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Advanced scintillation single crystals based on complex oxides with large atomic number

S.F. Burachas, L.L. Nagornaya, G.M. Onishchenko, L.A. Piven', E.N. Pirogov, V.D. Ryzhikov
STC for Radiation Instruments, Concern "Institute for Single Crystals", 310001, Kharkov, Ukraine.

Abstract. An improved production technology has been developed for scintillation single crystals based on complex oxides with large atomic number – bismuth germanate (BGO), gadolinium silicate (GSO), cadmium tungstate (CWO) and lead tungstate (PWO). Scintillators based on these crystals have good energy resolution and light output, high detection efficiency, they are not hygroscopic, have high radiation stability and mechanical strength. This makes it possible to use them as radiation detectors for high energy physics (PWO), in instruments for radiation and radioecological monitoring (BGO,CWO,GSO).

Keywords: crystal, technology, scintillator, instrument for radiation monitoring.

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An improved production technology has been developed for scintillation single crystals based on complex oxides with large atomic number – bismuth germanate (BGO), gadolinium silicate (GSO), cadmium tungstate (CWO) and lead tungstate (PWO). Scintillators based on these crystals have good energy resolution and light output, high detection efficiency, they are not hygroscopic, have high radiation stability and mechanical strength. This makes it possible to use them as radiation detectors for high energy physics (it covcern to PWO), in instruments for radiation and radioecological monitoring (BGO, CWO, GSO). The attainted results were published partly in the review paper [1] and articles [2-8]). Main parameters of these crystals are presented in the Table.

Experimental and theoretical studies were carried out and technological equipment and accessories based on them were improved, technological processes of preparation and processing of large-sized crystals were developed for these oxide crystals.

Mathematical modelling of thermal conditions and growth processes of oxide crystals with constant radii by Czochralski method have been carried out. The model accounts for thermal physical, hydrodynamic and physical parameters and properties of the system "crystallization unit with induction heating – melt – crystal". The mathematical model developed allowed to determine the nesessary technological parameters and regimes of growth, as well as design principles of the crystallization unit [1].

A choice and substantiation of physico-chemical methods for growing large CWO crystals were made with an account of their required quality [4, 6]. Physico-chemical studies of the system of crystal-forming oxides Gd_2O_3 - SiO_2 allowed to optimize the requirements for the incoming control of the oxides and to develop a technology of preparation and synthesis of the Gd_2SiO_5:Ce charge [6]. Analogous investigations of other crystals were carried out too. A possible concentration limit for the content of harmful impurities in this oxide crystals raw powder was established. With an account of the obtained data an additional purification of raw material (cadmium and tungsten oxides) was performed. Optimal technological processes for preparation of raw powder were developed [4].

Technology for rapid melting-on of the charge in the form of granules, pellets and tablets has also been developed, as well as a new method for melting-on of the charge, which allowed to prolong the specified life of iridium and platinum crucibles.

An optimized choice of the crystal seed orientation has been realized. Conditions and regimes of technological stages of seeding and beginning of growth of the crystal upper cone have been worked out enabling to suppress the formation of unit structure.

An improved design of the crystallizer based on iridium and platinum crucibles has been developed, which ensured the required initial overheating of the melt and allows to pull crystals up to 50 mm in diameter without

Table. Parameters of oxide scintillation single crystals developed at the STC RI

Type of crystal	Maximum size, mm	Light yield with respect to NaI(Tl), %	Energy resolution (^{137}Cs, ∅40×40mm), %	Price for 1 cm^3, US $
BGO	∅ 75×150 ∅ 55×230	12-14	10.5-13	10-17
CWO	∅ 60×150	33-40	8-10	9-20
GSO	∅ 55×180	20-25	9-11 28-30 (60 keV)	30-60
PWO	22×22×200	11-14 photoelecrons/MeV	less than 0.3% for particles of 50 GeV energy	2,5-4,5

macrodefects and with the minimum Tyndall scattering. A new crystallization unit, accounting for previous developments, has been created, with a possibility of its controlled movement along the vertical axis and azimuth, ensuring reliable visual control and optimum configuration of the thermal field during seeding and growth, as well as better uniformity of the thermal field during post-growth annealing and cooling of GSO crystals. For growth of these crystals we chose a computer-controlled industrial growth installation of the induction type "Crystal-3M", and the required technological equipment was developed. Technological conditions and regimes were determined for growth of GSO crystals of required dimensions, and the final growth stage – "freezing" of the crystal – has been developed.

We have also established the temperature range of thermochemical stability of PWO melt, in which crystals of high structural perfection could be obtained. The optimization of the growth conditions and the thermal treatment once allowed us to fabricate highly homogeneous and transparent PWO scintillation crystals with fast initial luminescence decay and minimized slow component [5, 7].

New methods of a thermal treatment of the crystal boules for scintillators in different surroundings and scintillators fabrication for scintillation units using a novel technique combining mechanical and thermal treatment have been developed. Corresponding modifications of technological equipment has been made.

This allows to obtain large oxide crystals of high spectrometric quality with the absorption coefficient in the region of the intrinsic luminescence 0.01-0.05 cm^{-1}. The technological yield of crystals is rather high.

