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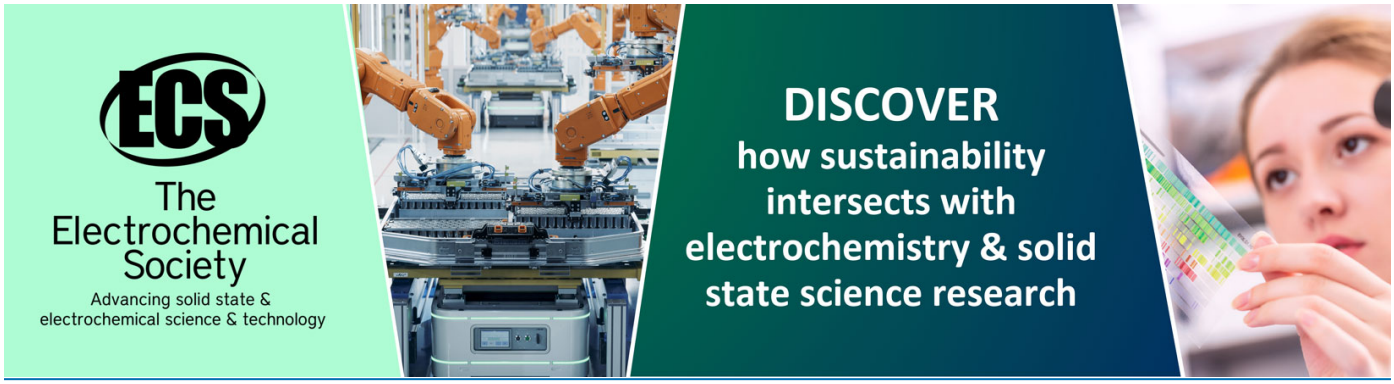

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Study of the New Glass and Glass Ceramic Stoichiometric and Gd^{3+}-loaded $BaO*2SiO_2$ (DSB:Ce) Scintillation Material for Future Calorimetry

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Abstract. In the last forty years, application of crystalline materials in ionizing radiation detectors has played a crucial role in the discovery of matter properties and promoted a continuous progress in the detecting technique. Further concepts of the detectors at HEP experiments will require an unique combination of the material features, particularly in case of collider experiments. Crucially important becomes a minimal level of radiation damage effects under the electromagnetic part of ionizing radiation and energetic hadrons as well: low deterioration of the optical transmission, low level of afterglow and low level of radioluminescence due to radio-nuclides being generated due to secondary nuclear reactions in the detector material itself. A systematic study of the radiation hardness of inorganic optical and scintillation materials have been performed. We concluded that both oxide and fluoride crystals which consist of atoms with atomic number less than 60 will be reasonably survivable in the irradiation environment of future experiments at colliders. In this study we focused on the study of cheap, capable for a mass production glass ($BaO*2SiO_2$) and DSB: Ce glass ceramics obtained from this glass. We also made this glass more heavy by admixing gadolinium oxide into the matrix. Glass with Gd^{3+} admixture possesses two times larger light yield than pure (BaO*2SiO2) glass and glass ceramics. Both types of the materials were produced as fibre and blocks of larger volume.

1. Introduction

In the last forty years, the application of crystalline materials in ionizing radiation detectors has played a crucial role in the discovery of matter properties and promoted a continuous progress in the detecting technique. Further concepts of the detectors at HEP experiments will require an unique combination of the material features, particularly in case of collider experiments. Crucially important becomes a minimal level of radiation damage effects in particular due to high fluences of energetic hadrons: low deterioration of the optical transmission, low level of afterglow and low level of radio-luminescence due to radio-nuclides being generated in nuclear reactions within the detector material. A systematic study of the radiation hardness of inorganic optical and scintillation materials have been performed during last three years [1,2]. We concluded that both oxide and fluoride crystals which consist of elements with atomic numbers below 60 will reasonably survive in the irradiation environment of future experiments at colliders. In this study we focus on material which is cheap and capable for mass production. We have explored glass ($BaO*2SiO_2$) and DSB: Ce glass ceramics, which was obtained in a further refining process [3,4] as well as heavier modifications by adding heavy rare earth ions. Transparent glass ceramics contains nano-sized particles of Ba_2SiO_5 which improve the scintillation properties. The phase diagram of the $BaO*SiO_2$ indicates several compounds. The melting point increases with the Ba content. However, to form glass the content of SiO2 should not be below 30 mol%. Moreover, special attention should be paid to keep up the stoichiometry of $BaO*2SiO_2$ which

can be obtained in the pure form in a narrow range of the SiO_2 content. The glass produced from stoichiometric compositions is radiation hard to hadronic irradiation [1]. The Ba-Si system is rather flexible to incorporate trivalent ions such as the rare earth ions Lu, Gd and Yb. However, trivalent ytterbium is known to be a strong quencher of the scintillation process in Ce^{3+} doped materials. Gadolinium Gd is not expensive and its presence in the crystalline material promote development of bright scintillation and a higher efficiency for absorption of γ-rays.

