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Paul Guss

Remote Sensing Laboratory, gusspp@nv.doe.gov

Ronald Guise

Remote Sensing Laboratory

Ding Yuan

National Security Technologies, LLC

Sanjoy Mukhopadhyay

Remote Sensing Laboratory

Robert O'Brien

University of Nevada, Las Vegas

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Authors

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Lanthanum halide nanoparticle scintillators for nuclear radiation detection

Paul Guss,^{1,a)} Ronald Guise,¹ Ding Yuan,² Sanjoy Mukhopadhyay,³ Robert O'Brien,⁴ Daniel Lowe,⁴ Zhitao Kang,⁵ Hisham Menkara,⁵ and Vivek V. Nagarkar⁶

¹Remote Sensing Laboratory, P.O. Box 98521, M/S RSL-48, Las Vegas, Nevada 89193, USA

²National Security Technologies, LLC, Los Alamos Operations, P.O. Box 809, M/S LAO/C320, Los Alamos, New Mexico 87544, USA

³Remote Sensing Laboratory—Andrews, Building 1783, Arnold Avenue Andrews AFB, Maryland 20762, USA

⁴University of Nevada, Las Vegas, 4505 S. Maryland Parkway, Las Vegas, Nevada 89154, USA

⁵Georgia Tech Research Institute, 925 Dalney St., Atlanta, Georgia 30332, USA

⁶RMD, Inc., 44 Hunt Street, Watertown, Massachusetts 02472, USA

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Nanoparticles with sizes <10 nm were fabricated and characterized for their nanocomposite radiation detector properties. This work investigated the properties of several nanostructured radiation scintillators, in order to determine the viability of using scintillators employing nanostructured lanthanum trifluoride. Preliminary results of this investigation are consistent with the idea that these materials have an intrinsic response to nuclear radiation that may be correlated to the energy of the incident radiation. © 2013 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4790867>]

I. INTRODUCTION

Scintillator materials are used to detect and, in some cases, quantify a gamma-ray's energy. Higher resolution scintillators are expensive, difficult to manufacture, and fragile. Low to moderate resolution scintillators are less costly, easier to manufacture, and more rugged, but they offer lower performance envelopes when compared to high resolution materials. At issue is whether the desirable qualities of each scintillator type can be combined to achieve high performance at low cost.

Recent studies suggest that nanocomposites may enable the use of scintillator materials such as cerium-doped lanthanum trifluoride ($\text{LaF}_3:\text{Ce}$) and cerium tribromide (CeBr_3) without requiring the growth of large crystals.¹ Nanostructured detectors may allow us to engineer immensely sized detectors of flexible form factors that will have a broad energy range and an energy resolution sufficient to perform isotopic identification. Furthermore, nanocomposites are easy to prepare and very low in cost. It is much less costly to use nanocomposites rather than grow large whole crystals of scintillator materials; with nanocomposites fabricated on an industrial scale, costs are even less. Nanostructured radiation scintillator detectors may improve quantum efficiency and provide vastly improved detector form factors. Quantum efficiencies, up to 60%, have been seen in photoluminescence from silicon nanocrystals in a densely packed ensemble.²

Further, nanocomposite detectors may offer an avenue to combine the advantages of both types of scintillator materials to overcome the disadvantages of each. It is hypothesized that “nanocrystals,” 2–5 nm in diameter, of certain inorganic scintillator materials, packed densely in plastic or inorganic solutions, can capture most of the x-ray and gamma-ray energies, thus offering nearly the performance of large crystals. The resulting mixture would also have the desirable features

of plastic or liquid scintillators.^{3,4} For instance, the chemical synthesis of the cerium-doped lanthanum halide nanoparticles is scalable,³ and large quantities of material can be produced at a time, unlike typical crystal sizes resulting from crystal growth processes such as the Bridgman process.

In order to create a new class of scintillator materials that combines good energy resolution, large size, and low cost, Del Sesto developed a large-scale synthesis of narrowly sized, distributed, <10 nm $\text{LaF}_3:\text{Ce}$ nanoparticles.³ In fact, nanoparticles of many candidate scintillating materials, such as $\text{LaF}_3:\text{Ce}$, or even CeBr_3 , can all be cast into transparent oleic acid or polymer composites with up to 60% scintillator volume loading. Preliminary experiments showed that the $\text{LaF}_3:\text{Ce}$ oleic acid-based nanocomposites exhibit a photopeak when exposed to ^{137}Cs source gamma radiation.³ In general, lanthanum halides show the promise of being useful over a wide energy range of x-rays and gamma rays. In fact, another motivation for this work was to investigate whether or not these nanostructured radiation scintillators may extend the gamma energy response on both the low- and the high-energy regimes. If true, this opens the prospect that x-rays and relatively high-energy activation prompt gamma rays may be simultaneously detected using one detector using nanocrystals such as nanostructured lanthanum tribromide, lanthanum trifluoride, or cerium tribromide.

