Ultra-precise half-lives in literature and evaluation issues

Balraj Singh
Department of Physics and Astronomy
McMaster University, Hamilton, Canada

IAEA-NSDD-2019
Vienna, April 8-12, 2019

Guidelines for Half-life evaluations: ground states and long-lived isomers

Appendix A in Summary Report of NSDD-2015 meeting: INDC(NDS)-0687

-the final uncertainty should not be lower than 0.01%.

By ultra-precise half-life, I mean <0.01% total uncertainty.

This issue came up at the IAEA-ICTP workshop October 2018, for A=218 ENSDF evaluation: half-life of ^{222}Rn decay to ^{218}Po:

2015Be07: PL-B 743, 526: 3.82146 (16 stat) (4 syst) d: 0.0044% total uncertainty.

2018Po10: ARI 40, 171 (2018): ^{99m}Tc: 6.00660 (5 stat) (17 syst) h: 0.003% total uncertainty.

I communicated with the first author Stefaan Pomme about this result. Value is valid for a chemical compound used in hospitals: TcO_4Na (sodium pertechnetate). Chemical effects could be 0.15%. ENSDF-2017 value is 6.0072(9) h; 0.015% uncertainty.

Questions:

Is lowest uncertainty of 0.01% in half-life justified based on available data?

How to evaluate half-life data in the presence of ultra-precise results?

Half-life evaluations

<u>Lowest recommended uncertainty</u>: bit of history.

1990Ho28: Pure Appl. Chem. 62, 941: Norman Holden at NNDC, BNL.

Evaluated $T_{1/2}$ of 30 nuclides from ^{3}H to ^{231}Pa, including ^{222}Rn.

"*In the review of nuclear data by the International Atomic Energy Agency[16], their general comment on uncertainties included a statement questioning the validity of any presently stated uncertainties of less than **0.1%** for half-lives*".

Ref 16: IAEA reference: C.W. Reich, R. Vaninbroukx, IAEA-TEC-DOC-336, 275 (1985):

Caption of Table 1 on p279 states: *"The Working Group questions the validity of any presently stated uncertainties of less than 0.1% for half-lives and <0.5% for other quantities".* Other quantities: alpha and gamma emission probabilities, …

Recollection from a US-NDP meeting at BNL about 15 years ago: responding to a question about lowest possible uncertainty in $T_{1/2}$, Charles Reich mentioned that the lowest uncertainty should not be less than 0.01%. Also literature scan of the last few years suggests uncertainties >0.01%.

After the ICTP-2018 workshop, I tried searching various databases whether one could justify such a limit from available data and evaluations.

Available Databases in the world for half-lives of nuclides

ENSDF (NDS): for all known nuclides: independent evaluation

NUDAT: for all known nuclides, most data from ENSDF

Nuclear Wallet Cards (2011): for all known nuclides, most data from ENSDF, some from current papers at the time.

DDEP: for 220 nuclides, including 20 isomers: independent evaluation.

NUBASE-2016: for all known nuclides, many values from ENSDF database, updated for papers after literature cut-off dates in ENSDF

Table of Isotopes 1996-99: for all known nuclides, mostly from the ENSDF database of those years.

Live chart of Nuclides (IAEA): for all known nuclides: from ENSDF.

Wall charts: Bechtel (old GE) (2010), Karlsruhe (2018), JAEA (2014): for all known nuclides, source of information not known; uncertainties not given.

I searched **ENSDF, DDEP and NUBASE-2016** databases for half-lives with uncertainties <0.01%.

Search of databases: ENSDF (Jan 2019 version)

Thanks to Marco Verpelli (IAEA-NDS) for retrieval of data from ENSDF

Nuclide	Half-life	Uncertainty in percent
^{210}Po	138.376(2) d	0.0014%
^{56}Mn	2.5789(1) h	0.0039%
^{124}I	4.1760(3) d	0.0072%
^{60}Co	1925.28(14) d	0.0073%
^{122}Sb	2.7238(2) d	0.0073%
^{198}Au	2.6941(2) d	0.0074%
^{131}I	8.0252(6) d	0.0075%
^{147}Pm	2.6234(2) y	0.0076%
^{222}Rn	3.8235(3) d	0.0078%
^{127}Xe	36.346(3) d	0.0083%
^{153}Sm	46.284(4) h	0.0086%
^{125}Sb	2.75856(25) y	0.0091%

17 cases: <0.01% (out of which 6 have uncertainties: 0.0091% to 0.0097%).

90 cases: 0.01 to 0.05%: very precise.

68 cases: 0.06 to 0.099%: precise.

^{210}Po (famous / infamous) nuclide on top of the list for the most precise half-life recommended in various databases for the last ~50 or so years, based on a value reported in 1964EiZZ (lab report), where the assigned uncertainty is only statistical. The systematic uncertainty could be much higher.

