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Development of enriched $^{106}\text{CdWO}_4$ crystal scintillators to search for double β decay processes in ^{106}Cd

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ABSTRACT

A cadmium tungstate crystal scintillator enriched in ¹⁰⁶Cd was developed for an experiment to search for double beta processes in ¹⁰⁶Cd. With this aim samples of cadmium with natural isotopic composition and enriched in ¹⁰⁶Cd were purified by vacuum distillation. Cadmium tungstate compounds were synthesized from solutions. The absolute isotopic composition of the enriched cadmium was accurately determined as 66.4% by thermal ionisation mass-spectrometry. A ¹⁰⁶CdWO₄ crystal boule was grown by the low-thermal-gradient Czochralski technique. The total irrecoverable loss of the enriched cadmium in all the stages of the crystal scintillator production does not exceed 2.3%. The produced ¹⁰⁶CdWO₄ crystal scintillator with mass of 216g exhibits good optical and scintillation properties.

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1. Introduction

Observations of neutrino oscillations indicate non-zero neutrino mass and provide important motivation for high sensitivity experiments to search for neutrinoless double beta $(0v2\beta)$ decay. Generally only half-life limits for $0v2\beta$ mode were obtained so far (see, e.g., reviews [1–5]). However, it is worth noting that in Ref. [6] a positive evidence of $0v2\beta$ decay of ⁷⁶Ge has been reported

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with $T_{1/2}$ at level of about 2×10^{25} yr; for some years, new experiments are in preparation both on and on other isotopes.

One of the most sensitive 2β experiments has been performed in the Solotvina Underground Laboratory with the help of cadmium tungstate crystal scintillators enriched in ¹¹⁶Cd [7,8]. The two neutrino mode of double β decay of ¹¹⁶Cd was observed with the half-life $T_{1/2} = (2.9^{+0.4}_{-0.3} \times 10^{19})$ yr, and the half-life limit on 0v2 β decay of ¹¹⁶Cd was set as $T_{1/2} \ge 1.7 \times 10^{23}$ yr at 90% C.L. It corresponds to an upper bound on the effective Majorana neutrino mass $\langle m_v \rangle \le 1.7$ eV [8]; this result is among the strongest world-wide limits on $\langle m_v \rangle$. The Solotvina experiment demonstrates that CdWO₄ crystals possess several important properties required for high sensitivity 2 β decay experiments,

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i.e. low level of intrinsic radioactivity, good scintillation characteristics, pulse-shape discrimination ability, low cost, and stability of operation.

Another application of CdWO₄ crystal scintillators is the search for double β processes in ¹⁰⁶Cd. The value of $Q_{2\beta}$ =(2770 ± 7) keV for ¹⁰⁶Cd [9] is one of the highest for double β^+ active nuclides and allows three modes of decay: 2 ϵ (double electron capture), $\epsilon\beta^+$ (electron capture with emission of positron) and $2\beta^+$ (decay with emission of two positrons). The isotopic abundance of ¹⁰⁶Cd in natural cadmium is (1.25 ± 0.06)% [2,10], which is relatively high in comparison with other potentially $2\beta^+$ active isotopes.

 ^{106}Cd is among the most widely studied $2\beta^+$ nuclides with a rather long history of investigations. Performed studies can be divided into two groups: (1) experiments using samples of Cd with external detectors for registration of the emitted particles, and (2) experiments with detectors containing Cd nuclei (the so called "source=detector" approach). The first method allows to collect more detailed information on 2ß processes in the most advanced experiments, tracks of emitted β particles, their individual energies and angle between the tracks can be measured. The second method measures only an energy release in a detector (very often even not distinguishing between β or γ or α particles as possible sources of this energy release), but it has generally higher efficiency for detection of 2β decays (up to 100%), and its use is preferable for the first reaching of the $T_{1/2}$ scale at which the 2β process can be observed. Summary of the previous searches for $2\hat{\beta}^+$ processes in ¹⁰⁶Cd is given in Table 1.

An advantage of experiments using detectors containing cadmium is the higher detection efficiency of the ¹⁰⁶Cd double β decay effects, especially in the case of the double electron capture. Therefore, the development of CdWO₄ crystal scintillators enriched in ¹⁰⁶Cd could be an important step to go towards the level of theoretical predictions for two neutrino mode of 2 ϵ and $\epsilon\beta^+$ processes: $T_{1/2} \sim 10^{21} - 10^{22}$ yr [16,21–24].

