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Optical Spectroscopy and Scintillation Properties of a Cerium Activated Cs_2NaYBr_6 Crystal

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Introduction

The quest for high performance scintillators is crucial in supporting the development of uncooled, advanced gamma ray spectroscopy and radiation detection applications. New scintillators should have high light yield, short lifetime, good proportionality and stopping power. Scintillators with these characteristics can ultimately replace benchmark scintillators such as NaI(Tl) and CsI(Tl) while providing better energy resolution. In this study, we explore a cerium doped Cs_2NaYBr_6 scintillator for radiation detection applications. The result of its optical and radio-luminescent properties are presented.

Crystal Growth and Characterization

The cerium doped Cs_2NaYBr_6 crystal was grown by a vertical Bridgeman technique. Cerium was used as an effective activator to produce higher light yield and faster response. A 10 mm sample of a cerium doped Cs_2NaYBr_6 single crystal (12.7 mm dia.) was used for scintillation characterization (***Fig. 1***). Luminescence and scintillator properties, including photo-excitation and emission, photoluminescence quantum yield, gamma energy spectrum, and radioluminescence decay time, and linearity response, were investigated.

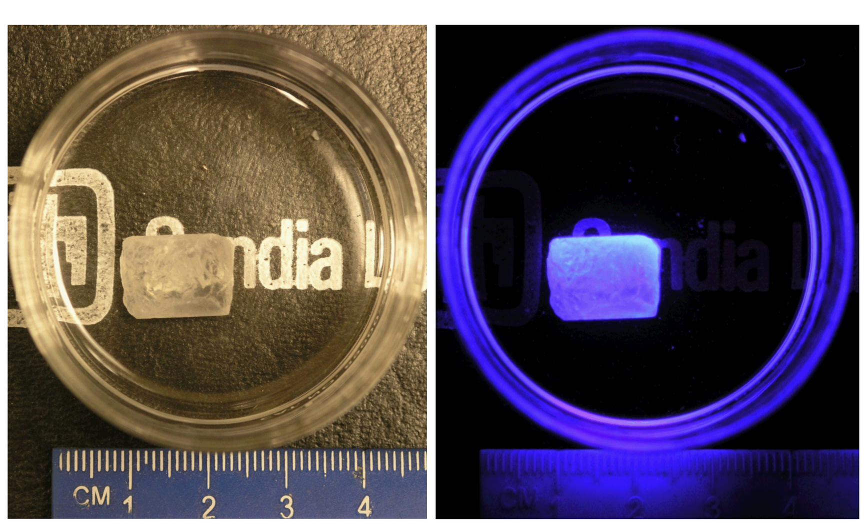

Fig. 1. The $Cs2NaYBr_6$ crystal used in radioluminescent characterization. Photographs on left and right are taken under ambient and black light conditions.

Structure and Thermal Properties

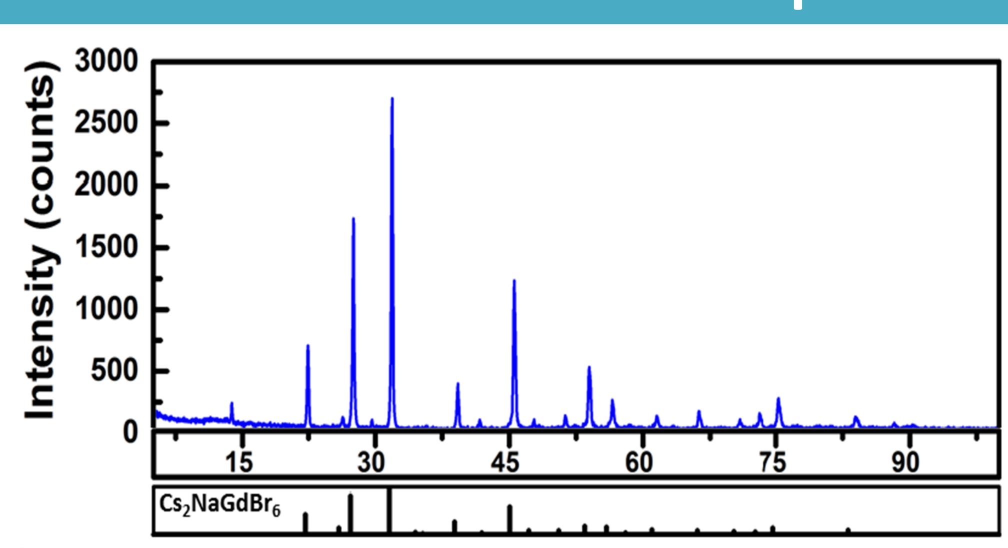

Fig. 2. X-ray powder diffraction pattern for the cerium doped Cs_2NaYBr_6.

