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Search for 2β decay of cerium isotopes with CeCl₃ scintillator

P. Belli^a, R. Bernabei^{a,b,1}, F. Cappella^{c,d}, R. Cerulli^e, F.A. Danevich^f, A. d'Angelo^{c,d}, A. Di Marco^e, A. Incicchitti^c, F. Nozzoli^{a,b}, V.I. Tretyak^f

^aINFN, Sezione di Roma Tor Vergata, I-00133 Rome, Italy

^bDipartimento di Fisica, Università di Roma "Tor Vergata", I-00133 Rome, Italy ^cINFN, Sezione di Roma, I-00185 Rome, Italy

^dDipartimento di Fisica, Università di Roma "La Sapienza", I-00185 Rome, Italy

^eINFN, Laboratori Nazionali del Gran Sasso, 67010 Assergi (AQ), Italy

^fInstitute for Nuclear Research, MSP 03680 Kyiv, Ukraine

Abstract

Double beta processes in ¹³⁶Ce, ¹³⁸Ce and ¹⁴²Ce have been searched for by exploiting the active source approach with the help of a 6.9 g CeCl₃ crystal as a scintillator for the first time at the Gran Sasso National Laboratory (LNGS) of the INFN (Italy). The total measurement time is 1638 h; even such a small exposure has allowed the achievement of some improved half-life limits on 2β decay processes in these isotopes at the level of $10^{16} - 10^{18}$ yr. In particular (90% C.L.): $T_{1/2}^{0\nu\varepsilon\beta^+}(^{136}\text{Ce}) \ge 8.8 \times 10^{16}$ yr, $T_{1/2}^{2\nu\varepsilon\beta^+}(^{136}\text{Ce}) \ge 2.4 \times 10^{16}$ yr, $T_{1/2}^{2\nu2K}(^{136}\text{Ce}) \ge 3.2 \times 10^{16}$ yr, $T_{1/2}^{2\nu2K}(^{138}\text{Ce}) \ge 4.4 \times 10^{16}$ yr, and $T_{1/2}^{2\nu2\beta^-}(^{142}\text{Ce}) \ge 1.4 \times 10^{18}$ yr. Possible future perspectives are also briefly addressed.

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Keywords: CeCl₃ scintillator; double beta decay; low background.

1 INTRODUCTION

The neutrinoless (0ν) double beta (2β) decay of atomic nuclei $(A, Z) \rightarrow (A, Z \pm 2) + 2e^{\mp}$ is forbidden in the Standard Model (SM) since it violates the lepton number by two units [1]; however, it is predicted in many SM extensions. The double beta decay experiments are considered the best way to determine an absolute scale of the neutrino mass, to probe the nature of the neutrino, to establish the neutrino mass hierarchy, to search for the existence of right-handed admixtures in the weak interaction, and to test some other effects beyond the SM. During the last two decades the experimental sensitivity to the $0\nu 2\beta^-$ decay mode reached levels up to $10^{23} - 10^{25}$ yr [1].

¹Corresponding author. *E-mail address:* rita.bernabei@roma2.infn.it (R. Bernabei)

In the SM the two-neutrino (2ν) double beta decay is an allowed rare process; to-date it is the rarest decay observed in direct laboratory experiments. It was detected only for 10 nuclides, and the corresponding half-lives are in the range of $10^{18} - 10^{24}$ yr [2, 3].

So far the experimental investigations in this field have been mainly focused on the $2\beta^-$ decays, while the results for double positron decay $(2\beta^+)$, electron capture with positron emission $(\varepsilon\beta^+)$, and capture of two electrons from the atomic shells (2ε) are much more modest (the most sensitive experiments gave limits on the 2ε , $\varepsilon\beta^+$ and $2\beta^+$ processes at level of $10^{17} - 10^{21}$ yr [2, 4, 5]). This is due to: i) the lower energy releases in the 2ε , $\varepsilon\beta^+$ and $2\beta^+$ processes with respect to those in the $2\beta^-$ decay (which imply lower probabilities for the processes and make difficult the background suppression); ii) the natural abundances of $2\beta^+$ isotopes (typically lower than 1% with only few exceptions). Nevertheless, efforts are in progress in this direction since the studies of the neutrinoless 2ε and $\varepsilon\beta^+$ decays could help to investigate a contribution of right-handed admixtures in weak interaction to the neutrinoless $2\beta^-$ decay [6].

An interesting case is the Cerium; in fact, it offers three double beta decaying isotopes: ¹³⁶Ce, ¹³⁸Ce and ¹⁴²Ce (see Table 1), and the recent development of new scintillating materials containing Ce also allows the exploitation of the efficient "source=detector" approach. In particular, the ¹³⁶Ce is a very interesting isotope since the high energy release allows the $2\beta^+$ decay mode, which is energetically possible only for 6 candidate-nuclei [2]. Moreover, some resonant neutrinoless 2ε captures in ¹³⁶Ce to the excited states of the ¹³⁶Ba are also energetically allowed (see e.g. [7]).

