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FIRST STUDY OF RADIATION HARDNESS OF LEAD TUNGSTATE CRYSTALS AT LOW TEMPERATURES

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Abstract

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The electromagnetic calorimeter of PANDA at the FAIR facility will rely on an operation of lead tungstate (PWO) scintillation crystals at temperatures near −25 °C to provide sufficient resolution for photons in the energy range from 8 GeV down to 10 MeV. Radiation hardness of PWO crystals was studied at the IHEP (Protvino) irradiation facility in the temperature range from room temperature down to −25 °C. These studies have indicated a significantly different behaviour in the time evolution of the damaging processes well below room temperature. Different signal loss levels at the same dose rate, but at different temperatures were observed. The effect of a deep suppression of the crystal recovery process at temperatures below 0°C has been seen.

Keywords: lead tungstate, radiation hardness, recovery, low temperature.

Аннотация

Семенов П.А., Узунян А.В., Давиденко А.М. и др. Исследование радиационной стойкости кристаллов вольфрамата свинца при низких температурах: Препринт ИФВЭ 2007–4. – Протвино, 2007. – 9 с., 7 рис., 2 табл., библиогр.: 15.

Электромагнитный калориметр эксперимента PANDA (FAIR) будет состоять из сцинтилляционных кристаллов вольфрамата свинца (PWO), находящихся при температурах близких к −25 °C, чтобы обеспечить регистрацию фотонов в диапазоне энергий от 8 ГэВ до 10 МэВ. Исследование радиационной стойкости кристаллов PWO проведено в ИФВЭ (Протвино) в диапазоне температур от комнатной температуры до −25 °C. Исследования показали существенное различие в поведении процессов повреждения при низких температурах по сравнению с комнатной температурой. Обнаружено различие в уровнях падений сигналов при одинаковой мощности дозы облучения, но при разных температурах. Выявлен эффект глубокого подавления восстановительных процессов в кристаллах при температурах ниже 0°C.

1. Introduction

Total absorption shower counters made of lead tungstate ($PbWO_4$ or PWO) scintillation crystals are very promising for development of high quality electromagnetic calorimeters [1]. They have already been used in the PRIMEX experiment at JLaB and manifested a brilliant π^0-mass resolution (1.5 MeV/c^2) [2]. The CMS electromagnetic calorimeter consisting of 62,000 PWO crystals in the barrel part and 15,000 in the endcap part is due to start operation at LHC soon [3]. Both the calorimeters work at room temperature.

ALICE, another calorimeter at LHC, will be the first PWO-based calorimeter in the world operating at a temperature decreased down to –25 °C. An increase of the light yield by a factor of 3 in comparison with operation at room temperature (+20 °C) is expected [4]. Moreover, the noise of the calorimeter electronics will be reduced. Both these effects will improve the energy resolution of the calorimeter. The same operation temperature of –25 °C is selected also for 20,000 PWO crystals in the PANDA calorimeter, a new experiment in the FAIR project at GSI, Germany [5].

Radiation hardness is another important issue in application of PWO scintillators in high energy physics experiments. It is well known that crystal scintillators suffer from radiation damage. This problem has been extensively studied over the last seven years at CMS [6,7] and BTeV, the Fermilab experiment which was terminated in 2005 [8,9,10]. However, all the studies of PWO radiation hardness have been carried out only at room temperature.

In this paper we present the results of the first investigation of PWO crystal radiation hardness at temperatures below 0 °C. The temperature was varied in the range from +20 °C down to –25 °C. The study was carried out for two different radiation loads of 20 and 2 rad/h (radiation loads for the PANDA endcap electromagnetic calorimeter are estimated to be up to 2–3 rad/h).

We used eight crystals produced by the "North Crystals" Company in Apatity (denoted by the letter "a" in this paper) and two more crystals produced by the Bogoroditsk Techno-Chemical Plant ("b"). The crystals which are marked as "b", as well as a1412 and a1434, were produced at the end of 2002. The other crystals were produced in 2005. The dimensions of the crystals were $\sim 22\times$ 22 mm^2 in cross section and 180 mm in length.

