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Development of improved radiation detector materials

Tailored processing new materials aimed at threat reduction, nonproliferation applications

Threat reduction and nuclear nonproliferation activities urgently require improved radiation detectors. The performance of these detectors can be significantly enhanced if the materials currently entrusted with the detection of radiation are replaced with optimized materials. The Materials Science and Technology (MST) and Materials Physics and Applications (MPA) Divisions at Los Alamos National Laboratory have unique expertise to provide this requisite optimization of radiation detector materials. Historically, MST has provided detector single crystals in specific geometries for hydrodynamic and subcritical testing. However, new capabilities expand the portfolio to encompass a variety of material forms (*e.g.* single crystals, transparent polycrystals and nanocomposite scintillators) as well as integrated methodologies to search for new compositions. This portfolio allows MST and MPA to combine an efficient search for and optimization of scintillator detector materials with a conversion of these compositions to innovate formats, which are appropriate for a specific application. By integrating the various *composition* and *form* activities, MST and MPA provide a new approach to detector materials development—an approach which is less reliant on intuitive trial and error.

Material form

Single crystal

The Structure/Property Relations Group (MST-8) is home to a robust single crystal growth facility, one with a capability unique in the United States to perform "traveling solvent" growth using optical float zone (TS-OFZ) and Czochralski (TS-CZ) methods. The traveling solvent method allows for the synthesis of compounds that cannot be grown via conventional single crystal methods. Many candidate detector materials fall under this category. The TS-OFZ method typically yields crystals in ~1-2 days while the TS-CZ method typically requires ~1-2 weeks (as TS-CZ crystals are much larger; see example above). In order to efficiently integrate a variety of developmental components into a coherent composition optimization process, research and development scale crystals are typically grown via the TS-OFZ method. However, as candidate detector materials are optimized, device scale crystals can be grown via the TS-CZ method. The Single Crystal Growth Laboratory (SCGL) has two each of the TS-OFZ and TS-CZ units with precision atmosphere control. Furnaces are also set up to perform crystal growths via the Bridgman and micropulling down methods. The micropulling down method allows for the growth of shaped crystals. Additionally, the SCGL has the capability to synthesize materials in inert atmospheres. Finally, in addition to growth, the SCGL houses equipment to fabricate detector materials into specific geometries, such as plates or pixels.

A Czochralski single crystal boule grown in MST-8's Single Crystal Growth Laboratory.

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Transparent polycrystalline ceramics

Single crystal growth is often expensive and the size of high-quality crystals is limited. If size or cost are more important than energy resolution for a particular special nuclear material detector application, then an alternative processing route to single crystal growth is needed. One such alternative is transparent polycrystalline ceramic scintillators, which can be processed from conventional ceramic forming techniques. These materials can therefore be produced in large-size scintillator panels with lower cost and high production rate. The Materials Technology-Metallurgy Group (MST-6) has successfully developed a series of transparent polycrystalline ceramics such as AlON, $MgAl_2O_4$, and Al_2O_3. Applying the success with transparent polycrystalline AlON, MST-6 researchers have demonstrated fabrication of polycrystalline $Lu_3Al_5O_{12}$:Ce scintillators and extended this polycrystalline ceramic technique to further develop an innovative nanosized grain polycrystalline $LaBr_3$:Ce. Polycrystalline scintillators have a much higher mechanical strength compared to single crystals and can therefore better withstand harsh environments. In addition, ceramic processing can provide a more uniform distribution of the activators and co-dopant. The challenge of polycrystalline ceramic scintillators is achieving transparency. However, recent synthesis developments by MST-6 researchers have helped make ceramic scintillators a viable detector material form.

Examples of transparent aluminum oxynitride (AlON) polycrystalline ceramics fabricated by MST-6 researchers.

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Nanocomposites

Another alternative approach to the development of radiation detector materials that bypasses some of the limitations of single crystals is to use composites of nanoscale materials. Utilizing nanophosphors and semiconductor quantum dots will allow production of large polymer or glass composites of the nanocrystals with an expanded variety of shapes and sizes attainable, as well as with tunable properties. Several groups, including Materials Chemistry (MPA-MC) and MST-8, are involved in various aspects of this research and development, which ranges from the design, synthesis and development of the nanocomposites, to design and fabrications of real-world devices and applications.

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New compositions

Rapid prototyping

MPA researchers have employed a rapid discovery method for the development of new radiation detector materials. Utilizing polymer-assisted-deposition techniques and automated procedures for the preparation of epitaxial films, high volumes of new compositions can be prepared and analyzed. The materials are prepared on single crystal substrates to ensure the formation of crystalline materials. The image at right shows a scintillator identified using the Los Alamos National Laboratory rapid through put materials discovery program.

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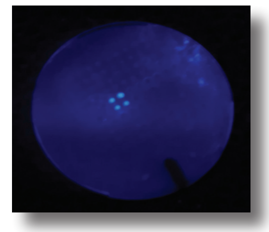

Candidate scintillator compounds synthesized via the rapid prototyping method.

Atomistic simulation

Currently it is not possible to predict the efficacy of particular scintillator compositions by using state-of-the-art simulation techniques alone. In an ideal situation new compositions would be identified theoretically, thereby reducing the necessity of expensive and time consuming experimental trials. However, MST-8 researchers have taken steps toward theoretical scintillator identification by using simulation techniques to identify atomic-scale phenomena that limit scintillator performance. In most cases, this modeling has been used to identify performance limiting defects. However, in other cases, new compositional regions of interest have been identified. An advantage of this type of modeling is that it can be used to simultaneously consider a wide range of compositions (see figure at right). For the greatest benefit, these simulations can be coupled to experimental synthesis and characterization activities, such as the rapid prototyping described above.

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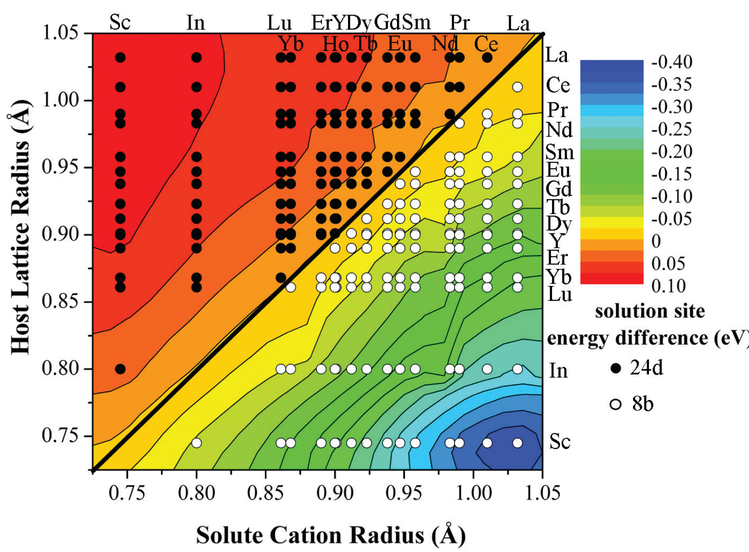

For activators such as Eu in RE_2O_3 rare earth oxide scintillators, a 24d site preference is desired for optimal scintillator performance. This contour map describes the calculated energetic site preference for activators in RE_2O_3 oxide scintillators, therefore identifiying regions of compositional interest.

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