2. The investigated samples

DSB glass and glass ceramics are obtained by glass production technology with successive thermal annealing. They can be produced in bulk and fibre shapes. The "mother" glass is prepared from the constituents Ba and Si and produced in a mold from the molten glass. Specification for purity of the initial ingredients is similar to a specification for crystalline scintillators. Afterwards it is annealed according to an optimized heating procedure to improve its properties. Gd salt is added in the glass at the stage of the raw material preparation.

Table 1. Some selected properties of the developed scintillation material. (*depends on the Gd content in the final glass; ** for maximum Gd content)

material	ρ g/cm^3	Z_{eff}	X_o cm	λ_{max} nm	cutoff undoped material nm
$(BaO*2SiO_2)$:Ce glass	3.7	51	3.6	440, 460	310
DSB:Ce	3.8	51	3.5	440, 460	310
$(BaO*2SiO_2)$:Ce glass heavy loaded with Gd	4.7 - 5.4*	58	2.2**	440, 460	318

Creation of the nano-crystallites occurs due to homogeneous seeding during thermal annealing. The composition does not contain special dopants acting as nucleation agents. Nano-structuring of the glass can be obtained at temperature in the range of 800-900 ºC in a short treatment and a sudden crystallization of the glass occurs. Further thermal treatment leads to an increase of the dimensions of crystallites up to microns and the ceramics becomes non-transparent. Table 1 summarizes some of the relevant parameters of the investigated different materials.

3. 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.

3.1. The luminescence properties

Some relevant physical properties of the investigated materials are listed in table 1. 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 heavy loaded with Gd^{3+} and surrounding the Ce^{3+} the luminescence centers remain the same as in $BaO*2SiO_2$ glass. Table 2 summarizes the measured kinetics of the luminescence measured at room temperature for the three different materials. The Gd loaded samples show slower kinetics in case of a smaller content of Gd. However, the kinetics is accelerated with the increase of concentration. At large Gd content it becomes an efficient transporter of the excitations avoiding capture of carriers by unlinked boundaries which occurs in the glass. To our expectation further increase of the Gd^{3+} content will lead to a shortening of the scintillation kinetics.

Table 2. Scintillation kinetics of the measured samples

Material	Decay constants and their fractions in the kinetics		
	Fast ns (%)	Middfast ns (%)	Slow ns (%)
$(BaO*2SiO_2)$:Ce glass	22 (12)	72(50)	450(38)
$(BaO*2SiO_2)$:Ce glass loaded with 7 weight% of Gd oxide		86(40)	330(60)
$(BaO*2SiO_2)$:Ce glass loaded with 20 weigth% of Gd oxide	50(19)	120(39)	400(40)

The light output of first samples of DSB:Ce measured with low energy α-and γ-sources [3, 4] 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 γ-rays. As an example, figure 1 shows the response to a ^{137}Cs source for two samples. The sample #1 contains 10 weight percent Gd_2O_3 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^2 and a thickness of 5 mm

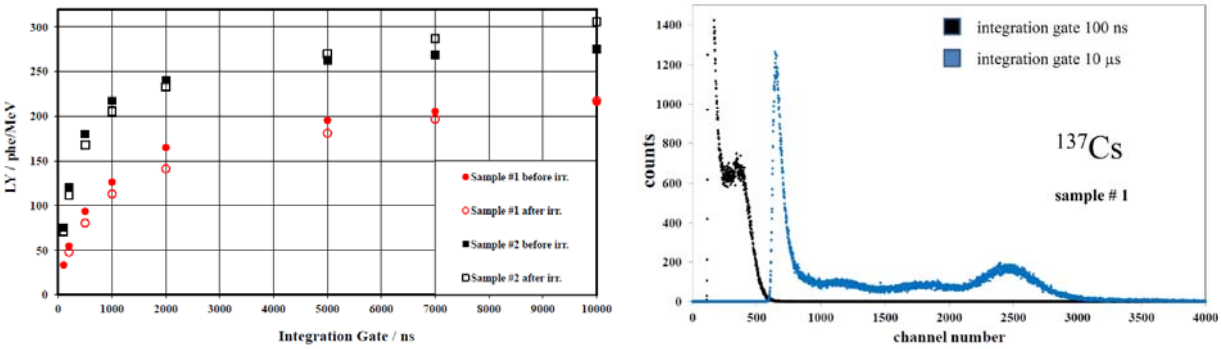

Figure 1. (left) Measured light output for two Gd loaded samples as a function of the integration gate of the output of the photomultiplier with bialkali photocathode. The response to a ^{137}Cs source was measured at room temperature. (right) Response of sample #1 to γ-rays from a ^{137}Cs source. The signals from the photomultiplier are integrated over a gate width of 100 ns or 10 μs, respectively.

