Prompt γ-ray techniques based on neutron capture in hydrogen for the detection of water in nuclear legacy environments: a review

Brendan Green[1, *], Damilola E. Folley[1], Stephen Croft[1], David I. Hambley[2], Andrew Kennedy[1], Robert Mills[2], Lee Packer[3] and Malcolm J. Joyce[1]

[1]School of Engineering, Lancaster University, Lancaster LA1 4YW, United Kingdom

[2]National Nuclear Laboratory, Sellafield, Cumbria, CA20 1PG, United Kingdom

[3]United Kingdom Atomic Energy Authority, Culham Science Centre, Abingdon, Oxfordshire, OX14 3DB
United Kingdom

*Corresponding author. School of Engineering, Lancaster University, Lancaster. *E-mail address:* b.j.green@lancaster.ac.uk (B. Green)

Abstract

Water present in nuclear legacy materials, such as spent nuclear fuel and fuel element debris, can impact both the chemical and structural stability of these materials. Subsequently, the suitability of these materials for disposal in geological repositories is degraded. Water ingress increases the potential for radioactive material to be dispersed and for neutronic properties to be augmented due to changes in neutron moderation by the constituent hydrogen. This risk necessitates the detection and quantification of water in what can be complex nuclear contexts. Neutron capture γ-ray analysis is of particular interest in this regard: this review provides a summary of the methods and equipment used to apply this to the problem of water assay in nuclear materials. Whilst relatively few directly relevant reports have been published concerning the measurement of water in spent nuclear fuel, the detection of hydrogen and the measurement of its characteristic 2.223 MeV γ-ray following the $^{1}H(n,\gamma)^{2}H$ reaction has been studied more extensively. This approach for detecting water in composite materials and its impact on PGAA of other materials is considered. The review concludes with the current applications of the capture γ-ray analysis technique and the potential of its use for water ingress assay nuclear materials at Fukushima Daiichi and Chernobyl.

Glossary

1F: Fukushima Daiichi Nuclear Power Station

AGR: Advanced Gas-cooled Reactor

BEGe: Broad-Energy Germanium

BWR: Boiling Water Reactor

ChNNP4: Chernobyl Nuclear Power Plant unit 4

FCM: Fuel Containing Material

FP: Fission Product

HPGe: High-Purity Germanium

LMNF: Lava-like Materials containing Nuclear Fuels

LWR: Light Water Reactor

MCNP: Monte Carlo N-Particle

MOX: Mixed-Oxide fuel

NPS: Nuclear Power Station

NSC: New Safe Containment

PFR: Prototype Fast Reactor

PGAA: Prompt γ-ray Activation Analysis

PWR: Pressurised Water Reactor

RPV: Reactor Pressure Vessel

SNF: Spent Nuclear Fuel

1. Introduction

1.1 Water in nuclear legacy environments

When nuclear fuel is discharged from a power reactor, fission products (FPs) and actinides from the neutron induced nuclear reactions render it highly radioactive and heat generating. These issues complicate the ease with which spent nuclear fuel (SNF) is handled and must be considered in the safe handling, storage, transport or processing of the fuel. The differing cooling and shielding requirements determine the precautions and ease of these operations. Temporary storage is employed to allow for the reduction of decay heat and radioactive emissions over several decades following reactor discharge. Often this has involved initial storage of the fuel intact under water in ponds which is an approach that has been adopted all over the world. To date, in the UK, no SNF has been disposed of permanently, and it exists either as intact fuel stored in water- or air-cooled containments, or as separated wastes from reprocessing,

Today, some 70 years after the first nuclear power reactors were built, many countries are finalising strategies for the permanent disposal of their high active waste and SNF. These usually involve consignment in a geological facility, either deep or near-surface. For this to be achieved successfully, proof of compliance of the waste with a disposal safety case will be required. This usually mandates that the materials destined for consignment in a disposal facility have been rendered unreactive chemically. Materials must be insoluble, and thus not vulnerable to corrosion which may lead to the dispersion of radioactivity earlier than the facility design specification allows. Measurements and other mitigating defences are also put in place, negating the potential for inadvertent nuclear criticality.

The strategy to store spent fuel in ponds has risked water ingress into fuel pins, where the can used to consign the fuel has failed, and penetration by water into the fuel itself, where the clad has been compromised because of corrosion or cracking. Water exacerbates the risk associated with chemical reactivity in SNF and introduces a source of moderation that either must be accounted for or excluded prior to disposal. Water in SNF presents an additional criticality safety risk which must be accounted for when considering long term dry storage. Examination of the quantities of water required for criticality in different storage conditions and general criticality safety has been, and continues to be, of utmost importance to the nuclear industry [1-3]. The strategy for SNF affected in this way is likely to be that it must be detected and that this fuel be diverted for alternative processing that removes the water.

The requirement for detection has resulted in a variety of possibilities for what has become known collectively in some contexts as *poolside monitoring*, which comprises a variety of potential techniques; relevant non-destructive methods include X-ray computer-aided tomography, gravimetric assessment, and neutron assay etc [4]. This review concerns one such method: *neutron capture γ-ray emission from hydrogen*. The structure of this review is as follows. First the motivation for measuring water content in SNF is established. Next the development of prompt gamma activation analysis (PGAA) methods is discussed before considering PGAA specifically for hydrogen detection. Finally, the apparatus and methods for detecting moisture and water as well as additional applications of PGAA were examined.

