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Status of LUMINEU program to search for neutrinoless double beta decay of ¹⁰⁰Mo with cryogenic ZnMoO₄ scintillating bolometers

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Abstract. The LUMINEU program aims at performing a pilot experiment on $0v2\beta$ decay of ¹⁰⁰Mo using radiopure ZnMoO₄ crystals enriched in ¹⁰⁰Mo operated as cryogenic scintillating bolometers. Large volume ZnMoO₄ crystal scintillators (~ 0.3 kg) were developed and tested showing high performance in terms of radiopurity, energy resolution and α/β particle discrimination capability. Zinc molybdate crystal scintillators enriched in ¹⁰⁰Mo were grown for the first time by the low-thermal-gradient Czochralski technique with a high crystal yield and an acceptable level of enriched molybdenum irrecoverable losses. A background level of ~ 0.5 counts/(yr keV ton) in the region of interest can be reached in a large detector array thanks to the excellent detectors radiopurity and particle discrimination capability, suppression of randomly coinciding events by pulse-shape analysis, and antico-incidence cut. These results pave the way to future sensitive searches based on the LUMINEU technology, capable of approaching and exploring the inverted hierarchy region of the neutrino mass pattern.

INTRODUCTION

Search for neutrinoless double beta $(0\nu_2\beta)$ decay is considered as the only experimentally viable approach to test the Majorana nature of neutrino and the lepton number conservation. The $0\nu_2\beta$ decay, if mediated by the mass mechanism, is able to measure the effective Majorana neutrino mass and to determine the hierarchy of neutrino masses. In addition, the process can be mediated by right handed currents in weak interactions, existence of massless (or very light) Nambu-Goldstone bosons (majorons), and other effects beyond the Standard Model [1, 2, 3, 4, 5]. The isotope ¹⁰⁰Mo is one of the most promising nuclei to search for $0\nu_2\beta$ decay thanks to the high energy of the decay ($Q_{2\beta} = 3034.40(17)$ keV [6]), comparatively high natural isotopic abundance ($\delta = 9.82(31)\%$ [7]) and the favorable theoretical predictions (see, e.g., [1, 2] and references therein). Here we report a status of the LUMINEU program (Luminescent Underground Molybdenum Investigation for NEUtrino mass and nature) which aims at preparing and construction of a next-generation $0\nu_2\beta$ decay experiment capable to explore the inverted hierarchy region of neutrino mass with cryogenic scintillating bolometers based on radiopure ZnMoO₄ crystals enriched in ¹⁰⁰Mo [8].

DETECTORS DEVELOPMENT

Properties of ZnMoO₄ crystal as cryogenic low temperature scintillator were checked for the first time in [9]. Large volume high optical quality ZnMoO₄ crystal scintillators from deeply purified molybdenum with mass larger than 0.3 kg were then developed in the framework of the LUMINEU project and their optical, luminescent, diamagnetic, thermal and bolometric properties were tested [10]. Cryogenic scintillating bolometers based on ZnMoO₄ crystals show high performance in terms of energy resolution (e.g., FWHM = 6 keV at 2615 keV was reached in [11]), powerful α/β particle discrimination capability (on the level of 15 σ) and a very low level of radioactive contamination [12]. The data on radioactive contamination of a LUMINEU ZnMoO₄ crystal scintillator with mass 334 g are presented in Table 1. The level of thorium and radium contamination fully satisfies the LUMINEU requirements.

TABLE 1. Radioactive contamination of LUMINEU ZnMoO₄ crystal scintillator [12].

Chain	Nuclide (sub-chain)	Activity (µBq/kg)
	¹⁹⁰ Pt	3.8 ± 1.2
²³² Th	²³² Th	≤ 2.3
	²²⁸ Th	≤ 5.3
²³⁵ U	²³⁵ U	≤ 3.3
²³⁸ U	²³⁸ U	≤ 1.8
	²²⁶ Ra	≤ 4.8
	²¹⁰ Po	1271 ± 22

A Zn¹⁰⁰MoO₄ crystal with a mass of 0.17 kg was grown from molybdenum enriched in ¹⁰⁰Mo [13] as an important step of the project. The output of the crystal boule was 84%, while the irrecoverable losses of enriched molybdenum were found to be $\approx 4\%$ in the total production cycle, which demonstrates an important advantage of the purification method and of the low-thermal-gradient Czochralski technique used for the crystal growing. Recently, a large enriched Zn¹⁰⁰MoO₄ crystal boule with mass ≈ 1.4 kg was grown (see Fig. 1) and two detectors were produced. Test of the detectors performance and radioactive contamination is in progress.

