On $^{146}$Nd, $^{144}$Sm and other unexplored $2\beta$ decay isotopes

Francesco Nozzoli

INFN-TIFPA, I-38123 Trento, Italy.

(Dated: June 26, 2018)

$^{146}$Nd is one of only four (over a total of 35 existing) $2\beta^-$ decay isotope candidates whose half-lives currently lack experimental limits. The $\alpha$ activity of the $^{146}$Sm daughter nuclide allows placement of limits on the $2\beta$ decay using the $^{146}$Nd/$^{142}$Nd abundance ratio ($T_{1/2}^{2\beta} \lesssim 3 \times 10^9$ yr) or direct search for $^{146}$Sm with accelerator mass spectrometers ($T_{1/2}^{2\beta} \gtrsim 4.5 \times 10^{19}$ yr). With a similar approach, a modest ($\sim 10^{13}$ yr) first limit on half-lives for the other unexplored $2\beta$ unstable isotopes and competitive limits (few $10^3$ yr) for $^{98}$Mo and $^{122}$Sn are also given. Finally, it is shown how the limit $T_{1/2}^{0\nu\beta^+} \gtrsim 10^{15}$ yr for the unexplored $^{144}$Sm $0\nu\beta^+$ decay may be obtained from the data of a GSO crystal scintillator.

PACS numbers: 23.40.-s, 14.60.Pq, 23.60+e

Keywords: Double Beta Decay; $^{146}$Nd; $^{144}$Sm; Low background experiments.

I. INTRODUCTION

Neutrinoless double beta decay ($0\nu2\beta$) process is currently of great interest, since it is closely related to fundamental aspects of elementary particle physics beyond the standard model [1]. In particular the investigation of $0\nu2\beta$ process offers information complementary to that given by neutrino oscillation experiments, possibly revealing the nature of the neutrino (Majorana or Dirac particle) and giving the absolute scale for the effective neutrino mass.

On the other hand, 2-neutrino double beta decay ($2\nu2\beta$) is a second-order process, which is not forbidden by lepton number conservation; however, the investigation of this process in as many nuclei as possible is very useful, since it gives information about the calculation of nuclear matrix elements both for the $2\nu2\beta$ and for the $0\nu2\beta$ processes [2].

Current experiments achieve impressive sensitivity to $0\nu2\beta$ – and the existing limits on the half-life for this process are in the $10^{24} - 10^{26}$ yr range, making it possible to test for the effective neutrino mass in the sub-eV range, depending on the considered isotope (see e.g. [3–8]). Moreover, most of these experiments are able to identify the $2\nu2\beta$ decay whose half-life lies in the $10^{19} - 10^{21}$ yr range for favorable isotopes (for a review see e.g. [9–10]).

Considering all of the 35 potential $2\beta^-$ and 22 potential $\epsilon\beta^+$ decay candidates, only eight nuclei still have no experimental limits on half-life [10–13]; this is generally due to a low transition energy ($Q_{\beta\beta}$), a low natural abundance, or to difficulties in obtaining a detector material containing the $2\beta$ active nuclei.

II. LIMITS FROM THE ABUNDANCE RATIO IN THE EARTH’S CRUST

A simple preliminary limit on the half-life of all unexplored $2\beta$ decay isotopes can be inferred from the daughter/parent abundance ratio in the Earth’s crust [14].

In particular, assuming that daughter isotope, $D(t)$, is stable (or that the known decay half-life is much longer than the Solar System lifetime), and that the $2\beta$ process is the dominant decay process for the parent isotope, $P(t)$, the growth equation for the system is:

$$\frac{dD(t)}{dt} = \frac{P(t)}{\tau_{\beta\beta}} + \frac{dD^{ext}}{dt} = \frac{P(t)}{\tau_{\beta\beta}}$$  \hspace{1cm} (1)

where $\frac{dD^{ext}}{dt}$ is the possible contribution coming from sources that are different from the $2\beta$ decay of the parent isotope, and this rate is positive since the daughter isotope is stable. Equation (1) can be integrated from the time of the Solar System formation ($t = 0$) to the present time ($t = T \approx 4.5$ Gyr) to obtain the half-life limit:

$$D(T) > P(T) \left( \frac{2}{\tau_{\beta\beta}} - 1 \right) \Rightarrow T_{1/2}^{\beta\beta} > \frac{\ln(2) \cdot T}{\ln \left( \frac{D(T)}{P(T)} + 1 \right)}$$

where it was assumed that additional production of the parent isotope, $P^{ext}$, is negligible with respect to the amount of parent isotope, $P(0)$, produced at Solar System formation time.

