Performance of DSB – a new glass and glass ceramic as scintillation material for future calorimetry

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Performance of DSB – a new glass and glass ceramic as scintillation material for future calorimetry

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Abstract. The application of crystalline materials in ionizing radiation detectors has played a crucial role in the discovery of the properties of matter. However, the experiences gathered at high intensity machines such as the LHC have indicated their limitations and underlined the requirements for materials being more tolerable to radiation damage in particular caused by energetic hadrons. Systematic studies of the radiation hardness of inorganic optical and scintillation materials propose both oxide and fluoride crystals composed of atoms with atomic numbers below 60. In this study we report on a cheap glass ($BaO*2SiO_2$) and DSB: Ce glass ceramics even capable for mass production. Admixing gadolinium oxide (Gd^{3+}) even provides a two times larger light yield. Both types of the materials can be produced in a fibre and bulk geometry. This paper summarizes the overall performance and reports on a first test of a 3x3 matrix of large volume samples exposed to energy-marked photons up to 180 MeV.

1. Introduction

Since the discovery of NaI(Tl) as an efficient inorganic scintillator the development and application of crystalline materials for ionizing radiation detectors has played an important role in the discovery of the properties of matter such as the detection of the Higgs boson. However, the observation of severe radiation damages in particular due to high energetic hadrons and highly ionizing secondary products will require in future, particularly in case of high luminosity collider experiments, a minimal or at least tolerable level of radiation damage to guarantee a minor deterioration of the optical transmission, a low level of afterglow and radio-luminescence due to radioactive nuclides being created in nuclear reactions within the detector material itself. We are performing a systematic study of the radiation hardness of inorganic optical and scintillation materials [1,2]. It appears that both oxide and fluoride crystals which consist of atoms with atomic numbers below 60 could withstand the irradiation environment of future experiments.

Among other materials we have studied ($BaO*2SiO_2$) and the glass ceramics (DSB: Ce) obtained from this glass [3, 4, 5] or as heavier versions by the admixture of heavy rare earth ions such as Lu, Gd or Yb. The transparent glass ceramics contains after an appropriate tempering process nano-sized particles of Ba_2SiO_5 being the origin of the scintillation properties of the material. In all cases a future mass production at low cost appears realistic taking advantage of the well developed technology for glass production.

The phase diagram of the $BaO*SiO_2$ system [5] indicates that there are several compounds possible. However, the melting point increases with the content of Barium and one should have at least 30 mol% of SiO_2 to form a glass. In general, special attention has to be paid maintaining the stoichiometric composition of $BaO*2SiO_2$ which can be obtained in the pure form only in a narrow range of SiO_2. As experimentally confirmed [1], glass produced from stoichiometric compositions is radiation hard with respect to hadronic irradiation.

2. The achieved performance of initially small samples

The optimization of the scintillation yield depends strongly on the crystallization process during the annealing phase. X-ray diffraction studies performed by the group of D. Rinaldi (SIMAU, Ancona, Italy) investigating samples doped with Ce, Tb and Dy, respectively, have confirmed the formation of crystalline structures at temperatures between 750-900 ºC. Beyond that temperature limit the system crystallizes as a mixture of $BaSi_2O_5$ and β-$BaSiO_3$ becoming opaque.

2.1. Luminescence properties

The basic parameters have been determined using small samples of a typical volume of a few cm^3. The luminescence yield was measured using phototubes (PMT) with a bialkali photocathode integrating the scintillation light up to a gate length of 10 µs. The transparent and optically polished DSB:Ce samples show a light yield on the level of 100 phe/MeV using low energy radioactive sources. Depending on the concentration of the additional loading with Gd_2O_3 the light output increases significantly up to a factor three. The maximum light yield is reached at integration gates of 4-10 µs. There is no indication of a strong temperature dependence of the luminescence in the range between -45 ºC and +18 ºC, respectively, except at large integration gates due to some slower components.

