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SEARCH FOR DOUBLE β DECAYS OF ^{96}Ru AND ^{104}Ru WITH HIGH PURITY Ge γ SPECTROMETRY

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Experiment to search for double β decay of ^{96}Ru and ^{104}Ru is in progress in the underground Gran Sasso National Laboratories of the INFN (Italy) with the help of ultra-low background high purity (HP) Ge γ spectrometry. After 2162 h of data taking with 473 g ruthenium sample in low-background set-ups with HP Ge detectors, new improved limits on 2β processes in ^{96}Ru and ^{104}Ru have been established on the level of 10^{18}–10^{19} yr.

Keywords: double beta decay, ^{96}Ru, ^{104}Ru.

Introduction

Studies of double beta (2β) decay give important information about properties of neutrino and weak interactions [1]. Measurement of neutrinoless (0ν) double beta decay could define the nature of neutrino (Majorana or Dirac) and the absolute scale of the effective neutrino mass. Neutrinoless mode of 2β decay is forbidden in the Standard Model (SM) as violating the lepton number conservation. At the same time, two-neutrino (2ν) mode is fully allowed in the SM. However, as a second-order weak process, 2ν2β decay occurs at extremely low rates. Experimental efforts during almost 75 years have mainly been dedicated to the investigations of $2\beta^-$ decay leading to the observation of the two neutrino $2\beta^-$ decay in 10 isotopes with half-lives in the range of 10^{18} - 10^{24} yr. Only limits on the level up to 10^{23} - 10^{25} yr were set in the most sensitive experiments on neutrinoless (0ν) $2\beta^-$ decay. The only one positive evidence of the $0\nu2\beta^-$ decay of ^{76}Ge has been reported with $T_{1/2} \approx 2 \cdot 10^{25}$ yr [2]. For 2ε, $\varepsilon\beta^+$ and $2\beta^+$ processes, even allowed in the Standard Model the neutrino accompanied mode is still not observed.

^{96}Ru is one of only six potentially $2\beta^+$ active nuclei [3]. The energy of 2β decay is $Q_{2\beta}$ = (2718 ± ± 8) keV [4]. The decay scheme of ^{96}Ru is presented in Fig. 1 [5, 6]. ^{96}Ru has considerably high natural abundance: δ = 5.54 % [7]. It should be also noted that in the 0ν2ε process with capture of one electron from the K shell and a second one from one of the L shells, the energy release of (2695 ± 8) keV is close to the energy of the excited level of ^{96}Mo with E_{exc} = = 2700 keV [5,6]. In case of 2ε capture from two L shells the energy release (2713 ± 8) keV is equal, within errors, to the energy of the other excited level 2713 keV. Such a coincidence could give a resonant enhancement of the neutrinoless KL and 2L capture as a result of energy degeneracy [8]. The possibility of the resonant 0ν double electron capture was discussed in [9 - 11] where an enhancement of the decay rate was predicted for the case of coincidence between the released energy and the energy of an excited state.

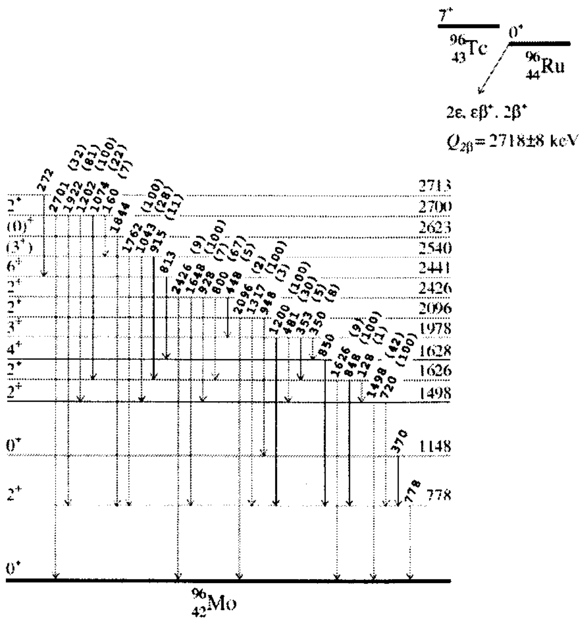

Fig. 1. Decay scheme of ^{96}Ru [5, 6]. Energies of excited levels and emitted γ quanta are in keV (relative intensities of γ quanta are given in parentheses). $Q_{2\beta}$ is the double beta decay energy.

