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Ten years of lead tungstate development

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Abstract

The CMS Collaboration at CERN has undertaken at the end of 1994, an ambitious R&D program on lead tungstate scintillating crystals for its electromagnetic calorimeter. All the parameters of this crystal have been extensively studied in order to optimize its performances in the context of the large hadron collider. Full-size crystals (23 cm in length, up to $2.5 \times 2.5\,\text{cm}^2$ in section) with the desired trapezoidal geometry can now be grown and mechanically processed with a yield in excess of 80%. A thorough investigation of the raw material preparation and of the growing conditions has led to a significant improvement in the optical transparency and in the light yield of the crystals. A detailed understanding of the light emission mechanism was established in 1995. A systematic analysis of the parameters influencing the structural quality of the crystals was at the origin of a considerable improvement of the radiation hardness of full-size crystals. All these progress has led several other experiments to select lead tungstate as the best scintillator for their detector. The conditions of these improvements will be discussed in the context of the mass production of nearly 80,000 crystals in a cost-effective way.

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1. Introduction

The development of a new scintillator with tight specifications for light yield, decay time and radiation damage, poses challenging problems to be solved by experts working in different fields of material science. This requires a multidisciplinary approach, with a good coordination of efforts and a well-organized support. If, in addition, a large production of several cubic meters has to be made in a few years only, additional problems have to be solved, related to production infrastructure, reproducibility of parameters, production yields and cost effectiveness.

In the last two decades, a new generation of high-energy physics (HEP) experiments has become a driving force for the development of new

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scintillators. This has started with bismuth germanate (BGO) for the L3 experiment at CERN [2] (Fig. 1) and caesium iodide (CsI either thallium doped or pure) for CleoII at Cornell [3], Crystal Barrel [4], KTeV at FermiLab (USA) [5], Belle at KEK (Japan) [6] and BaBar at SLAC (USA) [7], which were already known but could only be produced in small sizes and small quantities only. It became even more evident with barium fluoride (BaF_2) for TAPS [8] and GEM [9], cerium fluoride (CeF_3) as a candidate for CMS [10] and L3P [11] and finally lead tungstate ($PbWO_4$) for CMS at CERN[10], which were essentially developed for

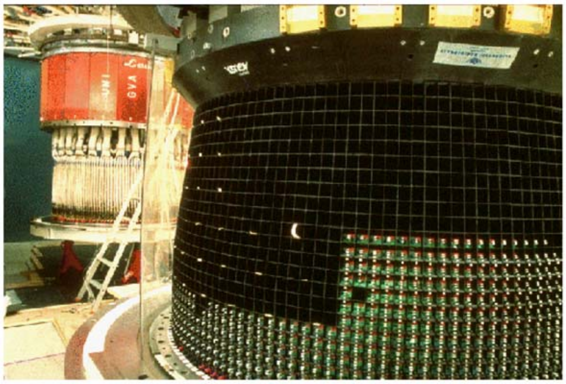

Fig. 1. Construction of the L3 BGO detector at CERN.

particle physics experiments. The difficult physics constraints and harsh experimental conditions impose very tight specifications to modern detectors. The size of the experiments and the high quantitative demand allowed to organize the R&D effort and production on a large scale (Table 1). This has been particularly illustrated by the work of the Crystal Clear Collaboration [1] which was able to create 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. At this level, the possibility to make use of the large production infrastructure installed during the cold war in former Soviet Union has been a key to the success. This has motivated our collaboration with ISTC [12] for the conversion of the Bogoroditsk Techno-Chemical Plant in Russia for the production of the 80,000 lead tungstate crystals (11 m^3, 90 tons) of the CMS experiment at CERN.

2. The conditions of scintillator development for HEP

The context of scintillator development for HEP is rather difficult. The market cannot be stabilized because of the rapidly evolving demand at each generation of the experiment. The very large size of the projects imposes a strong effort of develop-

