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Abstract. A search for double-β decay of osmium has been realized for the first time with the help of an ultra-low background HPGe γ detector at the underground Gran Sasso National Laboratories of the INFN (Italy). After 2741 h of data taking with a 173 g ultra-pure osmium sample limits on double-β processes in ^{184}Os have been established at the level of $T_{1/2} \sim 10^{14}$–10^{17} y. Possible resonant double-electron captures in ^{184}Os were searched for with a sensitivity of $T_{1/2} \sim 10^{16}$ y. A half-life limit $T_{1/2} \geq 5.3 \times 10^{19}$ y was set for the double-β decay of ^{192}Os to the first excited level of ^{192}Pt. The radiopurity of the osmium sample has been investigated and radionuclides ^{137}Cs, ^{185}Os and ^{207}Bi were detected in the sample, while activities of ^{40}K, ^{60}Co, ^{226}Ra and ^{232}Th were limited at the $\approx$ mBq/kg level.

1 Introduction

Neutrinoless double-β decay ($0\nu2\beta$) is one of the most promising probes for physics beyond the Standard Model. The process is sensitive to the lepton number violation, the nature of the neutrino (Majorana or Dirac particle), the absolute neutrino mass and the neutrino mass hierarchy (see [1–11] and references therein). In particular, the investigation of neutrinoless double-electron capture ($0\nu2\varepsilon$) and of electron capture with positron emission ($0\nu\varepsilon\beta^+$) could clarify the possible contribution of right-handed currents to the $0\nu2\beta^-$ decay rate [8,12].

Osmium contains two potentially double-β active isotopes: ^{184}Os (decay energy $Q_{2\beta} = 1453.7(0.6)$ keV [13]; isotopic abundance $\delta = 0.02(1)\%$ [14]; allowed decay channels: 2ε and $\varepsilon\beta^+$) and ^{192}Os ($Q_{2\beta} = 412.4(2.9)$ keV [15]; $\delta = 40.78(19)\%$ [14]; $2\beta^-$). The decay schemes of ^{184}Os and ^{192}Os are presented in figs. 1 and 2, respectively. There is a possibility of a resonant enhancement of the 0ν double-electron capture in ^{184}Os to a few excited levels of ^{184}W. The most promising of them is the level $(0)^+$ 1322.2 keV [16].

We have investigated the radiopurity of the osmium sample and realized the first search for double-β processes in ^{184}Os and ^{192}Os with the help of an ultra-low background HPGe γ spectrometer. Preliminary results of the experiment were presented in [17].

