Light outputs of yttrium doped BaF$_2$ crystals irradiated with neutrons

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ABSTRACT: The fast luminescence component of barium fluoride (BaF$_2$) crystals with a subnanosecond decay time can find wide application in particle physics and nuclear physics. However, the slow luminescence component with the 630 ns decay time could cause pile-up signals at a high rate environment. Doping of BaF$_2$ crystals with rare earth elements suppresses the slow emission component, but at the same time the radiation hardness of the crystals deteriorates. This work presents the results of studying crystal samples, both pure BaF$_2$ and those doped with yttrium in a proportion of 1 at.% Y, 3 at.% Y and 5 at.% Y, irradiated with a fast neutron fluence of about $2.3 \times 10^{14}$ n/cm$^2$. Their light output and decay kinetics were measured before and after irradiation. It is found that the light output loss of a pure BaF$_2$ crystal after irradiation is about 7%, and the light output loss of yttrium doped samples after irradiation is about two times higher. The measurement results demonstrate that after irradiation the fast component of each sample has a relative light output loss 2–3% larger than the slow one.

KEYWORDS: Radiation damage to detector materials (solid state); Calorimeters; Radiation-hard detectors

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1 Introduction

Barium fluoride (BaF$_2$) crystals have long been known as scintillators with a broad luminescence band with a peak at 310 nm and a decay time of about 630 ns. Interest in studies of barium fluoride crystals increased after luminescence was discovered in them in the early 1980s at the boundary of the ultraviolet and vacuum ultraviolet regions (190–225 nm) with a decay time of about 0.7–0.8 ns [1–3]. The fast luminescence component of BaF$_2$ allows it to be used as a fast scintillator in medical applications, in high energy physics and nuclear physics.

The Mu2e experiment, which is currently being prepared at Fermilab, uses CsI crystals in the electromagnetic calorimeter [4]. The calorimeter is made of 1348 CsI crystals $34 \times 34 \times 200$ mm$^3$ in size, arranged in two annular discs. The calorimeter should provide an energy resolution of about 5% at $E = 105$ MeV and a timing resolution better than 0.5 ns [5]. At the second phase of the experiment, it is planned to significantly increase the beam intensity, to use an electromagnetic calorimeter made of BaF$_2$ crystals to increase the speed of operation [6]. This will require reducing the signal integration time from current $\sim 200$ ns to several tens of nanoseconds. The requirements for the calorimeter with respect to the energy and timing resolution will remain the same, but the harsh radiation environment will require many studies of crystals, photosensors and electronics. A high fraction of the slow component (about 85%) in the total luminescence can cause pile-up of signals at a high beam rate. To solve this problem, various approaches are being investigated: 1) the use of thin films on an avalanche photodetector [7] to suppress luminescence above 280 nm, 2) the use of a thin multilayer filter on the quartz glass substrate between the crystal and the photodetector to suppress luminescence in the range 250–400 nm [8], 3) the use of a solar-blind photomultiplier with an aluminum-gallium nitride (AlGaN) photocathode [9], 4) suppression of the slow scintillation component by doping a BaF$_2$ crystal with yttrium [10].

It is known that doping of a BaF$_2$ with rare earth elements (La, Y, Lu, Sc) suppresses the slow component emission [11]. However, doping of crystals usually leads to a decrease in their radiation hardness [12]. In this paper, we present a comparison of the light outputs of pure and yttrium doped BaF$_2$ crystal samples before and after irradiation with a neutron beam.
2 Crystal samples

In total, four samples were selected for the study: one pure BaF$_2$ crystal and three samples doped with a rare earth element yttrium in a proportion of 1 at.\% Y, 3 at.\% Y and 5 at.\% Y. The samples were grown at SICCAS (China) by the Bridgman method. The samples were cut from large ingots and had a size of $1 \times 1 \times 1$ cm$^3$, all faces were optically polished. Light outputs and decay kinetics of crystal samples were measured before and after irradiation.

