New experimental limits on double-beta decay of osmium

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Abstract

Double-beta processes in $^{184}$Os and $^{192}$Os were searched for over 15851 h at the Gran Sasso National Laboratory (LNGS) of the I.N.F.N. by using a 118 g ultra-pure osmium sample installed on the endcap of a 112 cm$^3$ ultra-low-background broad-energy germanium detector. New limits on double-electron capture and electron capture with positron emission in $^{184}$Os were set at the level of $\lim T_{1/2} \sim 10^{16} - 10^{17}$

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In particular, the $2\nu 2K$ and $2\nu KL$ decays of $^{184}$Os to the ground state of $^{184}$W are restricted as $T_{1/2} \geq 3.0 \times 10^{16}$ yr and $T_{1/2} \geq 2.0 \times 10^{16}$ yr, respectively. A lower limit on the half-life for the double-beta decay of $^{192}$Os to the first excited level of $^{192}$Pt was set as $\text{lim } T_{1/2} = 2.0 \times 10^{20}$ yr at 90% C.L.

Keywords: $^{184}$Os, $^{192}$Os, Double-beta decay, Low-background gamma spectrometry

1 Introduction

Double-beta ($2\beta$) decay of atomic nuclei is a key process to study properties of neutrino and weak interaction, and to search for effects beyond the Standard Model of particles and interactions (SM). While the $2\beta$ decay with the emission of two neutrinos ($2\nu 2\beta$) is allowed by the SM and already observed in several nuclides with half-lives in the $(10^{18} - 10^{24})$ yr range [1], the neutrinoless mode of the decay ($0\nu 2\beta$) is forbidden in the framework of the SM since the process breaks the lepton number $L$ by two units [2, 3, 4, 5, 6] and probes the Majorana nature of the neutrino (the particle is equal to its antiparticle) [7, 8]. The $0\nu 2\beta$ decay remains unobserved despite more than seventy years of experimental attempts. The most sensitive experiments give limits on the $0\nu 2\beta$ decay half-life for several nuclei at the level of $\text{lim } T_{1/2} \sim (10^{24} - 10^{26})$ yr [9, 10, 11, 12, 13]. Assuming the mass mechanism of the $0\nu 2\beta$ decay by exchange of a virtual light Majorana neutrino, the region of half-life limits corresponds to effective Majorana neutrino mass limits in the range of $\langle m_{2\beta} \rangle \sim (0.1 - 0.5)$ eV. The range of $\langle m_{2\beta} \rangle$ limits is due to the possible quenching of the axial vector coupling constant $g_A$ (used to calculate the effective neutrino mass from the experimental half-life limits) [14, 15] and the uncertainty of nuclear-matrix-element calculations (see, e.g., discussion and references in [16]).

The experimental sensitivity to double-electron capture (2EC), electron capture with positron emission (EC$\beta^+$), and double-positron decay ($2\beta^+$) is substantially lower than the sensitivity of the experiments to search for $2\beta$ decay with electrons emission ($2\beta^-$). At the same time, the mechanisms of the neutrinoless 2EC, EC$\beta^+$ and $2\beta^+$ processes are the same as for the $0\nu 2\beta^-$ decay and thus, the investigations of the 2EC, EC$\beta^+$ and $2\beta^+$ decays can give essentially the same information about properties of the neutrino and
the weak interaction.

However, there are important arguments to develop experimental methods to search for the $0\nu 2\text{EC}$, $0\nu \text{EC}$, $\beta^+$ and $\beta^+$ decays owing to the potential clarification of the mechanism of the $0\nu 2\beta^-$ decay when observed: whether it is mediated by the mass mechanism with exchange by virtual light Majorana neutrinos or with a possible contribution of right-handed currents in the weak interaction \cite{17}. In addition there is an interesting possibility of a resonant $0\nu 2\text{EC}$ process that can increase the $0\nu 2\text{EC}$ decay probability up to the six orders of magnitude in case the parent and daughter atoms’ energies are degenerate \cite{18, 19, 20, 21, 22}.

The $^{184}\text{Os}$ isotope is one of the potentially $2\text{EC}$ and $\text{EC}$, $\beta^+$ decaying nuclides with the decay energy $Q_{2\beta} = 1452.8(7)$ keV \cite{23} and a rather small isotopic abundance $\delta = 0.02(2)$\% \cite{24}. A simplified decay scheme of $^{184}\text{Os}$ is shown in Fig. 1.