2 Measurements, results and discussion

2.1 Experiment

An ultra-pure osmium sample (more than 99.999% purity grade [18]) with a mass of 173 g was used in the experiment. The material was obtained using electron-beam melting of osmium powder with further purification by electron-beam zone refining. The estimated density of the metal is 23 g/cm^3 (the tabulated value is 22.57 g/cm^3 [19]). The sample contains 1.1×10^{20} nuclei of ^{184}Os and 2.2×10^{23} nuclei of ^{192}Os, assuming the natural isotopic composition [14]. The experiment was carried out at the Gran Sasso National Laboratories of the INFN (Italy) with the ultra-low background HPGe detector GeCris with a volume of 465 cm^3. The detector is shielded by lead ($\approx$ 25 cm) and copper ($\approx$ 10 cm). The FWHM energy resolution of the spectrometer is 2.0 keV
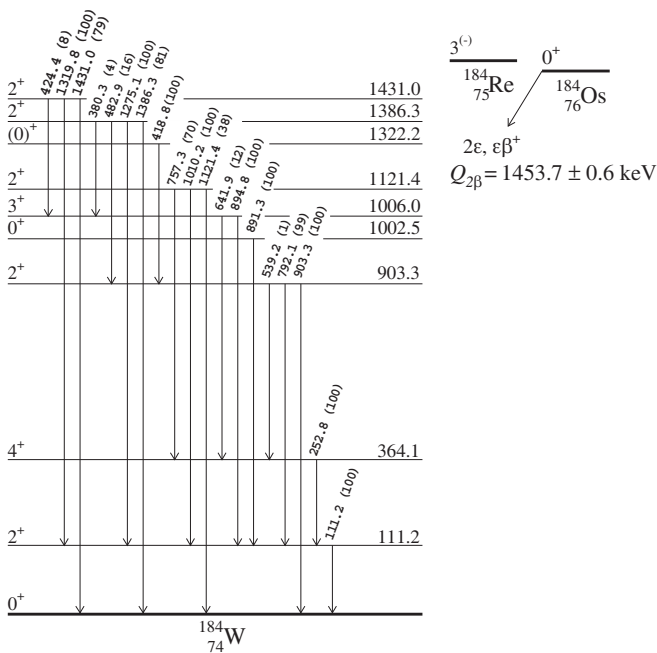
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Fig. 1. Decay scheme of ^{184}Os [20]. The energies of the excited levels and of the emitted γ quanta are in keV (relative intensities of γ quanta, rounded to percent, are given in parentheses).
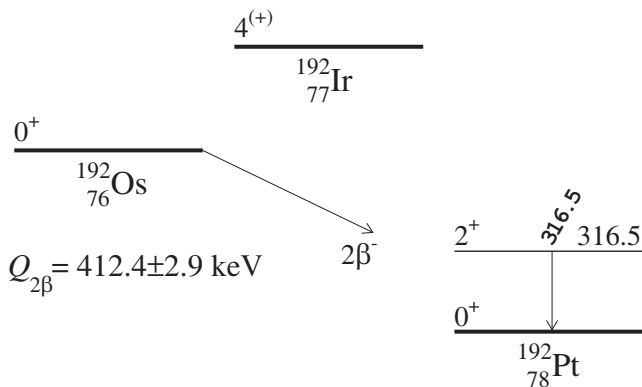

Fig. 2. Decay scheme of ^{192}Os [21].

for the 1333 keV γ quanta of ^{60}Co. The data with the sample were accumulated over 2741 h, while the background spectrum was taken over 1046 h. The spectra are presented in fig. 3.

2.2 Radioactive contamination of the osmium sample

The peaks in the energy distributions can be ascribed to γ quanta of U/Th daughters, ^{40}K, ^{60}Co, ^{137}Cs, ^{185}Os and ^{207}Bi. The response functions of the detector to decays of these nuclides as well as to the double-β processes in the osmium isotopes were simulated by EGSnrc [22] and GEANT4 [23,24] packages with initial kinematics given by the DECAY0 event generator [25]. Both simu-
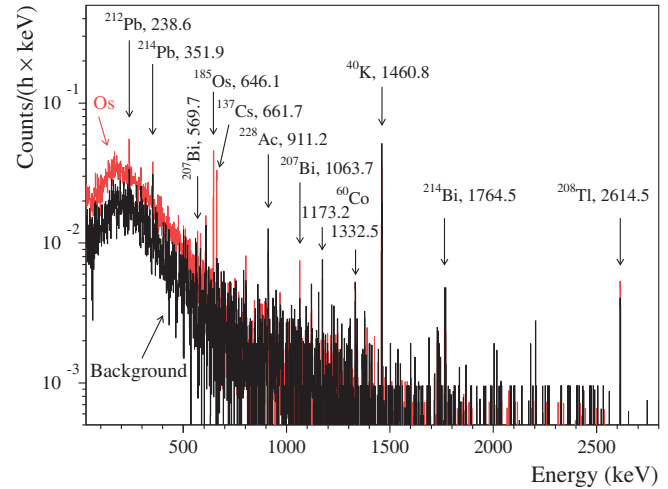

Fig. 3. (Color online) Energy spectra measured with the ultra-low background HPGe γ spectrometer with the osmium sample over 2741 h (Os) and without the sample over 1046 h (Background). The energies of the γ lines are in keV.

