Radiocarbon, Vol 61, Nr 5, 2019, p 1135–1142 DOI:10.1017/RDC.2019.26
Selected Papers from the 23rd International Radiocarbon Conference, Trondheim, Norway, 17–22 June, 2018

THE HALF-LIFE OF ^{14}C—WHY IS IT SO LONG?

Walter Kutschera

University of Vienna, Faculty of Physics – Isotope Physics, Vienna Environmental Research Accelerator (VERA), Währinger Str. 17, A-1090 Vienna, Austria

ABSTRACT. The half-life of radiocarbon (^{14}C) is 5700 ± 30 yr, which makes it particularly useful for dating in archaeology. However, only an exceptional hindrance of the beta decay from ^{14}C to ^{14}N—a so-called Gamow-Teller ß-decay—makes this half-life so long. A normal strength would result in a half-life of only a few days, completely useless for archaeological dating. The unusual hindrance is based on the nuclear structure of the two nuclei, resulting in strongly destructive interferences of the nuclear transition matrix element. Nuclear model calculation with great computational efforts have been performed in the literature to reproduce the very low transition probability. Here, we will attempt to describe the nuclear physics behind this most unusual half-life.

KEYWORDS: half-life, nuclear structure, radiocarbon dating.

INTRODUCTION

Development of ^{14}C Half-Life Measurements

In 1946, Willard Libby published a brief paper (Libby 1946), in which he discussed the possibility that radiocarbon (^{14}C) produced via the ^{14}N(n,p)^{14}C reaction in the atmosphere may enter the biosphere in detectable amounts. His estimate was based on the number of neutrons liberated in the free atmosphere by spallation of nuclei through cosmic-ray interaction (Korff and Hamermesh 1946). At that time the half-life of ^{14}C was known to be in the range from 10^3 to 10^5 yr (Ruben and Kamen 1941). Later in 1946, two half-life measurements of ^{14}C were published, 4700 ± 470 yr (Reid et al. 1946), and 5300 ± 800 yr (Norris and Inghram 1946), respectively. In a first attempt to detect cosmogenic ^{14}C in biological carbon, biological methane was extracted from a sewage disposal plant in Baltimore (Anderson et al. 1947). In order to get a measurable signal from the rare ß-decays of ^{14}C, the methane was isotopically enriched by thermal diffusion up to a factor 260. Later, with an improved ß-counting system, radiocarbon could be measured without enrichment, and an average activity value of 12.5 ± 0.2 counts per minute per gram of carbon (c/m/g) was found for contemporary wood samples from different regions around the world (Libby et al. 1949). In addition, wood samples from two tombs of the Old Kingdom in Egypt estimated to be around 4600 years old were measured. With a half-life of 5720 ± 47 yr (Engelkemeir et al. 1949), an expected ^{14}C activity of 7.15 ± 0.15 c/m/g was calculated, whereas the weighted mean of several ^{14}C measurements on the wood samples was 7.04 ± 0.20 c/m/g (Libby et al. 1949). The good agreement encouraged Libby to measure also younger samples of known age, leading to the well-known decay curve shown in Figure 1 (Arnold and Libby 1949, see Figure 1). This, then, was really the starting point of ^{14}C dating. In Figure 1, the decay curve was calculated with the ^{14}C half-life of 5720 ± 47 yr (Engelkemeir et al. 1949). Later, end and wall corrections of the gas counter were applied, which lowered the half-life to 5580 ± 45 yr (Engelkemeir and Libby 1950). In 1952, Libby proposed a half-life value of 5568 ± 30 yr (Libby 1952), as the weighted mean of three values, 5580 ± 45 yr (Engelkemeir et al. 1949; Engelkemeir and Libby 1950), 5589 ± 75 yr (Jones 1949), and 5513 ± 165 yr (Miller et al. 1950).

Corresponding author. Email: walter.kutschera@univie.ac.at.

