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NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH
Section A

Large volume $CdWO_4$ crystal scintillators

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Abstract

A low background scintillation spectrometer based on $CdWO_4$ crystals of large volume (100–200 cm^3) with improved energy resolution has been developed as a result of advances in crystal growth, optimization of light collection and design of a special electronic unit for amplification and shaping of the $CdWO_4$ signals. The energy resolution of crystals with volume 80–150 cm^3 is 10–12% for 662 keV γ-rays. A value of 7–8% at 662 keV, which is comparable to NaI(Tl), has been obtained with small samples (10–20 cm^3). The high purity of the $CdWO_4$ crystals (^{228}Th contamination is less than 8 $\mu Bq/kg$, that of ^{226}Ra less than 13 $\mu Bq/kg$) allows construction of unique spectrometers based on $CdWO_4$ crystals of large volume for environmental control, well logging and measurements of radioactive contamination in various samples with a sensitivity of 10^{-12} to 2×10^{-13} Ci/kg.

1. Introduction

During the last decade high-Z crystal scintillators have been developed stimulated by the practical needs of CT and experimental particle physics. They include bismuth germanate (BGO), $Bi_4Ge_3O_{12}$, gadolinium orthosilicate (GSO), Gd_2SiO_5, cadmium tungstate, $CdWO_4$, lead tungstate, $PbWO_4$, and others, whose distinguished property is a high detection efficiency for X- and γ-rays, as well as excellent operational characteristics (chemical resistance, non-hygroscopicity, etc.). These scintillators can be used for γ-ray registration in well logging applications and in low energy nuclear physics also.

The results of the elaboration of large volume $CdWO_4$ crystals and improvements of their spectrometric characteristics are summarized in this paper. This work has been performed since 1986 at the Institute for Nuclear Researches (Kiev) in cooperation with the Institute for Single Crystals (Kharkov), the Max Planck Institute (Heidelberg) and the Institute for Materials (Lvov).

The luminescence of $CdWO_4$ crystals was first reported in 1948 [1], and their application as scintillators in 1950 [2]. Since that time the scintillation, optical and mechanical properties of these crystals have been widely investigated [3–9]. The main characteristics of $CdWO_4$ are presented in Table 1. This material is non-hygroscopic and

Table 1
Main characteristics of $CdWO_4$ crystal scintillators

Characteristic [units]	Value	Ref.
Effective atomic number Z_{eff}	64	
Density [g/cm^3]	7.9	[2–8]
Melting point [K]	1598	[8]
Cleavage plane	⟨010⟩	
Hardness [Mohs]	4–4.5	
Hygroscopicity	No	
Wavelength of emission maximum[a] [nm]	470/540	[4,8]
Index of refraction	2.3	
Average decay time [μs]	12	[8]
Afterglow (after 3 ms) [%]	0.1	[5]
Light yield [% of NaI(Tl)]	25–40	[7,8]

[a] Two emission components were first observed in Ref. [4] with decay times of 5 and 20 μs and intensity maxima at 470 and 540 nm.

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chemically inert. The crystal has to be machined with care because of its cleavability. The important advantages of $CdWO_4$ crystal scintillators, apart from the high detection efficiency for X- and γ-rays mentioned before, are the relatively large light yield (about 40% of NaI(Tl)), radiation resistance and very slight temperature dependence of the scintillation output [3–9].

The main disadvantage of the $CdWO_4$ scintillator is the quite long decay time (12–15 ms), which restricts the value of its maximum counting rate. However, this is not really important in some applications such as, for example, the environmental measurement of radioactive contamination. Moreover, these crystals can be utilized successfully for the study of rare and forbidden processes in nuclear and particle physics. Indeed, the search for the double beta-decay of ^{116}Cd was carried out with the help of $CdWO_4$ scintillators enriched with ^{116}Cd to 83% [10]. In spite of the small volumes of the crystals used (10–20 cm^3), one of the best results on the neutrinoless double beta-decay was achieved with such a crystal [10]. To further advance this research and to improve the sensitivity of low level measurements of radioactivities it is necessary to use crystals of larger volume with better energy resolution. In principle, the high light yield of $CdWO_4$ crystals allows us to obtain an energy resolution that is comparable to that of NaI(Tl) but, in fact, an energy resolution of only 8% (FWHM at an energy of 662 keV) has been reached with a very small crystal (2 cm^3) [7]. For a $CdWO_4$ crystal with a volume of 12 cm^3 the best value was 9.1% (at 662 keV) and it was only 19% for a 50 cm^3 crystal [8]. Such extreme distortion of the energy resolution is a result of the self-absorption of the scintillation light [8].

