Journal of Physics: Conference Series **3130** (2025) 012020 doi:10.1088/1742-6596/3130/1/012020

Position-sensitive neutron detection via a capture γ-ray proxy for water assay in spent nuclear fuel

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Abstract. The potential to exploit 2.223 MeV capture γ rays for the position sensitive detection of neutrons as a proxy for water in spent nuclear fuel is described. Previously, we demonstrated from a modelling perspective that the extent to which water might be dispersed in spent nuclear fuel following interim storage in ponds might be implied via this proxy, as opposed to direct detection of the neutron(s) itself. Position-sensitive neutron detection via secondary gamma-ray production can be advantageous for the detection of media that attenuate neutrons whilst being relatively transparent to γ rays, especially, for example, in situations where hydrogenous liquids might enhance criticality risk in heterogeneous waste mixtures. However, precisely mapping hydrogen concentration based on neutron attenuation directly can be impeded by the indirect nature of most neutron-based interactions whilst the coincident signatures of neutrons associated with spontaneous fission that are de rigueur for safeguards assay are not always viable. In this work, an experimental testbed exploiting hydrogenous foams, for example, Nylon-12, as analogues for dispersed water in fuel-containing materials, and 316L stainless steel metal foam as analogues for the fuel-containing materials was developed. These materials were used to simulate the emission and transmission of γ rays in a mix of fuel and water. The results show that 2.223 MeV γ rays are a viable indicator of detecting water in spent nuclear fuel.

1. Introduction

The detection and characterisation of water in spent nuclear fuel (SNF) is essential for nuclear safety and regulatory compliance. Methods suggested have often depended on the direct detection of neutrons, which can be effective in some scenarios but pose potential challenges when handling SNF [1-3]. Some of the reported challenges include neutron attenuation properties [4], detector response in harsh radiation environments [5], and the complexity of neutron interactions [4]. This can make it difficult to detect hydrogenous materials like water that may pose criticality risks in heterogeneous waste mixtures. There is therefore a need for the advancement of detection techniques that offer improved sensitivity and accuracy for water detection and characterisation in SNF. Progress in the development of neutron detection techniques for various applications is well documented [4].

The H(n,γ) method [2] is a potential option for nuclear assay allowing for position-sensitive neutron detection using capture γ rays. The method exploits the fact that characteristic γ rays emitted can serve as distinct markers of neutron capture events. By analysing the observed spectrum of these γ rays, it is thus possible to determine the position and the number of neutron interactions that occurred. This approach helps in detecting and quantifying certain nuclides, such as hydrogen in water present in SNF assemblies and fuel debris. Binnersley et al. [1] demonstrated through simulations the detection of 2.223 γ rays in nuclear fuels in order to ascertain the presence and mass of water. Their approach was shown to be more practicable than direct detection of neutrons for this purpose.

In this preliminary study, we further investigate the concept of position-sensitive detection using a capture γ-ray proxy for water assay in SNF using experimental and simulation approaches. This is achieved using hydrogenous foams, such as Nylon-12, to replicate water and 316L stainless steel foams to represent fuel containing materials. These analogues were chosen for their neutron capture properties and mean free path close to SNF as well as their ability to produce the characteristics 2.223 MeV γ rays. By manipulating cell density and composition of foam analogues, we aim to achieve neutron propagation and capture in a controlled geometry using a californium-252 sealed source.