Another osmium isotope, the $^{192}\text{Os}$, is potentially $2\beta^-$ active with decay energy $Q_{2\beta} = 406(3)$ keV \cite{23} and isotopic abundance $\delta = 40.78(32)$\% \cite{24}. The decay scheme of $^{192}\text{Os}$ is shown in Fig. 2. As one can see from Figs. 1 and 2 some possible $2\beta$ decay processes in $^{184}\text{Os}$ and $^{192}\text{Os}$ are expected to be accompanied by emission of $\gamma$ quanta that can be identified by the $\gamma$-spectrometry method.

An experiment to search for rare decays of naturally occurring osmium nuclides with emission of $\gamma$ quanta has been carried out at the LNGS (depth of 3600 meters of water equivalent (m w.e.)) by using a sample of ultrapure osmium and ultra-low-background germanium $\gamma$ detectors of the STELLA facility \cite{27}. The data of the experiment’s first stage \cite{28, 29} were used to derive half-life limits at the level of $\lim T_{1/2} \sim 10^{14} - 10^{17}$ yr for $2\text{EC}$ and $\text{EC}$, $\beta^+$ processes in $^{184}\text{Os}$, and the $\lim T_{1/2} = 5.3 \times 10^{19}$ yr was reached for the $2\beta^-$ decay of $^{192}\text{Os}$ to the first $2^+$ $316.5$ keV excited level of $^{192}\text{Pt}$. However, the experiment’s sensitivity was limited by a rather poor detection efficiency due to self-absorption of $\gamma$ quanta in the osmium sample that was in the form of four metal ingots with diameter in the range of $(7–10)$ mm \cite{3}.

The poor detection efficiency was especially a troublesome issue for the main goal of the experiment, which was the search for $\alpha$ decay of $^{184}\text{Os}$ and $^{186}\text{Os}$ to excited levels of daughter nuclei with emission of $\gamma$ quanta of rather low energies $103.6$ keV and $100.1$ keV, respectively. To increase the detection

\footnotetext{2It should be noted that osmium is the densest naturally occurring element with the density of $22.587$ g/cm$^3$ \cite{30}.}
efficiency, the ingots were cut into thin slices with a thickness of (0.79 – 1.25) mm by using a method of electroerosion cutting. Moreover, the Os sample was placed on a specially developed ultra-low background broad-energy germanium (BEGe) detector with improved detection efficiencies and energy resolutions in the energy region from several keV to several hundreds keV. The optimization exhibited a substantial improvement of the experimental sensitivity and lower limits were set for the $\alpha$ decays of $^{184}$Os and $^{186}$Os to the first excited levels of daughter nuclei as $\lim T_{1/2}(^{184}$Os) = $6.8 \times 10^{15}$ yr and $\lim T_{1/2}(^{186}$Os) = $3.3 \times 10^{17}$ yr. It should be noted that the limits are already well above the theoretical predictions for the nuclides half-lives.
Figure 2: Decay scheme of $^{192}\text{Os}$ [26]. The $Q_{\beta\beta}$ value is from [23].

relative to the $\alpha$ decays [31].

A new stage of the experiment is in progress by using the Os sample directly placed on the crystal of the Ge detector as described in ref. [32] to further increase the detection efficiency of the low-energy $\gamma$-ray quanta expected in the $\alpha$ decays of $^{184}\text{Os}$ and $^{186}\text{Os}$ to the first excited levels of the daughter nuclei. On the other token, in the present study the data of the already completed measurements [31] are utilized to search for $2\beta$ processes in $^{184}\text{Os}$ and $^{192}\text{Os}$ with emission of X-ray and $\gamma$-ray quanta.

2 Experiment and data analysis

The production of the Os sample, the accurate determination of the Os sample isotopic composition, the low-background measurements and data analysis are described in detail in [31]. Here we briefly describe the main features of the experiment.

The Os sample was prepared from osmium metal obtained by electron-beam melting with further purification by electron-beam zone refining. The obtained ingots were cut (after the first stage of the experiment [28]) into thin slices with a thickness of $(0.79 - 1.25)$ mm by electroerosion cutting with a brass wire in kerosene.

The isotopic composition of the osmium sample was measured by using negative thermal ionization mass spectrometry. The isotopic concentration of $^{184}\text{Os}$ was determined as $0.0170(7)$% [31], with an accuracy essentially higher than that of the adopted reference value of $0.02(2)$% [24]. The $^{192}\text{Os}$ isotopic
abundance was measured as 40.86(5)% (the reference value is 40.78(32)% [24]).

The Os slices with a total mass of 117.96(2) g were assembled on the top and around of the BEGe detector endcap. The active volume of the detector is 112.5 cm$^3$; the endcap of the detector is made of 1.5 mm aluminium to increase the detection efficiency to low energy $\gamma$ quanta. The detector with the Os sample was shielded by layers of $\approx$ 5 cm thick high-purity copper and 20 cm thick lead. The experiment was running at the STELLA facility of the LNGS.