Summarizing the previous data [2–8] and the results of our tests [9] one can propose the following reasons for the deterioration of the energy resolution of large $CdWO_4$ crystals.

1. Self-absorption and defects of the crystal structure, which form inner scattering centers (optical inhomogeneities). These macrodefects can be caused by deviations from the optimum charge composition, by a significant amount of uncontrollable impurities in the mixture and also by temperature instabilities during crystal growth.

2. The shift between the maximum of the $CdWO_4$ scintillation emission and the spectral sensitivity maximum of standard photomultipliers.

3. Difficulties of light collection from $CdWO_4$ crystals because of the large index of refraction ($n = 2.3$).

4. Problems of output signal registration with standard electronics because of the long duration of the scintillation light pulse.

To overcome these difficulties, research and development was carried out on all mentioned points, which allowed, finally, improvement of the spectrometric characteristics of large volume $CdWO_4$ crystal scintillators.

2. Crystal growth, light collection and electronics

$CdWO_4$ single crystals were grown from oriented seeds by the Czochralski technique from a stoichiometric mixture of high purity CdO and WO_3 oxides. The crystal diameter was controlled during growth by the weight method. To avoid the appearance of micro-pores inside the crystal and, as a consequence, its optical inhomogeneity, the required thermal conditions (radial and axial temperature gradients and their stability) were provided by a specially constructed thermal unit. Heat screens and doubled corundum heat insulation were used. After growth the crystals were cooled slowly and annealing was performed at a constant temperature over about 80 h. The obtained single crystals were sufficiently transparent even for samples of 150 mm length and had a light yellow color.

The scintillation emission intensity of $CdWO_4$ has a maximum at 490 nm, which does not correspond to a maximum of the typical spectral sensitivity of photomultipliers (PMT). To find an appropriate photomultiplier a test of PMT types and individual tubes was carried out. The best result was obtained with Philips XP2412 photomultipliers, but some tubes of the FEY-139 type showed similar characteristics.

It is known that the best energy resolution can be obtained for samples of the correct geometric form (a cylinder is desirable) with a smooth side surface. The crystal surfaces have to be sandpapered, but not polished. The choice of reflecting material and wrapping is also important. For the crystals whose height was less than their diameter we obtained the best energy resolution using a few layers of 5 μm thick Teflon film. For long crystals, wrapping of the side surface with wrappings of different reflecting capability was used. The upper part of the crystals was covered with layers of Teflon film and an aluminized Mylar film to obtain the best reflection. Then, near the PMT photocathode, materials with reduced reflecting capability were used. This approach allows improvement of the uniformity of the light output of the crystal by means of a $\approx$10% reduction of the total light yield. The resulting improvement of the energy resolution by a factor of 1.2 was achieved due to a non-uniform wrapping for crystals with height greater than 50 mm. The light yield distribution along the axis of the crystals was measured with a collimated beam of γ-quanta from a ^{207}Bi source.

The large decay time is a serious drawback for the practical application of $CdWO_4$ scintillators. The decay constant, with a mean value of 12–15 μs, leads to a required full light collection time of about 30 μs, whereas standard spectrometric electronics imply pulses of a few μs. To solve this problem a special electronic unit was designed and realized as a double-width CAMAC module. This unit contains a leading edge trigger, fast linear amplifier, integrator and output gate. The integrator is opened for 30 μs after the moment when the input signal exceeds the discrimination level. Then, the output gate forms a short output signal with amplitude proportional to the collected charge. The combination of a fast amplifier and an integrator minimizes pile-up effects, providing complete collection of the emitted light during 30 μs. In tests with the ^{207}Bi γ-source no distortion of the energy resolution and no peak shift were found until a counting rate of 3000 counts/s. The electronic unit provides an energy threshold of about 30 keV and a dynamic range of 1:80 for γ-rays detected by $CdWO_4$ scintillators. A built-in pulse shape discrimination circuit allows rejection of the noise pulses and additionally reduces the energy threshold of the spectrometer.

3. Energy resolution and background measurements of $CdWO_4$ crystals

Results of energy resolution measurements with different large volume $CdWO_4$ crystals are presented in Table 2. Also shown is the energy resolution of a "standard" $CdWO_4$ crystal of small volume ($V = 10$ cm^3), which is the best published value – 7.5% (FWHM) at an energy of 662 keV (Fig. 1). The calibration spectrum shown in Fig. 2 demonstrates good energy resolution and peak-to-Compton ratio for a crystal with mass 1.1 kg, which exceed considerably the characteristics published previously.