II. EXPERIMENT

Nanostructured radiation scintillator detectors may lead to techniques to improve quantum efficiency and exploit vastly improved detector form factors compared to currently used inorganic scintillators. Nanocomposites can dramatically change the size, shape, and luminosity of materials and, by being easily accessible to coupling to photodiodes, can offer higher quantum efficiency for detecting light quanta. This compares to photomultiplier tubes (PMTs) that suffer from low quantum efficiency (1%–10%). In our initial investigation, nanoparticles of lanthanum halides and cerium

^{a)}Author to whom correspondence should be addressed. Electronic mail: gusspp@nv.doe.gov.

tribromide were suspended in oleic acid. It is anticipated that when a gamma ray strikes this material, its energy is absorbed by both the nanocrystals and the oleic acid, raising some atoms to a higher energy level. These atoms de-excite and give off their energy as optical photons in the visible and near-visible regions of the electromagnetic spectrum. A phototransducer, either a photomultiplier tube or an avalanche photodiode, can be employed to amplify and to convert the collected energies into electrical pulses.⁴ The number of optical photons generated is correlated to the energy level and intensity of the photon striking the material. A multichannel analyzer counts the optical photons, determines the energy level of the photon striking the material, and increases the count of photons of that energy level by one, ultimately creating a gamma-ray spectrum.

Nanoparticles with sizes <10 nm were fabricated, two scintillating detectors employing these nanoparticles were prepared, and the process of characterization of their optical, physical, and radiation detector properties was performed. The crystals used in this work were $\text{LaF}_3\text{:Ce}$ nanoparticles mixed with oleic acid. The experimental materials used and compared in the study are listed in Table I.

The modified oleic acid is a scintillator material, so it increases the amount of energy converted to a detectable signal. A polymer composite produced by Radiation Monitoring Devices, Inc. (RMD), was used to make initial measurements of the 5% $\text{LaF}_3\text{:Ce}$ -loaded and 25% $\text{LaF}_3\text{:Ce}$ -loaded samples shown in Figure 1.

The detectors for interfacing to the phototube and electronics were prepared as shown in Figure 2. For the measurements, a special transparent cup was employed to contain the nanoparticle and oleic acid mixture. Nanoparticle detector volume was ~ 2.5 ml with 0.5 in. diameter $\times$ 0.75 in. length. It was set upon a Hamamatsu R647 PMT, using a Hamamatsu E849-36 PMT base in conjunction with an ORTEC 456 High Voltage Power Supply. An ORTEC 450 Research Amplifier was employed for signal processing and a PC-104 based MCA was used to acquire spectra. A control detector made up of BC400 with 0.5 in. diameter $\times$ 0.75 in. length was used to obtain baseline data and establish stabilization and normalization information for the system.

A data acquisition test protocol was established, which included acquiring a spectra for each detector for laboratory background, ^{22}Na (3.22 μCi), ^{60}Co (3.78 μCi), ^{137}Cs (31.9 μCi), ^{241}Am (9.09 μCi), and ^{252}Cf (5.03 μCi). The ^{252}Cf data were acquired both with and without 1 in. lead shielding and with

FIG. 1. Detectors were prepared by RMD with upper and lower detectors containing 5% $\text{LaF}_3\text{:Ce}$ and 25% $\text{LaF}_3\text{:Ce}$ nanoparticle loading by mass, respectively.

and without several inches of paraffin shielding. The data were acquired, typically for 1000 s with the source at a distance of 8 cm from the detectors. Two source distances were used for the ^{137}Cs source, namely 8 cm and 16 cm. Because of the different shielding considerations for the ^{252}Cf , a constant distance of 18 cm was used for the acquisition of all data using this specific source. The initial results are reported here and are compared to both the modeling data and the data reported by Del Sesto.³

III. RESULTS

Figure 3 shows the gamma spectral data with the background subtracted from the first measurements made with 25% $\text{LaF}_3\text{:Ce}$ -loaded samples using the ^{241}Am (3 a), ^{22}Na (3 b), ^{137}Cs (3 c), and ^{60}Co (3 d) sources (ordered by lowest to highest photopeak energy). Figure 4 illustrates a slight energy dependence for the 25% $\text{LaF}_3\text{:Ce}$ detector, based on the normalized spectra acquired with the ^{137}Cs and ^{60}Co sources. The ^{252}Cf data (not shown) also indicated a good neutron response by these detectors.