![Energy spectrum measured with the Os sample by the ultra-low-background BEGe detector over 15851 h (solid line). The detector’s background energy spectrum over 1660 h, normalized to the Os sample’s measured time, is shown by circles. The energies of $\gamma$ peaks are in keV.](image)

The energy spectra measured with the Os sample over 15851 h and background data taken over 1660 h are presented in Fig. 3. The lower counting-rate in the spectrum with the sample below $\sim$ 0.4 MeV can be
explained by the very high density of osmium that absorbs effectively external radiations from the shielding materials surrounding the detector (mainly bremsstrahlung from $^{210}$Bi in the lead shield). The analysis of the $\gamma$ peaks observed in the spectra allowed us to estimate the Os sample radioactive contamination to be rather low, with only $^{40}$K and $^{137}$Cs detected (see Table 1 and [28] for details of the analysis). Other $\gamma$ peaks observed in the spectrum measured with the Os sample are statistically indistinguishable from the background. Thus only limits were set for $^{60}$Co and daughters of $^{232}$Th, $^{235}$U and $^{238}$U (a possible contamination of the sample by $^{241}$Am will be discussed in Section 2.1).

Table 1: Radioactive trace impurities in the Os sample.

| Decay chain | Radionuclide | Specific activity (mBq/kg) |
|-------------|--------------|---------------------------|
|             | $^{40}$K     | $11 \pm 4$                |
|             | $^{60}$Co    | $\leq 1.3$                |
|             | $^{137}$Cs   | $0.5 \pm 0.1$             |
|             | $^{241}$Am   | $\leq 5.6$                |
| $^{232}$Th  | $^{228}$Ra   | $\leq 6.6$                |
|             | $^{228}$Th   | $\leq 16$                 |
| $^{235}$U   | $^{235}$U    | $\leq 8.0$                |
|             | $^{231}$Pa   | $\leq 3.5$                |
|             | $^{227}$Ac   | $\leq 1.1$                |
| $^{238}$U   | $^{238}$U    | $\leq 35$                 |
|             | $^{226}$Ra   | $\leq 4.4$                |
|             | $^{210}$Pb   | $\leq 180$                |

The dependence of the energy resolution of the detector (full width at half maximum, FWHM, in keV) on the energy of $\gamma$-ray quanta ($E_\gamma$, in keV) was determined using the $\gamma$ peaks of $^{40}$K, $^{208}$Tl, $^{210}$Pb, $^{214}$Pb, and $^{214}$Bi observed in the data of the long-time measurements as following:

$$\text{FWHM (keV)} = 0.57(5) + 0.029(2) \times \sqrt{E_\gamma}.$$  (1)
2.1 Limits on 2EC and EC$\beta^+$ processes in $^{184}$Os

There are no peculiarities in the experimental data which could be ascribed to $2\beta$-decay processes in $^{184}$Os. Thus, we report the limits on the $^{184}$Os half-life relative to the different channels and modes of the $2\beta$ decay adopting the following formula:

$$\text{lim } T_{1/2} = \frac{N \cdot \ln 2 \cdot \eta \cdot t}{\text{lim } S},$$  \hspace{1cm} (2)

where $N$ is the number of $^{184}$Os nuclei in the sample ($6.35 \times 10^{19}$), $\eta$ is the detection efficiency, $t$ is the time of measurement, and $\text{lim } S$ is the upper limit on the number of events of the effect searched for which can be excluded at a given confidence level (C.L.).

The fastest decay of $^{184}$Os is theoretically expected to be the $2\nu$2EC, mainly absorbing the K and/or L electron shells. In case of $2\nu$2K, $2\nu$KL, and $2\nu$2L capture in $^{184}$Os, a cascade of X-rays and Auger electrons of the W atom is expected. However, the $2\nu$2L decay cannot be detected in the present experiment since the energies of the L X-rays of tungsten (7.4 keV–11.7 keV) are below the detector’s energy threshold.