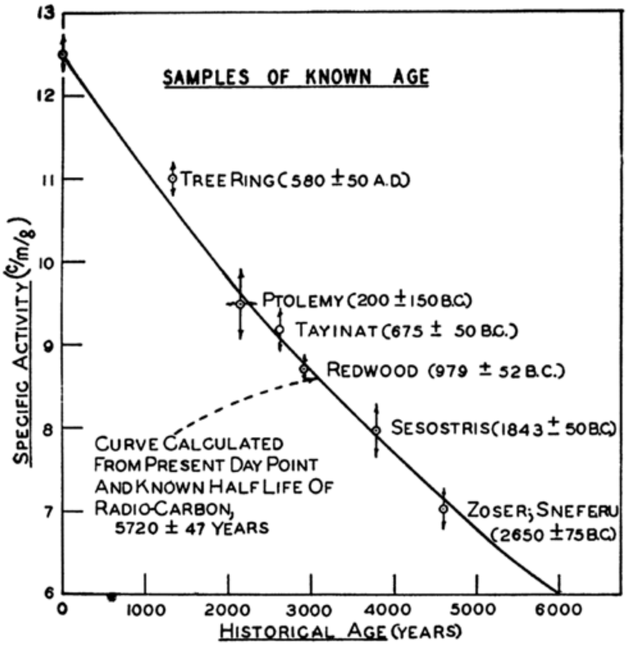

Figure 1 The first proof that ^{14}C dating does work (Arnold and Libby 1949).

However, at the Fifth Radiocarbon Conference in Cambridge 1962, a new value of 5730 ± 40 yr was agreed upon for the half-life of ^{14}C (Godwin 1962). This was based on the weighted mean of three new half-life measurements: 5760 ± 50 yr (Mann et al. 1961), 5780 ± 65 yr (Watt et al. 1961), and 5680 ± 40 (Olsson et al. 1962). It is interesting to note that this value is very close to the original half-life used for the decay curve in Figure 1, but is 2.9% longer than the "Libby half-life" of 1952.

Finally, in 1968 a new half-life measurement was published by Bella et al. (1968), and a weighted mean of 5700 ± 30 yr was calculated (Kutschera 2013) from the results of four half-life measurements: 5780 ± 65 yr (Watt et al. 1961), 5680 ± 40 yr (Olsson et al. 1962), 5745 ± 50 yr (Hughes and Mann 1964), and 5660 ± 40 yr (Bella et al. 1968).

In 2007, an effort was started (Roberts and Southon 2007) to measure the absolute ^{14}C/^{12}C ratio of the widely used ^{14}C standard material OX-I (Oxalic Acid I) by accelerator mass spectrometry (AMS). Once this ratio is known, a new measurement of the ß-decay rate of OX-I would yield an additional half-life value. For the time being, we recommend using 5700 ± 30 yr for the half-life of ^{14}C, which is also the value reported in the data base of the National Nuclear Data Center of Brookhaven National Laboratory (2018).

It is well known that the exact value of the ^{14}C half-life is not important for ^{14}C dating, since true ages can only be determined from the measured ^{14}C content with the help of ^{14}C calibration curves (Reimer et al. 2013). Simply speaking, ^{14}C dating means to determine the absolute age of an object by comparing the measured ^{14}C content with the one of an object of known age. However, a non-trivial condition for this procedure to work is that the object of unknown age has taken up ^{14}C from the same source as the object of known age used for the calibration. For deviation from this assumption, see e.g. Dee et al. (2010).

The exact value of the half-life would only matter if absolute dating with ^{14}C discussed some 20 years ago (Szabo et al. 1998) could be implemented. It would require to measure the ratio of ^{14}C/^{14}N*, where ^{14}N* is the radiogenic decay product of ^{14}C. This, however, seems quite impossible because one would have to identify the minute amount of ^{14}N* in the presence of the "ocean" of normal ^{14}N we live in.

THEORY OF ß-DECAY

The basic theory of ß decay was developed by Fermi (1934), shortly after the neutron was detected (Chadwick 1932a, 1932b), and Heisenberg described the nucleus as consisting of protons and neutrons (Heisenberg 1932). Fermi also accepted the hypothesis of Pauli put forward in 1930 in his famous letter to the "Radioactive Ladies and Gentlemen" at a meeting in Tübingen (Mößbauer 1998), that in ß-decay a neutrino with spin 1/2 ħ is emitted together with the electron, which was known to have also spin 1/2 ħ (Uhlenbeck and Goudsmit 1926). In essence, the half-life of a nucleus depends on the decay energy determined by the mass difference between parent and daughter nucleus, and on the transition matrix element depending on the nuclear structure of the two nuclei. In his ß-decay theory, Fermi already discussed the conditions for allowed and forbidden ß-decays, which were further developed by Gamow and Teller (1936).

