

Detection of 2.223 MeV γ -rays for Water Identification in Spent Nuclear Fuel Assemblies

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Abstract— The detection of 2.223 MeV γ -ray emissions resulting from neutron capture on hydrogen, to monitor water presence in spent nuclear fuel (SNF) assemblies is described. Water detection is crucial because its ingress into dry storage systems or other containment environments poses safety risks and potential criticality concerns. This study thus aims to explore the use of the characteristic 2.223 MeV γ -rays for reliable detection of water in SNF assemblies, with a focus on how neutron interactions, such as scattering and absorption, impact γ -ray production and detection in different fuel configurations. This approach will provide a non-invasive and accurate approach for nuclear monitoring and maintaining nuclear safety. To achieve this objective, a Mirion Technologies' GR5021 high-purity germanium (HPGe) detector has been used to measure γ -ray emissions from neutron capture events under controlled conditions. Californium-252 (Cf-252) neutron sources were used to provide a well-characterised neutron field representative of those in SNF environments. Various test configurations were created using high-density polyethylene (HDPE) slabs and 3D-printed metal analogues. The HDPE slabs serve as analogues to simulate the neutron moderation characteristics of water, whereas the metal slabs serve as analogues for fuel-containing materials. Results showed the consistent detection of 2.223 MeV events, with γ -ray counts responding consistently with expectations as material thickness was increased. Overall, the results suggest that the 2.223 MeV γ -ray has potential as a reliable and sensitive indicator for detecting water in SNF assemblies, with significant implications for nuclear safety and fusion reactor environments.

Keywords— neutron capture, hydrogen, water, γ ray, spent nuclear fuel.

I. INTRODUCTION

THE long-term management of spent nuclear fuel (SNF) remains a central priority in the nuclear industry's commitment to safety and environmental responsibility. A key aspect of both wet and dry storage systems is to ensure that containment integrity is maintained, where the unintended presence or absence of water might introduce significant safety anomalies. In dry casks, water ingress poses risks such as corrosion, radiolysis, and most importantly, a decrease in the reactivity margin due to the neutron moderation properties of hydrogen. Conversely, in pool storage systems, confirming the continued presence of water is essential for adequate shielding and effective decay heat removal [1, 2].

Among the various methods for monitoring water in spent nuclear fuel (SNF) environments, passive γ -ray spectroscopy is

a non-invasive and highly-sensitive possibility. A particularly distinctive signature is the 2.223 MeV γ -ray emitted when thermal neutrons are captured by hydrogen nuclei [3, 4]. Since neutrons are emitted from SNF via spontaneous fission and (α ,n) reactions, this γ -ray line has potential as a direct indicator of hydrogenous materials, such as water. Previous research [3,5,6] has demonstrated the feasibility of this approach for safeguards and verification applications, with studies showing its effectiveness in detecting water signatures in storage pools and intact fuel assemblies.

Despite these advances, much of the existing research has been limited to simulations or homogeneous configurations, with there being relatively little experimental validation in heterogeneous fuel-like systems. This gap is significant because realistic SNF consignments can comprise complex geometries and diverse material compositions, influencing neutron scattering, absorption, and subsequent γ -ray production, significantly [2]. Addressing this challenge is essential for both fission and, potentially, fusion safety applications. Progress in the development of neutron detection techniques for various applications, as well as the methods of γ -ray measurement of hydrogen are well documented [1,7].

In this study, we investigate the detection of 2.223 MeV γ -rays experimentally, as a proxy for water in SNF. Using a high-resolution HPGe detector and a ²⁵²Cf neutron source, we explore how different test materials, including high-density polyethylene (HDPE) and 3D-printed metal analogues, impact neutron capture rates and γ -ray emission under controlled conditions.