The response of the BEGe detector to the $2\nu$2K and $2\nu$KL decays of $^{184}$Os was simulated with the help of the EGSnrc package [33] assuming the following energies and intensities of X-ray from the K shell of W atom: 57.43 keV (K$\alpha_3$, 0.021%), 57.98 keV (K$\alpha_2$, 27.4%), 59.32 keV (K$\alpha_1$, 47.6%), 66.95 keV (K$\beta_3$, 5.33%), 67.24 keV (K$\beta_1$, 10.3%), 67.69 keV (K$\beta_5$, 0.24%), 69.07 keV (K$\beta_2$, 3.56%), 69.27 keV (K$\beta_4$, 0.54%) [34]. For L$_1$, L$_2$ and L$_3$ shells X-ray quanta with the mean energy value of 9.5 keV and intensity of 25% were simulated. Auger electrons and X-rays with smaller energy were not simulated, considering the very low probability of reaching the detector for such small energy electrons and X-ray quanta. The simulated energy distributions for the $2\nu$2K and $2\nu$KL decays of $^{184}$Os are shown in Fig. 4.

The energy spectrum accumulated with the Os sample was fitted by the simulated $2\nu$2K ($2\nu$KL) distribution plus a sum of several Gaussian functions to describe the X-ray peaks of Os, Pb, Bi and Po present in the spectrum (see Fig. 5). The fits in the energy interval (55–81) keV, where K X-rays of W are, return area of the effects searched for: $S = (−74 ± 86)$ counts $((−87 ± 83)$ counts) that is no evidence for the $2\nu$2K ($2\nu$KL) decay. The

\footnote{It should be noted that K X-ray peaks of Os are absent in the background data, while the Pb, Bi and Po X-ray peaks are clearly observed in both spectra.}
fits quality is very good: $\chi^2/\text{n.d.f.} = 61.5/81 = 0.759$ ($\chi^2/\text{n.d.f.} = 59.2/81 = 0.731$), where n.d.f. is number of degrees of freedom. According to the recommendations \cite{35}, 78 (65) counts should be accepted as $\lim S$ at 90% C.L.\footnote{All the half-life limits in the present work are given at 90% C.L.} Taking into account the detection efficiency simulated with the help of the EGSnrc package $\eta = 2.911\% (1.635\%)$, the following half-life limits were set for the $2\nu2K$ and $2\nu KL$ decays of $^{184}\text{Os}$ to the ground state of $^{184}\text{W}$:

$$T_{1/2}(2\nu2K) > 3.0 \times 10^{16} \text{ yr},$$

$$T_{1/2}(2\nu KL) > 2.0 \times 10^{16} \text{ yr}.$$
Figure 5: Energy spectra measured with the Os sample (solid histogram) and background data (circles) in the energy region where K X-rays of W are expected from the $2\nu$2K and $2\nu$KL decays of $^{184}$Os. The fit of the data by the background model is shown by solid line, while the excluded effect of the $2\nu$2K decay is presented by dashed line (the excluded distribution is multiplied by a factor 10 for better visibility). Note that the two spectra are not normalized to the time; the acquisition times are 15851 h and 1660 h, respectively.

the ones obtained in the previous work for the $2\nu$2K decay of $^{184}$Os [28] are given in Table 2.

Every 2EC transitions of $^{184}$Os to excited levels of $^{184}$W should be accompanied by X-rays of W, too. Thus, the half-life limits on the $2\nu$2K and $2\nu$KL decays of $^{184}$Os to the 111.2 keV excited level of $^{183}$W were estimated by using the already obtained limit $S$ values for the $2\nu$2K and $2\nu$KL decays to the ground state of $^{184}$W. The detection efficiencies for the decays calculated with the EGSnrc package and the event generator DECAY0 [36, 37].
Table 2: Half-life limits on $2\beta$ processes in $^{184}\text{Os}$ and $^{192}\text{Os}$.