The ^{14}C ß-decay, the Most Hindered Gamow-Teller Transition

Explaining the unusually long half-life of ^{14}C from first principles is a big challenge to nuclear theory. The spins of the electron and neutrino can couple antiparallel to 0 and parallel to 1. For allowed transitions (Fermi 1934), they transport no angular momentum. Therefore, in the anti-parallel mode they can connect parent nuclei with nuclear spin 0^+ to daughter nuclei with 0^+ states (so-called super-allowed Fermi transitions). In the parallel mode they can connect 0^+ or 1^+ states to 1^+ or 0^+ states, respectively (so-called allowed Gamow-Teller [GT] transitions)

The transition strength for a ß-decay is usually characterized by the Log *ft* value, which is the logarithm of the *ft* product expressed by the relation

$$\mathrm{Log}\, ft = \mathrm{constant}/|\mathrm{M_{GT}}|^2 \tag{1}$$

Here, *f* is the Fermi phase-space integral, a factor which can be calculated from the ß-decay energy, and *t* is the half-life. $|\mathrm{M_{GT}}|^2$ is the square of the Gamow-Teller matrix element, which contains the information about the nuclear structure of the parent and daughter states. The inverse dependence of Log *ft* on the GT matrix element means that large Log *ft* values are the result of small matrix elements, which then leads to long half-lives. To reproduce the ^{14}C half-life requires a reduction of the GT matrix element ($\mathrm{M_{GT}}$), which is in the order of 1 for normal GT strength, to the very small value of $\sim 2\times10^{-3}$.

The ß-decay from the 0^+ ground state of ^{14}C to the 1^+ ground state of ^{14}N fulfills the GT condition. As it happens, there exist a total of 714 GT transitions in all nuclei (Singh et al. 1998), with the majority of Log *ft* values lying between 4 and 6 (Figure 2). From the distribution of the GT transitions it can be seen that the $^{14}\mathrm{C}\rightarrow^{14}\mathrm{N}$ transition has the highest

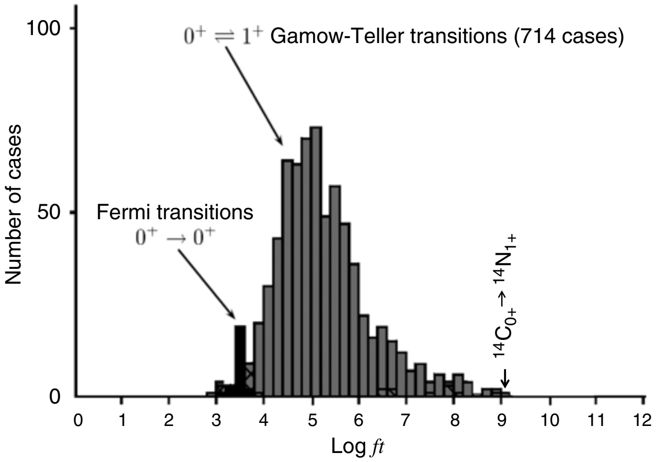

Figure 2 Distribution of Log *ft* values for Fermi and Gamow-Teller transitions (Singh et al. 1998). The Log *ft* value for the $^{14}C_{0+}$ to $^{14}N_{1+}$ transition is 9.04, and its position is marked with an arrow.

log *ft* value of 9.04, which also means that it is the most hindered GT transition (smallest GT matrix element, see below).

