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Cadmium Tungstate Scintillators with Excellent Radiation Hardness and Low Background

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Abstract

Single crystals of CdWO_4 were irradiated by ^{60}Co γ -rays for the accumulated doses in the range of 10^4 to 10^6 rad. Degradation in optical transmittance was less than 2%/cm for the doses up to 10^6 rad as far as the measurement was carried out a few to several tens hours after irradiation. The result is discussed in comparison with the larger damage reported previously. The intrinsic background spectrum indicates that radioactive impurities in CdWO_4 are much less than in GSO:Ce and BCO .

1. Introduction

Cadmium tungstate, CdWO_4 , has already been established as one of dense (density $\rho = 7.9 \text{ g/cm}^3$) and large light output (38% of NaI:Tl) scintillators and is widely used as detectors for X-ray computed tomography (X-CT). Characteristics of CdWO_4 are listed in Table 1 in comparison with the other typical scintillators. Scintillation in CdWO_4 was first observed in 1948 by Kroger[1] in a systematic study of the luminescence of various tungstate powders. Single crystals of CdWO_4 were grown by the Verneuil method and evaluated as a γ -ray detector[2]. Since then significant progress [3-8] has been made in the crystal growth, mostly by the Czochralski method, as well as in the study of scintillation characteristics.

As for the radiation hardness of CdWO_4 , only one measurement by Ishii et al.[8] has been published to the authors' knowledge. The radiation damage and its recovery with time in a 3-mm thick crystal were studied for the irradiation by 120 keV X-rays up to 2×10^5 rad. Degradation in luminescence was 20% (10%) at 2 hours after irradiation by 2×10^5 (2×10^7) rad. It recovered with a time constant as large as 20 hours. Almost complete recovery was observed within 100 hours.

Recently, we have measured the radiation damage of CdWO_4 due to low energy γ -rays from ^{60}Co . It is the main aim of the present paper to present the result of this measurement up to 10^6 rad, a much larger dose than used by Ishii et al.[8]. We have also measured the background energy spectrum in CdWO_4 , since there is ever increasing interest in γ -ray detectors with low intrinsic background. The ^{115}Cd isotope, which is abundant by 7.5 % in natural Cd, is one of good candidates [9] for the study of neutrino-

less double beta decay, one of important subjects in elementary particle physics.

2. Measurement

CdWO_4 crystals were grown by Furukawa Co. using the Czochralski method. Two rectangular crystals with a size of $1 \times 1.2 \times 4 \text{ cm}^3$ were cut from the ingots and polished to the optical grade on the whole surface. These ingots were grown in the course of studying the effects of different dopants on CdWO_4 . One of the test samples, No.1, was pure without dopants. Among various dopants added into the melt, cesium kept the crystal colourless as in the case without dopants. However, cesium did not enter the host lattice; the amount of cesium in the grown crystal was less than the detection sensitivity of 0.06 ppm (by weight) of the atomic absorption analysis. The No.2 sample was thus grown by adding cesium into the melt, although the crystal does not contain cesium. We included this crystal (No. 2) in the present test carried out at visible wavelengths, since this crystal showed sizably better optical transmission in the infrared region.

We observed the scintillation pulse shape for ^{137}Cs γ -rays on an oscilloscope, by mounting the crystal on a photomultiplier (Hamamatsu R329) with a bialkali photocathode. The decay constant was approximately $10 \mu\text{s}$ for both crystals. Optical transmission spectra were measured across the 1.2-cm thickness with a spectro-photometer (Hitachi 330) before and after irradiation. Excitation and emission spectra were measured with a fluoro-spectrophotometer (Shimadzu RF-510) in a configuration of the excitation and the emission at right angles to each other.

