Gadolinium Based Scintillators for Thermal Neutron Detection

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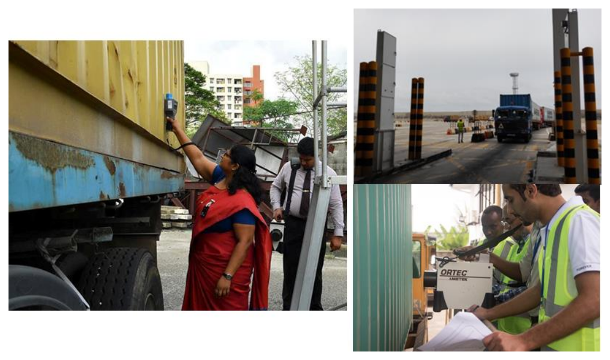
1.Introduction

Detection of illicit trafficking of nuclear material relies on the detection of the radiation emitted, for example, in plutonium detection one of the characteristic signatures derives from neutron emission. For this reason, neutron detectors offer an important role, particularly in detection systems used in nuclear security.



Most current neutron detection systems used in nuclear security use Helium-3 based technology. Helium-3 technology based detectors have high neutron detection efficiency, good gamma-ray discrimination, are non-toxic, and are considered the ‘‘standard’’ for neutron detection.

1.1. Why do we need alternative technology to replace Helium-3?

- Pure Helium-3 is required for Helium-3 based detectors.
- Helium-3 is a rare non-radioactive isotope of helium, which is a byproduct of tritium decay. No other mechanism of producing it is known at present.
- The growing demand for it already exceeds production in the next few years leading to an exponential increase of the price.

It is necessary to develop alternative detection systems based on technologies different from Helium-3 and new materials are needed to meet these challenges.

Alternative options: Boron fluoride based detectors
Semiconductor based detectors
Scintillator based detectors

1.2. Drawbacks of alternative technologies

- Boron fluoride is a toxic gas - not been used in nuclear security applications.
- Semiconductor crystals suffer from limitations in size - limit their detection efficiency.
- Boron lined gas filled proportional counters - relatively low detection efficiency.

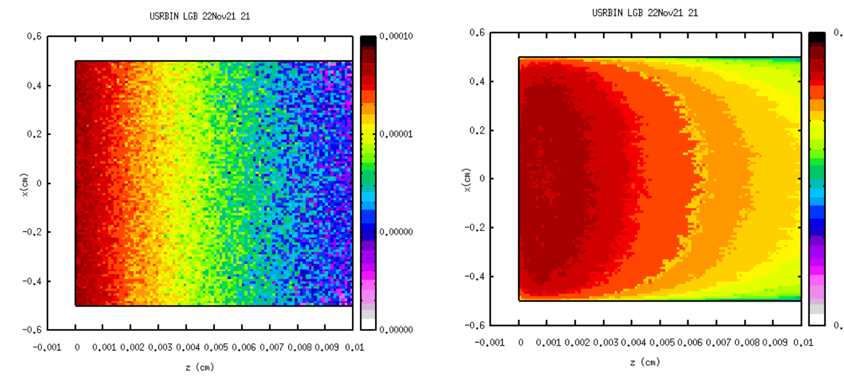
Therefore, scintillation neutron detectors can be a longer term alternative to Helium-3 technology. This research is focused on examining a possible method to engineer an effective scintillator for neutron detection.

2. Materials and Methods

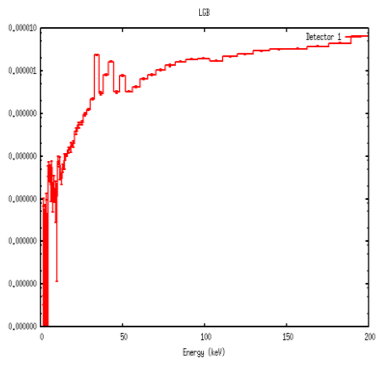
Lithium, gadolinium, and boron, all possess large neutron capture cross-section. An efficient new range of scintillators that contain these elements would be very promising. Under this research, we have developed scintillator layers based on $Li_6Gd(BO_3)_3:Eu^{3+}$, $Gd_2O_3:Eu^{3+}$ and $GdBO_3:Eu^{3+}$ phosphors.