The Moszkowski Plot

Moszkowski (1951) has given a rapid method of determining log $f_o t$ values in a graphical way (nomogram) by connecting the maximum kinetic energy of ß⁻ or ß⁺ particles, or the energy available for K-capture, with the half-life. Assuming a 100% decay to one final state (no branching to other states), the Log *ft* is then given by Log ft = log $f_o t$ + log (C) (Moszkowski 1951). Here, log (C) is a Coulomb correction, which is different for the three decay modes. For the ß⁻ decay of ^{14}C, this correction is small, log $(C) \sim 0.2$, and is therefore of minor importance in the following discussion. As can be seen from Figure 3, for the ^{14}C decay one obtains a log $f_o t$ value close to 9 by connecting the maximum electron energy of 156 keV with the known half-life of 5700 yr. Alternatively, with a log $f_o t$ value around 3.5, not uncommon for GT transitions of neighboring light nuclei (Singh et al. 1998), one obtains a half-life of only around 10 days. The major challenge for a theoretical description of the GT transition of ^{14}C is then to find the proper structure (wave function) for the parent and daughter states, which produce an exceptionally small GT matrix element required to produce the large Log *ft* value of the ^{14}C ß-decay.

ATTEMPTS TO CALCULATE THE HALF-LIFE OF ^{14}C

Overall, one can distinguish three different approaches for a theoretical description of nuclei. (1) In the traditional shell-model approach one starts with an inert core of nucleons filling a closed shell, and tries to describe the properties of nuclei by some effective interaction of the particle or holes outside the core (Inglis 1953). (2) Alternatively, light nuclei were also described by the interactions of pre-formed alpha particles (Hafstad and Teller 1938). (3) Finally, in the so-called *ab-initio* no-core shell model approach all nucleons are treated as active, requiring an enormous computational effort for systems like ^{14}C (Maris et al. 2011). A recent review covers the many theoretical efforts to describe the properties of light nuclei (Freer et al. 2018),

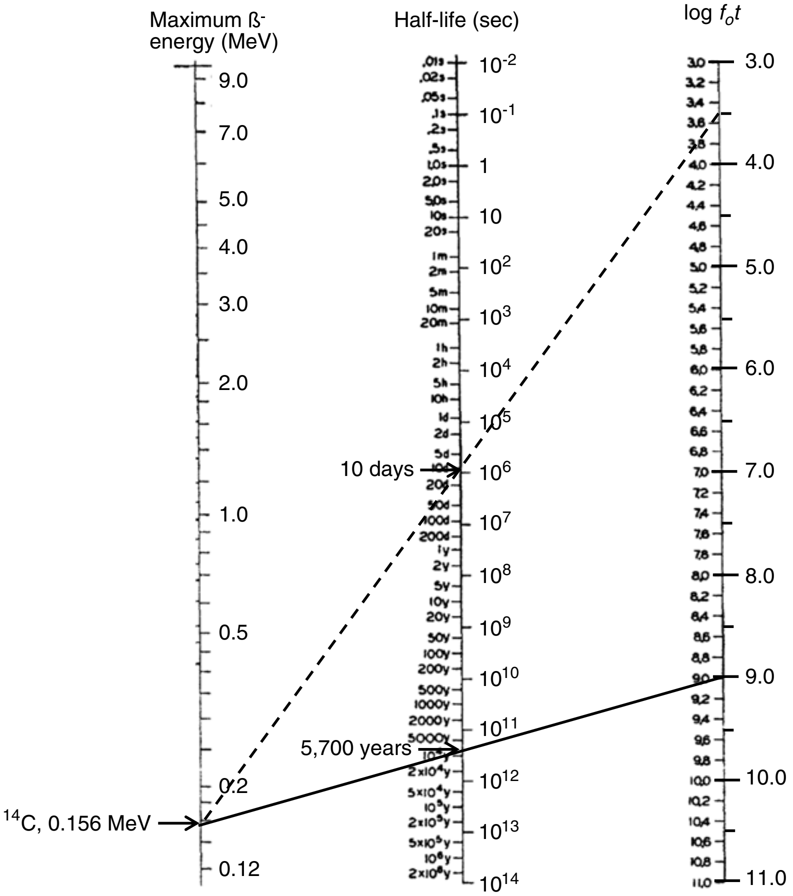

Figure 3 Graphical way to determine the half-life by connecting the maximum energy of the electrons emitted in the ß-decay of ^{14}C with the log $f_o t$ value. For clarity the figure is a simplified version of the original nomogram of Moszkowski (1951), which also included $ß^+$ and K-capture decays. The solid line connects the ^{14}C decay energy with the log $f_o t$ of 9, whereas the dashed line indicates the much shorter half-life, if the log $f_o t$ value would be 3.5.

indicating that methods (2) and (3) may eventually merge together in a "complete" description of nuclear properties.