2.2. Radiation hardness

The radiation hardness has been tested primarily by investigating the change of the optical transparency in the relevant wavelength region between 300 nm and 900 nm, respectively. The irradiation studies have been performed using low energy γ-rays from a strong ^{60}Co source at the radiation centre of the Justus-Liebig-University up to an integral dose of 100 Gy or using 150 MeV protons at a flux of $\sim 2\cdot 10^{11}$ protons/s/cm^2 up to an integral fluence of $5\cdot 10^{13}$ protons/cm^2 performed at the cyclotron facility of the University of Groningen (The Netherlands).

The un-doped mother glass shows a drastic reduction of the optical transparency for electromagnetic and even more in case of hadronic probes but the DSB:Ce and DSB:Ce/Gd samples indicate only a small reduction of the transmission. Irradiation studies with 1.2 MeV γ-rays up to an integral dose of 500 Gy have confirmed that the spontaneous recovery of damage can be drastically enhanced by stimulating with preferentially blue LED light (λ=460 nm). The transparency can be completely recovered within an illumination time of only 200-300 minutes investigated at room temperature. Even infrared light indicates already an acceleration of the recovery. Typical optical spectra are shown in ref. [1,6].

3. The first large volume samples

After the promising tests of small samples the involved Belarus supplier has delivered large rectangular blocks of 23 x 23 mm^2 cross section and a length up to 125mm. The samples are optically polished on all surfaces. The technology of the production of large pieces is presently limited by the inhomogeneity of the material due to the creation of bubbles which cannot be avoided in the oven used so far. Figure 1 illustrates the optically visible imperfection in the first Gd-loaded DSB:Ce which is randomly distributed. These bubbles have a strong impact on the optical transmission and consequently on the light collection as illustrated in figure 2.

The formation of bubbles and little cracks is even more pronounced in first attempts to produce round fibres of typically 1 mm diameter [6]. In a first test of 20 fibres of 50 mm length embedded into a Mo-structure and completely readout on one side via a small PMT of 19mm diameter (Philips XP1912) showed a response of ~ 80 phe for cosmic muons passing the detector module vertically.

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Figure 1. Picture of a large DSB:Ce block (23 x 23 x 125 mm^3) loaded with Gd.

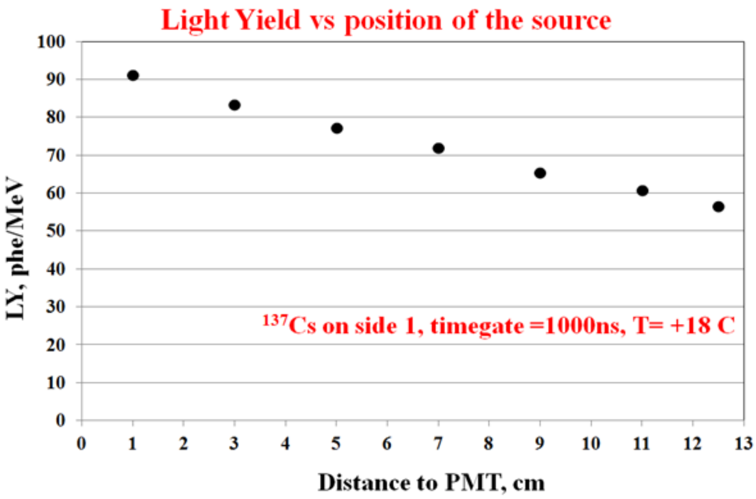

Figure 2. Position dependence of the collected light yield as a function of the position of the ^{137}Cs source along the length of the DSB:Ce/Gd scintillator block as shown in figure 1.

In spite of the inhomogeneity of the scintillator block, which was readout via a photomultiplier, a good response to low energetic radioactive sources and the energy loss of cosmic muons penetrating in horizontal and vertical direction could be obtained (see figure 3).

