Contribution to "Diamond Jubilee of RCA"

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How accurate are half-life data of long-lived radionuclides?

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Abstract: We have consulted existing half-life data available in Nuclear Data Sheets for radionuclides with $Z < 89$ in the range between 30 and 10^8 years with emphasis on their uncertainty. Based on this dataset, we have highlighted the lack of reliable data by giving examples for nuclides relevant for astrophysical, environmental and nuclear research. It is shown that half-lives for a substantial number of nuclides require a re-determination since existing data are either based on one single measurement, are contradictory or are associated with uncertainties above 5%.

Keywords: half-life; long-lived; nuclear data; radionuclide; uncertainty.

1 Introduction

Isotopes with long half-lives are of special interest for scientists in various fields of nuclear research, among others related to energy production, nuclear astrophysics or environmental applications:

- Fission products (e.g. ^{79}Se, ^{135}Cs) and minor actinides can have a high radiological impact on the environment during operation and severe accidents of nuclear power plants and on the residual heat production during storage of spent nuclear fuel. Correspondingly, high attention has to be paid in case of intermediate and/or final disposal of nuclear waste.
- The so-called "branching points" in the nuclear s-process nucleosynthesis (e.g. ^{60}Fe, ^{79}Se), radionuclides produced via the p-process (e.g. $^{148/150}$Gd, ^{146}Sm, ^{154}Dy) as well as several other long-lived isotopes, are of high relevance for the understanding of the element formation in the early solar system and the development of our universe.
- A couple of suitable isotopes (e.g. ^{10}Be, ^{14}C and ^{32}Si) can be used for nuclear dating of environmental samples in order to reconstruct climate changes, material circulations and other processes relevant in geoscience and climate research.

The precise knowledge of the nuclear properties, in particular half-lives, transition probabilities and branching ratios as well as cross sections of a variety of nuclear – mainly neutron- and charged-particle-induced – reactions of these isotopes is a precondition for the evaluation of scientific data in any of these research areas. But how reliable are the presently known data on the decay properties of these long-lived isotopes? In an astonishingly large number of cases, we are dealing with very old measurements, only a small number of determinations, large uncertainties or even contradictory results. In the following text, we aim to display an overview on existing half-life data, especially pointing out some of the recent measurements and the consequent necessity for re-evaluations. We are going to discuss the impact of inaccurate or wrong values, detect obvious lack of data, explain the reasons and give some ideas for future improvement, including also a brief sketch of own research activities for selected isotopes. It is important to note that the underlying work does not aim towards evaluation of the reported nuclear decay data but should rather give a systematic overview on the current knowledge about half-lives of long-lived isotopes. For a very recent publication on recommended half-life data of commonly used radionuclides and methods of their evaluation, the reader is referred to Ref. [1].

2 State-of-the-art

So far, more than 3000 radioactive isotopes are known with half-lives lasting from a few nanoseconds to billions of years. The definition of "short-lived" or "long-lived" strongly depends on the corresponding applications. While

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"short-lived cosmogenic radionuclides" can have half-lives of several million years, isotopes of super-heavy elements with atomic numbers above 104 count already as "long-lived" if they have half-lives of a couple of hours. In order to be able to make a reasonable pre-selection of isotopes that could be included in the current work, we have consulted existing half-life data reported by the Nuclear Data Sheets (NDS) for isotopes with $T_{1/2}$ > 1 year, i.e. beginning with ^{106}Ru. We excluded *a priori* all radionuclides that are considered primordial giving an upper limit of $T_{1/2}$ < 100 million years, i.e. ending with ^{244}Pu. Figure 1 shows a graphical illustration of the relative uncertainties (1σ) of half-lives of these radioisotopes. The abbreviation 'a' (annum) is further used for the unit of 'year'.

As can be seen from Figure 1a, for the majority of the plotted isotopes (94 out of 120) the half-life is known with less than 5% uncertainty. There are some notable exceptions that are partially denoted in the figure or are not given at all, i.e. for ^{186m}Re, ^{250}Cm and ^{248}Bk values have been reported with no uncertainty. It is also noteworthy that for the half-life range between 1 and 30 years the knowledge of half-life data is of considerably better quality than that of longer-lived isotopes. Out of 41 nuclides in this range, 34 are given with an uncertainty lower than 1% and the 12 most precisely known $T_{1/2}$ data originate from isotopes in this half-life range (with ^{60}Co, ^{147}Pm and ^{125}Sb leading the list). This is not surprising given the fact that decay counting techniques are commonly applied for isotopes whose half-life is shorter than the period of a human generation. Exceptions certainly exist, as for example for ^{101}Rh.

We observe an overall trend that for isotopes commonly encountered in fission or activation, i.e. on the neutron rich side of the line of stability, half-life data are generally better known and the respective number of $T_{1/2}$ determinations is larger. This is especially true for isotopes of the actinide series, where large effort has been put to improve decay data for the nuclear energy community [2]. As can be seen in Figure 1b, the vast majority of half-life data in the actinide region is more precise than that for lighter isotopes – out of 35 nuclides in this region, 29 come with an uncertainty lower than 1%. The half-lives of $^{238, 239, 240}$Pu and ^{241}Am are among the most 20 precise, although most of these nuclides live too long for practical decay counting. Noteworthy is the astonishing certainty about the ^{227}Ac half-life as well as the lack of sophisticated data for $^{247, 248}$Bk and ^{250}Cm.

Based on the pre-selection given above, we decided to omit the actinide series in the underlying work. We also decided to set a lower limit for half-lives in the range of a human generation, since it is mankind that defines the perception of time scale. Therefore, for our evaluation here, we make an arbitrary cut at 30 years, which is certainly as good as any other, and call these isotopes "long-lived". In total, 54 radionuclides with half-lives matching these

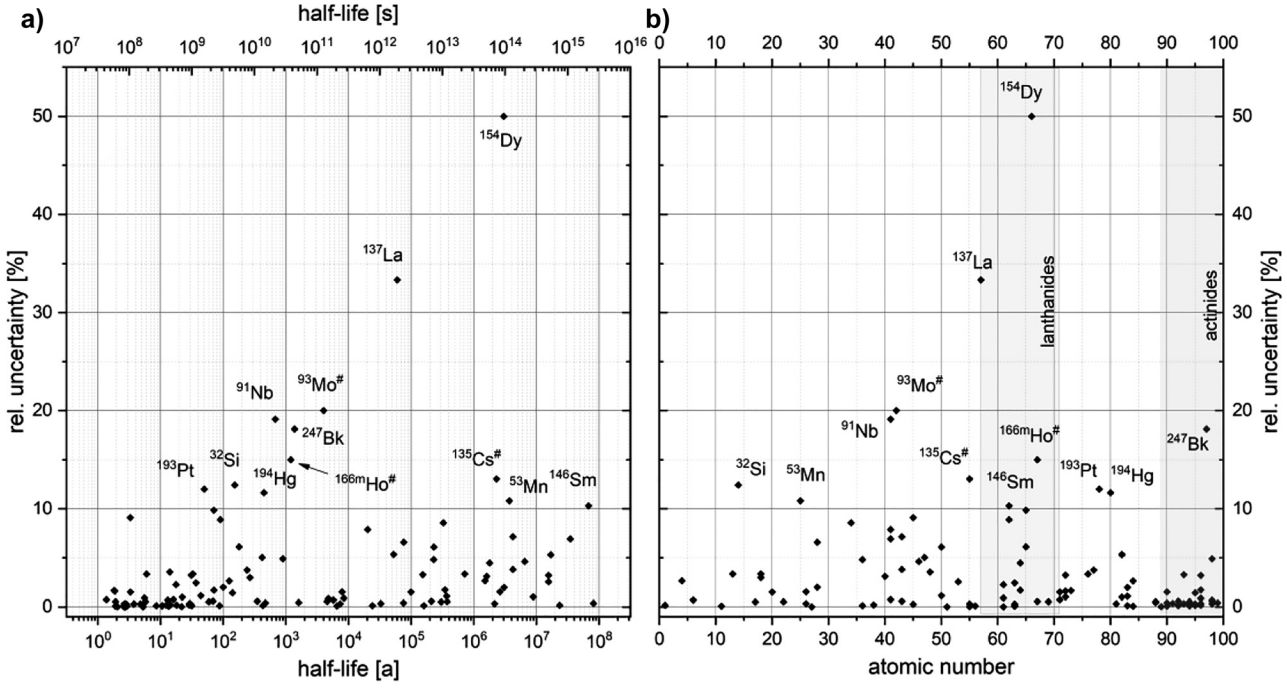

Figure 1: Relative uncertainties (1σ) of half-lives for artificial radioisotopes with $T_{1/2}$ > 1 a as function of a) $T_{1/2}$ and b) Z of the respective isotope. Data based on entries in Nuclear Data Sheets. For isotopes marked with a hash symbol (#), new half-life determinations have been reported.

criteria exist, with atomic masses from 10 (^{10}Be) to 226 (^{226}Ra), with a half-life range from ^{137}Cs (30.08 a) to ^{146}Sm ($6.8{\cdot}10^7$ a). A considerable amount of them (23) have given uncertainties for the half lives of 5% or more. In some cases, only one single measurement is reported and a considerable number of measurements are older than 50 years. In Table 1, a summary of these isotopes included in our review is given. All data are taken from the respective Nuclear Data Sheets up to volume 177 and were updated according to new measurements reported in literature until 31st of October 2021, new entries in the Evaluated Nuclear Structure Data File (ENSDF [3]) and values recommended by the Decay Data Evaluation Project (DDEP [4]). Uncertainties are stated in concise notation. We do not claim here absolute timeliness and completeness, which would be beyond the scope of the present contribution.