II. EXPERIMENTAL METHOD

Experiments were conducted with a ²⁵²Cf source stored in 1 m × 1 m × 1 m water. Hydrogenous and metal analogues were used to simulate water dispersed in fuel-containing materials. High-density polyethylene (HDPE), with a density of 0.96 g/cm³, served as a standard moderator material. The hydrogen atomic number density in the HDPE was 4.13 × 10²² atoms/cm³. For the metal-based analogues, two different zinc-based materials were studied: zinc resin slabs measuring 15 cm × 21 cm × 6 cm, with a density of 3.60 g/cm³, comprising 88.2% zinc and 11.8% resin, produced through advanced manufacturing processes. Additionally, commercially available natural zinc sheets (Metal Sheets Ltd, UK), made of 99.5% pure zinc with

the remaining 0.5% consisting of Cu, Ti, Al, and other trace elements such as Fe, Cd, Sn, Mn and Mg, were investigated as a target sample. The zinc sheet dimensions were 300 mm × 200 mm × 6 mm.

To detect the capture γ -rays emitted from the sample materials described above, a portable GR5021 high-resolution reverse-electrode germanium (REGe) detector (Mirion Technologies, CANBERRA, Olen) with a crystal size of 90 mm \varnothing × 165 mm and a CP5-PLUS-F-RDC-4 cryostat was used. The data acquisition was controlled by Genie 4.0 Software (Mirion Technologies). To ensure accurate energy assignments, the detector was calibrated using a standard range of point sources comprising ^{60}Co , ^{137}Cs , ^{241}Am , ^{22}Na , and ^{152}Eu . The energy resolution of the detector was evaluated as 1.91 keV (FWHM) at 1.3 MeV.

To reduce background radiation, the REGe detector, albeit resistant to neutron damage, was shielded with lead blocks and positioned perpendicular to a water-filled tank housing a ^{252}Cf neutron source with an activity of 3.7 MBq and a neutron emission rate of approximately 4.3×10^5 n/s. For each test configuration, the target samples were placed 20 cm from the detector and positioned 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 54000 s.

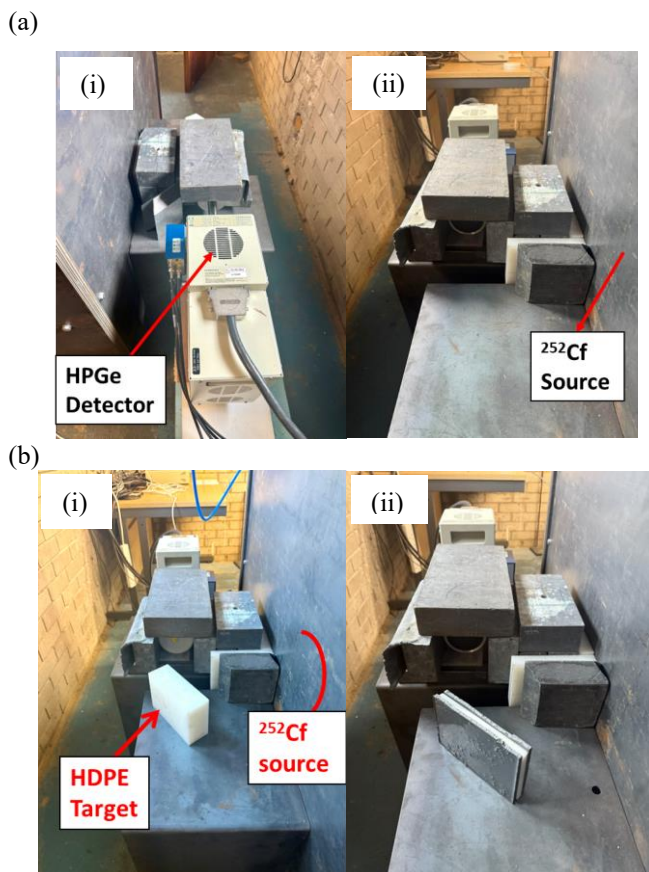
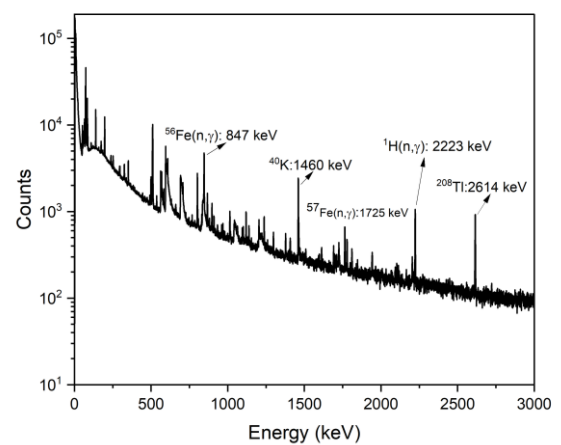