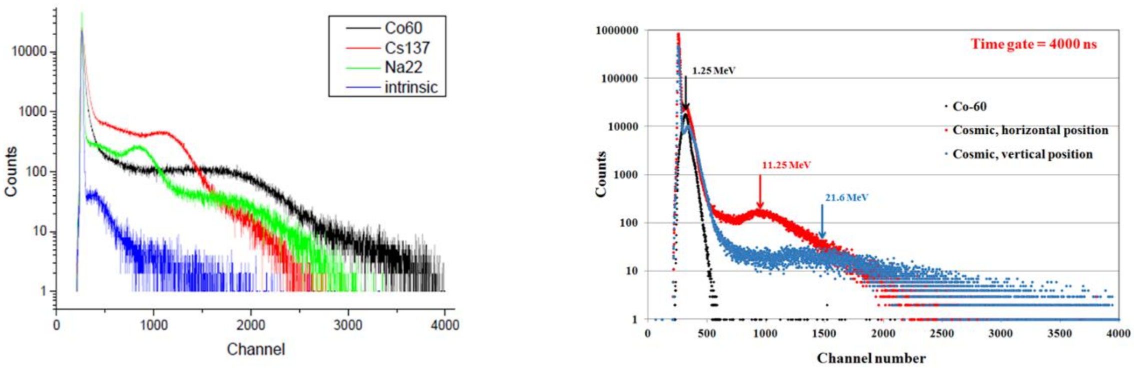

Figure 3. Response of a large volume DSB:Ce/Gd scintillator block to low energy γ-rays (left) and cosmic muons (right) penetrating the detector in longitudinal or transversal direction. The light output was detected at room temperature with a photomultiplier tube (Hamamatsu R2059-01) and integrated over a gate length of 4 μs.

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4. Quality and response of a set of 10 large volume DSB:Ce detector blocks

For a first calorimetric test of DSB:Ce, a set of 10 large volume blocks (20 x 20 x 100 mm^3) has been delivered in 2017 and carefully characterized with respect to homogeneity, light yield and radiation hardness. Figure 4 shows the samples when illuminated with UV-light.

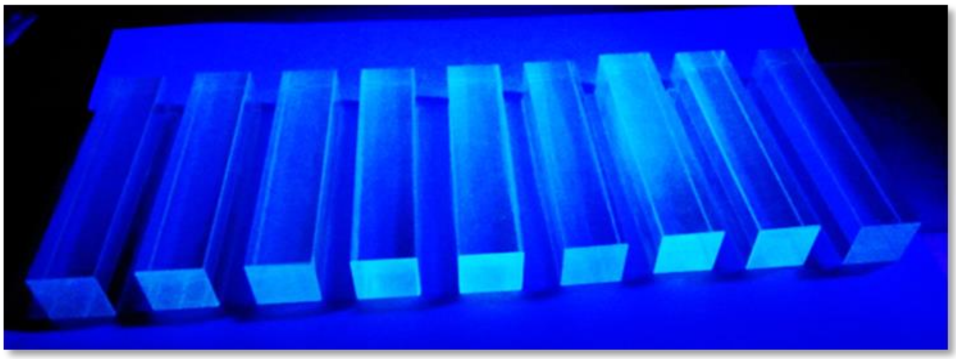

Figure 4. Photograph of the batch of 9 large DSB:Ce blocks (20 x 20 x 100 mm^3) illuminated with UV-light delivered in 2017. All surfaces are optically polished. Part of the UV-light is obviously scattered due to bubbles as clearly seen in sample #7 (counted from the left).

In spite of the improved technology, most of the samples contain areas with bubbles at different concentration having impact on the transport of the scintillation light. The figures 5 and 6 provide an impression of the homogeneity of the 10 large size samples with respect to the light yield as a function of integration time and its dependence on temperature. In the latter case, the three selected samples show within the accuracy of the measurement no temperature dependence at all between -25ºC and +20ºC, respectively. The variation of the light yield of the different samples is related to the concentration of bubbles. As shown in figure 6, the dependence of the collected light from the location of the γ-source shows in principle a similar slope for all investigated blocks.

