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Research activity with different types of scintillation materials

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Abstract. Nowadays there is a growing interest and demand in the development of new types of scintillation materials for experimental high energy physics. Future detector developments will focus on cheap, fast, and radiation hard materials, especially for application in collider experiments. The most recent results obtained by the Giessen group in close cooperation with colleagues from different institutes will be presented. The new start of the mass production of high quality lead tungstate crystals (PbWO₄, PWO) for electromagnetic calorimetry was started by the company CRYTUR (Turnov, Czech Republic). We will present a detailed progress report on the research program of lead tungstate performed in the last two years. The latest results in the development of LuAG:Ce, YAG:Ce and LYSO:Ce inorganic fibers, grown by the micro pulling down method and cut with the heated wire technique as well as new glass ceramics material BaO*2SiO₂ (DSB) doped by Ce and Gd will be presented. In addition, different samples of the organic plastic scintillator EJ-260 produced by the company Eljen Technology (Sweetwater, USA) have been characterized. The study has focused on the change of performance after irradiation with 150 MeV protons up to an integral fluence of 5 10¹³ protons/cm² as well as with a strong 60 Co gamma-source accumulating an integral dose of 100 Gy.

1. A new production of PbWO₄ crystals

The lead tungstate (PbWO₄, PWO) scintillation crystal is one of the most widely used scintillation materials in electromagnetic calorimetry in modern accelerator experiments [1]. It combines a unique set of physical and scintillation properties, which allow the construction of an affordable compact electromagnetic calorimeters (EMC) of high quality. 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 [2] due to the high photon energies to be expected. The PANDA EMC will be based on an improved quality (PWO-II) for increased light yield and radiation hardness [1]. The operating temperature of the PANDA EMC will be -25° C that allows to increase the light yield by a factor of four by reducing the quenching of luminescence but blocks the statistical recovery of radiation damage during operation. Crystals for CMS and partly for the PANDA calorimeter were produced by the Czochralski method at Bogoroditsk Technological Chemical Plant (BTCP, Bogoroditsk, Russia) until its shut down some years ago. 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 in 2014 a research program to develop the technology for

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mass production of PWO-II crystals in close collaboration with Giessen University (Germany) and RINP (Minsk, Belarus). The test results of one of the first PWO crystals with dimensions of $2 \times 2 \times 20$ cm³ are presented on Figs. 1 and 2. The longitudinal optical transmittance was measured using a Varian Cary 4000 spectrophotometer. Transversal optical transmittance was measured with a Hitachi 3200 spectrophotometer. The radiation hardness of the sample can be characterized by the radiation induced coefficient which is defined as $dk = Ln[T_b/T_a]/d$, where T_b and T_a are the optical transmittances before and after irradiation, correspondently, and d is the thickness of the PWO sample. The irradiation was performed at the Radiation Center of Justus-Liebig-University (Giessen, Germany) with a set of 60 Co γ -sources. The dose of 30 Gy corresponds to the value, which will be reached in the most forward part of the PANDA EMC after half year of the operating time. The light yield was measured using a calibrated Hamamatsu R2059-01 photomultiplier and a 137 Cs γ -source. The obtained results confirm already now that the technological approach of CRYTUR will allow to produce lead tungstate crystals with properties very close to the PWO-II specifications of the PANDA experiment at FAIR (Darmstadt, Germany). As a next step, a pre-production of crystals in PANDA geometry will be started and characterized using a set of additional and technically optimized furnaces.







Figure 2. The transversal radiation induced coefficient measured at different positions of the 20 cm long crystal (left figure) compared to the longitudinal coefficient (right figure) after an integral γ -dose of 30 Gy.

2. Inorganic fibers made of LuAG:Ce, YAG:Ce and LYSO:Ce

Compared to conventional organic plastic scintillator fibers, inorganic fibers provide a series of advantages, like a significantly higher light yield, a more effective interaction with electromagnetic probes and a higher resistance to radiation damage. Due to these advantages and due to the high granularity provided by the fiber geometry, there are a lot of applications for these fibers in high

energy physics experiments as well as in medical applications. Especially the implementation of new concepts in sampling calorimeters like dual readout calorimetry and the use in PET scanners are under investigation.