During the last years, it became obvious that a number of half-live values recommended by Nuclear Data Sheets have to be re-evaluated due to recent measurements showing considerable disagreement with values reported earlier. Prominent examples are ^{60}Fe [20, 21, 78], ^{126}Sn [42], ^{79}Se [79, 80], ^{135}Cs [48] and ^{146}Sm [81] with deviations up to 60% in comparison to former measurements. Considering this experience, more data have to be expected to be questionable, especially in cases, where only one measurement exists or where the measurements are very old, or both. In particular, some of the works from the early 1950 and 1960s suffer from incomplete descriptions of the measurement performance and barely documented estimates of uncertainties, making the results doubtful for critical evaluations. However, this does not automatically mean that old data are wrong. For instance, the most recent measurements of the ^{146}Sm half-life [81] is not consistent with two previous determinations, but is in good agreement with older data within the uncertainties, making it hard to judge what data to trust (see chapter 3.1.2). Similar situations are found for ^{126}Sn and ^{135}Cs. In other cases, newer studies reproduced the half-life values measured at earlier times, but with significantly improved uncertainties. Prominent examples are ^{182}Hf [82], ^{166m}Ho [61], ^{41}Ca [83], ^{129}I [46] or ^{151}Sm [53, 54].

The lack of reliable decay data for a considerable number of long-lived isotopes urgently calls for an improvement. However, a simple new measurement is not always the solution of the problem. In particular in cases, where already several, but contradicting half-life values exist (for instance for ^{32}Si or ^{135}Cs, see the following chapters) it is futile to merely add one more single measurement to this data landscape, which would rather complicate the situation further. Instead, an extended set of new determinations with highest accuracy is needed, from which a new, recommended value can be deduced towards acceptance as the consensually recommended value. Such important efforts are currently undertaken in the frame of the Decay Data Evaluation Project [4].

It is crucial that newly generated half-life data are obtained by as much as possible independent and complementary methods with a minimum deviation of the final values within the envisaged uncertainty. Otherwise, the community would end up with a similar data base, with the marginal difference of one more data point. The consequence of this would be – like before – that researchers might pick the half-life value that fits best to their experimental findings. This practice gives the impression that the scientific results suffer from some arbitrariness and, hence, are less reliable. Uses of various half-life values also exacerbate comparison of results. A harmonization using a commonly accepted half-life with low uncertainty is imperatively needed. These requirements are extremely challenging and depend substantially on the sample quality. Exploring possible reasons for this unsatisfying situation, several problems have to be faced.

At first, measurements of half-lives by following the decay are nearly impossible for isotopes with half-lives of more than a few hundreds of years due to detection system instabilities. Instead of that, the experiment has to be performed via the direct method making use of Eq. (1):

$$T_{1/2} = \frac{N \cdot \ln 2}{A} \tag{1}$$

where A is the activity of the isotope and N the number of its atoms. This implies that two very accurate and independent measurements (both of the activity A and the number of atoms N) have to be performed, with a sample containing sufficient amount of the wanted radionuclide in highest purity and very precise sample preparation methods allowing to trace back the corresponding sub-samples. Both measurements can be – depending on the nature of the isotope under study – very challenging. In particular, in studies from the early 1950 or 1960s, very often, theoretical predictions for the number of atoms were used, deduced from cross sections, implantation probabilities, annual deposition rates or other estimations. Some of the early Accelerator Mass Spectrometry (AMS) measurements applied for the determination of N suffered from insufficient knowledge on the total transmission, resulting in an essential underestimation. Activity measurements on the other hand often have to face several problems due to unknown or uncertain branching ratios and transition probabilities or are generally challenging for radionuclides that emit low energetic radiation. In various of these cases, radio-chemically pure samples are required, because any

Table 1: List of isotopes matching the criteria of half-life (30 a < $T_{1/2}$ < 10^8 a) and atomic number (Z < 89). Data extracted from Nuclear Data Sheets; new determinations and alternative evaluations are given in the comments.

Nuclide	$T_{1/2}$ [a] according to NDS[a]	Relative 1σ uncertainty [%]	NDS reference[a]	Year of latest measurements	No. of measurements	Comments
^{10}Be	1.51(4)·10^6	2.65%	[5]	2010	17	1.386(16)·10^6 a [6] 1.388(18)·10^6 a [7]
^{14}C	5.73(4)·10^3	0.53%	[8]	1972	18	DDEP & ENSDF: 5.70(3)·10^3 a
^{26}Al	7.17(24)·10^5	3.35%	[9]	1984	4	DDEP: 7.17(24) 10^5 a
^{32}Si	153(19)	12.42%	[10]	2015	10	159.4(56) a [11][b] ENSDF: 157(7) a
^{36}Cl	3.013(15)·10^5	0.50%	[12]	1966	4	DDEP: 3.02(4)·10^5 a
^{39}Ar	268(8)	3.00%	[13]	1996	3	276(3) a [14]
^{42}Ar	32.9(11)	3.34%	[15]	1965	1	
^{41}Ca	9.94(15)·10^4	1.51%	[16]	2012	8	DDEP: 1.002(17)·10^5 a
^{44}Ti	59.1(3)	0.51%	[17]	2006	11	DDEP: 60.0(11) a
^{53}Mn	3.7(4)·10^6	10.81%	[18][c]	1971	4	
^{60}Fe	2.62(4)·10^6	1.53%	[19]	2017	5	2.50(12)·10^6 a [20] 2.72(16) and 2.69(28)·10^6 a [21]
^{59}Ni	7.6(5)·10^4	6.58%	[22]	1994	6	DDEP: 7.6(5)·10^4 a
^{63}Ni	100.1(20)	2.00%	[23]	2008	6	101.2(15) a [24][b] DDEP: 98.7(24) a ENSDF: 101.2(15) a
^{79}Se	3.27(28)·10^5	8.56%	[25]	2014	11	DDEP: 3.56(40)·10^5 a
^{81}Kr	2.29(11)·10^5	4.80%	[26]	1964	2	
^{93}Zr	1.61(5)·10^6	3.11%	[27]	2010	4	DDEP: 1.61(6)·10^6 a
^{91}Nb	680(130)	19.12%	[28]	1982	1	
^{92}Nb	3.47(24)·10^7	6.92%	[29]	1978	2	
^{94}Nb	2.03(16)·10^4	7.88%	[30]	2012	4	2.04(4)·10^4 a [31]
^{93}Mo	4.0(8)·10^3	20.00%	[27]	2021	2	4.839(63)·10^3 a [32]
^{97}Tc	4.21(16)·10^6	3.80%	[33]	1998	2	
^{98}Tc	4.2(3)·10^6	7.14%	[34]	1973	4	
^{99}Tc	2.111(12)·10^5	0.57%	[35]	1984	4	DDEP: 2.115(11)·10^5 a
^{107}Pd	6.5(3)·10^6	4.62%	[36]	1969	2	
^{108m}Ag	418(21)	5.02%	[37]	2018	5	448(27) a [38] 437.7(88) a [39][b,d] DDEP & ENSDF: 438(9) a
^{121m}Sn	43.9(5)	1.14%	[40]	2002	4	
^{126}Sn	2.30(14)·10^5	6.09%	[41]	2009	6	2.345(71)·10^5 a [42] 2.33(10)·10^5 a [43] 1.980(57)·10^5 a [44]
^{129}I	1.57(4)·10^7	2.55%	[45]	2018	6	1.614(12)·10^7 a [46] DDEP: 1.61(7)·10^7 a See also Ref. [1]
^{135}Cs	2.3(3)·10^6	13.04%	[47]	2016	4	1.6(6) and 1.3(2)·10^6 a (2σ) [48]
^{137}Cs	30.08(9)	0.30%	[49]	2020	>20	DDEP: 30.05(8) a See Ref. [1] for new data
^{137}La	6(2)·10^4	33.33%	[49]	1956	1	
^{146}Sm	6.8(7)·10^7	10.29%	[50]	2012	5	See also [51]
^{151}Sm	90(8)	8.89%	[52]	2015	3	96.6(24) a [53][d] 94.6(6) a [54][d] DDEP: 94.7(6) a
^{150}Eu	36.9(9)	2.44%	[55]	1993	3	
^{148}Gd	71.1(12)	1.69%	[56]	2003	4	
^{150}Gd	1.79(8)·10^6	4.47%	[55]	1966	3	
^{157}Tb	71(7)	9.86%	[57]	1983	4	
^{158}Tb	180(11)	6.11%	[56]	1984	3	
^{154}Dy	3.0(15)·10^6	50.00%	[58]	1971	4	
^{163}Ho	4570(25)	0.55%	[59]	1988	3	