1.2 The motivation for moderated neutron capture γ-ray water assay in SNF

In 2022, Binnersley et al. [5] used MCNP simulations to demonstrate three possible approaches to measuring water present in intact spent nuclear fuel: neutron detection and two approaches based on PGAA with neutrons, with and without moderation. The three methods presented attempt to interrogate the quantity of water by non-destructive means, thereby presenting an opportunity to assess water content, *in situ*. Multiple spent fuel types were considered including advanced gas-cooled reactor (AGR), mixed oxide (MOX) fuel from the Dounreay Prototype Fast Reactor (PFR) and high burn-up MOX fuel from a Gen III, light water reactor (LWR-MOX). The study considered a variety of fuel types allowing the applicability of each method on different fuel geometries and radionuclide content to be explored.

Method 1 exploited fast neutrons incident on spent fuel with neutron detectors positioned behind the fuel to record the test results. In these simulations AGR fuel was subject to monoenergetic 14 MeV neutrons and MOX and LWR-MOX assemblies to monoenergetic 2.45 MeV neutrons. No moderation was placed between the neutron source and target assembly. For the AGR fuel, variation of the water content exhibited a considerable drop in the transmission of fast neutrons from a level of 10 g to 200 g of water. The fast neutron flux through PFR and LWR-MOX fuels exhibited a limited change until the water mass had reached between 300 g and 400 g. Due to the high burn-up in LWR-MOX fuel, the high neutron emission from the fuel impeded the effectiveness of this technique significantly.

Methods 2 and 3 were both based on PGAA of the emission of a 2.223 MeV γ-ray following neutron capture in the $^{1}H(n,\gamma)^{2}H$ reaction. To detect the emitted γ-rays, the simulations used broad energy germanium (BEGe) detectors positioned around the fuel assembly. Method 2 did not moderate the 2.45 MeV neutrons from the source. The photon detection rates from the AGR assembly and the PFR assembly were equivalent to the background rate below 200 g and 2000 g of water, respectively. From these observations it was clear that, in the absence of moderation, neutrons did not produce a signal of sufficient magnitude to identify water, particularly in the PFR case. The detector and fuel assembly arrangements for methods 1 and 2 are reproduced in Figure 1. The AGR assembly was surrounded by 10 detectors whilst the PFR assembly used 9.

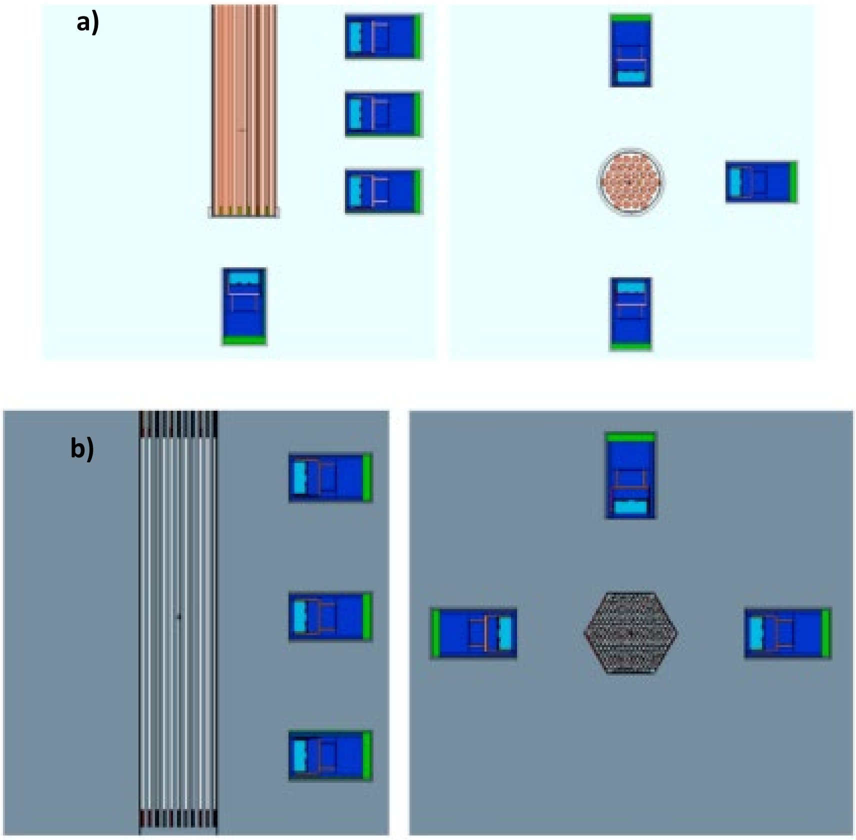

Figure 1: a) MCNP representation of Ge detectors (in blue) around the AGR welded assembly model and b) MCNP representation of Ge detectors around the PFR assembly. These arrangements were used in methods 1 and 2 by Binnersley et al. [5] with an unmoderated neutron source. Figure taken from [5].

Method 3 introduced a cylindrical graphite sleeve to serve as a moderator around the fuel assembly as this was considered a feasible adjustment to a potential monitoring facility. The simulation geometry of the AGR assembly surrounded by the graphite moderator is seen in Figure 2. This improves the neutron capture cross-section for hydrogen to the thermal neutron cross-section of 0.3326 +/- 0.0007 b [6, 7] over and above that in method 1. As a result, method 3 yielded a detection rate increase of 20 to 60 times greater than the unmoderated case, allowing the 2.223 MeV γ-ray signal to be observed down to water masses of 10 g [5]. The signal-to-background ratio was greater than 0.1 when the water mass was ≥ 50 g, whereas the unmoderated system required a water mass ≥ 200 g to achieve a similar response.

