

PAPER • OPEN ACCESS

Progress in the Development of the Lead Tungstate Crystals for EM-Calorimetry in High-Energy Physics

To cite this article: R. W. Novotny *et al* 2017 *J. Phys.: Conf. Ser.* **928** 012031

View the [article online](#) for updates and enhancements.

Related content

- [Growth of Crack-Free 3-Inch-Diameter Lithium Tetraborate Single Crystals by Czochralski Method](#)
Ryuichi Komatsu, Tadashi Sugihara and Satoshi Uda
- [Characterization of \$\text{YbNi}_4\(\text{P}_{1-x}\text{As}_x\)_2\$, \$x = 0, 0.2\$ single crystals grown by Czochralski method](#)
K Kliemt and C Krellner
- [Bulk Single Crystal Growth of \$\text{Bi}_{12}\text{TiO}_{20}\$ by the Czochralski Method](#)
Yasunori Okano, Hiroyuki Wada, Tsuguo Fukuda et al.



240th ECS Meeting ORLANDO, FL

Orange County Convention Center Oct 10-14, 2021

Abstract submission deadline extended: April 23rd

SUBMIT NOW

Progress in the Development of the Lead Tungstate Crystals for EM-Calorimetry in High-Energy Physics

R. W. Novotny¹, K.-T. Brinkmann¹, A. Borisevich², V. Dormenev¹, J. Houzvicka³, M. Korjik², and H.-G. Zaunick¹

¹Justus-Liebig-Universität Giessen, II. Physikalisches Institut, Heinrich-Buff-Ring 16 D 35392 Giessen, Germany

²Institute for Nuclear Problems, Bobruiskaya 11, 220030, Minsk, Belarus

³CRYTUR, spol. s.r.o., Palackeho 175, CZ-511 01, Turnov, Czech Republic

Email: r.novotny@exp2.physik.uni-giessen.de

Abstract. Even at present time there is a strong interest and demand for high quality lead tungstate crystals (PbWO_4 , PWO) for electromagnetic (EM) calorimetry. PWO is implemented into the EM calorimeter of the CMS-ECAL detector at LHC [1] and required for the completion of the PANDA EMC [2] and various ongoing detector projects at Jefferson Lab. The successful mass production of PWO using the Czochralski method was stopped after bankruptcy of the Bogoroditsk Technical Chemical Plant (BTCP) in Russia as major producer so far. The Shanghai Institute of Ceramics, Chinese Academy of Science (China) was considered as an alternative producer using the modified Bridgman method. The company CRYTUR (Turnov, Czech Republic) with good experience in the development and production of different types of inorganic oxide crystals has restarted at the end of 2014 the development of lead tungstate for mass production based on the Czochralski method. An impressive progress was achieved since then. The growing technology was optimized to produce full size samples with the quality meeting the PANDA-EMC specifications for PWO-II. We will present a detailed progress report on the research program in collaboration with groups at Orsay and JLab. The full size crystals will be characterized with respect to optical performance, light yield, kinetics and radiation hardness.

1. Introduction

The lead tungstate (PbWO_4 , PWO) scintillation crystal is one of the most widely used scintillation materials in electromagnetic calorimetry in modern accelerator experiments. It combines an unique set of physical and scintillation properties, which allow the construction of an affordable compact electromagnetic calorimeter (EMC). The first generation of mass production of PWO-I crystals was used in the electromagnetic calorimeter of the CMS detector at LHC. The technology of mass production was optimized to obtain detector units with high radiation hardness and less stringent requirements to the light yield [1] due to the high photon energies to be expected. The production technology of the next generation of lead tungstate, PWO-II for the PANDA EMC, was optimized to increase the light yield without degradation or even with achieving improvement of the radiation hardness [2]. The operating temperature of the PANDA EMC will be -25°C which enhances the light



yield by a factor of four due to the reduction of the quenching of luminescence but blocks statistical recovery of radiation damage during operation. The crystal material is considered as well as a candidate for some future experiments [3,4].

Crystals for CMS and partly for the PANDA calorimeter were produced by Czochralski method by the Bogoroditsk Technological Chemical Plant (BTCP, Bogoroditsk, Russia) until its shutdown some years ago. As an alternative producer for mass production of PWO crystals, Shanghai Institute of Ceramics, Chinese Academy of Science (SICCAS, Shanghai, China) was considered. It produces lead tungstate crystals by the modified Bridgman method.

The company CRYTUR (Turnov, Czech Republic) has a good experience in the development and production of different types of inorganic oxide crystals and has initiated a research program to develop the technology of mass production of lead tungstate crystals.

Table 1. The relevant parameters defining the PWO quality required for the PANDA-EMC. The parameter dk describes the radiation induced coefficients calculated as $dk = [\ln(T_b/T_a)]/d$, where T_b , T_a are the optical transmittances before and after irradiation, correspondently, and d is the thickness of the PWO sample. LY is the abbreviation for light yield, which is measured in units of photoelectrons per MeV, phe/MeV, deposited energy.