2. Materials and methods

The experimental setup utilises hydrogenous and porous metal foams as analogues to simulate dispersed water and fuel-containing materials. A common moderator material included in the study is high-density polyethylene ($[C_2H_4]_n$) with a density of 0.96 g/cm^3. Mesh analogues of Nylon-12 ($[C_{12}H_{23}NO]_n$) containing a high concentration of hydrogen atoms and produced by additive manufacturing processes were also explored. The hydrogen atoms serve as neutron moderators and capture agents, similar to the neutron interaction characteristics of water molecules. Whilst solid Nylon-12 has a density of 1.01 g/cm^3, the mesh structure was designed to alter the effective density. This was done to accurately represent the scattering and absorption characteristics observed in SNF assemblies. The hydrogen density of the HDPE and Nylon-12 is 4.13×10^{22} and 3.60×10^{21} atoms/cm^3 respectively. A 316L stainless steel metal foam composed of Fe, Cr, Ni, Mo, Mn, Si, N, P, C, and S in varying proportions [6] having a mass of 56.4 g, 85 mm × 85 mm × 25 mm dimension, and density 0.44 g/cm^3 was also used as a suitable metal analogue to simulate the neutronic behaviour in spent nuclear fuel. The use of 316L stainless steel foam was considered as a material of choice in this study due to its compositional similarity to SNF clad materials [7].

To detect capture γ rays emitted from candidate target samples containing hydrogen, a thallium-doped sodium iodide (NaI(Tl)) scintillator (Bicron, Netherlands) with a crystal size of 80.9 mm ∅ × 222.3 mm L was used. The detector was connected to a laptop via USB to a digital photomultiplier tube base. The measurement was controlled by MAESTRO Multichannel Analyser Emulator Software (Ametek Ortec, USA) with the high voltage set to 850 V. To ensure accurate energy assignments, the detector was calibrated using standard point sources comprising ^{60}Co, ^{137}Cs, and ^{152}Eu. The energy resolution of the detector was evaluated as 6.97% at 662 keV, satisfying the manufacturer's requirements of <7.5% for optimal efficiency. The detector was shielded with lead blocks to reduce background radiation and positioned perpendicular to a water-filled tank in which a ^{252}Cf neutron source (5.05 MBq and 5.89×10^5 n/s) is housed. Afterwards, the target samples were angled at 45° from the source tank to optimise the detector's efficiency in capturing the γ rays. All measurements were taken for a live

time of 86400 s, except for energy calibrations which took 300 s for each of the radioactive sources used.

GEANT4 simulations [8] were performed to model the detector response to γ rays emitted from neutron capture events. The simulations used material parameters, detector geometry, and absolute energy deposition methods to anticipate the detector response to 2.223 MeV γ rays. A

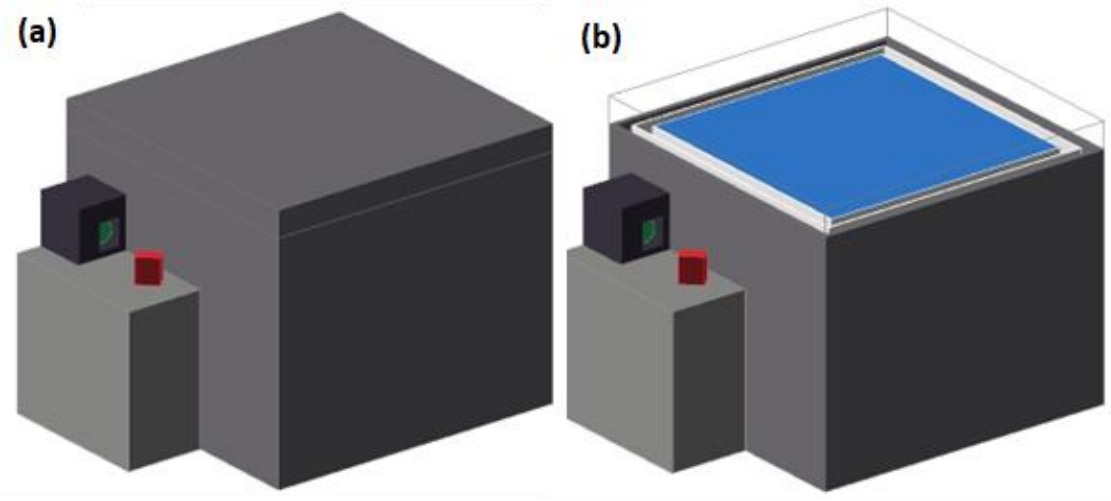

Figure 1. GEANT4 3D model of the experimental set-up. Fig. 1(a) shows the steel lid on the tank whilst Fig 1(b) affords a view into the construction of the tank. A NaI(Tl) detector (green) is shielded within a lead block (black). The target (red) material is selected and balanced on a stainless-steel stand (light grey). The tank is comprised of a steel (dark grey) security layer, a double-walled fibreglass (white) tank, and an internal volume filled with water (blue).