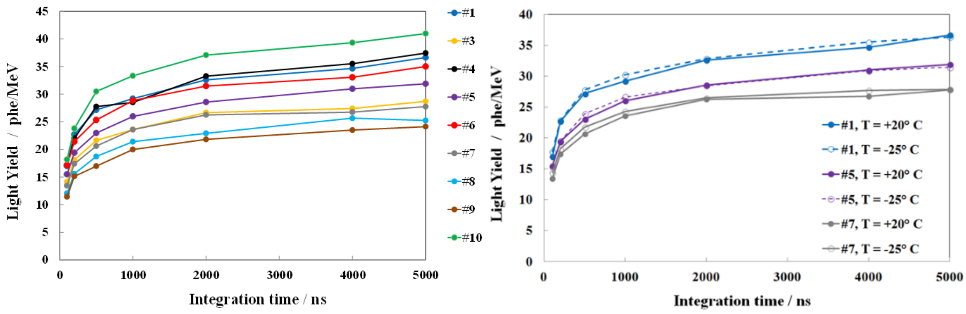

Figure 5. Light yield dependence on the integration time at room temperature (left) and in comparison at two extreme temperatures measured for large size DSB:Ce detector blocks.

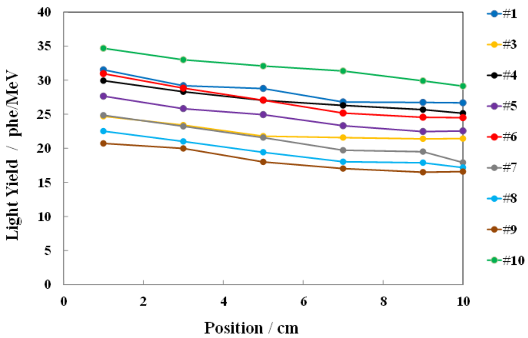

Figure 6. Dependence of the collected light yield on the relative position of the γ-source.

5. The response to high energy photons

In order to test the applicability of DSB:Ce as modules for electromagnetic calorimetry, a matrix of 9 blocks has been assembled as a prototype in spite of the short length of 10 cm compared to the radiation length of $X_0 \sim 3.5$cm. Each detector element has been individually wrapped in Teflon, optically coupled to a PMT (Philips XP1911) using silicon grease and mechanically fixed using light-tight black shrinking tube. The 9 modules are assembled as a 3 x 3 matrix. All units are readout individually by a VME-based DAQ system, recording an energy and time information each for an on- and off-line analysis.
The experiment was performed at the A2-Taggings-Facility at the MAMI accelerator at the University of Mainz. The tagging system provides energy marked Bremsstrahlung photons generated by a 450 MeV electron beam hitting a radiator. Photons in the range between 21.8 MeV up to 180.7 MeV have been selected with an intrinsic energy width of ~ 1.2 MeV. The collimated photon beam was hitting the detector module at a distance of 15 m with a beam spot of ~ 10 mm in diameter. An event to be recorded required the coincidence between the 15 selected tagger channels and the central module of the matrix. The timing information of the tagger was used in the off-line analysis to exclude random coincidences. Figure 7 shows photographs of the front and side view of the test matrix. A plastic scintillator placed in front was used as a veto to suppress electrons/positrons due to conversion of Bremsstrahlung photons in air between the radiator and the test matrix.

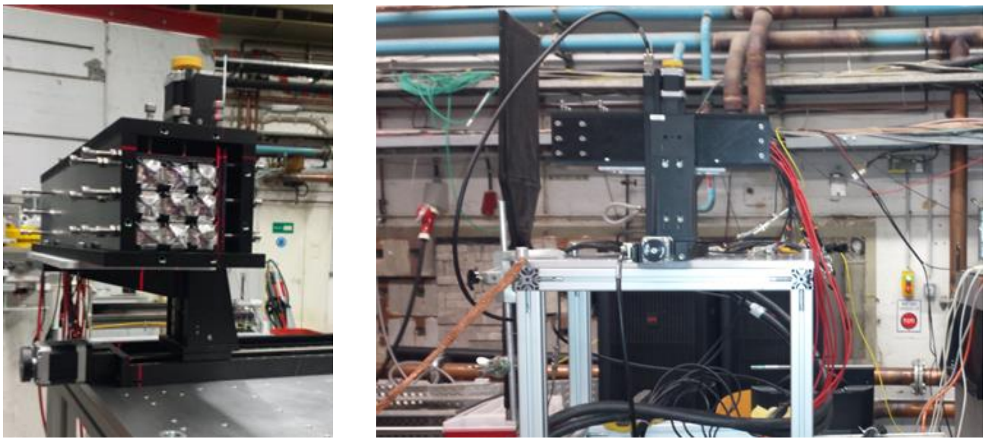

Figure 7. Photographs of the experimental setup at MAMI at Mainz. Front view of the 3x3 matrix (left) and side view of the arrangement showing the plastic veto in front (right). The photon beam was hitting from the left side.

