

# Study of the New Glass and Glass ceramic Stoichiometric and Gd-loaded $\text{BaO} \cdot 2\text{SiO}_2$ (DSB) Scintillation Material for Future Calorimetry, and Studying of properties of the plastic scintillator EJ-260 under the irradiation with 150 MeV protons and 1.2 MeV gamma-rays

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In the last decades crystalline inorganic scintillation material has played a dominant role in calorimetry in medium and high energy physics experiments. Future detector developments will have to focus on cheap, fast, and radiation hard materials, especially for application in collider experiments. In order to increase the radiation hardness with respect to hadronic damage systematic studies of scintillation materials with lower effective nuclear charge have been initiated in particular with the LHC upgrade [1] and the ILC Program [2]. In particular, a significant increase in luminosity has to be considered. The present study has been based on the glass material  $\text{BaO} \cdot 2\text{SiO}_2$  (DSB) using different activators such as Ce or Gd ions. The production of various samples in different shapes takes advantage of optimized sintering processes and the established technology of glass production. We will report on test results of a large set of DSB samples with volumes of 1-2 cm<sup>3</sup> as well as large size samples up to a length of 12 cm or thin fibers, respectively. The investigation has been focusing on light output, the scintillation kinetics, optical transmission and radiation hardness with respect to irradiations with a high dose due to gamma-rays or 150 MeV protons.

[1] The CERN Large Hadron Collider: Accelerator and Experiments, Vol. 1-2, CERN, Geneva, 2009.

[2] International Linear Collider Technical Design Report, Tokyo, Geneva, Chicago –12 June 2013.

## Summary

### I. INTRODUCTION

In the last forty years, application of crystalline materials in detectors for ionizing radiation detectors has played a crucial role in the discovery of the properties of matter and promoted a continuous progress in the detecting technique. Future concepts of the detectors at HEP experiments will require an unique combination of the material features, particularly in case of luminosity collider experiments. A minimal and tolerable level of radiation damage under the electromagnetic part of ionizing radiation and due to energetic hadrons as well will become mandatory. That requires low deterioration of the optical transmission, a low level of afterglow and radio-luminescence due to radioactive nuclides to be created due to nuclear reactions in the detector material. From systematic studies of the radiation hardness of inorganic optical and scintillation materials during the last three years we concluded that both oxide and fluoride crystals which consist of atoms with atomic number below 60 will be favorable.

In this study we focus on the study of glass ( $\text{BaO} \cdot 2\text{SiO}_2$ ) and glass ceramics (DSB: Ce/Gd) obtained from this glass and their heavier modifications which are obtained by the admixture of heavy rare-earth ions. The transparent glass ceramics contains nano-sized particles of  $\text{Ba}_2\text{SiO}_5$  which improve scintillation properties of the resulted material. In all cases mass-production at a low cost appears realistic.

### II. THE INVESTIGATED SAMPLES

DSB glass and glass ceramics are obtained by glass production technology with successive thermal annealing. It can be produced in bulk and fiber shapes. The “mother”glass is prepared from the constituents Barium and Silicon and produced in a mold from the molten glass mass. Specifications for the purity of the initial ingredients is similar to the production of single crystalline scintillators. Afterwards it is annealed according to an optimized heating procedure to improve its properties. Gadolinium salt is added to the glass at the stage of the raw material preparation already.

The creation of the nano-crystallites occurs due to homogeneous seeding during thermal annealing. The composition does not contain special dopants acting as nucleation agents. Nucleation occurs by creation of ordered nano-sized parts of the glass. It is interesting to note that in the stoichiometric composition ordering is carrying on due to changes of the distances between atoms and angles of boundaries, so no mass transfer occurs. Therefore, no spores or other imperfections at the partial crystallization of the “mother”glass are to

be expected. Nano-structuring of the glass is obtained at temperatures in the range 800-900 degC during the treatment short in time. However, when the temperature is sufficiently high, an avalanche of crystallization is initiated in the glass. The glass ceramics becomes non transparent because it is composed of many micro-sized particles strongly scattering light.

### III. EXPERIMENTAL RESULTS

The spectroscopic and scintillation properties were measured with a Cary1E Varian spectrophotometer, a custom made luminescence spectrometer SDL-2 and a start-stop scintillation kinetics spectrometer. The spectra of the signal amplitudes using various commercial photomultipliers, such as XP20202 or R2059-1, were recorded with commercial electronics and DAQ systems.