Structural refinement based on X-ray powder diffraction data (***Fig. 2***) indicates that Cs_2NaYBr_6:Ce^{3+} has a cubic elpasolite structure (space group Fm-3m (225)) with a lattice parameter of 11.3003(3) Å, which is consistent with our embedded ion model prediction. Differential scanning calorimetry (DSC) measurement showing the melting point of this new compound is at 702 °C, and the enthalpy change associated with the melting process is 59.65 J/g (***Fig. 3***).

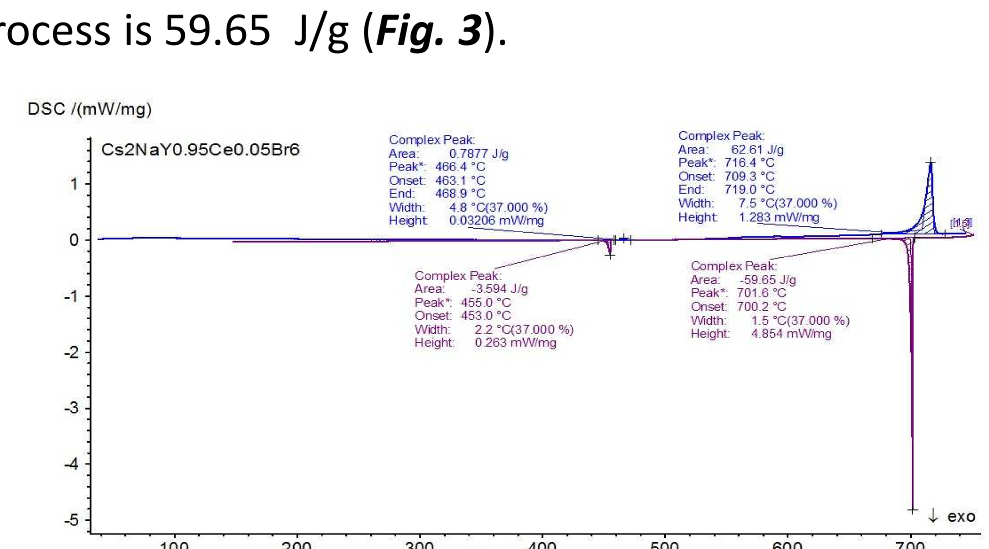

Fig. 3. The DSC measurement for the cerium doped Cs_2NaYBr_6. Data were collected under flowing argon with a heat and cooling rate of 5°C/min.

Optical Characterization

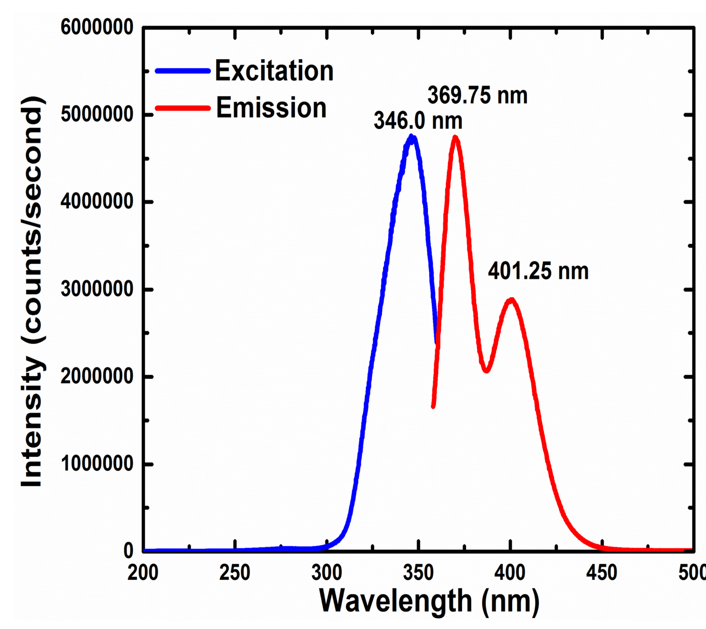

Fig. 4. Photoluminescence for the cerium doped Cs_2NaYBr_6.

Excitation and emission spectra reveal a relatively small 23.75nm Stokes shift (***Fig. 4***). The small Stoke shift could potentially cause self-absorption in large crystals, which makes them more vulnerable to temperature change as strong self-absorption at high temperatures can diminish the scintillator performance. The double emission peaks on the right are characteristics of the 5d to 4f transition for Ce^{3+} activator. The cerium doped Cs_2NaYBr_6 crystal has a quantum yield of 41.7%.

Photoluminescence decay was measured by a time-correlated-single photon counting technique, using a pulsed 340 nm LED as the light source. The optical decay can be best fitted by two exponentially decay components (***Fig. 5***). The first and second lifetimes are 31.46 and 20.46 ns, respectively. While the first, 31.46 ns, component dominates the decay behavior (71.12%) the second component occupied almost three tenths of the decay population (28.88%).

