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Radioactive contamination of ^{7}LiI(Eu) crystal scintillators

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ABSTRACT

The radioactive contamination of two 26 g samples of low background lithium iodide crystal scintillators doped by europium and enriched in ^{7}Li to 99.9% (^{7}LiI(Eu)) has been investigated by scintillation method at the sea level, and by ultra-low background HPGe γ spectrometry deep underground. No radioactive contamination was detected. In particular, the contamination of the crystal scintillators by ^{226}Ra and ^{228}Th does not exceed 1 mBq/kg, and the activity of ^{40}K is less than 0.5 Bq/kg.

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1. Introduction

The europium doped lithium iodide (LiI(Eu)) scintillator is known since about 70 years [1]. Single LiI(Eu) crystals, in particular enriched in ^{6}Li, are used for a long time for efficient neutron detection (see e.g. Refs. [2–5]). The main properties of LiI(Eu) crystal scintillators are listed in Table 1.

Two decades ago the prospects of LiI(Eu) as a solar neutrino detector were considered [11]. Recently, LiI(Eu) scintillators were proposed to search for the resonant capture of axions possibly emitted in the solar *pp*-cycle by excited ^{7}Li [12]. Taking into account the high natural isotopic abundance of ^{7}Li (92.41% [13]), different Li-containing targets have been already used in several solar axion experiments (see Ref. [14] and references therein). In order to search for solar axions, one of the main requirements for the targets is the achievement of a level of radioactive contamination as low as possible. The radiopurity plays an important role also in other applications of scintillation detectors, including neutron detection.

The radioactive contamination of two samples of lithium iodide crystal scintillators doped by europium and enriched in ^{7}Li was tested by using two different low background techniques: (1) as a scintillator in a low background set-up at sea level; (2) with the help of ultra-low background HPGe γ spectrometry deep underground.

2. Experiments, data analysis and results

2.1. Production of the ^{7}LiI(Eu) scintillators

Two ^{7}LiI(Eu) single crystals ($\oslash 20 \times 20$ mm, with masses of ≈ 26 g each one) grown by the Bridgman–Stockbarger method [15] in the Institute of Scintillation Materials (Kharkiv, Ukraine) were used in the present study. The enrichment of the lithium used for the crystals growth in ^{7}Li was 99.9%; the concentration of the europium in the initial charge was 8×10^{-3} wt%. Due to the high hygroscopicity of the LiI, the ^{7}LiI(Eu) crystals were housed in oxygen free high conductivity (OFHC) copper containers with an external size of $\oslash 26 \times 32$ mm. The opposite sides of the containers were made of quartz windows $\oslash 24 \times 6$ mm. The lateral surface of the crystals was surrounded by light-reflector made of annealed magnesium oxide to improve the light collection. The total masses of the packed scintillators No. 1 and No. 2 (see Fig. 1) are 53.6 and 51.6 g, respectively.

2.2. Low background measurements with ^{7}LiI(Eu) scintillation detector at sea level

The radioactive contamination of the ^{7}LiI(Eu) crystal No. 1 was measured by using the low background set-up installed at

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Table 1
Properties of LiI(Eu) crystal scintillators.

Property	Value	Reference
Density (g/cm^3)	4.08	[6]
Melting point (K)	719	[6]
Index of refraction	1.96	[6]
Hygroscopic	Very	[1,6]
Wavelength of emission maximum (nm) at 300 K	470–475	[2,7]
Light yield (photons/MeV)	$(1.1\text{–}1.5) \times 10^4$	[5,8,9]
Light output (% of NaI(Tl)) for γ rays	30–40	[5,8,10]
Scintillation decay time (μs) under γ ray excitation at 300 K	1.2–1.4	[2,7]