Table 1. Radioactive impurities in the osmium sample. The upper limits are presented at 90% CL, the uncertainties are given with 68% CL. The reference date for activities is October 2011.

Chain	Nuclide	Activity (mBq/kg)
^{232}Th	^{228}Ra	≤ 2.0
	^{228}Th	≤ 2.3
^{238}U	^{226}Ra	≤ 0.6
	^{40}K	≤ 1.9
	^{60}Co	≤ 0.1
	^{137}Cs	1.9 ± 0.3
	^{185}Os	3.0 ± 0.3
	^{207}Bi	0.4 ± 0.1

lations gave consistent results[1]. Some excess of events in the 662 keV peak of ^{137}Cs was observed with an activity (1.9 ± 0.3) mBq/kg. The radioactive ^{185}Os (electron capture decay, $T_{1/2} = 93.6$ d [26]) was also detected in the sample with an activity (3.0 ± 0.3) mBq/kg. We assume that this radionuclide was generated before the installation of the sample into the set-up by capture of neutrons on ^{184}Os and by spallation processes induced by cosmic rays on heavier osmium isotopes. The presence of ^{207}Bi at the level of (0.4 ± 0.1) mBq/kg can be explained by contamination of the sample. The activities of ^{137}Cs, ^{185}Os and ^{207}Bi, as well as upper limits on ^{40}K, ^{60}Co and U/Th daughters are presented in table 1.

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[1] For example, the detection efficiency to the γ quanta with an energy of 418.8 keV, expected to be emitted in the resonant $0\nu2\varepsilon$ process in ^{184}Os, is 3.14% (3.12%) with EGSnrc (GEANT4).
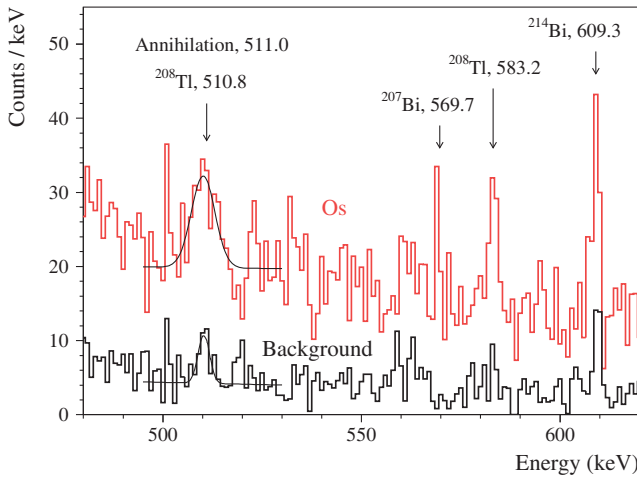
Fig. 4. (Color online) Part of the energy spectra accumulated with the osmium sample over 2741 h (Os sample) and without the sample over 1046 h (Background) in the vicinity of the annihilation peak. Fits of the peaks are shown by the solid lines.

2.3 Search for $\varepsilon\beta^+$ and 2ε processes in ^{184}Os

We do not observe any peaks in the energy distribution accumulated with the osmium sample which could indicate double-β activity of ^{184}Os or ^{192}Os. Therefore only lower half-life limits ($\lim T_{1/2}$) can be set according to the formula

$$\lim T_{1/2} = N \cdot \eta \cdot t \cdot \ln 2 / \lim S,$$

where N is the number of potentially 2β unstable nuclei, η is the detection efficiency (in which the yields of the specific γ quanta in accordance with the scheme of the decay, fig. 1, are included), t is the measurement time, and $\lim S$ is the upper limit on the number of events of the effect searched for which can be excluded at a given confidence level (CL). All the half-life limits and the values of $\lim S$ are presented in this paper at 90% CL.

