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FY05 LDRD Final Report Nanomaterials for Radiation Detection

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Auspices Statement

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FY05 LDRD Final Report
Nanomaterials for Radiation Detection
LDRD Project Tracking Code: 04-ERD-107
Tzu-Fang Wang, Principal Investigator

Abstract

We have demonstrated that it is possible to enhance current radiation detection capability by manipulating the materials at the nano level.

Introduction/Background

Fabrication of three-dimensional (3-D) nanomaterial composite for radiation detection has great potential benefits over current semiconductor- and scintillation-based technologies because of the precise control of material-radiation interaction and modulation of signal output. It is also a significant leap beyond current 2-D nanotechnology. Moreover, since we are building the materials using a combination of top-down and bottom-up approaches, this strategy to make radiation detection materials can provide significant improvement to radiation-detection technologies, which are currently based on difficult-to-control bulk crystal growth techniques.

We are applying this strategy to tackle two important areas in radiation detection: gamma-rays and neutrons. In gamma-ray detection, our first goal is to employ nanomaterials in the form of quantum-dot-based mixed matrices or nanoporous semiconductors to achieve scintillation output several times over that from NaI(Tl) crystals. In neutron detection, we are constructing a 3-D structure using a doped nanowire "forest" supported by a boron matrix and evaluating the detection efficiency of different device geometry with simulation.

Research Activities/Results/Technical Outcome (for gamma-ray detection)

We first decided to use nanoporous silica as supporting the matrix and to infiltrate it with quantum dots and laser dyes. This material, appears to constitute a matrix of choice: it is sturdy, inert, transparent, and, more importantly, the size of the nano-cavities can be tailored to the size of the host laser dye molecules and quantum dots by performing a slow HF (hydrofluoric acid) etch. We demonstrated that the matrix, once etched, cleaned and dried, was transparent in the visible range. We infiltrated the prepared nanoporous glass with quantum dots and also with laser dyes and we characterized the emission and absorption properties of the new composite materials (see Figure 1). Such materials enable the study of the radiation interactions with the nanomaterials as well as of the effect of the Stokes Shift (wavelength shift between absorption and emission). The visible wavelength output is crucial in improving the resolution of the detector since there is a factor of 3 in the quantum efficiency of photomultiplier tube (for UV output of most scintillators) and photodiode that operate in the visible range. We also studied alpha particle interaction with dyes and CdSe quantum dots embedded in nanoporous silica matrices.

We also studied alpha particle interaction with dyes and CdSe quantum dots embedded in nanoporous silica matrices. We found that though the absorption and emission spectra of rhodamine B dye-filled and quantum dots-filled silica are similar

(Figure 1), the energy spectra (Figure 2) recorded using a photomultiplier-based system show very different behavior upon $^{243-244}\text{Cm}$ alpha source irradiation. We are currently combining Monte Carlo simulation codes, TRIM¹⁾, and DETECT 2000²⁾ to understand the photon transport property of these systems.

Figure 1. Emission and absorption spectra for nano-porous glass filled with (a) CdSe quantum dots and (b) rhodamine B dye.

Figure 2. Detection counts versus "Energy" (channels) plots using a photomultiplier-based system with $^{243-244}\text{Cm}$ alpha source on nano-porous glass filled with (a) CdSe quantum dots and (b) rhodamine B dye. Background is shown in black.

Research Activities/Results/Technical Outcome (for neutron detection)

We investigated a 3D semiconductor pillar structures embedded with neutron converter materials (^{10}B) for thermal neutron detection. (Figure 3a) We accomplished two tasks: First, we combined TRIM codes and Monte Carlo simulation to simulate the relation of detector geometry to its neutron detection efficiency. We found that the pillar semiconductor structured neutron detector can perform 3x to 20x better than planar structured detector (2-5% efficiency). Our simulation results also verify that detectors with nanometer sized pillars could have 30% detection efficiency than its micron sized counter part. (Figure 3b) Second, we have successfully demonstrated the fabrication of 3D nanometer sized pillar system of various diameters using deep reactive ion etching³⁾ and nanosphere lithography (Figure 4). Moreover, we can fabricate pillars with sub micron size and 1:10 aspect ratio in this regime. (Figure 5.)

Figure 3. (a) Schematic of solid state neutron detector with proposed pillar semiconductor platform embedded with ^{10}B . (b) Graph of simulated neutron detection efficiency versus etch depth of silicon pillar structures for evaluating proposed neutron detector geometry in the micron and nanometer sized regime.

Figure 4. Scanning electron micrographs of sub-micron silicon pillars of different diameters made on silicon wafer using polystyrene sphere masks tailored by oxygen reactive ion etching and deep reactive ion etching "Bosch process" technique. The top views and cross-sections of resulting samples with 30, 60, 90, 120 and 150 secs of oxygen etching time are: (a) & (f), (b) & (g), (c) & (h), (d) & (i), and (e) & (j) respectively. The scale bars are 750nm. Processing done in Center for Micro and Nano Technology (CMNT, LLNL).

Figure 5. Scanning electron micrograph of silicon nanopillars of *ca.* 200nm with 1:9.5 aspect ratio made by nanosphere lithography and deep reactive ion etching technique. Processing done in Center for Micro and Nano Technology (CMNT, LLNL).

Exit Plan

We are seeking support from NNSA, NA-22 and DHS through a) strengthening effort in radiation detection technology development; b) enabling technology for fieldable and affordable radiation detectors.

References

- 1) J. F. Ziegler, J. P. Biersack, and U. Littmark, The stopping and range of ions in solids, Pergamon press, New York (1985).
- 2) G. F. Knoll, T. F. Knoll, and T. M. Henderson, IEEE Trans. Nucl. Sci. **35**, 872-875 (1988).
- 3) M. A. Blauw, T. Zijlstra, and E van der Drift, Balancing the etching and passivation in time-multiplexed deep dry etching of silicon *J. Vac. Sci. Technol. B* **19** 2930-4 (2001).

Record of Inventions

"Semiconductor nano-materials for radiation detection", Sonia Létant, Chin Li Cheung, and Tzu-Fang Wang, IL-11433. Filed on 10/04/2004.

"Semiconductor nano-matrix for neutron detection", Chin Li Cheung, Rebecca J. Welty, Catherine Reinhardt, and Tzu-Fang Wang, IL11463. Filed on 12/16/2004.

Presentations

"Fabrication of Nanopillars by Nanosphere Lithography," Barry Cheung, Rebecca Welty, Cathy Reinhardt & Tzu-Fang Wang. Poster presentation in Symposium J of 2005 Spring Materials Research Society meeting. March 28-April 1, 2004.

"Roadmap for High Efficiency Solid-State Neutron Detectors," R.J. Nikolic, C.L. Cheung, C.E. Reinhardt, and T.F. Wang, SPIE - International Symposium on Integrated Optoelectronic Devices, Photonics West, Boston, MA, Oct. 2005.

Publications

"Fabrication of nanopillars by nanosphere lithography," C.L. Cheung, R.J. Welty, C.E. Reinhardt, and T.F. Wang, Nanotechnology, accepted for publication, to appear vol. 17, 2006.

"Study of porous glass doped with quantum dots or laser dyes under alpha irradiation," S.E. Létant and T.F. Wang, submitted to Applied Physics Letters, 2006