Method 1 demonstrated a better sensitivity to the presence of water for water masses over 100 g when compared to method 2. However, the detection of γ-rays produced by moderated neutrons explored in method 3 demonstrated the greatest sensitivity across all masses of water simulated. Method 3 also yielded the greatest disparity between the presence and absence of water in the fuel assembly. All three methods are expected to have difficulty measuring small quantities of localised water inside pin voids. The higher intrinsic neutron emission rate of the PWR-MOX fuel is predicted to limit the effectiveness of all techniques explored. Pulsing the neutron source might improve sensitivity by allowing the response from delayed decay products to be suppressed to yield a relatively uncontaminated neutron capture γ-ray spectrum. Pulse subtraction may also reduce the impact of the higher neutron emission rates in PWR-MOX by, for example, a neutron chopper or electronically pulsed D-D neutron generator.

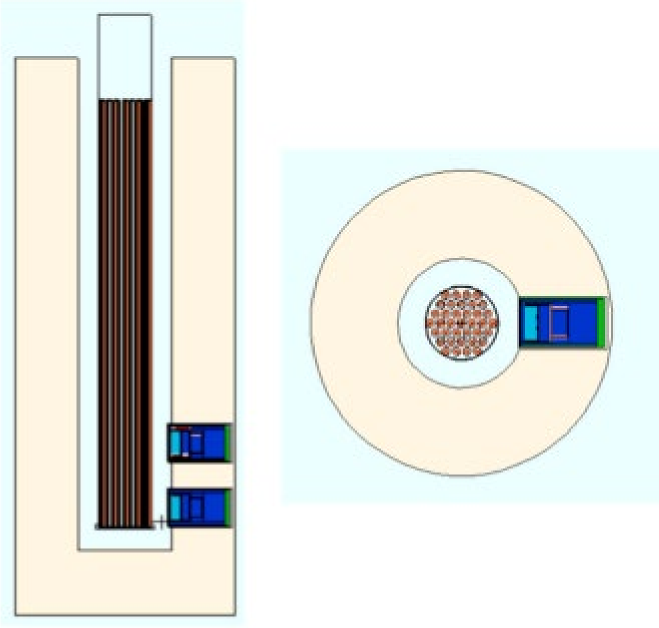

Figure 2: Welded AGR assembly surrounded by graphite moderator with two germanium detectors to count gamma rays produced by PGAA [5], elevation (left) and plan view (right). Figure taken from [5].

Simulations by Binnersley et al. [5] demonstrated that water detection by PGAA provides greater sensitivity to small quantities of water than fast neutron scattering. The characteristic 2.223 MeV photon produced by the $^{1}\text{H}(n,\gamma)^{2}\text{H}$ reaction is used to indicate the presence of the constituent hydrogen in water with the potential to enhance the approach with Compton suppression. The single γ-ray emission from hydrogen in an area of the spectra where there is little potential contamination is a further advantage, when compared to the more complex spectra of other isotopes. As a technique for material analysis, the PGAA response is relatively independent of the physical form of the target isotope, i.e., liquid or solid. PGAA is non-destructive, and measures close to the total elemental content due to the highly penetrative nature of neutrons and γ-rays, given the relative isotopic abundance of hydrogen is known, *a priori*, or can be assumed.

It is expected that the gamma continuum from SNF would make detection of a 2.223 MeV γ-ray signal challenging. Binnersley et al. do not consider the passive gamma emissions resulting from the decay of FPs and spontaneous fissions from plutonium, curium and californium. The prompt spectrum generated in MCNP5 by Binnersley et al. shows significantly fewer counts above 2 MeV allowing for the observation of 2.223 MeV γ-rays from hydrogen [5]. Figure 3 shows a typical spectrum from a PWR fuel assembly which has been cooled for 12 years and has a burn-up of 47 GWd/tU [8]. The potential detection limit of hydrogen amongst the SNF gamma continuum is therefore dependent on the burn-up of the fuel material and the resulting presence of FPs and heavy isotopes.

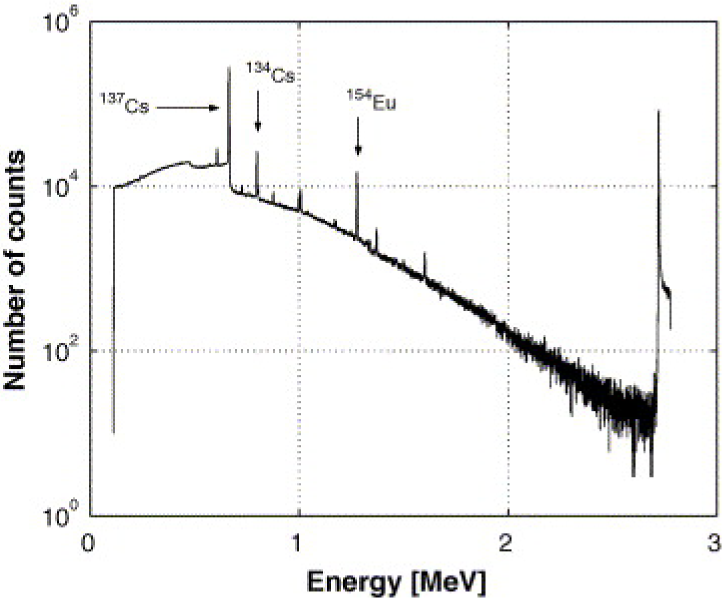

Figure 3: Gamma spectrum of a PWR fuel assembly with a burn-up of 47 GWd/tU and a cooling time of 12 years [8]. Figure taken from [8].

2. Neutron-capture γ-ray emissions from hydrogen

2.1 First observations

Almost 100 years since the initial discovery of γ-ray emission by neutron capture, the use of PGAA and specifically the measurement of the 2.223 MeV γ-ray from capture on hydrogen could prove an important tool for a variety of applications.