This geochemical limit for $2\beta$ decay is therefore based on the $D/P$ ratio measured in the Earth’s crust [14].

The limit is very conservative, since it is related to the requirement that the daughter nuclei not be overproduced, with respect to the parent ones, during the Solar System life because of the $2\beta$ decay.

Table 1 summarizes the limits on $2\beta$ half-lives that can be obtained for some $2\beta$ unstable nuclei. The limits obtained are very modest, ranging between a few $10^8$ yr and a few $10^9$ yr. However, $2\beta$ processes for these nuclei are constrained here for the first time.

It is important to note that for the $^{80}$Se – $^{80}$Kr case, losses of Kr gas from the Earth’s crust to the atmosphere
are possible \[13\], therefore the ratio \(\frac{\alpha}{\beta}\) may be underestimated in the Earth’s crust and the obtained limit might not be conservative.

On the other hand, since the \(^{146}\text{Sm}\) has a relatively short lifetime and “immediately” decays into \(^{142}\text{Nd}\), the limit of \(^{146}\text{Nd}\) 2\(\beta\) decay is related to the relative isotope abundance of the same atomic species, which is much better measured\[20\] than the absolute abundance of different atomic species in the Earth’s crust.

Finally, for \(^{98}\text{Mo} – ^{98}\text{Ru}\) and \(^{122}\text{Sn} – ^{122}\text{Te}\) 2\(\beta\) decay, the current existing limits were calculated in \[11\] on the basis of the photographic emulsion measurements of \[18\] with corrections on the decay energy and the natural abundance of the isotopes. In the case of \(^{98}\text{Mo} – ^{98}\text{Ru}\) and \(^{122}\text{Sn} – ^{122}\text{Te}\), the average \(\frac{\alpha}{\beta}\) ratios in Earth’s crust are \(6.5 \times 10^{-5}\) and \(2.5 \times 10^{-4}\), respectively. The limits \(T_{1/2}^{98}\text{Mo} > 5 \times 10^{13}\) yr and \(T_{1/2}^{122}\text{Sn} > 1.3 \times 10^{13}\) yr, obtained with the abundance ratio approach, are at the level of the existing limits based on photographic emulsion analysis \[11\]–\[13\].

The approach described above, based on Earth’s crustal abundance ratio, could be useful also to set lower limits on half-lives for other unexplored rare/exotic nuclear decay processes \[26\], \[27\], such as rare \(\alpha\) decay \[28\], rare \(\beta\) decay \[21\], \[30\] and cluster decay \[21\], \[32\].

### III. THE \(^{146}\text{Nd} 2\beta\) DECAY PROCESS

Among all the 2\(\beta\) decay candidates, \(^{146}\text{Nd}\) (17.2 \% isotopic abundance) is the nucleus having the lowest \[^Q\beta\beta\] value (70.2 keV) and it is still unexplored. The theoretical prediction for 2\(\nu\)2\(\beta\) decay of \(^{146}\text{Nd}\) gives the hopelessly-high half-life \(T_{2\nu}^{146}\text{Nd} = 2.1 \times 10^{31}\) yr in the pseudo-SU(3) model \[^34\]; this is due to the very low available phase space. However, for the same reason (assuming \(\langle m_\nu \rangle = 1\) eV), 0\(\nu\)2\(\beta\) decay of \(^{146}\text{Nd}\) is expected to be a factor \(\sim 10^3\) more probable than the standard 2\(\nu\) process \[^35\]. In this latter case, in fact, \(T_{0\nu}^{146}\text{Nd} = 1.18 \times 10^{28}\) yr is expected \[^34\].

Despite the very high expected half-lives, his feature offers additional interest to the investigation of \(^{146}\text{Nd}\) double beta decay.