One positron can be emitted in the $\varepsilon\beta^+$ decay of ^{184}Os with an energy up to (431.7 ± 0.6) keV. The annihilation of the positron should give two 511 keV γ's leading to an extra rate in the annihilation peak. To estimate $\lim S$ for the decay, the energy spectra were fitted in the energy interval (495–530) keV (see fig. 4). There are (92 ± 24) events in the 511 keV peak in the data accumulated with the osmium sample. The area of the annihilation peak in the background spectrum is (24 ± 10) counts, corresponding to (63 ± 26) counts after normalising to the exposure of the osmium sample. The difference in the areas of the annihilation peak with and without sample $((29 \pm 35)$ counts) gives no indication of the effect. In accordance with the Feldman-Cousins procedure [27] we should take $\lim S = 86$ counts which can be excluded at 90% CL. Taking into account almost the same detection efficiency for $2\nu (\eta = 9.0\%)$ and $0\nu (8.9\%)$ modes, we have obtained the following limit on the half-life of ^{184}Os for the 2ν and 0ν modes of $\varepsilon\beta^+$ decay (see also table 2 where the energies of γ quanta used in the analysis, the detection efficiencies
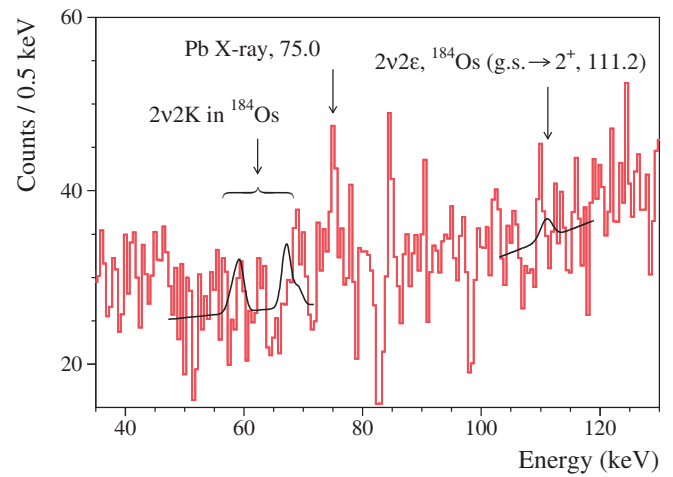

Fig. 5. (Color online) The low-energy part of the energy spectrum accumulated with the osmium sample over 2741 h. The excluded effects of the $2\nu2K$ process in ^{184}Os with the half-life $T_{1/2} = 1.9 \times 10^{14}$ y, and of the $2\nu2\epsilon$ transition to the excited 2^+ level of ^{184}W with an energy of 111.2 keV with the half-life $T_{1/2} = 3.1 \times 10^{15}$ y are shown by the solid lines.

and the values of $\lim S$ are given):

$$T_{1/2}^{(2\nu+0\nu)\varepsilon\beta^+}\left(^{184}\text{Os, g.s.} \to \text{g.s.}\right) \geq 2.5 \times 10^{16} \text{ y.}$$

The $\varepsilon\beta^+$ decay of ^{184}Os is also allowed to the first excited level of ^{184}W with an energy of 111.2 keV. The strongest restrictions on the decay were obtained by analysis of the annihilation peak. Taking into account slightly different detection efficiencies for 2ν (9.0%) and 0ν (8.8%) modes, we have obtained the following limits:

$$T_{1/2}^{2\nu\varepsilon\beta^+}\left(^{184}\text{Os, g.s.} \to 111.2\,\text{keV}\right) \geq 2.5 \times 10^{16} \text{ y,}$$

$$T_{1/2}^{0\nu\varepsilon\beta^+}\left(^{184}\text{Os, g.s.} \to 111.2\,\text{keV}\right) \geq 2.4 \times 10^{16} \text{ y.}$$