Neutron interactions with nuclei can be categorised as either *scattering* or *absorption*, with two primary sub-categories under the latter: *capture* and *fission*, with radiative capture being of particular interest to this review. Thermal neutrons usually have a higher probability of capture by a target nucleus than fast neutrons which is reflected in the typical $1/v$ trend in the corresponding total neutron cross-section, where v is the neutron speed. Upon capture, the nucleus occupies an excited state with a γ-ray photon emitted typically within ~10^{-12} seconds and with an energy equivalent to the binding energy of the neutron [9]. Where the excited state has a much higher neutron binding energy, multiple γ-ray photons might be emitted to stabilise the nucleus via several intermediate states. The corresponding energy spectrum is characteristic of the isotopic composition of the target material in question. Therefore, an inherent advantage of PGAA is the ability to identify and measure quantities of different isotopes simultaneously given an effective and capable detector.

Radiative emissions from the bombardment of hydrogen by thermal neutrons were first documented in 1934 shortly after the discovery of the neutron [10]. Observations were made in two different laboratories just a few months apart [11, 12]. Building on measurements of neutron scattering in paraffin wax and liquid hydrogen, in which radiation was seen to be emitted at 120°-180° to the direction of incident neutrons, Lea [11] identified the radiation at these extreme angles to be γ-ray photons. At the time, the mechanism for this release of γ radiation had not been published or developed widely. However, Lea hypothesised correctly that the neutron combining with the proton of the hydrogen nucleus resulted in a γ-ray photon. Its energy was equivalent to the mass defect between deuterium and protium. Additionally, a reasonable estimate of the energy of the photon was made between 2-4 MeV based on a lead thickness of 3.4 cm necessary to absorb the emitted photons.

Fermi et al. [12] came to a similar conclusion following the irradiation of a wide range of elements by neutrons from a sealed glass tube containing beryllium powder and radon, a radiochemical (alpha,n) neutron source. Observations made following exposure to this source confirmed neutron capture in the nuclei of target materials. Many irradiated isotopes underwent α or β^- decay with no instance of β^+ decay recorded. However, Fermi et al. did not observe any γ-rays when irradiating hydrogen with neutrons most likely due to the neutrons being too high in energy to interact with hydrogen via capture interactions.

2.2. Prompt gamma activation analysis

Following the discovery of neutron capture γ emissions, but prior to the advent of research reactors, PGAA was done with radiochemical neutron sources placed in hydrogenous moderating materials such as paraffin [13]. When using a reactor, target materials are either placed inside the reactor and capture γ-rays observed via a collimator with a detector outside. More often a beam of neutrons is drawn from the reactor and a sample irradiated outside. Whilst radiochemical neutron sources such as californium-252 (^{252}Cf) generally have a lower neutron flux than a reactor, they can render mobile applications feasible. Gladney [13] provides a detailed review utilising PGAA and the techniques for measuring a variety of elements including the development and improvement of PGAA apparatus for the detection of hydrogen.

In 1966, Isenhour and Morrison [14] discussed a modulation technique for PGAA induced by moderated neutrons from a reactor with considerable detail of the apparatus they developed, being one of the first research groups to utilise γ-ray spectra from neutron capture reactions [15, 16]. Whilst others had used PGAA for material analysis, research optimising the technique had been limited otherwise. Several disadvantages surrounding current PGAA systems and the use of unmodulated and uninterrupted neutron sources were identified by Isenhour and Morrison including: spectrometry equipment with high resolution but without the required sensitivity for detailed material analyses; scintillation crystals that improved sensitivity but with reduced resolution; and the complication by other sources of γ-rays such as the inelastic scattering of fast neutrons [14].

A schematic diagram of the apparatus assembled by Isenhour and Morrison [14] to solve the issues they identified is shown in figure 4a with the detector and shield assembly shown in detail in figure 4b. Like Hamermesh and Hummel [17], the neutron chopper used a slotted cadmium sheet to control the bursts of thermal neutrons to the target. Magnetic pickups mounted perpendicular to the chopper edge experienced a change in magnetic flux as the slots passed them, sending a signal to a multichannel analyser (MCA). This allows the MCA to switch between memory storage to record prompt γ-ray emissions and background emissions separately to ensure the electronics are synchronised with the rotational frequency of the chopper. At the maximum chopper operating speed of 1000 cycles per second, 99% of delayed γ-rays from reactions with a half-life greater than 1 ms were excluded.