Early on, it was realized that the exceptionally long half-life of ^{14}C would require a nuclear structure of the involved nuclei, which go beyond the description with standard shell model approaches (Inglis 1953). One of the first attempts was the introduction of a non-central tensor force in addition to the central force and ordinary spin-orbit interaction (Jancovici and Talmi 1954). Although some reduction of the GT matrix element for the ^{14}C ß-decay was achieved with this method, it was not able bring it down to the required very small value. In more recent times, a thorough study of the GT strength in the mass-14 nuclei (Negret et al. 2006) discussed a variety of theoretical attempts and came to the conclusion that it might be necessary to include cluster structures of nuclei in addition to shell-model descriptions. For example, the famous "Hoyle state" in ^{12}C at an excitation energy of 7.65 MeV and a spin of 0^+ is such a cluster state (Freer and Fynbo 2014). The existence of this

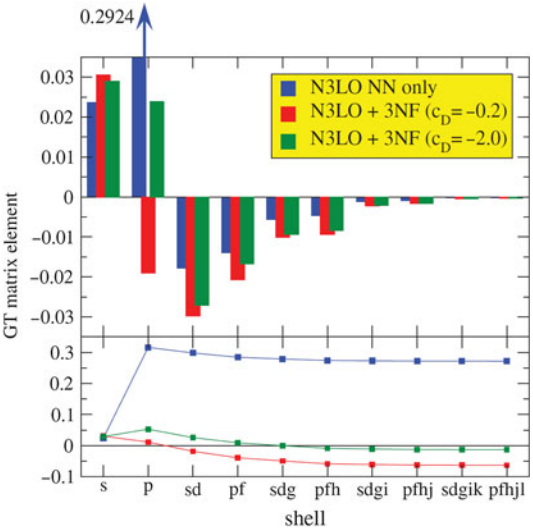

Figure 4 Summary of the contributions to the GT matrix elements for the ^{14}C ß-decay. The figure is reproduced from Maris et al. (2011). Only the calculation including three-body interactions with a relatively large coupling constant (C_D = –2.0, green symbols) is able to result in a sufficiently small GT matrix element (~2×10^{-3}) to reproduce the long half-life (green summary curve in the lower panel; please see electronic version for color figures).

state was predicted by Hoyle (1954) to understand the ^{12}C abundance in stars, and requires a production through the triple-alpha process. Great strides have been undertaken to improve a shell-model approach to explain the GT strength in the ^{14}C ß-decay (Holt et al. 2008). Although a substantial reduction of the GT matrix element was achieved by fine-tuning the nucleon-nucleon interaction with a proper interaction potential, the introduction of three-nucleon interaction seem to be necessary to reproduce the very small GT Matrix element in ^{14}C (Holt et al. 2009). This approach was pursued in an extraordinary large computational effort by an *ab-initio* shell- model calculation with all 14 nucleons active (Maris et al. 2011). This group found a sufficiently small GT matrix element (~2×10^{-3}) to reproduce the long half-life of ^{14}C by including three-nucleon forces as well. The result of this calculation is summarized in Figure 4, taken from the work of Maris et al. (2011). It should be noticed that the three-nucleon force (3NF) requires a proper value for the low-energy coupling constant C_D to cancel the contributions from the two-nucleon interaction, described here with the next-to-next-to-next-to-low-order (N3LO) approach, resulting in the vanishing small GT matrix element. In a way, C_D is a fitting constant and not entirely derived from first principles. Nevertheless, the work of Maris et al. (2011) for the first time arrived at a GT matrix element small enough to explain the long half-life of ^{14}C.