3 Irradiation of crystals

The crystal samples were irradiated in channel #3 of the IBR-2M [13] pulsed reactor at the Frank Laboratory of Neutron Physics, JINR. The reactor operates with a pulse frequency of 5 or 10 Hz and a pulse duration of 200–300 $\mu$s. Channel #3 was specifically built to study the effect of neutron and gamma radiation on materials. The neutron flux density immediately after the water moderator is $\sim 10^{16}$ n/(cm$^2$×s) per pulse or $\sim 10^{13}$ n/(cm$^2$×s) on average over time [14]. All four crystal samples were placed together about 5 m from the water moderator and received the same radiation dose. A nickel wire placed with the samples was used to measure the fast neutrons (E>1 MeV) fluence from the wire induced activity.

Irradiation of the samples was carried out during the working cycle of the reactor, which lasted 14 days. From the activity induced on the nickel wire it was found that about $2.3 \times 10^{14}$ n/cm$^2$ (E > 1 MeV) passed through the samples during the irradiation run.

4 Setup for the crystals characterization

The light outputs of the crystal samples were measured before and after neutron irradiation with a $^{22}$Na gamma source at room temperature. The block diagram of the test setup is shown in figure 1. Two back-to-back outgoing 511 keV gammas from the $^{22}$Na source hit the crystal under tests and an additional trigger crystal made of cerium doped yttrium orthosilicate YSO (Y$_2$SiO$_5$:Ce) of the same size, and generated coincidence triggers.

![Figure 1](image.png)

Figure 1. The block diagram of the setup for crystals characterization.

The samples were wrapped with a double layer of 0.1 mm thick Teflon film. The signals from the BaF$_2$ samples were registered by a Hamamatsu R2059 photomultiplier tube (PMT) while the
signals from the YSO crystal were registered by a Hamamatsu R2496 PMT. No optical grease was used between the crystals and the PMT photocathodes. Signals from the R2059 PMT were recorded by the 10 bit 2 Gs/s CAEN NDT5751 Digitizer. Signals from the R2496 PMT were amplified by the CAEN N979B unit and sent to the NDT5751 Digitizer to generate a trigger signal. The threshold value set in the Digitizer for the trigger counter to generate a coincidence signal corresponded to an energy loss in the YSO crystal of about 100 keV. The total signals from the BaF$_2$ samples were integrated within 2 $\mu$s. The fast luminescence component of the samples was measured during the first 20 ns and the slow one after 20 ns. The light output of the samples was estimated from the full absorption peaks of 511 keV gammas. The systematic errors in measuring the positions of the full absorption peaks did not exceed 1.5%. The measuring channel was calibrated using a single electron peak by illuminating the PMT photocathode with low-intensity light from a light emitting diode. The calibration spectrum is shown in figure 2. The spectrum was fitted with an exponential function describing the noise and two Gaussians describing the pedestal and single electron events. Most of the events in the spectrum fall into the pedestal. The average number of photoelectrons in the spectrum is $\mu = 0.086$.

![Figure 2](figure2.png)

**Figure 2.** Spectrum from the PMT illuminated with a very low intensity light from a light emitting diode.

The R2059 PMT has a quantum efficiency of about 16–17% in the fast luminescence region (200–220 nm) and about 23–24% in the slow luminescence region (310 nm). The measured light outputs of the fast and slow luminescence of the samples were not corrected for the difference in the quantum efficiency of the PMT.
5 Results and discussion

As mentioned above, the light output of the samples was estimated from the full absorption peaks of 511 keV gammas from the $^{22}$Na source. The spectra of the signals from the samples before and after neutron irradiation due to $^{22}$Na excitation are presented in figure 3. The left column of figure 3 shows the spectra of the total signals from the pure and yttrium doped BaF$_2$ samples while the central and right columns depict the spectra of the slow and fast luminescence components respectively. The spectra correspond, from top to bottom, to the samples with an yttrium fraction of 0%, 1 at.%%, 3 at.% and 5 at.% respectively. The solid blue lines show the spectra taken before irradiation, and the dashed red lines present the spectra of the irradiated samples. Figure 4 depicts kinetics of the signals from the same samples before (solid lines) and after (dashed lines) irradiation. Here the light outputs are shown as a function of the integration time.