BACKGROUND CONDITIONS IN EXPERIMENT WITH Zn¹⁰⁰MoO₄ DETECTORS

To estimate background in conditions of an experiment with $Zn^{100}MoO_4$ scintillating bolometers, a Monte Carlo simulation of 48 $Zn^{100}MoO_4$ detectors (with a mass 495 g, size 0.000×40 mm each) in the EDELWEISS set-up is in progress (some preliminary results can be found in [14]). We simulated contamination of $Zn^{100}MoO_4$ crystals and the nearest to the detectors materials (copper holders, polyethylene terephthalate (PTFE) clamps, reflective foil) by 238 U and 232 Th daughters. We have also simulated cosmogenic nuclides in the $Zn^{100}MoO_4$ crystals and the copper holder. The results of the simulation are summarized in Table 2, which reports also the assumed contaminations. A rejection factor of 99.9% was applied every time when a β event is emitted in sequence with an α event within the



FIGURE 1. Boule of $Zn^{100}MoO_4$ crystal with mass ≈ 1.4 kg produced from molybdenum isotopically enriched in ¹⁰⁰Mo.

time resolution of the bolometer. We also assumed suppression of the bulk and surface ²⁰⁸Tl (²²⁸Th chain) by a factor 10^3 applying the delayed anti-coincidence with a prompt α event from ²¹²Bi [15]. Finally, anticoincidence rejection of background thanks to the multi-detector structure was taken into account. The simulated background energy spectrum and its main components are shown in Fig. 2.

TABLE 2. Monte Carlo simulated background contributions in an experiment with $Zn^{100}MoO_4$ scintillating bolometers installed in the EDELWEISS set-up.

Source of background	Activity (µBq/kg)	Background, counts/(yr keV kg)
²²⁸ Th in Zn ¹⁰⁰ MoO ₄ crystals	10	8.6×10^{-6}
²²⁶ Ra in Zn ¹⁰⁰ MoO ₄ crystals	10	3.1×10^{-8}
⁸⁸ Y in Zn ¹⁰⁰ MoO ₄ crystals	0.3	6.3×10^{-7}
⁵⁶ Co in Zn ¹⁰⁰ MoO ₄ crystals	0.06	6.2×10^{-5}
²³² Th in Zn ¹⁰⁰ MoO ₄ surface layer	0.5	1.2×10^{-5}
²³⁸ U in Zn ¹⁰⁰ MoO ₄ surface layer	2.4	1.5×10^{-4}
²²⁸ Th in copper holders	20	1.3×10^{-6}
²²⁶ Ra in copper holders	70	1.5×10^{-7}
⁵⁶ Cu in copper holders	0.2	6.6×10^{-5}
²²⁸ Th in PTFE clamps	100	9.6×10^{-6}
²²⁶ Ra in PTFE clamps	60	7.5×10^{-7}
²²⁸ Th in reflective foil	100	7.5×10^{-5}
²²⁶ Ra in reflective foil	60	2.1×10^{-5}
Random coincidence of ¹⁰⁰ Mo $2\nu 2\beta$ events	8130	$\approx 10^{-4}$
Total		$\approx 5 \times 10^{-4}$

As it was pointed out in [16] random coincidence of $2\nu 2\beta$ events can be the main background in bolometric $0\nu 2\beta$ experiments with ¹⁰⁰Mo due to the poor time resolution of cryogenic detectors. However, the background can be reduced to the level of $\approx 10^{-4}$ counts/(yr keV kg) with the help of pulse-shape discrimination [17]. Therefore, a total background counting rate $\approx 5 \times 10^{-4}$ counts/(yr keV kg) can be reached, which corresponds to 3 counts/(yr ton) in the region of interest (assuming 6 keV window centered at the $0\nu 2\beta$ peak position).

CONCLUSIONS

Cryogenic scintillating bolometers based on large volume (~ 0.3 kg) ZnMoO₄ crystals, developed in the framework of the LUMINEU project, show excellent performance: a few keV energy resolution, 15 sigma alpha/beta particle discrimination power at the $Q_{2\beta}$ value of ¹⁰⁰Mo. Radioactive contamination of ZnMoO₄ crystal scintillators is very



FIGURE 2. Energy spectra of 48 Zn¹⁰⁰MoO₄ detectors in EDELWEISS set-up simulated with the help of GEANT4 package. (Inset) The main components of background are shown: (1) surface contamination of $Zn^{100}MoO_4$ crystals, (2) radioactive contamination of the reflecting foil surrounding the crystal, (3) bulk contamination of $Zn^{100}MoO_4$ crystals, contamination of copper holders (4) and PTFE clamps (5).

low thanks to the deep purification of molybdenum: activity of 228 Th and 226 Ra does not exceed 5 μ Bq/kg. Enriched Zn¹⁰⁰MoO₄ crystals with a mass of 0.17 kg and 1.4 kg were grown for the first time by the low-thermal-gradient Czochralski technique. The production cycle provided a high yield of the crystal boule (more than 80% of the initial charge) and an acceptable level of irrecoverable losses of molybdenum ($\approx 4\%$). A background level of ~ 0.5 counts/(yr keV ton) in the region of interest can be reached in a large detector array thanks to the excellent detectors radiopurity and particle discrimination capability, suppression of randomly coinciding events by pulse-shape analysis, and anticoincidence cut. It should be stressed that also Li₂MoO₄ cryogenic scintillating bolometers were developed and tested as promising detectors for a high sensitivity experiment with ¹⁰⁰Mo [18]. These results pave the way to future sensitive searches based on the LUMINEU technology, capable of exploring the inverted hierarchy region of the neutrino mass pattern. The LUMINEU activity is part of the CUPID project [19, 20], a proposed bolometric tonnescale experiment to be built as a follow-up to CUORE and exploiting as much as possible the CUORE infrastructures.

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