First search for $2\beta^+$ and $\varepsilon\beta^+$ processes in ^{96}Ru was performed in 1985 on the surface of the Earth [12]. A Ru sample of 50 g was measured over 178 h with two low-background HP Ge detectors (110 cm^3 each, operating in coincidence). The limits on $2\beta^+$ and $\varepsilon\beta^+$ decay to the ground state (g.s.) and excited

† Deceased

levels of ^{96}Mo were in the range of $T_{1/2}$ = $= 10^{16}$ - 10^{17} yr. New improved limits on different channels and modes of 2β processes in ^{96}Ru as well as the first limit on ^{104}Ru → ^{104}Pd* decay were obtained recently in our preliminary experiment [13]. Here we report new results of the experiment derived from improved statistics.

Experiment

Ruthenium of 99.99 % grade in form of tablets ≈ ∅16 × 5 mm (with mass ≈ 8.5 g each) produced by powder metallurgy was provided by Heraeus [14]. The total mass of the sample is 473 g. The ruthenium was measured at the Laboratori Nazionali del Gran Sasso (average overburden of ≈ 3600 meters water equivalent) with the HP Ge detector GeCris (468 cm^3) during 986 h, and in the low-background set-up GeMulti with four HP Ge detectors (≈ 225 cm^3 each) during 1176 h. Energy spectra without sample were accumulated over 1046 h with GeCris and 7711 h with GeMulti. Typical energy resolution of the detectors is 2 keV at 1332 keV line of ^{60}Co. The spectra normalized on the time of measurements are presented in Fig. 2.

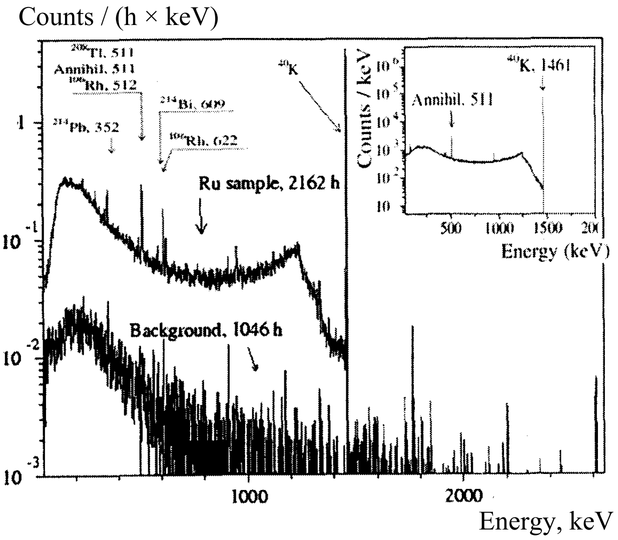

Fig. 2. Sum energy spectra accumulated with the ruthenium sample over 2162 h (Ru sample) and without sample over 1046 h (Background) by the ultra-low background HP Ge γ spectrometry. Inset: simulated spectrum of ^{40}K. The energies of γ lines are in keV.

There is a clear difference between the spectra mainly due to contamination of the ruthenium sample by ^{40}K. The activity of ^{40}K in the sample is (3.3 ± 0.6) Bq/kg. Some excess was also observed in gamma peaks of ^{214}Bi (in equilibrium with ^{226}Ra). Besides, a peak with energy 622 keV and area S = = (215 ± 25) counts was detected (Fig. 3). We suppose this peak is due to presence of radioactive ^{106}Rh, produced by the decay of cosmogenic ^{106}Ru in