Up to now, the possible 2β decays of Ce isotopes were searched for only in 4 experiments: three of them exploited the active source approach with CeF₃ [8, 9] and GSO(Ce) crystals [10], and one the passive source approach with HPGe detector using the same CeCl₃ as in the present work [11]. The processes were not detected and only limits on half lives were set; for ¹³⁶Ce and ¹³⁸Ce they were in the range of $10^{15} - 10^{17}$ yr; for ¹⁴²Ce they were in the range of $10^{17} - 10^{19}$ yr.

In this paper we report the results obtained in the investigation of the 2β processes in Ce isotopes using for the first time the CeCl₃ crystal as a scintillator to realize the "source=detector" approach.

Table 1 summarizes the main information about the possible 2β decays in the CeCl₃ crystal.

	Energy	Isotopic	Decay	Number of nuclei
Transition	release	abundance	channels	in 100 g of $CeCl_3$
	$({\rm keV})$ [12]	(%) [13]		crystal
$^{136}\text{Ce}{\rightarrow}^{136}\text{Ba}$	2419(13)	0.185(0.002)	$2\varepsilon, \varepsilon\beta^+, 2\beta^+$	4.52×10^{20}
$^{138}\text{Ce}{\rightarrow}^{138}\text{Ba}$	693(10)	0.251(0.002)	2ε	6.13×10^{20}
$^{142}\text{Ce}{\rightarrow}^{142}\text{Nd}$	1416.7(2.1)	11.114(0.051)	$2\beta^-$	2.72×10^{22}

Table 1: Potentially 2β active nuclides present in CeCl₃ crystals.

2 EXPERIMENTAL SET-UP AND MEASUREMENTS

2.1 CeCl₃ crystal scintillator

In recent years scintillating materials containing Cerium have attracted much attention in the developments of new fast scintillators with high density, large light yield, perfect linearity and good energy resolution for applications in fundamental and applied nuclear and particle physics, and in several other fields. In fact – due to the electric dipole allowed 5d-4f transitions in the atomic shell – the Ce³⁺ ions yield fast (10–100 ns) scintillation in the 300–500 nm wavelength range [14, 15]. Scintillating crystals, where Ce is not a dopant but a main constituent, were also studied such as CeF₃, CeBr₃, and CeCl₃.

In particular, the detector used in the present measurements is a cylindrical $\otimes 13 \times 13$ mm CeCl₃ crystal (6.9 g mass), housed – because of its hygroscopicity – in a combined plastic and copper container ($\otimes 16 \times 20$ mm external size). Previous measurements were carried out on the detector radiopurity by using a HPGe detector deep underground at LNGS [11], and main scintillation features were studied in ref. [16].

The present data taking has been performed by equipping the crystal with a $\bigcirc 7.6 \times 10$ cm Tetrasil-B light guide to further reduce the contribution to the background of the used 3" low background photomultiplier (PMT). The detector has been installed deep underground ($\simeq 3600$ m w.e.) in the low background DAMA/R&D set-up [5, 17] at LNGS. It was surrounded by Cu bricks and sealed in a low radioactive air-tight Cu box continuously flushed with high purity nitrogen gas (stored deeply underground for a long time) to avoid presence of residual environmental Radon. The copper box was surrounded by a passive shield made of 10 cm of high purity Cu, 15 cm of low radioactive lead, 1.5 mm of cadmium and 4/10 cm polyethylene/paraffin to reduce the external background. The whole shield has been closed inside a Plexiglas box, also continuously flushed by high purity nitrogen gas.

The signals from the PMT have been recorded – over a time window of 4000 ns and a sampling time of 1 GSample/s – by a 8 bit DC270 Acquiris transient digitizer (TD).

2.2 Calibration

The energy scale and the energy resolution of the CeCl₃ scintillator detector have been measured by using the following γ sources: i) ¹³³Ba (81 keV and 356 keV); ii) ²²Na (511 keV and 1275 keV); iii) ¹³⁷Cs (662 keV); iv) ⁶⁰Co (1173 keV and 1333 keV); see Fig. 1. The linearity of the energy calibration is shown in Fig. 2.

The dependence of the energy resolution of the CeCl₃ detector on the energy can be fitted by the function: $\sigma_{\gamma}/E_{\gamma} = a + b/\sqrt{E_{\gamma}}$, where E_{γ} is the energy of the γ quanta in keV; in particular, for the present assembling: $\sigma_{\gamma}/E_{\gamma} = 0.0121 + 0.237/\sqrt{E_{\gamma}}$.



Figure 1: Energy distributions measured by the CeCl₃ detector in the assembling with light guide when using the following γ sources: (top left) ¹³³Ba; (top right) ²²Na; (bottom left) ¹³⁷Cs; (bottom right) ⁶⁰Co. See text.



Figure 2: Linearity of the energy response of the CeCl₃ scintillator as measured with the γ sources.

2.3 Pulse shape discrimination capability

The pulse shape discrimination (PSD) capability between α and $\gamma(\beta)$ particles has been investigated in ref. [16] with the crystal directly coupled to the PMT; the pulses, recorded by a 2.5 GSample/s TD over a time window of 500 ns, were analysed and a good discrimination

capability was found for the CeCl₃ crystal.