The isotope \(^{146}\text{Nd}\) is a potentially \(\alpha\) radioactive nucleus (\(Q_\alpha = 1.182\) MeV). However, since the expected half-life for this decay channel is expected to be order \(10^{34}\) yr \[^36\], this contribution can be neglected with respect to the 2\(\beta\) process.

The schema of \(^{146}\text{Nd} 2\beta\) decay to \(^{146}\text{Sm}\) is shown in Fig.1. A peculiarity of the \(^{146}\text{Nd} 2\beta\) decay is the \(\alpha\) activity of the daughter: \(^{146}\text{Sm} (T_{\alpha}^{146}\text{Sm} = 68\) Myr, \(Q_\alpha = 2529\) keV \[^16\], \[^37\].

The half-life of \(^{146}\text{Sm}\) was recently re-determined in \[^16\] to be 68\(\pm\)7 Myr, which is \(\sim 30\%\) shorter than previous estimation (103\(\pm\)5 Myr) \[^37\]. Due to the relatively short half-life of \(^{146}\text{Sm}\), the presence of this isotope in natural samples is expected to be very small. Therefore, an alternative approach for the investigation of 0\(\nu\)2\(\beta\) decay of \(^{146}\text{Nd}\) is to search for the \(^{146}\text{Sm} \alpha\) peak. This approach, with respect to the direct investigation of the 2\(\beta\) spectra, would be advantaged by the reduction of the electromagnetic background by possible pulse shape analysis and it would avoid the difficulties posed by the small \(Q_{\beta\beta}\). On the other hand, despite its smallness, the possible presence of relic \(^{146}\text{Sm}\) from Solar System formation or from cosmogenic activation would offer an unavoidable background contribution.

In fact, assuming equilibrium, the ratio of 2\(\beta\) produced \(^{146}\text{Sm}\) over \(^{146}\text{Nd}\) is given by:

\[
\frac{^{146}\text{Sm}^{(\beta\beta)}}{^{146}\text{Nd}} \simeq \frac{T_{0\nu}^{146}\text{Nd}}{T_{1/2}^{1/2}^{146}\text{Sm}} \sim 10^{-20} \left(\frac{m_\nu}{1\text{eV}}\right)^2.
\]

On the other hand, relic \(^{146}\text{Sm}\) from Solar System formation is expected to be in the range:

\[
\frac{^{146}\text{Sm}^{(\beta\beta)}}{^{146}\text{Nd}} = \frac{^{146}\text{Sm}^{(0)}}{^{146}\text{Nd}} e^{-\frac{T_{\alpha}^{146}\text{Sm}}{10^{17} - 10^{23}}},
\]

The large uncertainty in the latter evaluation is obtained by considering the two more recent determinations of \(^{146}\text{Sm}\) lifetime: \(T_\alpha = \frac{T_{\alpha}^{146}\text{Sm}}{10^{17} - 10^{23}}\). The ratio \(^{146}\text{Sm}^{(\beta\beta)} / ^{146}\text{Nd}\) is expected to be of order \(\sim 1\%\) \[^16\], \[^22\], and the ratio \(^{146}\text{Sm}^{(0)} / ^{146}\text{Nd}\) \(\sim 3\%\) \[^14\] has been considered.

Despite the possible high backgrounds provided by relic \(^{146}\text{Sm}\) or by cosmogenic \(^{146}\text{Sm}\) production due to neutron spallation/capture, a cautious lower limit on the \(^{146}\text{Nd} 2\beta\) decay half-life can be obtained. It is important to note that the possible experimental detection of the \(^{146}\text{Sm} \text{“background” would be itself a very interesting physical result} \[^38\] giving additional information on \(^{146}\text{Sm} \text{half-life, on nuclear synthesis at the time of Solar System formation} \[^16\], \[^22\], \[^23\], \[^29\] and/or on the early crust-mantle differentiation process} \[^24\].

As an example the limit of \(^{146}\text{Sm}^{147}\text{Sm} < 10^{-11}\) was obtained in \[^16\] by analyzing the blank spectrum of
\[ {\text{nat}}\text{Sm} \] with the accelerator mass spectrometer (AMS). Considering Eq. 2, the limit \( T_{1/2}^{146\text{Nd}} > 4.5 \times 10^{19} \text{ yr} \) can be inferred[40].