| Transition | Level of daughter nucleus (keV) | $E_{\gamma}$ (keV) | Detection efficiency | $\text{lim } S$ | Experimental limits, $T_{1/2}$ (yr) at 90% C.L. |
|------------|---------------------------------|-------------------|---------------------|-----------------|-----------------------------------------------|
|            |                                 |                   |                     |                 | Present work | Previous result |
| $^{184}\text{Os} \rightarrow ^{184}\text{W}$ | | | | | |
| $2\nu$2K | g.s. | 57 – 69 | 2.911% | 78 | $\geq 3.0 \times 10^{16}$ | $\geq 1.9 \times 10^{14}$ |
| $2\nu$KL | g.s. | 57 – 69 | 1.635% | 65 | $\geq 2.0 \times 10^{16}$ | |
| $2\nu$2K | $2^+ 111.2$ | 57 – 69 | 3.487% | 78 | $\geq 3.6 \times 10^{16}$ | $\geq 3.1 \times 10^{15}$ |
| $2\nu$KL | $2^+ 111.2$ | 57 – 69 | 1.959% | 65 | $\geq 2.4 \times 10^{16}$ | $\geq 3.1 \times 10^{15}$ |
| $2\nu$EC | $2^+ 111.2$ | 111.2 | 0.340% | 37 | $\geq 7.3 \times 10^{15}$ | $\geq 3.1 \times 10^{15}$ |
| $2\nu$2EC | $2^+ 903.3$ | 903.3 | 1.230% | 4.9 | $\geq 2.0 \times 10^{17}$ | $\geq 3.2 \times 10^{16}$ |
| $2\nu$EC | $0^+ 1002.5$ | 891.3 | 2.397% | 6.8 | $\geq 2.8 \times 10^{17}$ | $\geq 3.8 \times 10^{17}$ |
| $2\nu$2EC | $2^+ 1121.4$ | 757.3 | 0.802% | 6.2 | $\geq 1.0 \times 10^{17}$ | $\geq 6.9 \times 10^{16}$ |
| $2\nu$KL | $(0^+)$ 1322.2 | 903.3 | 1.056% | 4.9 | $\geq 1.7 \times 10^{17}$ | |
| $2\nu$2L | $2^+ 1386.3$ | 1275.1 | 0.967% | 26 | $\geq 3.0 \times 10^{16}$ | |
| $2\nu$2L | $3^+$ 1425.0 | 903.3 | 0.518% | 4.9 | $\geq 8.4 \times 10^{16}$ | |
| $2\nu$2L | $2^+ 1431.0$ | 1319.8 | 1.002% | 18 | $\geq 4.4 \times 10^{16}$ | |
| $0\nu$2K | g.s. | 1313.1 – 1314.5 | 1.838% | 9.0 | $\geq 1.6 \times 10^{17}$ | $\geq 2.0 \times 10^{17}$ |
| $0\nu$KL | g.s. | 1370.5 – 1373.8 | 1.827% | 11 | $\geq 1.3 \times 10^{17}$ | $\geq 1.3 \times 10^{17}$ |
| $0\nu$2L | g.s. | 1427.9 – 1433.1 | 1.833% | 20 | $\geq 7.3 \times 10^{16}$ | $\geq 1.4 \times 10^{17}$ |
| $0\nu$2K | $2^+ 111.2$ | 1201.9 – 1203.3 | 1.911% | 20 | $\geq 7.6 \times 10^{16}$ | $\geq 3.3 \times 10^{17}$ |
| $0\nu$KL | $2^+ 111.2$ | 57 – 69 | 1.584% | 65 | $\geq 1.9 \times 10^{16}$ | |
| $0\nu$2EC | $2^+ 903.3$ | 903.3 | 1.019% | 4.9 | $\geq 1.7 \times 10^{17}$ | $\geq 2.8 \times 10^{16}$ |
| $0\nu$2EC | $0^+ 1002.5$ | 310.6 – 312.0 | 3.773% | 14 | $\geq 2.1 \times 10^{17}$ | $\geq 3.5 \times 10^{17}$ |
| $0\nu$2EC | $2^+ 1121.4$ | 757.3 | 0.736% | 6.2 | $\geq 9.4 \times 10^{16}$ | $\geq 6.4 \times 10^{16}$ |
| $0\nu$KL | $(0^+)$ 1322.2 | 903.3 | 1.045% | 4.9 | $\geq 1.7 \times 10^{17}$ | $\geq 2.8 \times 10^{16}$ |
| $0\nu$2L | $2^+ 1386.3$ | 1275.1 | 0.966% | 26 | $\geq 3.0 \times 10^{16}$ | $\geq 6.7 \times 10^{16}$ |
| $0\nu$2L | $(3^+)$ 1425.0 | 903.3 | 0.517% | 4.9 | $\geq 8.4 \times 10^{16}$ | |
| Resonant $0\nu$2L | $2^+ 1431.0$ | 1319.8 | 1.005% | 18 | $\geq 4.4 \times 10^{16}$ | $\geq 8.2 \times 10^{16}$ |
| $2\nu$EC$\beta^+$ | g.s. | 511 | 7.526% | 58 | $\geq 1.0 \times 10^{17}$ | $\geq 2.5 \times 10^{16}$ |
| $2\nu$EC$\beta^+$ | $2^+ 111.2$ | 511 | 7.271% | 58 | $\geq 1.0 \times 10^{17}$ | $\geq 2.5 \times 10^{16}$ |
| $0\nu$EC$\beta^+$ | g.s. | 511 | 7.403% | 58 | $\geq 1.0 \times 10^{17}$ | $\geq 2.5 \times 10^{16}$ |
| $0\nu$EC$\beta^+$ | $2^+ 111.2$ | 511 | 7.191% | 58 | $\geq 9.9 \times 10^{16}$ | $\geq 2.4 \times 10^{16}$ |
| $^{192}\text{Os} \rightarrow ^{192}\text{Pt}$ | | | | | |
| $2\beta^-(2\nu+0\nu)$ | $2^+ 316.5$ | 316.5 | 4.820% | 45 | $\geq 2.0 \times 10^{20}$ | $\geq 5.3 \times 10^{19}$ |
are slightly higher than those relative to the g.s. transitions, leading to a slightly higher half-life limits on the decays (the limits are presented in Table 2).