A method for measuring scintillation and background characteristics of these crystals was elaborated. Except scintillation characteristics optical, spectral-luminescent, kinetic ones and radiation stability of this oxide crystals were investigated [2, 4, 5]. On the base of studying these characteristics we have developed some kinds of detectors and instruments for radiation monitoring.

Some batches of detectors BGO and PWO of our fabrication for high energy physics projects CMS and ALICE were supplied to CERN and PSI (Switzerland) and tested with good results. Some thousands of BGO, CWO, GSO scintillators were supplied to several firms of Netherlands, Germany, France, Italy, Japan, USA, Czech Republic, Russia.

On the basis of these crystals scintillators have been developed which found broad application in high energy physics, tomography, spectrometry and radiometry. The unique combination of their properties – high atomic number and density, good energy resolution, high light yield, conditions for K-resonance, non-hygroscopicity - allowed to create a number of new radiation control instruments with improved sensitivity, detection efficiency and reliability.

New instrument, alpha-gamma radiometer-spectrometer RK-AG-02, comprising a compact detection unit (DU) based on GSO scintillator and a data processing unit (DPU) using a microcomputer is designed for measurements of activity of Am-241 and other transuranic radionuclides (TUR) [8]. It can be used as a part of a radioecological laboratory for radioecological express sample control of soil, agricultural products, etc. under real radiation conditions.

The alpha-gamma radiometer is based on the principle of detection of Am-241 gamma-radiation (E_g = 59.5 keV) and alpha-radiation of TUR. The detection unit, due to the K-resonance on Gd atoms of the GSO scintillator, ensures high efficiency of gamma-radiation detection, as well as highly efficient detection of alpha-radiation. The measurement modes (gamma or alpha) are realized using plug-in units attached to the DU from the side of the GSO scintillator, formation of "windows" – measurement channels – in the spectrometric amplifier of the DU electronic sub-unit, and the DPU software.

The experimental instrument was tested under real radiation conditions in the Chernobyl explosion zone together with ORTEC and LP-4900B type spectrometers.

In comparison with them, our spectrometer displayed better technical characteristics, being at the same time less expensive.

With our instrument, measurement errors of Am-241 and TUR activities did not exceed 20%. The required duration of measurement was 10-100 times shorter with the same number of pulses.

Another new developments of STC RI are multi-purpose spectrometric scintillation detection units BDEG-40T for gamma-radiation, which are based on 40x40 mm^2 BGO (or CWO) scintillators and FEU-176 type PMT. They are equipped with a built-in spectrometric amplifier and high voltage transformer. These instruments are very efficient as parts of radiometric and spectrometric equipment of radiological laboratories. Its amplitude resolution is 11-13 (9-11)%, efficiency of registration for ^{137}Cs more then 2-3 times exceeds analogous values for alkalihalide scintillation crystals.

The basic technical decisions of crystal BGO, CWO, GSO, PWO production technology and instruments of radiation monitoring have been protected by more then 30 patents. Industrial production of the above-described instruments is being launched by our Center, and they can be supplied to possible customers.

References

1. S.F. Burachas, L.I. Gerasimchuk, E.N. Pirogov et al., Physico-chemical principles of germanoeulitine single crystals synthesis and growth (Review article) // *Functional materials,* V. 3, № 3, pp. 373-388, (1996).
2. S.F. Burachas, V.G. Bondar', K.A. Katrunov et al., Growth and optical perfection of gadolinium silicate and cadmium tungstate crystal scintillators // *Inorganic materials*, V. 32, № 12, pp. 1525-1527, (1996).
3. S.F. Burachas, H.H. Gutbrod, V.D. Ryzhikov, V.I. Man'ko et al., Peculiarities of grown $PbWO_4$ scintillator crystals for application in high-energy physics // *Journal of crystal growth*, V. 186, pp. 175-180, (1998).
4. L.L. Nagornaya, S.F Burachas, Yu. Ya. Vostretsov, V.D. Ryzhikov, Studies of ways to reduce defects in $CdWO_4$ // *Journal of crystal growth*, V. 198/199, pp. 877-880, (1999).
5. S. Burachas, L. Nagornaya, H. Gutbrod, V. Ryzhikov et al., Lead tungstate $PbWO_4$ crystals for high energy physics // *Journal of crystal growth*, V. 198/199, pp. 881-884, (1999).
6. S. Burachas, V. Bondar', V. Krivoshein et al., Synthesis and melting of raw charge for oxide crystal growth by Czochralsky method, *Abstracts of third International Conference "Single crystal growth, strength problems, and heat mass transfer"*, Obninsk, Russia, September 21-24, 1999, p. 83.
7. S. Burachas, V. Bondar', B. Grinev et al., Lead tungstate scintillation crystals optimized for high-energy physics applications, *Abstracts of fifth International Conference on Inorganic Scintillators and their Applications*, Moscow, August 16-20, 1999, p.134.
8. Ryzhikov V.D., Burachas S.F., Pirogov E.N. et al, Advanced alpha-gamma radiometer-spectrometer based on GSO scintillator and handheld PC, *Abstracts of Nuclear Science Symposium-99*, October 14-16, 1999, Seattle, USA.