#### A. Luminescence properties

Three types of material have been investigated, the basic Ce-doped glass, the DSB:Ce and the glass heavily doped with Gd ions. One can state that the luminescence spectra of the three materials are similar and consist of two bands peaking near 440 and 460 nm. The intensities of these bands vary from material to material. However, the band at the longer wavelength dominates in glass materials whereas the component at shorter wavelength shows an increase of the relative intensity when the material is partly crystallized. The similarity of the luminescence spectra confirms that the luminescent centers have the same origin in all samples. It indicates that even in a glass heavily loaded with Gd<sup>3+</sup> and surrounding the Ce<sup>3+</sup> the luminescence centers remain the same as in BaO\*2SiO<sub>2</sub> glass.

#### B. Light output of first samples

The light output of first samples of DSB:Ce measured with low energy gamma-sources was on the level of 100 phe/MeV using a standard photomultiplier with bialkali photocathode and an integration time of 4μs. Similar tests with samples loaded with Gadolinium Oxide show a significantly increased light yield and efficiency for obtaining even a photo peak for gamma-rays.

The sample #1 contains 10 weight percent Gd<sub>2</sub>O<sub>3</sub> and sample #2 the double loading. In both cases 0.5 weight percent of Ce are added. Both samples have a cross section of 10x10 mm<sup>2</sup> and a thickness of 5 mm. In the first case asymptotic values of 200 phe/MeV are obtained for the longest integration gate. Sample 2 even reaches values close to 300 phe/MeV.

The irradiation with gamma-rays (<sup>60</sup>Co) with an integral dose of 100 Gy confirms for both samples the high radiation hardness of the optical transmission. Even the response to <sup>137</sup>Cs after irradiation shows no significant shift of the photo peak.

The strong increase of the efficiency and light output with increasing Gadolinium concentration can be explained by the formation of a Gd<sup>3+</sup> subzone within the inorganic compound which promotes diffusion of electronic excitations towards the Ce<sup>3+</sup> ions or avoid capture of carriers by boundaries within the glass. The samples show a significantly slower kinetics at smaller content of Gd. Obviously, the kinetics is accelerated with an increase of the concentration.

#### C. First production of DSB:Ce fibers

The physical properties of DSB:Ce allow a wide spectrum of shapes. Besides the casting of blocks and larger bulks the technology of fiber production of glasses can be applied. However, there is in addition the thermal annealing process which has to be studied in detail with respect to the achievable quality of fibers.

There has been a first attempt to produce round fibers of typically 1 mm diameter and an overall length of 200mm. The confirmation of the luminescence mechanism in the fiber has been investigated on one hand by a direct measurement of the emitted light after excitation with a LASER light of 325nm. One observes both components but with a stronger attenuation within the fiber for the component at shorter wavelength.

A direct measurement of the light output created by passing electrons from a <sup>90</sup>Sr source recorded in coincidence with a plastic scintillator has delivered on average 1.5 phe with a photomultiplier with bialkali photocathode. In addition, the observed light output is position dependent due to a strong light attenuation. However, the illumination by a red LASER indicates macro defects within the fibers. Therefore, additional research is needed to optimize the manufacturing process for fibers.

As a first application a small prototype of a sampling calorimeter module containing 4x5 50 mm long fibers in a matrix made of Molybdenum of 1mm wall thickness has been tested with cosmic muons. The common readout of all 20 fibers with a photomultiplier provides a signal corresponding to > 100 detected photoelectrons.

### IV. CONCLUSION AND OUTLOOK

The status report of the development of the new material DSB:Ce presents even at a very early stage very promising features for future applications in calorimetry. The first results on samples loaded with Gadolinium indicate new options for optimizing the efficiency, the light yield and the decay kinetics. So far even samples with a volume such as 23x23x120mm<sup>3</sup> have been produced. Such blocks might be used to cut quadratic fiber with sufficient homogeneity.

**Author:** NOVOTNY, Rainer Willi (Justus-Liebig-University)

**Co-authors:** Dr BORISEVICH, A. (INP Minsk); Dr ZAUNICK, Hans-Georg (University Giessen); Prof. BRINKMANN, Kai-Thomas (University Giessen); Dr KORJIK, Mikhail (INP Minsk); Mr ZIMMERMANN, Sebastian (University Giessen); Dr DORMENEV, Valery (University Giessen)

**Presenter:** NOVOTNY, Rainer Willi (Justus-Liebig-University)

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