In the case of the $2\nu2K$ capture in ^{184}Os, a cascade of X-rays and Auger electrons with individual energies up to 69.5 keV is expected. The most intense X-ray lines of tungsten are [26]: 58.0 keV (the yield of the X-ray quanta is 27.4%), 59.3 keV (47.0%), 67.0 keV (5.4%), 67.2 keV (10.3%) and 69.1 keV (3.6%). To derive a limit on the decay, the energy spectrum was fitted in the energy interval (47–72) keV by the model consisting of five Gaussians (to describe the expected X-ray peaks) plus a polynomial function of the first degree (background). The fit provides the total area of the $2\nu2K$ effect (21 ± 26) counts, which corresponds to $\lim S = 64$ counts (see fig. 5). The detection efficiency of the whole effect in this case is calculated as $\eta = \Sigma_i\, \eta_i$, where η_i are the efficiencies for the specific X lines with energies from 58.0 keV to 69.1 keV. Taking into account $\eta = 0.05\%$[2], one can calculate the following

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[2] We have calculated the efficiency of the set-up to detect the X-rays by using both the EGSnrc and GEANT4 codes, which again give consistent results.

Table 2. Half-life limits on 2β processes in ^{184}Os and ^{192}Os.

Process of decay	Decay mode	Level of daughter nucleus (keV)	E_γ (keV)	Detection efficiency	lim S	Experimental limit at 90% CL (y)
^{184}Os $\rightarrow ^{184}$W						
$\varepsilon\beta^+$	2ν	g.s.	511	9.0%	86	$\geq 2.5 \times 10^{16}$
$\varepsilon\beta^+$	0ν	g.s.	511	8.9%	86	$\geq 2.5 \times 10^{16}$
$\varepsilon\beta^+$	2ν	2^+ 111.2	511	9.0%	86	$\geq 2.5 \times 10^{16}$
$\varepsilon\beta^+$	0ν	2^+ 111.2	511	8.8%	86	$\geq 2.4 \times 10^{16}$
$2K$	2ν	g.s.	58–69	0.05%	64	$\geq 1.9 \times 10^{14}$
2ε	2ν	2^+ 111.2	111.2	0.076%	5.8	$\geq 3.1 \times 10^{15}$
2ε	2ν	2^+ 903.3	903.3	2.3%	17	$\geq 3.2 \times 10^{16}$
2ε	2ν	0^+ 1002.5	891.3	4.5%	2.8	$\geq 3.8 \times 10^{17}$
2ε	2ν	2^+ 1121.4	757.3	1.5%	5.2	$\geq 6.9 \times 10^{16}$
$2K$	0ν	g.s.	1314.1–1315.3	4.1%	5.0	$\geq 2.0 \times 10^{17}$
KL	0ν	g.s.	1371.5–1374.7	4.1%	7.4	$\geq 1.3 \times 10^{17}$
$2L$	0ν	g.s.	1428.9–1433.9	4.2%	7.1	$\geq 1.4 \times 10^{17}$
$2K$	0ν	2^+ 111.2	1202.9–1204.1	4.2%	2.7	$\geq 3.3 \times 10^{17}$
2ε	0ν	2^+ 903.3	903.3	2.0%	17	$\geq 2.8 \times 10^{16}$
2ε	0ν	0^+ 1002.5	891.3	4.1%	2.8	$\geq 3.5 \times 10^{17}$
2ε	0ν	2^+ 1121.4	757.3	1.4%	5.2	$\geq 6.4 \times 10^{16}$
Resonant $2K$	0ν	$(0)^+$ 1322.2	903.3	2.0%	17	$\geq 2.8 \times 10^{16}$
Resonant KL	0ν	2^+ 1386.3	1275.1	2.1%	7.5	$\geq 6.7 \times 10^{16}$
Resonant $2L$	0ν	2^+ 1431.0	1319.8	2.1%	6.1	$\geq 8.2 \times 10^{16}$
^{192}Os $\rightarrow ^{192}$Pt						
$2\beta^-$	$2\nu + 0\nu$	2^+316.5	316.5	3.0%	27	$\geq 5.3 \times 10^{19}$

half-life limit:

$$T_{1/2}^{2\nu 2K}(^{184}\text{Os, g.s.} \rightarrow \text{g.s.}) \geq 1.9 \times 10^{14}\,\text{y}.$$