CONCLUSION

From the early days of ^{14}C dating, the long half-life was both beneficial for archaeology and perplexing from a nuclear theory point of view. Over the years, great efforts were made to understand nuclear forces well enough to be able to describe the properties of light nuclei from first principles. Even though this task is not finished, and further improvements are to be expected (Freer et al. 2018), it seems obvious that the parent nucleus ^{14}C has a very different nuclear structure as compared to the daughter nucleus ^{14}N to which it decays. Perhaps the inhibition of the decay has to do with one of them having more cluster structure than the other, requiring a total rearrangement of all nucleons. The great effort of theorists to understand nuclear forces in all details may one day lead to a complete understanding of the most unusual half-life of ^{14}C. Whatever the outcome of these efforts, the long half-life of ^{14}C remains to be a true gift of Nature to archaeology and many other fields of the environment at large including the atmosphere, biosphere, hydrosphere, lithosphere, and cryosphere (Kutschera 2013).

ACKNOWLEDGMENTS

The author acknowledges helpful discussions with Alex Brown, Martin Freer, John Southon, and Johannes van der Plicht. Constructive comments by two referees are also gratefully acknowledged.

REFERENCES

Anderson EC, Libby WF, Weinhouse S, Reid AF, Kirshenbaum AD, Grosse AV. 1947. Natural radiocarbon from cosmic radiation. Physical Review 72:931–936.

Arnold JR, Libby WF. 1949. Age determination by radiocarbon content: checks with samples of known age. Science 110:678–680.

Bella F, Alessio M, Fratelli P. 1968. A determination of the half-life of ^{14}C. Il Nuovo Cimento 58B: 233–246.

Chadwick J. 1932a. Possible existence of a neutron. Nature 129(3252):312.

Chadwick J. 1932b. The existence of a neutron. Proceedings of the Royal Society A 136 (830): 692–708.

Dee MD, Brock F, Harris SA, Bronk Ramsey C, Shortland AJ, Higham TFG, Rowland JM. 2010. Investigating the likelihood of a reservoir offset in the radiocarbon record for ancient Egypt. Journal of Archaeological Science 37: 687–693.

Engelkemeir AG, Hamill WH, Inghram MG, Libby WF. 1949. The half-life of radiocarbon (C^{14}). Physical Review 75:1825–1833.

Engelkemeir AG, Libby WF. 1950. End and wall corrections for absolute beta-counting in gas counters. Review of Scientific Instruments 21: 550–554.

Fermi E. 1934. Versuch einer Theorie der ß-Strahlen. I. Zeitschrift für Physik 88:161–177.

Freer M, Fynbo H. 2014. The Hoyle state in ^{12}C. Progress in Particle and Nuclear Physics 78:1–23.

Freer M, Horiuchi H, Kanada-En'yo Y, Lee D, Meißner U-G. 2018. Microscopic clustering in light nuclei. Reviews of Modern Physics 90: 035004, 1–46.

Gamow G, Teller E. 1936. Selection rules for the ß-disintegration. Physical Review 49:895–899.

Godwin H. 1962. Radiocarbon dating, fifth international conference. Nature 195:943–945.

Hafstad LR, Teller E. 1938. The alpha-particle model of the nucleus. Physical Review 54: 681–692.

Heisenberg W. 1932. Über den Bau der Atomkerne. I. Zeitschrift für Physik 77(1–2):1–11.

Holt JW, Brown GE, Kuo TTS, Holt JD, Machleidt R. 2008. Shell model description of the ^{14}C dating ß decay with Brown-Rho-Scaled NN interaction. Physical Review Letters 100: 062501.

Holt JW, Kaiser N, Weise W. 2009. Chiral three-nucleon interaction and the ^{14}C-dating ß decay. Physical Review C 79:054331, 1–11.

Hoyle F. 1954. On nuclear reactions occuring in very hot stars. I. The synthesis of elements from carbon to nickel. Astrophysical Journal Supplement 1:121–146.

Hughes EE, Mann WB. 1964. The half-life of carbon-14: comments on the mass-spectrometric methods. International Journal of Applied Radiation and Isotopes 15:97–100.

Inglis DR. 1953. The energy levels and the structure of light nuclei. Reviews of Modern Physics 25(2):390–450.

Jancovici B, Talmi I. 1954. Tensor forces and the ß decay of C^{14} and O^{14}. Physical Review 95: 289–291.

Jones WM. 1949. A determination of the half-life of carbon 14. Physical Review 76(7):885–889.

Korff SA, Hamermesh B. 1946. The energy distribution and number of cosmic-ray neutrons in the free atmosphere. Physical Review 69:155–159.