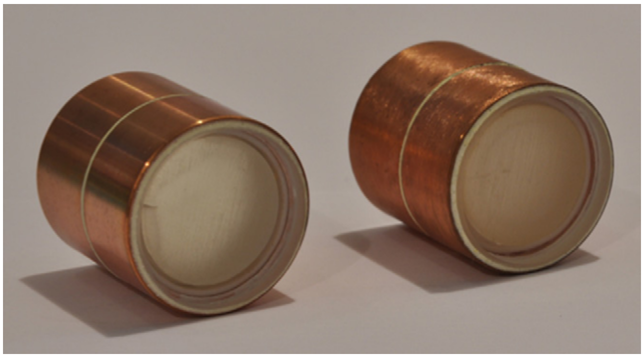

Fig. 1. The enriched ^{7}LiI(Eu) crystal scintillators. (For interpretation of the references to color in this figure caption, the reader is referred to the web version of this paper.)

sea-level in the Institute for Nuclear Research (INR, Kyiv, Ukraine). The scintillator was optically coupled to a 3 in. photomultiplier tube (PMT) Philips XP2412 through a high purity polystyrene light-guide (⊘66 × 120 mm) wrapped in Mylar foil. The window of the scintillator opposite to the light-guide was covered with Mylar foil to improve the light collection. Dow Corning Q2-3067 couplant was used to provide the optical contact. The detector was surrounded by a passive shield made of OFHC copper (5–12 cm thick), and lead (5 cm).

An anti-muon veto counter was installed above the set-up. The detector consists of polystyrene based plastic scintillator (50 × 50 × 8) cm viewed by a low background PMT FEU-125 (Ekran Optical Systems, Russia) with an effective diameter of the photocathode ≈ 15 cm. The anti-muon shield suppressed the background caused by cosmic rays by a factor ≈ (2–3) at the energy ≈ 4 MeV. However, the cosmic rays still provide some background in the ^{7}LiI(Eu) scintillator due to the relatively small spatial angle where the anti-muon counter covers the detector.

An event-by-event data acquisition system was able to record the pulse shape of the ^{7}LiI(Eu) scintillator over a time window of 102 μs (by using a 20 MS/s 12 bit transient digitizer [16]), the arrival time of the signals (with an accuracy of 0.3 μs), and the signals amplitude by a peak sensitive analog-to-digital converter.

The energy scale and the energy resolution of the detector were measured with ^{137}Cs and ^{207}Bi γ sources. The dependence of the energy resolution on energy can be approximated by a function: $\mathrm{FWHM(keV)} = \sqrt{39(3) \times E_\gamma}$, where E_γ is the energy of the γ quanta in keV.

The energy spectrum measured with the ^{7}LiI(Eu) scintillator over 126 h is presented in Fig. 2. The background is caused mainly by external γ quanta from radioactive contamination of the set-up, in particular of the PMT by potassium, thorium and uranium, and by cosmic rays. The response functions of the detector to the external γ quanta were simulated with the GEANT4 package [17] and the event generator DECAY0 [18]. Two exponential functions were adopted to describe the contribution from the cosmic rays. The spectrum measured with the ^{7}LiI(Eu) detector was fitted in the energy interval (220–4400) keV by the model composed by the mentioned components. The result of the fit ($\chi^2/\mathrm{n.d.f.} = 319/197 = 1.62$, where n.d.f. is number of degrees of freedom) is presented in Fig. 2 together with the main components of the background.

