of 4.25.