Table 1
Crystal calorimeters in the world

	Crystal Ball	CLEO II	L3	BaBar	Belle	L∗ Lol	CMS Eol	GEM Eol	L3P Eol	ALICE	CMS
Where	SPEAR	CESR	LEP	SLAC	KEK	SSC	SSC	LHC	LHC	LHC	LHC
When	1972	Late 1980's		1999						2007	
Beam	e^+e^-	e^+e^-	e^+e^-	e^+e^-	e^+e^-	pp	pp	pp	pp	ion ion	pp
Beam energy	4	6	100	9 + 3.1 GeV	8 + 3.5	20	20	8	8	5.5 A TeV	7
Crystal	NaI(TI)	CsI(TI)	BGO	CsI(TI)	CsI(TI)	BaF_2	BaF_2	CeF_3	CeF_3	$PbWO_4$	$PbWO_4$
Number (k)	0.7	7.8	11.4	6.8	8.8	26	15	45	100	18	77
Length (X_0)	16	16	21.5	16	16	24.5	24.5	25	25	22	25
Photodetector	PMT	Si PD	Si PD	Si PD	Si PD	V4T	V4T	Si PD	VPD	Si PD	APD/V3T
$B[T]$	0	1.5	0.5	1	1	0.75	0.8	4	1	$\leqslant 0.5$	4
f_{BC} (MHz)	1.3	2.8	0.091	238	10–508	60	60	67	67	8	40
				New! Asymmetric b-factory		Proposals				New! Large Hadron Collider	

ment and production in a relatively short period of time. Unfortunately, the benefit of these efforts is very often lost if the next experiment requires another scintillator with improved performances. For a long time NaI(Tl) was the only candidate because the most important parameter was a high light yield to be able to read out the signals from low-energy particles with standard electronics. Then, the increasing size of the experiments and the necessity of having a good granularity of the detectors enabled research on higher density materials. That was the era of BGO with a very high density of 7.13 but rather moderate light yield and CsI, with a smaller density of 4.51 but much higher light yield, which seems to have been a good compromise, as it is the only scintillator to have been used in at least 5 large size detectors so far. Unfortunately, the hunt for very rare events imposed to build accelerators of higher luminosity, puts new constraints for short decay time of the scintillators. The requirements for high density have been further increased, whereas the one for high light yields has been somehow reduced because of the increased energy of incoming particles and the emergence of a new type of photodetectors like avalanche photodiodes. These new requirements triggered a strong R&D effort on BaF_2, CeF_3 and $PbWO_4$.

Another difficulty for this activity is the complexity of the decision mechanisms in HEP. As new technologies are needed for every new generation of experiment, an important R&D effort has to be made for a proof of feasibility and a good understanding of cost issues, before any approval can be made. This takes usually several years during which no firm commitments can be made and some conditions can change. At least two bad experiences were seen in the recent past:

- the large effort of several years for the development of large-size radiation-hard blocks of BaF_2 crystals was suddenly stopped by the decision to stop the SSC program in the US.
- Similarly, the spectacular developments of avalanche photodiodes has led the CMS Collaboration to finally prefer lead tungstate crystals with a lower light yield but higher density, to the well-developed Cerium Fluoride CeF_3, in order to build a more compact and less expensive detector.

The uncertainty of future markets for these new scintillators is another problem for the crystal producers. At least in the first phases of the development the prospects for other applications than physics experiments are not well known. If the situation has been rather good for BGO in the scientific, industrial and medical domains, reasonably good for CsI with several physics experiments and some commercial applications, it is still very unclear for BaF_2, CeF_3 and $PbWO_4$.

On top of these difficulties, the more and more severe budgetary constraints impose strong limits on the production costs of the scintillators which are only partially compensated by the financial support during the R&D phase.

Keeping in mind all these difficulties, a proper strategy has been set up for the development and the production of the large quantity of lead tungstate crystals of the CMS electromagnetic calorimeter.

3. Strategy for the CMS calorimeter

3.1. General considerations

The first and probably most important action in the beginning of such a challenging project is to clearly define the objectives. This includes a strict definition of a list of realistic specifications to be reached by the crystal, in order to guarantee the physics performance of the detector without over-designing. The understanding of the cost driving factors and the study of the methods to reduce their impact on the final cost has to be included in the R&D program at the same level as the fight for improved technical parameters. Finally, the preparation of the production infrastructure must be included in the overall program with a detailed analysis of all the production aspects: procurement of raw materials, equipment, manpower and safety. In the case of CMS a program in 3 phases has been set up with 3 years R&D, 2 years pre-production and more than 5 years production periods.

A proper funding must be defined for each phase, in full agreement with the crystal producers. It is important that the losses are minimized in the case of a modification or even a stop at any stage of the program. For the first time in the history of HEP, CMS has organized a well-defined support during the R&D and the pre-production phase funded by CERN with the help of the International Science and Technology Center (ISTC). This long-term effort associated to a non-negligible risk must be shared with well-selected industrial partners. The possibility to make use of the large production infrastructure installed during the cold war for the growth of non-linear crystals for military applications has played an important role in the selection of the Bogoroditsk Techno-Chemical Plant (BTCP).