I have looked through data for all the other nuclides in this table, and there seem evaluation issues, combined with the fact that precise half-lives for several nuclides have been corrected by NIST (2014Un01: ARI 87, 92), e.g. for ^{198}Au, uncertainty should be at least 0.0003 d, or 0.011%.

Search of databases: DDEP

7 cases with uncertainties <0.01%.

Nuclide	Half-life	Uncertainty (%)
^{210}Po	138.3763(17) d	0.0012%: same in ENSDF
^{153}Sm	1.92855(5) d	0.0026%: 0.0086% in ENSDF
^{110m}Ag	249.78(2) d	0.0080%: 0.016% in ENSDF
^{125}Sb	2.75855(25) y	0.0091%: same in ENSDF
^{95}Zr	64.032(6) d	0.0094%: same in ENSDF
^{133}Xe	5.2474(5) d	0.0095%: same in ENSDF
^{54}Mn	312.19(3) d	0.0096%: 0.06% in ENSDF

Search of databases: NUBASE-2016

21 cases with uncertainty <0.01%, 15 are the same as in ENSDF, including 0.0014% for ^{210}Po.

Nuclide	Half-life	Uncertainty (%)	Comment
^{51}Cr	27.7010(11) d	0.0040%	From ENSDF-2006. Value is 27.704(4) d in ENSDF-2017, 0.014% uncertainty
^{222}Rn	3.8215(2) d	0.0052%	From 2015Be07. 0.0078% in ENSDF-2011
^{18}F	109.739(9) min	0.0082%	W. avg. of 109.770(18) (2014Un01), 109.722(12) (2010Ga04), 109.748(21) (2004Sc04). I get the value as 109.739(14), 0.013%. 109.77(5) min in ENSDF from NP-A(1995) eval
^{99m}Tc	6.0067(5) h	0.0083%	From ENSDF-2011. Value is 6.0072(9) h in ENSDF-2017, 0.015% uncertainty
^{10}C	19.3009(17) s	0.0088%	W. avg. of 19.2969(74) (2016Du10), 19.282(11) (2009Ba06, not 2009Ba04), 19.310(4) (2008Ia01). I get the value as 19.3047(63), 0.033%. 2016Du10 have another value of 19.3009(17). Combining this, WA is 19.3017(25) s, 0.013% 19.290(12) s in ENSDF from NP-A(2004) eval

^{210}Po half-life evaluation

ENSDF-2014, 2003, 1992, 1981: 138.376(2) d (1964EiZZ). Same in NUBASE-2016.
DDEP (2014): 138.3763(17) d (1964EiZZ); "..there is no reason to doubt this value and the published uncertainty". 1990Ho28 evaluation: 138.4(1) d

Since 1976 or so, in all the database, value is taken from a lab. report:

1964EiZZ: MLM-1209, p11 (1964): 138.3763(17) d, 0.0012% uncertainty.

Recent papers 2012Do08 and 2014Po01 have only nominal values (study of change in decay constatnt)

Measurements in 1964EiZZ, uncertainties are labeled as internal probable errors.

138.3749(6) (1954 to 1958 data)

138.3832(11) (1963 to 1964 data)

138.3726(15) (1964 data)

Authors WA=138.3763(17) d.

Above data are a discrepant set: I get WA=138.3763(25) d, but with reduced $\chi 2$ =25.

Unweighted average=138.3769(32) d.

1954Ei20 (PRC 96, 719):from the same lab as 1964EiZZ:

138.391(23)

138.401(12)

138.408(14)

138.4059(66)

138.410(24)

138.314(24)

Authors' weighted average=138.4005(58) d, clearly stating that uncertainty is statistical.

I get WA=138.4007(85) d, with reduced $\chi 2$ =2.8, fairly reasonable.

^{210}Po half-life: evaluation

Using all the 9 data points (6 from 1954Ei20 and 3 from 1964EiZZ), I get WA=138.3766(16) but with reduced $\chi 2$ =11. Unweighted average=138.3845(100).

Other measurements:

1953Cu46 (same lab MLM as 1954Ei20, α counting): 138.374(32) d

1953Gi10 (NBS): 138.39(14) d

1949Be54: 138.30(14) d

1936Sa01: 139.6(14) d

1931-Dorabialska: 137.6(6) d

1927-Da Silva: 140.2 d

1920Cu01: 140 d

1912Sc01: 136.5(3) d

Using 138.3845(300) (1954Ei20,1964EiZZ, assuming total uncertainty 3 times the statistical uncertainty);
138.374(96) (1953Cu46 quoted uncertainty of 0.032 increased 3 times);

138.39(14) (1953Gi10); 138.30(14) (1949Be54),

one gets WA=138.381(30) d, where the first value gets a weight of 84%.