Table 1: (continued)

Nuclide	$T_{1/2}$ [a] according to NDS[a]	Relative 1σ uncertainty [%]	NDS reference[a]	Year of latest measurements	No. of measurements	Comments
^{166m}Ho	1.20(18)·10^{3}	15.00%	[60]	2012	3	1.1326(39)·10^{3} a [61][d] DDEP: 1.133(8)·10^{3} a
^{178m2}Hf	31(1)	3.23%	[62]	1973	1	
^{182}Hf	8.90(9)·10^{6}	1.01%	[63]	2005	4	
^{186m}Re	2·10^{5}	None	[64]	1972	1	
^{192m2}Ir	241(9)	3.73%	[65]	1970	1	
^{193}Pt	50(6)	12.00%	[66]	1971	3	
^{194}Hg	447(52)	11.63%	[67]	1981	4	
^{202}Pb	5.25(28)·10^{4}	5.33%	[68]	1981	2	
^{205}Pb	1.70(9)·10^{7}	5.29%	[69]	1958	1	
^{207}Bi	31.55(4)	0.13%	[70]	2020	7	31.22(17) a [71] DDEP: 32.9(14) a See also Refs. [1, 72]
^{208}Bi	3.68(4)·10^{5}	1.09%	[73]	1964	2	
^{210m}Bi	3.04(6)·10^{6}	1.97%	[74]	1976	3	
^{209}Po	125.2(33)	2.64%	[75]	2015	3	DDEP: 115(13) a
^{226}Ra	1.600(7)·10^{3}	0.44%	[76]	1966	6	DDEP: 1.600(7)·10^{3} a

[a]Reference according to Nuclear Data Sheets; for ^{10}Be and ^{14}C data are from Nuclear Physics A. [b]Data has been included in the Evaluated Nuclear Structure Data File (ENSDF) [3]. [c]The ^{53}Mn half-life stated in reference [18] contains a misprint (3.74(4) instead of 3.7(4)·10^{6} a according to Ref. [77]). [d]Data has been included in evaluation by DDEP [4].

other radiation source in the sample would influence the measurement result. Such samples are hard to produce and normally require mass separation after or before neutron or charged particles irradiations. A very good overview on how half-lives can be measured, the problems to be solved and important conclusions can be found in Ref. [84].

Another reason for scarce half-life data is the availability of sufficient sample material. Studying the original literature, it becomes obvious that in many cases the amount of material available was extremely limited. The half-life determinations for ^{53}Mn, for instance, were performed with only 10^{11}–10^{13} atoms in total [77, 85, 86]. Very often, the isotopes are very exotic and can hardly be produced by conventional means to obtain sufficient sample material. Prominent examples are neutron-rich isotopes such as ^{32}Si and ^{60}Fe or α-emitters of the lanthanide series such as ^{150}Gd and ^{154}Dy. International collaborations of metrology institutes with entities capable of producing and isolating these exotic long-lived radionuclides are prerequisite to enable measurements for improving the situation in the current nuclear data landscape.

In the following, we highlight some examples for radioisotopes with insufficient, incorrect or contradicting half-life data, with emphasis on the impact in various fields of science.

3 Selected examples

3.1 Nuclear astrophysics applications

3.1.1 p-process examples: ^{91}Nb (680(130) a), ^{92}Nb (3.47(24)·10^{7} a) and ^{94}Nb (2.03(16)·10^{4} a)

Around 35 heavy, proton-rich isotopes occurring during different stellar burning processes bypass reaction paths like charged-particle induced fusion and neutron-capture reactions. A series of photo-disintegration reactions occurring in supernovae, called the p-process, in combination with proton capture were proposed to explain the observed abundance pattern. However, the models fail in case of the light p-nuclei ^{92,94}Mo and ^{96,98}Ru [87, 88], these being the p-nuclei with the highest isotopic abundance. Since recent theories state that a chain of proton captures can lead to the production of ^{92}Mo, the main uncertainty for that production scenario is the cross section of the ^{91}Nb(p, γ) reaction and its inverse reaction for the destruction of ^{92}Mo. With respect to this, the ^{91}Nb(p, γ)^{92}Mo reaction might be one of the key reactions to explain the isotope production in this mass region. The exact knowledge on the decay rate of ^{91}Nb (and, as explained below, of the other long-lived Nb isotopes) is one of the preconditions to correctly evaluate the situation.

The adopted value of the ^{91}Nb half-life of 680(130) a [28] originates from only one measurement [89]. The sample was prepared by the (n, p) reaction on natural molybdenum metal and following chemical separation of the produced niobium from the molybdenum matrix. The activity of ^{91}Nb was determined by X-ray measurements on a low-energy HPGe detector. The number of atoms of the produced niobium isotopes was measured by surface ionization mass spectrometry as ratios between ^{91}Nb/^{92}Nb and ^{91}Nb/^{94}Nb, thus the result is partially dependent on the half-lives of $^{92/94}$Nb.

^{92}Nb is a p-process isotope decaying into ^{92}Zr. Establishing a ^{92}Nb–^{92}Zr chronometer would give the possibility to provide information on early Solar System evolution and insights into the p-process nucleosynthesis. The adopted value for its half-life, $3.47(24)\cdot10^7$ a [29] is a weighted average of the two so far existing measurements. In 1977, authors in Ref. [90] performed a measurement as a combination of surface ionization mass and γ-ray spectrometry with a ^{92}Nb sample obtained from fast neutron reactions on natural molybdenum metal. Two half-life values were obtained from this experiment. The first one, $3.3(5)\cdot10^7$ a, originates from mass spectrometric data dependent on the half-life of ^{94}Nb. The second one, $3.2(6)\cdot10^7$ a, was obtained from the number of atoms determination by dilution analysis. In the work of Ref. [91], the ^{93}Nb(n, 2n)^{92}Nb reaction on niobium foils was utilized. The amount of ^{92}Nb in the sample was calculated based on neutron flux measurements and cross sections for the nuclear reaction, whereas the activity was determined by γ-ray spectrometric measurements, leading to a half-life value of $3.6(3)\cdot10^7$ a.

The half-life of ^{94}Nb has been determined with an uncertainty lower than 2% by Ref. [92] using a neutron-activated Nb sample and a combination of γ-ray spectrometry for the determination of ^{94}Nb activity and multi collector inductively coupled plasma mass spectrometry (MC-ICP-MS) for the number of ^{94}Nb atoms. The obtained value, $2.04(4)\cdot10^4$ y is in agreement with the latest literature value from 1959 of $2.03(16)\cdot10^4$ a [93] and is compatible with the two earlier obtained values: $1.8(4)\cdot10^4$ a [94], $2.2(5)\cdot10^4$ a [95] within their given uncertainties. With the recent measurement by authors of Ref. [92], a sufficiently confirmed and, thus, reliable value for the half-life of ^{94}Nb is now available.

A new attempt to re-determine the half-life of ^{91}Nb is currently ongoing at PSI. At the Physikalisch Technische Bundesanstalt (PTB) in Braunschweig, Germany, an enriched ^{92}Mo target had been irradiated with protons in the energy range of 12–20 MeV to obtain around 10^{16} atoms of ^{91}Nb via the reaction ^{92}Mo(p, 2p)^{91}Nb and ^{92}Mo(p, pn)^{91}Mo/^{91}Nb [96]. The radiochemical separation of the wanted isotope from the target material has been performed successfully [97]. After sufficient decay of the co-produced ^{91m}Nb, part of this sample is ready for the half-life determination.

3.1.2 Radio-Lanthanides – ubiquitous in stellar nucleosynthesis

The region of lanthanide isotopes offers an especially fascinating scientific field for nuclear astrophysics, because most if not all of the nucleosynthesis processes of heavy elements are represented in this region of the chart of nuclides: s-process for nuclides such as ^{148}Sm or ^{150}Sm, r-process for nuclides such as ^{150}Nd or ^{154}Sm, p-process for ^{144}Sm and ^{146}Sm, the rest of the nuclides likely being synthesized through more than one process. A long-term research program was launched at PSI to produce and separate sufficient amounts of ^{137}La, $^{157/158}$Tb, $^{148/152}$Gd, ^{154}Dy and ^{146}Sm from irradiated Ta samples [98]. The corresponding half-live measurements are currently ongoing.

^{137}La $(6(2)\cdot10^4$ a$)$

The odd-odd neutron-deficient heavy nuclide ^{138}La is one of the rarest Solar System species. In spite of its very small abundance (^{138}La/^{139}La $\approx 10^{-3}$), ^{138}La is underestimated in all the p-process nucleosynthesis calculations performed so far. This results from an unfavourable balance between its main production by ^{139}La(γ, n)^{138}La and its main destruction by ^{138}La(γ, n)^{137}La [99]. For the evaluation of the relevant nuclear reactions, determining the mass ratios in this region, the destruction of ^{137}La by neutron capture and/or decay is therefore of paramount importance.

