Radiation Measurements 111 (2018) 27–34

Contents lists available at ScienceDirect

Radiation Measurements

journal homepage: www.elsevier.com/locate/radmeas

Quantum-dot doped polymeric scintillation material for radiation detection

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A B S T R A C T

Scintillation and luminescence enhancement were observed in a Quantum Dot-doped scintillating polymer, facilitating energy transfer with polymer medium and scintillating dye. 2,5-diphenyloxazole (PPO) and Cadmium Sulfide (CdS) Quantum Dots (QD) with 418 nm emission coated with oleic acid were dispersed in a polyvinyl toluene (PVT) matrix with transparency retained. Under UV excitation a sharp emission was detected at 426 nm from minimal QD loading with possible indication of QD energy transfer. Scintillation enhancement was observed with different radioactive sources in PVT-PPO/QD polymer with a photomultiplier detector setup. The response to gamma and beta radiation was investigated with differences in the energy spectrum and a count increase observed with increased QD loading.

1. Introduction

The development of scintillation materials has gained significant traction in the fields of high energy physics, medical imaging, and nuclear security due to advances in material fabrication (Bertrand et al., 2014, 2015). Current composition of scintillators lies with either organic or inorganic material each suited for different application. Inorganic crystalline scintillators are widely used due to their superior energy resolution and stopping power for gamma radiation. However, they are limited by their scalability and hygroscopic property thus requiring protection and careful handling. To fill the shortcomings of inorganic crystalline scintillators, organic scintillating polymers are implemented instead. Organic polymers tend to have poor gamma stopping power due to its low Z characteristics which results in poor energy resolution, but may provide high sensitivity for charged particle detection (Torrisi, 1997; Ghergherehchi et al., 2010; Quaranta et al., 2002; Miramonti, 2002). Organic scintillators have faster response time and are scalable compared to inorganic detectors. Another advantage of implementing organic scintillators is the integration of various doping agents into the medium to raise scintillation efficiency, sensitivity, and allow for controlling the emission wavelength through energy transfer (Sandler and Tsuo, 1963; Destruel et al., 1989). Wavelength emission is an important factor to consider due to the quantum efficiency of the detector. Different detectors such as Photomultiplier tubes (PMT) or Silicon Photomultipliers (SiPM) have different detection wavelengths for optimal performance. To accommodate their parameters, wavelength matching is required between scintillators and detectors where wavelength shifters are often added to enhance optimization (Buzhan et al., 2003). In most cases the quantum efficiency peak of a PMT lies within most inorganic scintillator's emission in the blue and low UV region as opposed to SiPM where it lies in the red and infrared (IR) wavelength region making coupling with traditional scintillator problematic. In such cases wavelength shifters such as fluorescent nanoparticles may be used in a similar manner as fluorescence dyes by doping it into the plastic scintillator. Iridium complexes as well as other luminescence emitters like lanthanides at minimal loading has been demonstrated to enhance scintillation through energy transfer with wavelength shifting effects (Sguerra et al., 2104; Campbell and Crone, 2007). Semiconductor nanocrystals such as quantum dots are ideal dopants which can be employed into a scintillating polymer. Owing to their unique emission and energy transfer properties they might achieve similar effects as more conventional doping agents.

Quantum dots are well studied in a wide range of applications from medical imaging (Juzenas et al., 2008; Chan et al., 2002) to solar cell devices and are gradually branching into radiation detection (Nozik, 2002; Kongkanand et al., 2008; Sargent, 2012). Due to their specific wavelength emission controlled by size control and high quantum yield they serve as a popular choice in photo devices. Studies have been done

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https://doi.org/10.1016/j.radmeas.2018.02.008
Received 16 August 2017; Received in revised form 15 December 2017; Accepted 27 February 2018
Available online 02 March 2018
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on colloidal QD in thin transparent matrices for radiation detection with poor results under gamma radiation but with possible implementation as an alpha particle detector (Létant and Wang, 2006; Manickaraj et al., 2013; Dai et al., 2002; Wagner et al., 2012). Although the QD is a moderate Z material its stopping power is not sufficient at small loading and it tends to have poor scintillating property in a polymer by itself (Yao et al., 2010). In a recent study by Liu et al. it was shown that a polymer loaded with 60% by mass of quantum dots provided enough stopping power to observe a photoelectric peak from gamma radiation with a secondary dye (Liu et al., 2017). However, the high loading of QD may affect stability of the scintillator itself. Despite its poor performance as a scintillator, QDs can be paired with other luminescence materials through the use of non-radiative Förster Resonance Energy Transfer (FRET) possibly improving the performance of the material.

