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MATERIALS RESEARCH HIGHLIGHT

Development of nanocomposite scintillators

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New scintillator materials are in high demand to assist in non-proliferation and counter-proliferation. One application for these materials includes the protection of borders and ports from the introduction of nuclear materials. In order to create a new class of scintillator materials that combines good energy resolution, large size and low cost, we have developed a large-scale synthesis of narrowly size-distributed <10nm cerium-doped lanthanum halide nanoparticles, $Ce:LaX_3$, where X = F or Br. $Ce:LaBr_3$ is one of the brightest known scintillator materials. The synthesis of the cerium-doped nanoparticles is scalable to large quantities, and the particles can be cast into transparent oleic acid or polymer composites with up to 60% scintillator volume loading. Preliminary experiments show that the $Ce:LaF_3$ oleic acid based nanocomposites exhibit a photopeak when exposed to ^{137}Cs source γ-radiation.

Background

Radiation detecting materials are necessary in many applications of modern technology, ranging from homeland security to more efficient and precise radiographical imaging methods. Modern-day x-ray or γ radiation detectors typically consist of single-crystal materials, which are either restricted in size due to production technique limitations or which need to be cooled to liquid-nitrogen temperatures to achieve high energy resolutions, as with high-purity germanium (HPGe).

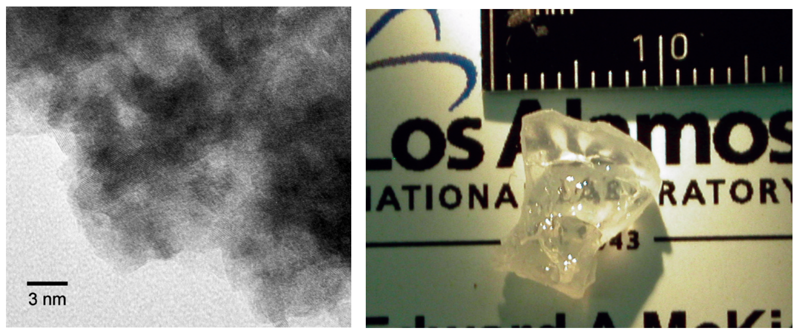

Figure 1. Transmission electron microscopy image of the $Ce:LaF_3$ particles (left) and a 1cm thick oleic-based nancomposite of $Ce:LaF_3$ with nearly 30% volume loading of the phosphor.

New materials which exhibit the energy resolution between that of Tl:NaI, a widely used scintillator, and HPGe, which operate at ambient temperatures, and which could be cast into a variety of shapes and sizes would advance the area of radiation detection and imaging.

An important approach to developing such detector materials that bypasses the limitations of single crystals is using composites of nanoscale materials. Utilizing nanocrystals of known scintillator and detector materials will allow for the production of large composites of the nanocrystals with an expanded variety of attainable shapes and sizes. Preliminary measurements indicate that nanophoshpor materials may have higher energy resolution than their respective bulk single crystal materials due to greater light output of nanocrystals, which translates to greater scintillation efficiency.[1]

The known scintillators of cerium-doped lanthanum halides, $Ce:LaX_3$, where X = F or Br, were chosen for development into nanocomposites. $Ce:LaBr_3$ is one of the brightest scintillators known[2] and the short lifetime (about 30ns) is advantageous in materials for fast and efficient detection of gamma-ray radiation.

Synthesis of nanoparticles and nanocomposites

Nanoparticles of the $Ce:LaF_3$ scintillator are easily synthesized via precipitation reaction in an aqueous ethanol solution in the presence of oleic acid. The resulting nanoparticles have low dispersity and are <10nm in diameter, as seen in the TEM image in Figure 1. The oleic acid can act as a matrix for the $Ce:LaF_3$. The result is a gel-like composite containing up to 60% by volume of the $Ce:LaF_3$ particles. Some samples maintain transparency in up to 1cm thick gels, as seen in one example in Figure 1. The emission of the $Ce:LaF_3$ oleic nanocomposite is shifted approximately 20nm as compared to single crystalline $Ce:LaF_3$, as predicted for nanoscale crystals.

The $Ce:LaF_3$ particles have also been embedded into a polymer composite loaded at 8% and 15% by weight. The polymer composites are slightly opaque and we are in the process of modifying the composites to increase this transparency.

