Preliminary Results from an Investigation into Nanostructured Nuclear Radiation Detectors for Non-Proliferation Applications

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Nanoparticle Detector

The principle is that “nanocrystals,” crystals 2 to 5 nm in diameter, of certain inorganic scintillator materials can capture most of the energy from γ 's, thus offering nearly the performance of single large crystals, if packed densely enough in plastic. The resulting mixture also has the desirable features of plastic. The crystals are mixed in a modified polystyrene plastic, a scintillator material, so it increases the amount of energy converted to a detectable signal.

Property Contrast

Background: $\text{LaF}_3:\text{Ce}$ at 5% nanocrystals with DI- water

Property	Crystal Detector	Nanostructured Detector
Cost	\$300/cc	\$1/cc
Configuration	Fixed solid crystal	Plastic; May be formed into large sheets to increase sensitivity
Temperature	Ambient for Low resolution crystals; Low for High resolution crystals	Ambient
Photoluminescence	2-120 K-photons/Mev [300-530 nm]	~70 K-photons/Mev [397-782 nm]
Resolution	<1% Low Temp; 2-8% Ambient Temp for ^{137}Cs (662 keV)	15% achieved for ^{241}Am (60 keV)
Efficiency	PMTs suffer from low quantum efficiency (1–10%)	<60%

Fig. 7. Photopicks observed from small $\text{LaF}_3:\text{Ce}$ pieces under irradiation from ^{241}Am (dots) and ^{57}Co (diamonds).

Motivation

1. Economics - Nanoparticle detectors offer the possibility of an economical detection system, with the promise of high sensitivity.
2. Efficiency - Nanoparticle detectors offer the possibility of achieving high quantum efficiencies and sensitivity.
3. Energy Range - Nanoparticle detectors may even offer the possibility of broader energy spectral ranges.

Goal

To demonstrate the possibility of measuring both low-energy X-rays and relatively high-energy gamma rays simultaneously using the nano-structured nuclear radiation detectors.

Technical Accomplishments – List of Detectors

Figure	Detector		Properties		Comment
	Nanoparticle	Type	Volume	Concentration	
Figure 1	CdSe/ZnS	Core Shell	1 cc	1.40%	epoxide-amine polymer
Figure 2	CdSe/ZnS	Core Shell	1 cc	2.80%	epoxide-amine polymer
Figure 3	CdSe:S	Quantum Dot	1 cc	0.20%	epoxide-amine polymer
Figure 4	CdTe/ZnS	Core Shell	1 cc	0.01%	
Figure 5	CdTe/ZnS	Core Shell	1 cc	0.05%	
Figure 6	CdTe/ZnS	Core Shell	1 cc	0.11%	
Figure 7	CdTe/ZnS	Core Shell	1 cc	0.43%	
Figure 8	CdTe/ZnS	Core Shell	1 cc	0.90%	
Figure 9	CdTe/ZnS	Core Shell	1 cc	2%	
Figure 10	ZnSe:Mn	Quantum Dot	1 cc	1%	polystyrene matrix
Figure 11	ZnSe:Mn	Quantum Dot	1 cc	2%	polystyrene matrix
Figure 12	ZnSe:Mn	Quantum Dot	1 cc	5%	polystyrene matrix
Figure 13	D1	Core Shell	<1 cc	0.90%	Surfactant
Figure 14	D2	Core Shell	<1 cc	0.43%	Surfactant
Figure 15	D3	Core Shell	<1 cc	0.11%	Surfactant
Figure 16	D4	Core Shell	<1 cc	0.05%	Surfactant
Figure 17	D5	Core Shell	<1 cc	0.01%	Surfactant
Figure 18	D6	Core Shell	<1 cc	0.90%	Surfactant
	BC400	Plastic	<1 cc		Surfactant

Technical Accomplishments – List of Detectors

Figure 1. Nanocomposite 1.4% CdSe/ZnS.

Figure 4. Nanocomposite 0.01% CdTe/ZnS.

Figure 7. Nanocomposite 0.43% CdTe/ZnS.

Figure 10. Nanocomposite 1% ZnSe:Mn.

Figure 13. Nanocomposite D1 .

Figure 16. Nanocomposite D4.

Figure 2. Nanocomposite 2.8% CdSe/ZnS.