The production of cadmium tungstate crystal scintillators enriched in ¹⁰⁶Cd involves a number of steps: the purification of the isotopically enriched cadmium, the synthesis of the ¹⁰⁶CdWO₄ compound, the growth of the crystals, the production of as much larger scintillation element to have a maximal number of ¹⁰⁶Cd nuclei in a low background 2β experiment. To obtain crystals with good scintillation characteristics, it is necessary to control the purity of the initial materials and of the compound for the crystal growth at a level of 0.1–1 ppm for a range of elements. A careful control of the enriched material is necessary on all the stages of the cadmium tungstate crystals production taking into account the high cost of the enriched cadmium 106 isotope.

In this paper we describe the main stages of the 106 CdWO₄ crystal development: the purification of cadmium metal samples, the production of cadmium tungstate powder, the crystal growing, the control of all initial materials purity, production of cadmium tungstate crystal, and the measurements of the optical and scintillation characteristics of the produced 106 CdWO₄ scintillator. Some preliminary results of these investigations were reported in [25,26,].

2. The production of cadmium tungstate crystal scintillators

2.1. The purification of cadmium

The purification of the cadmium metal samples was performed by vacuum distillation [27]. The vacuum in a graphite apparatus during the distillation process was kept at the level of $\approx 10^{-5}$ torr. A sample of natural cadmium of weight ≈ 0.5 kg was used to develop the technology of the purification. The cadmium for the R&D was taken of a rather low purity grade, ≈ 99.8 %, because some

Table 1

Previous experiments on searches for $2\beta^*$ processes in 106 Cd.

Short description	$T_{1/2}$ limit, yr	Year [Ref.]
Thin ^{nat} Cd samples between	$\sim\!10^{15}~(0\nu2\beta^{*})$	1952
photographic emulsions, 1344 h exposure underground (567 m depth) ^a	$\sim\!10^{15}(0\nu\epsilon\beta^*)$	[11]
Wilson cloud chamber at the Earth level, 30 g of ^{nat} Cd foil (110 mg/ cm ²), 12 352 acceptable photographs, 1 2 h total exposure	$6\times 10^{16}~(0\nu 2\beta^+)$	1955 [12]
Two Nal scintillators in coincidence, Earth level, 1.5 kg ^{nat} Cd sample, 72 h, search for coincidence annihilation 511 keV γ 's ($2\beta^+$, $\epsilon\beta^+$) or for coincidence X-rays (2ϵ)	$\begin{array}{l} (2.2-2.6)\times 10^{17} \\ (2\beta^{*}) \\ (4.9-5.7)\times 10^{17}(\epsilon\beta^{*}) \\ 1.5\times 10^{17}(2\nu 2\epsilon) \end{array}$	1984 [13]
¹¹⁶ CdWO ₄ crystal scintillator (122 g, enriched in ¹¹⁶ Cd to 83%), Solotvina underground laboratory (1000 m w.e.), 2982 h	$\begin{array}{l} (0.5 - 1.4) \times 10^{18} \\ (0v2\beta^{+}) \\ (0.3 - 1.1) \times 10^{19} \\ (0v\epsilon\beta^{+}) \\ 5.8 \times 10^{17} \ (2v2\epsilon) \end{array}$	1995 [14]
^{nat} CdWO ₄ crystal scintillator 1.046 kg, Gran Sasso underground laboratory (3600 m w.e.), 6701 h	$\begin{array}{l} 2.2 \times 10^{19} (0 \vee 2\beta^{+}) \\ 9.2 \times 10^{17} (2 \vee 2\beta^{+}) \\ 5.5 \times 10^{19} (0 \vee \beta^{+}) \\ 2.6 \times 10^{19} (2 \vee \beta^{+}) \end{array}$	1996 [15]
331 g ^{nat} Cd sample, HP Ge detector 120 cm ³ , Modane underground laboratory (4800 m w.e.), 1137 h, search for deexcitation and annihilation γ 's	$\begin{array}{c} 2.5 \times 10^{19} \ (216 \beta^{+}) \\ 1.0 \times 10^{19} \ (2\beta^{+}) \\ (6.6 - 8.1) \times 10^{18} \ (\epsilon\beta^{+}) \\ (3.5 - 6.2) \times 10^{18} \ (2\epsilon) \end{array}$	1996 [16]
0.5 g ^{nat} CdTe bolometer (10 mK),	$1.4\times 10^{16}~(0\nu\epsilon\beta^{*})$	1997
Two Nal scintillators in coincidence, ¹⁰⁶ Cd sample (enriched in ¹⁰⁶ Cd to 68%) 154g, Gran Sasso underground laboratory (3600 m w.e.), 4321 h	$\begin{array}{l} (1.6\!-\!2.4)\!\times\!10^{20} \\ (2\beta^*) \\ (1.1\!-\!4.1)\!\times\!10^{20}(\epsilon\beta^*) \\ (3.0\!-\!7.3)\!\times\!10^{19}(2\epsilon) \end{array}$	[17] 1999 [18]
Three ¹¹⁶ CdWO ₄ crystal scintillators (330 g, enriched in ¹¹⁶ Cd to 83%) and ^{nat} CdWO ₄ 454 g, Solotvina underground laboratory (1000 m w.e.), 14 183 b	$\begin{array}{l} (0.5-1.4)\times 10^{19} \\ (2\beta^{*}) \\ (0.1-7.0)\times 10^{19} \left(\epsilon\beta^{*}\right) \\ (0.6-8.0)\times 10^{18} \left(2\epsilon\right) \end{array}$	2003 [8]
COBRA experiments, sixteen 1 cm ³ ^{nat} CdZnTe semiconductor detectors (total mass 104 g), Gran Sasso underground laboratory (3600 m w e) 5546 h	$\begin{array}{l} (0.9-2.7)\times 10^{18} \\ (2\beta^{*}) \\ (4.6-4.7)\times 10^{18}(\epsilon\beta^{*}) \\ 1.6\times 10^{17}(2\epsilon) \end{array}$	2009 [19]
TGV experiment, 32 planar HP Ge detectors (\sim 17 cm ³ each), 16 thin ¹⁰⁶ Cd samples (75% enrichment, 13.6 g total mass), 4800 b	$\begin{array}{l} (1.0-1.2)\times 10^{20} \\ (2\beta^{*}) \\ (0.8-1.1)\times 10^{20} \left(\epsilon\beta^{*}\right) \\ (0.4-1.7)\times 10^{20} \left(2\epsilon\right) \end{array}$	2009 [20]