The only half-life determination for this radionuclide performed so far is more than 60 years old. Authors in Ref. [100] irradiated 0.88 g of natural CeO_2 in the ORNL graphite reactor for an extended period to yield $5.4\cdot10^{15}$ atoms of ^{137}La. After chemical purification, the La activity was determined using a Xe-filled proportional counter measuring the K-shell X-ray activity. A half-life value of $6(2)\cdot10^4$ a was obtained considering the thermal neutron capture cross section of ^{136}Ce, the neutron flux and chemical yield of the separation.

While the authors in Ref. [100] state, that the major source of uncertainty is the neutron capture cross section of ^{136}Ce, there appears to be a major disagreement still nowadays on the exact value of this quantity. While the Atlas of Neutron Resonances [101] recommends a value of $\sigma(^{136}\text{Ce(n, g)}^{137g+m}\text{Ce}) = 4.25(18)$ b according to the measurement of Ref. [102], a more recent determination [103] found a value of 7.64(63) b, in agreement to the value used

by Ref. [100]. Beyond that, the burn-up of ^{137}La during irradiation was not considered in Ref. [100] and the exact half-life and neutron capture cross section of this isotope remain unknown.

$$^{146}\mathbf{Sm} \quad (\mathbf{6.8(7)\cdot 10^7 \ a})$$

Among the p-process nuclides, ^{146}Sm is especially interesting in view of evidence for live ^{146}Sm in the Early-Solar System, established by isotopic anomalies of Nd in meteoritic material [104–106]. Earth crust samples themselves display an interesting anomaly in their ^{142}Nd isotopic abundance compared to bulk meteoritic material (chondrites) with a systematic difference [107, 108]. The α-decay of ^{146}Sm to ^{142}Nd constitutes in this case a suitable Solar-System clock, assuming that the half-life is known with sufficiently high precision. A summary of the half-life measurements available in literature can be found in Table 2.

Two values were reported in early 1950 and 1960s, with $5{\cdot}10^7$ a [109] and $7.5(15){\cdot}10^7$ a [110], respectively. Ref. [111] performed a new measurement in 1966 using isotope dilution mass spectrometry, yielding in a considerably longer half-life of $1.026(48){\cdot}10^8$ a, which was confirmed by Ref. [112] with excellent agreement. These authors used bombardment of ^{147}Sm with deuterons for sample production and determined the number of ^{146}Sm atoms via decay of ^{146}Eu. The most recent and simultaneously recommended half-life value, $6.8(7){\cdot}10^7$ a, based on direct measurement of the ^{146}Sm/^{147}Sm α-activity and atomic ratios [81], is shorter than the former ones and, interestingly, confirms the earlier value reported by Ref. [110]. This half-life value implies higher initial ^{146}Sm abundance in the early solar system than previously estimated. Terrestrial, Lunar, and Martian planetary silicate mantle differentiation events dated with ^{146}Sm-^{142}Nd converge to a shorter time span and in general to earlier times, due to the combined effect of the new ^{146}Sm half-life for such planetary processes, enhancing the importance of an accurate knowledge of the ^{146}Sm half-life value, see also Ref. [51]. The situation urgently calls for further measurements of the ^{146}Sm half-life.

Table 2: Reported measurements of the ^{146}Sm half-life.

Year of measurement	Value	Reference	Comment
1953	$5{\cdot}10^7$ a	[109]	Yield estimations relative to ^{145}Sm, α counting with emulsion films
1964	$7.5(15){\cdot}10^7$ a	[110]	
1966	$1.026(48){\cdot}10^8$ a	[111]	Isotope dilution MS, α counting with ionization chamber
1987	$1.031(45){\cdot}10^8$ a	[112]	Number of atoms via decay of ^{146}Eu, α spectrometry
2012	$6.8(7){\cdot}10^7$ a	[81]	AMS/α spectrometry relative to ^{147}Sm
	$6.8(7){\cdot}10^7$ a	[50]	Recommended value according to NDS

$$^{154}\mathbf{Dy} \quad (\mathbf{3.0(15)\cdot 10^6 \ a})$$

^{154}Dy is a pure α-emitter with a half-life of approx. 3 million years. It decays via a decay chain (^{150}Gd, ^{146}Sm) to stable ^{142}Nd, which directly influences the abundance of the natural isotopic composition of stable Nd and thus also the ^{146}Sm/^{142}Nd chronometer. Furthermore, ^{154}Dy is important in the synthesis of ^{142}Nd, ^{146}Sm, ^{152}Gd, and ^{158}Dy due to the nuclear reactions ^{154}Dy(α, γ)^{158}Er, ^{154}Dy(γ, α)^{150}Gd and ^{154}Dy(γ, 2n)^{152}Dy [88]. The half-life of ^{154}Dy remains associated with the largest relative uncertainty throughout the chart of nuclides despite the fact that similar isotopes like ^{150}Gd, which half-life is known with much lower uncertainty, are similarly difficult to obtain in sufficient quantity and radio-isotopic purity.

An overview of existing half-life determinations on ^{154}Dy is given in Table 3. The recommended value in NDS is based on the evaluation performed by Ref. [113] that took into account measurements performed by Ref. [114] and the revised values reported by Refs. [115, 116]. Based on the existing dataset, the knowledge about the ^{154}Dy half-life can be regarded as being rather vague. A re-determination of the ^{154}Dy half-life has been recently performed at PSI and will be published soon [117].

3.1.3 s-process: examples

$$^{60}\mathbf{Fe} \quad (\mathbf{2.62(4)\cdot 10^6 \ a})$$

The s-process nuclide ^{60}Fe can – due to the high-energetic γ-rays of its daughter nuclide ^{60}Co – be detected

Table 3: Half-life measurements of ^{154}Dy.

Year of measurement	Value	Reference	Comment
1961	10^6 a	[115]	Theoretical yield estimations, α-counting with ionization chamber
1965	$2.9(15){\cdot}10^6$ a	[114]	Yield estimation relative to ^{153}Dy/α spectrometry
1967	$10(4){\cdot}10^6$ a	[118]	
1971	10^7 a	[116]	Yield estimation relative to ^{153}Dy/α spectrometry
	$3.0(15){\cdot}10^6$ a	[58, 113]	Recommended value according to NDS

directly in space and is, therefore, an indicator for supernovae explosions. The evaluation of stellar processes requires the knowledge of all production and destruction paths, and one of these is the radioactive decay.

Until 2009, only one reliable half-life measurement had been performed by Ref. [119], resulting in a value of $1.49(27)\cdot10^6$ a, being considerably larger than the previous rough estimate of $5\cdot10^5$ a by Ref. [120]. The authors produced the sample by irradiating Cu with 191 MeV protons at the Brookhaven Linac Isotope Producer (BLIP) followed by a radiochemical separation of Fe from the irradiated matrix material. The final sample contained some 10^{14} atoms of ^{60}Fe. The activity was determined by following the ingrowth of the decay product ^{60}Co after a very careful separation of the initially produced ^{60}Co. The number of atoms was determined using AMS. The experimenters had to fight with problems of ^{60}Ni contamination in the AMS system stemming from Ni-containing components and additionally report possible losses of ^{60}Fe atoms due to unknown reasons. Attempts to determine the transmission by use of a ^{60}Co beam failed, so that the authors admit that their value could be too low. This thesis proved to be true when authors in Ref. [78] repeated the measurement with a factor 10 more material, extracted from an irradiated Cu beam dump of the high-power 590 MeV proton accelerator at PSI. Similar to the approach by Ref. [119], the activity was determined by the ingrowth of ^{60}Co, but the number of atoms was determined by Multi-Collector Inductively-Coupled-Plasma Mass Spectrometry (MC-ICP-MS). In contrary to AMS, this method allows determining absolute values for the content of isotopes in a sample. The obtained value of $2.62(4)\cdot10^6$ a differed with respect to the one reported before nearly by a factor of two. Two further measurements, also performed with material from PSI, yielded $2.50(12)\cdot10^6$ a [20] and $2.69(28)\cdot10^6$ a [21], respectively. Authors in Ref. [20] also used the ingrowth of ^{60}Co for the activity measurement, but AMS for the number of atoms, in which they measured the ^{60}Fe relative to ^{55}Fe, thus circumventing the difficulties with the absolute transmission. Authors in Ref. [21] measured the activity using the γ-ray of ^{60m}Co, which is in secular equilibrium already after a couple of hours after separation. This is possible only with very old samples, because the simultaneous production of ^{55}Fe during irradiation of the copper, resulting in a huge background caused by Bremsstrahlung at lower energies. The number of atoms in this work was determined by AMS relative to the values obtained in Ref. [20]. The present weighted mean half-life value substantially improves the reliability of ^{60}Fe as an important chronometer for astrophysical applications. A summary of the performed measurements is given in Table 4.

Table 4: Reported measurements of the ^{60}Fe half-life.