IV. LIMITS FROM Sm-DOPED DETECTORS

As an alternative to the search for \(^{146}\text{Sm}\) with AMS, investigation of \( 2\beta^- \) decay of \(^{146}\text{Nd}\) could be possible by a direct search for the presence of the \(^{146}\text{Sm} \alpha\) peak in detectors containing Sm. Currently, there exists no appropriate Sm-based scintillator or bolometer. A search for \(^{146}\text{Sm}\) could still be pursued in existing detectors that have been doped with Sm or containing Sm as a contaminant. As an example, a ZnWO\(_4\) scintillating bolometer doped with enriched \(^{146}\text{Sm}\) was used in [17] to detect the \(^{148}\text{Sm} \alpha\) decay (\( T_{1/2}^{148} = 6.2^{+1.2}_{-1.1} \times 10^{15} \text{ yr} \) and \( Q_{\alpha}^{148} = 1987.3 \pm 0.5 \text{ keV} \)). Despite the enrichment procedure, the faster \(^{147}\text{Sm} \alpha\) decay (\( T_{1/2}^{147} = 1.06 \times 10^{11} \text{ yr} \)) is clearly visible with a peak at \( Q_{\alpha}^{147} = 2310.5 \text{ keV} \). On the other hand, just two events are observed in the 2.5 MeV region where \(^{146}\text{Sm}\) is expected. Isotopic composition measurements on enriched \( \text{Sm}_2\text{O}_3 \) powder are tabulated in [17]. After enrichment, the \(^{147}\text{Sm}\) was depleted by a factor 0.06 (from 14.99\% to 9.1\%) and \(^{148}\text{Sm}\) was depleted by a factor 0.02 (from 3.07\% to 0.96\%). Considering the enrichment of \(^{148}\text{Sm}\) by a factor 8.5 (from 11.24\% to 95.54\%), it is possible to estimate that the depletion factor for the \(^{146}\text{Sm}\) should be in the range 0.03 - 0.04.

Therefore, from the measurement reported in [17], a limit of \( 2 \times 10^{-8} \) on \(^{146}\text{Sm} \) natural abundance can be inferred; this implies \( T_{1/2}^{146\text{Nd}} > 3.5 \times 10^{15} \text{ yr} \). A similar limit (\( \lesssim 10^{-7} \)) on the natural abundance of \(^{146}\text{Sm}\) can be obtained by considering the spectrum of \( \text{Li}_6\text{Eu}(\text{BO}_3)_3 \) [41]. Also in this case, the \(^{147}\text{Sm} \alpha\) peak due to natural Sm contamination is clearly visible but the \(^{146}\text{Sm}\) peak is absent. It is important to note that the limits obtained with this technique can be greatly improved if a specific enrichment in \(^{146}\text{Sm}\) is pursued. In particular, considering the ZnWO\(_4\) used by [17], an improvement of a factor \( \sim 250 \) could be obtained by enriching the sample in isotope 146 instead of isotope 148. A further factor 100 improvement can be obtained by taking data for a period of a few years (the results of [17] are based on an exposure of 364 h). This would provide sensitivity to improve the AMS-based limit on \(^{146}\text{Nd} \) \( 2\beta^- \) decay by a factor \( \sim 10 \).

V. A LIMIT FOR \(^{144}\text{Sm} \) \( 0\nu\beta^+ \) DECAY

\(^{144}\text{Sm}\) is one of only four \( e\beta^- \) decay isotope candidates without experimental limits on half-life (see Table 1). As above, this section discusses the possible sensitivity to \(^{144}\text{Sm} \) \( 0\nu\beta^+ \) decay offered by investigation with detectors containing traces of Sm. In particular, the concentration of \(^{144}\text{Sm}\) nuclei in the detector can be inferred through the \( \alpha \) activity of \(^{147}\text{Sm}\).