Range of $T_{1/2}$ limits corresponds to values given for the transitions to the ground state or to excited levels of ¹⁰⁶Pd. More detailed information on limits for specific levels and 2 β modes can be found in original papers (see also [1]). COBRA and TGV experiments are still running.

 a To our knowledge, this was the first underground experiment in history of investigations of 2β decay.

part of the enriched ¹⁰⁶Cd had a similar level of contamination. A sample of pure cadmium with mass of 463 g was obtained.

The samples of cadmium enriched in 106 Cd to 66.4% (see Section 3.1) were purified in the same way. At this stage the irreplaceable losses¹ were found to be at the level of 1.8%; while

¹ The irreparable losses of enriched cadmium were mainly due to leak of cadmium vapors from the graphite apparatus, and their subsequent condensation on the walls of the chamber. The cadmium oxide condensate was practically impossible to collect. Some part of the material was also lost due to etching before the distillation procedure.

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Table 2											
The contamination of o	cadmium wi	ith natural	isotope	abundance	and	enriched in	1 ¹⁰⁶ Cd	before an	d after the	purificatio	on.

Element (natural composition)	Concentration in natura	l Cd (ppm)		Concentration in ¹⁰⁶ Cd (ppm)			
	Before purification	ion After purification		Before purification After purification		tion	
	ICP-MS	ICP-MS	L-MS	ICP-MS	ICP-MS	L-MS	
Ni	30	0.3	< 0.2	0.6	0.6	< 0.2	
Cu	47	0.3	< 0.2	5	0.7	0.5	
Fe	0.4 ^a	0.17 ^a	< 0.5	1.3 ^a	0.4 ^a	< 0.4	
Mg	30	< 0.5	< 0.05	12	< 0.5	< 0.05	
Mn	0.2	0.1	< 0.3	0.1	0.1	< 5	
Cr	0.2	0.1	< 1	9	< 0.5	< 0.1	
V	< 0.005	< 0.005	< 0.08	< 0.005	< 0.01	< 0.08	
Со	0.3	< 0.003	< 1	0.02	< 0.01	< 0.1	
K	8	< 5	0.7	11	< 10	0.04	
Pb	1000	3	< 1	270	8	< 0.3	
Th	< 0.001	< 0.001		< 0.001	< 0.001		
U	< 0.001	< 0.001		< 0.001	< 0.001		

^a Concentration of iron was measured by AAS.

the polluted residue of the cadmium (where impurities have concentrated) was at the level of 5.6%.

