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$^{arch}PbMoO_4$ scintillating bolometer as detector to searches for the neutrinoless double beta decay of ^{100}Mo

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Abstract. The $^{arch}PbMoO_4$ scintillating crystal has been produced from archaeological lead for the first time. The advanced technique for deep purification of lead against chemical impurities was used resulting in 99.9995% purity level of final material. The $^{arch}PbMoO_4$ crystal was characterized by means of cryogenics bolometric measurements and demonstrates excellent performances as a scintillating bolometer. The energy resolution (0.3% at 1462 keV of ^{40}K), the high light yield (5.2 keV/MeV for γs, and 1.2 keV/MeV for α particles) and the highly efficient particle identification achieved with this detector, representing the high quality of the crystal. As a final proof for the feasibility of the $^{arch}PbMoO_4$ crystal as a promising detector to search for the neutrinoless double β-decay of ^{100}Mo, the crystal should be produced using the LTG Czochralski technique to prevent the possible contamination during the crystal growth and to increase the production yield.

1. Introduction

The claim of the Heidelberg-Moscow group in 2001 [1] on the neutrinoless double β-decay ($0\nu2\beta$) observation on ^{76}Ge, impelled numerous experimental efforts in order to probe this result using different nuclei as well as various experimental techniques [2–9]. ^{100}Mo is one of the most promising isotopes to search for the $0\nu2\beta$ decay due to its high energy transition ($Q_{\beta\beta}$ = 3035 keV [10,11]), the comparably high natural isotopic abundance (9.67% [12]), the possibility to be highly enriched by the ultra-speed gas-centrifuge technology (up to 99.5%) and the reasonable price for such type of enrichment (in the scale of $100/g). The high $Q_{\beta\beta}$ leads to a large phase space integral, which leads to a higher probability of the $0\nu2\beta$ decay. From an experimental point of view, the $Q_{\beta\beta}$ value higher than of 2615 keV line of ^{208}Tl results in a lower background induced by the natural radioactivity. The use of crystals embedding the isotope of interest operated as scintillating bolometers is one of possible way to realize a very sensitive experimental approach. This technique provides a high experimental sensitivity due to the combination of an excellent energy resolution with a high detection efficiency and the active particle discrimination.

In last decade a different crystals containing molybdenum were tested, demonstrating their feasibility as scintillating bolometers, for example Li_2MoO_4 [13], $CaMoO_4$ [14], $ZnMoO_4$ [15], $Li_2Zn_2(MoO_4)_3$ [16]. The $PbMoO_4$ crystals were also considered as a possible target-material for a next generation experiment aimed to detect the $0\nu2\beta$ decay of ^{100}Mo [17,18]. Whereas the high quality $PbMoO_4$ crystals can easily be grown by well-known technologies, its application in this field is so far limited by the presence of a high activity of ^{210}Pb in freshly produced lead, with values that range from few [19] to thousands Bq/kg [20]. ^{210}Pb is a decay product of ^{238}U decay chain, which is naturally present in all rocks and ores. During the melting of the lead-containing ore, ^{210}Pb concentrates in the lead metal, while all the other radioactive nuclides belonging to ^{238}U chain (^{234}U, ^{230}Th and ^{226}Ra) are segregated to the slag, since they are chemically very different with respect to Pb. This results in a high purity of lead with respect to natural radioactive nuclides, but ^{210}Pb isotope remains embedded. Eventually ^{210}Pb activity decreases with a half-life of 22.3 years, thus in a metal which was melted hundreds years ago its activity is negligible. In fact, the extremely low level of intrinsic radioactive contamination of archaeological lead, with a limit less than few mBq/kg of ^{210}Pb, has demonstrated by several works [21–23]. So, only archaeological lead can be a suitable material for production of the low-background $PbMoO_4$ crystals.

The main target of the present work is to investigate the performance of the $^{arch}PbMoO_4$ crystal produced using of archaeological lead, as a promising scintillating bolometer to search for the $0\nu2\beta$ decay of ^{100}Mo.

2. Crystal production

The general requirements on chemical purity of the initial materials to produce high-quality $PbMoO_4$ crystals impose limitations on transition metals concentration (Ti, V, Cr, Mn, Fe, Co, Ni) less than 1 ppm and on some other elements (Mg, K, Mn, Bi, Sn, Mg, Al, Cu) less than 5 ppm. Higher concentration of these impurities will significantly degrade the scintillating properties of the crystal (light output, energy resolution, and intensity of afterglow). With respect to bolometric characteristics this particular requirement is necessary to achieve the extremely low concentration of point defects (vacations and interstitial atoms) and others kind of defects like a color centers and its associates, dislocations and blocks. The specific type of defect in the crystal will determine the dominant mechanism of phonon scattering and consequently will change the heat capacity that will spoil the energy resolution in heat channel of the bolometer.

