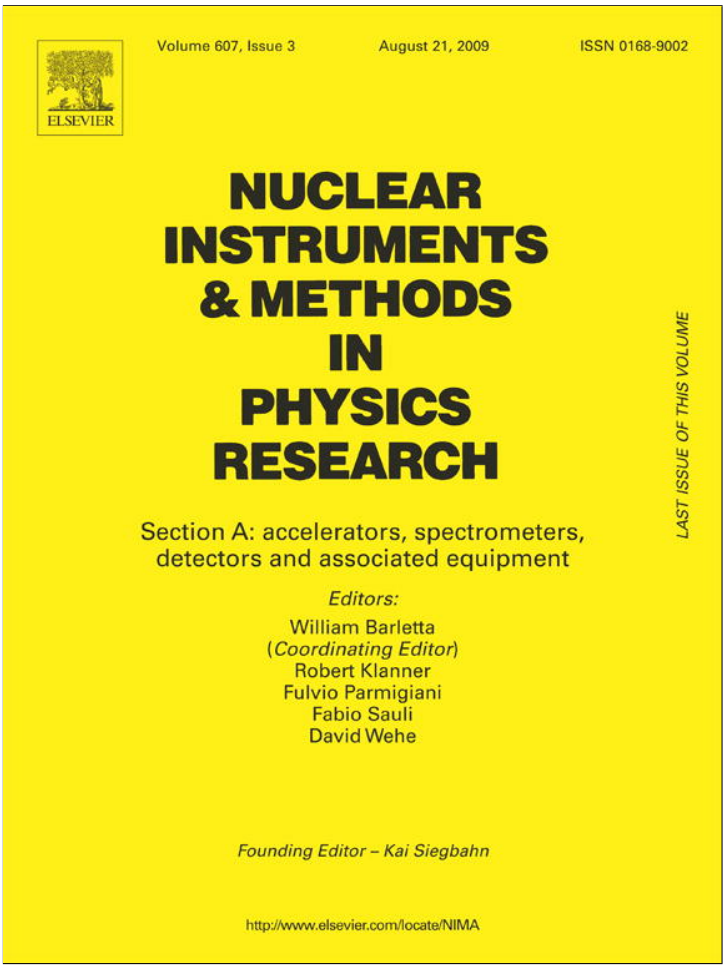

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Intrinsic radiopurity of a Li_2MoO_4 crystal

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ABSTRACT

The radiopurity of a lithium molybdate (Li_2MoO_4) single crystal, a potential detector for the search for the neutrinoless double beta decay of ^{100}Mo, was studied deep underground at the Laboratori Nazionali del Gran Sasso (LNGS) (Italy). Data were collected with a crystal sample with a mass of 33.95 g, using a high purity germanium (HPGe) detector with a total measurement time of 1240.3 h. With the exception of slight pollution with ^{40}K ((170 ± 80) mBq/kg), no other radioactive contaminants were found; the derived upper limits are: <20 mBq/kg for ^{238}U chain, <30 mBq/kg for ^{232}Th chain, <4 mBq/kg for ^{137}Cs and <8 mBq/kg for ^{60}Co.

1. Introduction

The radiopurity of a lithium molybdate crystal is investigated here mainly in relation to its possible use as a potential detector in the search for neutrinoless double beta (2β) decay of ^{100}Mo. Neutrinoless (0ν) 2β decay of atomic nuclei is forbidden in the standard model (SM) but predicted in many SM extensions [1]. It occurs if the neutrino is a Majorana particle ($\nu = \bar{\nu}$) with non-zero mass. Recent experimental observations of the oscillation of solar, atmospheric, reactor and accelerator neutrinos (see f.i. summary [2]) provide evidence that the neutrino is indeed a massive particle. This fact further increases the importance of the search for 2β0ν decay.