2.1. Simulation

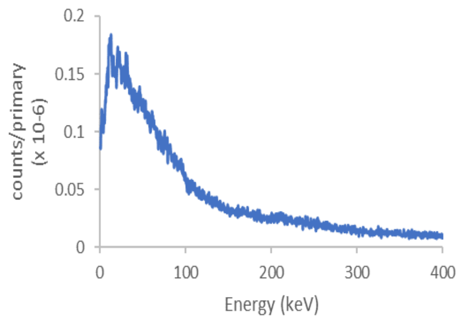
FLUKA/FLAIR codes have been chosen to simulate the response of the Gd based scintillator to thermal neutrons, e.g. $Li_6Gd(BO_3)_3:Eu^{3+}$:



Particle fluence (GeV/cm^3 per primary)

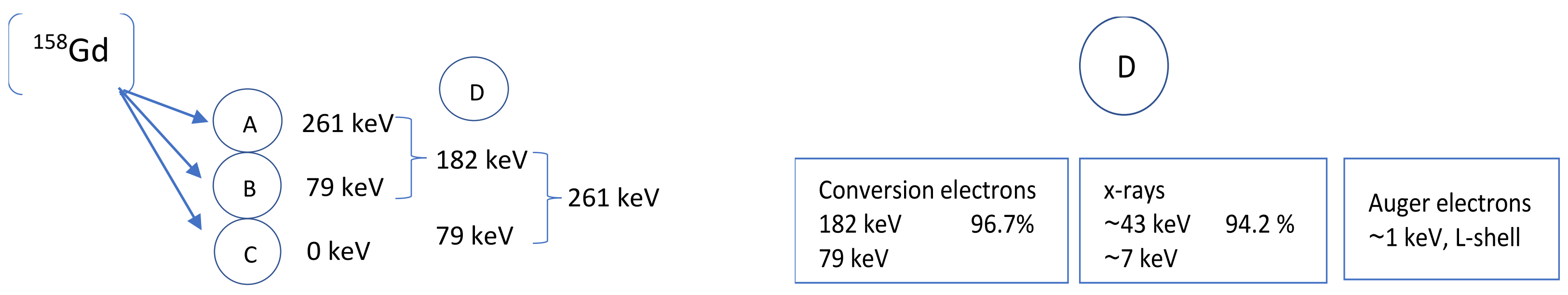


Kinetic energy of particles crossing the surface



Simulated pulse height spectra

$$^{157}Gd + ^{1}n_0 \longrightarrow ^{158}Gd^* \rightarrow ^{158}Gd + \gamma + \text{conversion electrons} + \text{X-rays (29-182 keV)} + \text{Auger electrons}$$



Conversion electrons
182 keV 96.7%
79 keV

x-rays
~43 keV 94.2 %
~7 keV

Auger electrons
~1 keV, L-shell

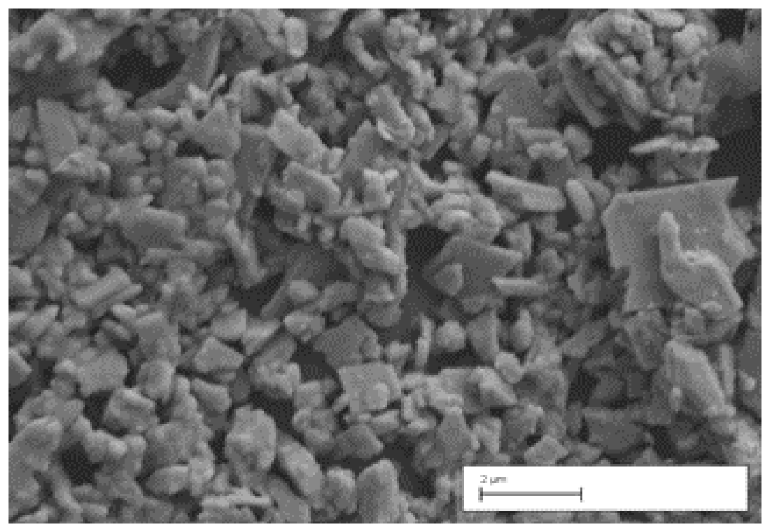
2.2. Development of phosphor as scintillator for neutron detection

To synthesise the $Li_6Gd(BO_3)_3:Eu^{3+}$ phosphor stoichiometric amounts of $Gd(NO_3)_3$ and $Eu(NO_3)_3$ stock solutions were added to a beaker and heated to 60˚C while stirring. To the solution were then added the required amounts of Li_2CO_3 and H_3BO_3, when the solution became clear the reaction was complete.