Irradiation with ^{60}Co γ -rays was carried out at Japan Atomic Energy Research Institute (JAERI). Five cycles of irradiation, followed by transmission measurement, were carried out to cover the accumulated dose[10] from 10^4 to 10^6 rad by increasing the dose by a factor of 10 per cycle. Each irradiation period took 1.5, 0.9, 3, 65 and 147 hours for the accumulated dose of 10^4 , 10^5 , 10^6 and 10^7 rad, respectively, by mounting the samples at different distances from the ^{60}Co source. The irradiated samples were sent to KEK and the transmission measurement was carried out at 218, 36, 81, 37 and 52 hours after irradiation by 10^4 , 10^5 , 10^6 , 10^7 and 10^8 rad, respectively. To avoid possible annealing by ultra-violet (UV) light as seen in CsI:Tl [11], PbF_2 [12], BaF_2 [13], etc., we paid attention for safety lest a sample should be exposed to strong UV-light, including the sunshine, during any course of the irradiation-measurement cycles. Each crystal was wrapped with aluminium foil except during a short period for transmission measurement which took about 15 minutes per measurement per sample.

To measure the intrinsic background energy spectrum in CdWO_4 , we used another large crystal of 4.5 cm in diameter and 2 cm in thickness. The energy scale was calibrated with γ -emitting isotopes. The crystal was mounted on a 2-inch photomultiplier (Hamamatsu R329) by inserting a 5 cm diameter, 10 cm long acrylic light guide between the crystal and the photomultiplier in order to suppress the background γ -rays coming from the photomultiplier. Measurement was carried out in a box shielded with 5-cm thick Cu (OFHC) and 10-cm thick Pb.

3. Result

Transmission spectra

Transmission spectra of the two samples are given in Figs. 1a (for No. 1) and 1b (No. 2) for different accumulated doses, i.e. 0 (before irradiation), 10^6 and 10^7 rad. The results for 10^6 (10^6 and 10^7) rad, which were close to those for 10^6 (10^6) rad within 1%, are not shown in the figures for simplicity. The spectra for different doses were normalized at 600 nm, where the transmittance was the same to within the reproducibility of 1%. The degradation in transmittance was saturated above 10^6 rad and was less than two percent up to 10^7 rad. It should not be difficult to monitor and correct the degradation of this small amount in actual γ -ray detectors, since the degradation should usually occur slowly. We found that the recovery in the period from 52 hours to 21 days after irradiation by 10^6 rad was not sizable.

Excitation and emission spectra

Excitation and emission spectra are presented in Fig. 2 for No. 1 crystal; the result for No. 2 was almost the same. The excitation and the emission peaks sit at 330 and 470 nm, respectively. After irradiation by 10^6 rad, the excitation-emission spectra remained the same as before to within an ambiguity as large as 10 % which comes from the reproducibility error in mounting the crystal on the spectro-fluoro-photometer.

Background energy spectrum in CdWO₄

Figure 3 shows a background energy spectrum accumulated for 2×10^6 s. The energy resolution was roughly 34 % (20 %) in FWHM at 0.551(1.275) MeV. We have not seen any clear peaks or structures which are usually seen below ~ 2 MeV in other materials such as GSO:Ce (Gd₂SiO₅:Ce)[14] and BGO (Bi₄Ge₃O₁₂) [15]. To compare the background level of CdWO₄ with GSO:Ce and BGO, we

made similar measurement for a GSO:Ce(0.5 %) crystal (grown by Hitachi Chemical) with 5 cm diameter and 3 cm thickness and a BGO crystal (grown by NKK) with 5 cm diameter and 2 cm thickness. When compared typically at 1 MeV, the background level of 0.03 counts/h/cm²/keV in CdWO₄ (see Fig. 3) was about 1/8 of that in GSO:Ce and 1/2 of that in BGO, although substantial reduction of background, typically by a factor of 1/4 at 1 MeV, has been obtained[15] in BGO grown from specially selected raw material. The above result indicates that the radioactive impurities such as U and Th series should be much less in CdWO₄ than in GSO:Ce and BGO. Since the present CdWO₄ is of the ordinary grade with respect to the radioactive background, there may be a possibility for further reduction by selecting the raw material. CdWO₄ may be useful in experiments which require low intrinsic background.

3. Summary and discussions

The above-mentioned result can be summarized as follows:

- (1) Single crystals of CdWO₄ show excellent radiation hardness against low energy γ -rays, as far as the measurement is carried out a few to several tens hours after irradiation. Degradation in optical transmittance was less than 2 %/cm for the doses up to at least 10^6 rad.
- (2) Adding Cs into the melt neither improved nor degraded the crystal quality at visible wavelengths, including radiation hardness, optical transmittance, excitation-emission characteristics, etc.
- (3) The background energy spectrum shows no clear peaks due to radioactive impurities such as U and Th series. The background level was typically 0.03 counts/h/cm²/keV at 1 MeV, which is about 1/8 of GSO:Ce and 1/2 of BGO. This result indicates that CdWO₄ can be used as γ -ray detec-

tors with low background.