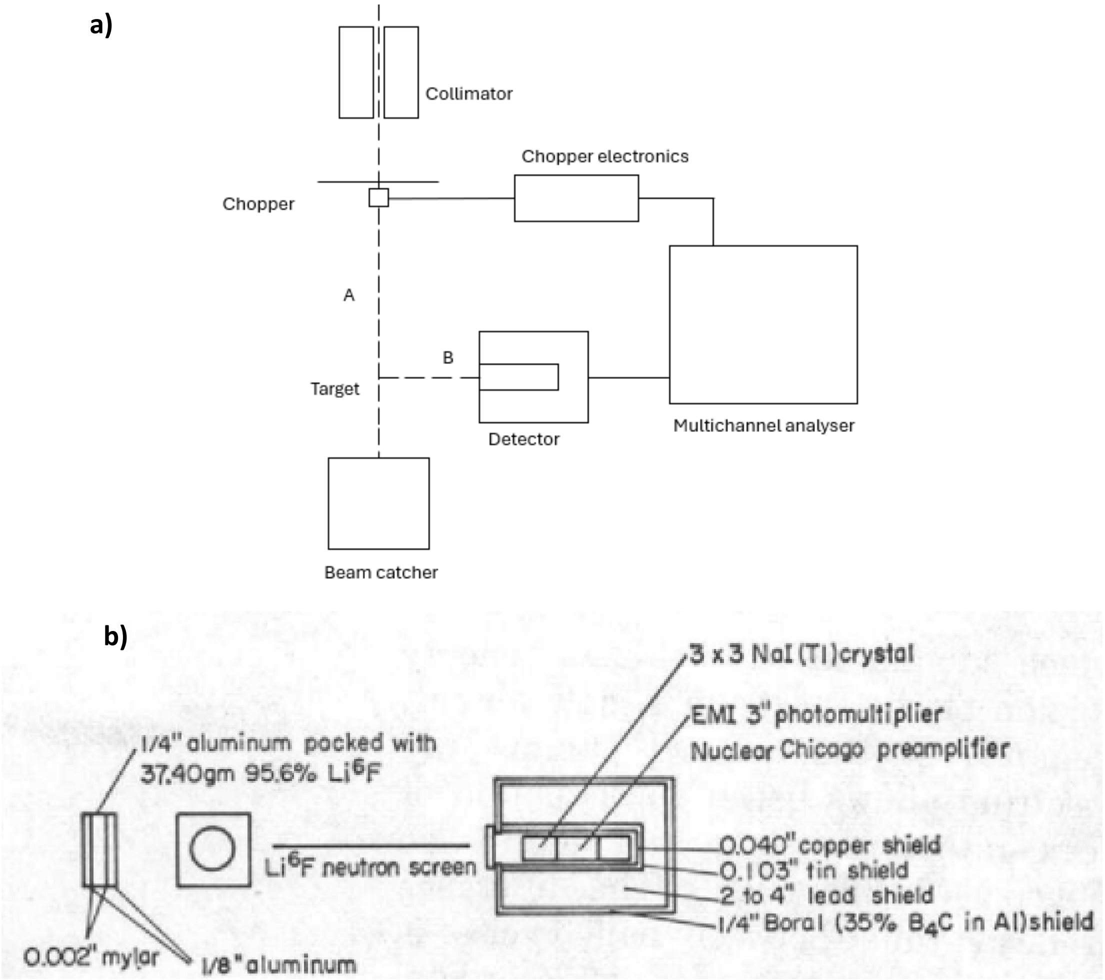

Figure 4: Apparatus used by Isenhour and Morrison [14] with the system arrangement shown in (a) and (b) a close-up of the detector at point B in figure 4a with a ^{6}LiF window to protect the NaI(Tl) crystal from neutron damage. Reprinted (adapted) with permission from [14]. Copyright 2025 American Chemical Society (figure 4a has been recreated for clarity).

Hamermesh and Hummel [17] also attempted to address these issues by taking multiple spectra of the different combinations of a target material and a cadmium shield, the latter to reduce the influence of neutrons from the source. The four permutations with and without the cadmium shutter and sample result in four spectra which can be subtracted and added from one another to remove background influences. Cadmium between the reactor and target inhibits the neutron capture reaction as it absorbs thermal neutrons whilst allowing fast neutrons and γ-rays to pass.

A schematic diagram of the corresponding apparatus is reproduced in figure 5 depicting the small ^{6}LiF window below the sample used to absorb scattered thermal neutrons whilst permitting a narrow beam of capture γ-rays to interact with the NaI scintillator-based detector. Additionally, as identified by Isenhour and Morrison [14], the method ignores the accumulation of delayed γ-ray products from short half-life isotopes. The latter only have a negligible impact on the observed γ-ray spectrum when the half-life of all products is very long with respect to the duration of the count. Otherwise (and in most real-world scenarios), the spectrum observed is not due entirely to neutron capture reactions in the target material.

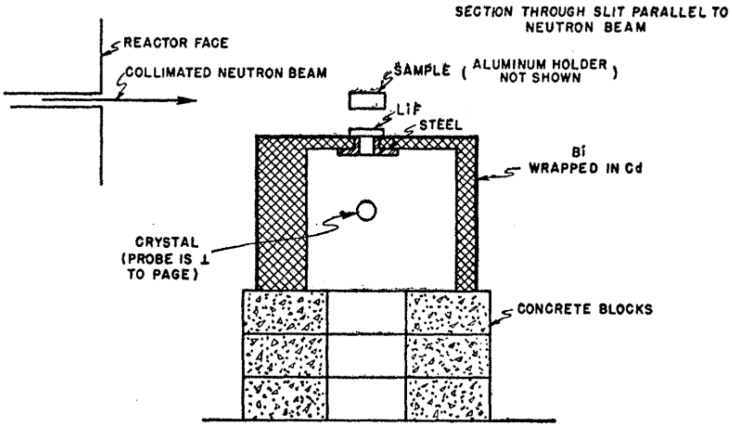

Figure 5: Detector arrangement used by Hamermesh and Hummel [17] with a NaI(Tl) γ-ray detector within a bismuth shield 8” in thickness on the reactor side and 4” thick elsewhere. The bismuth is covered in cadmium to reduce thermal neutron capture by bismuth. Figure taken from [17].

Later in 1966, Isenhour and Morrison [18] demonstrated the effectiveness of their modulation technique by measuring the γ-ray spectrum produced following the irradiation of boron-10 by thermal neutrons. For target materials that may produce isotopes with shorter half-lives, the capture γ-ray spectrum will become obscured by unwanted photons [14]. Modern semiconductors and electronics may allow for improved energy resolution and sensitivity. Results with PGAA also suffered due to a decrease in neutron flux of six orders of magnitude when the chopper was in use and that, with their current apparatus, measurement of delayed γ-ray production was more reliable. Isenhour and Morrison acknowledged that in all cases but two of those irradiated by thermal neutrons, PGAA provided a greater inherent sensitivity. With improvements to collimation geometry and the greater neutron fluxes from larger reactors would improve PGAA accuracy.