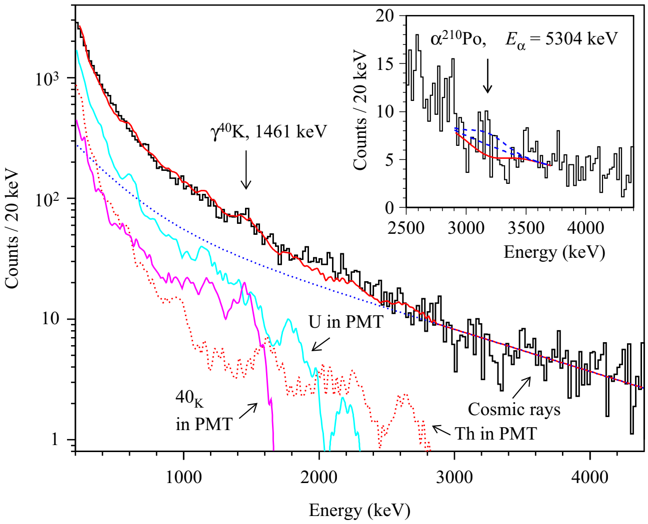

Fig. 2. The energy spectrum measured with the ^{7}LiI(Eu) scintillation detector over 126 h in the low background set-up at the sea level. The fit of the spectrum in the range (220–4400) keV is shown together with the main components of the background. (Inset) High energy part of the spectrum where α peak of ^{210}Po is expected. Fit of the data is shown by the solid line, while the exponential background model and a peak of ^{210}Po excluded at 90% CL are shown by the dashed lines. (For interpretation of the references to color in this figure caption, the reader is referred to the web version of this paper.)

There are no clear peculiarities in the spectrum which could be ascribed to internal trace radioactivity of the ^{7}LiI(Eu) crystal. Therefore, only limits on the contamination of the scintillator by possible radionuclides: ^{40}K, ^{90}Sr–^{90}Y, ^{137}Cs, ^{152}Eu, ^{154}Eu, ^{210}Bi (daughter of ^{210}Pb from the ^{238}U family, which is expected to be out of equilibrium with ^{238}U and ^{226}Ra) were set on the basis of the experimental data. With this aim decays of the expected radionuclides in the ^{7}LiI(Eu) crystal were simulated with the GEANT4 package [17] and the event generator DECAY0 [18]. Then the spectrum was fitted in the energy interval (220–4400) keV by the model composed by the background components (^{40}K, ^{232}Th and ^{238}U in the PMT, and cosmic rays) plus – in turn – the distribution of each considered possible internal radioactive contamination to be estimated. The limits on the activities of the radionuclides are given in Table 2.

A search for the decays of ^{228}Th (daughter of ^{232}Th) in the scintillator was performed with the help of the time-amplitude analysis.[1] To estimate the activity of ^{228}Th, the following sequence of α decays of its daughters was selected: ^{220}Rn ($E_\alpha = 6288$ keV, $T_{1/2} = 55.6$ s)$\rightarrow^{216}$Po ($E_\alpha = 6778$ MeV, $T_{1/2} = 0.145$ s)$\rightarrow$ ^{212}Pb. Taking into account that the α/β ratio in LiI(Eu) scintillator is about 0.6 [10], the energy intervals were chosen as (2900–5200) keV for α particles of ^{220}Ra, and (3100–5500) keV for ^{216}Po. The time interval was taken as (0.01–0.2) s (the efficiency of events selection in the time window is 57%). There are no peculiarities in the selected spectra which can be interpreted as α peaks of ^{220}Rn and ^{216}Po; thus, they were fitted by a straight

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[1] The method of the time-amplitude analysis is described in detail e.g. in Refs. [20,21].

line (which represents the background model) and a Gaussian at 3773 keV (4067 keV) with FWHM = 384 keV (398 keV) to describe an α peak of ^{220}Rn (^{216}Po). The fits give areas of the peaks $S = (-2.1 \pm 3.4)$ counts (^{220}Rn) and $S = (0.7 \pm 1.4)$ counts (^{216}Po). One can combine the values to obtain an averaged value of the area $S = (0.3 \pm 1.3)$ counts, which gives no indication of the effect searched for. Using the procedure proposed by Feldman and Cousins [22,23], we took 2.4 counts as an area of the effect which can be excluded at 90% confidence level (CL), and we set the following limit on the activity of ^{228}Th in the crystal: ≤ 0.4 mBq/kg.