Thus, as a result of the fulfilled R&D on the growth and annealing of high quality crystals, selection of the proper PMT, choice of the complex wrapping and use of special electronics, the energy resolution of large volume $CdWO_4$ crystals has improved to a level that can be compared to the characteristics of NaI(Tl) scintillators.

The intrinsic radioactive impurities of crystal scintillators ultimately determine the sensitivity of the low background installations using these scintillators. Therefore, the background of the developed $CdWO_4$ crystals was measured to test their feasibility for a planned large-scale experiment on the double beta-deçay of ^{116}Cd [10] and also for ultra-low radioactivity detection. The measurements were carried out at the Solotvina Underground

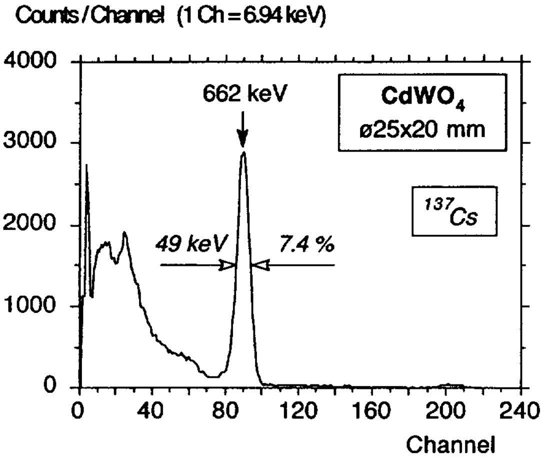

Fig. 1. Gamma-ray spectrum of ^{137}Cs on a "standard" $CdWO_4$ crystal of ∅25 × 20 mm.

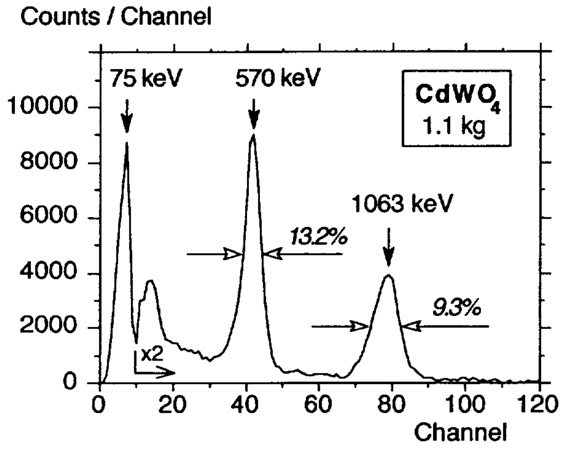

Fig. 2. Calibration γ-ray spectrum of ^{207}Bi measured with $CdWO_4$ scintillators of ∅54 × 65 mm.

Table 2
Energy resolution of $CdWO_4$ scintillators

$CdWO_4$ crystal		Energy resolution [%]		
Dimensions (volume)	Weight [kg]	570 keV	662 keV	1063 keV
∅40 × 30 mm (37.5 cm^3)	0.3	9.7	8.5	6.7
∅50 × 37 mm (73 cm^3)	0.6	11.1	9.8	7.6
∅54 × 65 mm (149 cm^3)	1.1	13.2	11.4	9.3
∅54 × 95 mm (217 cm^3)	1.7	14.9	13.8	11.1
∅25 × 20 mm (10 cm^3)	0.08	8.8	7.5	6.4

Laboratory built in a salt mine at a depth of more than 1000 mwe [11] where the cosmic muon flux is suppressed by a factor of 10^4. Due to the low radioactive contamination of the salt (NaCl), the natural γ background is 30–70 times lower than in chambers built of common materials [11].

In the low background installation a $CdWO_4$ crystal of volume 149 cm^3 was viewed by the PMT through a quartz light-guide 50 cm long. A passive shielding of OFHC copper (5 cm), mercury (7 cm) and lead (15 cm) surrounds the $CdWO_4$ scintillator to reduce the external background. The data acquisition system consists of a microcomputer, magnetic tape recorder and CAMAC crate with electronic modules. The energy calibration was performed with ^{207}Bi.