Experimental searches in various isotopes have been performed in the past 60 y, almost all with negative results; only Ref. [3] has positive evidence for the observation of the 2β0ν decay of ^{76}Ge with $T_{1/2} = 2.23 \times 10^{25}$ y been reported. New experiments are currently forthcoming on ^{76}Ge and other isotopes; their results are anticipated with great interest.

Among the 35 nuclei candidates for $2\beta^-$ decay [4], ^{100}Mo is one of the best choices because of its high transition energy of $Q_{2\beta} = 3034.40(17)$ keV [5] and its relatively high natural isotopic abundance of 9.824(50)% [6]. Molybdenum isotopes can also be enriched using relatively inexpensive ultra-speed centrifuge technology. The high $Q_{2\beta}$ value leads to a large phase space integral of the decay, i.e., to a higher probability of the 2β0ν decay. From an experimental point of view, $Q_{2\beta}$ values greater than the highest relevant energy of the natural gamma radioactivity (2615 keV γ line of ^{208}Tl) are also preferable. They ensure lower backgrounds, helping to distinguish the faint signal expected.

The best actual limit for the 2β0ν decay of ^{100}Mo has been given by the NEMO-3 experiment, where ≃ 7 kg of enriched ^{100}Mo was used. After 389 d of taking data, an upper limit of $T^{0\nu}_{1/2} > 4.6 \times 10^{23}$ y at 90% C.L. was reached [7]. At the same time, the allowed two neutrino (2ν) double beta decay of ^{100}Mo was measured with high statistics: with 219,000 observed events, the derived half-life is $T^{2\nu}_{1/2} = 7.1 \times 10^{18}$ y [7].

Notwithstanding these relevant results, two items could be improved in future experiments: the quite low efficiency of detection of the 2β0ν signal (near 14%) and the relatively poor energy resolution (near 0.3 MeV at the $Q_{2\beta}$ value). A configuration where the detector is also the source containing the ^{100}Mo nuclei could ensure almost 100% efficiency in detecting the 2β0ν signal. One kg of ^{100}Mo would then be equivalent to 7 kg of ^{100}Mo in the NEMO-3 experiment.

Recently, $CaMoO_4$ crystal scintillators were characterized as possible detectors for 2β decay investigations of ^{100}Mo [8]. One other promising material is lithium molybdate. It should be also noted that in such a detector, in addition to ^{100}Mo, ^{92}Mo and ^{98}Mo could also be studied for their possible 2β decays.

Intrinsic radioactive contamination is an extremely an important characteristic of detectors used in searches for rare nuclear decays because it is directly related to the achievable sensitivity of experiments. As a first step in characterizing the

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Li_2MoO_4 material, we present here the results of the investigation of the radioactive contamination of a Li_2MoO_4 crystal. Such results, to our knowledge, are reported for the first time.

2. Li_2MoO_4 crystals

The lithium molybdate (Li_2MoO_4) was obtained by a solid-state synthesis technique from MoO_3 and Li_2CO_3 powders (both of 99.5% purity) with subsequent recrystallization from water solutions. Transparent single crystals of up to $\varnothing 25 \times 35$ mm in size were grown using the Czochralski method with a drawing speed of 4 mm/h. The room temperature X-ray powder diffraction (XRD) pattern was indexed according to the lattice parameters of the trigonal system (space group $R\bar{3}$). The structure is isotypic with phenacite (Be_2SiO_4). The density calculated from the XRD pattern (3.07 g/cm^3) is slightly different from the hydrostatic density (3.02 g/cm^3). This could be due to the presence of some defects in the crystal lattice. The material is soluble in water and weakly hygroscopic; the melting point is $(701 \pm 2)^\circ$C [9]. The luminescence characteristics were investigated at the temperatures of 10 and 300 K in [10]. At 300 K, no luminescence was found. It was observed at 10 K instead, with a maximum of the emission spectrum at 580 nm.