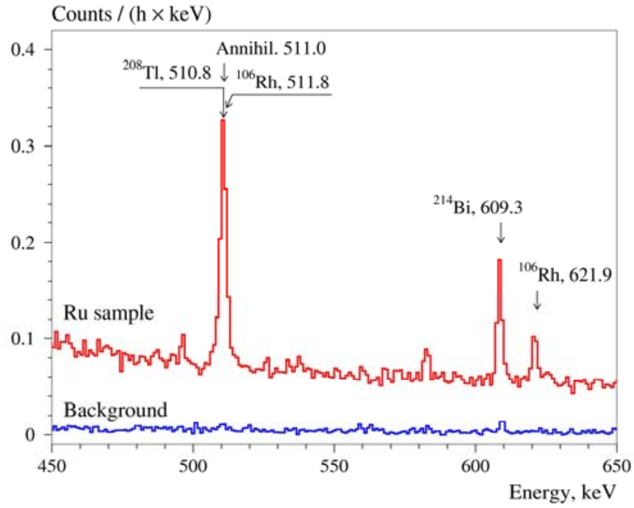

Fig. 3. Fragment of sum energy spectra accumulated with the ruthenium sample over 2162 h (Ru sample), and without sample over 1046 h (Background) by ultra-low background HP Ge γ spectrometers. The energies of γ lines are in keV.

the sample. Activities of ^{40}K, ^{106}Ru, and ^{226}Ra, as well as upper limits on activities of possible contamination of ruthenium by ^{60}Co, ^{137}Cs, U/Th daughters were determined in our previous work [13].

Search for double β processes in ^{96}Ru and ^{104}Ru

Two positrons can be emitted in the $2\beta^+$ decay of ^{96}Ru with the total energy of (674 ± 8) keV. Annihilation of these positrons will give four 511 keV γ's leading to extra rate in the annihilation peak. A part of the spectrum around 511 keV is shown in Fig. 3. In the experimental distribution measured with the Ru sample during 2162 h, there are (1673 ± 71) events in the 511 keV peak. The main source of the 511 keV line is decay of ^{40}K in the sample which produces an annihilation peak through the creation of e^+e^- pairs by 1461 keV γ quanta and small β^+ decay branch. A simulated spectrum of ^{40}K is shown in inset of Fig. 2, where the peak at 1461 keV together with single escape (950 keV), double escape (439 keV) and annihilation (511 keV) peaks can be recognized. The activity of ^{40}K (3.3 ± 0.6) Bq/kg corresponds to (1049 ± 38) counts in the annihilation peak. Some contribution to the annihilation peak give also decays of ^{208}Tl and ^{106}Rh. The difference between the observed and the expected number of counts (173 ± 102) can be ascribed to the $2\beta^+$ decay of ^{96}Ru searched for; it gives no evidence for the effect. In accordance with the Feldman - Cousins procedure [15] recommended by the Particle Data Group [16], the effect cannot be greater than 320 counts at 90 %

of confidence level (C.L.). Taking into account the number of ^{96}Ru nuclei in the sample (N = 1.56 × × 10^{23}) and the registration efficiency (9.6 %), we have estimated a limit on the half-life of ^{96}Ru relatively to $2\beta^+$ decay (sum of 2ν and 0ν modes) as (here and below all the limits are given at 90 % C.L.):

$$T_{1/2}\ (2\beta^+(2\nu+0\nu),\ \text{g.s.} \rightarrow \text{g.s.}) \geq 7.2 \cdot 10^{18}\ \text{yr}.$$

A limit on the $\varepsilon\beta^+$ decay of ^{96}Ru to the ground state of ^{96}Mo was estimated in the same way taking into account the slightly lower detection efficiency for this channel:

$$T_{1/2}\ (\varepsilon\beta^+(2\nu+0\nu),\ \text{g.s.} \rightarrow \text{g.s.}) \geq 4.6 \cdot 10^{18}\ \text{yr}.$$

There are no other peculiarities in the spectrum which could be ascribed to the double β processes in ^{96}Ru and ^{104}Ru. Therefore we can set only lower half-life limits on the decays searched for. To estimate numbers of events, the experimental energy spectrum was fitted in different energy intervals by the sum of components representing the background (internal ^{40}K, ^{207}Bi, U/Th, external γ from the details of the set-up) and the expected models for 2β processes in ^{96}Ru simulated by using the EGS4 code [17] with the initial kinematics given by the DECAY0 event generator [18]. The fits allow us to set limits on the processes of 2β decay in ^{96}Ru presented in the Table.