The traditional client–producer relationship must be replaced by a more effective spirit of collaboration. A mutual understanding of the different constraints on both sides has to be built in the necessary respect of a certain level of confidentiality to protect the long-term interests of the producers. This sociological 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 solving the difficult equation of maximization of happiness on both sides: best performances for lowest cost on the client side, versus best profit and possibility to attract other clients on the producer side.

3.2. Organization of the R&D

An important characteristic in the field of material science is that it requires a multidisciplinary approach. The users (in our case high-energy physicists) define a set of desired performances which determine the goal to be reached. The crystal producers bring the technology and their experience in organizing mass production with maximum yield and optimized cost. A group of experts are also needed in different fields like solid state physics, spectroscopy, chemistry, trace element analysis, to help producers to reach the specifications set by the users. Some of the required expertise may exist in the production centers, but in most of the cases one has to open the collaboration to outside laboratories. One difficulty is to select these groups not only for their expertise but also for their ability to understand the specific spirit of their contribution. They have on one hand to understand the user's requirements, and on the other hand to help solving problems in an industrial context and not only for their academic interest. This is a long-term work, and the experience gained in previous large projects as well as R&D efforts in the frame of officially supported groups like the Crystal Clear Collaboration at CERN [1] are playing a crucial role in organizing these contacts.

Another problem comes from the difficulty of the measurements in the field of material sciences, which very often requires heavy equipment with scheduled access spread in different parts of the world. This is the case for synchrotron radiation sources, radiation facilities, EPR and ESR systems and, to a lesser extent, for Thermo-luminescence and elaborated spectroscopic devices. The time needed to perform and analyze the results of the experiments are long. This is why a specific organization had to be made in order to reduce the feedback loop with the producer. For each problem (radiation hardness), or quantity to be improved (light yield), experts are asked to propose a few tests to identify the parameters involved in this problem. Once these parameters are known, they are systematically scanned by the producer in order to find the best optimization. At this stage, a two-level feedback loop is organized, one 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 laboratories for a full control and understanding of the process. Once a significant improvement seems to have been made, it has to be confirmed on a statistical basis on a set of at least 10 full-size crystals in the conditions of mass production. This approach reduces as much as possible the time needed to solve a problem. However, one has to count about one year for each important step in the development of a new material. This is the time it took the CMS Collaboration to grow crystals of the required dimensions in 94, to suppress slow components at the end of 95, and more recently to make significant progress in radiation hardness of lead tungstate crystals [13].

3.3. Cost optimization

One important aspect of these developments is the cost optimization. All the R&D effort must be driven by cost considerations. It is not sufficient to solve a problem with non-affordable solutions. This is why the R&D as well as the production strategy are developed as a function of the existing infrastructure in the production centers. It is better to suppress one or two metals in 4N raw materials than to have to buy 5N oxides. Optimizing the orientation of the crystal is certainly cheaper than developing specific machines for cutting fragile crystals. The maximization of the yields at each stage of the production is one of the key objectives of the R&D.

As potential future markets are uncertain, the production infrastructure has to be organized as much as possible with R&D funds in order to not impinge too much on the production cost of the crystals. This is also the role of the R&D in developing production technologies that are as simple as possible, minimizing the power consumption, and with a high degree of automatization in order to reduce manpower costs.

4. Progress on lead tungstate

This systematic approach has been followed for the development of lead tungstate crystals for the CMS experiment at CERN.

The very specific requirements of the scintillating crystals for the Electromagnetic Calorimeter at the CERN Large Hadron Collider CMS experiment have been the subject of intensive research and development over the last ten years. At the start of these studies it was by no means clear that the very high purity of raw material or the special and harsh requirements regarding the radiation hardness of these crystals could be met at all. None of the most experienced manufacturers in the field was at that time anywhere close to being able to deliver the quality of crystals needed. An intensive long-term R&D effort was therefore undertaken by a scientific research consortium including the international CRYSTAL CLEAR Collaboration [1], members of the CERN-CMS experiment, the Minsk Institute of Nuclear Problems and the Bogoroditsk Techno-chemical Plant. It operated under the umbrella and with the active help of the International Science and Technology Center (ISTC) and with generous financial support from the European Union as one of the major funding Parties to the ISTC Programs.