Another approach to search for 2EC transitions of $^{184}$Os to excited levels of $^{184}$W is based on the analysis of the experimental data in a region where $\gamma$ peaks after de-excitation of $^{184}$W are expected. For example, Fig. 6 shows a part of the energy spectrum measured with the Os sample where a $\gamma$ peak with the energy 111.2 keV after the 2$\nu$2EC decay of $^{184}$Os to the 111.2 keV excited level of $^{184}$W is expected. The data were fitted by a

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5 This happens due to a rather big internal electron conversion coefficient $\alpha = 2.57$ for the $\gamma$ quanta emitted in de-excitation of the 111.2 keV excited level of $^{184}$W. As a result, additional K X-rays are coming from the internal conversion process of the 111.2-keV level de-excitation.

6 It should be stressed that limits obtained in such a way are valid for capture from any shells, not only for 2K or KL decays.
simple model consisting in a linear function (to describe the background) and a Gaussian function centered at 111.2 keV with the width calculated by applying the formula as the effect searched for. The fit returns a peak area \((-2.1 \pm 23.7)\) counts that gives \(\lim S = 37\) counts at 90% C.L. (the excluded peak is shown in Fig. 6 too). The detection efficiency for \(\gamma\) quanta with energy 111.2 keV in the decay was simulated by using the EGSnrc package and the event generator DECAY0 obtaining \(\eta = 0.340\%\). This substantially smaller detection efficiency (in comparison to the X-ray expected in the 2\(\nu\)K and 2\(\nu\)KL decays) can be explained by the rather big internal electron conversion coefficient \(\alpha = 2.57\) for the 111.2-keV \(\gamma\)-transition. As a result, the half-life limit for the 2\(\nu\)EC decay of \(^{184}\)Os to the 111.2 keV excited level of \(^{184}\)W is \(\lim T_{1/2} = 7.3 \times 10^{15}\) yr.

The half-life limits for the decays of \(^{184}\)Os to several excited levels of \(^{184}\)W were obtained in a similar way. The results of the analysis are presented in Table 2.

In the neutrinoless 2EC process in \(^{184}\)Os from K and L shells to the ground state of \(^{184}\)W the energy excess is supposed to be taken away by a bremsstrahlung \(\gamma\) quanta with an energy \(E_\gamma = Q_{23} - E_{b1} - E_{b2}\), where \(E_{bi}\) are the binding energies of the captured electrons on the atomic shells of the daughter W nuclide. For example, to estimate the half-life limit for the 0\(\nu\)2K decay of \(^{184}\)Os, the experimental spectrum measured with the Os sample was fitted in the energy interval 1300 – 1325 keV by the sum of a straight line (background) and a Gaussian-shaped peak with a width determined using the formula (1). The peak position was varied within the energy interval \((1313.8 \pm 0.7)\) keV determined by the accuracy of the \(Q_{23}\) value \([23]\). The maximum peak area returned by the fits \((2.3 \pm 4.1)\) counts, see Fig. 7 (a)) was considered, leading to \(\lim S = 9.0\) counts and \(\lim T_{1/2} = 1.6 \times 10^{17}\) yr. In the case of the 0\(\nu\)KL and 0\(\nu\)2L decays the ranges of the intervals in which the peaks searched for may appear were slightly larger due to the different values of the binding energies on the L\(_1\), L\(_2\) and L\(_3\) shells of the W atom: 12.1 keV, 11.5 keV and 10.2 keV, respectively. The parts of the energy spectrum measured with the Os sample in the regions of interest, the fitting curves, and the excluded peaks are shown in Fig. 7.