Year of measurement	Value	Reference	Comment
1957	$5\cdot10^5$ a	[120]	
1984	$1.49(27)\cdot10^6$ a	[119]	AMS/γ-spectrometry of ^{60}Co ingrowth
2009	$2.62(4)\cdot10^6$ a	[78]	ICP-MS/γ-ray spectrometry of ^{60}Co ingrowth
2015	$2.50(12)\cdot10^6$ a	[20]	AMS (^{60}Fe/^{55}Fe ratio)/γ-ray spectrometry of ^{60}Co ingrowth
2017	$2.69(28)\cdot10^6$ a	[21]	AMS from Ref. [20]/γ-ray spectrometry of ^{60}Co from ^{60m}Co decay
	$2.62(4)\cdot10^6$ a	[19]	Recommended value according to NDS

The impact of this corrected ^{60}Fe half-life is manifold. The measurement of the neutron capture cross section [121] at stellar energies performed in 2009 had to be corrected according to the new half-life because the number of atoms in the sample was determined by activity measurements. A very high impact has to be expected for all results obtained so far by AMS. Since a certified ^{60}Fe standard material was missing for a long time, all relative measurements used surrogates produced in nuclear reactions with heavy ions [122], at ISOLDE [123] or the sample material from the half-life measurement of Ref. [119]. New standard material, characterized by MC-ICP-MS has been produced from the PSI copper beam dump and was already used for studies demonstrating the occurrence of a super nova explosion 2.6 million years ago [124–126]. For further progress in this field, the correct value of the ^{60}Fe half-life is definitely a key parameter.

^{186m}Re $(2\cdot10^5$ a$)$

The isotope ^{186m}Re is of particular interest from the astrophysical point of view, since a new s-process path of ^{185}Re(n, γ)^{186m}Re(n, γ)^{187}Re could change the accuracy of a ^{187}Re-^{187}Os cosmo-chronometer [127]. The 149 keV level excited state of ^{186}Re decays by internal transition to the ground state with an accepted half-life of $2\cdot10^5$ a originating from one measurement without given uncertainty of the value [128]. The measurements were performed on neutron irradiated rhenium metal. Specific activity of ^{186m}Re in the sample was determined by combination of mass spectrometry and γ-ray spectrometry on Ge(Li) detector yielding a value of $2\cdot10^5$ a. An additional measurement with beta counting of the equilibrated ground state of ^{186}Re with a proportional counter yielded a half-life of $1.7\cdot10^5$ a as presented by authors in Ref. [128].

3.2 Cosmogenic radionuclides

3.2.1 Closing the nuclear dating gap – ^{32}Si (153(19) a) and ^{39}Ar (268(8) a)

The radioactive isotopes ^{32}Si with a half-life of 153(19) a [10] and ^{39}Ar with 268(8) a [13] are cosmogenic nuclides that are produced in the upper atmosphere by bombardment of cosmic rays on argon. They are the only two available radionuclides with the potential to fill the dating gap between the relatively short-lived ^{210}Pb ($T_{1/2}$ = 22.20(22) a) on the one side and the longer-lived ^{14}C ($T_{1/2}$ = 5730(40) a) on the other side, thus allowing to understand environmental processes such as glacier dynamics, ocean and atmospheric circulation, sedimentation in lakes and oceans or groundwater flow in the recent past (100–1000 years), see Figure 2. The main drawback of both, ^{32}Si and ^{39}Ar, is the lack of accurate knowledge on their half-lives and their extreme low abundance in environmental samples.

Authors in Ref. [129] performed a critical evaluation of the data pool for ^{32}Si given in Figure 3. They calculated an average value of 144 years with >10% uncertainty by applying several mathematical treatments (for details see the descriptions and references in Ref. [129]). Nevertheless, only three of the data points actually meet the estimated uncertainty band. Moreover, the weighted average value does not necessarily represent the best estimator in this case, and the associated uncertainty is questionable. The ENSDF database recommends a ^{32}Si half-life of 157(7) a, which is mainly impacted by the most recent measurement performed in the frame of a PhD thesis in 2015 [11]. It is of high importance to perform additional careful measurements in order to increase certainty about the true ^{32}Si half-life.

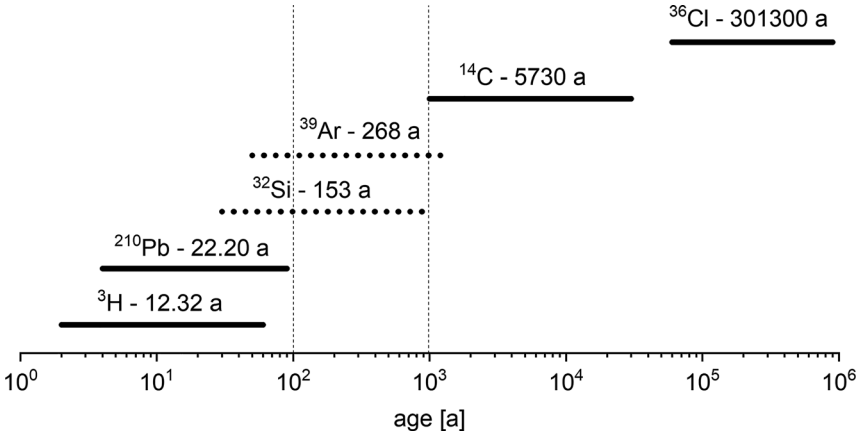

Figure 2: Radionuclides for nuclear dating up to 10^6 years.

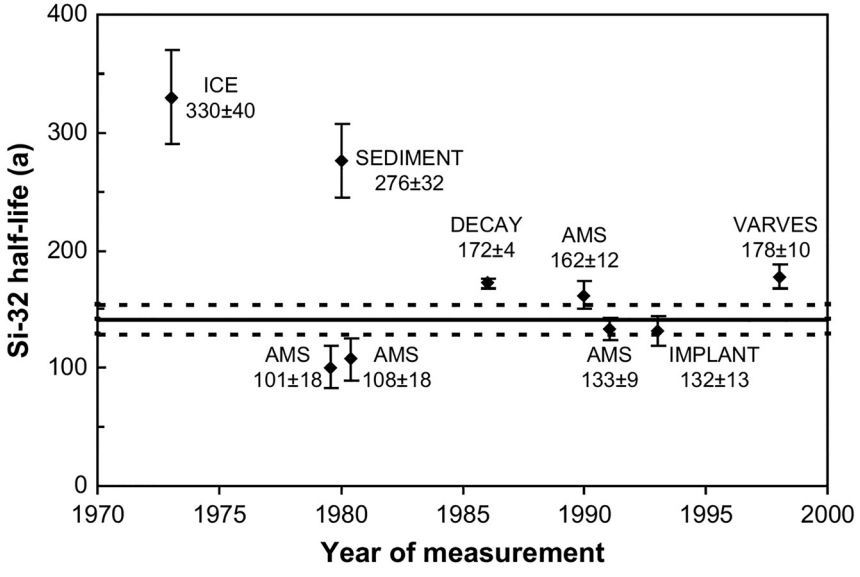

Figure 3: Compilation of ^{32}Si half-life determinations. The solid line represents the mean value of 144 years of all the determinations with exception of the two early stratigraphic measurements made on ice and sediment. The broken lines represent the uncertainty of ±11 years of the mean value. Figure and caption taken from Ref. [129], reproduced with kind permission of Elsevier.

The situation is slightly different for ^{39}Ar. The currently applied value for its half-life is based on two measurements, both performed more than 50 years ago, yielding 265(30) a [130] and 269(3) a [131], respectively. Nuclear Data Sheets recommends the adopted latter value with an enlarged uncertainty [13]. A third publication, claiming 276(3) a [14] has not been included in the evaluation, maybe due to the fact that the authors announced further improvement of the measurement, but they never published it. The nearly perfect agreement of the two measurements pretends a high degree of reliance, and the value is therefore willingly accepted in the community. However, in the performed work, mixtures of ^{37}Ar, ^{39}Ar and ^{42}Ar isotopes were used, produced by both spallation and neutron activation. While the mass spectrometry used to determine the number of atoms seems to be feasible, the activity determination suffers from the difficulty to distinguish between the two long-lived pure β-emitters ^{39}Ar and ^{42}Ar with nearly the same β-energy. Moreover, contamination of the samples with natural Ar can hardly be quantified. Therefore, a re-measurement of the ^{39}Ar half-life is a precondition before this isotope can seriously be considered as a dating tool. For the shorter-lived ^{42}Ar isotope, the accepted half-life value of 32.9(11) a nowadays still relies on one single measurement reported in literature [131].

A joint project involving several Swiss institutes (PSI, CHUV-IRA, ETHZ, Laboratory Spiez) as well as the PTB in Germany is currently underway to re-determine the ^{32}Si half-life by utilizing several independent methods for measuring both the activity and the number of atoms. High quality samples with activities up to 20 MBq were obtained by irradiation of Vanadium metal for two years in the target of the PSI neutron facility SINQ and subsequent radiochemical separation and purification [132]. By means of this, we hope to overcome the obstacle of the inconsistent database and, thus, pave the way towards the implementation of ^{32}Si as a nuclear dating tool in environmental research.

