Lead Tungstate Scintillation Material Development for HEP Application

E. Auffray[1] and M. Korzhik[2](✉)

[1] CERN, Geneva, Switzerland
[2] Institute for Nuclear Problems, Belarus State University, Minsk, Belarus
mikhail.korjik@cern.ch

Abstract. Here we describe the story of $PbWO_4$ scintillator material development. The role of Crystal Clear Collaboration in all stages of R&D is described. In spite of the fact that lead tungstate is widely used in high energy physics instrumentation, its development still ongoing. Resent findings show that crystal is suitable for fast timing in collider experiments.

Lead tungstate ($PbWO_4$, PWO) scintillation material technology is a result of the efforts of the large international multidisciplinary scientific community, driven by Crystal Clear Collaboration (CCC) [1, 2]. Used since more than one century, the renaissance of the systematic study of inorganic scintillation materials began with the development of the new colliders in the mid 1980s. Extensive investigations on the development of new scintillation materials were required, in parallel with CCC at CERN for LHC experiments, also under the Superconducting Super Collider program (SSC) in the USA, KEK in Japan and the UNK program (Protvino) in Russia. At that time High Energy Physics was a driving force in the development of new scintillators because of the high level of performance that was required in particle physics detectors and of the large volumes needed. CCC experts led the development of the material in collaboration with other CERN hosted collaborations, namely CMS (Compact Muon Solenoid) and ALICE (A Large Ion Collider Experiment) in the frame of the Large Hadron Collider (LHC) program [3]. This development resulted in the procurement of nearly 110 tons of Lead Tungstate scintillating crystals in a period of about 10 years.

In fact, most of the experts who were involved in PWO development and production have extensive experience with the development and the mass production of BGO scintillation crystals for the L3 experiment [4] at the CERN Large Electron Positron (LEP) collider in the eighties.

Crystal Clear was created by the active role of Dr. P. Lecoq, who was a driving expert for BGO development for S. Ting's L3 experiment at LEP and who initiated RD18 at CERN in frame of DRDC with the participation of experts from 15 countries.

First PWO samples, which showed scintillation properties, were produced by cooperation of Institute for Nuclear Problems (INP, Minsk, Belarus) and Institute of Single Crystals (ISC, Kharkov, Ukraine) [5]. First results of the beam test, which were performed at IHEP (Protvino, Russia) in collaboration with LAPP (Annecy, France),

M. Korzhik and A. Gektin (eds.), *Engineering of Scintillation Materials and Radiation Technologies*, Springer Proceedings in Physics 200,
https://doi.org/10.1007/978-3-319-68465-9_3

caused widespread interest at the CRSTAL2000 Conference in Charmonix (France) in 1992. The participants of the first working meeting at CRYSTAL2000, dedicated to PWO study, are in Fig. 1.

PWO was fond to be the heaviest single crystalline material at that time allowing construction of the high granularity compact detectors. The CMS experiment chosen PWO scintillation crystal to build homogeneous electromagnetic calorimeter (ECAL) to meet the performance criteria for the discovery of the 125 GeV Higgs boson through —its decay into two gamma. The outstanding energy resolution of a homogeneous scintillating crystal-based calorimeter is mandatory to isolate the very narrow Higgs peak at this energy above a huge background in the invariant mass distribution of the two photons. The ALICE experiment has decided to build a PWO based photon detector (PHOS) to resolve the high multiplicity events generated by heavy ions collisions.

Crystal Clear Collaboration was able to organize a multidisciplinary effort to make the best use of cross-fertilization between different categories of experts and industry to develop suitable scintillators at an industrial scale. The bridge between CERN based collaborations and the world largest producer of synthetic crystalline materials, Bogoroditsk Technical Chemical Plant (BTCP) from Bogoroditsk, Russia has been installed and worked for 15 years. Figure 2 shows leading investigators of the Workshop dedicated to production of the radiation hard crystals at BTCP in September 1999.