Parameter	Limit ^a
optical transmittance at 360 nm	$\geq 35 \%$
optical transmittance at 420 nm	$\geq 60 \%$
optical transmittance at 620 nm	$\geq 70 \%$
dk (420 nm)	$< 1.1 \text{ m}^{-1}$
LY at $T = +18^\circ \text{C}$ within 100 ns integration time gate	$\geq 16 \text{ phe/MeV}$
LY (100 ns)/LY(1000 ns) at $T = +18^\circ \text{C}$	$> 90 \%$

The aim of the R&D program was to obtain full size (up to 20 cm) lead tungstate crystals produced again by Czochralski method according to the PANDA EMC specifications as listed in table 1. This paper reports on results of the ongoing investigations.

2. Survey based on small samples

In the first stage, 11 series of lead tungstate ingots were grown under different conditions with respect to the atmosphere and the appropriate selection and concentration of dopants. In each case two small samples were cut from the top and bottom part of the ingot to study the homogeneity of the crystal quality. The samples have different shapes but the thicknesses are close to 1 cm with the two opposite sides optically polished to perform transmittance and light yield measurements. Besides the optical and luminescence properties the radiation hardness was investigated by determining the change of the transmission after γ -irradiation (^{60}Co) with an integral dose of 30 Gy at a dose rate of $\sim 2 \text{ Gy per minute}$ measured at room temperature using a Varian Cary 4000 spectrophotometer. The light yield was measured using a calibrated Hamamatsu R2059-01 photomultiplier at a temperature of $+18^\circ \text{C}$ and for selected samples at -25°C , respectively. TYVEK paper was used as reflector material.

As expected from previous experiences, the growth in $\text{N}_2 + \text{O}_2$ atmosphere and the doping with a sufficient amount of La/Y ions provided the best transmission characteristics near and below the emission wavelength of 420 nm as well as a good radiation hardness.

3. First results of large size crystals

As a crucial test of the new developed and installed furnace, full size ingots have been grown. The very first ingot showed a yellow coloring due to over-doping but the next ones were clear and visually transparent but slightly distorted due to an insufficient guiding of the pulling rod. The next obtained

ingots have been used to produce the first large size crystals with dimensions of 20x20x200 mm³ or 16x16x100mm³, respectively. The tests were primarily focusing on the reliability of the furnace. The crystals were initially grown in argon and finally in a N₂ atmosphere. As raw material not anymore used PWO-II crystals produced at BTCP have been chosen, since the optimization of the crystal production at CRYTUR will be based on the identical raw material selected and already pre-mixed in a specific stoichiometric composition optimized for BTCP. All samples were optically polished. The rectangular shape was chosen to allow a reliable investigation of the homogeneity of the crystal by measuring the optical transmittance and other parameters even transversal at different positions along the length of the crystal.

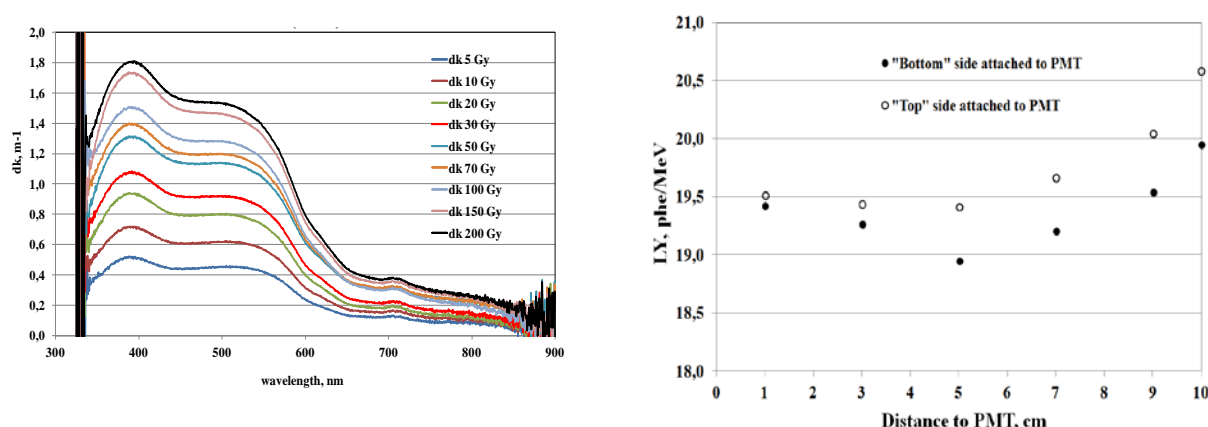


Figure 1. (left) Distribution of the longitudinal induced absorption coefficient of a 100 mm PWO crystal exposed to an integrated dose of up to 200 Gy. (right) The light yield measured as a function of position relative to the photo sensor for both crystal orientations. The positions "Top" and "Bottom" were randomly assigned to opposing ends of the crystal.