The sensitivity of the experiment to the $2\nu 2\varepsilon$ decay of ^{184}Os to the 111.2 keV excited level of ^{184}W is estimated from the fit of the energy spectrum accumulated with the osmium sample in the vicinity of the energy 111.2 keV. The fit gives an area for the γ peak of (-4.8 ± 6.1) counts, that leads to lim $S = 5.8$ counts (see fig. 5). The detection efficiency for the 111.2 keV γ quanta, taking into account the fact that the transition from the 111.2 keV excited level to the ground state of ^{184}W is efficiently converted to electrons (the experimental coefficient of conversion is $\alpha_{total} = 2.57$ [20]), was calculated to be 0.076%. Generally speaking, due to the large coefficient of the conversion to electrons, the sensitivity of the experiment should be different for $2K$, KL and $2L$ captures. For example, since both K electrons are already captured in the $2K$ process, the conversion from the K shell is prohibited in the subsequent 111.2 keV deexcitation process. The coefficients of conversion from different atomic shells can be

calculated with the BrIcc program [28][3]. However, the excited 111.2 keV level of ^{184}W has quite a long half-life of 1.251 ns [20] compared to the characteristic atomic relaxation time of $\sim 10^{-15}$ s (see, *e.g.*, [29]). Thus, we consider that the relaxation processes in the W atomic shell after the initial $2\nu 2\varepsilon$ capture are finished before the 111.2 keV deexcitation and we can apply the value $\alpha = 2.57$ for all $2\nu 2\varepsilon$ captures. This gives

$$T_{1/2}^{2\nu 2\varepsilon}(^{184}\text{Os, g.s.} \rightarrow 111.2\,\text{keV}) \geq 3.1 \times 10^{15}\,\text{y}.$$

To estimate limits on the $2\nu 2\varepsilon$ decay of ^{184}Os to the 2^+ 903.3 keV, 0^+ 1002.5 keV and 2^+ 1121.4 keV excited levels of ^{184}W, the energy spectrum accumulated with the osmium sample was fitted in the energy intervals where intense γ peaks from the de-excitation process are expected. For instance, we have obtained the following limit on the $2\nu 2\varepsilon$ decay of ^{184}Os to the excited 903.3 keV level of ^{184}W by the fit of the data in the energy interval (890–910) keV where the 903.3 keV γ peak is expected:

$$T_{1/2}^{2\nu 2\varepsilon}(^{184}\text{Os, g.s.} \rightarrow 903.3\,\text{keV}) \geq 3.2 \times 10^{16}\,\text{y}.$$