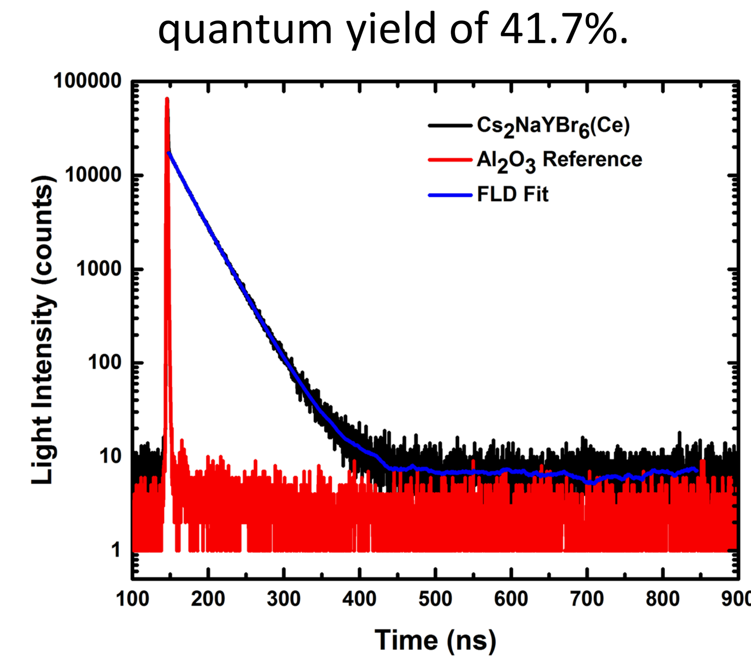

Fig. 5 Photoluminescence decay measurement for the cerium doped Cs_2NaYBr_6.

There is no evidence to suggest the presence of afterglow for this new compound as all decay components are within 50 ns. With a short photoluminescence lifetime, this new material is promising to have a fast response time when excited with gamma rays.

Scintillation Properties

Radioluminescence decay measured by a coincidence technique was fitted with two components.(***Fig. 6***). The first component is 43.95 ns which makes up most of the decay events (99.98%), while the second component is much slower with a decay time of 537.7 ns which only made up 0.02% of the decay events. The cause of this long lasting afterglow is still not clear and will be studied in the future.

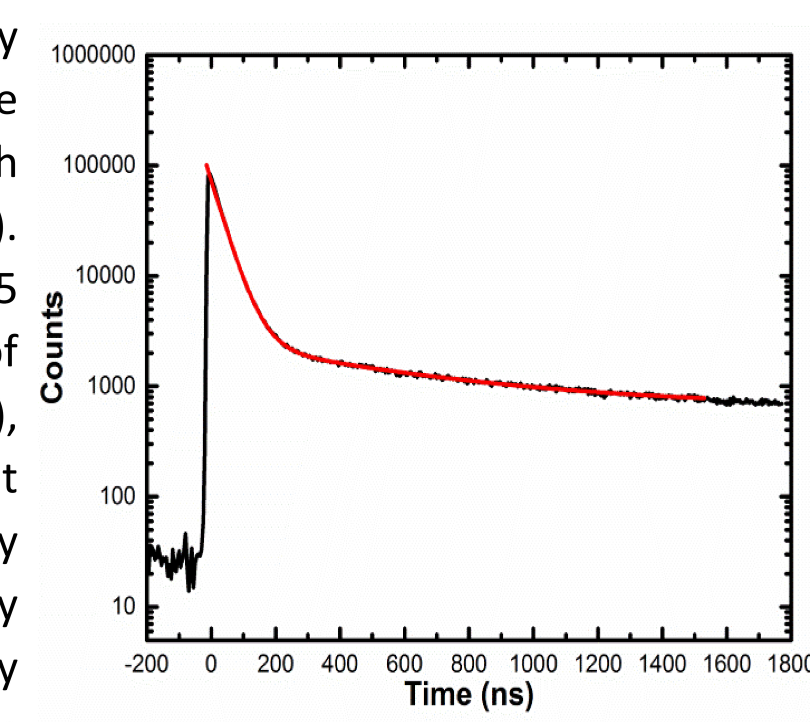

Fig. 6. Radioluminecence decay measurement for the cerium doped Cs_2NaYBr_6. Cs-137 is used for the coincidence measurement. (Chi^2 = 0.9835).

