Radiation Monitoring Devices Inc.
44 Hunt Street
Watertown, MA 02472

A Fast, High Light Output Scintillator for Gamma Ray and Neutron Detection

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Principal Investigator: Gerald Entine, Ph.D.
Contributing Scientists: Kanai S. Shah, M.S.
Leonard Cirignano, M.S.
Jarek Glodo, Ph.D.
Edgar V. Van Loef, Ph.D.

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Submitted to:

Dr. Jehanne Simon-Gillo, SC-23
U.S. Department of Energy, SC-64
19901 Germantown Road
Germantown, MD 20874-1290
(via FedEx)

1. Introduction

In radiation detection systems employing scintillation crystals for the detection of high-energy particles such as γ-rays or neutrons, the scintillator often limits the performance of the system. Ideally, a scintillator should have a high light yield for good energy and position resolution, a fast response time for good time resolution and high density and atomic number Z for efficient γ-ray detection. The latter property is of less importance in the case of (thermal) neutron detection. Obviously, there are many other criteria such as transparancy of the material to its own emission, ease of crystal growth, etc. However, there is no commercially available material that meets all these criteria and the choice for a particular scintillator is often a compromise among these and other factors.

Recently, the scintillation properties of a new scintillator, $RbGd_2Br_7$:Ce were published by Dorenbos *et. al.* [1] and Guillot-Noël *et. al.* [2]. It was shown that $RbGd_2Br_7$:10% Ce has a very high light output (~ 56,000 photons/MeV), a fast principle decay (~ 43 ns), a linear response of light yield vs. energy, and good γ-ray detection efficiency. Furthermore, the presence of Gd makes this material a possible candidate for thermal neutron detection, as many of the Gd isotopes but especially ^{157}Gd have high cross sections for thermal neutron absorption (^{157}Gd: σ = 255,000 barns) in combination with a high Q value of 8 MeV. So far, however, only very small crystals (< 0.1 cm^3) of $RbGd_2Br_7$:Ce were grown.

In view of the attractive properties of $RbGd_2Br_7$:Ce for γ-ray and thermal neutron detection, and the lack of larger volume crystals, the goal of the Phase I project was to perform a rigorous investigation of the crystal growth of this exciting material and explore its capabilities for γ-ray and thermal neutron detection. The Phase I research turned out to be very successful. All technical objectives were met and in many cases exceeded expectations. We were able to produce large (>1 cm^3) $RbGd_2Br_7$:Ce crystals with excellent scintillation properties and demonstrated the possibility to detect thermal neutrons. As far as we are aware, our Phase I experiment was the first to demonstrate thermal neutron detection with $RbGd_2Br_7$:Ce. Clearly, the feasibility of the proposed research was adequately proven.

The Phase II research builds on the successful results obtained during Phase I. Phase II will initially focus on optimizing the $RbGd_2Br_7$:Ce growth process to produce high quality, larger volume $RbGd_2Br_7$:Ce crystals. We will continue to use the versatile Bridgman technique. During this process, crystal growth parameters will be adjusted for optimal growth conditions. Our goal is to produce high quality $RbGd_2Br_7$:Ce crystals of size 1”x 1”x 1” (~16 cm^3). We will work on packaging aspects that allow efficient light collection and prevent crystal degradation. We will study and measure emission spectra, light yield, scintillation decay, energy and time resolution. The effects of variation in Ce concentration on the scintillation properties of $RbGd_2Br_7$:Ce will be examined in detail. Comprehensive γ-ray spectroscopic and imaging studies will be conducted. Also, optimization of $RbGd_2Br_7$:Ce for thermal neutron detection will be addressed. Our initial studies will determine the optimal geometry of the $RbGd_2Br_7$:Ce crystals for neutron detection. For thermal neutron detection experiments, we will produce large area, thin samples (≤ 0.5 mm thick) in order to minimize γ-ray sensitivity without compromising thermal neutron detection efficiency. Thermal neutron detection will be conducted with samples coupled to photomultiplier tubes (PMTs) and silicon avalanche photodiodes (APDs). Finally, thermal neutron imaging studies will be performed by coupling $RbGd_2Br_7$:Ce scintillators to APD arrays available at RMD.