2.1. Fibers grown by the micro pulling down method

During the last years a R&D program for the optimization and characterization of fibers grown by the micro pulling down method has been performed. The crystal melt is heated up in a crucible with a radio frequency heater and the fiber is pulled down on a seed through a small opening in the bottom of the crucible. The quality of the fibers produced by Fibercryst (Lyon, France) could be increased significantly by an optimization of the pulling speed, the temperature gradient and the Cerium concentration and distribution within the fiber [3]. Fig. 3 shows the improvement of the attenuation coefficient of the scintillation light in typical fibers of 1-2 mm diameter, starting at approximately 1.0 cm⁻¹ in 2009 down to 0.02 cm^{-1} in 2014 [3,4,5].



Figure 3. Improvement of the attenuation coefficient of LuAG:Ce fibers grown by the micro pulling down method over the last years. The green line shows the attenuation value measured for a rectangular rod cut from a large crystal and polished on all side faces [3].

2.2. Fibers cut with the heated wire method

As an alternative to fibers grown by the micro pulling down method, fibers cut by the heated wire technique from a conventional crystal, produced by the company CRYTUR (Turnov, Czech Republic) are currently under investigation. Fig. 4 shows several $1.0 \times 1.0 \times 100 \text{ mm}^3$ large rectangular LuAG:Ce fibers.



Figure 4. Picture of three $1.0 \times 1.0 \times 100 \text{ mm}^3$ large rectangular LuAG:Ce fibers cut by the heated wire technique.

The position dependent light yield of these fibers, measured by a SiPM with a cell size of 50 μ m is shown in the left part of figure 5, while the right part of the figure shows a comparison to fibers grown by the micro pulling down method. The fibers show typical attenuation coefficients between 0.03cm⁻¹ and 0.06 cm⁻¹ which is comparable to typical Ø 2 mm fibers and even slightly better than the values of Ø 1 mm fibers grown by the micro pulling down method. The detected light yield, measured with α -particles of an ²⁴¹Am source, shows comparable values for both fiber types.

As an application of the rectangular fibers, a first sampling calorimeter prototype, consisting of four modules with dimensions of $13 \times 13 \times 100 \text{ mm}^3$ has been tested with a tagged photon beam in the energy range from 50 MeV up to 800 MeV in the A2 hall of the MAMI accelerator in Mainz. The single modules consisted of a tungsten array with $1.0 \times 1.0 \times 100 \text{ mm}^3$ large rectangular LuAG:Ce fibers inserted in the holes between the 1 mm thick tungsten layers. The readout of the modules, shown in Fig. 6 has been performed with PMTs with a bi-alkali photocathode coupled to the rear face of the modules via a light guide. The achieved relative energy resolution for the single modules, with a beam interacting centrally in each module is shown in the right part of Fig. 6.



Figure 5. Light yield of different samples of $1 \times 1 \times 100 \text{ mm}^3$ large rectangular LuAG:Ce fibers by the cut heated wire technique (left). Right: comparison of the fibers (rod 1-3) with fibers grown by the micro pulling down method.

Figure 6. Picture of a sampling calorimeter module equipped with rectangular $1.0 \times 1.0 \times 100$ mm³ large rectangular LuAG:Ce fibers (left) and the energy resolution of the 4 modules measured with a tagged photon beam.

3. DSB:Ce - A new scintillator material

Recently, a new scintillation material, DSB:Ce³⁺, was announced [6]. It can be produced as glass or nano-structured glass ceramics with application of standard glass production technology with successive thermal annealing. When doped with Ce³⁺, the material can be applied as scintillator. The light yield of luminescence is near 100 ph/MeV. Un-doped material has a wide optical window from 4.5eV and can be applied to detect Cherenkov light. Several samples with dimensions of 15x15x7 mm³ have been tested for damage effects under irradiation with gamma-quanta and 150 MeV protons. Results of the tests of one of these samples are presented in Fig.7. No significant damage after both type of irradiations was observed.