The highest sensitivity to the 0\(\nu\)2EC decays of \(^{184}\)Os to the excited levels of \(^{184}\)W (denoted here as \(E^*\)) was achieved either searching for \(\gamma\) peaks with certain energy emitted after de-excitation of the excited levels (as e.g. for the transitions to the levels \(2^+\) 903.3 keV and \(2^+\) 1121.4 keV) or analysing the data in the energy region where the bremsstrahlung \(\gamma\) quanta with the
Figure 7: Parts of the energy spectrum measured with the Os sample where bremsstrahlung $\gamma$ peaks from the $0\nu 2K$ (a), $0\nu KL$ (b), and $0\nu 2L$ (c) captures in $^{184}$Os to the ground state of $^{184}$W are expected. The fits of the data are shown by solid lines, while the excluded peaks are presented by dashed lines. The horizontal lines (above the arrows labelling the energy of the peaks searched for) show the energy uncertainty due to the error of the $Q_{2\beta}$ value of $^{184}$Os plus the difference of the L$_1$, L$_2$ and L$_3$ shells binding energies.

energy $E_\gamma = Q_{2\beta} - E^* - E_{b1} - E_{b2}$ are expected (the case of decays to the levels 2$^+ 111.2$ keV and 0$^+$ 1002.5 keV). The results of the analysis are again presented in Table 2.

Neutrinoless double-electron captures to the excited levels (0)$^+$ 1322.2
keV and $2^+ 1386.3$ keV have been considered as resonant ones \cite{28}. However, double-electron capture to the excited level $(0)^+ 1322.2$ keV is energetically favoured from K and L shells\footnote{Although with a lower probability, capture from higher shells (M, N, O) is also allowed.}, while the $2^+ 1386.3$ keV level favours the electron capture from the L shell. The transitions are far from resonant condition taking into account the rather big difference $\Delta = Q_{23} - E^* - E_{b1} - E_{b2}$: $\Delta = [(49.0 - 50.9) \pm 0.7]$ keV for the $0\nu$KL to the 1322.2-keV level, and $\Delta = [(42.3 - 46.1) \pm 0.7]$ keV for the $0\nu$2L transition to the 1386.3-keV level. Nevertheless, the decays were restricted analysing the experimental data in the energy regions where intense $\gamma$ peaks from the decays are expected. In addition, a half-life limit on the kinematically allowed $0\nu$2L transition of $^{184}$Os to the $(3)^+ 1425.0$ excited level of $^{184}$W with $\Delta = [(3.6 - 7.4) \pm 0.7]$ keV was set in the present work (the results of the analysis are presented in Table 2). Only the transition to the $2^+ 1431.0$ keV level from the L shells remains a candidate for the resonant decay taking into account the region of $\Delta$ values from -3.1 keV to 2.1 keV. A search for the resonant $0\nu$2L transition was realized analysing the experimental energy spectrum gathered with the Os sample in the energy region where a peak with energy 1319.8 keV is expected. However, the obtained limit (see Table 2) is weaker than the previous result due to the rather low detection efficiency for high energy $\gamma$-ray quanta of the used BEGe detector.

The electron capture with positron emission in $^{184}$Os should lead to the emission of two annihilation $\gamma$ quanta with energy 511 keV, causing an extra counting rate in the annihilation peak. The energy spectra taken with the Os sample and the background data were fitted in the energy interval (490 – 540) keV with a model constructed from a 511 keV peak and a linear function to describe background. The peak’s width was a free parameter of the fit to take into account a typically bigger width of the annihilation peak due to the Doppler broadening. The fits of the spectra measured with the Os sample and of the background data in the vicinity of the annihilation peak are shown in Fig. 8. There are $(346 \pm 32)$ counts in the 511 keV peak in the data gathered with the Os sample, while the annihilation-peak area in the background spectrum is $(60 \pm 13)$ counts, which leads to the residual peak area $(−227 \pm 128)$ counts and to the limb $S = 58$ counts after the normalization of the expected number of background counts to the time of measurement with the Os sample. Taking into account the detection efficiency for the annihilation $\gamma$ quanta (slightly different depending on the decay mode: 2$\nu$
or $0\nu$, see Table 2, one can obtain the half-life limits for the $2\nu EC\beta^+$ and $0\nu EC\beta^+$ decays of $^{184}$Os to the ground state of $^{184}$W presented in Table 2. Moreover, limits on the $2\nu EC\beta^+$ and $0\nu EC\beta^+$ decays to the 111.2 keV excited level of $^{184}$W were obtained by using the annihilation-peak analysis, too.

![Energy Spectra](image)

**Figure 8:** Part of the energy spectra measured with the Os sample in the vicinity of the 511 keV annihilation peak. The background data are presented by dots. The fits of the data are shown by the solid lines. Note that the two spectra are not normalized to the time; the acquisition times are 15851 h and 1660 h, respectively.