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[3] For the total conversion coefficient it gives a value of 2.57, in good agreement with the experimental result [20].
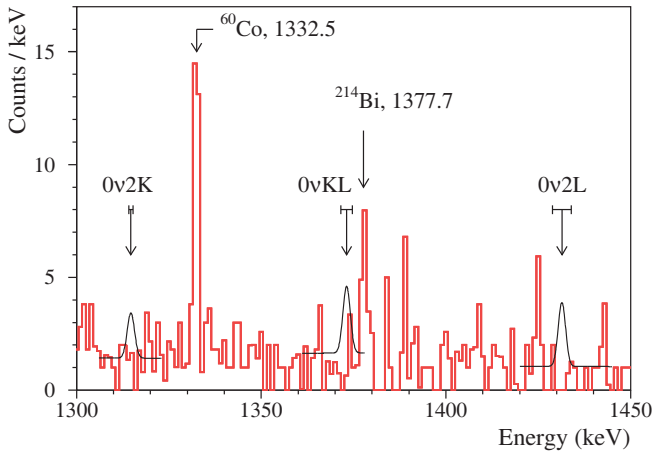
Fig. 6. (Color online) Part of the energy spectrum where the peaks from the $0\nu2\varepsilon$ captures ($2K$, KL and $2L$) in ^{184}Os to the ground state of ^{184}W are expected. The peaks that are excluded at 90% CL are shown by the solid lines.
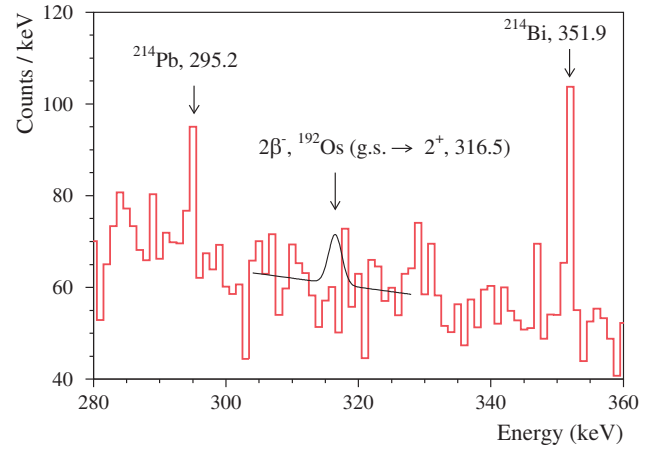

Fig. 7. (Color online) Part of the energy spectrum where the peak from the 2β decay of ^{192}Os to the first excited level of ^{192}Pt is expected. The excluded —at 90% CL— peak is shown by the solid line.

The limits obtained for the $2\nu2\varepsilon$ decay of ^{184}Os to the excited levels of ^{184}W are presented in table 2.

In the neutrinoless double-electron capture in addition to X-rays we suppose that only one bremsstrahlung γ quantum is emitted [30] to take away the rest of the energy, which in the 2ν process is taken by the neutrinos. In the case of the 0ν double-electron capture from K and L shells, the energy of the γ quanta is expected to be equal to $E_\gamma = Q_{2\beta} - E_{b1} - E_{b2}$, where E_{bi} are the binding energies of the captured electrons on the K and L atomic shells of tungsten [26]: $E_K = 69.5$ keV, $E_{L_1} = 12.1$ keV, $E_{L_2} = 11.5$ keV, $E_{L_3} = 10.2$ keV. Therefore, the expected energies of the γ quanta for the $0\nu2\varepsilon$ capture in ^{184}Os to the ground state of ^{184}W are $E_\gamma = (1314.7 \pm 0.6)$ keV for $2K$, $E_\gamma = (1373.1 \pm 1.6)$ keV for KL and $E_\gamma = (1431.4 \pm 2.5)$ keV for $2L$. There are no clear peaks in the data with these energies (see fig. 6). We have estimated the values of $\lim S$ by fitting the data in the energy intervals with the simple model: a Gaussian function (to describe the peaks searched for) plus a polynomial function (continuous background). Taking into account the calculated efficiencies to detect γ quanta with energies of $\approx (1.31-1.43)$ MeV ($\approx 4.1\%-4.2\%$), we set the following limits on the processes searched for:

$$T_{1/2}^{0\nu2K}(^{184}\text{Os, g.s.} \rightarrow \text{g.s.}) \geq 2.0 \times 10^{17} \text{ y},$$

$$T_{1/2}^{0\nu KL}(^{184}\text{Os, g.s.} \rightarrow \text{g.s.}) \geq 1.3 \times 10^{17} \text{ y},$$

$$T_{1/2}^{0\nu2L}(^{184}\text{Os, g.s.} \rightarrow \text{g.s.}) \geq 1.4 \times 10^{17} \text{ y}.$$

The $Q_{2\beta}$ energy of ^{184}Os allows also the population of several excited levels of ^{184}W. The limits obtained for the processes of $0\nu2\varepsilon$ decay to the ground and to the excited levels of the daughter nuclei are presented in table 2.