Considering as an example the 635 g GSO scintillator of Ref. [19], the expected contamination from Sm is at the level of \( \sim 8 \) ppm [19]; moreover, since the GSO of Ref. [19] is 5.4 cm \( \times \) 4.7 cm \( \odot \), the detection efficiency for an internal 511 keV \( \gamma \) quanta is \( \epsilon \gtrsim 0.5 \) (for 511 keV in GSO, \( \mu_\rho \sim 0.1 \text{ cm}^2/\text{g} \)).

Therefore, considering the limit \( T_{0\nu\beta^+}^{160\text{Gd}} > 1.3 \times 10^{21} \text{ yr} \) for \(^{160}\text{Gd} \) nuclei given in Ref. [19] (which has a \( Q_{\beta\beta}^{160\text{Gd}} \approx 1730 \text{ keV} \) very similar to \( Q_{\beta\beta}^{144\text{Sm}} \approx 1781 \text{ keV} \)), a limit for \(^{144}\text{Sm} \) nucleus \( 0\nu\beta^+ \) decay half-life \( T_{0\nu\beta^+}^{144\text{Sm}} \gtrsim 10^{15} \text{ yr} \) can be deduced.

Finally, the sensitivity to \( 0\nu\beta^+ \) decay of \(^{144}\text{Sm}\) could be greatly improved by requiring coincidences between near detectors, thanks to the 511 keV \( \beta^-\)-annihilation \( \gamma \) quanta.

VI. CONCLUSIONS

Limits for half-lives of \( 2\beta^- \) processes in 10 isotopes (see table 1) are set here for the first time; this completes the picture of \( 2\beta^- \) decay tables presented in Ref. [12]. The limits obtained from the absolute abundance ratio of elements in the Earth’s crust are generally very modest (at level of \( \sim \) Gyr); however, for some elements, such as \(^{98}\text{Mo}\) and \(^{122}\text{Sn}\), this approach gives limits of a few \( 10^{13} \) yr that are at the level of existing experimental ones. Moreover, for \(^{146}\text{Nd}\) and \(^{144}\text{Sm}\) isotopes, half-life limits of \( T_{1/2}^{2\beta^-} \gtrsim 4.5 \times 10^{19} \text{ yr} \) and \( T_{1/2}^{2\beta^+} \gtrsim 10^{15} \text{ yr} \), respectively, can be obtained from existing experimental data taken with accelerator mass spectrometers or with Sm-containing detectors.

ACKNOWLEDGMENTS

I wish to acknowledge all the colleagues involved in useful discussions for this work and some other ones [42,43].
[1] Particle Data Group, C. Patrignani et al., Chin. Phys. C 40 (2016) 100001.
[2] J. Engel and J. Menendez, Rep. Prog. Phys. 80 (2017) 046301.
[3] C. Alduino et al., Phys. Rev. C. 93 (2016) 045503.
[4] J.B. Albert et al., Nature 510 (2014) 229.
[5] M. Agostini et al., Nature 554 (2017) 47.
[6] A. Gando et al., Phys. Rev. Lett. 117 (2016) 082503.
[7] R. Arnold et al., Phys. Rev. D. 89 (2014) 111101(R).
[8] O. G. Polischuk et al., AIP Conf. Proc. 1894 (2017) 020018; [arXiv:171103854].
[9] R. Henning, Rev. Phys. 1 (2016) 29.
[10] A. Barabash, Nucl. Phys. A 935 (2015) 52; [arXiv:1702.06340].
[11] V.I. Tretyak and Yu.G. Zdesenko, At. Data Nucl. Data Tables 61 (1995) 43.
[12] V.I. Tretyak, Yu.G. Zdesenko, At. Data Nucl. Data Tables 80 (2002) 83.
[13] V.I. Tretyak et al., Europhys. Lett. 69 (2005) 41; [arXiv:nucl-ex/0404016].
[14] David R. Lide, ed., CRC Handbook of Chemistry and Physics, 88th Edition (Internet Version 2008), CRC Press/Taylor and Francis, Boca Raton, FL.
[15] O.K. Manuel, J. Phys. G Nucl. Part. Phys. 17 (1991) S221.
[16] N. Kinoshita et al., Science 335 (2012) 1614.
[17] N. Casali et al. J. Low. Temp. Phys. 184 (2016) 952.
[18] J.H. Fremlin and M.C. Walters, Proc. Phys. Soc. A 65 (1952) 911.
[19] F.A. Danevich et al., Nucl. Phys. B (Proc. Suppl.) 48 (1996) 235; F.A. Danevich et al., Nucl. Phys. A694 (2001) 375; [arXiv:nucl-ex/0011202].
[20] The $^{146}\text{Sm}/^{142}\text{Nd}$ ratio is used as normalization in the Sm-Nd geochronology [21]; some anomalies observed in the abundance ratios of Nd isotopes relative to $^{142}\text{Nd}$ have been attributed to the possible contribution of $^{146}\text{Sm}$ [22][24]. Moreover, variations in the abundances of Nd isotopes in erupted lava are correlated with the volume of volcanic eruptions [25].
[21] see e.g. G.J. Wasserburg et al., Geochim. et Cosmochim. Acta 45 (1981) 2311.
[22] A. Prinzhofer et al., Astrophys. J. 344 (1989) L81.
TABLE I. Half-life limits for unexplored (or poorly explored) $2\beta$ processes; the average Earth's crust abundance of parent-daughter nuclei is considered [14].