We will continue our collaboration with Dr. William Moses at LBNL and Dr. James Lund at SNL during Phase II. Dr. Moses will help to optimize and characterize the fundamental and scintillation properties. Dr. Lund will evaluate $RbGd_2Br_7$:Ce for γ-ray and thermal neutron detection and will utilize these scintillators in his instrumentation for stored nuclear material monitoring. Moreover, Canberra Industries (Meriden, CT) - a leading firm in nuclear detector and instrumentation - agreed to collaborate with us during Phase II as well in Phase III. Canberra will investigate the properties of $RbGd_2Br_7$:Ce for gamma ray and neutron detection instrumentation and will consider incorporating $RbGd_2Br_7$:Ce in its products. Canberra will use its own resources for these evaluation efforts.

Figure 1. Photographs of a large $RbGd_2Br_7$:Ce crystal grown at RMD.

2. Recent Progress

During the past 6 months we have concentrated our efforts on the optimization of the crystal growth and the investigation of the scintillation properties. Recently, we have made significant progress with sample packaging, which is very important due to hygroscopic nature of the material.

2.1. Crystal growth

Figure 1 shows an example of RGB crystals grown at RMD. For optimization purposes we have grown a few 10 - 20 mm diameter crystals.

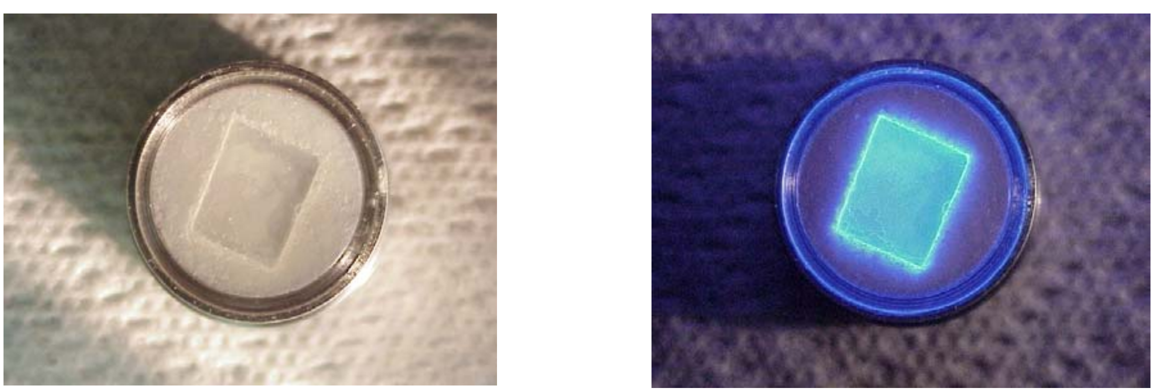
2.2. Packaging

Due to hygroscopic nature of the material, sample encapsulation in a moisture-free environment is crucial. Originally, the packaged samples were wrapped with a Teflon tape (reflector) and encapsulated in epoxy. Despite our efforts to seal the container from water and air, residual moisture left on the Teflon wrapping could still lead to sample deterioration. Use of reflective epoxy instead of Teflon tape was no options since it led to light loss and worse energy resolution. Recently we have developed a new metal case packaging technique that overcomes these problems.

Initial results with this new approach shows that the crystals exhibit a very small light loss (due to reflection on the case window) while preserving a good energy resolution. As shown in Fig. 2, the sample is placed in a metal can filled with reflective powder. The can is closed with a glass window.

2.3. Characterization of Bridgman Grown $RbGd_2Br_7$:Ce Crystals

Scintillation and spectroscopic properties of $RbGd_2Br_7$:Ce were evaluated. The samples were rectangularly shaped and a few mm thick.



(a) (b)

Figure 2. Encased $RbGd_2Br_7$:Ce crystal under (a) white light and (b) ultraviolet light illumination.