Figure 7. Optical transmittance spectra of the small sample of DSB:Ce³⁺ material before and after irradiations with 150 MeV protons and low energy γ -rays

The first full sized (12.5 cm length) sample of DSB:Ce³⁺ was produced at the end of 2015. A relatively large amount of uniformly distributed macro-defects were observed. The properties of the sample were measured in the same manner. Results of the light yield measurements at different temperatures as well as transversal and longitudinal optical transmittances are presented in Fig. 8. The relatively large differences of the slow component at different temperatures have to be further studied. The transversal optical transmittance shows satisfied uniformity along the sample. Relatively low

value of the longitudinal transmittance in comparison with transversal values can be explained by Rayleigh scattering on the macro-defects. Therefore, further optimization of the production technology of large volume samples of $DSB:Ce^{3+}$ is needed.



Figure 8. Test results of a 12.5 cm long $DSB:Ce^{3+}$ sample. Light yield dependence on integration time at different temperatures (left). Longitudinal and transversal transmittance spectra (right).

4. Radiation hardness of the organic scintillator EJ-260

Plastic scintillators play a significant role in the construction of large area detectors in high energy physics experiments. Future concepts for detectors at HEP experiments, particularly at collider facilities, will require an unique combination of the material features and affordable price. Crucially important will be a minimal level of radiation damage effects under the electromagnetic part of the ionizing radiation as well as due to high energy hadrons. It requires a tolerable deterioration of the optical transmittance, a moderate level of the damage of the scintillation mechanism itself and a minor contribution of the radio-luminescence due to radio-nuclides which might be generated by nuclear reactions in the material of the detector. Here we report on first results of the study of commercially produced plastic scintillation material EJ260 (Eljen Technology, Sweetwater, USA). This bright, fast, green light emitting scintillator has properties similar to other products available on the market such as BC428 and NE103, respectively. The samples have approximate dimensions of $5.0 \times 5.0 \times 1.26$ cm³. The two large opposite sides of each sample were optically polished.

4.1. Irradiation with ⁶⁰Co gamma-quanta

The original surfaces of the sample 081015Q02 were unfortunately mechanically damaged during the preparations for the tests. Therefore, we tried to perform optical re-polishing procedure. The thickness of all samples after the polishing procedure was reduced to the value of 9.8 mm.



Figure 9. Optical transmittance spectra of a sample 081015Q02. The green curve is the distribution obtained before the mechanical damage of the surfaces, black and red curves are data measured after the re-polishing procedure and before and after irradiation with γ -rays, respectively.

Measurements of the optical transmittance of the sample after the repolishing procedure and before and after irradiation with the ⁶⁰Co γ -source (integral dose =100 Gy) were performed. The results in comparison to data obtained before the repolishing are presented in Fig. 9, which shows a significant loss of the optical transmittance after the repolishing procedure but no considerable losses due to γ -irradiation.

4.2. Irradiation with 150 MeV protons

For irradiation tests with a 150 MeV proton beam at KART-KVI (Groningen, Netherlands), one sample was cut in two parts with similar dimensions (2.5x5.0x1.26 cm³). One piece of the scintillator was placed in front of a 4.5 mm thick lead block and was numbered "1". The second piece was placed behind and numbered as "2". The transversal size of the lead block was significantly larger than both scintillators. The assembly of two scintillator layers with a lead sheet in between was chosen to simulate a single section of a sampling calorimeter. The assembly "scintillator 1/lead/scintillator 2" was irradiated with 150 MeV protons impinging perpendicular to the front face of scintillator 1 with an integral fluence of $5 \cdot 10^{13}$ protons/cm². The scintillator samples were separated from the lead block six hours after the end of irradiation. Since the samples showed a relatively high level of radiation induced activity they had to be stored for two months at a temperature below -25°C. Receiving the samples back, the optical transmittance was measured again as well as after an additional irradiation with γ -rays of a ⁶⁰Co-source with an integral dose of 100 Gy (1.5 Gy/min.). The transmittance spectra before and after the irradiation procedures as well as spectra of the radiation induced coefficient of both samples after irradiation with 150 MeV protons and additional γ -rays are presented in Fig. 10.



Figure 10. Optical transmittance spectra (left) and spectra of the radiation induced coefficient (right) of both samples before and after irradiations with 150 MeV protons and low energy γ -rays.

The radiation induced spectra do not indicate any difference with respect to the positioning of the scintillator relative to the lead block. Secondary and forward emitted reaction products with lead do not cause additional damage in the sample placed behind the absorber block. The irradiation with a dose of 100 Gy of γ -rays does not induce a visible additional damage effect in any of the samples.

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