The crystal specifications data given by manufactures before the irradiation are provided in Table 1. The data show that these samples are typical examples of PWO scintillation crystals

with similar transmittance and light yield however they differ by the doped elements and the growth number (number of a crystal growing in the same crucible without cleaning of it).

Table 1. PWO crystal specifications data before irradiation.

Serial number	Doped element	Concentration, (ppm)	Growth number	Light Yield (phe/MeV)	Transmittance at 420 nm (%)	Transmittance at 632 nm (%)
a15608	Gd	80	1	10.9	69.0	73.9
a15517	Gd	80	1	10.5	71.3	74.0
a15851	Gd	80	1	11.0	71.3	73.9
a1412	Y	100	5	9,2	64.3	72.6
a1434	Y	100	4	9.0	65.6	73.1
a15500	Y	90	1	12.0	70.2	74.0
a16618	La	38	1	11.2	71.9	74.0
a16628	La/Y	22/20	1	12.3	71.3	74.0
b389	N/A*	N/A	N/A	10.3	70.5	72.9
b404	N/A	N/A	N/A	8.9	66.8	71.9

∗Data not available

2. Experimental procedure

A radioactive source ^{137}Cs emitting 661 KeV photons and having activity of $5 \cdot 10^{12}$ Bq was used to irradiate the crystals. Five crystals were irradiated simultaneously during each exposition. A commercial dosimeter, DKS-AT1123, was used to measure the dose rate (in air) in proximity to the crystals. The accuracy is better than 20% [12]. The simulated transverse radiation dose rate profile in crystal is shown in Fig. 1 [13].

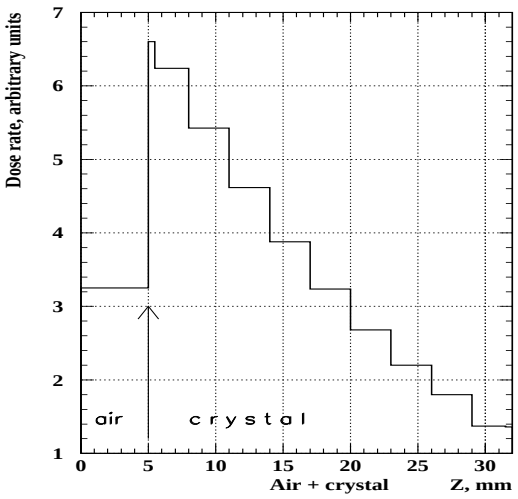

Figure 1. Transverse dose rate profile for crystal under ^{137}Cs gamma irradiation. The dose rate was estimated both in air and crystal, as indicated.