In the present work the possibility to apply the PSD to the production data collected with the CeCl₃ crystal in the present assembling (light guide and a 1 GSample/s TD) has been investigated. In particular, we use the Optimal Digital Filter Method [18]. For each signal f(t), the numerical characteristic of its shape (shape indicator, SI) was defined as SI = $\sum f(t_k) \times P(t_k) / \sum f(t_k)$, where the sum is over the time channels k, starting from the origin of signal and averaging up to 70 ns. The $f(t_k)$ is the digitized amplitude (at the time t_k) of a given signal. The weight function P(t) was defined as: $P(t) = \{f_\alpha(t) - f_\gamma(t)\}/\{f_\alpha(t) + f_\gamma(t)\}$, where $f_\alpha(t)$ and $f_\gamma(t)$ are the reference pulse shapes for α particles and γ quanta, respectively, obtained by summing up shapes of a few thousand γ or α events. To build the γ reference pulse we considered the data collected with the ²²Na and ⁶⁰Co sources, while for the α we considered the data collected in the production runs in the 1.5 – 2.3 MeV energy range², where the α contribution to the experimental energy distribution is dominant (see later). The distributions of the shape indicator for the two classes of events (energies above 1 MeV in γ scale) are depicted in Fig. 3; they are well described by Gaussian functions. Although there is still a statistical



Figure 3: (Color online) - Shape Indicator distributions of the events collected above 1 MeV: α particles (red, lower SI), γ particles (blue, higher SI).

separation between the two populations in the present assembling of the $CeCl_3$ crystal, it is poorer than in ref. [16]; this can be ascribed to the presence of the light guide, which reduces the far UV component of the scintillation light, and to the smaller sampling frequency. Thus, cautiously in the following analyses we will not profit of it.

2.4 Internal contaminations of the CeCl₃

In ref. [16] the residual contaminations in the $CeCl_3$ crystal were investigated and discussed in details; moreover, the activity of the $CeCl_3$ detector (including housing) was also previously

²The α energies are given in terms of calibrations with γ sources, that is in MeV electron equivalent. See also later in the figures.

investigated in ref. [11] on the basis of HPGe γ spectrometry. The present larger exposure and the used approach improve such an investigation in some aspects.

The energy distribution, measured deep underground in the present low background set-up during the 1638 h of data taking, is shown in Fig. 4.



Figure 4: Energy distribution measured deep underground during 1638 h live time inside the multicomponent low background passive shield; the superimposed curve (color online) is the overall background fit estimated as in Figs. 9, 11 of ref. [16], where also the main components were shown. The background fit is well in agreement with the experimental data with exception just of the lowest energy bins; for this reason in the determination of the results on the double electron capture decay channels of ¹³⁶Ce and ¹³⁸Ce isotopes this fit has not been applied.

The measured background has a similar behaviour as in [16] and is dominated by the internal residual contaminants; in particular, the events below $\simeq 1400$ keV are mainly due to the radioactive decays of ¹³⁸La and to the β decays of radioactive isotopes in the ²³⁵U chain, to which the α particles observed above $\simeq 1400$ keV belong. This has also allowed the determination of the relative light yield for α particles as compared with that for γ quanta – the so called α/β ratio³: $\alpha/\beta = 0.070 + 0.050 \times E_{\alpha} - 0.0026 \times E_{\alpha}^2$, where E_{α} is the energy of the α particles in MeV (E_{α} between 5.5 and 7.5 MeV).

In particular, the energy distribution of Fig. 4 has been analysed in order to more precisely determine the 235 U and 138 La content in the crystal, following the same procedures as in ref. [16] where the energy spectrum was compared with Monte Carlo expectations; the values obtained here are given in Table 2. The contents of 238 U and of 232 Th have been determined by analysing time correlated events in the TD time window (4000 ns) as described in the following.

During the present live time 14 double pulses events have been identified; two examples of them have already been shown in Fig. 7 of ref. [16]. These double pulses events cannot be ascribed to random coincidence; in fact, considering the mean rate measured above the energy threshold of $\simeq 50$ keV and the effective TD time window ($\simeq 3650$ ns), the expected number of random coincidences during the present data taking is: 8.3×10^{-5} .

The measured events can mainly be ascribed to $\alpha - \alpha$ time correlated events produced by α decay of ²¹⁹Rn into ²¹⁵Po followed by ²¹⁵Po α decay ($T_{1/2} \sim 1.781$ ms) into ²¹¹Pb (isotopes of the ²³⁵U chain). In particular, the first pulse in each event can be due to the ²¹⁹Rn α decay

³The α/β ratio is defined as the ratio of α peak position in the energy scale measured with γ sources to the energy of α particles (E_{α}). Because γ quanta interact with the detector by β particles, we use the more convenient term " α/β " ratio.

(79.4% α of 6.819 MeV, 12.9% α of 6.553 MeV and 7.5% α of 6.425 MeV), while the second one can be due to the ²¹⁵Po α decay ($\simeq 100\% \alpha$ of 7.386 MeV). Moreover, the number of the measured time correlated events (14) is in agreement with the expected number of $\alpha - \alpha$ events in the effective *TD* time window: 16 $\alpha - \alpha$ events, which can be derived from the ²³⁵U measured contamination in the detector (see Table 2). Thus, upper limits on Bi-Po events can be set.