Kutschera W. 2013. Application of accelerator mass spectrometry. International Journal of Mass Spectrometry 349–350:203–218.

Libby WF. 1946. Atmospheric helium three and radiocarbon from cosmic radiation. Physical Review 69:671–672.

Libby WF. 1952. Half-life of radiocarbon. In: Libby WF, editor. Radiocarbon dating. Chicago & London: University of Chicago Press. p. 34–42.

Libby WF, Anderson EC, Arnold JR. 1949. Age determination by radiocarbon content: world-wide assay of natural radiocarbon. Science 109:227–228.

Mann WB, Marlow WF, Hughes EE. 1961. The half-life of carbon-14. International Journal of Applied Radiation and Isotopes 11: 57–67.

Maris P, Vary JP, Navrátil P, Ormand WE, Nam H, Dean DJ. 2011. Origin of the anomalous long lifetime of ^{14}C. Physical Review Letters 106:202502.

Miller WW, Ballentine R, Bernstein W, Friedman L, Nier AO, Evans RD. 1950. The half-life of carbon fourteen and a comparison of gas phase counter methods. Physical Review 77(5): 714–715.

Mößbauer RL. 1998. History of Neutrino Physics: Pauli's Letters. In: Altmann M, Hillebrandt W, Janka H-T, Raffelt G, editors. Proceedings of the Workshop on Neutrino Astrophysics at Ringberg Castle, 20–24 Oct 1997, Sonderforschungsbereich 375 Astroteilchenphysik, Technische Universität München: 3–5.

Moszkowski SA. 1951. A rapid method of calculating log (ft) values for ß-transitions. Physical Review 82:35–37.

Negret A, Adachi T, Barrett BR, Bäumer C, van den Berg AM, Berg GPA, von Brentano P, Frekers D, De Frenne D, Fujita H, Fujita K, Fujita Y, Grewe EW, Haefner P, Harakeh MN, Hatanaka K, Heyde K, Hunyadi M, Jacobs E, Kalmykov Y, Korff A, Nakanishi K, Navrátil P, von Neumann-Cosel P, Popescu L, Rakers S, Richter A, Ryezayeva N, Sakemi Y, Shevchenko A, Shimbara Y, Shimizu Y, Tameshige Y, Tamii A, Uchida M, Vary J, Wörtche HJ, Yosoi M, Zamick L. 2006. Gamow-Teller strength in the A = 14 multiplet: a challenge to the shell model. Physical Review Letters 97:062502.

Norris WD, Inghram MG. 1946. Half-life determination of carbon (14) with a mass spectrometer and low absorption counter. Physical Review 70:772–773.

Nuclear Data Center, Brookhaven National Laboratory. 2018. Nuclear structure and decay data. Available at https://www.nndc.bnl.gov/nudat2/.

Olsson IU, Karlen I, Turnbull AH, Prosser NJD. 1962. A determination of the half-life of C^{14} with a proportional counter. Arkiv för Fysik 22(14):237–255.

Reid AF, Dunning JR, Weinhouse S, Grosse AV. 1946. Half-life of C^{14}. Physical Review 70:431.

Reimer PJ, et al. 2013. IntCal13 and Marine13 radiocarbon age calibration curves 0–50,000 years cal BP. Radiocarbon 55(4):1869–1887.

Roberts ML, Southon JR. 2007. A preliminary determination of the absolute ^{14}C/^{12}C ratio of OX-I. Radiocarbon 49(2):442–445.

Ruben S, Kamen MD. 1941. Long-lived radioactive carbon. Physical Review 59:349–354.

Singh B, Rodriguez JL, Wong SSM, Tuli JK. 1998. Review of Log ft values in ß decay. Nuclear Data Sheets 84:487–563.

Szabo J, Carmi I, Segal D, Mintz E. 1998. An attempt at absolute ^{14}C dating. Radiocarbon 40(1):77–83.

Uhlenbeck GE, Goudsmit S. 1926. Spinning electrons and the structure of spectra. Nature 117:264–265.

Watt DE, Ramsden D, Wilson HW. 1961. The half-life of carbon-14. International Journal of Applied Radiation and Isotopes 11:68–74.