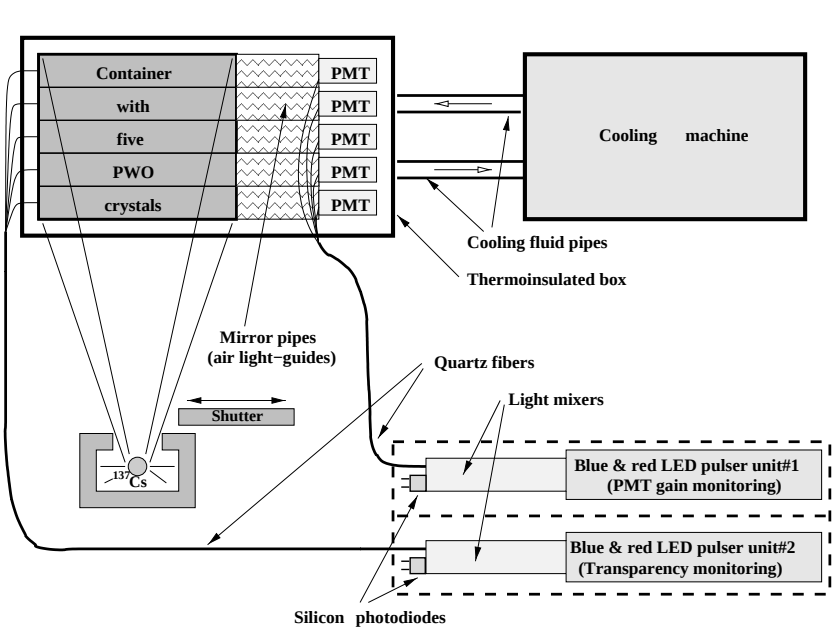

Figure 2. Layout of the gamma irradiation setup. The details are in the text.

Layout of the gamma irradiation setup is depicted in Fig. 2. Cryothermostat LAUDA RC6CP was exploited to stabilize and control the temperature of the crystal set under irradiation. Cooling power of 300 W provided by this cryothermostat was sufficient to maintain the constant temperature for a long time with the absolute accuracy of 0.1 °C. Glycol aqueous solution circulating between the LAUDA bath and the heat exchanger plate, where the crystals were installed, was used to maintain the crystals at a required temperature. The bases of the photomultiplier tubes (PMTs), which were installed outside the heat exchanger, were the main heat sources inside the crystal compartment. To make the temperature distribution more uniform, a permanent gas flow inside the compartment was provided using a fan. Dry nitrogen gas was supplied to the compartment to avoid problems with moisture gathering. To monitor the temperature of the crystals, several thermosensors (type PT100 and PT1000), were installed on the heat exchanger plate and directly on the crystals. To monitor variation of the light transmittance of the crystals due to irradiation as well as possible PMT gain instabilities, we used an LED-based monitoring system. This system enabled us to investigate the radiation-induced absorption of the crystals in two different spectral regions, in the vicinity of 450 nm and 640 nm. The system consisted of two similar modules. Each module contained blue or red LEDs with individual drivers, light mixer, one photodiode and fiber bundle. The system was similar to that described in more detail in [11].

A direct current (DC) method was used to test the radiation hardness of the crystals. The mean current value of the PMT through a 10 kOhm resistor was measured by sigma-delta ADC. This signal was proportional to the luminescence intensity and, consequently, to the light output of the crystal. The change of this signal provided an information on the changes in the light output invoked by irradiation. Simultaneously, the LED based monitoring system provided information on variation in crystal light transmittance. Since the level of the DC signal was

low (a few microamps), sensitive measurement devices were installed in a close proximity to the photodetectors. Digitized signals were transferred further to a remote computer using the RS485 protocol.

Ten crystals (two groups, five crystals in each group) were irradiated at the dose rate of 20 rad/h (and 2 rad/h) at different temperatures. Each cycle lasted from 50 to 300 hours. Before each cycle, the crystals were exposed to the daylight at room temperature to recover after the previous cycle of irradiation. The recovery was tested by a reference measurement of the light output at +20 °C. We observed that the crystals restore their initial light output within 5% after four days of such recovery.

3. Results

As it might be expected, cooling of the scintillation crystals increases their light output. Decrease of temperature from +20 °C down to –20 °C resulted in an increase in the PMT signal corresponding to the light output by approximately a factor of 3. The distribution of this enhancement factor for all 10 samples under study is presented in Fig. 3. The PMT current as a function of temperature for five crystals from one group is presented in Fig. 4. Four cycles of measurements at four different temperatures, +20 °C, +5 °C, –10 °C, and –22 °C, were performed.

The curves in Fig. 4 are just a guide for the eye. Before the irradiation, the light output at different temperatures of all five samples was the same within the experimental error. After the irradiation, the temperature dependence of the light output became less pronounced. The trend of this dependence was the same for all crystals, but the differences in absolute values were much higher than those before irradiation. As the radiation hardness of the crystals is slightly different, they loose different portions of their initial light output after the same period of irradiation at the same dose rate.