A Bi-Po event from ²³²Th chain is given by the β decay of ²¹²Bi into ²¹²Po followed by the ²¹²Po α decay ($T_{1/2}=299(2)$ ns; 100% α of 8.785 MeV), while a Bi-Po event from ²³⁸U chain is given by the β decay of ²¹⁴Bi into ²¹⁴Po followed by the ²¹⁴Po α decay ($T_{1/2}=164.3(2.0) \ \mu$ s; \simeq 100% α of 7.687 MeV). The detection efficiency in the effective TD time window is $\simeq 100\%$ for Bi-Po events of the ²³²Th chain and 1.5% for those of the ²³⁸U chain.

Considering the expected $\alpha - \alpha$ events from the ²³⁵U residual contamination as background with respect to the Bi-Po events, the Bayesian approach [19] allows the estimation of the upper limit on the number of Bi-Po events: 6.61 (90% C.L.) from both ²³⁸U and ²³²Th chains. Thus, the activities: $a_{212}B_{i}-212P_{o} \leq 0.16 \text{ mBq/kg}$ and $a_{214}B_{i}-214P_{o} \leq 11 \text{ mBq/kg}$ (90% C.L.) are derived; then, considering the chains in equilibrium, the upper limits on the ²³²Th and on the ²³⁸U contaminations are achieved (see Table 2).

Table 2: Radioactive contamination of the $CeCl_3$ crystal. The upper limits (90% C.L.) arise from the analysis of the Bi-Po events. The chains have been considered in equilibrium.

Decay	Nuclide	Activity (mBq/kg)	Contamination
Chain			from the chain
²³² Th	²²⁸ Th	< 0.16	< 40 ppt
^{235}U	²²⁷ Ac	=284(2)	= 3.52(0.03) ppb
^{238}U	226 Ra	< 11	< 0.9 ppb
	¹³⁸ La	=862(31)	=948(35) ppb

The obtained limits and values are compatible and more precise than those of ref. [11, 16] with the only exception of the 238 U residual contamination, which was significantly higher in the HPGe measurements [11]; this can be ascribed – as already discussed also in ref. [16] – to a few mBq 238 U residual contamination in the housing materials (copper, optical window, etc.) which are of commercial quality.

3 The investigation on the 2β decay modes

As discussed in the following, there are no clear peculiarities in the energy distribution measured by the $CeCl_3$ detector, which could be interpreted as double beta decay of Cerium isotopes. Therefore, only lower half-life limits 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, t is the measuring time, and $\lim S$ is the number of events of the effect searched for which can be excluded at a given confidence level. The values of η in the considered energy interval, of S/η (see later) and of $\lim S$ for each considered decay mode are summarized in Table 3.

The response functions of the CeCl₃ detector for the 2β processes have been simulated with the help of the EGSnrc code [20]; the initial kinematics of the particles emitted in the decays

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Transition	Decay	Decay	Energy	Efficiency	S/η	S_{lim}/η
	channel	mode	range (keV)	η		90%(68%) C.L.
$^{136}\text{Ce} \rightarrow ^{136}\text{Ba}$	$2\beta^+$	0ν	366-384	48%	$-(99 \pm 81)$	55(16)
		2ν	596-840	17%	$-(197 \pm 393)$	465(221)
	$\epsilon \beta^+$	0ν	1328-1504	54%	$-(53 \pm 55)$	46(16)
		2ν	200-1000	86%	$-(102 \pm 160)$	171(75)
	2K	0ν	56-92	79%	(58 ± 48)	137(106)
		2ν	56-92	87%	(53 ± 44)	125(97)
$^{138}\text{Ce} \rightarrow ^{138}\text{Ba}$	2K	0ν	56-92	73%	(64 ± 44)	151(117)
		2ν	56-92	87%	(53 ± 44)	125(97)
$^{142}Ce \rightarrow ^{142}Nd$	$2\beta^{-}$	0ν	1320-1496	84%	$-(26 \pm 36)$	36(15)
		2ν	200-1000	85%	$-(92\pm158)$	176(80)

Table 3: Values of the interesting quantities for the calculation of the $T_{1/2}$ limits on the various decay channels (see later). The overall uncertainty on each η value does not exceed 5%.

has been generated with the DECAY0 event generator [21]. Similar procedure has been used to evaluate the background contribution from the measured residual contaminations.

Preliminarily the experimental sensitivity to each considered double beta decay mode is estimated by using the so-called 1σ approach where the excluded number of the events due to the effect searched for is estimated simply as the square root of the number of background counts in the chosen energy window; if the expected signal is a peak the considered energy interval is $\pm 1 \sigma$ around the expected signal peak. Notwithstanding its simplicity, this method gives the right scale of the experimental sensitivity. Then, a suitable refined determination of the experimental limit is performed.