Fig. 1 Participants of the first working meeting at CRYSTAL2000, dedicated to PWO study. From left to right: Dr. M. Korzhik (INP BSU), Dr. L. Nagornaya (ISC), Prof. M. Issi (KEK), J.-P. Peignex (LAPP), M. Kobayashi (KEK)

Fig. 2 Leading experts of the CERN-BTCP workshop dedicated to PWO radiation hardness at the raw material production unit, September 1999. From left to right: A. Dosovitskiy (NeoChem), E. Auffray (CERN), P. Lecoq (CERN), A. Annenkov (BTCP), M. Korzhik (INP BSU)

A guideline of the CCC relationship with the crystal maker was quite different from traditional client-producer relationship, because as of the beginning it was a more effective mutual collaboration. A collective understanding of the different constraints on both sides was built to protect the long term interests of the HEP community and producer. This aspect is very important and if it takes generally several years to be fully integrated, it contributes to a large extent to the success of the operation. Such challenging projects cannot be successfully realized without satisfying the interest of both sides: a guarantee for the best physic performances on the scientific side, versus stable long term production and possibility to attract other clients on the producer side.

The step by step strategy to improve crystal properties was chosen and finally it led to the creation of the world largest crystal growth facilities allowing production of the crystals for LHC and other experiments. The search for Higgs boson imposed to build higher luminosity collider, what introduced a requirement for a short decay time of the scintillation. Due to expected large mass of the Higgs boson, equivalent 125 GeV, the light yield (LY) of the scintillation material was not a mandatory, however a minimal value of the LY non-uniformity along 25Xo long homogeneous scintillation element was crucially important for a whole set of crystals, what dictated perfect transmission of the crystal in the spectral range of scintillation.

As the first step, crystal coloration was decreased and then completely removed at the mass production. It was managed by fine adjustment of the crystal growth atmosphere and not isovalent doping of the material, allowing affective compensation of the cation vacancies in the crystal.

As a second step, slow components of scintillation kinetics were suppressed in scintillation. It was found, that slow scintillations are originated by Mo impurity presence in the crystal. Raw materials were carefully specified, their purification was organized, which allowed drastic decrease of slow component and emission of 95% of scintillation light in100 ns gate.

Finally, production technology was optimized in such a way that more than 98% of the produced crystals showed very good tolerance to electromagnetic part of ionizing radiation, which guaranteed an excellent performance of Electromagnetic Calorimeter (ECAL) and Photon Scintillation Detector (PHOS) in a harsh irradiation environment of collider experiments.

These essential tasks were resolved in 1995–1997 due to results of different CCC groups, first of all combining of the beam tests performed by CMS and ALICE and the measurements in the field of material sciences, which require very often sophisticated equipment with scheduled access spread in different parts of the world. This is the case for synchrotron radiation sources at Saclay and Hamburg, radiation facilities in Belarus, Russia, France, Germany, EPR and ENDOR in Germany and Check Republic, thermo-luminescence and elaborated spectroscopic devices in overall the Europe. CCC network made very short time needed to perform and analyze the results of the measurements. Moreover, specific organization was made, in order to reduce the feedback loop with the producer. For each problem (slow scintillation, radiation damage, coloration) experts of CCC Institutes were asked to propose a few tests to identify the parameters involved in this problem. Once these parameters were known, they were systematically investigated by the producer in order to find the best technological solution. At this stage, a multilevel level feedback loop was organized, with one, then with a few simple tests made in the vicinity of the production center to allow quick reactions, and another one with more in depth studies in specialized facilities for a full control and understanding of the process. Once a significant improvement has been made, it has to be confirmed on a statistical basis on a set of a few full size crystals in the conditions of mass production. This approach for a spread efforts reduced significantly the time needed to solve a problem. Up to now, this approach is used by CC Collaboration to develop materials for HEP applications and medical imaging as well. In a relatively short time the nomenclature of PWO crystal ingots became available (Fig. 3) to produce any types of homogeneous scintillation elements for HEP applications.