Important progress have been obtained and is illustrated in Fig. 2. The first objective was to grow ingots large enough to extract crystals with the required dimensions: 23 cm in length, $2.2 \times 2.2\,cm^2$ front section and $2.5 \times 2.5\,cm^2$ back section. It took several months to obtain crystals of these dimensions with a good mechanical stability and therefore a high cutting and polishing yield.

A second problem was the suppression of slow components in the light emission of $PbWO_4$. It was observed in 1995 that as a consequence of the optimization for a higher light yield slow components at a few percent level in the crystals were produced. A thorough investigation was made, which led to the conclusion that a combination of molybdenum contamination and bad annealing conditions could introduce such components.

A very critical parameter for lead tungstate crystals is their ability to survive in the high-radiation environment of the Large Hadron Collider (LHC) in the CMS experiment, with the

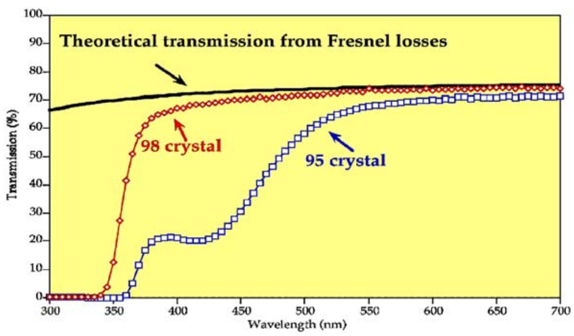

Fig. 2. Progress on optical transparency of crystals.

minimum variations of collected light as a function of time. This problem is difficult to solve, as it implies a perfect modelization of the radiation conditions in the LHC machine, and a very good understanding of the chemistry of defects in this crystal. All these aspects have been systematically investigated and impressive progress has been made.

The very promising results of the first phase of the R&D program (1996–1998) induced the collaborating Institutes to continue the ISTC program and to further develop the necessary technologies, including the implementation of stringent quality control methods and special automated measuring equipment. This second R&D phase, financially supported on a 50/50 basis by the European Union and CERN-CMS, has led to excellent results and set the grounds for the mass production phase, in which the quality of the mass production technologies is being demonstrated on a large scale. This is also the opportunity to bridge to the commercial market. It consists in developing a "western like" commercial culture in the company. This has several aspects : reliability of production, training of staff, excellent managerial structure, quality insurance policy, installation of a modern communication system, development of a network of commercial contacts.

We are now in the last phase of this program. In order to fulfill the very tight schedule of the Large Hadron Collider project, a total of 77,000 crystals, 23 cm long, representing a volume of 11 m^3 and a mass of 90 tons, has to be completed before the start of LHC. In spite of the fact that more than 150 Czochralski ovens are involved in this production, a large effort had to be developed to increase the productivity in order to reach this goal. The main action has been to progressively increase the diameter of the ingots from 38 to 65 mm and then to 85 mm in order to be able to extract not only one but two and up to four crystals per ingot, respectively. The production rate which is presently of 1000 crystals per month could then progressively increase to 1500 per month with 65 mm ingots and 2200 crystals per month with 85 mm ingots (Fig. 3).

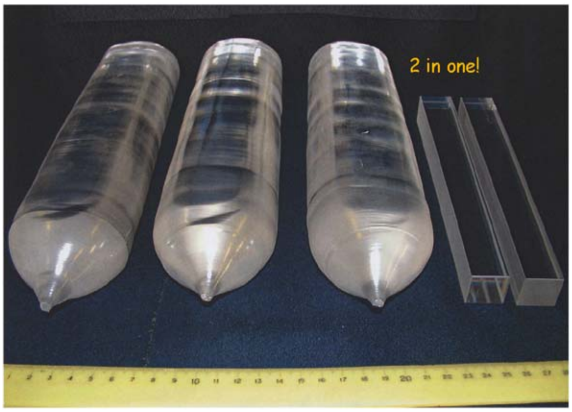

Fig. 3. 65 mm ingots with 2 CMS barrel crystals cut per ingot.

5. Other experiments using lead tungstate

The large and successful effort invested by CMS on the development of lead tungstate crystals has led several other experiments to choose this crystal for their detector.

The ALICE experiment at CERN is a dedicated heavy-ion experiment at LHC for the study of the initial phase of the collision of heavy nuclei via the direct production of single photons and di-photons. It will also look for signals of chiral-symetry restoration and jet-quenching as a probe of deconfinement.

