Development and Experimental Assessment of Periodic-Nanostructure Coatings for Improved Detection Properties of High-Refractive-Index Inorganic Scintillators

Marek Flaska[1, a)], Stuti Surani[1], Roman Samulyak[1], Federico Scurti[1], and Douglas Wolfe[2]

[1]*Ken and Mary Alice Lindquist Department of Nuclear Engineering, Hallowell Building*
The Pennsylvania State University
University Park, PA 16802, USA
[2]*Associate Vice President for Research, Director of Strategic Initiatives, 304 Old Main*
The Pennsylvania State University
University Park, PA 16802, USA

[a)]*Corresponding author: mflaska@psu.edu*

Abstract. Inorganic scintillators are widely used in various applications of gamma counting and spectroscopy, such as nuclear nonproliferation and safeguards, and medical, space, or industrial applications. Typically, they have a good energy resolution, stable performance, somewhat low cost, and relatively high gamma detection efficiency. However, many inorganic scintillators have high refractive indices and therefore suffer from significant light losses due to the light phenomenon called the total internal reflection (TIR). This research project investigates using optimized, two-dimensional, periodic nanostructures called photonic crystals (PHCs) to aid in recovering some of the TIR-driven light loss. From the practical point of view, PHCs create, via constructive interference of electromagnetic light waves, an improved optical coupling between a scintillator and photosensor for the trapped light, thereby improving the overall light extraction, collection, and consequently the light output (LO) of the scintillator-photosensor assembly. Improving the LO of an inorganic scintillator leads to enhanced energy and time resolutions, thereby allowing for an extended range of radiation detection applications. PHCs can be optimized in terms of their shape, dimensions, and material composition to maximize the scintillator's LO. This project consists of two phases: The simulation phase for optimization simulations of PHCs, and the manufacturing phase for manufacturing the optimized PHCs and characterizing the improvement in LO and energy resolution through radiation measurements. During the first phase, a simulation toolkit was built to optimize the PHC geometry for a given inorganic scintillator. Optimization simulations were performed for LYSO, BGO, and $LaBr_3$ inorganic scintillators coated with Si_3N_4 and TiO_2 PHC materials. The LYSO and BGO scintillators were chosen primarily for their lower cost and non-hygroscopic nature, which greatly simplifies the manufacturing process. $LaBr_3$, the gold standard among inorganic scintillators for gamma spectroscopy, is the ultimate choice for PHC treatment due to its excellent energy resolution (~ 2% at 662 keV), despite being hygroscopic and expensive. The final simulation results show an ideal LO improvement of up to 57%, 130%, and 58% for LYSO, BGO, and $LaBr_3$, respectively, for individually optimized PHC geometries. During the second project phase, the PHC geometry optimized during the first phase for the LYSO-Si_3N_4 combination was manufactured via electron beam (e-beam) lithography. LYSO was chosen for the PHC-coating efforts, because it has higher light yield and energy resolution than BGO and is hygroscopic, which simplifies the PHC-manufacturing process. In the future, the manufacturing process will be modified for different scintillator materials, including BGO, NaI, and $LaBr_3$. The PHC-manufacturing method was developed for the LYSO scintillator material with Si_3N_4 PHCs through the e-beam lithography and ion etching. Following the manufacturing efforts, radiation measurements were performed for the PHC-coated LYSO scintillators using a ^{137}Cs gamma source. The radiation measurements show a LO improvement of up to 28% and up to 13% in energy resolution for the best performing PHC coating. Future work will include further improving the overall quality of the manufactured PHCs and incorporating experimentally observed defects in the simulations. With an improved energy resolution, some inorganic scintillators will become competitive enough when compared to room-temperature semiconductor detectors, which would result in cheaper gamma-spectroscopy systems available for detection applications that require deployment of many systems, such as nuclear nonproliferation and safeguards applications. Assessing improvements in timing resolution will also be included in the future work; medical applications such as PET scanners based on LYSO or LSO scintillators would also benefit from optimized PHC coating due to an improved timing resolution. Finally, improving the overall performance of inorganic scintillators by using optimized PHC coatings could have a global impact for many inorganic-scintillator-based applications that would benefit from higher detection efficiency, energy resolution, or timing resolution.