PANDA Collaboration Experiment at FAIR (GSI, Darmstadt) is the second largest lead tungstate based electromagnetic calorimeter [6] which is under construction now. Development is led by Justus Liebig University of Giessen (Germany), CCC member since the beginning of the collaboration. The physics goals of the experiment require the energy threshold ~20 meV. For this reason, crystals require very ambitious specification.

In fact, most of the requirements, except the light yield, were achieved at the PWO development in nineteen's. An extended R&D program was initiated to improve the light yield combined with operation at low temperatures, such as T = −25 °C. These efforts resulted in the development of the new generation of lead tungstate crystal technology, later called PWO-II. The production technology of the next generation of lead tungstate, was optimized to increase the light yield without degradation of the radiation hardness. For the moment PWO scintillators of PANDA specification are

Fig. 3 Different types PWO ingots which were under production at BTCP since 1999 year

produced by CRYTUR [7]. In a relatively short time CRYTUR reached the quality of PWO-II crystals for full size elements and can be a reasonable supplier of the lead tungstate scintillation elements for High Energy Experiments.

Recently, in spite the 20 years story of the PWO study, new properties of the PWO luminescence were discovered by CCC Institutes: INP BSU (Minsk) and Vilnius University (Vilnius) in cooperation with CERN [8]. This study was initiated by the coming LHC upgrade project in order to significantly increase its luminosity. Time resolution of PWO-based calorimetric detectors becomes a crucial issue for the detector exploiting at a high pile up conditions. Lead tungstate crystal, according to a classification described in [9], is a self-activated scintillator. Its luminescence band with maxim 420 nm occurs due to electronic transition in host oxy-anionic complexes WO_4^{2-} and, similar to other tungstate of scheelite structure, is temperature quenched. Due to this reason scintillation is fast and exhibits a short emission decay time. Together with regular luminescence centres, the oxygen vacancy-based defect centres WO_3, exist in the crystal and emit green luminescence with maximum near 500 nm. Use of the femtosecond laser sources for excitation and advanced streak camera for registration recently allowed to measure precisely a rise time of PWO photo-luminescence (PL) [8]. The initial part in the kinetics of the PL intensity spectrally integrated within the entire band (400–600) nm was measured. It was found that PL rise and initial decay approximately follow the shape of the instrumental function, i.e., they are instrumentally limited. This evidences that the PL rise time in PWO scintillation crystals is considerably shorter than 2 ps. Taking into account that thermalization process of free carriers after ionization has roughly the same duration, we concluded that rise time of scintillation will be at the level of a few ps.

Interesting, both regular WO_4^{2-} and defect-related WO_3 luminescence centers show the same leading edge of the luminescence build up. This is an indication that no intermediate recapturing processes are involved in the energy transfer processes. This is consistent with the low thermo-activation energy of the trap, based on WO_3 defect. At the 254 nm interband excitation, when electrons are excited from a valence band, the initial stage of decay kinetics is characterized by two decay constants: $\tau_1 = 5.9$ and $\tau_2 = 824$ ps, excitation. At the ns domain, kinetics follows to PL decay with the time constant of 6–10 ns, which is observed in routine start–stop type measurements of PWO luminescence kinetics.

New results on PWO luminescence are obtained under photo-excitation when most of the pairs of the carriers remain geminate. Under ionizing radiation, the pairs are disconnected during the thermalization process. Nevertheless, a dramatic increase in the rise time of the scintillation pulse at a high energy deposit due to ionization is not expected. The high energy deposit in the material of inorganic scintillator is typical in the high-energy physics experiments, like in LHC experiments. At the registration of particles with energy more than 100 GeV it exceeds several GeV within a time of less than 1 ns. So the density of free carriers, even within a few ps, becomes high enough to provide prompt coupling of opposite carriers for recombination. For this reason, we do not expect a substantial increase of the scintillation rise time compared to photoluminescence rise time and estimate it at the level of less than few ps.

Recent results show that the fast rise of luminescence in PWO scintillators is short enough to reach sub-50-picosecond readout, which is targeted for the future scintillator detectors at High Luminosity LHC and Future Circular Colliders.

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