As a starting material was used a small piece of the lead ingot from the load carried by a Roman Empire age ship sunk near Sardinia [24]. A new advanced technology of vacuum distillation, which described in details in [25], was used for the deep purification of this lead metal. This purification technique is allowed to achieve purity grade of lead more than 99.9995% starting from 99% in the initial metal. The lead oxide (PbO) was produced by means of several sequential chemical transformations: the dissolution of purified lead in a weak nitric acid solution followed by the neutralization of the acid solution by ammonia, and the lead hydroxide $Pb(OH)_2$ precipitated at the end. This substance was washed several times by ultra-pure water and centrifuged. Final product was obtained by annealing of the lead hydroxide in a quartz crucible at 550°C for more than 24 hours.

The $^{arch}PbMoO_4$ single crystal was grown by the conventional Czochralski technique in a platinum crucible (∅40×40 mm) under argon atmosphere. In order to prevent the imbalance of the $PbMoO_4$ charge components due to MoO_3 evaporation during the crystal growth, a small excess of MoO_3 (1.0%) was added to stoichiometric mixture of PbO and MoO_3 powders before the charge synthesis. The total mass of loaded charge into crucible was 350 g, resulting in a single crystalline boule of 175 g (∅20×90 mm). It indicates that the production yield is about 50% from the charge. Finally, a 57.2 g cylindrical crystal with dimensions about ∅20×30 mm was cut from the boule and used as a detector.

3. Low temperature measurements

The $^{arch}PbMoO_4$ crystal was operated over 19 hours as a scintillating bolometer at mK-temperatures, in a $^3He/^4He$-dilution refrigerator at Laboratori Nazionali del Gran Sasso (Assergi, Italy). When an interacting particle deposits energy in the crystal volume, a large fraction of energy is converted to heat, while the rest produces scintillation light. The heat and light signals are simultaneously recorded as

temperature increasing by the Germanium Neutron Transmutation Doped thermistors (Ge-NTD) on the main crystal and on the light detector (LD), which is also operated as a bolometer. The main crystal and the LD were housed in a copper structure described in [26]. A reflecting foil (3M VM2002) was used to surround the detector setup, leading to an increase in the overall light collection efficiency. More details on the light detector, which consists of a high purity Ge wafer (∅44.5·0.3mm), can be found in [27]. According to the nature of the interaction, α or β/γ particles will generate different amount of light for the same energy released in the volume of main crystal. This peculiar feature allows to strongly reducing the background, therefore improving detector sensitivity.

The heat and light signals were converted into voltage pulses by Ge-NTD thermistors, were amplified and fed into a 18-bit ADC unit. The trigger of the $^{arch}PbMoO_4$ crystal was software generated while the LD is independently acquired in coincidence with the former. The entire waveform of each voltage pulse is sampled and acquired. The time window has a width of 512 ms sampled with 1024 points. The amplitude and the shape of the voltage pulse were then determined off-line making use of the Optimal Filter technique [28]. Detailed information on the electronic system and data acquisition can be found in [29–31].

The amplitude of the heat and light signals is energy-calibrated by means of known calibration sources. The light detector was calibrated using a ^{55}Fe X-ray source permanently facing the LD. An energy resolution (FWHM) of 590 eV was evaluated for the X-ray doublet of 5.9 and 6.5 keV. While the heat channel is energy-calibrated with α peaks of internal radioactive contamination by the daughter nuclides of U/Th natural decay chains, using a second order polynomial with zero intercept as a calibration curve.