3- dimensional model of the experimental setup is shown in Fig. 1. The source resides inside the inner water filled fibreglass tank and an outer fibreglass tank acts as a bund. A security steel box surrounds the arrangement. There are two air gaps as indicated. The ^{252}Cf source is moved to the face of the tank closest to the target when in use.

3. Results

3.1 Detection of 2.223 MeV with and without HDPE

Fig. 2 shows the spectra produced experimentally from a (n,γ) reaction when the source is exposed. Fig. 2(a) shows the spectra produced when there was no target placed in front of the neutron tank. A 2.223 MeV γ-ray which emanates from water in the ^{252}Cf source tank is detected from the ^{1}H(n,γ)^{2}H reaction. The result indicates that the 2.223 MeV γ rays are a viable indicator for detecting water in SNF. Subsequent measurements with 140 mm × 100 mm × 50 mm HDPE block used as a target sample gave similar spectra shown in Fig. 2(b). The 2.223 MeV peak is again observed. Without the HDPE target, a net peak area of 30986 counts of 2.223 MeV γ rays at 0.39 counts/s was observed. The addition of HDPE gave 81527 counts at a higher count rate of 1.024 counts/s.

3.2 Detection of 2.223 MeV in foam analogues with moderation

Fig. 3 shows the spectra of the (n,γ) interaction in Nylon-12 and 316L stainless steel analogues.

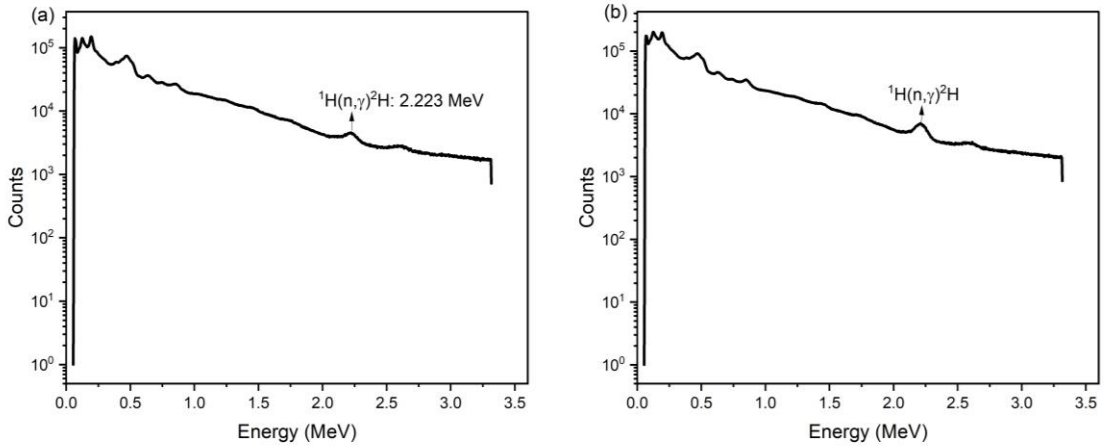

Figure 2. γ-ray spectra of 2.223 MeV corresponding to $^{1}H(n,\gamma)^{2}H$ reaction, which emanates from water (a) without HDPE and (b) with HDPE exposed to a ^{252}Cf source