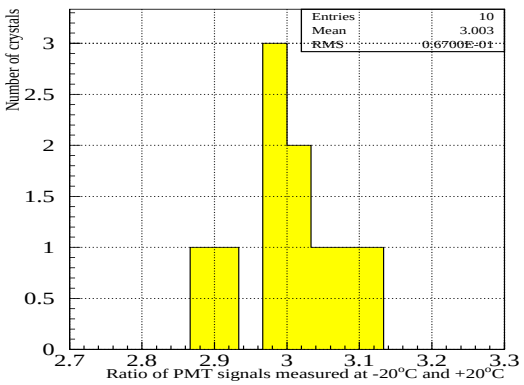

Figure 3. Distribution of the ratio of PMT signals measured at $-20°C$ and $+20°C$ among all ten crystals.

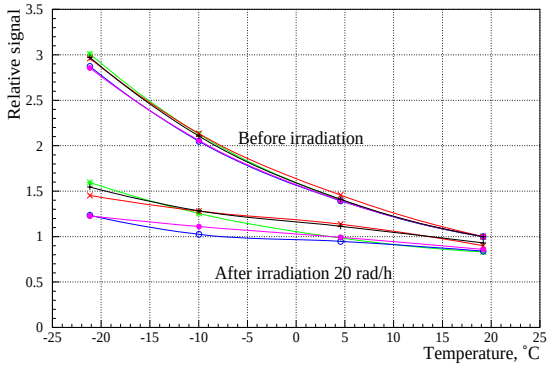

Figure 4. Relative light output as a function of crystal temperature. Five PWO samples were measured before irradiation and after irradiation over 80 hours at the dose rate of 20 rad/h.

To test the time evolution of the variation of the light output, we measured the PMT DC current as a function of time when the crystals were subjected to persistent irradiation at a constant dose rate.

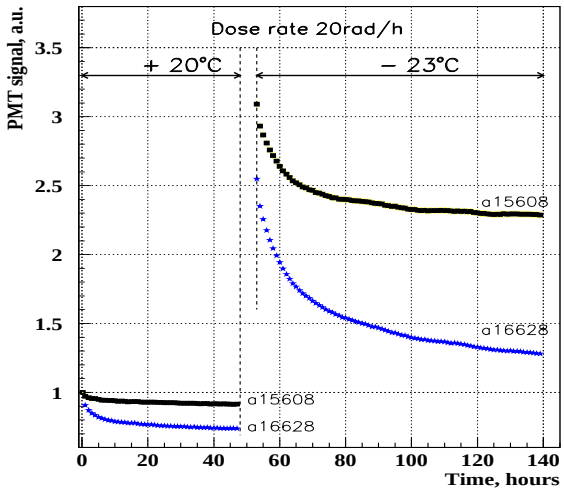

Figure 5. Time evolution of PMT signal of two samples (a15608 and a16628) subjected to irradiation at dose rate of 20 rad/h. Periods when constant temperatures were maintained (+20 °C and –23 °C) are indicated by horizontal arrows.

Typical curves of light output change under irradiation at the dose rate of 30 rad/h are shown in Fig. 5. Initially, +20 °C temperature was maintained. The signal dropped within the first 20 hours and approached practically a constant level afterwards. The drop was different for different crystals. As discussed above, decrease of temperature down to –23 °C caused an increase of the signal by a factor of approximately 3. The further irradiation caused a decrease of the signal. The signal also tends to a constant value. However, the time necessary to reach the constant value is longer, more than 300 hours, instead of 30 h at +20 °C. The next set of measurements was performed at the dose rate of 2 rad/h, which is about the maximal dose rate expected for the crystals in calorimeter for PANDA experiment. The same groups of the crystals were irradiated at +20 °C and –20 °C.