Initial measurements of the unloaded and 8% $Ce:LaF_3$ loaded sample shown above have been made using ^{137}Cs sources. These measurements show the expected Compton edge in the polymer only sample,

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and the Compton edge and photo-peak expected in the nanophosphor composite. Figure 3 shows an energy spectrum. The red plot is the measured polymer-only spectrum and the black plot is the spectrum from the nanocomposite scintillator. As the development of this material continues the energy resolution is expected to improve and the photo-peak-to-Compton ratio will become greater at higher loadings.

Utilizing precipitation methods in an ionic liquid solvent rather than an aqueous solvent nanoparticles of Ce:$LaBr_3$ have also been synthesized. The particles are < 10nm in size, and fairly monodisperse. The TEM image in Figure 4a clearly shows resolved lattice spacings in individual nanocrystals. Emission from the cerium in the nanoparticles is again Stokes shifted by about 10nm relative to bulk single crystals, as seen also in Figure 4b.

We are currently in the process of scaling up the synthesis of the Ce:LaF_3 nanoparticles to near-kilogram quantities per reaction. Development of polymer-based nanocomposites is being optimized to increase volume loadings of the nanophosphor while maintaining transparency in composites greater than 1cm thick. Additionally, a more efficient route using conventional solvents is being pursued to synthesize the Ce:$LaBr_3$ nanophophors with smaller particle sizes and higher yields.

Acknowledgement

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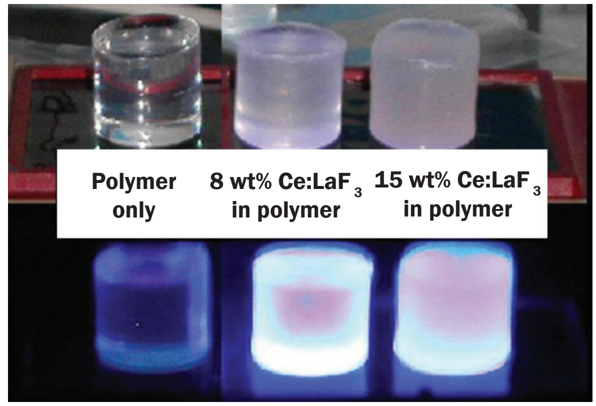

Figure 2. Polymer with Ce:LaF_3 nanoparticles at 8% and 15% by weight (top), and under UV-light excitation (bottom).

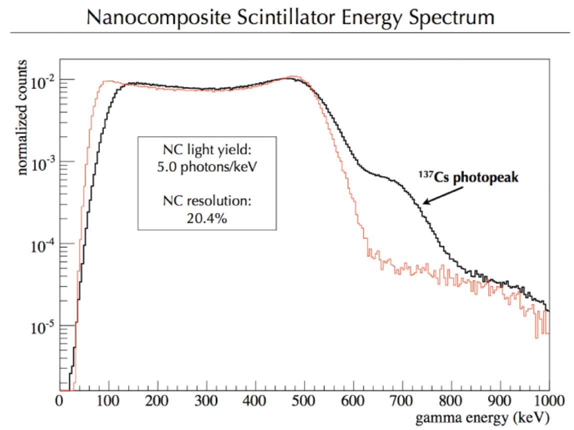

Figure 3. Energy spectrum of polymer only, and 8% CeF_3: polymer nanocomposite exposed to ^{137}Cs γ irradiation.

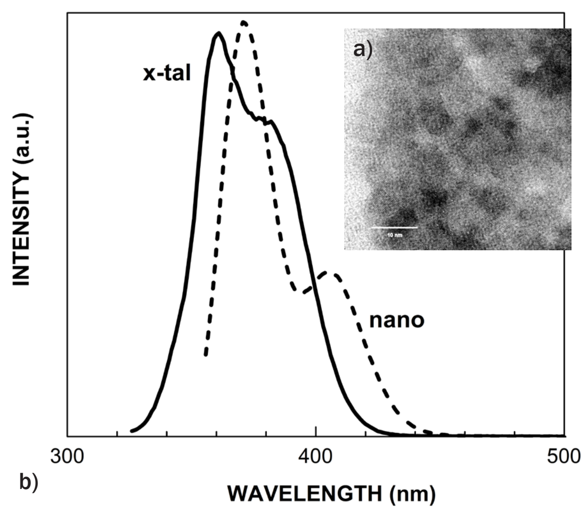

Figure 4. a, inset) TEM image of Ce:$LaBr_3$ nanocrystals, and b) emission spectrum of the nanocrystals compared to bulk single crystals.