3.2.2 Longer-lived dating radionuclides

$$\mathbf{^{81}Kr \quad (2.29(11)\cdot 10^{5} \quad a)}$$

The nuclide ^{81}Kr is produced by cosmogenic induced proton and neutron spallation reactions in the atmosphere on stable Kr isotopes [133]. Due to its stable concentration in the atmosphere it is a desirable dating nuclide for the time range between $5{\cdot}10^4$ a and $2{\cdot}10^6$ a.

Two half-life determinations exist up to date. In an early investigation by authors in Ref. [134], neutron irradiated NaBr was used as source for ^{81}Kr, where its activity was determined using a proportional counter and isotopic composition by mass spectrometry. While a very limited amount of experimental data was made public, a half-life of $2.1(5){\cdot}10^5$ a has been deduced. In the work of Ref. [135], the ^{81}Kr sample was prepared by neutron irradiation of natural krypton gas. The number of ^{81}Kr atoms was determined by combination of integrated current measurements during ^{82}Kr mass separation and subsequent mass spectrometric measurements of the $^{82/81}$Kr atomic mass ratio. The activity of the sample was measured using a NaI detector equipped with a beryllium window for detection of the 12 keV bromine K-shell X-rays. The deduced half-life of $2.13(+16{-}26){\cdot}10^5$ a was calculated as an average of six different measurements taking into account the uncertainty of the K-shell electron capture branching ratio for ^{81}Kr. The random and systematic uncertainty were reported to be around 2% and 3%, respectively. The currently accepted value of $2.29(11){\cdot}10^5$ a was adjusted by the evaluator taking into account an updated K-shell fluorescence yield and electron capture ratio [26].

$$\mathbf{^{107}Pd \quad (6.5(3)\cdot 10^{6} \quad a)}$$

The isotope ^{107}Pd is used as a chronometer in the ^{107}Pd/^{107}Ag system for the dating of iron meteorites [136]. The ^{107}Pd is produced in nuclear reactors being a long-lived fission product decaying by emission of β-particle. Two measurements of its half-life have been performed so far. In the most recent work performed by Ref. [137], the half-life of ^{107}Pd was obtained from specific activity measurements of a ^{107}Pd sample combined with the determination of its isotopic composition. About 160 μg of palladium were chemically separated from uranium fission products and then cooled down for 14 years before the measurement. The relative abundance of ^{107}Pd was determined by mass spectrometry. Activity measurements were performed on three aliquot samples counted by an internal proportional counter. The derived half-life value $6.5(3){\cdot}10^6$ a is an averaged value from activity measurement of the three aliquots. In an earlier measurement of Ref. [138], ^{107}Pd was separated from uranium fission products and its specific activity was measured on a proportional counter. The total ^{107}Pd isotopic ratio was assumed to be 20% of total determined of Pd amount. Three half-life values were estimated from three different samples yielding 6.6, 7.3 and $8.5{\cdot}10^6$ a. An additional independent measurement of the ^{107}Pd half-life with lower uncertainty would improve the accuracy of the ^{107}Pd/^{107}Ag chronometer and other dating techniques using this isotope.

^{202}Pb $(5.25(28)\cdot10^{4}$ a) and ^{205}Pb $(1.70(9)\cdot10^{7}$ a)

Long-lived isotopes of Pb have been occasionally used for dating of meteorites and other chronology applications. The $^{202/205}$Pb pair has been employed in precision mass spectrometry to correct for instrumental mass fractionation [139]. The isotope ^{205}Pb is also interesting for astrophysical research since it is produced only via s-process nucleosynthesis [140]. Moreover, the inverse β-decay of ^{205}Tl to ^{205}Pb could serve as probe for the solar neutrino flux [141].

Two publications have been reported on the determination of the ^{202}Pb half-life. Authors in Ref. [142] produced ^{202}Pb by deuteron bombardment of natural Tl with subsequent chemical separation and estimated a half-life of $3\cdot10^{5}$ a based on several assumptions regarding the cross section of the ^{203}Tl(d,3n)^{202}Pb reaction and X-ray emission yields. In a more detailed investigation by Ref. [143], a stacked target consisting of Tl_2O_3 and Al monitor foils was irradiated with 52 MeV protons and ^{202}Pb subsequently extracted by liquid-liquid extraction and anion exchange. The ingrowth of the ^{202}Tl activity was then followed for 150 d to estimate the total activity concentration of the obtained ^{202}Pb solution. With the addition of mass spectrometry, a half-life of $5.25(28)\cdot10^{4}$ a was obtained, which remains the accepted value up to date [68].

Authors involved in the first measurement of ^{202}Pb have later also reported on the determination of the ^{205}Pb half-life [144]. They performed extended neutron irradiations of enriched ^{204}Pb with subsequent chemical purification to enable X-ray counting and mass spectrometric analysis of the obtained samples. The reported value of the partial L-electron capture half-life was $3.0(5)\cdot10^{7}$ a, which was later adjusted by the evaluator taking into account an updated L-shell fluorescence yield and the electron capture branching ratio [69]. The currently accepted value of $1.70(9)\cdot10^{7}$ a therefore, relies on only one single experimental determination performed in 1958 and is associated with a highly questionable uncertainty budget.

3.3 Nuclear energy and safety

^{93}Mo $(4.0(8)\cdot10^{3}$ a)

The isotope ^{93}Mo is mainly produced in nuclear power plants by activation of molybdenum rich steels used for the primary cooling circuit and the reactor vessel. The ^{93}Mo is one of the long-lived radioisotopes in the low and medium level radioactive waste being the predominant radioactive dose contributor after the shorter-lived isotopes have decayed. Its currently accepted half-life of $4.0(8)\cdot10^{3}$ a originates from one single indirect determination. In the work of Ref. [145], ^{93}Mo was produced by 21 MeV deuteron bombardment of niobium metal. With the assumption that the cross section for (d, 2n) reactions for odd-even nuclei is constant in the mass range A = 85–109, the number of produced ^{93}Mo atoms was estimated from the integral current of irradiation. After chemical separation, the activity of ^{93}Mo was measured by means of X-ray spectrometer using the K-shell emission lines of Nb. The deduced half-life of $3.0(5)\cdot10^{3}$ a was later adjusted by the evaluator to the currently accepted value of $4.0(8)\cdot10^{3}$ a taking into account updated K-shell X-ray emission probability for the EC decay of ^{93}Mo [27].

In a very recent re-determination performed at PSI, ^{93}Mo was extracted and thoroughly purified from proton irradiated Nb. Liquid scintillation counting of the ^{93}Mo activity concentration coupled with mass spectrometry of the obtained sample yielded a new half-life value of 4839(63) a [32].

^{79}Se $(3.27(28)\cdot10^{5}$ a)

The fission product ^{79}Se is a pure low-energetic β-emitter produced in nuclear fission with a rather low cumulative fission yield of 0.048%. However, due to its comparable long half-live of more than 10^{5} years it is one of the few radionuclides remaining for evaluation of the long-term radiological impact in final repositories. Moreover, ^{79}Se is – similar to ^{60}Fe – one of the branching points in the slow stellar element synthesis processes. For both application fields, the exact knowledge of the half-life is crucial.

The half-life measurements of ^{79}Se are a very good example for the dilemma, which can arise by performing new measurements. Up to the early 1990s only two rough estimations existed, which were extremely contradicting: $\geq7.0\cdot10^{6}$ a [146] and $\leq6.5\cdot10^{4}$ a [138], respectively. The latter value was later corrected to $\leq6.5\cdot10^{5}$ a by the NDS evaluator [147] after detecting a calculation error in the work of Ref. [138], but this did not really solve the problem. Several measurements were then performed from 1995 on, resulting in an very inconsistent dataset. The work by Ref. [80] gives a very good overview on the difficult situation and ends with the remarkable sentence: “Responsible authorities in charge of compendia should carefully review and evaluate especially the more up-to-date literature on the ^{79}Se half-life in order to come to an agreement as to which value internationally should be officially accepted and presented in tables.”