The underlying conditions for FRET requires the donor and acceptor agents to be dipole orientated, spatially distanced, and have overlapping absorption and emission spectra at the resonance condition (Förster, 1959). Intramolecular Excitation Energy Transfer such as FRET has already demonstrated its applications in bio sensors with QD and proteins. Resonance energy transfer with luminescence QD as the donor have been well documented through bio-tagging and bio-probes over the past decade (Clapp et al., 2006). More recently QD were paired with nanoparticles and scintillating dyes for radiation scintillation utilizing this technique with QD as the acceptor. A study based on x-ray detection fabricated CeF3/ZnO lanthanide QD composite demonstrating an increase in luminescence by a factor of 4 (Sahi and Chen, 2013). Campbell and Crone reported on the scintillation properties of a MEH-PPV (poly[2-methoxy-5-(2′-ethylhexloxy)-p-phenylene vinylene) and CdSe/ZnSe QD polymer composite thin film using an electron beam from transient cathode luminescence as a surrogate for beta radiation (Campbell and Crone, 2006). Luminescence was detected from 3 keV transient cathode electrons with energy transfer observed under UV excitation, exhibiting one order of magnitude higher light output. Proton scintillation was reported for core-shell CdSe/ZnS QD doped with PPO dye polymer hinting FRET luminescence under 45 MeV proton beam however with minute scintillation effect under x-ray excitation (Park et al., 2014). To further understand the effect of FRET more studies are needed on QD incorporated scintillating system.

In this paper, we investigate possible Förster Resonance Energy Transfer occurring between organic scintillating dye and QD in bulk plastic polymer under high energy excitation. Non-radiative energy transfer arises between the polymer medium and scintillating dye due to fulfillment of FRET criteria however this transfer is unclear with the addition of QD. Energy transfer between organic dyes and inorganic nanoparticles through FRET under radiation have not been extensively studied and only been recently demonstrated at extreme conditions (Liu et al., 2017). Here we experimentally show scintillation enhancement of a PVT-PPO polymer when doped with CdS QD along with its effects in emission spectroscopy and collected energy spectra from ionizing radiation interactions.

2. Materials and methods

2.1. Experimental details and sample preparation

Scintillating polymers were fabricated through standard thermal polymerization method with monomer styrene. 2,5-diphenyloxazole (99%), 4-Methylstyrene (96% with 3,5-di-tert-butylcatechol), silica gel 28–200 mesh, and benzoyl peroxide (97%) were purchased from Sigma-Aldrich. Cadmium sulfide quantum dots with emission at 418 nm in a toluene solution coated with oleic acid ligands were received from Tetramer Technologies, LLC. The QD were prepared for inclusion into the polymer by removing the toluene solvent with a methanol wash. The precipitated QD were stored in the dark and dried overnight in a vacuum desiccator, purging excess solvent. 4-methylstyrene from Sigma-Aldrich was prepared by removal of the polymerization inhibitor with 0.15 M NaOH through solvent extraction. The extracted styrene was further processed through column distillation with silica gel, the purified styrene was stored in a freezer, if it was not immediately used. 3 mL of distilled styrene was mixed with the desired amount of QD (from 0 up to 5 mg) and sonicated for 5 min ensuring dispersion. 15 mg of benzoyl peroxide and 120 mg of PPO was added to the solution and further sonicated for 20 min. The solution is thermally polymerized at 75 °C under nitrogen. The solution was stirred for the first hour to ensure uniformity until viscosity effects were observed. After 72 h, the polymerized sample is cooled and removed from the mold. The same method was used to fabricate all samples. The samples were fabricated to a standard dimension of Ø 2.2 × 0.65 cm shown in Fig. 1.

Fig. 1. PVT-PPO/QD (left) and PVT-PPO (right) samples, bottom picture shows the luminescence under UV lamp excitation at around 250 nm.

2.2. Instrumentation and measurements

Fabricated samples were characterized by their absorption and fluorescence spectra as well as radio-luminescence response. Fluorescence spectra from UV to visible light were measured by a Cary Eclipse Fluorescence Spectrometer, excitations were carried out at 250 nm and 280 nm by a Xenon flash lamp with an internal PMT for detection. UV/Vis absorption and transmission was measured by a dual-beam Olis-upgraded Cary 14 Spectrophotometer with deuterium and tungsten lamps for UV and visible light emission, respectively. The absorption and transmission spectra were measured from 300 nm to 700 nm with the lamp switch from UV to visible light set at 350 nm.

Scintillation pulses were captured with an EMI-9886 PMT with peak quantum efficiency at 380 nm at 30% connected to a Canberra Osprey® digital tube base unit. The output data was digitized and processed by ProSpect® software on a PC. A plastic sample holder for the polymer scintillator was attached to the aperture of the PMT centering the sample. No optical gel was used to couple the scintillator material to the PMT. The detector setup was placed in a light-tight box to reduce the background noise. A radioactive source was placed directly on the aperture of the holder exposed to the sample. Six different calibration sources were used based on their gamma and beta emission properties. Their respective activities, at the time of the measuremens, are listed in Table 1. For the gamma measurements sealed button sources containing Am-241, Cs-137, and Co-60 were used. For these measurements, the alpha or beta component was blocked by a taped sealed glass slide inserted between the source and the polymer. For the beta measurements C-14, P-32, and Sr-90 sources were used. The C-14 and Sr-90 sources were sealed button sources while the P-32 source was obtained by neutron activation of natural phosphor using the UCI TRIGA® reactor. For the Sr-90 source Y-90, generated from the decay of Sr-90, was present and in secular equilibrium with, and hence of equal activity as, the Sr-90. Each sample was counted for a certain amount of time

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