Half-life limits on 2β processes in ^{96}Ru and ^{104}Ru isotopes.
The energies of the γ lines, which were used to set the $T_{1/2}$ limit, are listed in column 4

Process of decay	Decay mode	Level of daughter nucleus, E, keV	E_γ, keV	$T_{1/2}$, yr		
				Experiment		Theory
				Present work	[13]	
^{96}Ru → ^{96}Mo						
$2\beta^+$	0ν + 2ν	g.s.	511	>7.2 · 10^{18}	>3.9 · 10^{18}	5.1 · 10^{26} - 5.1 · 10^{29} [19 - 24]
$\varepsilon\beta^+$	0ν + 2ν	g.s.	511	>4.6 · 10^{18}	>2.5 · 10^{18}	7.8 · 10^{21} - 2.3 · 10^{29} [19 - 26]
	2ν	2^+ 778	778	>1.8 · 10^{19}	>5.8 · 10^{18}	-
	0ν	2^+ 778	778	>1.8 · 10^{19}	>5.6 · 10^{18}	-
	0ν+2ν	0^+ 1148	778	>1.7 · 10^{19}	>5.3 · 10^{18}	3.8 · 10^{29} - 2.0 · 10^{30} [19, 22]
2K	0ν	g.s.	2670-2686	>2.1 · 10^{19}	>1.2 · 10^{19}	2.8 · 10^{34} [19]
KL	0ν	g.s.	2687-2703	>1.1 · 10^{19}	>6.7 · 10^{18}	-
2L	0ν	g.s.	2705-2721	>2.3 · 10^{19}	>1.2 · 10^{19}	-
2ε	2ν	2^+ 778	778	>1.9 · 10^{19}	>7.2 · 10^{18}	-
		0^+ 1148	778	>1.8 · 10^{19}	>6.4 · 10^{18}	5.2 · 10^{27} [19]
		2^+ 1498	778	>1.6 · 10^{19}	>4.5 · 10^{18}	-
		2^+ 1626	848	>1.9 · 10^{19}	>1.3 · 10^{19}	-
	0ν	2^+ 778	778	>1.9 · 10^{19}	>7.0 · 10^{18}	-
		0^+ 1148	778	>1.8 · 10^{19}	>6.1 · 10^{18}	-
		2^+ 1498	778	>1.6 · 10^{19}	>4.2 · 10^{18}	-
		2^+ 1626	848	>1.8 · 10^{19}	>1.2 · 10^{19}	-
Resonant KL	0ν + 2ν	2700	1922	>2.2 · 10^{19}	>5.0 · 10^{18}	-
Resonant 2L	0ν + 2ν	2713	813	>5.1 · 10^{19}	>1.3 · 10^{19}	-
^{104}Ru→^{104}Pd						
$2\beta^-$	0ν + 2ν	2^+ 556	556	> 1.0 · 10^{20}	> 3.5 · 10^{19}	6.2 · 10^{28} - 1.8 · 10^{29} [27, 28]

Transitions to the excited levels of ^{96}Mo with energies of 2700 and 2713 keV are the most interesting because of a possible resonant enhancement of the process. Analyzing the energy intervals where γ peaks from the de-excitation of the 2700 and 2713 keV levels of ^{96}Ru are expected, the following limits on the resonant double electron capture in ^{96}Ru from K and L, and from two L shells were set:

$$T_{1/2}\ (\text{KL}(2\nu + 0\nu),\ \text{g.s.} \rightarrow 2700\ \text{keV}) \geq 2.2 \cdot 10^{19}\ \text{yr}.$$

$$T_{1/2}\ (\text{2L}(2\nu + 0\nu),\ \text{g.s.} \rightarrow 2713\ \text{keV}) \geq 5.1 \cdot 10^{19}\ \text{yr}.$$