3.1 Double β processes in ¹³⁶Ce

As already mentioned above, the ¹³⁶Ce can decay through $2\beta^+$, $\varepsilon\beta^+$ and 2ε . The used CeCl₃ crystal contains $N_{136} = 3.12 \times 10^{19}$ ¹³⁶Ce nuclei. The physics for the ¹³⁶Ce is rather rich. Two positrons could be emitted in its $2\beta^+$ decay with energy up to 375 keV; thus, only the ground state of ¹³⁶Ba can be populated. The annihilation of these positrons will give rise to four γ 's of 511 keV. In the $\varepsilon\beta^+$ decay, not only the ground state but also the first excited level of ¹³⁶Ba at 818.5 keV can be populated. Finally, in the 2ε capture, many excited 0⁺ and 2⁺ levels of ¹³⁶Ba can be populated with the subsequent emission of different γ quanta in the de-excitation process.

The experimental sensitivity to the $2\nu 2\beta^+$ decay mode has preliminarily been determined by the 1σ approach. In this case one gets: $S/\eta \leq 422$ events in the energy interval 596 – 840 keV, which is the energy interval of interest for the decay mode (see Fig. 5); this gives a sensitivity: $T_{1/2}^{2\nu 2\beta^+} \geq 9.6 \times 10^{15}$ yr (68% C.L.). Then, a refined analysis is made: the energy distribution is fitted in the energy range 596 – 840 keV by the background model, which takes into account the measured residual contaminations, and the expected signal behaviour as evaluated by EGSnrc code. A $\chi^2/n.d.f. = 60.0/60 = 1.00$ is obtained (see Fig. 5). This approach gives a total number of events, which could be ascribed to the $2\nu 2\beta^+$ decay: $S/\eta = -(197\pm 393)$ counts. This corresponds (in accordance with the Feldman-Cousins procedure [22]) to $\lim S/\eta = 465(221)$ counts at 90%(68%) C.L., giving for the considered process the half-life limit:



Figure 5: (Color online) (Left) Energy distribution around 698 keV where it is expected the signal from the $2\nu 2\beta^+$ decay mode of ¹³⁶Ce following the detection of the two positrons and of one of the four γ 's emitted in the annihilation process. The dashed line (red) shows the best-fit curve and the histogram presents the experimental data; there is also shown (in dot-dashed blue) the obtained 90% C.L. limit multiplied by a factor twenty. (*Right*) Energy distribution around 375 keV where it is expected the signal from the $0\nu 2\beta^+$ decay mode of ¹³⁶Ce when the annihilation γ 's escape the detector. The dashed line (red) shows the best-fit curve and the histogram presents the experimental data; there is also shown (in dot-dashed blue) the obtained 90% C.L. limit multiplied by a factor ten.

In the case of the $0\nu 2\beta^+$ decay mode the maximum sensitivity has been obtained in the energy region around 375 keV. The 1σ approach in the energy interval 366–384 keV gives $S/\eta \leq$ 93 events which corresponds to: $T_{1/2}^{0\nu 2\beta^+} \geq 4.4 \times 10^{16}$ yr (68% C.L.). Then, the experimental data in the 320-436 keV energy interval has been fitted following the same procedure described above, obtaining a $\chi^2/n.d.f. = 38.1/28 = 1.36$ (see Fig. 5). The total number of events which corresponds to $N/\eta = -(99 \pm 81)$ counts which corresponds to $N/\eta = 55(16)$ counts at 90%(68%) C.L. Thus, the following half-life limit is obtained:

$$T_{1/2}^{0\nu2\beta^+}(^{136}\text{Ce}) \ge 0.74(2.53) \times 10^{17} \text{ yr} \qquad 90\%(68\%) \text{ C.L.}$$

In the $\varepsilon\beta^+$ decay, various particles are emitted: positron, X rays and Auger electrons from de-excitations in atomic shells, and γ quanta and/or conversion electrons from de-excitation of a daughter nucleus and from positron annihilation.

The maximum sensitivity to study the $2\nu\varepsilon\beta^+$ decay is obtained in the energy interval 200 - 1000 keV (see Fig. 6). The 1σ approach in this energy interval gives: $S/\eta \leq 164$ events, and $T_{1/2}^{2\nu\varepsilon\beta^+} \geq 2.5 \times 10^{16}$ yr (68% C.L.). Then, the energy spectrum has been fitted in the same energy range according to the aforementioned procedure, obtaining a $\chi^2/n.d.f. = 257/199 = 1.29$ (see Fig. 6). The total number of events which could be ascribed to the $2\nu\varepsilon\beta^+$ decay mode is: $S/\eta = -(102 \pm 160)$ counts which corresponds to $\lim S/\eta = 171(75)$ counts at 90%(68%) C.L. and, thus, to:



Figure 6: (Color online) (Left) Energy distribution in the 200 – 1000 keV energy interval where it is expected the signal from the $2\nu\varepsilon\beta^+$ decay mode of ¹³⁶Ce following the detection of one positron and of the X-rays and Auger electrons. The dashed line (red) shows the best-fit curve and the histogram presents the experimental data; there is also shown (in dot-dashed blue) the obtained 90% C.L. limit multiplied by a factor fifty. (*Right*) Energy distribution around 1397 keV where it is expected the signal from the $0\nu\varepsilon\beta^+$ decay mode of ¹³⁶Ce when the annihilation photons escape the detector. The dashed line (red) shows the best-fit curve and the histogram presents the experimental data; there is also shown (in dot-dashed blue) the obtained 90% C.L. limit multiplied by a factor three.