The activity of the ^{226}Ra in the scintillator was estimated by using the same approach to analyze the fast decaying chain ^{214}Bi ($Q_\beta = 3270$ keV [24], $T_{1/2} = 19.9$ m) → ^{214}Po ($E_\alpha = 7687$ keV, $T_{1/2} = 164.3$ μs) → ^{210}Pb. For the first couple (^{214}Bi → ^{214}Po) all events within (100–3300) keV were used as triggers (the efficiency to detect β decay of ^{214}Bi in the detector was calculated to be 94% by using the GEANT4 package), while a time interval (80–500) μs (59% of ^{214}Po decays) and an energy window (3300–7500) keV were set to select α events from ^{214}Po. Only 3 "second" events were selected with energies in the energy interval (4400–5000) keV (α peak from decay of ^{214}Po is expected at the energy 4612 keV with FWHM = 424 keV). To be on the safe side, one can adopt the procedure of Refs. [22,23] considering three detected events and zero background (estimated number of random coincidences is 1×10^{-3}) obtaining 7.4 counts as an effect excluded at 90% CL; thus, a limit ≤ 1.1 mBq/kg is derived for the ^{226}Ra activity in the crystals.

In order to estimate the activity of ^{210}Po (which is expected to be in the scintillator out of equilibrium with ^{226}Ra), the energy spectrum was fitted in the energy interval (2900–3700) keV by an exponential function (which represents the background model) and a Gaussian at 3180 keV with FWHM = 352 keV (expected peak). The fit (see inset in Fig. 2) gives an area of the expected peak (-21 ± 25) counts, which gives no indication of the effect. According to Refs. [22,23], we took 23 events which can be excluded at 90% CL. Using this value we set a limit on the activity of ^{210}Po in the scintillator ≤ 2 mBq/kg.

The summary on limits obtained by the analysis of the experimental data accumulated at the sea level in the low background scintillation set-up is presented in Table 2.

Table 2
Radioactive contamination of ^{7}LiI(Eu) crystal scintillators measured in scintillation mode and by HPGe γ spectrometry. Upper limits are given at 90% CL. The radioactive contamination of LiF(W) [12,14] and SrI$_2$(Eu) [19] crystal scintillators are presented for comparison.

Chain	Nuclide	Activity (mBq/kg)				
		^{7}LiI(Eu)			LiF(W)	SrI$_2$(Eu)
		No. 1 Scint.	No. 1 HPGe	No. 2 HPGe	[12,14]	[19]
^{232}Th	^{228}Ac		≤ 88	≤ 92	$\leq (1.6-20)$	≤ 68
	^{228}Th	≤ 0.4	≤ 28	≤ 58	$\leq (0.6-20)$	6 ± 2
^{238}U	^{226}Ra	≤ 1.1	≤ 19	≤ 39	$\leq 20-3.3(8)$	110 ± 50
	^{210}Pb	≤ 480				≤ 280
	^{210}Po	≤ 2				≤ 60
	^{40}K	≤ 365	≤ 541	≤ 177	$\leq (5.1-66)$	≤ 200
	^{60}Co		≤ 14	≤ 8	$\leq (0.3-2)$	≤ 16
	^{90}Sr–^{90}Y	≤ 160				≤ 90
	^{137}Cs	≤ 22	≤ 25	≤ 37	$\leq (0.2-5)$	53 ± 11
	^{152}Eu	≤ 650	≤ 47	≤ 54		≤ 108
	^{154}Eu	≤ 1430	≤ 42	≤ 51		≤ 67

2.3. Measurements with ultra-low background HPGe γ spectrometer

The radioactive contamination of the ^{7}LiI(Eu) detectors was also tested with the help of an ultra-low background HPGe detector (GeBer, 244 cm^3) installed deep underground (≈ 3600 m w.e.) at the Gran Sasso National Laboratories of the INFN (Italy). To suppress the external background the detector is surrounded by a passive shield made of the OFHC copper (≈ 10 cm thick), low radioactivity lead (ca. 20 cm), and borated polyethylene (≈ 10 cm). The set-up is enclosed in a Plexiglas box and continuously flushed with high purity nitrogen to remove environmental radon. The energy resolution of the GeBer spectrometer is 2.1 keV for the 1332 keV γ line of ^{60}Co.