The contamination of the cadmium samples was measured by inductively coupled plasma mass-spectrometry (ICP-MS) and laser mass-spectrometry (L-MS). The concentration of iron was checked with the help of atomic absorption spectroscopy (AAS). The results of the measurements are presented in Table 2. One can see only the lead ICP-MS results for the purified samples do not agree with the L-MS data. Generally a semi-quantitative method used in the ICP-MS measurements allows to reach a reasonable accuracy on the level of \approx 30%, while an estimated accuracy of the L-MS data is on the level of \approx 50%. The significant disagreement of lead concentration can be explained by instrumental memory effect due to previous lead matrix samples analyzed on the apparatus.

Therefore, the cadmium samples were purified two orders of magnitude for Ni, Cu, Mg, Co, Pb. The purity of cadmium after purification has been found to be satisfactory in order to produce high quality CdWO₄ crystal scintillators [28–30].

2.2. The synthesis of the CdWO₄ and 106 CdWO₄ compounds

The raw materials to grow the CdWO₄ crystal scintillators were produced by the NeoChem company (Moscow, Russia). All the operations were realized by using quartz or polypropylene labware, materials with low level of radioactive contaminations. Reagents of high purity grade (concentration of any metal less than 0.01 ppm) were used. Water, acids and ammonia were additionally distilled by laminar evaporation in quartz installation.

The high cost of enriched ¹⁰⁶Cd limits the choice of the methods for its additional purification. Recrystallization methods, typically used for cadmium salts purification, cannot be applied due to the low outcome of the final product ($\approx 85\%$). Therefore, after dissolving the metallic cadmium in nitric acid, purification was realized by coprecipitation on a collector. At the same time additional recrystallization was performed to purify ammonium para-tungstate used as tungsten source. Solutions of cadmium nitrate and ammonium para-tungstate were mixed and then heated to precipitate cadmium tungstate

$Cd(NO_3)_2 + (NH_4)_2WO_4 = CdWO_4 + 2NH_4NO_3.$

Then the $CdWO_4$ sediment was rinsed and filtered. Finally the compound was dried and annealed.

The same procedure was used to produce cadmium tungstate compound from enriched ¹⁰⁶Cd. 336.3 g of ¹⁰⁶CdWO₄ compound

Table 3							
The contamination	of the	e CdWO ₄	and	¹⁰⁶ CdWO ₄	compounds	determined	by
ICD MS							

Element (natural	l Concentration (ppm)					
composition)	Requested by crystals producer	Measured in CdWO ₄ powder	Measured in ¹⁰⁶ CdWO ₄ powder			
Ni	≤ 0.5	≤ 0.05	≤ 0.05			
Cu	≤ 0.5	≤ 0.05	≤ 0.05			
Fe	≤ 2	≤ 0.4	≤ 0.8			
Mg	≤ 2	≤ 0.1	≤ 0.1			
Mn	≤ 2	≤ 0.05	≤ 0.05			
Cr	≤ 2	≤ 0.01	≤ 0.01			
V	≤ 2	≤ 0.01	≤ 0.01			
Со	≤ 2	≤ 0.05	0.09			
K		≤ 5	≤ 5			
Pb		≤ 0.3	0.04			
Th		≤ 0.1	≤ 0.05			
U		\leq 0.01	\leq 0.007			

The contamination by Fe was measured by AAS.

were obtained from 104.92 g of enriched ¹⁰⁶Cd in form of metal. Taking into account the isotopic composition of the enriched material, the produced ¹⁰⁶CdWO₄ powder contains 102.01 g of cadmium (therefore the produced compound contains 97.23% of the initial enriched cadmium). The returned contaminated residue (including also samples used for different analyses) in total contains 2.67 g of enriched cadmium (2.54%), while the irreplaceable losses of enriched material were only 0.24 g (0.23%).