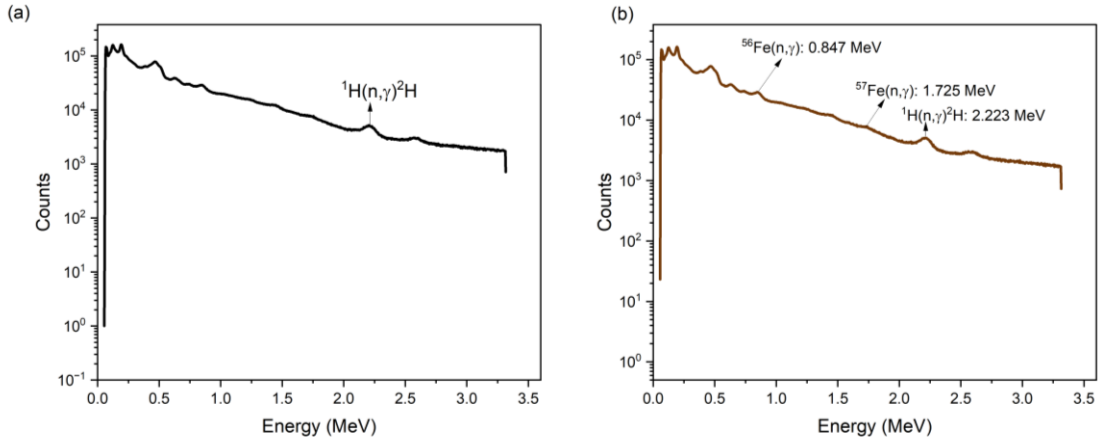

Figure 3. γ-ray spectra corresponding to (a) Nylon-12 and (b) 316L stainless steel foams.

Both spectra reveal a similar pattern of peaks and show the 2.223 MeV γ-ray line produced by H(n, γ) interactions. A net peak area of 43344 counts of 2.223 MeV γ rays of hydrogen was observed in Fig. 3(a) when a Nylon-12 target was used with an HDPE as a moderator. For the 316L stainless steel material (Fig. 3(b)), a peak area of 44424 counts was recorded at a rate of 0.56 cps. It is evident that the count rates recorded using the foams are much lower than those recorded using the HDPE block, which is attributed to the size of the target material used. In Fig. 3(b), below 2.2 MeV, ^{56}Fe isotope is seen along the 847 keV and 1.725 MeV γ lines [9]. This is due to the presence of heavy metals such as Fe, Cr, and Ni in the 316L stainless steel foam.

3.3 Simulation of 2.223 MeV in foam analogues

Geant4 simulations were carried out modelling the experimental set-up used in this work. 100 million neutron decays from ^{252}Cf were simulated. Using an idealised NaI(Tl) scintillator with HDPE, Nylon-12 and 316L stainless steel targets, the number of γ rays incident on the detector is recorded.
Fig. 4 shows the full energy peak gamma spectra observed using each target material. The