$$T_{1/2}^{2\nu\varepsilon\beta^+}(^{136}\text{Ce}) \ge 2.4(5.4) \times 10^{16} \text{ yr} \qquad 90\%(68\%) \text{ C.L.}$$

For the $0\nu\varepsilon\beta^+$ decay, the 1σ approach was applied in the energy interval 1371 - 1423 keV ($\pm 1 \sigma$ around the expected signal peak at 1397 keV), however in this case the tail of the nearby composite background structure affect this estimate. Anyhow, in this way we obtain $S/\eta \leq 26$ events, which corresponds to $T_{1/2}^{0\nu\varepsilon\beta^+} \geq 1.6 \times 10^{17}$ yr (68% C.L.). Then, the energy spectrum was fitted in energy range 1328 - 1504 keV following the aforementioned procedure, obtaining a $\chi^2/n.d.f. = 17.1/21 = 0.81$ (see Fig. 6). The total number of events which could be ascribed to the $0\nu\varepsilon\beta^+$ decay mode is $S/\eta = -(53 \pm 55)$ counts; this corresponds to $\lim S/\eta = 46(16)$ counts at 90%(68%) C.L., and to:

$$T_{1/2}^{0\nu\varepsilon\beta^+}(^{136}\text{Ce}) \ge 0.88(2.53) \times 10^{17} \text{ yr} \qquad 90\%(68\%) \text{ C.L}$$

As regards the case of 2ε decay, we have studied just the case of the double electron capture from K shell (binding energy: 37.44 keV). In particular, the $2\nu 2K$ gives a structure around \simeq 75 keV (sum of the binding energies of the two electrons from K shell). The 1σ approach in the energy range 72 - 78 keV ($\pm 1 \sigma$ around the expected signal peak) gives $S/\eta \leq 39$ events which corresponds to: $T_{1/2}^{2\nu 2K} \geq 1.0 \times 10^{17}$ yr (68% C.L.). Then, the energy spectrum was fitted in the energy range 56 - 92 keV by the background model (here – due to the small energy window and the quite smooth energy spectrum – it is assumed as a straight line) and the signal simulated with EGSnrc code (see Fig. 7); a $\chi^2/n.d.f. = 7.4/6 = 1.2$ is obtained. The total number of events which could be ascribed to the $2\nu 2K$ decay mode is: $S/\eta = (53 \pm 44)$ counts which corresponds to $\lim S/\eta = 125(97)$ counts at 90%(68%) C.L., and to:



Figure 7: (Color online) Energy distribution around 75 keV where it is expected the signal from the $2\nu 2K$ decay mode of ¹³⁶Ce following the detection of the X-rays and Auger electrons as well as the signal from the $0\nu 2K$ decay mode. The horizontal dashed line (red) shows the best-fit curve and the histogram presents the experimental data; there is also shown in dot-dashed (blue) line the obtained 90% C.L. limit multiplied by 2 on the $2\nu 2K$ and on $0\nu 2K$ decay modes (both for ¹³⁶Ce and ¹³⁸Ce).

In the $0\nu 2\varepsilon$ process, in addition to the particles described above some other particle(s) should be emitted to take away the energy released instead by the two neutrinos in the $2\nu 2\varepsilon$ process. Usually a (bremsstrahlung) gamma quantum is assumed, and in the following we also suppose deexcitation by one γ quantum. Its energy will be equal to $E_{\gamma} = Q - E_{b1} - E_{b2}$, where E_{b1} and E_{b2} are the binding energies of the first and of the second captured electron on the corresponding atomic shells. For the ¹³⁶Ce this bremsstrahlung gamma quantum will have energy at least of 2344 keV. In the present experimental set-up the low mass of the used CeCl₃ crystal gives a very low peak efficiency for the detection of this gamma quantum of the order of 0.01%, which makes its detection very difficult. However the $0\nu 2K$ can also be identified by the peak at $\simeq 75$ keV, taking into account the different detection efficiency with the respect to the $2\nu 2K$ process. In particular, rescaling by efficiencies the result of the previous fit we obtain for the $0\nu 2K$ decay mode: $S/\eta = (58 \pm 48)$ counts; this corresponds to $\lim S/\eta = 137(106)$ counts at 90%(68%) C.L. and to:

$$T_{1/2}^{0\nu 2K}(^{136}\text{Ce}) \ge 3.0(3.8) \times 10^{16} \text{ yr} \qquad 90\%(68\%) \text{ C.L.}$$

3.2 Double electron capture in ¹³⁸Ce

As already mentioned in Introduction, the ¹³⁸Ce isotope can decay only through 2ε with Q = 693(10) keV. Its natural abundance is $\delta = 0.251\%$; thus, the used CeCl₃ crystal contains $N_{138} = 4.23 \times 10^{19}$ nuclei.