2.3.1. X-ray excited optical luminescence

X-ray excited optical luminescence spectra of $RbGd_2Br_7$:Ce crystals were recorded in reflection mode using a Philips X-ray tube with a Cu anode. Typically, the anode was operated at 40 kV and 20 mA. The scintillation light is dispersed by a McPherson 0.2-meter monochromator and detected with a Hamamatsu R2059 photomultiplier tube (PMT) having a quartz window.

The X-ray excited optical luminescence spectrum of a large $RbGd_2Br_7$:Ce crystal is shown in Fig. 3. Other crystals show similar luminescence spectra under X-ray excitation. The emission is due to Ce^{3+} $5d \rightarrow 4f$ transitions, peaking at 430 nm, which is well suited for bi-alkali PMTs.

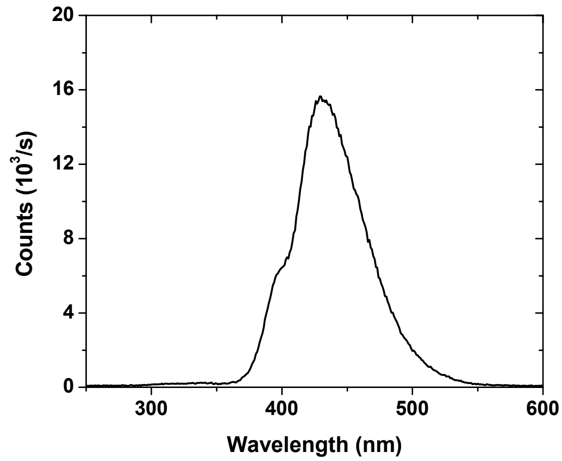
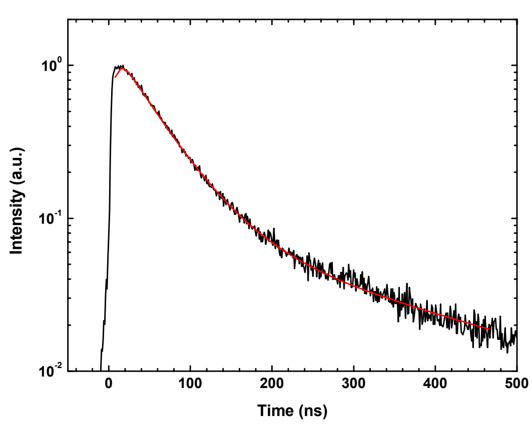


Figure 3. X-ray excited optical luminescence spectrum of $RbGd_2Br_7$:Ce.

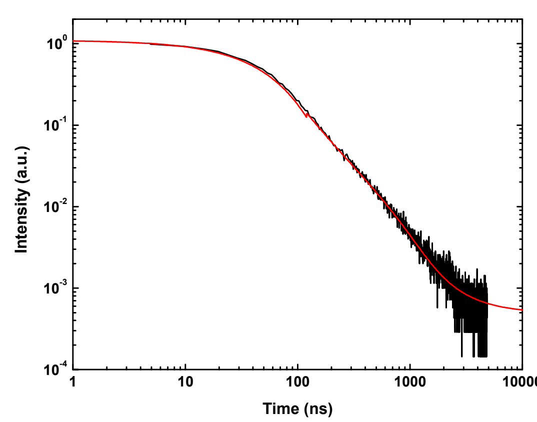
2.3.2. Scintillation decay

The scintillation decay time spectra of $RbGd_2Br_7$:Ce under ^{137}Cs excitation are shown in Fig. 4. A least-square fit of the decay curves is shown in red as well. Several decay components can be distinguished. The first component has a decay time of 52 ± 3 ns, which represents 70% of the total light yield. The second decay component has a slightly longer decay time of 320 ± 40 ns, which represents 25% of the total light yield. Additionally, a rise time of about 6 ns is observed. These results are similar to those obtained during the Phase I project and show that the scintillation of $RbGd_2Br_7$:Ce is very fast.