3. PGAA for water and hydrogen detection

3.1 Hydrogen detection

Garbrah and Whitley [19] studied hydrogen detection via PGAA using apparatus similar to that used in the earlier work by Hamermesh and Hummel (figure 5) and the later work of Heurtebise and Lubkowitz [20]. As presented in figure 6, a collimated neutron beam was extracted from a reactor to strike the target in front of the scintillator. A 2” hole in the top of the detector shielding was used to collimate γ-rays to the 3” × 3” NaI(Tl) crystal through a ^{6}LiF window. A cadmium shutter can be moved in front of the neutron beam to absorb thermal neutrons and the same four spectra recorded by Hamermesh and Hummel [17] were also recorded here to remove background γ-ray emissions.

The reactor used by Garbrah and Whitley provided a neutron flux of 10^6 n $cm^{-2}s^{-1}$ and the accurate detection of hydrogen at concentrations > 10 wt% and poor accuracy at concentrations of < 1 wt%. Hydrogen detection in organic materials was demonstrated to within 2% or better of the accepted value for hydrogen content. At a neutron flux of 10^6 n $cm^{-2}s^{-1}$, water content, and by extension hydrogen content, in materials such as soil and concrete are better determined by neutron scattering due to the high scattering cross-section of hydrogen.

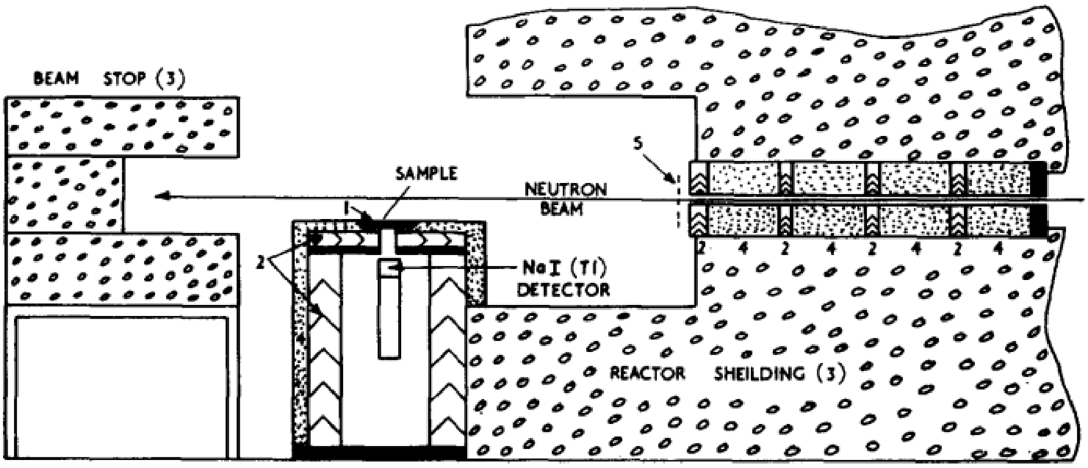

Figure 6: Arrangement of the PGAA detection system used by Garbrah and Whitley [19]. The NaI(Tl) γ-ray detector is protected by a lead-borated wax housing. Numbered materials are as follows: 1) Lithium-6 fluoride; 2) lead; 3) concrete; 4) borated wax; and 5) cadmium shutter. Figure taken from [19].

The simultaneous detection of carbon, nitrogen and hydrogen was demonstrated by Gladney et al. [21] using a Ge(Li) detector and a thermal neutron column from the Los Alamos Omega West Reactor. Beryllium was used for the sample holder due to its small neutron absorption cross-section when the neutron column was evacuated. The thermal neutron flux at the sample is about 2×10^{11} n $cm^{-2}s^{-1}$ with the neutron capture γ-rays travelling 6 m via a series of collimators to the Ge(Li) detector [22]. 1 g samples were irradiated for one hour and detection limits of 1000 µg, 500 µg, and 5 µg were achieved for concentrations of carbon, nitrogen and hydrogen, respectively [21]. PGAA is effective at detecting trace amounts of hydrogen due to the emission of a single, high-energy, characteristic γ-ray after neutron capture has occurred. Detection limits for nitrogen and carbon are higher due to the spectra being more complex comprising multiple lines, thus increasing their risk of contamination by background capture events.

A major aid in recent material analysis by thermal neutron PGAA is the availability of comprehensive and complete data sets covering the induced radioactivity by thermal neutron bombardment. Analysis of PGAA spectra has been made easier with the information provided by Molnár et al. [7, 23] from the Budapest PGAA research group and is available in a comprehensive, online database. Additionally, this database builds on and improves previous work which was incomplete and contained contaminant lines that did not result from neutron capture emissions.

Hydrogen can be detected in the μg range in many materials, although the detection limit is typically restricted by the contribution of hydrogen in the ambient background [24]. However, isotopes with γ-ray emission energies near the 2.223 MeV energy for hydrogen may suggest that more hydrogen is present than is the case. From the expanded PGAA database [7], Biegalski et al. [24] found several prompt capture γ-rays emitted by germanium in the range of 2.215 MeV to 2.240 MeV. As reported recently, most high-energy applications utilise cryogenically cooled Ge detectors after the early success of Ge and lithium-drifted Ge detectors. γ-ray emissions from Ge with similar energies to the γ-rays emitted following the $^{1}H(n,\gamma)^{2}H$ reaction are consequently of concern to the reliability of hydrogen detection systems that employ germanium radiation detectors.