As can be seen in figure 3 and figure 4, doping of the BaF$_2$ crystals with yttrium leads to decrease of the total signals. The main reason for the decrease in the total signal is the suppression of the slow emission component. In the unirradiated samples the total signal drops by approximately a factor of 2.8, 4.5 and 4.8 in the 1 at.%%, 3 at.% and 5 at.% yttrium doped samples respectively compared to the pure BaF$_2$ crystal. At the same time, the slow emission components in the same
Doped samples were suppressed by the 3.6, 7.4 and 7.8 respectively compared to the slow emission of the pure BaF$_2$ crystal. The fast emission in the sample doped with 1 at.% Y practically did not change in comparison with the fast emission of the pure BaF$_2$ crystal, while the fast emission components of the 3 at.% and 5 at.% yttrium doped samples decreased by about 2.5% and 8% respectively.

**Figure 4.** Light outputs of the pure and yttrium doped BaF$_2$ samples before (solid lines) and after (dashed lines) irradiation as a function of the integration time.

Light outputs of all samples (in ph.e/MeV) before and after irradiation are presented in figure 5. The fast and slow emission components are shown together with the total signals. Note that the total and slow emission light yields are shown on the left vertical scale while the fast emission component is on the right scale.

It is known that rare earth doped BaF$_2$ crystals usually lose their radiation hardness compared to the pure BaF$_2$. Our data confirm this. This can be seen qualitatively from the spectra in figure 3 and from the light outputs of the samples before and after irradiation shown in figure 5. The light output loss of the total signals, fast or slow components of each sample was estimated as $1 - \frac{LY_{irr}}{LY_0}$, where $LY_0$ and $LY_{irr}$ are the light outputs (total, fast or slow component) of each sample before and after irradiation. The light output loss of each sample after irradiation for the total signals, slow and fast emissions is shown in figure 6. One can see that at our irradiation dose the light output loss of the pure BaF$_2$ crystal is about 7%. The light output loss of the yttrium doped samples is approximately two times higher than that of the pure BaF$_2$ sample. Another interesting observation is that in all yttrium doped samples the light output loss of the fast emission component is 2–3% higher than that of the slow emission.

Loss of the crystal light yield leads to a decrease in the energy resolution of the calorimeter. However, given that a neutron fluence of about $10^{13}$ n/cm$^2$/year is expected in the Mu2e-II calorimeter,
our measurements show that neutrons will not lead to dramatic losses in light yield and energy resolution of yttrium doped BaF$_2$ crystals.

Thus, our data confirm the suppression by several times of the slow luminescence component of yttrium doped BaF$_2$ crystals. However, the loss of the light output of the yttrium doped samples turned out to be two times higher than that of the pure BaF$_2$ crystal. Special attention should be paid to the fact that in the yttrium doped samples the loss of the light output of the fast luminescence component is 2–3% higher than that of the slow component.

6 Conclusion

Light outputs of a pure BaF$_2$ crystal and three samples doped with yttrium in a proportion of 1 at.%, 3 at.% and 5 at.% were measured before and after irradiation in a neutron beam. All samples were irradiated in the neutron beam of the IBR-2M pulsed reactor at JINR, Dubna. The total fast neutron ($E > 1$ MeV) fluence that passed through the samples was about $2.3 \times 10^{14}$ n/cm$^2$.

At this level of irradiation, the light output loss of the pure BaF$_2$ crystal is about 7%, while the loss of the light output of the fast component is about 1% greater than that of the slow one. The results show that the light output losses of both the fast and the slow emission component in the yttrium doped samples after irradiation are approximately two times higher than those in the pure BaF$_2$ crystal. It should be noted that the fast emission component of each yttrium doped sample after irradiation has a relative light output loss 2–3% higher than the slow one. Undoubtedly, additional studies of light yield losses in a wider range of radiation doses are needed.
Figure 6. Light output loss of the pure and yttrium doped BaF$_2$ samples after irradiation depending on the fraction of yttrium.

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