The 9 detector modules have been calibrated during the experiment by directing the photons in the center of each unit and recording the energy deposition for the lowest photon energies. Alternatively, under the same detector settings cosmic muons have been recorded when penetration vertically the matrix. A coincidence with a plastic paddle on top was used as a trigger. Figure 8 shows the recorded response of the 9 units before a relative calibration.

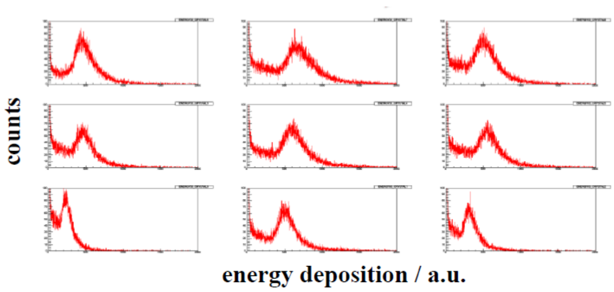

Figure 8. The response of the 3x3 matrix composed of DSB:Ce blocks to cosmic muons penetration vertically the modules.

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After the relative calibration the contribution of each responding unit can be used to reconstruct the electromagnetic shower. The minimum energy threshold of all neighboring detectors to be considered in the reconstruction has been set to ~ 2 MeV. Under these conditions, the maximum hit multiplicity varies between 4 at the lowest energy of 21.8 MeV up to 6 for 180.7 MeV. Figure 9 shows for two selected photon energies the energy distribution among the matrix when the photon hit the central module.

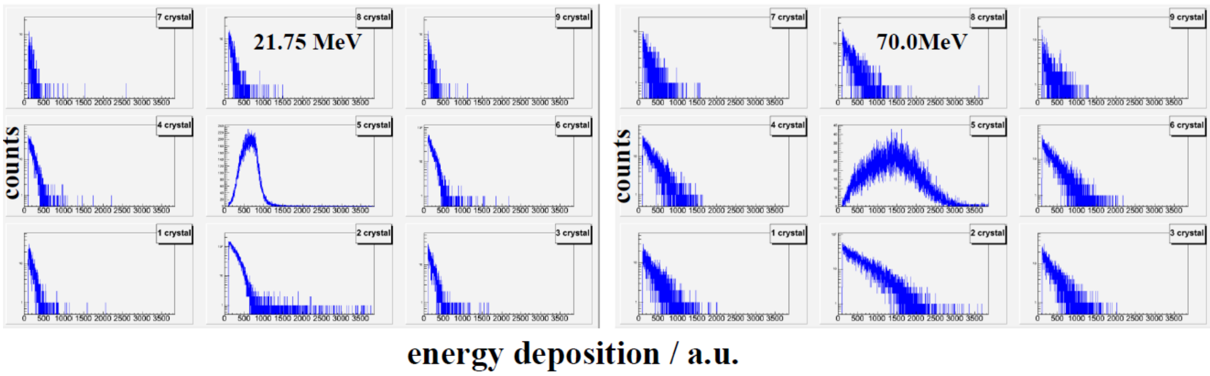

Figure 9. Response of the 3x3 matrix to energy marked photons of 21.8 MeV and 70.0 MeV, respectively. The shown energy depositions in the neighboring units are normalized to the central module.

Due to the total size of the active volume of the matrix with respect to the radiation length (X_0~3.5cm) as well as the Moliére Radius (R_M~3.8cm) the containment of the shower is very limited as expected even at low photon energies. In addition, the inhomogeneity of the 9 blocks and the observed position dependent light collection will have a strong impact on the achievable energy resolution.
Figure 10 shows for two selected photon energies the reconstructed line shapes of the central unit, the total energy deposition in the 8 neighboring elements and the overall reconstructed sum. At the lowest energy a Gaussian like shape can be obtained. However, already at an energy of 70 MeV the shower leakage is dominating.