Fig. 6 shows that after initial drop the signal in all expositions tends to reach a constant value. The higher is the dose rate, the lower is the constant level and the faster this constant level is reached. As demonstrated also in Fig. 6, decrease of temperature enhances the initial signal drop and delays the process of approaching a constant light output level.

We also observed striking differences in recovery of the initial optical transmittance at the room and low temperatures. Almost no recovery is observed at –25 °C (see the first time period indicated in Fig. 7). Meanwhile, when the temperature is fixed at +20 °C, we can see a noticeable growth of the crystal transmittance (the second time period in Fig. 7).

The results of irradiation over 80 hours of the ten crystals at 2 and 20 rad/h dose rate for the two temperatures, +20 °C and –20 °C, are collected in the Table 2. We have not seen any significant difference in radiation hardness of the ten crystals dependent on the doped element (see Tables 1 and 2) neither on the manufacturer. Though we might notice that crystal a15608 is the most radiation hard.

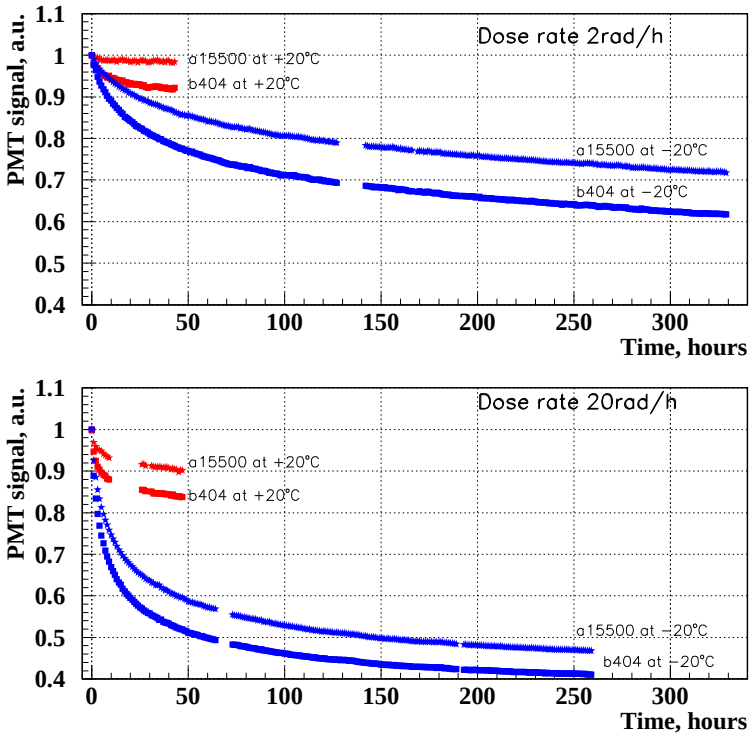

Figure 6. Time evolution of external light yield of two PWO crystals measured under 2 and 20 rad/h irradiation dose rate at temperatures of –20 °C and +20 °C, as indicated.

Table 2. Decrease of PMT DC signal in [%] as a result of irradiation over 80 hours at the two dose rates and the two temperatures for the ten crystals.

Serial number	2 rad/h at +20 °C	2 rad/h at –20 °C	20 rad/h at +20 °C	20 rad/h at –20 °C
a15608	2	13	9	30
a15517	1	16	8	43
a15851	5	21	15	51
a1412	1	17	16	54
a1434	5	20	18	44
a15500	2	18	10	45
a16618	5	23	20	50
a16628	6	26	26	60
b389	5	18	11	36
b404	8	27	16	52