We have studied the 2ε decay of the ¹³⁸Ce analysing the case of the K shell electron capture. In fact, the process of relaxation in the atomic shell of the ¹³⁸Ba after the $2\nu 2K$ capture in ¹³⁸Ce is exactly the same as for the ¹³⁶Ce. Thus, we can use our previous calculations for the ¹³⁶Ce just correcting for the different number of ¹³⁸Ce nuclei. It gives the following limit:

$$T_{1/2}^{2\nu 2K}(^{138}\text{Ce}) \ge 4.4(5.7) \times 10^{16} \text{ yr} \qquad 90\%(68\%) \text{ C.L.}$$

The $0\nu 2K$ capture in ¹³⁸Ce is also similar to the $0\nu 2K$ process in ¹³⁶Ce. However, the energy of the emitted additional γ quantum (618 keV) is lower for the ¹³⁸Ce than for the ¹³⁶Ce (2344 keV). This leads to slightly lower efficiency for the detection of the 75 keV peak. The final values for the $T_{1/2}$ limit is the following:

$$T_{1/2}^{0\nu 2K}(^{138}\text{Ce}) \ge 3.6(4.7) \times 10^{16} \text{ yr} \qquad 90\%(68\%) \text{ C.L.}$$

3.3 2β decays of ¹⁴²Ce

As already mentioned in Introduction, the ¹⁴²Ce isotope can decay through $2\beta^-$ with Q = 1416.7 keV. The natural abundance is $\delta = 11.114\%$, thus the used CeCl₃ crystal contains $N_{142} = 1.87 \times 10^{21}$ nuclei.

In the $2\beta^-$ decay the particles emitted are electrons and neutrinos (in the mode 2ν); thus, to detect this process with a good efficiency it is needed that the decaying isotope is inside the detector itself as in the present case.

The signal expected for the $2\nu 2\beta^-$ decay mode is continuous with a maximum around 400 keV (see Fig. 8, left). The maximum sensitivity to study this decay mode is achieved in the



Figure 8: (Color online) (Left) Energy distribution in the 200 – 1000 keV energy interval where it is mainly expected the signal from the $2\nu 2\beta^-$ decay mode of ¹⁴²Ce following the detection of the electrons. The dashed line (red) shows the best-fit curve and the histogram presents the experimental data; there is also shown (in dot-dashed blue) the obtained 90% C.L. limit multiplied by a factor fifty. (*Right*) Energy distribution around 1417 keV where the signal from the $0\nu 2\beta^-$ decay mode of ¹⁴²Ce is expected. The dashed line (red) shows the best-fit curve and the histogram presents the experimental data; there is also shown (in dot-dashed blue) the obtained 90% C.L. limit multiplied by a factor fifty. (*Right*) Energy distribution around 1417 keV where the signal from the $0\nu 2\beta^-$ decay mode of ¹⁴²Ce is expected. The dashed line (red) shows the best-fit curve and the histogram presents the experimental data; there is also shown (in dot-dashed blue) the obtained 90% C.L. limit multiplied by a factor three.

energy interval 200 – 1000 keV (see Fig. 8, left). The 1 σ approach in this energy interval gives: $S/\eta \leq 165$ events, and $T_{1/2}^{2\nu 2\beta^-} \geq 1.5 \times 10^{18}$ yr (68% C.L.). Then, the energy spectrum has been fitted in the same energy range by the background model and by the $2\beta^-$ signal simulated with EGSnrc code, obtaining a $\chi^2/n.d.f. = 257/199 = 1.28$ (see Fig. 8, left). The total number of events which could be ascribed to the $2\nu 2\beta^-$ decay mode is: $S/\eta = -(92 \pm 158)$ counts which corresponds to $\lim S/\eta = 176(80)$ counts at 90%(68%) C.L. and, thus, to:

$$T_{1/2}^{2\nu 2\beta^-}(^{142}\text{Ce}) \ge 1.4(3.0) \times 10^{18} \text{ yr} \qquad 90\%(68\%) \text{ C.L.}$$

For the $0\nu 2\beta^-$ decay, the 1σ approach was applied in the energy interval 1391 - 1443 keV $(\pm 1 \sigma \text{ around the expected signal peak at 1417 keV})$, however in this case the tail of the nearby composite background structure affects this estimate. Anyhow, in this way we obtain $S/\eta \leq 19$ events, which corresponds to $T_{1/2}^{0\nu 2\beta^-} \geq 1.3 \times 10^{19}$ yr (68% C.L.). Then, the energy spectrum was fitted in the energy range 1320 - 1496 keV by the background model and the signal simulated with EGSnrc code, obtaining a $\chi^2/n.d.f. = 15.3/21 = 0.73$ (see Fig. 8, right). The total number of events which could be ascribed to the $0\nu 2\beta^-$ decay mode is $S/\eta = -(26 \pm 36)$ counts; this corresponds to $\lim S/\eta = 36(15)$ counts at 90%(68%) C.L., and to:

$$T_{1/2}^{0\nu2\beta^-}(^{142}\text{Ce}) \ge 0.67(1.62) \times 10^{19} \text{ yr} \qquad 90\%(68\%) \text{ C.L.}$$

4 Conclusions

A search for double beta processes in Cerium isotopes has been realized by using for the first time a small CeCl₃ crystal scintillator to realize the "source=detector" approach. In spite of the small exposed mass (6.9 g) and of the limited time of measurements (1638 h) some new improved half-life limits have been obtained as summarized in Table 4, where also the results of the previous most sensitive experiments are given for comparison.