Figure 5. Nanocomposite 0.05% CdTe/ZnS.

Figure 8. Nanocomposite 0.9% CdTe/ZnS.

Figure 11. Nanocomposite 2% ZnSe:Mn.

Figure 14. Nanocomposite D2.

Figure 17. Nanocomposite D5.

Figure 3. Nanocomposite 0.2% CdSeS

Figure 6. Nanocomposite 0.11% CdTe/ZnS.

Figure 9. Nanocomposite 2% CdTe/ZnS.

Figure 12. Nanocomposite 5% ZnSe:Mn.

Figure 15. Nanocomposite D3.

Figure 18. Nanocomposite 0.9% CdTe/ZnS surfactant.

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Technical Accomplishments–Solid Nanoparticle Detector

The 0.9% CdTe/ZnS nanocomposite detector is typically approximately 1 cm in diameter and 1 cm in length. It is not well polished. This small size geometry must be fit onto the $\frac{3}{4}$ " photomultiplier tube (PMT) with an excellent light coupling and placed inside a light-tight enclosure.

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Technical Accomplishments- Solid Nanoparticle Detector Mount

Aluminum detector mount is shown on left. The outer protective cap is shown in the middle. Photomultiplier tube is shown on the right.

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Technical Accomplishments- Solid Nanoparticle Detector Mount

Photomultiplier tube with inner cap assembled is shown on left. Full assembly has the detector seated inside the inner cap, which is coupled by optical grease to the photomultiplier tube, and has the outer protective cap cover inserted over the inner cap containing the detector, as is shown on the right.

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Technical Accomplishments

- Stepped through plan for data acquisition
- Definitely determined an energy correlation to the spectra for different types of nanoparticle detectors and different concentrations of nanoparticle
- Nano-issues appeared with MCNPX
- Ran into unexpected results with several surface roughness case studies

Technical Accomplishments – BC400

This is a ^{137}Cs spectrum (with background subtraction) acquired with a plastic BC400, 12.5 mm diameter by 19 mm long detector using the new detector assembly. Inset: MCNPX calculation of simulated spectrum is shown.

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^{241}Am , ^{137}Cs , ^{60}Co , and the Background Spectra Subtracted

0.2 % CdSe:S Quantum Dot Detector

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^{241}Am , ^{137}Cs , ^{60}Co , and the Background Spectra Subtracted

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Technical Accomplishments – 2.8% CdSe/ZnS

Background-subtracted spectrum of ⁶⁰Co source with both 1173- and 1332-keV γ lines (blue) and of ¹³⁷Cs with 662-keV γ line (black) taken with detector 2, the 2.8% CdSe/ZnS core shell nanoparticle detector. Inset: MCNPX calculation of simulated spectra is shown.

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Technical Accomplishments – 2.8% CdSe/ZnS

Raw spectra of ^{60}Co source with both 1173- and 1332-keV γ lines (aqua) and of ^{241}Am with 60-keV γ line (blue) taken with 2.8% CdSe/ZnS core shell nanoparticle detector.

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Spectra of ⁶⁰Co source taken with 0.2% CdSe:S and 1%, 2%, and 5% ZnSe:Mn quantum dot nanoparticle detectors.

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Geometry for the MCNPX calculation of spectrum of ^{137}Cs source (with 662-keV γ line) taken with a CdSe nanoparticle detector, with 8% of the mass composed of CdSe nanoparticles.

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Geometry for the MCNPX calculation for detector with 1-mm CdSe millispheres. In this case, millispheres are nearly to scale, and correctly project the relative abundance of material of CdSe to compose 2.8% of the mass of the detector

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MCNPX calculation of spectrum of ^{137}Cs source (with 662-keV line) taken with detector 2, the 2.8% CdSe/ZnS core shell nanoparticle detector with a polystyrene matrix, with 100 nm CdSe nanospheres, compared to a background spectrum

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MCNPX Calculation versus Data

Data

MCNPX

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MCNPX Calculation versus Data

Data

MCNPX

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^{241}Am , ^{137}Cs , ^{60}Co , and the Background Spectra Subtracted

Top: Count Rate (Counts/s); Bottom: Probability Spectrum (Counts/y)

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^{241}Am , ^{137}Cs , ^{60}Co , and the Background Spectra Subtracted

Top: Count Rate (Counts/s); Bottom: Probability Spectrum (Counts/y)

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Results

– Solid Nanoparticle detector

Measured Spectra with and without a 31.9- μ Ci ^{137}Cs Source.