3. Measurements and data treatment

The Li_2MoO_4 crystal under investigation had a mass of 33.95 g, and its dimensions were $\varnothing 22 \times 33$ mm. It was measured in an ultra-low background setup with a GePV detector, one of the HPGe detectors placed deep underground in the Laboratori Nazionali del Gran Sasso (LNGS) (3800 m w.e.) [11]. The active volume of the detector is 364 cm^3. The experimental setup is surrounded by a passive shield, which consists of lead (25 cm) and, in the innermost part, oxygen-free high conductivity copper (10 cm). The setup is also flushed continuously with almost radon-free nitrogen and is isolated from the surrounding atmosphere. The Li_2MoO_4 sample was measured for 1240.3 h; the background of the HPGe detector was measured for 732.2 h. The sample spectrum together with the background spectrum for comparison is presented in Fig. 1.

All peaks in the spectrum of the Li_2MoO_4 crystal belong to natural and man-made radioactivity, i.e., ^{40}K, ^{60}Co, ^{137}Cs and nuclides in chains of ^{232}Th and ^{238}U. Comparing the rates of the peaks in the Li_2MoO_4 spectrum with those in the background, they are equal within statistical uncertainties. Some additional activity was found only for ^{40}K: $A = (170 \pm 80)$ mBq/kg.

The specific activities of the nuclides were calculated using the formula $A = (S_s/t_s - S_b/t_b)/(y \cdot \eta \cdot m)$, where S_s (S_b) is the area of a peak in the sample (background) spectrum; t_s (t_b) is the time of the sample (background) measurement; y is the yield of the corresponding γ line [12]; η is the efficiency of the full peak detection and m is the mass of the sample. The full energy peak efficiencies were calculated with the GEANT4 package [13]. The upper limits were obtained applying the Feldman–Cousins procedure [14]. All results are summarized in Table 1.

4. Discussion and conclusions

As already stated in the introductory part, lithium molybdate contains three potentially 2β decaying Mo isotopes. Their natural abundances were precisely determined in recent measurements [6] as: 14.5246(15)% for ^{92}Mo, 24.391(18)% for ^{98}Mo, and 9.824(50)% for ^{100}Mo. Energy releases $Q_{2\beta}$ are given in [15] as

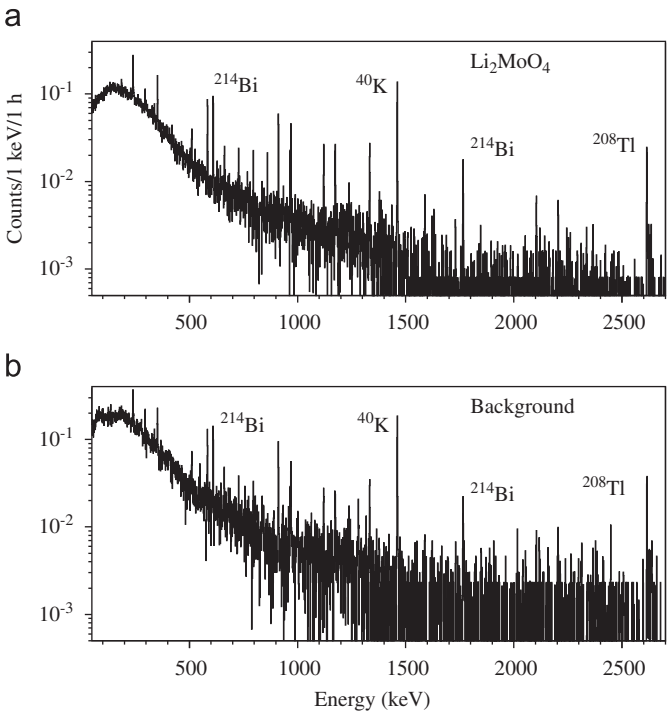

Fig. 1. Spectra measured underground in the LNGS with the low-background HPGe detector 364 cm^3: (a) Li_2MoO_4 sample during 1240.3 h; (b) background of the detector during 732.2 h.