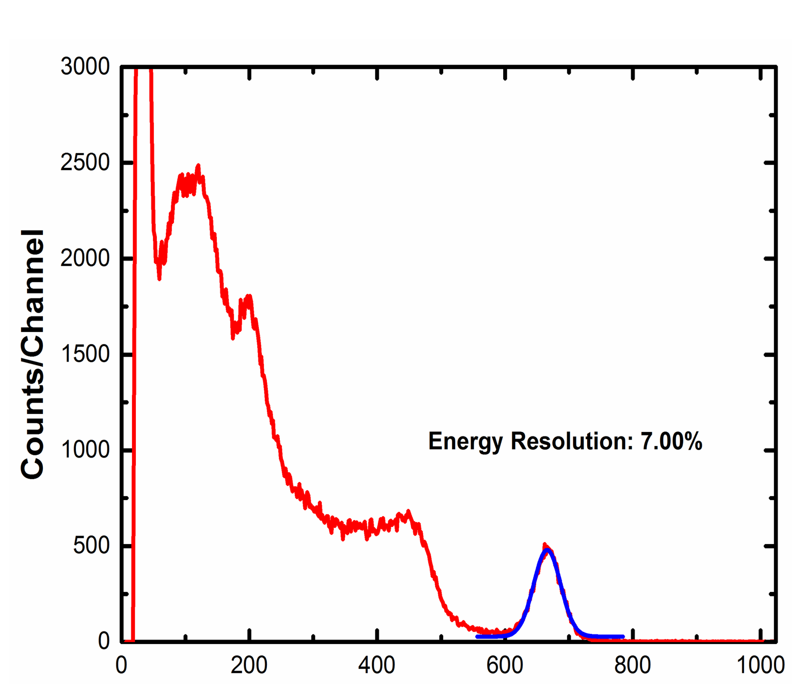

Fig. 7. Pulse Height Spectra for the cerium doped Cs_2NaYBr_6. Sample was irradiated by a Cs-137 source.

The total energy peak of the Cs-137 gamma ray was fitted with a Gaussian distribution (***Fig. 7***). An energy resolution value of ~7% is estimated for this new compound, which is compatible to the NaI(Tl) and better than CsI(Tl) and BGO scintillators.

Scintillation Properties Cont.

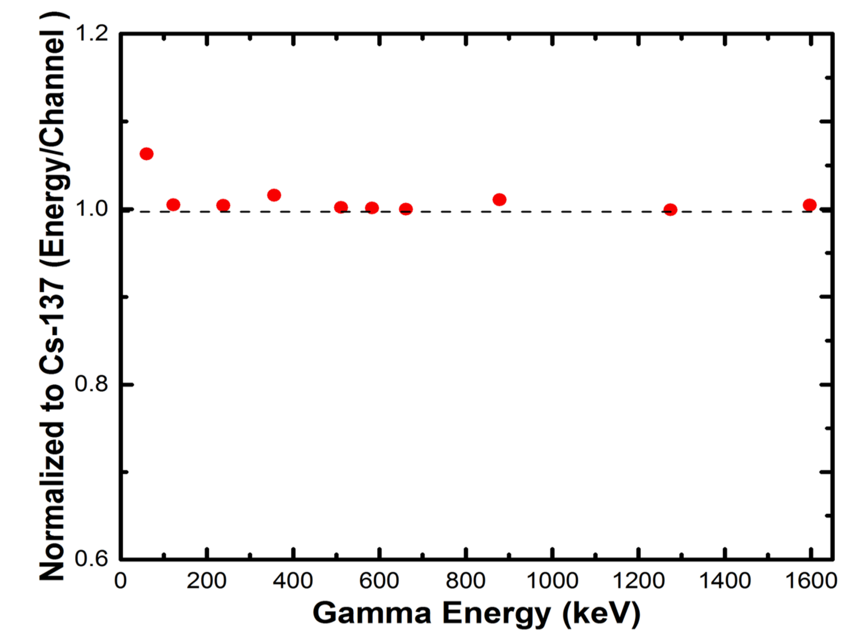

Fig. 8. Linearity measurement for the cerium doped Cs_2NaYBr_6. Data were collected from different isotopes and normalized to Cs-137 .

This new compound exhibits good proportionality at high energy range (>500 keV) and starts to loose linearity at low energies (***Fig. 8***). A good proportional response is a general trait for the elpasolite halide crystals. A linearity of 6.0% is observed at the low energy range for this crystal, which is better than many well established scintillators such as NaI:Tl (14%), CsI:Tl (10%), and LSO (11%). The linearity response can be further improved with a higher quality crystal through a refined growth process and the arts of detector package techniques.

Conclusion

A cerium doped Cs_2NaYBr_6 crystals was grown using a vertical Bridgeman technique for optical and radioluminescent characterization. Optical excitation and emission spectra indicate that cerium is an effective activator for this new compound. In addition, cerium produces a fast response time for optical and radioluminescence responses. The energy resolution at 662 keV for this crystal is ~7%, with a satisfactory linearity response in the 62 keV to 1597 keV energy range. These preliminary results suggest that with improved crystal quality this new compound is promising for gamma ray spectroscopy applications.

Acknowledgements

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