The contamination of the cadmium tungstate compound produced from natural and enriched cadmium was measured by ICP-MS, while the concentration of iron was analyzed by AAS. The results of the measurements are presented in Table 3. The level of purity of the compounds satisfies the requirements to raw materials for cadmium tungstate crystals growth [28–30].

2.3. The growth of 106 CdWO₄ crystal

The growth of the CdWO₄ crystals from the specially purified cadmium with natural isotopic abundance by Czochralski process was described in [25,26]. The cadmium tungstate crystal from the enriched ¹⁰⁶Cd was grown by the low-thermal-gradient Czochralski technique [30–32] in platinum crucible $\emptyset 40 \times 100$ mm². The stoichiometry of the initial material was checked by gravimetric method. This procedure includes high temperature decomposition of CdWO₄ powder in hydrogen atmosphere with P. Belli et al. / Nuclear Instruments and Methods in Physics Research A 615 (2010) 301-306



Fig. 1. Boule of ¹⁰⁶CdWO₄ single crystal grown by the low-thermal-gradient Czochralski process (left; the scale is in centimeters and inches). Scintillation element ¹⁰⁶CdWO₄ with mass of 215.8 g ($\approx \emptyset 27 \times 50 \text{ mm}^2$) produced from the boule (right; the scale is in centimeters). The side surface of the scintillation element was made diffuse with the help of fine-grain polishing paper to improve the uniformity of the scintillation light collection.

following extractive distillation of cadmium. The Cd/W ratio in the initial ¹⁰⁶CdWO₄ compound was determined to be (1.003 \pm 0.002). Then, the Cd/W ratio in the charge was adjusted to an optimal value for CdWO₄ crystal growth by adding a small amount of cadmium oxide (¹⁰⁶CdO). The mass of the initial ¹⁰⁶CdWO₄ charge was 265 g.

A boule of 231 g was grown (see Fig. 1, left), that is 87.2% of the mass of the initial charge. The growth direction was parallel to the crystal axis [010]. The length of the boule was 60 mm, diameter of the cylindrical part was 27 mm. The crystal has practically neither coloration nor defects, which imperfections are typical for CdWO₄ crystals. The irreplaceable losses of cadmium (mainly due to evaporation from melt) were only 0.78 g (0.3%). Therefore, the total loss of enriched cadmium on the stages of purification (1.8%), powder production (0.2%) and crystal growth (0.3%) was at the level of 2.3%.

A scintillation element with mass of 215.8 g ($\approx \emptyset 27 \times 50 \text{ mm}^2$) was produced from the grown ¹⁰⁶CdWO₄ crystal boule by cleavage (with the help of a sharp knife) of the top and bottom conic parts on cleavage plane. The loss of material was practically negligible (< 0.01%) at this stage. To maximize the number of the ¹⁰⁶Cd nuclei available for a double β experiment, no other treatment of the crystal was done (Fig. 1, right). Only the side surface of the crystal was made diffuse, using fine-grain polishing paper, to improve uniformity of the scintillation light collection. As a result, the loss of enriched material at this stage was very low $\approx 0.03\%$.

3. The characterization of ¹⁰⁶CdWO₄ crystal

3.1. The absolute isotopic composition of the enriched ¹⁰⁶Cd

The isotopic composition of the two metal samples of enriched ¹⁰⁶Cd was measured in a VG 354 thermal ionization mass spectrometer in the Isotope Science Laboratory at Curtin University (Perth, Australia). The absolute isotopic compositions of these samples were then determined by calibrating the mass spectrometer with the international reference material for cadmium—BAM I012 (Bundesanstalt für Materialforschung und -prüfung), in order to calculate the isotope fractionation factor for cadmium. In this experiment no significant isotope correction factor was required, so that the absolute isotopic composition of the ¹⁰⁶Cd samples is presented in Table 4.

The absolute isotope abundance for ^{106}Cd is (66.40 \pm 0.05)%, whilst β active ^{113}Cd has an absolute isotope abundance of (3.935 \pm 0.003)%. The atomic weight of the ^{106}Cd samples is

 Table 4

 The absolute isotopic composition of the ¹⁰⁶Cd samples (%).

