

Comparative analysis of scintillators for detecting hydrogen capture γ rays

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Abstract— A comparative analysis of thallium-doped sodium iodide, europium-doped strontium iodide and cerium bromide scintillators for the detection of 2.223 MeV γ rays, indicative of hydrogen presence in spent nuclear fuel via neutron capture on hydrogen is presented. The detection of this full-energy peak might serve as a proxy for water ingress in spent nuclear fuel which is important for the assessment of spent fuel storage integrity and safety. Experiments have been performed to explore hydrogenous foams as analogues for dispersed water in fuel-containing materials. Each scintillator's efficiency, energy resolution, and potential application in a field setting are evaluated to determine the most effective detection system for monitoring hydrogen levels.

Index Terms— Hydrogen, neutron capture, spent nuclear fuel, gamma analysis.

I. INTRODUCTION

THE detection and analysis of various elements in spent nuclear fuel (SNF) is important in the nuclear industry. The presence of hydrogen in SNF assemblies is potentially indicative of water ingress, which is important because water in this environment is safety concern and introduces risks for long-term storage [1]. Neutron capture on hydrogen results in the emission of a characteristic 2.223 MeV γ ray via neutron activation analysis (NAA). The principle of NAA usually involves irradiating a target sample with neutrons and subsequently measuring the γ rays emitted by the activated nuclei. Most NAA experiments require high-energy γ -ray detectors, and often scintillators are used for this purpose.

The selection of an appropriate scintillation detector is essential for precise identification of 2.223 MeV γ ray and the performance of such detector is largely dependent on the scintillation material used. Thallium-doped sodium iodide (NaI(Tl)) has been used extensively in γ -ray spectroscopy for decades and remains highly suitable detector choice due to its simplicity and proven performance. In recent years, several new scintillators have been developed: Europium-doped strontium iodide (SrI₂[Eu]) and cerium bromide (CeBr₃) are some of the commonly-used scintillators in γ -ray spectroscopy due to their low cost, superior energy resolution and high-efficiency detection capabilities [2, 3]. In this research we explore the capabilities of thallium-doped sodium iodide, europium-doped strontium iodide and cerium bromide scintillators experimentally in detecting the 2.223 MeV γ ray of

hydrogen in spent nuclear fuel.

II. MATERIALS AND METHOD

The experimental measurements employed NaI(Tl), SrI₂(Eu), and CeBr₃ scintillators (Scionix, Netherlands) to detect capture γ rays emitted from candidate target samples containing hydrogen. The scintillators were each connected to a laptop via USB to a digital photomultiplier tube base. The measurements were controlled by MAESTRO Multichannel Analyser Emulator Software (Ametek Ortec, USA) and BrightSpec Multichannel Analyser (BrightSpec NV/SA, Belgium), and MCA4A for NaI(Tl) and SrI₂(Eu) and CeBr₃ scintillators respectively. The scintillators were calibrated using standard point and volume sources comprising ⁶⁰Co, ¹³⁷Cs, ¹⁵²Eu, and ²²⁶Ra to provide accurate energies are measured. Measurements were carried out on samples of high-density polyethylene (HDPE) foams ($\rho=0.96$ g/cm³) and mesh analogues of Nylon-12 ($\rho=1.01$ g/cm³).

The scintillators were positioned perpendicular to the target samples placed in front of a water-filled tank in which a ²⁵²Cf neutron source is housed. All measurements were done for 86400 s, except for energy calibrations which were done for 300 s for each of the radionuclides. GEANT4 simulations were performed to model the response of each detector to γ rays emitted from neutron capture events. The simulations used material parameters, detector geometry, and energy deposition methods to anticipate each detector's reaction to 2.223 MeV γ rays. A 3-D model of the experimental setup of the three scintillators is shown in Fig. 1.

III. RESULTS AND DISCUSSION

Fig. 2 shows the spectra of 2.223 MeV γ rays from the ¹H(n, γ)²H reaction using the three scintillators described above. Evidently, CeBr₃ shows the best energy resolution to detect the 2.223 MeV γ ray in the 1 MeV-6 MeV range. In comparison, the NaI(Tl) and SrI₂(Eu) scintillators are in the 1 MeV-3.2 MeV and 1 MeV-2.4 MeV range. A comparison of the detectors measuring the 2.223 MeV full-energy peak when no target is placed in front of the neutron tank is shown in Fig. 2 (a) arising from water in the neutron tank.

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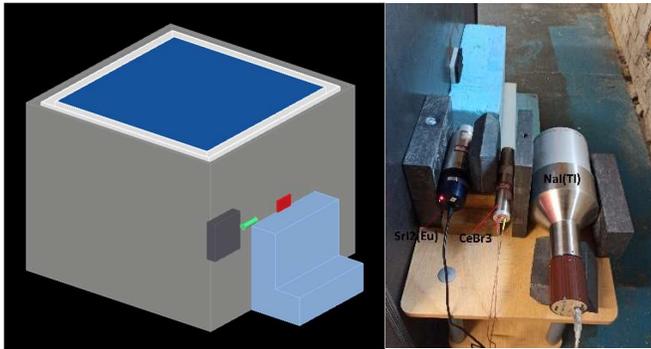


Fig. 1. GEANT4 3D model (left) of the experimental setup (right). Grey = neutron tank made of EN8 steel, blue = water, white = fibreglass, light blue = polystyrene blocks for sample support, red = target samples (HDPE and nylon-12), green = detector, dark grey = lead block(s) to shield detectors. The scintillators between the lead blocks correspond to $\text{SrI}_2(\text{Eu})$, CeBr_3 , and $\text{NaI}(\text{Tl})$ scintillators.