The background spectrum of $CdWO_4$ (149 cm^3) measured in this installation for 107 h is shown in Fig. 3. There are no peaks and only one peculiarity exists – the sharp increase below an energy of 320 keV which is the spectrum of the fourth-forbidden β-decay of ^{113}Cd (natural abundance 12%) with a half-life of $T_{1/2} = 9.3 \times 10^{15}$ yr and endpoint energy 319 keV [12]. The background rate in the energy region 2–3 MeV is ≈5 counts/yr/keV/kg, which is close to that of ultra-low background HP Ge detectors [13]. It is known that the background of the best scintillation setups, based on NaI(Tl) and others, is larger than for semiconductor detectors by one to two orders of magnitude [14]. The very low value of the background rate obtained in our tests is evidence of the high intrinsic purity of the grown crystals. Very small limits of radioactive contamination were calculated from the background spectra: ^{228}Th, 8 μBq/kg; ^{226}Ra, 13 μBq/kg; ^{40}K, 4 mBq/kg; ^{137}Cs, 0.3 mBq/kg; and ^{90}Sr, 3 mBq/kg. Such high purity is an important advantage of the developed $CdWO_4$ crystal scintillators. For example, it was found earlier that BGO crystals contain an activity of 500–600 mBq/kg of ^{207}Bi [15] and subsequently cannot be used in low background experiments.

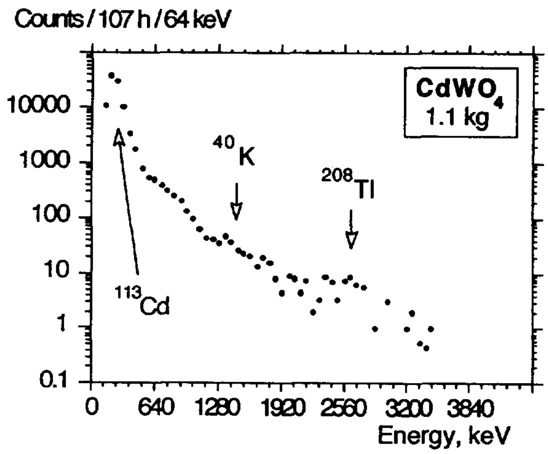

Fig. 3. Background spectrum of a $CdWO_4$ crystal (⌀54 × 65 mm) recorded during 107 h in the low background installation located at the Solotvina Underground Laboratory.

The spectrum in Fig. 3 was also used to estimate the sensitivity of a low background spectrometer with a $CdWO_4$ crystal (149 cm^3) for measuring the radioactive contamination in the environment. The detection efficiency for γ-rays emitted by a 1 kg sample of standard earth was calculated using the CERN code GEANT 3.14 [16]. It was calculated that the sensitivities for the detection of, for example, ^{40}K and ^{208}Tl are 1.2×10^{-12} and 2×10^{-13} Ci/kg, respectively (for a measuring time of 24 h and with a statistical accuracy of 30%). This sensitivity corresponds to the characteristics of the best low background HP Ge spectrometer [13]. In fact, the latter installations provide more complete spectrometric information due to the better energy resolution. But taking into account the simplicity, operational requirements and lower cost, $CdWO_4$ low background spectrometers are preferable for some applications in low level activity measurements.

4. Conclusions

A low background scintillation spectrometer based on large volume $CdWO_4$ crystals (100–200 cm^3) with improved energy resolution has been developed as a result of advances in crystal growth, optimization of light collection and design of a special electronics unit for amplification and shaping of the $CdWO_4$ signals. The energy resolution of crystals with volume 80–150 cm^3 is 10–12% for 662 keV γ-rays. A value of 7–8% at 662 keV has been obtained with small samples (10–20 cm^3), which is comparable to NaI(Tl). The special electronics unit provides constant characteristics of the spectrometer for counting rates up to 3000 counts/s.

The measurements carried out in the Solotvina Underground Laboratory gave the following limits for the internal contamination of the $CdWO_4$ crystals: ^{228}Th, 8 μBq/kg; ^{226}Ra, 13 μBq/kg; ^{40}K, 4 mBq/kg; ^{137}Cs, 0.3 mBq/kg; and ^{90}Sr, 3 mBq/kg. The background rate of $CdWO_4$ crystals of large volume (149 cm^3) is 5 counts/yr/keV/kg in the energy region 2–3 MeV, which is similar to the corresponding feature of the best low background HP Ge detectors. The sensitivity for the detection of ^{40}K and ^{208}Tl contamination in a 1 kg sample of standard earth is 1.2×10^{-12} and 2×10^{-13} Ci/kg, respectively (for a measuring time of 24 h and with a statistical accuracy of 30%). This allows construction of unique spectrometers based on large volume $CdWO_4$ crystals for environmental control, well logging and measurements of radioactive contamination in various samples.

Moreover, considerable progress can be attained with large volume $CdWO_4$ crystals in the planned experiments to study the double beta-decay of ^{116}Cd and other rare decays.

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