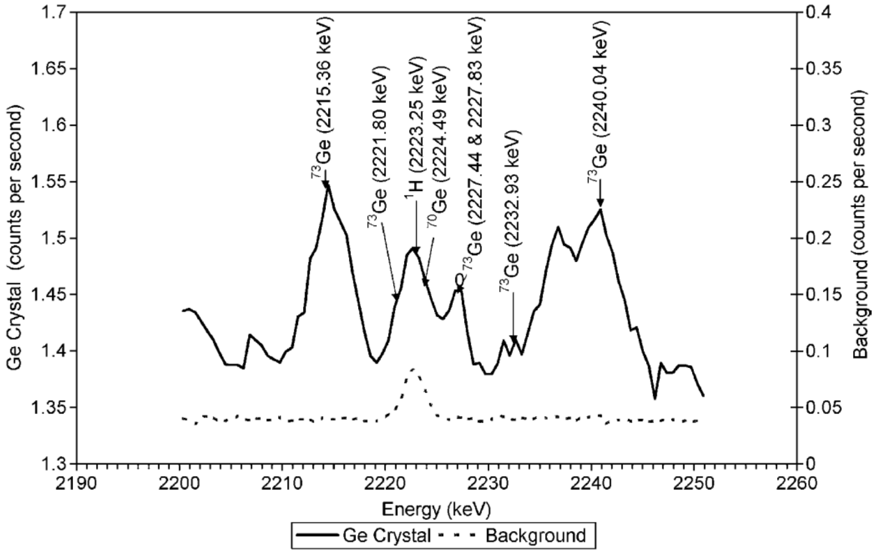

Figure 7: Emission spectra of prompt γ-rays from germanium and hydrogen neutron capture demonstrating the need to shield germanium detectors from thermal neutrons to ensure a clear hydrogen emission peak is obtained [24]. Figure taken from [24].

Figure 7 shows the γ-ray emission energies from germanium neutron capture with a ^{73}Ge and ^{70}Ge emission line within the FWHM of the hydrogen emission peak. Experimentally, germanium emission γ-rays constitute between 7% and 8% of the area under the 2.223 MeV peak [24]. The interference on the hydrogen spectrum by Ge emissions is a direct function of the neutron flux incident upon the high-purity germanium (HPGe) detector. Shielding the HPGe detector therefore serves two important purposes: it prevents neutron damage to the crystal and prevents interference by γ-ray emissions at energies near to 2.223 MeV. ^{6}LiF windows are used across γ-ray collimators as they are relatively transparent to energetic signature γ-rays but absorb thermal neutrons due to the high capture cross-section of lithium-6.

Detection of explosives hidden in vehicle compartments or luggage by X-ray or vapour sensing of volatile compounds is challenging. This is due to the small size of detonator elements and relative ease with which vapour ‘scents’ are masked [25]. PGAA constitutes an alternative for detecting the common elements in explosives, i.e., H, C, N, O and Cl, due to the high penetrating power of neutrons and ability to detect the isotopes of these light elements. Hydrogen concentrations were measured by PGAA for the detection of concealed explosives and the ratio of nitrogen and carbon to hydrogen was assessed to minimise false positives. Measurements were performed at the 30 MW thermal neutron PGAA facility of the Korea Atomic Energy Research Institute. The PGAA system consisted of a HPGe detector surrounded by eight BGO detectors with two NaI(Tl) scintillators as a Compton suppression device. The HPGe detector was placed 25 cm away from the sample and the neutron flux incident on the sample was 1.4×10^8 n $cm^{-2}s^{-1}$.

Melamine ($C_3H_6N_6$) was used as an analogue for an explosive in these measurements. Measurements taken with this PGAA facility were in close agreement with established values for hydrogen content. However, hydrogen content below 10 mg/kg in samples of approximately 30 mg was measured as higher than the accepted value whilst the measured value fell below the true value for samples of mass near to 130 mg. The count rates at different masses of melamine are reproduced in Figure 8. A sensitivity to hydrogen of 3 cps/mg of H and a detection limit of 6 µg were demonstrated. The uncertainty in these results derives primarily from statistical errors, detection sensitivity and background effects. Longer counting times and reduction of the hydrogen background by placing the sample in a vacuum and protecting the shielding from neutron capture may act to reduce the uncertainty.

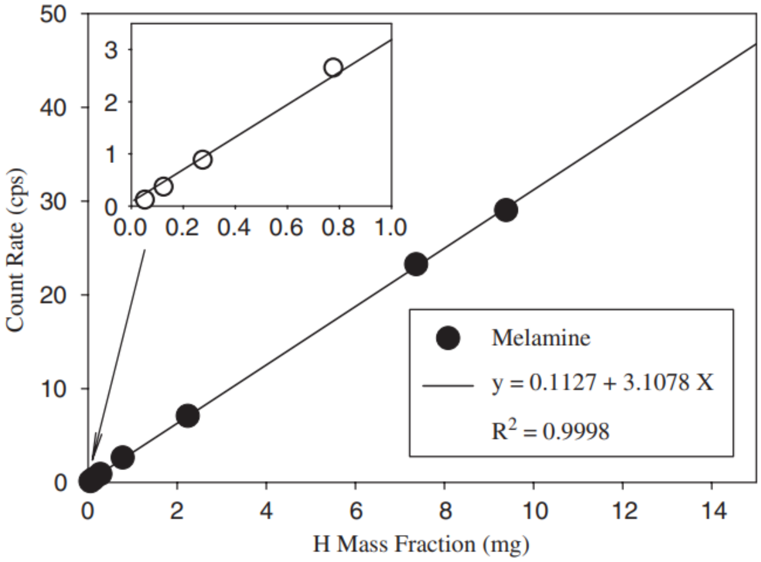

Figure 8: Measured count rate from thermal neutron irradiation of different masses of melamine ($C_3H_6N_6$). Following up the trend line from (0,0) in terms of increasing mass the melamine samples targeted were 1.1, 2.6 5.7, 16.2, 46.7, 101.81 and 153.5 mg [25]. Figure taken from [25].