Another isotope of ruthenium, ^{104}Ru, is unstable relatively to $2\beta^-$ decay ($Q_{2\beta}$ = 1301 keV). The natural abundance of ^{104}Ru is comparatively high: η = 18.62 %. We set a limit on the double β^-

transition of ^{104}Ru to the 2^+_1 excited level of ^{104}Pd with the energy of 556 keV. The fit in the energy interval (547 - 572) keV gives the following limit:

$$T_{1/2}\ (2\beta^-(2\nu + 0\nu),\ \text{g.s.} \rightarrow 556\ \text{keV}) \geq 2.3 \cdot 10^{19}\ \text{yr}.$$

All the limits on double β decays of ^{96}Ru and ^{104}Ru obtained in the present work are summarized in the Table. The results of the previous study [13] are given for comparison.

Conclusions

The measurements performed over 2162 h with a 473 g sample of ruthenium with the help of ultra-low background HP Ge γ spectrometers were used to set new limits on double β processes in ^{96}Ru in the range of $T_{1/2} \sim 10^{18-19}$ yr, which improve limits set in our previous measurements [13]; the values are two-three orders of magnitude higher than those obtained in the previous experiment [12].

R&D to purify the material from ^{40}K is in progress. Reducing the potassium contamination by at least one order of magnitude and increasing the exposure up to one-two years × kg could allow to improve the sensitivity of the experiment ≈ 1 order of magnitude in terms of half-life.

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ПОШУК ПОДВІЙНОГО β-РОЗПАДУ ЯДЕР ^{96}Ru ТА ^{104}Ru ЗА ДОПОМОГОЮ НАДНИЗЬКОФОНОВИХ НАПІВПРОВІДНИКОВИХ ГЕРМАНІЄВИХ γ-СПЕКТРОМЕТРІВ

П. Беллі, Р. Бернабей, Ф. Каппелла, Р. Черуллі, Ф. А. Даневич, С. д'Анджело, А. Інчікітті, М. Лаубенштейн, О. Г. Поліщук, Д. Проспері, В. І. Третяк

Експеримент з метою пошуку подвійного β-розпаду ядер ^{96}Ru та ^{104}Ru проводиться у підземній Національній лабораторії Гран Сассо Національного інституту ядерної фізики (Італія) за допомогою наднизькофонових HP Ge γ-спектрометрів. Після 2162 год накопичення даних зі зразком рутенію масою 473 г отримано нові обмеження на періоди напіврозпаду для 2β-процесів в ядрах ^{96}Ru та ^{104}Ru на рівні 10^{18} - 10^{19} р.

Ключові слова: подвійний β-розпад, ^{96}Ru, ^{104}Ru.

ПОИСК ДВОЙНОГО β-РАСПАДА ЯДЕР ^{96}Ru И ^{104}Ru С ПОМОЩЬЮ СВЕРХНИЗКОФОНОВЫХ ГЕРМАНИЕВЫХ γ-СПЕКТРОМЕТРОВ

П. Белли, Р. Бернабей, Ф. Каппелла, Р. Черулли, Ф. А. Даневич, С. д'Анджело, А. Инчикитти, М. Лаубенштейн, О. Г. Полищук, Д. Проспери, В. И. Третяк

Эксперимент по поиску двойного β-распада ядер ^{96}Ru и ^{104}Ru ведется в подземной Национальной лаборатории Гран Сассо Национального института ядерной физики (Италия) с помощью сверхнизкофоновых HP Ge γ-спектрометров. После 2162 ч накопления данных с образцом рутения массой 473 г получены новые ограничения на периоды полураспада для 2β-процессов в ядрах ^{96}Ru и ^{104}Ru на уровне 10^{18} - 10^{19} лет.

Ключевые слова: двойной β-распад, ^{96}Ru, ^{104}Ru.

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