Most of the limits obtained in the present work are higher than the limits resulted in the previous stage of the experiment [28]. The largest improvement (more than two orders of magnitude) was achieved for the $2\nu 2K$ decay, while the $2\nu KL$ process was investigated for the first time. Also the $2\nu$ double-electron capture to the $(0)^+ 1322.2$ keV, $2^+ 1386.3$ keV and $2^+ 1431.4$ keV excited levels of $^{184}$W were studied for the first time. However, the sensitivity of the present experiment is worse for some decay channels.
emitting high energy $\gamma$ quanta because of the lower detection efficiency of the used BEGe detector.

2.2 Limit on $2\beta^-$ decay of $^{192}$Os to the first excited level of $^{192}$Pt

There is no evidence in the data for a peak at the energy 316.5 keV, expected in the $2\beta^-$ decay of $^{192}$Os to the first $2^+$ 316.5 keV excited level of $^{192}$Pt. To estimate a limit $S$ value for the 316.5 keV peak area, the energy spectrum taken with the Os sample was fitted by a model consisting of a Gaussian peak at energy 316.5 keV (with the width determined by formula (1)) and a linear function to describe the background. The best fit ($\chi^2$/n.d.f. = 31.9/58 = 0.550) achieved in the energy interval (307 – 322) keV returns as peak area: $(8.1 \pm 22.5)$ counts, which corresponds to limit $S = 45$ counts. The fitting

![Energy Spectrum](image)

Figure 9: Part of the energy spectrum measured with the Os sample where a peak with energy 316.5 keV from the $2\beta^-$ decay of $^{192}$Os to the first $2^+$ 316.5 keV excited level of $^{192}$Pt is expected. The fit of the data is shown by the solid line, while the excluded $\gamma$ peak is presented by the dashed line.
curve and the excluded peak are shown in Fig. 9. With detection efficiency of $\eta = 4.820\%$ and $1.526 \times 10^{23}$ number of $^{192}$Os nuclei in the sample, the half-life limit $T_{1/2} \geq 2.0 \times 10^{20}$ yr was obtained, valid for both the $0\nu$ and $2\nu$ decay modes. This limit improves of 4 times the result of the previous experiment (see Table 2).

3 Conclusions

Double-beta processes in $^{184}$Os and $^{192}$Os were searched for over 15851 h using an ultra-low-background broad-energy germanium detector with an active volume of 112 cm$^3$, optimized for low-energy $\gamma$-ray spectrometry. The sample of ultra-pure osmium with a mass of 118 g and with a thickness of $(0.79 - 1.25)$ mm was placed on the detector end-cap. The experiment was carried out in the STELLA facility of the LNGS.

New improved half-life limits on most of the $2\beta$ decay channels of $^{184}$Os were set at the level of $\lim T_{1/2} \sim 10^{16} - 10^{17}$ yr at 90% C.L. A particular progress was achieved for the $2\nu2K$ and $2\nu KL$ processes in $^{184}$Os (the $2\nu KL$ decay was analyzed for the first time) thanks to the substantial improvement of the detection efficiency with the thin Os sample and the use of the BEGe detector with high detection efficiency and good energy resolution to X-ray quanta expected in the decays. The half-lives of the $^{184}$Os $2\nu2K$ and $2\nu KL$ decays were measured to be: $T_{1/2} \geq 3.0 \times 10^{16}$ yr and $T_{1/2} \geq 2.0 \times 10^{16}$ yr, respectively. The newly determined lower half-life limit on the $2\beta^-$ decay of $^{192}$Os to the first excited level of $^{192}$Pt, $\lim T_{1/2} = 2.0 \times 10^{20}$ yr, improves the previous limit by 4 times.

The results of the present experiment are very far from theoretical estimates of the $^{184}$Os decay probability. While there are no estimates of the half-lives for the two-neutrino $2EC$ or $EC\beta^+$ processes, the existing calculations for the half-life of $^{184}$Os concerning the $0\nu2EC$ decay to the 1322.2 keV level of $^{184}$W are at level of $T_{1/2} \sim 10^{28} - 10^{31}$ yr (assuming the mechanism of the decay by exchange of a virtual light Majorana neutrino with $\langle m_{2\beta} \rangle = 0.1$ eV) [21, 38].

The sensitivity of the experiment could be improved by $4 - 6$ orders of magnitude by using osmium enriched in the $^{184}$Os isotope and by increasing the sample mass and the number of BEGe detectors, and further reducing the background level (e.g., by utilization of the background-reduction technique applied in the GERDA experiment [9]). Surprisingly, such an experiment
looks practically realistic despite the very low natural isotopic abundance of $^{184}$Os. Osmium isotopes could be enriched by gas centrifugation [39], at present the only viable technology to produce large amounts of isotopically enriched materials.

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