The ALICE PHOS calorimeter consists of 17920 PWO crystals $22 \times 22 \times 180\,mm^3$ organized in 5 modules of 3584 crystals each. Special production facilities have been installed in Apatity, Russia, for the procurement of these crystals grown by Czochralski method. The detector will be operated at –25 °C and read out with avalanche photodiodes [14].

The BTeV experiment at FermiLab is a fixed target experiment to study quark flavor physics, in particular, the rare decays of b-flavored particles as the source of CP violation. About 10,000 slightly tapered crystals, with dimensions $(27–28) \times (27–28) \times 220\,mm^3$ will be assembled in a wall perpendicular to the beam axis. The potential production sites are Bogoroditsk, Apatity, Shanghai and Beijing. The production is expected to take place in the 2006–2008 period [15].

MECO will be installed on the AGS at Brookaven National Laboratory. It is a highsensitivity experiment (2×10^{-17}) which will address rare symmetry violating process by looking at muons converting to electrons in the field of a nucleus. About 2300 PWO crystals, $30 \times 30 \times 120\,mm^3$ will be used for this experiment [16].

The PrimEx experiment at Jefferson Laboratory will use a wall of 1200 PWO crystals $20.5 \times 20.5 \times 180\,mm^3$ readout by PMT for a precision measurement of the π° lifetime via the Primakov effect [17].

The Photon Ball is to be installed into the ANKE magnetic spectrometer at COSY, Jülich. It will study the nucleon structure via the direct measurement of neutral mesons. It is a compact hermetic ball of 876–1100 tapered PWO blocks of 120 mm length readout by 15 mm quartz light guide and fine mesh Hamamatsu 5505 PMT [18].

The PANDA experiment is a multipurpose detector for the antiproton storage ring at GSI. It will study charmonium, glueball, strangeness and hypernuclei spectroscopy. The favoured technical option so far is based on 7200 PWO crystals, $35 \times 35 \times 150\,mm^3$ readout by avalanche photodiodes.

6. Conclusions

Because of the high level of performance required in particle physics detectors and the large volumes needed, HEP is a driving force in the development of new scintillators. Through the example of the CMS electromagnetic calorimeter being built at CERN in the frame of its Large Hadron Collider program, this paper describes the strategies developed for the R&D and the procurement of nearly 100 tons of lead tungstate scintillating crystals in a period of about 10 years. This project is now well under way as about one-third of the crystals have been produced so far and the detector is in its assembly phase.

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References

[1] R&D proposal for the study of new fast and radiation hard scintillators for calorimetry at LHC: Crystal Clear Collaboration, CERN/DRDC P27/91-15, project RD-18.
[2] L3 Collab., Nucl. Instr. and Meth. A 289 (1990) 35.
[3] Y. Kubota, et al., The CleoII detector, CLNS 91/1122, 1991.
[4] The Crystal Barrel Collaboration.
[5] R.E. Ray, The KTeV pure CsI calorimeter, Proceedings Of Fifth International Conference on Calorimetry in High-Energy Physics, World Scientific, NJ, September 1994, p. 110.
[6] The Belle Collaboration, Technical design report, KEK Report 95-1, April 95.
[7] The BaBar Collaboration, BaBar Technical Design Report, SLAC-R-95-457.
[8] R. Novotny, et al., Nucl. Instr. and Meth. A 262 (1987) 340.
[9] GEM Letter of Intent, SSCL SR-1184, November 1991.
[10] CMS Technical Proposal, CERN/LHCC 94-38, December 1994.
[11] L3P Letter of Intent, CERN/LHCC 92-5, 1992.
[12] ISTC: International Science and Technology Center.
[13] A. Annenkov, et al., Nucl. Instr. and Meth. A 426 (1999) 486.
[14] ALICE Collaboration Technical Proposal, CERN/LHCC/95-71, 1995.
[15] The BteV proposal, March 2002, BteV-doc-316.
[16] MECO Collaboration, A proposal to the National Science Foundation to construct the MECO and KORPIO experiments, 1999, http://meco.ps.uci.edu/.
[17] A precision measurement of the neutral pion lifetime via the Primakof effect, PrimEx Conceptual Design Report, Jefferson Lab, March 3, 2000.
[18] S. Barsov, et al., Nucl. Instr. and Meth. A 462 (2001) 364.