Early studies suggest that nanocomposite scintillator material may even be able to discriminate between neutrons and gamma rays. Some simulations supported this possibility.⁴ Los Alamos National Laboratory (LANL) investigators assert that basic physics calculations also support this possibility.⁴ For instance, when neutrons interact with the hydrogenous material, free protons are generated. When gamma rays interact with the material, free electrons are generated. The energy signatures for the two processes are different in magnitude and in time. The experiments performed in this investigation confirmed this possibility and demonstrated the ability of the material to detect neutrons and to differentiate them from gamma rays.

Early in this work, the data acquired was benchmarked using a variety of different parameters, such as detector-to-source distances, source strengths, source types, and geometries. At this point, there is sufficient confidence in the predictive capacity of the models that they may be used for guidance in fine-tuning parameters for future nanoparticle detector developments. The Monte Carlo N-Particle eXtended⁵ (MCNPX) models were started by simulating the

TABLE I. Investigated nanoparticle detectors.

Detector	Specifications	Known properties
BC400	1 cc plastic scintillator, $\sim 65\%$ anthracene	$\lambda_{\text{max}} \cong 423$ nm
5% $\text{LaF}_3\text{:Ce}$	1 ml oleic acid with 5% by mass $\text{LaF}_3\text{:Ce}$ nanoparticles <10 nm diameter	Detector nanoparticles with diameter <10 nm $\lambda_{\text{max}} \cong 380$ nm
25% $\text{LaF}_3\text{:Ce}$	1 ml oleic acid with 25% by mass $\text{LaF}_3\text{:Ce}$ nanoparticles <10 nm diameter	Detector nanoparticles with diameter <10 nm $\lambda_{\text{max}} \cong 380$ nm

FIG. 2. Detector setup for studying the nanocomposite detector is shown. (Left) The oscilloscope, power supply, and electronics are shown in the upper part of the photo, while the (liquid) detector assembly is shown below, resting on top of the photomultiplier tube, wrapped in black tape to make the assembly light tight. Signal and high-voltage cables are connected to the detector. (Right) The detector assembly is to the right of the ^{60}Co source.

FIG. 3. These are the first background-subtracted spectra for the 25% $\text{LaF}_3\text{:Ce}$ nanocomposite detector. Significantly, they not only demonstrate an excess in counts when background is removed but in addition they demonstrate a different spectral shape for each different individual radiation source, which holds open the promise of performing some level of spectral discrimination and isotopic identification with nanocomposites.

FIG. 4. Background-subtracted normalized measurement of a different response for ^{60}Co and ^{137}Cs sources using the 25% $\text{LaF}_3\text{:Ce}$ detector, which appears even more clearly, when this difference is measured for a BC-400 detector. A slight energy dependence for the 25% $\text{LaF}_3\text{:Ce}$ detector is evident.

LANL 8% $\text{LaF}_3\text{:Ce}$ nanoparticle systems, which have indicated the potential for detection of ^{137}Cs .³

Initial measurements of an energy spectrum³ for the unloaded and 8% $\text{LaF}_3\text{:Ce}$ -loaded samples were first acquired by the LANL group, using ^{137}Cs sources. Their measurements show the expected Compton edge in the polymer-only sample, and the Compton edge and photopeak expected in the nanophosphor composites that LANL produced.³ Their data were used as a check on the MCNPX model (Figure 5) we used to model our detectors.