Table 4: Half-life limits on 2β processes (ground state to ground state transitions) in Cerium isotopes at 90%(68%) C.L. Theoretical values for 0ν mode are given for the effective neutrino mass of 1 eV. More theoretical results can be found in compilations [2].

Transition	Decay	Decay	Exp. $T_{1/2}$ limit (yr)		Theoretical
	channel	mode	Present work	Previous results	$T_{1/2} ({ m yr})$
$^{136}\text{Ce} \rightarrow ^{136}\text{Ba}$	$2\beta^+$	0ν	$> 0.7(2.5) \cdot 10^{17}$	$> (6.9) \cdot 10^{17} [9]$	$2.4 \cdot 10^{29} [6]$
					$2.7 \cdot 10^{29} \ [23]$
		2ν	$> 0.9(1.8) \cdot 10^{16}$	$> 1.8 \cdot 10^{16} [10]$	$5.2 \cdot 10^{31} \ [6]$
	$\varepsilon \beta^+$	0ν	$> 0.9(2.5) \cdot 10^{17}$	$> 3.8 \cdot 10^{16} [10]$	$4.7 \cdot 10^{26} \ [6]$
					$4.0 \cdot 10^{26} \ [23]$
		2ν	$> 2.4(5.4) \cdot 10^{16}$	$> 2.6 \cdot 10^{15} [11]$	$9.2 \cdot 10^{23} \ [6]$
					$6.0 \cdot 10^{23} \ [24]$
	2K	0ν	$> 3.0(3.8) \cdot 10^{16}$	$> 6.0 \cdot 10^{15} [10]$	—
		2ν	$> 3.2(4.2) \cdot 10^{16}$	$> 2.7 \cdot 10^{16} [8]$	$3.2 \cdot 10^{18} - 6.4 \cdot 10^{19} \ [25]$
					$(3.2 - 5.1) \cdot 10^{21} [26]$
					$9.6 \cdot 10^{21} \ [24]$
$^{138}\text{Ce} \rightarrow ^{138}\text{Ba}$	2K	0ν	$> 3.6(4.7) \cdot 10^{16}$	$> 1.9 \cdot 10^{15} [11]$	_
		2ν	$> 4.4(5.7) \cdot 10^{16}$	$> 3.7 \cdot 10^{16} [8]$	$\geq 2.1 \cdot 10^{26} \ [27]$
$^{142}Ce \rightarrow ^{142}Nd$	$2\beta^{-}$	0ν	$> 0.7(1.6) \cdot 10^{19}$	$> (1.5) \cdot 10^{19} [9]$	$2.8 \cdot 10^{24} \ [28]$
		2ν	$> 1.4(3.0) \cdot 10^{18}$	$> 1.6 \cdot 10^{17} [10]$	$2.2 \cdot 10^{20} - 4.2 \cdot 10^{21}$ [28]
					$2.3 \cdot 10^{23} \ [29]$

This supports the interest in realizing larger mass and longer exposure deep underground with new CeCl₃ crystal scintillators exploiting the active source technique. A further relevant topic is the future preliminary selection of all the materials and of the growing/handling procedures as well as the development of chemical/physical purification techniques in order to improve the radiopurity of the crystal and housing materials. In particular, the ¹³⁸La contamination could be related mainly with the growing process; in fact, the crystal used here was realized just after the growth of a LaCl₃ scintillator. Moreover, the use of a larger *TD* sampling frequency and of different optical read-out should be considered in order to have the possibility to effectively apply the PSD for internal α background reduction when of interest. In addition, the $2\beta^+$ and $\varepsilon\beta^+$ processes can give strong signature when coincidences with other close detectors are investigated. It should be also stressed an advantage of much better energy resolution of CeCl₃ crystal scintillator (FWHM $\approx 5\%$ at 662 keV γ line of ¹³⁷Cs) in comparison with f.e. CeF₃ crystal scintillators ($\approx 18\%$ at the same energy) [8].

Further improvements in sensitivity can be reached by using enriched ¹³⁶Ce, increasing the detection efficiency by using larger CeCl₃ detector, and developing CeCl₃ scintillators with lower level of radioactive contamination. An experiment involving ≈ 100 kg of crystals enriched in ¹³⁶Ce to 20% (5 × 10²⁵ nuclei of ¹³⁶Ce) could reach over 5 years of measurements the half-life sensitivity $T_{1/2} \approx 10^{25}$ yr (supposing zero background). Such a sensitivity could contribute to our understanding of the neutrino mass mechanism and right-handed currents in neutrinoless processes [6]. Particular interest is offered by the $2\nu 2\varepsilon$ decay mode of the ¹³⁶Ce, whose half-life is expected to be in the range $10^{18} - 10^{22}$ yr by theoretical estimates [2]; thus, this process should be observed in such an experiment.

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