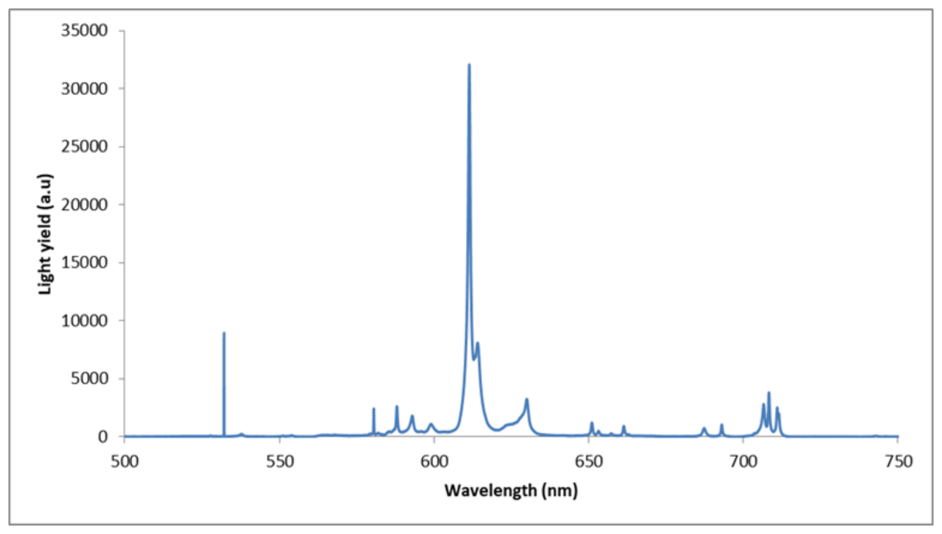
$$Gd(NO_3)_3.H_2O + 3Li_2CO_3 + 3H_3BO_3 + Eu(NO_3)_3.6H_2O \rightarrow Li_6Gd(BO_3)_3:Eu^{3+} + \text{nitric vapour} + H_2O + CO_2$$

The precipitate was then washed with deionized water several times and filtered at the pump. The precipitates were then dried at 60˚C in an oven, the resulting soft white phosphor precursor powders were annealed at 550 ˚C for sixteen hours producing the luminescent phosphor powder. To investigate the relationship between luminescence and firing temperature the samples were fired at two different temperatures (550˚C and 800˚C). K-bar printing was used to prepare thin films from the powders.

The luminescence spectrum of the $Li_6Gd(BO_3)_3:Eu^{3+}$ was obtained using Laser-induced spectroscopy (laser is at 532 nm with 1% filter).



SEM image

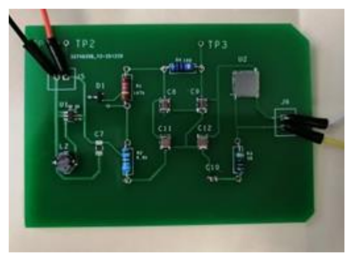


PL spectra

The synthesized phosphor samples show a red emission due to Eu^{3+}.

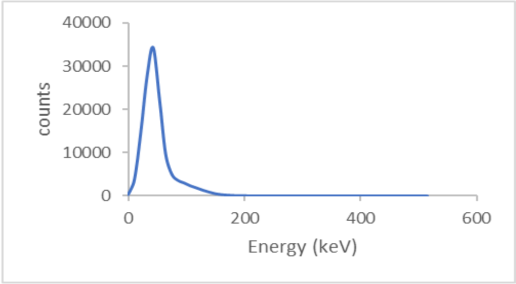
2.3. Electronics

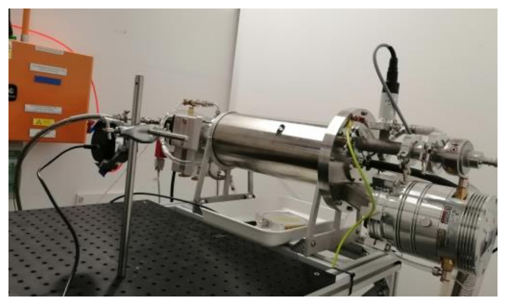
6×6 mm² SiPM from Broadcom is employed in the readout circuit design.



3. Testing with neutrons

A Deuterium-Deuterium (D-D) source which emits average energy at about 2.5 MeV is used as a neutron source. A high density-polyethylene (HDPE) moderator was used for converting fast neutrons emitted from D-D to thermal neutrons.





The fabricated Gd-based thin layer produced a pulse height spectrum with features that are attributable to Gd K x-ray emission, following a neutron capture as shown in the figure.

4. Conclusions and Acknowledgements

Simulated and measured K X-ray spectra are demonstrated to be in good agreement and indicate that the experimentally observed Gd K X-ray lines are due to neutron capture reactions. The results, for this small test samples, showed that Gd-based scintillators are promising scintillators for handheld applications.

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