In Figure 4, we summarize the existing data about the ^{79}Se half-life. The values of around $1\cdot10^{6}$ a $1.1(2)\cdot10^{6}$ a [148] and $1.12(17)\cdot10^{6}$ a [149] were both determined using

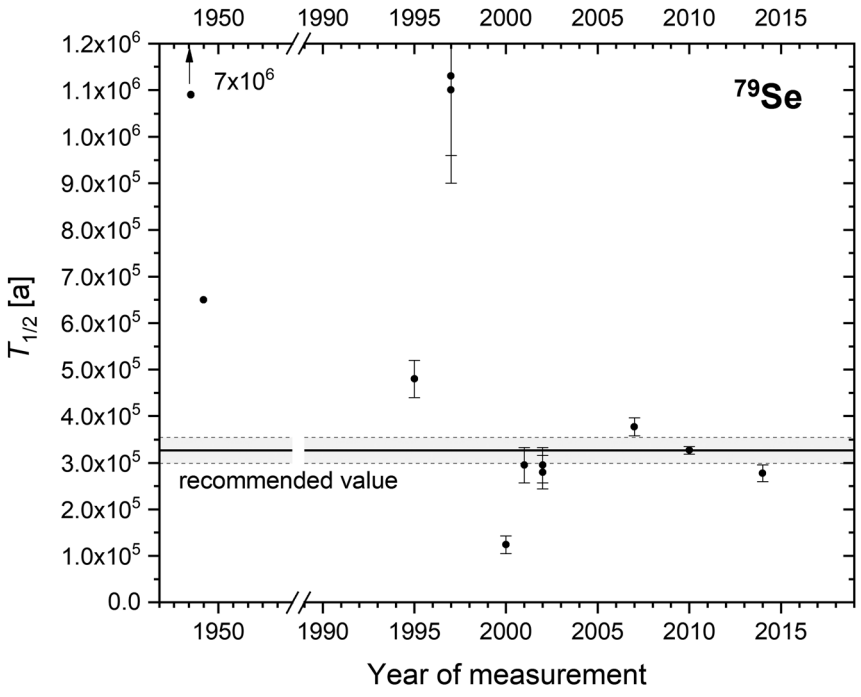

Figure 4: Overview of ^{79}Se half-life measurements with the weighted mean taken from Ref. [25] and its 1σ standard deviation.

AMS and liquid scintillation counting (LSC). Authors in Ref. [150] obtained 1.24(19)·10^5 a using projectile X-ray (PX)-AMS, a value which is a factor 10 lower than reported by Ref. [148]. This group repeated their measurement in 2001 and 2002, obtaining now two identical – however a factor 3 lower – values of 2.95(38)·10^5 a, using both AMS and PX-AMS [151, 152]. Interestingly, authors in Ref. [153] came up with a second value in the same year, now a factor 3 higher and confirming the value reported by Refs. [151, 152] (2.80(36)·10^5 a). From these findings, it seems to be clear that both the 1·10^6 a and the 1.24(19)·10^5 a values can be ruled out. Another value of 4.8(4)·10^5 a, reported by Ref. [154], used LSC and fission yield estimates relative to ^{137}Cs and ^{90}Sr to determine the number of atoms in their sample.

The three measurements from 2001 to 2002 would have given a consistent picture, but in 2007 [79] published a value of 3.77(19)·10^5 a, obtained using liquid scintillation counting and ICP-MS. The next value reported in 2010 by Ref. [80], 3.27(8)·10^5 a, where the number of atoms was also determined by ICP-MS, has the lowest uncertainty. It is in agreement with the values obtained in 2001/2002 by Refs. [151, 152] but not with any other. Because of an extremely careful sample treatment as well as highly precise and accurate measurements, both of the activity and the number of atoms, the value of Ref. [80] seemed the most reliable. However, further measurements were necessary to confirm the value. Unfortunately, the most recent work by Ref. [155] (2.78(18)·10^5 a) using AMS, confirms again the value of Ref. [151, 152]. The authors of this work discuss a probably wrong evaluation of the atomic abundances in the Refs. [79, 80] determinations due to erroneous correction of the ^{79}Br interference in the ICP-MS measurement. On the other hand, AMS measurements generally suffer from an insufficient knowledge of the total transmission. It is conspicuous that the AMS values underestimate the number of ^{79}Se atoms during measurement, whereas an overestimation using ICP-MS cannot be excluded. However, the weighted mean value given in the Nuclear Data Sheets in 2016 of 3.27(28)·10^5 a [25], calculated based on the measurements of Refs. [79, 80, 155], seems to be reasonable with respect to the shortcomings of the earlier determinations. Nonetheless, further measurements are necessary for confirmation.

^{135}Cs $(2.3(3)\cdot 10^6\ \mathrm{a})$

The isotope ^{135}Cs is one of the long-lived fission products, which might have a high impact on long-term storage of nuclear waste. Caesium as a member of the alkaline elements is highly soluble in water solutions, posing a special risk of contamination of the biosphere. Due to this outstanding water solubility, on the other hand, anthropogenic-produced Cs isotopes (e.g. the ratio of ^{135}Cs/^{137}Cs) can be used to study the time line of oceanographic flow, for dating of sediments and to reconstruct the history of nuclear contamination events. Moreover, ^{135}Cs is, like most of the long-lived fission products, a branching point in the s-process. For all these applications, the exact as possible knowledge of the half-life is a pre-condition.

Table 5: Half-life measurements of ^{135}Cs.

Year of measurement	Value	Reference	Comment
1949	$2.1(7)\cdot10^6$ a	[156]	Proportional counter/fission yield calculated from ^{135}Xe
1949	$2.9(3)\cdot10^6$ a	[157]	Proportional counter/fission yield calculated from ^{135}Xe
1955	$2.0(2)\cdot10^6$ a	[158]	Proportional counter/fission yield calculated from ^{135}Xe; γ-ray spectrometry of ^{137}Cs
2016	$1.6(6)\cdot10^6$ a	[48]	LSC/AMS
	$1.3(2)\cdot10^6$ a		LSC/ICP-MS
	$2.3(3)\cdot10^6$ a	[47]	Recommended value according to NDS

Very astonishingly, there are only a few measurements so far, including a recent one from 2016 by Ref. [48], which are in obvious disagreement. Table 5 provides a summary of the reported half-life determinations of ^{135}Cs.

The main problem associated with half-life determinations of this isotope is very probably the radionuclidic purity of the sample. Caesium isotopes are formed in nuclear reactors via the decay of the corresponding Xe isotopes and ^{137}Cs ($T_{1/2}$ = 30.08(5) a) is one of the main dose contributors in spent nuclear fuel. Furthermore, ^{135}Cs is always accompanied by ^{134}Cs ($T_{1/2}$ = 2.0652(4) a). All three isotopes are β-emitters with similar energies, making liquid scintillation counting problematic for accurate activity determinations. Authors in Ref. [48] tried to avoid contamination of ^{135}Cs by using a special technique for collecting Xe from a nuclear reactor after several hours of decay. This allows increasing the abundance of ^{135}Cs by orders of magnitude. Selective Cs adsorption was used to guarantee a high purification factor from other disturbing β-emitters like ^{90}Sr and ^{90}Y. Moreover, the authors used two independent methods for the determination of the number of atoms. Both determinations yielded half-life values significantly lower than previously reported in literature. Considering the manifold impact areas of the ^{135}Cs half-life, the situation urgently calls for further independent measurements.

^{126}Sn $(2.30(14)\cdot10^5$ a$)$

The tin isotope ^{126}Sn is mostly produced during nuclear fission in nuclear power plants. Due to its considerably long half-life, it is an important nuclide with respect to processing and waste management of irradiated fuel. Six half-life measurements have been performed so far, all these works used samples extracted from fission products of ^{235}U as a starting material. The currently accepted value of $2.30(14)\cdot10^5$ a in Ref. [41] originates from the weighted average of two measurements $2.07(21)\cdot10^5$ and $2.5(2)\cdot10^5$ a, both performed in 1996. In the work of Ref. [159], ^{126}Sn was produced by neutron irradiation of ^{235}U target in the nuclear reactor and the Sn fraction was chemically separated. The number of ^{126}Sn atoms in the sample was calculated based on the fission yield for ^{126}Sn, and the number of fissions was monitored through ^{90}Sr and ^{137}Cs produced in the uranium target. Finally, the activity of separated ^{126}Sn was measured by γ-ray spectrometry using a HPGe detector. A similar approach was chosen by authors in Ref. [160], but accelerator mass spectrometry was applied for the determination of the number of ^{126}Sn atoms, yielding $2.07(21)\cdot10^5$ a. A thermal ionization mass spectrometry (TIMS) measurement by authors of Ref. [42] on the same sample, however, yielded a revised half-life of $2.345(71)\cdot10^5$ a.

Further independent measurements undertaken by authors in Refs. [43, 44] obtained contradicting results that are either confirming or disproving the currently recommended value, see Table 6. This situation significantly complicates the judgement about the ^{126}Sn half-life certainty. Additional careful determinations using alternative activity measurements in combination with multiple mass spectrometric techniques are highly recommended.

^{194}Hg $(447(52)$ a$)$

The long-lived isotope ^{194}Hg, decaying by EC to the γ-ray emitter ^{194}Au, is considered to be one of the major dose contributors in spallation neutron sources operating with mercury such as the Spallation Neutron Source (SNS) in Oakridge (USA) or the Japan Proton Accelerator Research Complex (J-PARC) in Riken (Japan). Accelerator waste from these facilities containing mercury will be of major concern during decommissioning and final storage. Additionally, the formation of volatile mercury isotopes is a considerable point of attention in accelerator driven subcritical reactors, in which ^{194}Hg is formed by charged particle induced spallation of Pb and/or Bi.

The very recently revised half-life value of 447(52) a substantially changed over the past 60 years and still nowadays remains subjected to considerable uncertainty. While early investigations indicated a half-life of around 130 d [162], which was confirmed by authors in Ref. [163], the former research group reported a revised value of 600 d

Table 6: Half-life measurements of ^{126}Sn.