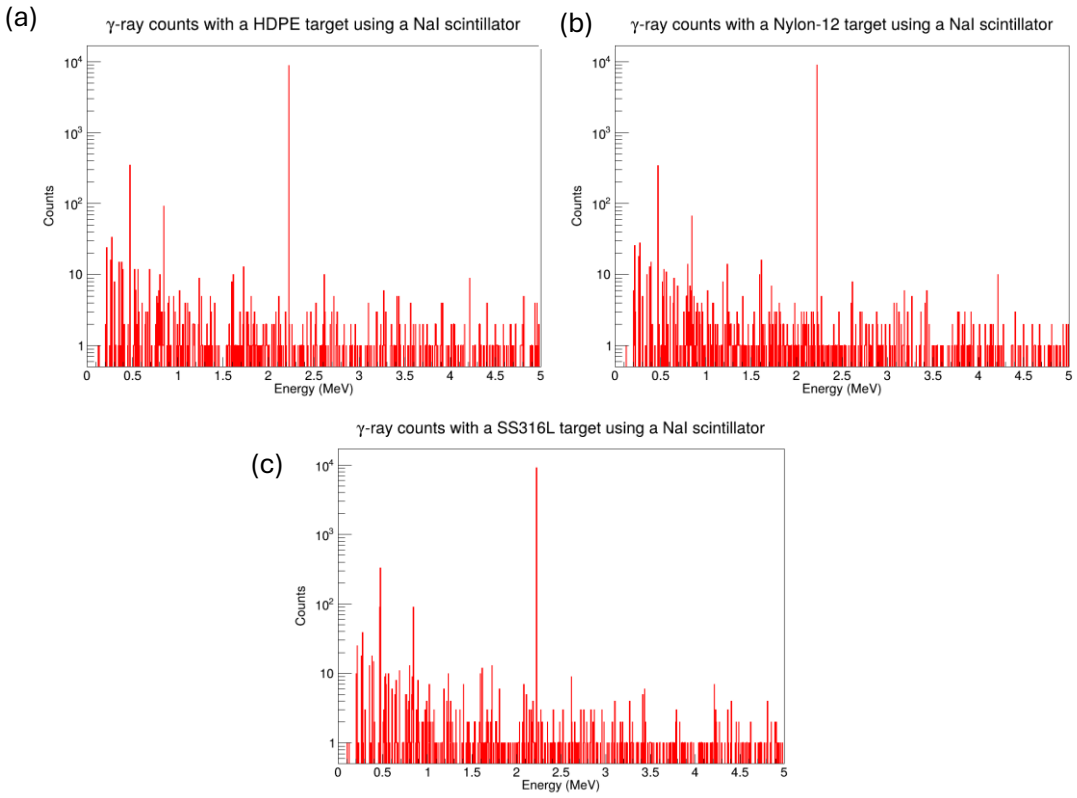

Figure 4. Simulated γ-ray energy spectra from (n,γ) reaction in (a) HDPE (b) Nylon-12 (c) 316L Stainless steel targets. The bin size for each data plot equals the number of channels in the experimental data.

number of 2.223 MeV γ-rays observed during the simulations were 8910, 8924, and 9078 for HDPE, Nylon-12, and 316L respectively. Similarly to the experimental measurements, the small target means a limited number of neutrons strike the target. This results in only minor differences between measurements, suggesting that most 2.223 MeV γ-rays originate from the water surrounding the source. Additionally, the mass attenuation coefficient of lead for photons is lowest at 2.2 MeV further supporting the water in the tank as the origin of the 2.223 MeV γ-rays [10].

4. Conclusion

The potential use of 2.223 MeV capture γ rays for position-sensitive detection via neutron capture in SNF has been studied. The use of a NaI(Tl) scintillation detector is effective for detecting the characteristics 2.223 MeV γ rays. However, its spatial resolution is inherently limited due to factors such as standardised crystal dimensions and photomultiplier size,

scattering, modest signal-to-noise ratio (energy-resolution), etc. These limitations can hinder the ability to resolve fine spatial details within spent nuclear fuel (SNF) matrices for neutron capture events. Nylon 12, with its 2.223 γ-ray peak successfully simulates hydrogen presence in water, whereas the 316L stainless steel reflects the structural components of SNF. This supports the methodology of this study in using these analogues to better understand neutron-γ capture in SNF. This preliminary study aimed to develop an end-to-end capability, combining experimental and simulation methods. These measurements will be used to optimise future work, not least free from the constraints of the ^{252}Cf inside the tank. Currently, it is unclear how the 2.223 MeV γ peak in the experimental and simulated work contributes to (n,γ) reactions in the water-filled tank. Future research will focus on improving the experimental testbed by replacing the scintillator with an HPGe detector and exploring additional analogues with similar hydrogen densities close to water. More ^{252}Cf point sources will be utilised to improve neutron propagation. Simulations will be run to examine various fuel assemblies in heterogeneous arrangements, considering the complete response function of the detector.

Acknowledgements

This work was funded as part of the Engineering and Physical Sciences Research Council United Kingdom's grant EP/X038327/1. We also acknowledge support from the Nuclear Decommissioning Authority for providing PhD funding to B Green.

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