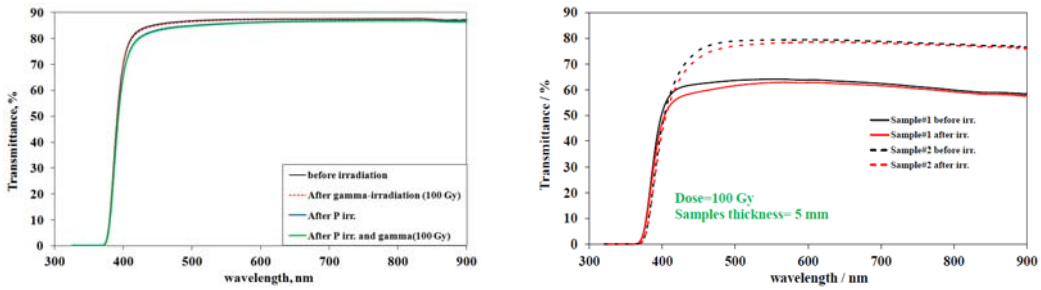

Figure 2.
(left) Optical transmission of a DSB:Ce sample before and after irradiation with γ-rays and 150 MeV protons. (right) Change of the optical transmission of two Gd loaded samples after γ-irradiation.

3.2. The radiation hardness

There have been done extensive studies of the different samples with respect to radiation hardness using either γ-rays (^{60}Co) or even a high fluence of 150 MeV protons provided by the KVI at the University of Groningen (The Netherlands). In particular, DSB:Ce after thermal treatment shows an

excellent radiation hardness. The samples have been irradiated with an integral dose of 100 Gy of γ-rays and an integral fluence of $5 \cdot 10^{13}$ protons/cm^2. The impact on the optical transmission is documented in figure 2 (left). The change of optical transmission after γ-irradiation of the two samples differently loaded with Gd is shown in figure 2 (right).

3.3. First production of DSB:Ce fibers and large size blocks

There have been first experiences in producing up to 200 mm long fibers as well as blocks up to 120 mm length. Both geometries show the expected luminescence. However, inhomogeneities and cracks, in particular in case of the fibers, limit light collection and cause short attenuation lengths. First samples are shown as photographic pictures in figure 3.

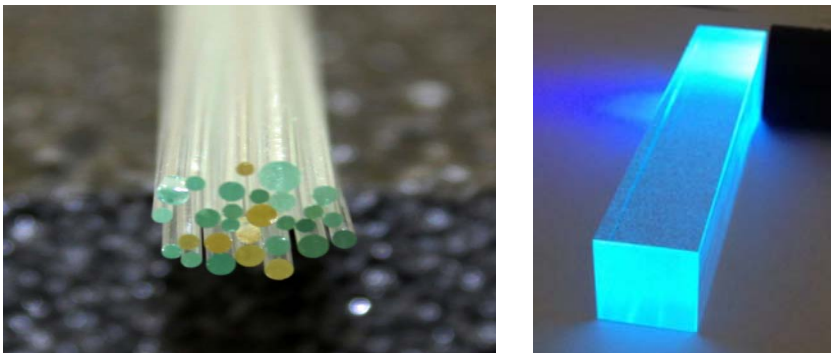

Figure 3. (left) Photograph of a bundle of 200 mm long fibers made of DSB:Ce with cross sections of ~ 1mm. (right) First large volume sample made of DSB:Ce. The rectangular block with a volume of 23x23x120 mm^3 has been polished on all surfaces is is illuminated with a UV-lamp.

4. Conclusions and outlook

The status report on the development on new radiation hard scintillator material presents even at a very early stage promising features for future applications in calorimetry. The first results of samples loaded with Gadolinium indicate new options for optimizing the efficiency, the light yield and the decay kinetics. However, there are still significant improvements of the manufacturing process necessary to avoid inhomogeneities, defects and inclusions of bubbles.

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