The samples No. 1 and No. 2 were measured over 788 and 500 h, respectively, whereas the background was accumulated over 3047 h. The energy spectra of the ^{7}LiI(Eu) sample No. 1 and of the background, both normalized to the time of measurements are shown in Fig. 3. As one can see, the spectrum measured with the ^{7}LiI(Eu) sample No. 1 is practically indistinguishable from the background. A similar counting rate was observed for the sample No. 2. The activities of the most probable γ emitting radionuclides in the ^{7}LiI(Eu) detectors (it should be stressed that the measurements are sensitive to the radioactive contamination of the detectors as a whole, including the copper container, quartz windows, glue and reflecting material) were estimated using the following formula:

$$A = \left(\frac{S_{sample}}{t_{sample}} - \frac{S_{bg}}{t_{bg}}\right) \cdot \frac{1}{y \cdot \eta \cdot m} \qquad (1)$$

where S_{sample} (S_{bg}) is the area of a peak in the sample (background) spectrum; t_{sample} (t_{bg}) is the time of the sample (background) measurements; y is the yield of the corresponding γ line [25]; η is the efficiency of the full absorption peak detection; m is the mass of the sample. The detection efficiencies were calculated using the GEANT4 code [17].

As an example we consider here the analysis of the 1460.8 keV γ line of ^{40}K. The area of the peak in the spectrum measured with the ^{7}LiI(Eu) sample No. 1 [background] is (375 ± 22) counts [(1406 ± 43) counts]. Taking into account the yield of the γ quanta: $y = 10.67\%$ and the detection efficiency: $\eta = 1.23\%$,

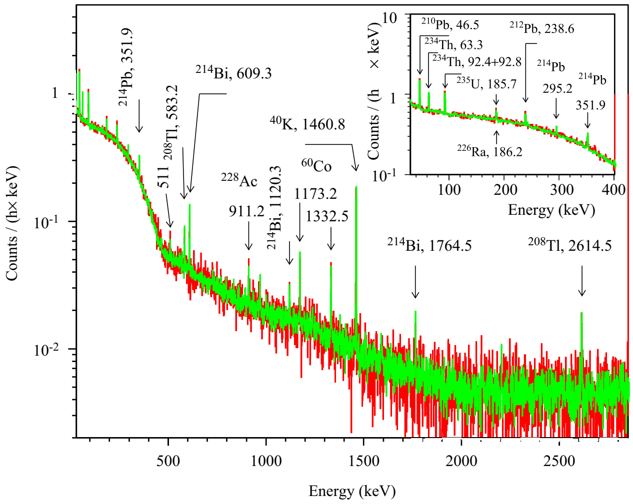

Fig. 3. Energy spectrum accumulated with the ^{7}LiI(Eu) sample No. 1 (red histogram) over 778 h by ultra-low background HPGe γ spectrometer. The background spectrum measured without sample over 3047 h (green histogram) is also shown. (Insert) Low energy part of the spectra. The energies of γ lines are in keV. (For interpretation of the references to color in this figure caption, the reader is referred to the web version of this paper.)

we have obtained the limit ≤ 541 mBq/kg at 90% CL on the activity of ^{40}K in detector No. 1.

The limits on the radioactive contamination of the detectors in ^{60}Co, ^{137}Cs, ^{152}Eu, ^{154}Eu and ^{232}Th/^{238}U daughters were obtained in the same way. The results are summarized in Table 2, where the data of the recent studies of two crystal scintillators, – comparable in chemical properties – LiF(W) and SrI_2(Eu), are given for comparison.

3. Conclusions

The radioactive contamination of two samples of ^{7}LiI(Eu) crystal scintillators has been tested using a low background scintillation counting method at the sea level in the Institute for Nuclear Research (Kyiv, Ukraine), and with the help of ultra-low background γ spectrometry deep underground in the Gran Sasso National Laboratories of the INFN (Italy). No radioactivity was detected in the detectors on the level of sensitivity ~ 1 mBq/kg for ^{226}Ra, ^{228}Th, ^{210}Po, ~ 10 mBq/kg for ^{60}Co, ^{137}Cs and ^{152,154}Eu, $\sim 0.1-1$ Bq/kg for ^{40}K, ^{90}Sr and ^{210}Pb.

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