The excellent radiation hardness summarized above in (1) is apparently different from the observation by Ishii et al.[8] (see Section 1). We cannot exclude a possibility of a sizable radiation damage and its quick recovery. One of the possible scenarios is as follows: degradation in transmittance increases with the accumulated dose but saturates around 10^{15} rad. The damage recovers with a time constant as short as 10-20 hours. Another scenario is that the present CdWO_4 crystals should have much higher radiation hardness than those of [8], i.e. absence of any sizable radiation damages even during irradiation. From a practical viewpoint, however, the conclusion is the same for both scenarios. CdWO_4 can be used in the high level radiation field even in the first scenario of the fast recovery of a sizable damage, since the accumulation of the dose should be usually much slower than the recovery.

CdWO_4 is denser (the radiation length $X_0=1.06$ cm) than BaF_2 [16] ($X_0=2.1$ cm) and GSO:Ce [17] ($X_0=1.38$ cm), two typical crystals with very high radiation hardness. Moreover CdWO_4 is easier to handle (no hygroscopicity) than BaF_2 and much cheaper than GSO:Ce in the raw material cost. CdWO_4 could be used as detectors or monitors for intense X-ray or γ -ray sources. If the slow decay constant ($\tau \sim 10 \mu\text{s}$) can be tolerated. Besides the application as scintillators, CdWO_4 can be used as transparent viewing windows in high radiation field, for example, around reactors.

After completion of the present work, we have noticed a recent work by Doychil[18]. Luminescence degradation in small CdWO_4 crystals (2-mm wide and

3-mm thick) due to 120 kV X-ray irradiation and its recovery with time were studied similarly as in the work by Ishii et al.[8]. A sizable degradation in luminescence (14% at 10^5 R) was qualitatively similar as in [8], while insufficient recovery (50-60%) even in 121 hours were different from both the present work and [8].

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Figure captions

- Fig. 1 Transmission spectra of CdWO_4 crystals, (a) No. 1 and (b) No. 2, for different accumulated doses. Solid curve: before irradiation, dotted one: 10^6 rad, chained one: 10^8 rad.
- Fig. 2 Excitation-emission spectra of CdWO_4 , No. 1 sample.
- Fig. 3 Background spectrum of CdWO_4 shielded by 5-cm thick Cu and 10-cm thick Pb.

Table 1 Characteristics of typical scintillators.

Quantity	NaI:TI	CsI:TI	CdWO ₄	BGO	BaF ₂	GSO:Ce
Afterglow(%/msec)	0.5-5.0/3	0.1-0.8/6	0.005/3	0.005/3		
Absorption coeff. (cm ⁻¹) [*]	2.22	3.24	7.68	9.98	3.13	5.74
Thickness to absorb 99%(mm) [*]	20.7	14.2	6	4.6	14.7	8.0
Thickness to absorb 90%(mm) [*]	10.4	7.1	3.0	2.3	7.4	4.0
Density (g/cm ³)	3.67	4.53	7.90	7.13	4.89	6.71
Radiation length X ₀ (cm)	2.59	1.86	1.06	1.12	2.03	1.38
Decay constant (ns)	230	1050	5000	300	0.6/600	30-60
Peak emission (nm)	415	550	470	480	220/310	440
Light yield (relative)	100	85	38	7-10	5/16	20
Index of refraction [*]	1.85	1.80	2.3	2.15	1.56	1.85
Peak excitation (nm)	290	300	330	280	---	310, 350
Hygroscopicity	strong	slight	no	no	little	no
melting point (°C)	651	621	1272	1050	1354	1900
Radiation resistivity (rad)	10 ³	10 ³	10 ³	10 ⁻⁸	10 ⁻⁷	10 ³
Hardness (Mohs)	2	2	4-4.5	5	3	5.7
Cleavage	(100)	none	(010)	none	(111)	(100)

*) at 150 kev +) at the wavelength of the peak emission.

Fig. 1a

Fig. 1b

Fig. 2

Fig. 3