Measurements performed on a millisecond time scale under pulsed X-ray excitation (50 ns pulse width) show that $RbGd_2Br_7$:Ce exhibits a small level of afterglow. The intensity at 6 ms after the excitation pulse is almost 2 orders of magnitude lower than in the case of CsI:Tl.



(a)



(b)

Figure 4. Scintillation decay time spectra of $RbGd_2Br_7$:10% Ce with a time scale of (a) 500 ns and (b) 10 µs

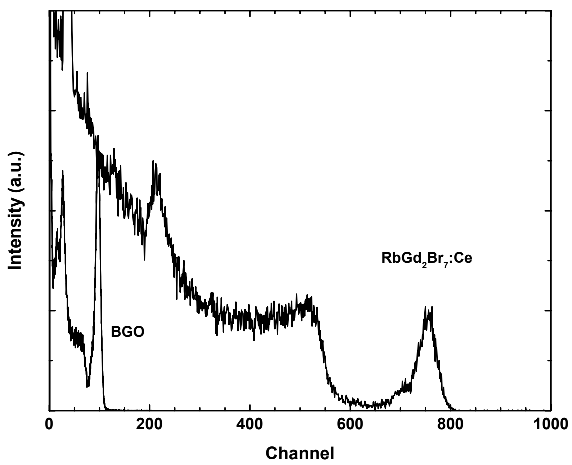


Figure 5. Pulse height spectrum of $RbGd_2Br_7$:10% under ^{137}Cs γ-ray excitation, obtained with a Hamamatsu R2059 PMT.

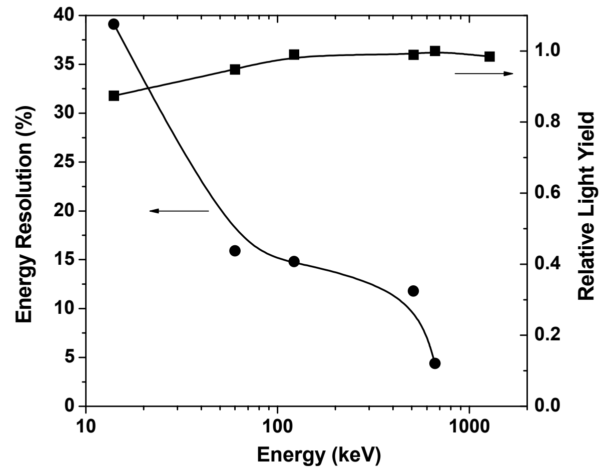


Figure 6. Energy resolution (●) and relative light yield (■) of $RbGd_2Br_7$:10% Ce as function of γ-ray energy.

2.3.3. Pulse Height Spectra and Energy Resolution

Figure 5 shows the pulse height spectra of $RbGd_2Br_7$:Ce and BGO under 662 keV ^{137}Cs γ-ray excitation obtained with a Hamamatsu R2059 PMT. Compared to BGO, the light yield of $RbGd_2Br_7$:Ce is 7.8 times larger. If we take 7500 ph/MeV as the absolute light yield of BGO [3], the absolute light yield of $RbGd_2Br_7$:Ce is 58500 ph/MeV. This value is not corrected for the difference in PMT quantum efficiency between BGO and $RbGd_2Br_7$:Ce, but is of the same order as the value for the light yield that we measured previously. The energy resolution R [full width at half maximum (FWHM) over peak position] of the 662 keV full absorption peak is $4.6 \pm 0.3\%$.

Using additional radioactive sources such as ^{241}Am, ^{57}Co, and ^{22}Na, we obtained the energy resolution and relative light yield of $RbGd_2Br_7$:Ce as function of γ-ray energy shown in Fig. 6. The $RbGd_2Br_7$:Ce crystals show a very good proportional response (light yield as function of γ-ray energy) that is better than measured for BGO [4] and comparable to that of YAP:Ce [4,5].

3. Future work

During the next period we will continue to investigate growth procedures to obtain high quality, large volume $RbGd_2Br_7$:Ce crystals. We also plan to build a radiation detector utilizing a $RbGd_2Br_7$:Ce crystal.

4. References

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