The shown sample of 100 mm length was free of visible imperfections or scattering centers. The optical performance showed only small variations along the crystal. However, the absolute value of the longitudinal transmission is significantly below the typical distribution of a former BTCP crystal. But the position of the absorption edge is nearly identical. The distribution of the induced absorption coefficient shows the typical behavior and the absolute value of dk at 420nm just meets the requirement to stay below 1.1m⁻¹. In order to test the homogeneity and the overall yield of luminescence, the light yield has been measured at room temperature for different locations of the ¹³⁷Cs source. On average, the observed light yield appears to reach the required limit even when taking into account the shorter crystal length. Figure 1 illustrates the already achieved radiation hardness and light yield.

4. The present performance of full size rectangular crystals

The most recent ingots show already a significant overall improvement with respect to colouring and overall size, which was achieved by modifications and optimizations of the growing technology and the fine tuning of the doping. Several rectangular crystals of 200 mm length and a cross section of 20x20 mm² have been produced in nearly identical quality with respect to all inspected performance parameters.

Figure 2 shows a photograph of one of these ingots confirming the clear transparency and the very symmetric shape. The luminescence yield measured at room temperature reaches a value of 17.7 phe/MeV for an integration gate of 100 ns. This value correspond to more than 92% compared to the collected yield within 1μs. Even the response at a temperature of -25°C does not indicate any slow contributions.



Figure 2. Photograph of one of the most recently grown PWO ingots (serial number 81).

Figure 3 documents the achieved radiation hardness for a sample optimized for the light yield. The distributions show the induced absorption coefficient in longitudinal and transverse direction after imposing an integral dose of 30 Gy (^{60}Co). The transverse measurement confirms as well the homogeneity of the distribution of dopants.

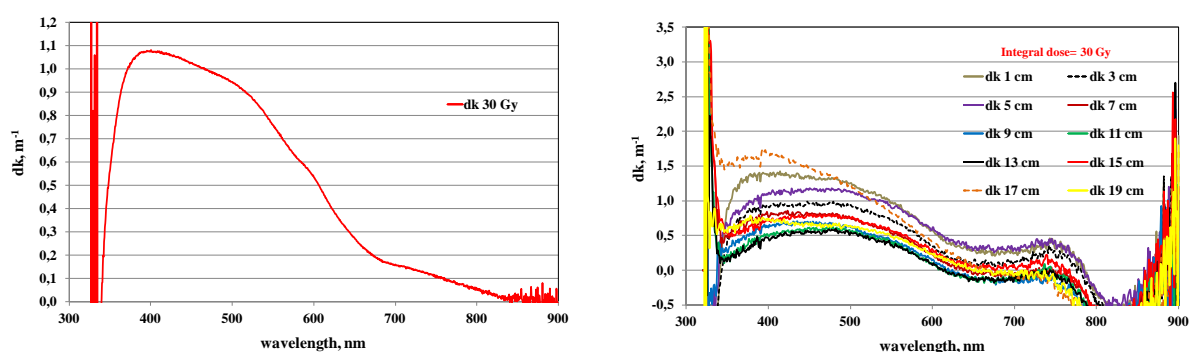


Figure 3. The longitudinal and transverse induced absorption coefficient as a function of wavelength measured at room temperature after imposing an integral dose of 30 Gy (^{60}Co). The results are determined for a $20 \times 20 \times 200 \text{ mm}^3$ full size crystal (# 79).

5. Conclusions and outlook

The development and manufacturing of PWO crystals was successfully restarted by the company CRYTUR. The obtained results confirm the first milestone to produce crystals very close to the PWO-II quality as reached by the former producer BTCP in spite of significant modifications of the technology. As a next step, the production of tapered crystals has been started as required for the barrel section of the PANDA-EMC. A pre-production of ~ 100 crystals will provide the input for any further optimization and to determine the overall conditions for mass production of the remaining 8500 crystals for PANDA.

References

- [1] E. Auffray et al., “Status of PWO crystal production for the electromagnetic calorimeter of CMS and of its construction,” Nucl. Instr. and Meth. in Phys. Res. A, vol. 537, Issues 1-2, pp. 373-378, Jan. 21, 2005.
- [2] A. Borisevich et al., “Lead tungstate crystal with increased light yield for the PANDA electromagnetic calorimeter,” Nucl. Instr. and Meth. in Phys. Res. A, vol. 537, Issues 1-2, pp. 101-104, Jan. 21, 2005.
- [3] Deeply Virtual Compton Scattering with CLAS at 6 GeV, Experimental Proposal, 2001 (http://www.jlab.org/exp_prog/proposals/01/PR01-113.pdf).
- [4] M. Battaglieri, et. al. (CLAS Collaboration), The CLAS12 Forward Tagger, Technical Design Report, 2012 (<http://www.ge.infn.it/batta/jlab/fttdr.2.0.pdf>).