Adjusting maximum weight of 50% for a value implies 0.067 uncertainty in the first value,

gives WA=138.372(50) d (0.036% uncertainty, instead of unrealistic 0.0014%). Need new measurement.

^{222}Rn half-life evaluation

ENSDF-2011, 1996, 1987, 1977: 3.8235(3) d (1972Bu33)

DDEP-2007: 3.8238(8) d. NUBASE-2016: 3.8215(2) d (2015Be07)

2015Be07: 3.82146 (16 stat) (4 syst) d, 0.0044%

Systematic uncertainty of 0.0010% surprisingly low!

Integral gamma-ray measurement (>6 keV) using NaI(Tl) detector: contains all the activities of ^{238}U or ^{226}Ra progeny from ^{222}Rn to ^{206}Pb. No discussion about pileup and system stability over long counting periods.

Comment from Ronald Colle, Senior researcher at NIST:

"…*The experiment is very, very complex and involves measurement of the progeny with some clearly undefined geometry, where the radon will certainly undergo diffusion in the olive oil bath. Yet, the authors contend that the "systematic" component of their uncertainty is four times less than just what they term the "statistical" part".*

Measurements, uncertainties are statistical.

3.82157(32) d

3.82134(30) d

3.82169(32) d

3.82124(35) d

WA=3.82146(16); uncertainty is 0.00030 if lowest value in the set is used.

Systematic uncertainty (?); could be the same as the statistical, or higher.

For averaging with other data use 3.82146(42) d; 0.011% uncertainty

^{222}Rn half-life evaluation: available data over 117 years.

2018Ap01: 3.81(12) d: nominal value.
2015Be07: 3.82146(42) d
2004Sc07 (PTB): 3.8195(30) d
1995Co34 (NIST): 3.8224(18) d
1972Bu33: 3.82351(34) d: integral gamma: no discussion of systematic uncertainties.
1990Ho28 evaluation adjusted the uncertainty to 0.00170.
1958Sh69: 3.83(3) d
1956Ma64: 3.82290(27) d: 1990Ho28 evaluation adjusted the uncertainty to 0.00170.
1956Ro31: 3.825(4) d
1955To07,1951To25: 3.825(5) d
1924Cu01: 3.823(2) d
1923Bo01: 3.825(4) d; 1921-Bothe: 3.811 d (same first author)
1913-Rutherford: 3.847 d; 1903-Rutheford: 3.71 d
1910-M. Curie: 3.85 d
1907-Rumelin: 3.747 d
1905-Sackur: 3.863 d
1904-Bumstead: 3.896 d
1902-P. Curie: 3.987 d

^{222}Rn half-life evaluation

WA of values from 1923 to 2015: 3.82177(42) d; with weight=79% for 2015Be07 value. Reducing its weight to 50%, i.e. adjusting the uncertainty to 0.00081 in 2015Be07, gives WA=3.82220(81) d; 0.021% uncertainty.

Conclusions:

1. Search of available data up to ~2017 for half-lives justifies 0.01% minimum uncertainty.

2. More precise values may be coming as e.g. 2018Po10 for ^{99m}Tc for a certain chemical of Tc, with uncertainty budget carefully analyzed and documented. In an e-mail of Dec 21, 2018, Stefaan Pomme (Geel) mentioned that highly precise measurements are coming up for ^{22}Na and ^{134}Cs from their lab.

3. General tendency among ENSDF evaluators to pick up the most precisely quoted result in literature, rather than critically going through all the available data critically. DDEP evaluators do better in terms of literature coverage, but their policy of rejecting values based on (1850s') Chauvenet's criterion is questionable. Choice of 50% maximum weight, and choice of adopted uncertainty not lower than the lowest in the experimental data, not consistent among both the NSDD and DDEP evaluators. Extra care is needed in the evaluation of half-lives quoted to 0.01-0.05% (and of course <0.01%) uncertainty, rather than simply copying what was done in previous evaluations, when no newer data are available. ^{210}Po and ^{222}Rn are good examples, where evaluations have suffered from not so good a judgement.

Wishful thinking !

Even though half-lives of nuclear ground states and long-lived isomers (<1 s or so) are basic to nuclear physics and applications, it seems strange to me that this world yet does not seem to have a consistent set of values throughout the chart of known nuclides. Available evaluated data in different databases (and places) seems somewhat of a hodge-podge.

Seems a need to create a database for half-lives of ground states and long-lived isomers, preferably by two evaluators, who can devote a year or so to this activity, with complete compilations covering all available literature and with careful analysis of uncertainties, to supply final recommended values with meaningful uncertainties, under the umbrella of a reputed organization.

Above exercise would be incomplete without following up on new data, and updating the database, where needed, at regular intervals.