Measurements with the 5% and 25% $\text{LaF}_3\text{:Ce}$ loaded samples shown in Figure 1 have been made using ^{137}Cs sources. Figure 5(a) shows the energy spectra. The blue (solid) plot is the measured 5% $\text{LaF}_3\text{:Ce}$ spectrum and the black (dotted) plot is the spectrum from the 25% $\text{LaF}_3\text{:Ce}$ nanocomposite scintillator. These measurements show the expected Compton edge, while both the Compton edge and photopeak appeared in the LANL spectra for the nanophosphor composites that LANL has produced.³ In Figure 5(b), a preliminary MCNPX calculation compares spectra for both a 5% and a 25% $\text{LaF}_3\text{:Ce}$ loaded sample. The blue (solid) plot is an energy spectrum calculated for 5% $\text{LaF}_3\text{:Ce}$, and the black (dotted) plot is the energy spectrum calculated for 25% $\text{LaF}_3\text{:Ce}$ polymer nanocomposite exposed to ^{137}Cs (100- μCi) γ irradiation. As the development of this material continues, the energy resolution is

FIG. 5. (a) Energy spectrum for ^{137}Cs (31.9- μCi) gamma irradiation of both 5% and 25% $\text{LaF}_3\text{:Ce}$ (10 nm spheres) polymer nanocomposite.¹ The blue (solid) line is the measured 5% $\text{LaF}_3\text{:Ce}$ spectrum and the black (dotted) line is the 25% $\text{LaF}_3\text{:Ce}$ spectrum. ^{137}Cs Compton edge and scattering is evident. (b) MCNPX model prediction show good agreement between both 5% and 25% $\text{LaF}_3\text{:Ce}$ loaded samples. The blue (solid) line is the measured 5% $\text{LaF}_3\text{:Ce}$ spectrum and the black (dotted) line is the 25% $\text{LaF}_3\text{:Ce}$ spectrum when the detector is exposed to ^{137}Cs (100- μCi) γ irradiation.

expected to improve and the photopeak-to-Compton ratio will become greater at higher loadings.

From the simulation results shown in Figure 5(b), an extrapolation is made using MCNPX for an 86% by mass nanoparticle content of $\text{LaF}_3\text{:Ce}$. Two methods of calculation are compared, and results are shown for $\text{LaF}_3\text{:Ce}$ in Figure 6. In one calculation, the atoms in the detector are distributed homogeneously, and the other calculation is performed with nano structures containing the $\text{LaF}_3\text{:Ce}$. This was done for the various materials (CdSe and $\text{LaF}_3\text{:Ce}$). These calculations indicated that the ratio between the photopeak and the Compton edge changes with material type (ratio is ~ 4 for $\text{LaF}_3\text{:Ce}$ but is ~ 3 for CdSe). The calculation made using actual nanoparticles of $\text{LaF}_3\text{:Ce}$ usually resulted in a closer replication of our actual data, which indicates the importance of the approach used in setting up the geometry for nanocomposite systems when modeling with MCNPX.

IV. ANALYSIS

For an ideal scintillator, the energy resolution, R , is given by

$$R = \left[\frac{\Delta E}{E} \right]_{FWHM} = 2.35 \times \sqrt{\frac{(1 + v(M))}{(N \times \epsilon_t)}}, \quad (1)$$

where N is the average number of photons generated at a given energy E , $v(M)$ is the variance in the multiplication factor of the PMT (for a typical 10-stage PMT with a gain of 2×10^6 , $v(M)$ is approximately 0.08), and ϵ_t is the average transport efficiency.⁶ $\Delta E/E$ is measured experimentally, $v(M)$ is a known constant, and combined histories and transport simulations can provide an estimate of ϵ_t . The only unknown parameter then is N , the number of photons generated in the material under an incident gamma energy. Létant and Wang⁶ determined a value of the average transport efficiency ϵ_t of 0.063 ± 0.002 for their cadmium selenide nanoparticle with zinc sulfide shell (CdSe/ZnS core shell quantum dot) nanocomposite detector. By inserting ϵ_t in the energy resolution formula discussed above, the average photon output of the quantum dot–glass material that Létant and Wang studied under 59-keV gamma-ray irradiation was estimated to be close to 4210 (while the number of photons generated by a sodium iodide [NaI:Tl] crystal under the same conditions was only 2600). For higher energies, the projected light output would be about 70 600 photons at 1 MeV, assuming the quantum dot medium has a linear response, which is a factor 1.75 better than the NaI:Tl crystal for which N is typically 40 000.⁶ However, the data for our initial measurements reported in Table II are consistent with a lower efficiency and a lower light output typical of nanoparticle detectors that is not seen in BC-400 detectors. This may be due to the combination of the self-shielding effect contributed by the inorganic solution and the high concentration of the suspended nanocrystals. All of the other particulars depicted in Table II, i.e., scaling with particle density and with source strength, are consistent with expectation.