2.4 Resonant double-electron capture in ^{184}Os

The neutrinoless double-electron capture of ^{184}Os to the excited levels of ^{184}W with energies of 1322.2 keV,

1386.3 keV and 1431.0 keV may occur with higher probability due to a possible resonant enhancement. The double capture on the $(0)^+$ 1322.2 keV level was considered as the most promising with a half-life in the range $(7 \times 10^{26}-2 \times 10^{27})$ y for a Majorana neutrino mass of 1 eV [16][4]. Limits on resonant double-electron capture in ^{184}Os from K and L shells were obtained by analyzing the experimental spectrum in the energy intervals where the strongest γ lines from the de-excitation of these levels are expected. For example, the greatest sensitivity to the resonant $2K$ capture in ^{184}Os to the excited $(0)^+$ level of ^{184}W with an energy of 1322.2 keV is obtained by searching for the de-excitation γ-ray with an energy 903.3 keV, yielding the limit

$$T_{1/2}^{Res\ 0\nu2K}(^{184}\text{Os, g.s.} \rightarrow 1322.2 \text{ keV}) \geq 2.8 \times 10^{16} \text{ y}.$$

Limits on the resonant double-electron captures to the excited levels 2^+ 1386.3 keV and 2^+ 1431.0 keV were obtained in a similar way. The half-life limits on the resonant 2ε processes in ^{184}Os are presented in table 2.

2.5 Double-β^- decay of ^{192}Os

To set a limit on the $2\beta^-$ transition of ^{192}Os to the 2^+ 316.5 keV excited level of ^{192}Pt, the energy spectrum was fitted by a straight line (which represents the background model) and a Gaussian at 316.5 keV. The energy spectrum in the vicinity of the peak is presented in fig. 7. Taking into account the detection efficiency ($\eta = 3.0\%$ both for the 2ν and 0ν modes of the decay), we have obtained the following limit:

$$T_{1/2}^{(2\nu+0\nu)2\beta^-}(^{192}\text{Os, g.s.} \rightarrow 316.5 \text{ keV}) \geq 5.3 \times 10^{19} \text{ y}.$$

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[4] However, following a recent high-precision measurement of the Q value for the double-β decay of ^{184}Os [13], the authors have concluded that the half-life of the transition exceeds 1.3×10^{29} y.

3 Conclusions

The radiopurity of an osmium sample has been investigated and the first experiment to search for 2β processes in ^{184}Os and ^{192}Os was carried out by using ultra-low background HPGe γ spectrometry. After 2741 h of data taking with a 173 g ultra-pure osmium sample limits on double-beta processes in ^{184}Os have been established at the level of $T_{1/2} \sim (10^{14}$–$10^{17})$ y. Possible resonant neutrinoless double-electron captures in ^{184}Os to the excited 1322.2 keV, 1386.3 keV and 1431.0 keV states of ^{184}W are bound at the level of $\lim T_{1/2} \sim 10^{16}$ y. The $2\beta^-$ decay of ^{192}Os to the first excited level of ^{192}Pt is restricted to $T_{1/2} \geq 5.3 \times 10^{19}$ y at 90% CL.

The osmium sample has subsequently been placed in a well-type ultra-low background HPGe detector especially designed for low energy γ-ray spectrometry[5]. In such a way we expect to improve the sensitivity of the experiment at least to the $2\nu2K$ decay, which is the most probable double-beta process in ^{184}Os. Further progress can be achieved by using osmium enriched in ^{184}Os, although this will require new enrichment techniques to be developed.

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[5] The main goal of this stage of the experiment is to search for α decay of ^{184}Os and ^{186}Os to the excited levels of their daughter nuclei (see [17] where an indication for α decay of ^{184}Os to the first excited level (103.5 keV) of ^{180}W with the half-life $T_{1/2} \sim 5 \times 10^{14}$ y was obtained).