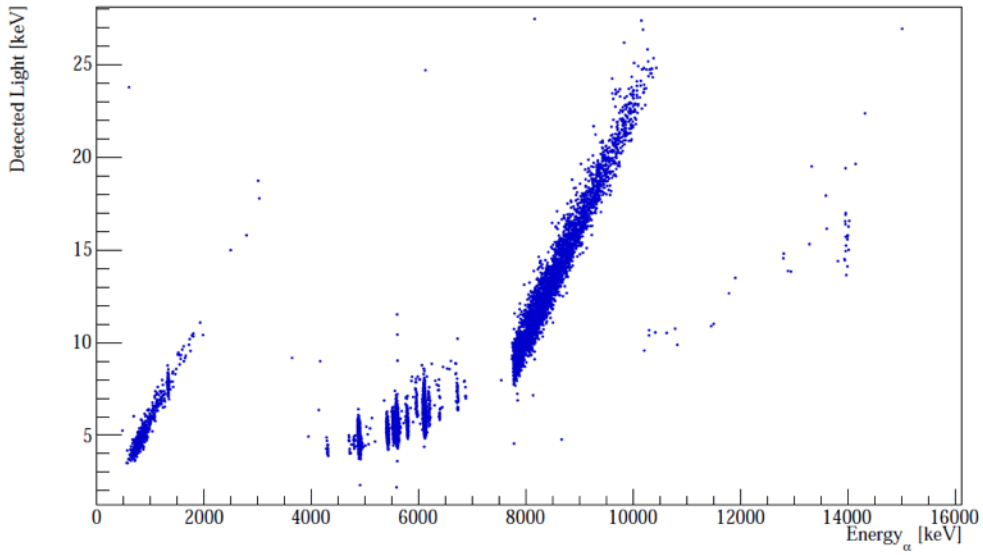

Figure 1. Light-vs-Heat scatter plot corresponding to 19 hours of background measurement with the 57.2 g $^{arch}PbMoO_4$ scintillating crystal, working as a scintillating bolometer. One can recognize several well separated events distributions caused by different type of particles (see description in text below).

The total acquired statistics is shown in Fig. 1, where the detected light is shown as a function of the energy detected by the $^{arch}PbMoO_4$ crystal. The nature of the interacting particles is clearly understood by the different Light Yield (LY), defined in this context as the ratio of the energy measured in the light detector (in keV) to the energy measured in the heat channel of the $^{arch}PbMoO_4$ crystal absorber (in MeV). From Fig. 1, one can see several easily distinguishable distributions induced by detection of different type of particles: a) band in energy range 0.5-3.0 MeV related to β/γ induced interactions (single β/γ events); b) structure between 4 MeV to 7 MeV represent α peaks of internal conta-

mination from U/Th natural decay chains (single α events); c) highly populated band from 8 to 11 MeV is caused by ^{212}Bi-^{212}Po coincidence events (sum of β and α events); d) pure populated distribution from 10 to 15 MeV induced by coincidence of two single α events due to internal contamination from U/Th natural decay chains. The pure β/γ events are characterized by LY of about 5.2 keV/MeV, while pure α's have lower LY on the level of 1.2 keV/MeV.

This $^{arch}PbMoO_4$ crystal demonstrates the outstanding bolometric characteristics. Offering perfect discrimination between α and β/γ events due to big difference in LY, the detector also has an extremely good energy resolution in heat channel both, for α particles (FWHM = 8.4 keV or 0.2% at E_α = 4870 keV of ^{226}Ra) and for γ's (FWHM = 5.1 keV or 0.3% at E_γ = 1462 keV of ^{40}K). Such excellent energy resolution clearly indicates the high perfection of the crystalline structure and the low defects concentration.

4. Internal α contamination

The "pure" α events were selected from entire statistics by applying cut on the amount of emitted light (i.e. LY < 2.0 keV/MeV) to reject β/γ events, and on deposited energy (i.e. < 7.0 MeV) to select only single α events, with the overall efficiency close to 100%. The spectrum of α events obtained applying these cuts is presented in Fig. 2.

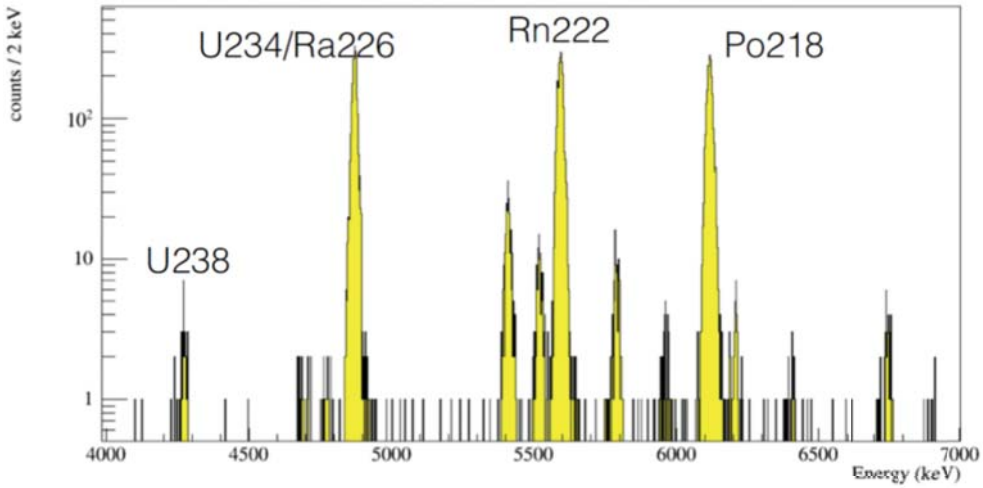

Figure 2. The selected "pure" α events from entire statistics collected with the $^{arch}PbMoO_4$ crystal 57.21 g of mass, during 19 hours of background measurements. The events are attributed to internal contaminations of the crystal by daughter α active nuclides of U/Th decay chains.