$106 \qquad \qquad 66.40 \pm 0.05 \qquad \qquad 1.25 \pm 0.06$
$108 \qquad \qquad 0.658 \pm 0.004 \qquad \qquad 0.89 \pm 0.03$
110 5.06 ± 0.01 12.49 ± 0.18
111 4.83 ± 0.01 12.80 ± 0.12
112 8.85 ± 0.03 24.13 ± 0.21
113 3.935 ± 0.003 12.22 ± 0.12
114 8.77 ± 0.03 28.73 ± 0.42
116 1.497 ± 0.008 7.49 ± 0.18

The isotopic composition of natural cadmium is given for comparison.

 (108.03 ± 0.02) as compared to the BAM-I012 value of $(112.411\pm0.002).$

As was demonstrated in [33], the production of CdWO₄ crystals does not change the isotopic abundance of cadmium. Therefore, we can assume that the isotopic composition of cadmium in the grown ¹⁰⁶CdWO₄ crystal scintillators is the same as in the measured metal samples presented in Table 4.

3.2. The luminescence under ultraviolet and X-ray excitation

The photoluminescence (PL) spectrum of a sample of the ¹⁰⁶CdWO₄ crystal was measured with a single monochromator MDR-23 at the room temperature. Photoluminescence was excited by a nitrogen laser (337.1 nm wavelength). The emission spectrum of the crystal is presented in Fig. 2. A wide (\approx 240 nm) PL band in the visible region with a maximum at 492 nm was observed in agreement with the results obtained by other authors for CdWO₄ crystals [34–36].

The luminescence under X-ray excitation was studied with BHV7 tube with rhenium anode (20 kV, 20 mA) at the room temperature. The light from the sample $\emptyset 10 \times 3 \text{ mm}^2$ was detected in the visible region by photomultiplier FEU-106 (sensitive in the wide wavelength region of 300–800 nm). The spectral measurements were carried out by using high-aperture monochromator MDP-2. The X-ray emission spectrum (see Fig. 2) shows a maximum intensity at the same wavelength 493 nm in agreement with the results obtained with CdWO₄ crystals [37–40].

3.3. The transmittance of ¹⁰⁶CdWO₄ crystal

The transmittance of the $^{106}CdWO_4$ scintillation element $\varnothing\,27\times50\,mm^2$ was measured in the spectral range 300–700 nm

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Fig. 2. Emission spectra of 106 CdWO₄ crystal under ultraviolet (PL) and X-ray (RL) excitation. Spectra were corrected for instrument response.



Fig. 3. The optical transmission curve of $^{106}\text{CdWO}_4$ crystal 50 mm long measured with 1.6 mm sample in reference beam.

using a SPECORD UV-vis spectrophotometer along the long (50 mm) axis of the crystal. A thin (1.6 mm) sample of a CdWO₄ crystal was placed in the reference beam of the instrument to correct the reflection losses. The results of the optical transmission measurements shown in Fig. 3 demonstrate that the sample exhibits good transmission properties in the relevant wavelength range 430–700 nm of the CdWO₄ emission spectrum.

From the data we have derived the attenuation length of the material (60 ± 7) cm at the wavelength of the emission maximum (490 nm). This is a significant improvement in comparison with earlier CdWO₄ samples exhibiting noticeable absorption in short-wavelength part of the visible light caused by yellowish coloration, defects and inclusions [40–43]. A likely explanation of such high optical quality of the material is the deep purification at the stage of the enriched cadmium distillation and of the ¹⁰⁶CdWO₄ compound synthesis, and also the advantages of the low-thermal-gradient Czochralski technique to grow CdWO₄ single crystals.

3.4. The scintillation properties of the $^{106}CdWO_4$ crystal scintillator

To measure the scintillation properties, the scintillation element was optically coupled with the help of Dow Corning



Fig. 4. The energy spectra of ¹³⁷Cs, ²⁰⁷Bi and ⁶⁰Co γ quanta measured by the ¹⁰⁶CdWO₄ scintillation crystal \emptyset 27 × 50 mm².

Q2-3067 optical couplant to 3" photomultiplier (PMT) Philips XP2412, and surrounded by a reflecting cup \emptyset 50 mm × 50 mm, made of dielectric mirror foil from 3 M [44], having specular reflectivity 98% in the visible region. The measurements were carried out with 10 µs shaping time of ORTEC 575 spectroscopy amplifier to collect most of the charge from the anode of the PMT. The crystal was irradiated by γ quanta of ⁶⁰Co, ¹³⁷Cs, and ²⁰⁷Bi sources. Fig. 4 shows the pulse amplitude spectra measured with the CdWO₄ crystal scintillator.