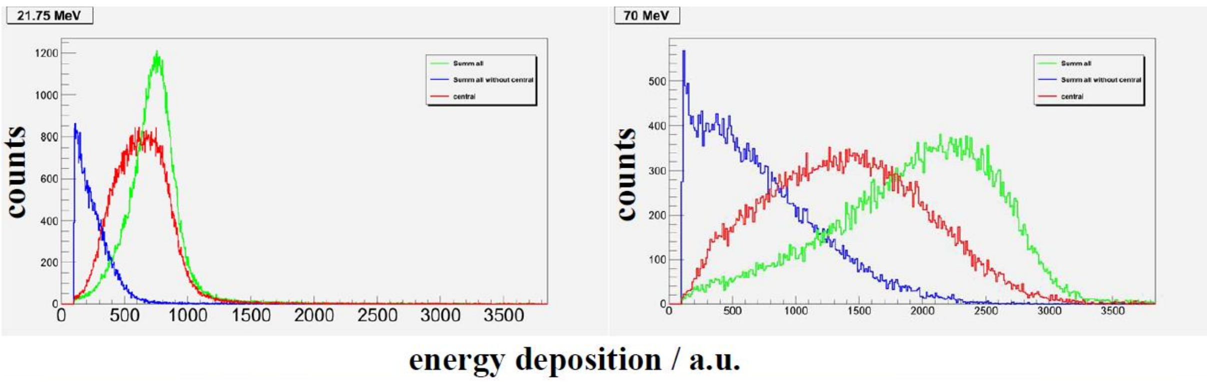

Figure 10. The reconstructed line shapes of the response of the matrix to photons of 21.8 MeV and 70.0 MeV. The green histogram represents the reconstructed total photon energy in arbitrary units.

For a better understanding the experimental data have been compared to GEANT4 simulations representing the identical geometry as well as threshold conditions. However, the simulation does not consider the inhomogeneity of the scintillators, the photon statistics of the scintillation process or light collection.

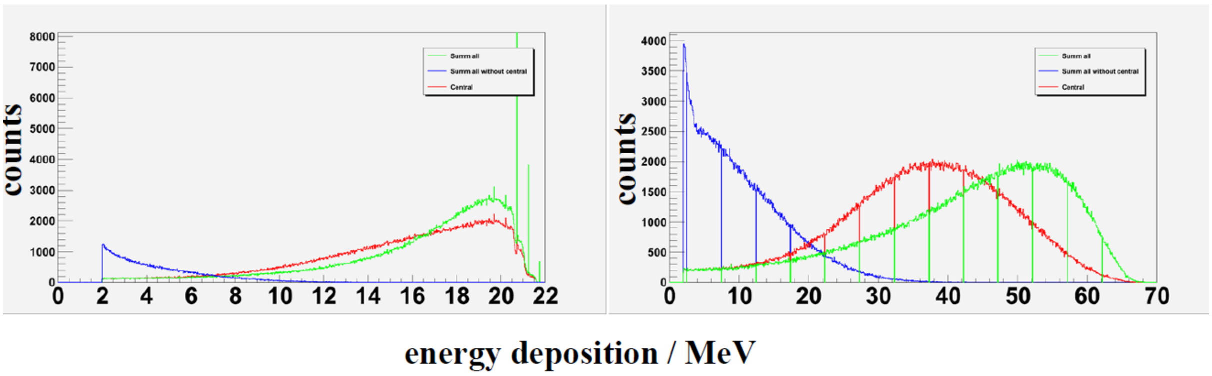

Figure 11. Simulation of the line shapes of the response of the matrix to photons of 21.8 MeV and 70.0 MeV based on GEANT4. The green histogram represents the reconstructed total photon energy.

Figure 11 shows for the same photon energies the response under ideal conditions. The distributions are given in absolute energies. The overall trend of the line shapes can be reproduced but the achieved energy resolutions are far from the simulated values. The comparison of the experimental and simulated energy resolutions is presented in figure 12.