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Results

– Solid Nanoparticle detector

Source	Intensity (μCi)	Count Rate versus Source (cps)		
		2.8%	1.4%	
		CdSe/ZnS (cps)	CdSe/ZnS (cps)	BC400 (cps)
		16 mm D $\times$ 4 mm L	16 mm D $\times$ 4 mm L	12.5 mm D $\times$ 19 mm L
Background		1.63	1.29	1.27
^{60}Co (4 cm)	3.78	23.74	18.09	
^{137}Cs (4 cm)	31.9	12.69	20.72	170.22
^{137}Cs (8 cm)	31.9	9.29	10.54	
^{241}Am (4 cm)	9.09	0.2	2.4	

BC400 and Nanodetector performance comparisons.

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Results – Solid Nanoparticle detector

Detector	Detector		Properties		Comment	Count Rate with Source (Counts/s)				
	Nanoparticle	Type	Volume	Concentration		²⁴¹ Am 9.09 µCi 4 cm	¹³⁷ Cs 31.9 µCi 4 cm	¹³⁷ Cs 31.9 µCi 8 cm	⁶⁰ Co 3.78 µCi 4 cm	Background
Detector 1	CdSe/ZnS	Core Shell	1 cc	1.40%	epoxide-amine polymer	2.4	20.72	10.54	18.09	1.29
Detector 2	CdSe/ZnS	Core Shell	1 cc	2.80%	epoxide-amine polymer	0.2	12.69	9.29	23.74	1.63
Detector 3	CdSe:S	Quantum Dot	1 cc	0.20%	epoxide-amine polymer	0.07	16.47	11.57	16.64	3.01
Detector 4	CdTe/ZnS	Core Shell	1 cc	0.01%						
Detector 5	CdTe/ZnS	Core Shell	1 cc	0.05%						
Detector 6	CdTe/ZnS	Core Shell	1 cc	0.11%						
Detector 7	CdTe/ZnS	Core Shell	1 cc	0.43%						
Detector 8	CdTe/ZnS	Core Shell	1 cc	0.90%		0.09	52.95	16.69	33.74	7.34
Detector 9	CdTe/ZnS	Core Shell	1 cc	2%						54.13
Detector 10	ZnSe:Mn	Quantum Dot	1 cc	1%	polystyrene matrix	0.07	64.82	20.7	49.4	2.59
Detector 11	ZnSe:Mn	Quantum Dot	1 cc	2%	polystyrene matrix	3.97	61.46	25.76	50.68	2.64
Detector 12	ZnSe:Mn	Quantum Dot	1 cc	5%	polystyrene matrix	0.05	683.39	471.66	20.88	9.25
Detector 13	D1	Core Shell	<1 cc	0.90%	Surfactant					
Detector 14	D2	Core Shell	<1 cc	0.43%	Surfactant					
Detector 15	D3	Core Shell	<1 cc	0.11%	Surfactant					
Detector 16	D4	Core Shell	<1 cc	0.05%	Surfactant					
Detector 17	D5	Core Shell	<1 cc	0.01%	Surfactant					
Detector 18	D6	Core Shell	<1 cc	0.90%	Surfactant					
Detector 19	BC400		1 cc	0%			170.22			1.27

Discussion

Initial results indicate that BC400 performs better:

- This is our first run for nanoparticle detectors
- The nanoparticle detectors do demonstrate an ability to distinguish different γ -ray energies and sources
- They also demonstrate a neutron response
- Efficiency increases with increasing nanoparticle content
- Spectroscopic fidelity increases with decreasing nanoparticle content

Summary

Nanocomposite detectors are cheap, easy to fabricate, and respond to nuclear radiation. We have established correlations between energy spectra γ -ray energy. We have not observed significant improvement compared to BC400, but need more work to determine how best to obtain isotopic correlations and to optimize light yield.

Recommendations for future direction

- Refine MCNPX model to improve match to experimental data
- Adjust MCNPX parameters to improve detector performance
- Make assessment of modeling results for future detector developments

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