Fig. 1. The experimental setup for the detection of the 2.223 MeV γ -ray from (a) background radiation when the ^{252}Cf source is exposed, (i) with the detector shielded in a lead cave arrangement, (ii) with the end-cap view and source position from the tank, and (b) from the target sample placed at 45° from the source tank to optimise the detector's efficiency in capturing the γ -rays. The HDPE target in (i) is replaced with a zinc resin (ii) sandwiched with a 10 mm-thick HDPE. The acquisition time for each measurement is 15 hours.

III. RESULTS

Fig. 2 presents the γ -ray energy spectra obtained from the experimental measurements. Fig. 2a shows the background energy spectrum from the (n, γ) reaction when the source is exposed. A 2.223 MeV γ -ray peak due to the water in the ^{252}Cf source tank is detected arising from the $^1\text{H}(n,\gamma)^2\text{H}$ reaction. Isotopes of ^{56}Fe and ^{57}Fe are observed in the spectrum and are thought to be due to the material composition of the source tank and the steel stand for the target in agreement with previous reports [8]. Other isotopes, e.g., ^{40}K , ^{208}Tl are evident from background radiation in the environment. Fig. 2(b) shows the spectrum measured from a 5 cm-thick HDPE slab when placed in front of the neutron source. A 2.223 MeV γ -ray peak is observed resulting in a peak area and intensity approximately twice that recorded in Fig. 2a without a target material.

(a)



(b)

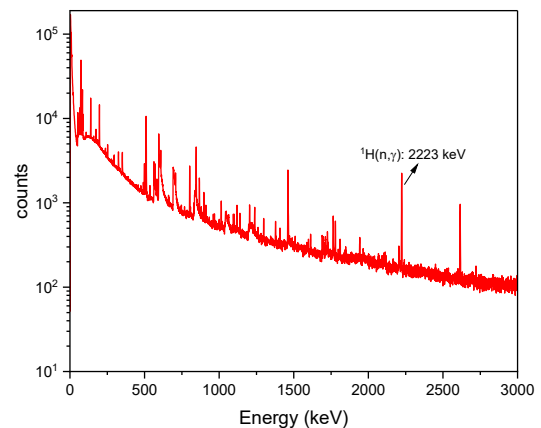


Fig. 2. γ -ray spectra of 2.223 MeV from (a) a background measurement (b) a 5-cm thick, 14 cm × 10 cm HDPE target, following 15 hours neutron exposure.

Fig. 3 shows the spectra produced when the 3-D analogues, that is, Zn resin and Zn sheets, were exposed to the neutron source. Fig. 3a shows the spectrum for the Zn resin which is consistent with that of Fig. 2. However, a reduction in counts is observed for the 2.223 MeV γ -ray peak in comparison with the HDPE spectra. This is as a result of the material composition and geometry of the target. Fig. 3b gives the spectra of the analogues at varying thicknesses. Each of the analogues was moderated with 10 mm of HDPE.