Table 1
Radioactive contamination of the Li_2MoO_4 crystal measured with HPGe spectrometry.

Chain	Nuclide	Activity (mBq/kg)
^{232}Th	^{228}Ac	≤ 32
	^{212}Pb	≤ 24
	^{208}Tl	≤ 12
^{238}U	^{214}Pb	≤ 20
	^{214}Bi	≤ 21
	^{40}K	$= 170(80)$
	^{60}Co	≤ 8
	^{137}Cs	≤ 4

1649(4) keV for ^{92}Mo and 113(6) keV for ^{98}Mo; for ^{100}Mo it was recently accurately measured [5] as 3034.40(17) keV.

^{100}Mo is one of the most investigated nuclei among all 2β decay candidates. In geochemical measurements, its half-life was determined to be $(2.1 \pm 0.3) \times 10^{18}$ y [16]. Two neutrino 2β decay has been observed in several direct experiments (see the summary in, e.g., [4,17]), with the most precisely measured half-life obtained in the NEMO-3 studies $T^{2\nu}_{1/2} = (7.1 \pm 0.5) \times 10^{18}$ y [7]. The decay to the excited 0^+_1 level (1130 keV) of ^{100}Ru was also detected with a $T_{1/2}$ of around 6×10^{20} y (see [4,17–19] and references therein). The best up-to-date limit on the $2\beta0\nu$ mode to the ground state of ^{100}Ru is $T^{0\nu}_{1/2} > 4.6 \times 10^{23}$ y [7]; some other possibilities (neutrinoless decays with emission of Majorons and transitions to different excited levels of ^{100}Ru) were also sought [18,20].

At the same time, 2β processes in ^{92}Mo and ^{98}Mo were investigated to a much lesser extent. For the 2ε and εβ$^+$ decays of

^{92}Mo, the $T_{1/2}$ limits are at the level of $10^{18}-10^{20}$ y; for ^{98}Mo, a quite poor limit of 10^{14} y is reported [4,21].

In addition to the background due to cosmic rays, which can be reduced by choosing a deep underground site, and due to natural and man-made radionuclides (^{40}K, ^{60}Co, ^{137}Cs, and U/Th chains), cosmogenically induced activities produced in the Li_2MoO_4 crystals during their stay on the Earth's surface could be dangerous. These activities were calculated with the program COSMO [22], assuming a 1 month exposure period on the Earth's surface and a deactivation time of 1 y deep underground. It was found that among a total of 69 induced radionuclides with $T_{1/2}>25$ d in Li_2MoO_4, none could make a large contribution to the background at the energy of the ^{100}Mo neutrinoless 2β decay. This is related to the high $Q_{2\beta}$ value of ^{100}Mo. This also makes the requirements for the radiopurity of the materials close to the detector less stringent.

In conclusion, the concentrations of radionuclides in a Li_2MoO_4 single crystal have been measured by means of ultra-low background HPGe γ spectrometry at the LNGS. A small amount of ^{40}K was found (170(80) mBq/kg). It could be related with rather poor level of purity of the initial materials (99.5%) used for the crystal production. Only upper limits could be set for the radioactivity concentrations of the ^{238}U chain (<20 mBq/kg), the ^{232}Th chain (<30 mBq/kg), ^{137}Cs (<4 mBq/kg) and ^{60}Co (<8 mBq/kg). These low levels of contamination together with the observation of the luminescence scintillation of Li_2MoO_4 at 10 K [10] give hope for an experiment in which the lithium molybdate could be used as scintillating detector in the search for the 2β decays of Mo isotopes in a "source = detector" approach. A first test of a Li_2MoO_4 crystal at ≈ 10 mK has shown an encouraging result: both scintillation and phonon signals were observed [23]. Further investigations of Li_2MoO_4 are planned.

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