Year of measurement	Value	Reference	Comment
1962	10^5 a	[161]	Estimated from ^{235}U fission yields
1996	$2.5(2)\cdot10^5$ a	[159]	γ-ray spectrometry/^{235}U fission yield estimation
1996	$2.07(21)\cdot10^5$ a	[160]	γ-ray spectrometry/AMS
1999	$2.345(71)\cdot10^5$ a	[42]	TIMS on sample of Ref. [160]
2005	$2.33(10)\cdot10^5$ a	[43]	γ-ray spectrometry/ICP-MS
2009	$1.980(57)\cdot10^5$	[44]	γ-ray spectrometry/ICP-MS
	$2.30(14)\cdot10^5$ a	[41]	Recommended value according to NDS

[164]. Subsequent publications on this subject gave 146(6) d [165], 700(100) d [166] and 426(50) d [167] (Figure 5).

Later experiments showed evidence that the half-life of ^{194}Hg is considerably longer. Measurements done by Ref. [168] with efforts to minimize possible evaporation losses of mercury yielded a half-life in the range between 90 and 540 a. It was not until around 1980 before investigations with improved techniques were undertaken. Analysing production yields of Hg isotopes produced via ^{197}Au(p, xn)$^{195-x}$Hg reactions, authors in Ref. [169] deduced a half-life of 367(55) a, which supersedes the value of 260(40) a reported earlier by the same group. Finally, researchers in Ref. [170] mass separated ^{194}Hg produced by spallation of 600 MeV protons on liquid lead at CERN ISOLDE. The activity of three collected samples was measured using a Ge(Li) detector, the total number of atoms was deduced from current measurements using Faraday cups during implantation. They deduced a half-life of 520 a, with a random and systematic uncertainty of 20 and 25 a, respectively.

The recommended half-life of ^{194}Hg given by Nuclear Data Sheets is a weighted average of measurements of Refs. [169, 170], whereas both reported values were adjusted taking into account adopted emission probabilities of the 328.5 keV line in ^{194}Au decay [67]. New independent measurements of the ^{194}Hg half-life are urgently required in order to increase the confidence in the stated value and its uncertainty.

^{209}Po (125.2(33) a)

Until 2005 the only known half-life value for ^{209}Po of 102(5) a was measured by Ref. [171] relative to the well-known ^{208}Po half-life using a source produced by proton irradiation of Bi. The measurement was based both on the activity and mass ratios determination of the two isotopes. In 2007, colleagues from NIST reported for the first time on

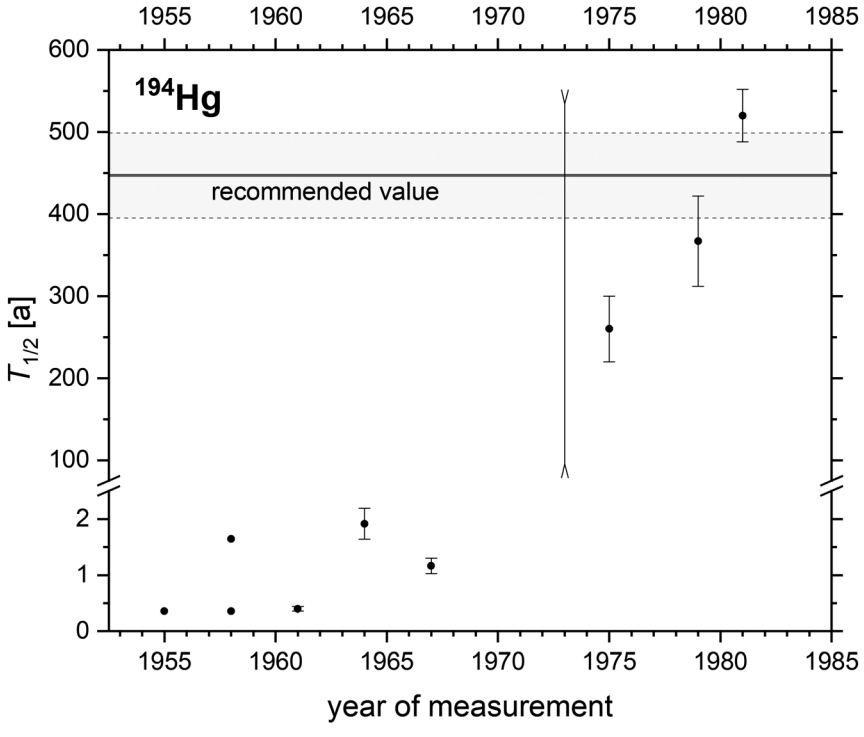

Figure 5: Overview of ^{194}Hf half-life measurements with the weighed mean as given by Nuclear Data Sheets [67].

a possible deviation of up to 25% from this adopted value due to observed inconsistencies in the regular examination of their standard materials over a wide time span [172]. In the following years, extended studies with large sets of samples were performed using liquid scintillation counting. The authors derived an improved value of 125.2(3.3) a [173], based on measurements dating back to 1993. Authors in Ref. [174] performed a complementary decay measurement using α-spectrometry with PIPS detectors over a time span of 400 days, yielding 120(6) a. This measurement was primarily aimed to solve the problem with the two contradicting former values. The authors announced to continue the experiment in order to get results that are more precise. The currently accepted value of 125.2(33) a is a weighted average of both most recent determinations [75].

^{209}Po is mainly used as calibration standard for alpha particle energy and alpha-emission rate measurements, and as low-level tracer and separation yield monitor in radiochemical procedures for environmental measurements and geophysical studies. Many environmental data on ^{210}Po rely on the determination relative to ^{209}Po. The fact that the former ^{209}Po half-life value of 102(5) a has to be considered as erroneous has an immense impact on a huge number of data generated over more than 50 years. Another independent measurement, maybe using the approach based on Eq. (1), would be of substantial benefit for the nuclear science community and beyond.

4 Conclusions

This review article makes no claim to completeness. In view of the large number of nuclides and the correspondingly necessary measurements, this is not even approximately possible. Instead, we wanted to point on the problems and the resulting consequences by means of some examples. For selected radionuclides, we discussed recent measurements and their impact in more detail. Our general intention is to sensitize the nuclear data community for the urgent need of reliable decay data and the difficulties associated with the performance of new measurements. Especially in cases where several conflicting measurements exist, assessing the situation is sometimes difficult and deriving a reliable value is practically impossible. Examples for this are ^{32}Si or ^{79}Se. On the other hand, we highlight examples, where remarkable progress could be achieved recently, e.g. ^{60}Fe and ^{93}Mo. It becomes clear, that the importance of the quality of the available sample cannot be emphasized enough. In particular, EC nuclides with no accompanying α, β or γ-ray emission (^{93}Mo, ^{157}Tb) are particularly challenging for the activity determination, i.e. they must be present in radio-chemically and isotopically ultra-pure form. This can be achieved either by dedicated production routes (e.g. ^{93}Mo, ^{137}La) or by a follow-up mass separation, as has been performed for the recent measurement of the ^{53}Mn half-life at PSI [to be published]. As an outcome of this review, we present here the inventory of radioisotopes shortlisted from Table 1, which half-life values urgently call for re-measurement. The selection of the shortlisted radionuclides is given in Table 7 and was based on the following criteria:

- The recommended value is based on one measurement only and is not confirmed by a new measurement performed after the last issue of NDS.
- The uncertainty of the recommended half-life value is higher than 5% unless a new confirming value with a lower uncertainty was reported.
- The two most recent independent determinations are not compatible within the stated uncertainty boundaries.

Table 7: Radionuclides recommended by the authors for a half-life re-determination based on different criteria. Several criteria might apply for the same nuclide.

Nuclide	Reason for re-measurement
^{42}Ar, ^{91}Nb, ^{137}La, ^{178m2}Hf, ^{186m}Re, ^{192m2}Ir, ^{205}Pb	Only one measurement
^{32}Si, ^{53}Mn, ^{59}Ni, ^{79}Se, ^{92}Nb, ^{98}Tc, ^{135}Cs, ^{146}Sm, ^{157}Tb, ^{158}Tb, ^{154}Dy, ^{193}Pt, ^{194}Hg, ^{202}Pb	Uncertainty >5%
^{97}Tc, ^{107}Pd, ^{121m}Sn, ^{126}Sn, ^{207}Bi, ^{208}Bi, ^{210m}Bi	Values not within stated uncertainty boundaries

Another problem, not directly addressed in this paper, is the reliability of the presented uncertainties attributed to the half-life measurements. Many of the half-life measurements from the 50 and 60s have poorly documented or incomplete uncertainty budgets thus calling for the re-measurement. Therefore, the 5% threshold value arbitrarily used in this paper might not be an appropriately suited criterion.

Similar considerations as for the half-lives have also to be undertaken for branching ratios and emission probabilities, which are mandatory for using radiation measurement techniques for basic nuclear research, i.e. for cross section and half-life measurements in general, but also for the quantitative determination of the radionuclide content in technical, artificial and environmental samples. Considerable effort has still to be put in force in order to improve knowledge of nuclear data for long-lived radionuclides.

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