V. DISCUSSION

The work by Walters,² suggests an intriguing aspect of nanocomposites. Walters examines the photoluminescence decay rates for dense silicon nanocrystal ensembles in this work. From a comparison of the experimental photoluminescence decay rates to the expected spontaneous emission rate,

FIG. 6. MCNPX model of the energy spectrum for ^{137}Cs gamma irradiation of a 86% $\text{LaF}_3\text{:Ce}$ (10 nm spheres) polymer nanocomposite, for calculated assuming both the case of an homogeneous mix of the atoms in the detector and for the case that the $\text{LaF}_3\text{:Ce}$ crystals are distributed in spherical nanostructures.

TABLE II. Count rate for 25% LaF₃:Ce, 5% LaF₃:Ce, and BC400 detectors.

Background subtracted count rate versus source (cps)				
Source	Intensity (μ Ci)	25% LaF ₃ :Ce (cps)	5%LaF ₃ :Ce (cps)	BC400 (cps)
Background		4.7	2.6	1
²² Na (8 cm)	3.22	2.5	1.8	23.3
⁶⁰ Co (8 cm)	3.78	11.2	7.9	34.4
¹³⁷ Cs (8 cm)	31.9	10.4	7	193.6
¹³⁷ Cs (16 cm)	31.9	5.3	4.1	52.95
²⁴¹ Am (8 cm)	9.09	0.07	...	...
²⁵² Cf (18 cm)	5.03	2.6	1.7	0.6

an internal photoluminescence quantum efficiency as high as $59\% \pm 9\%$ is found for such ensembles. Taken together with the work of Létant and Wang,⁶ this suggests nanocomposites have a potential for high quantum efficiency and good energy resolution for gamma-ray spectroscopy.²

As the development of this material continues, the energy resolution is expected to improve and the photopeak-to-Compton ratio may become greater at higher loadings. These measurements indicate that the expected Compton edge and photopeak may be present in the new nanophosphor composites that RMD will produce. For example, using a porous VYCOR[®] (Ref. 7) with CdSe/ZnS core shell quantum dots, Létant has obtained signatures of the ²⁴¹Am photopeak with energy resolution as good as thallium-doped sodium iodide.⁶ Létant even reported a factor 2 improvement in energy resolution of an un-optimized nanoporous glass-quantum dot composite material over a standard NaI crystal, using the 59-keV line of an Americium gamma source. Létant's detectors were small but had a very impressive alpha response.⁶ Additionally, McKigney⁸ fabricated a nanocomposite scintillator comprised of cerium doped lanthanum trifluoride (LaF₃:Ce) embedded in an organic matrix material. McKigney measured photopeaks for ²⁴¹Am and ⁵⁷Co. His photopeak measurements demonstrate the proof-of-principle of the nanocomposite scintillator concept. McKigney⁸ concludes the right next step is to disperse the nanoparticle in a scintillating matrix material, such as standard plastic scintillator. McKigney⁸ suggests the synthesis of nanophosphors of brighter scintillators and fabrication of these into a composite. These results are consistent that some limited spectroscopy is possible using nanocomposites, particularly if combined with clever algorithm schemes such as implemented by Symetrica.^{9–15}

VI. CONCLUSION

Two nanocomposite detectors, one of 5% LaF₃:Ce nanoparticle loading by mass and one of 25% LaF₃:Ce nanoparticle loading by mass were prepared. The first results of the detectors' response using these specific detector materials were compared to models and to prior nanocomposite detector data. In general, the agreement between data and models was good. It was determined that these nanocomposites' response to radiation is significant. Nanocomposites are sensitive to both neutrons and photons (γ - and X-rays). The first production nanocomposite detectors prepared for this work are weaker in terms of energy response than BC400 and

have yet to match the performance of plastic detectors. The 25% LaF₃:Ce nanoparticle loading by mass detector had better performance compared to the 5% LaF₃:Ce nanoparticle loading by mass detector, yet still roughly less than half the sensitivity (efficiency) of the BC400 detector of comparable size. It was also shown that in order to produce the most accurate models of detector performance, accurate definition of the nanostructure geometry is helpful. Key points are that nanocomposite detectors are cheap, easy to fabricate, and respond to nuclear radiation. More work is required, however, to determine how best to obtain isotopic correlations and to optimize light yield.

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