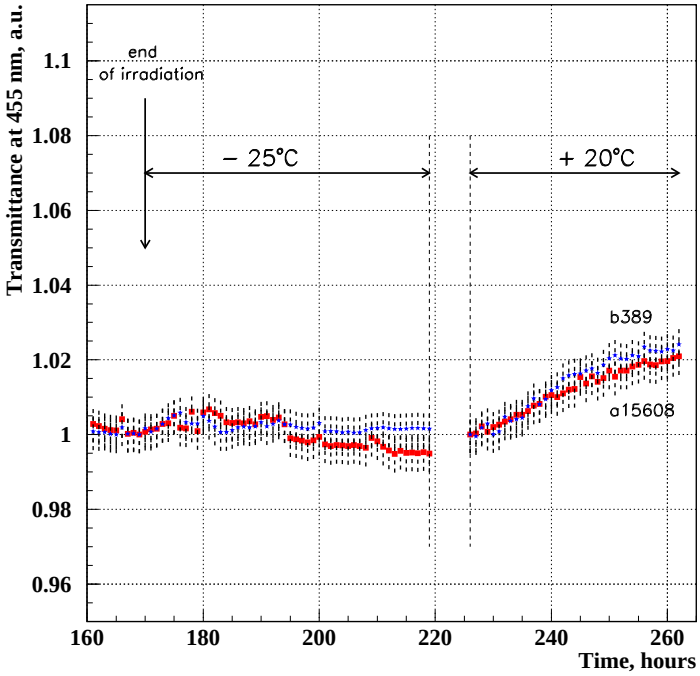

Figure 7. Time evolution of transmittance at 450 *nm* after exposition to irradiation. End of irradiation is marked by a vertical arrow; the periods of constant temperatures (–25 °C and +20 °C) are marked by horizontal arrows. The transmittance values at the beginning of recovery at –25 °C and +20 °C were normalized to one.

4. Discussion

It is most probable that the increase of light output at low temperatures is caused by decreased loss of excitation via non-radiative recombination. Meanwhile, the lower radiation hardness and suppressed recovery after irradiation, which were observed at low temperatures, are of considerable importance in application of PWO scintillation crystals as scintillators in cooled detectors. As it is generally accepted, the changes of light output of PWO crystals subjected to irradiation are caused not by any variations in emission efficiency but rather by radiation-induced absorption. Peculiarities of these features of radiation hardness will be studied in more detail in further publications. There are currently two approaches explaining the optical properties of PWO crystal under irradiation [6,14,15].

According to one approach [6], transmission damage occurs in crystal when valence electrons are trapped in metastable states around the crystal defects. The irradiation of the crystals creates color centers which absorb light in particular spectral regions and cause reabsorption of the PWO emission. When the rate of color centers production (proportional to the irradiation dose rate) equals the natural temperature-dependent recovery rate, the crystal light output reaches a saturation level (quasi-plateau). Thus, the temperature dependent features of irradiation-induced absorption and recovery of the initial transmittance after termination of the irradiation can be interpreted by the shift of the balance between production and recovery of the color centers.

According to another approach [14,15], the results presented above are in agreement with interpretation of radiation-induced changes in light output by structural changes in inclusions of variable-valency tungstate oxides WO_{3-x} [15]. Since tungsten has variable valency, irradiation invokes changes in tungsten valency, rearrangement of oxygen ions in the clusters WO_{3-x}, and, consequently, in change of absorption by these clusters. The irradiation-induced changes in the structure of WO_{3-x} clusters proceed slower at decreased temperatures, and the thermal energy is insufficient to facilitate the rearrangement of the initial cluster structure, which is necessary for recovery of initial optical transmittance.

5. Summary

To the best of our knowledge, this is the first study of radiation hardness of PWO scintillation crystals at temperatures decreased below the room temperature. Using a selection of 10 similar commercial PWO crystals produced by different manufactures we demonstrate that the radiation-induced decrease of light output proceeds slower at lower temperatures and the initial output does not recover after termination of irradiation when the crystal temperature is maintained at –25 °C. Nevertheless, the constant value of light output, which is reached after certain time under crystal irradiation at –25 °C, is still considerable higher than that at the room temperature. Thus, cooling of PWO detectors is beneficial, though the long time evolution of the light output of the crystal under irradiation at decreased temperature (of the order of hundreds hours) should be taken into account in application of these detectors.

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