Figs 2(b)-(d) show the 2.223 MeV γ ray in HDPE and nylon-12. The total number of 2.223 MeV γ rays obtained is identical with and without the targets used. This agrees with the same observation by Nauchi *et al.* [4] in their neutron capture measurements using a pure germanium detector.

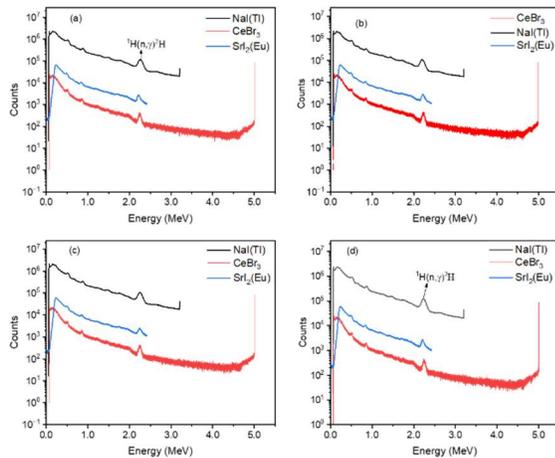


Fig. 2. γ -ray spectra for the detection of 2.223 MeV from the $^1\text{H}(n,\gamma)^2\text{H}$ reaction in (a) water (b) $10 \times 14 \times 5$ (cm) of HDPE (c) $10 \times 6 \times 1$ (cm) of Nylon-12 (d) $10 \times 6 \times 1$ (cm) of HDPE and Nylon-12. The HDPE was included here to serve as a moderator of the neutron field.

Fig. 3 shows the linear attenuation coefficients for the three scintillators. These data, generated from the NIST database, were normalized by the densities of the detector materials. The scintillators all have similar detection efficiencies within the energy range of 0.01 – 3 MeV.

Fig. 4. shows the simulation results of CeBr_3 γ -ray counts in HDPE and Nylon-12. The average energy of the HDPE and Nylon-12 spectra were 2.169 MeV and 2.153 MeV respectively. The total hits on the detector were 5987 and 5901 for the HDPE and nylon-12 targets respectively.

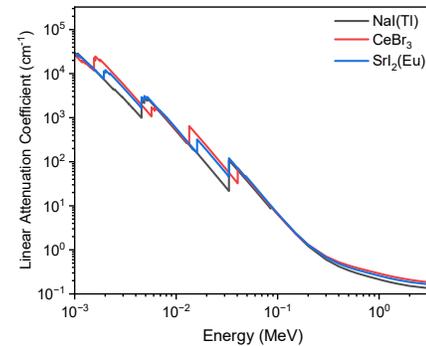


Fig. 3. Linear attenuation coefficients as a function of photon energy for $\text{NaI}(\text{Tl})$, $\text{SrI}_2(\text{Eu})$ and CeBr_3 scintillators. The data for this plot have been derived from XCOM [5].

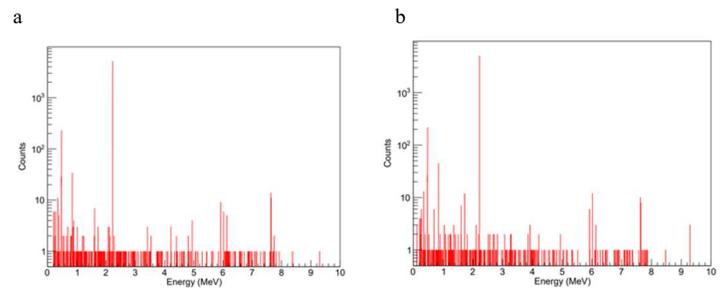


Fig. 4. Simulated energy spectra for (a) Nylon-12 and (b) HDPE targets. Each simulation contained 10 million neutron production events.

IV. CONCLUSION

The comparative analysis of $\text{NaI}(\text{Tl})$, $\text{SrI}_2(\text{Eu})$, and CeBr_3 scintillators for detecting 2.223 MeV γ rays from hydrogen capture reactions shows that each detector has unique performance characteristics for γ spectroscopy tasks. $\text{NaI}(\text{Tl})$ exhibits moderate resolution and detection efficiency compared to $\text{SrI}_2(\text{Eu})$. CeBr_3 offers better energy resolution and stability than the other two scintillators, which suggests this is the most effective in detecting 2.223 MeV and higher-energy γ rays. Future research will focus on developing strategies to optimise the scintillators to better enhance the detection capabilities of hydrogen in spent nuclear fuel.

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