| Decay channel | Parent-Daughter | $N_{\text{daughter}}$ | $T_{1/2}^{2\beta}$ limit (yr) | Note |
|---------------|-----------------|-----------------------|--------------------------------|------|
| $2\beta^-$    | $^{80}$Se - $^{80}$Kr | $9 \times 10^{-5}$ | $3.5 \times 10^{13}$ | a   |
| $2\beta^-$    | $^{86}$Kr - $^{86}$Sr | $2 \times 10^6$ | $2 \times 10^9$ |   |
| $2\beta^-\rightarrow \alpha$ | $^{204}$Hg - $^{204}$Pb | 33 | $8.8 \times 10^8$ |   |
| $2\beta^-\rightarrow \alpha$ | $^{146}$Nd - $^{146}$Sm - $^{142}$Nd | 1.58 | $3.3 \times 10^9$ |   |
| $2\beta^-\rightarrow \alpha$ | $^{146}$Nd - $^{146}$Sm | - | $4.5 \times 10^{19}$ |   |
| $2\beta^-\rightarrow \alpha$ | $^{146}$Nd - $^{146}$Sm | - | $3.5 \times 10^{15}$ |   |
| $2\beta^-$ | $^{98}$Mo - $^{98}$Ru | $6.5 \times 10^{-5}$ | $5 \times 10^{13}$ |   |
| $2\beta^-\rightarrow \alpha$ | $^{122}$Sn - $^{122}$Te | $2.5 \times 10^{-4}$ | $1.3 \times 10^{13}$ |   |
| $\epsilon\beta^+ + 2\epsilon$ | $^{144}$Sm - $^{144}$Nd | 46 | $8 \times 10^8$ |   |
| $0\nu\epsilon\beta^+$ | $^{144}$Sm - $^{144}$Nd | - | $10^{15}$ |   |
| $2\epsilon$ | $^{152}$Gd - $^{152}$Sm | 152 | $6 \times 10^8$ |   |
| $\epsilon\beta^+ + 2\epsilon$ | $^{162}$Er - $^{162}$Dy | 273 | $5.5 \times 10^8$ |   |
| $2\epsilon$ | $^{164}$Er - $^{164}$Dy | 26 | $10^9$ |   |
| $\epsilon\beta^+ + 2\epsilon$ | $^{168}$Yb - $^{168}$Er | 226 | $5.7 \times 10^8$ |   |
| $\epsilon\beta^+ + 2\epsilon$ | $^{174}$Hf - $^{174}$Yb | 210 | $5.8 \times 10^8$ |   |

\[ ^{a} \] Losses of Kr from the Earth's crust are possible [15]. Limit is not conservative.

\[ ^{b} \] Daughter nucleus potentially $\alpha$ radioactive ($T_{\text{daughter}}^{\alpha} > T_{1/2}^{144\text{Nd}} = 2.3 \times 10^{15}$ yr [12]).

\[ ^{c} \] Parent nucleus potentially $\alpha$ radioactive ($T_{\text{parent}}^{\alpha} > T_{1/2}^{152\text{Gd}} = 1.1 \times 10^{14}$ yr [12]).