An energy resolution 10.0% (FWHM) was obtained with the detector for the 662 keV γ line of ¹³⁷Cs; this energy resolution is quite high considering the elongated and slightly irregular shape of the crystal, that leads to a substantial non-uniformity of the light collection. It should be mentioned that even better energy resolution: 8.1% for the 662 keV γ line of ¹³⁷Cs, was obtained with a $\emptyset 22 \times 29 \text{ mm}^2$ CdWO₄ scintillator produced from purified cadmium with natural isotopic composition [25,26] and regular cylindrical shape.

4. Conclusions

A cadmium tungstate crystal scintillator was produced from cadmium enriched in ¹⁰⁶Cd for an experiment to search for double beta processes in ¹⁰⁶Cd. Samples of cadmium (with natural isotopic composition and enriched in ¹⁰⁶Cd) were purified to ppm level by vacuum distillation. The losses of enriched cadmium were found to be 1.8%; the part of polluted rest is 5.6%. The absolute isotopic composition of the enriched ¹⁰⁶Cd sample was measured as (66.40 ± 0.05)% using a thermal ionization mass spectrometer. The cadmium tungstate powders (natural and enriched) were synthesized from additionally purified solutions of cadmium nitrates and ammonium para-tungstate. The yield of the reaction (in case of ¹⁰⁶CdWO₄) was 97.23% with losses of cadmium only 0.23%. The level of purity of the compounds,

measured by ICP-MS and by AAS, satisfies the requirements to raw materials for growing cadmium tungstate crystals.

A boule of ¹⁰⁶CdWO₄ crystal scintillator with mass of 231 g (87% of the initial raw material) was grown by the low-thermalgradient Czochralski method. The crystal has practically neither coloration nor defects, which are typical for CdWO₄ crystals. The loss of enriched cadmium (mainly due to evaporation from melt) was at the level of 0.3%. The total irrecoverable loss of the enriched cadmium on the stages of purification, raw material production, crystal growth, and scintillation element production does not exceed 2.3%. To our knowledge only four crystal scintillators have been grown using enriched isotopes. The pioneering work of der Mateosian and Goldhaber to search for neutrinoless double β decay of ⁴⁸Ca by using enriched ⁴⁸Ca and 40 Ca CaF₂(Eu) crystal scintillators is worthy of mention [45]. The third case was a cadmium tungstate crystal enriched in ¹¹⁶Cd produced to search for double β decay of ¹¹⁶Cd [7,8,46].

The emission spectra of ¹⁰⁶CdWO₄ crystal were studied under ultraviolet and X-ray excitation. We have found the wavelength of the peak emission maximum at $\approx 490 \text{ nm}$ under X-ray and ultraviolet excitation, in agreement with the literature data for cadmium tungstate crystals produced from natural cadmium.

The results of the optical transmission measurements demonstrate that the ¹⁰⁶CdWO₄ crystal has good transmission properties in the wavelength range 450-700 nm comparable with the best reported in the literature. The attenuation length of the material is $\approx 60 \text{ cm}$ at the wavelength of the emission maximum 490 nm. The energy resolution 10.0% (FWHM) was measured by the 106 CdWO₄ scintillator \emptyset 27 × 50 mm² (mass of 216g, produced from the boule by cut of the top and bottom conic parts on cleavage plane) with the 662 keV γ quanta of $^{137}\text{Cs.}$ The good optical and scintillation properties of the crystal are due to the improved quality of the material as a result of the high level purification both by cadmium distillation and by 106 CdWO₄ compound synthesis, as well as due to the advantage of using

the low-thermal-gradient Czochralski technique to grow the crystal. Altogether these results demonstrate that the $^{106}\mbox{CdWO}_4$ crystal scintillator has a good prospect as a detector to search for double beta processes in ¹⁰⁶Cd. Preparation of an experiment

to search for double β decay of ¹⁰⁶Cd with the help of the developed ${\rm ^{106}CdWO_4}$ crystal scintillator is in progress in the DAMA R&D set-up at the Gran Sasso National Laboratories of the INFN (Italy).

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