As one can see from Fig. 2 and values listed in Tab. 1, the $^{arch}PbMoO_4$ crystal is extremely highly contaminated by the radioactive nuclides from ^{226}Ra sub-chain of ^{238}U chain, as well as with daughter nuclides from ^{235}U and ^{232}Th chains. Such level of internal contamination is absolutely unexpected, considering all efforts dedicated to the production of the high chemical and radioactive purity PbO and MoO_3 powders. We can only suppose that contamination incident is occurred during the crystal growth.

As was mentioned above (see Sec. 2), the $^{arch}PbMoO_4$ crystal was grown by the conventional Czochralski technique, where an opened Pt-crucible with loaded charge was covered by several layers of the aluminum oxide ceramics to provide a thermal insulation. In our opinion, the huge radioactive contamination of the crystal can be the result of the contact between the melted charge and the small piece of the aluminum oxide ceramics. This assumption matches with our experience gained in the framework of the LUCIFER/CUPID-0 project [32,33], devoted to the development of the low-background $Zn^{82}Se$ crystals starting from the high purity Zn and the enriched ^{82}Se isotope. A huge activity

of nuclides from ^{226}Ra sub-chain (about 0.2 kBg/kg) and ^{40}K (1 kBq/kg) was induced by a 0.3 g piece of aluminum oxide ceramics incorporated in a certain batch of the ZnSe powder. Assuming the same radiopurity of aluminum oxide ceramics in both cases (i.e. Zn^{82}Se powder production and ^{arch}PbMoO$_4$ crystal growth), the internal contamination of the ^{arch}PbMoO$_4$ crystal is compatible with activity caused by a piece of ceramics with mass of about 30 mg, fallen into the crucible during the crystal growth.

Table 1. Internal contamination of the 57.21 g ^{arch}PbMoO$_4$ crystal.

Chain	Nuclide	Activity [mBq/kg]
^{232}Th	^{232}Th	10±7
	^{228}Th	99±9
	^{224}Ra	86±8
	^{220}Rn	21±5
	^{212}Bi	49±7
^{238}U	^{238}U	38±6
	^{234}U	98±9
	^{230}Th	13±3
	^{226}Ra	2310±42
	^{222}Rn	2370±42
	^{218}Po	2340±42
	^{210}Po	266±16
^{235}U	^{235}U	14±3
	^{227}Th	17±4
	^{223}Ra	30±5
	^{219}Rn	5±2
	^{211}Bi	30±5

5. Discussion

Finally we would like to stress several aspects of the presented results. First of all, the technological issue of the radiopure crystal production. A possible solution to produce a low-background high-quality crystals is to use the Low Thermal Gradient (LTG) Czochralski technique [34,35], which operates with the closed Pt-crucible, that completely eliminates risks of contamination incident. Another very important advantage of this technique is the small temperature gradient at the level of 1 K/cm, which is one-two order of magnitude lower in comparison to the conventional Czochralski method. It leads to an absence of intensive evaporation of charge component from the melt and the losses did not exceed the level of 0.5% of the initial charge. Moreover the LTG Czochralski technique allows to crystallize up to 90% of the loaded charge (already achieved with BGO [36], $CdWO_4$ [37,38] and $ZnMoO_4$ [15] crystals), which is crucial in case of costly material, like enriched isotopes or highly purified archaeological lead. It should be also stressed that, despite the low molar fraction of Mo in the $PbMoO_4$ compound (26%) the number of Mo nuclei in 1 cm^3 of crystal is the same ($0.11 \cdot 10^{23}$) as for

the Li_2MoO_4 crystal, that has highest molar fraction of Mo (55%). This is the result of higher density of the $^{arch}PbMoO_4$ crystal (6.83 g/cm^3), in comparison to the Li_2MoO_4 crystal (3.03 g/cm^3). Moreover, the highest *Z* and density of the $^{arch}PbMoO_4$ crystal will increase the detection efficiency of the high energetic electrons produced in the 0ν2β decay of ^{100}Mo, taking in consideration the detector with the same dimensions. Finally, we do not see any drawbacks in the application of the $^{arch}PbMoO_4$ crystal produced from the archaeological lead as detector to search for the 0ν2β decay of ^{100}Mo, especially taking into consideration its excellent bolometric performances.

As a next step we are going to grow the $^{arch}PbMoO_4$ crystal from the highly purified archaeological lead and molybdenum oxide, using the LTG Czochralski technique to prevent possible contamination incident during crystal growth and to increase significantly the production yield. It will allow us to produce a high-quality large-volume $^{arch}PbMoO_4$ crystal with high performance as a low-background scintillating bolometer.

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