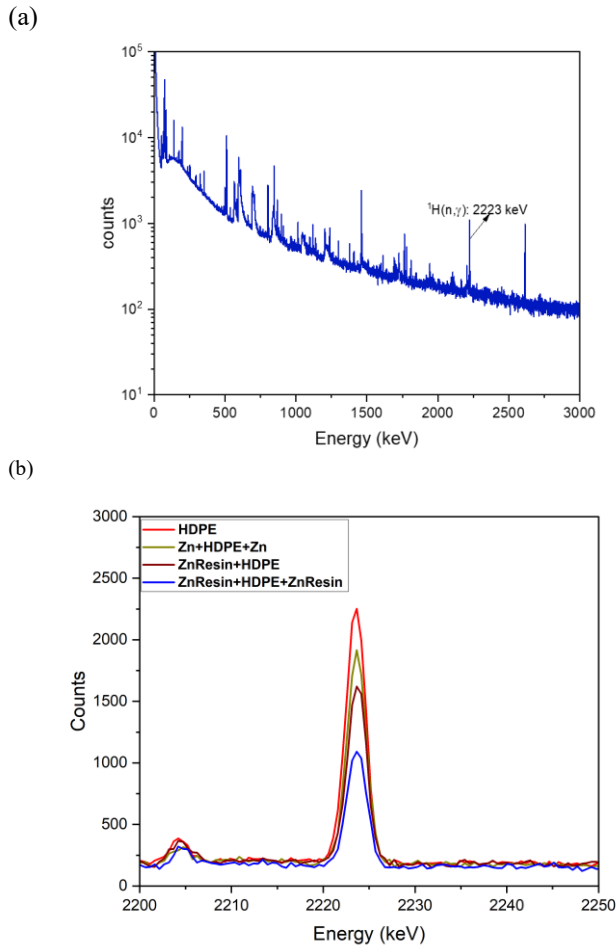


Fig. 3. (a) γ -ray spectra of 2.223 MeV corresponding to 10 mm-thick of Zn resin (105 mm \times 150 mm) sandwiched with 10 mm of HDPE (b) 2.223 MeV γ -ray peak of fuel analogues at varying thicknesses. The dimensions of the Zn sheet are 300 mm \times 200 mm \times 6 mm.

Fig. 4 presents the measured energy spectra of HDPE at various thicknesses. The observed increase in the 2.223 MeV γ -ray peak arises from neutron moderation in hydrogen-rich HDPE, where elastic scattering slows neutrons from the ^{252}Cf source into the thermal range, leading to capture and emission of the 2.223 MeV line. At very large thicknesses, the peak intensity is expected to approach saturation due to self-shielding and neutron leakage.

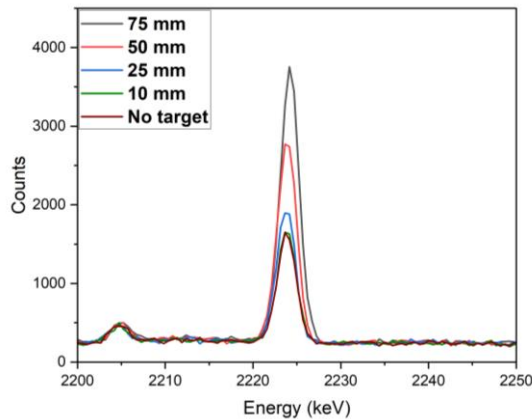


Fig. 4. γ -ray spectra of the 2.223 MeV γ -ray peak for various thicknesses of HDPE. The data have been truncated to show the regions of the 2.223 MeV γ -ray peak of hydrogen.

IV. CONCLUSIONS

The detection of the characteristic 2.223 MeV γ -ray from neutron capture of hydrogen in material analogues under consideration for SNF has been demonstrated. Consistent trends were observed across the measurements, confirming the reliability of the experimental approach. The variations in the γ -ray peak intensity with material thickness highlights the sensitivity of the technique to material composition and geometry. These results validate the use of hydrogenous and metal analogues for investigating neutron moderation and γ -ray production processes relevant to SNF scenarios.

Future research will focus on extending this study through simulations of SNF assemblies with varying water content, representing more realistic storage and accident conditions. In addition, systematic investigations will quantify the effect of foam cell density on neutron moderation and γ -ray emission. Experimental benchmarking using analogues representative of SNF will provide validation for computational results. Geometry-specific measurements using a number of bare ^{252}Cf neutron point sources will be conducted to further refine test configurations.

V. ACKNOWLEDGMENT

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