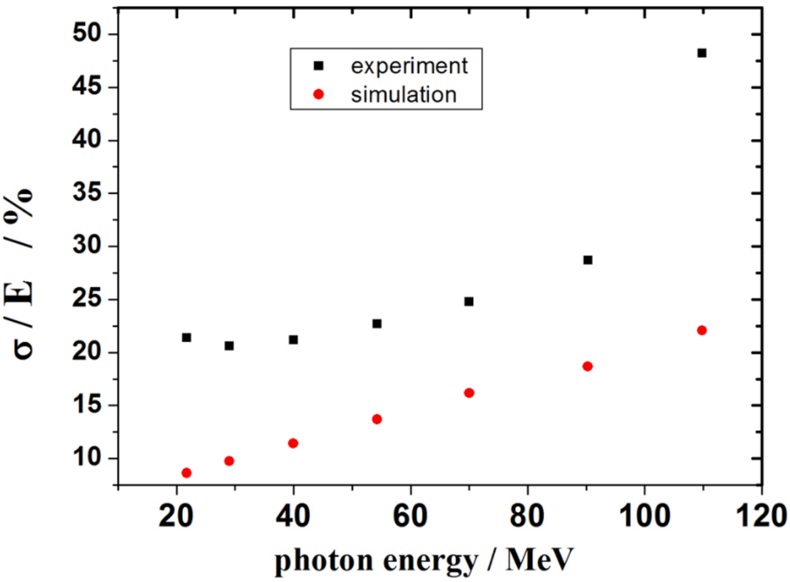

Figure 12. The achieved experimental energy resolution in comparison to simulations performed with GEANT4.

The discrepancy between experiment and simulation can be primarily explained by the limited photon statistics and the inhomogeneity of the units. It appears, as confirmed by the simulation, that the energy resolution is dominated by the shower leakage with the exception at the two lowest photon energies.

Besides the energy response, the timing properties of calorimeter elements are of increasing interest. The individual units have been readout via photomultipliers and the time response has been measured using a Constant Fraction Discriminator. In a separate run the photon beam has been directed between two units and the relative timing has been deduced for different incident photon energies. Figure 13 documents that, without further optimization, time resolutions below 300 ps for a single unit have been achieved. The observed plateau is an artefact due to the incomplete energy deposition in the only two coincident modules.

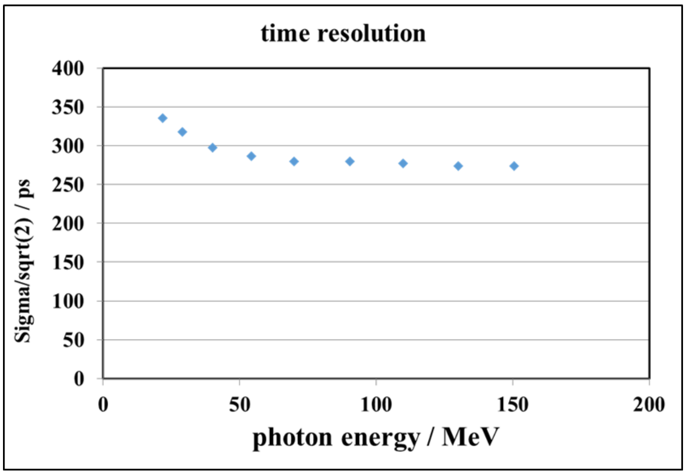

Figure 13. Experimental time resolution of a single unit of the matrix as a function of the incident photon energy**.**

6. Conclusion and outlook

The paper represents a status report on the investigations of a new glass ceramics DSB:Ce/Gd. The studies are focusing on the overall performance of the scintillation process, the quality of the material and the radiation hardness. For the first time, this material has been implemented in a test module for a calorimetric application. The results are very promising and the scintillator could be very well considered as active material in future radiation hard sampling calorimeters.
Nevertheless, the next steps require a significant improvement of the material by removing the bubbles. In addition, the availability of fibres either pulled from the melt or by cutting out of large blocks would further expand the spectrum of applicability.

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