3.2 Hydrogen detection by cold neutrons

Cold neutrons have energies less than thermal neutrons (<0.025 eV) and allow the inverse proportionality of neutron energy with capture cross-section to be exploited. Cold neutrons for analytical purposes can therefore provide greater sensitivity for the detection of trace elements in a target sample. A detection limit of < 5 mg/kg of hydrogen was achieved at the NIST Centre for Neutron Research in most materials whilst a detection limit of < 2 mg/kg was acquired by measuring a silicon sample [26].

A spallation neutron source installed in the Paul Scherrer Institute (Switzerland) was built to provide both thermal and cold neutrons to target samples [27]. At the end of one cold neutron guide, a PGAA facility is located. A plan view of this facility is seen in Figure 9. The cold neutron beam passes through a focusing neutron lens, striking the target placed inside an aluminium box. The neutron focusing lens is comprised of many polycapillary fibres which bend toward a focal point smaller than 1 mm diameter target area at a flux of 3×10^{10} n/cm^2s, allowing the target to be scanned.

Capture γ-rays are observed by Ge crystals surrounded by NaI(Tl) and BGO scintillators for Compton suppression. Photon emissions from each Compton suppression unit are logged by two PMTs and subtracted from the measured germanium spectra providing peak-to-background improvements for energies below 2 MeV. It is believed that with this arrangement, and careful precautions to avoid the presence of hydrogen in surrounding and detector materials, a hydrogen detection limit of 1 µg was achievable.

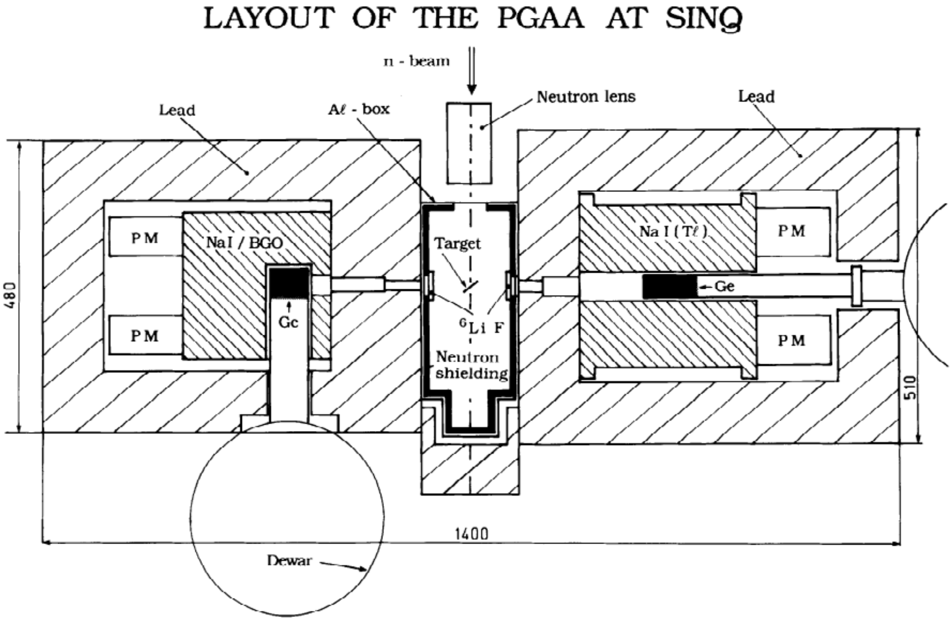

Figure 9: Cold neutron PGAA setup at the Paul Scherrer Institute (Switzerland) with two Ge detectors surrounded by NaI/BGO scintillators for Compton suppression [27]. Figure taken from [27].

Semiconductor materials with hydrogen embedded have altered electrical properties when compared to their pure counterparts [26]. Hydrogen fractions of silicon carbide and cerium aluminate semiconductors were analysed by bombarding solid samples with a cold neutron beam, generated by passing thermal neutrons through liquid hydrogen at 20 K. The PGAA detector is seen in figure 10 using a Ge crystal and BGO Compton suppression shield. Lead and cadmium shielding also surround the germanium detector, limiting detector exposure to background and stray reactor radiation. The neutron beam is collimated to a diameter of 2 cm or smaller where it strikes the target, and the emitted γ-rays are collimated toward the Ge detector.

Due to size constraints imposed by this facility, the maximum sample size is just 6 cm in diameter limiting the geometries that can be tested. As discussed above, a detection limit of less than 2 mg/kg of hydrogen in silicon carbide was achieved using this apparatus. When compared to other analytical methods used in this study, such as Rutherford backscattering and forward recoil elastic spectrometry, PGAA measured higher levels of hydrogen in silicon. This is due to the highly penetrative properties of neutrons and γ-rays which can measure both surface and internal hydrogen.

Using the apparatus in figure 10, PGAA was also effective for the detection of hydrogen in many materials used for hydrogen storage [28]. Hydrogen embrittlement of materials is a major concern for the transport and long-term storage of hydrogen. Therefore, an effective, non-destructive method of determining low levels of hydrogen such as cold neutron PGAA is necessary to understand the integrity of hydrogen-carrying infrastructure. PGAA was used successfully to measure the formation of $CaSiH_x$

compounds following the introduction and removal of hydrogen, as well as the uptake of hydrogen by fuel cell materials.

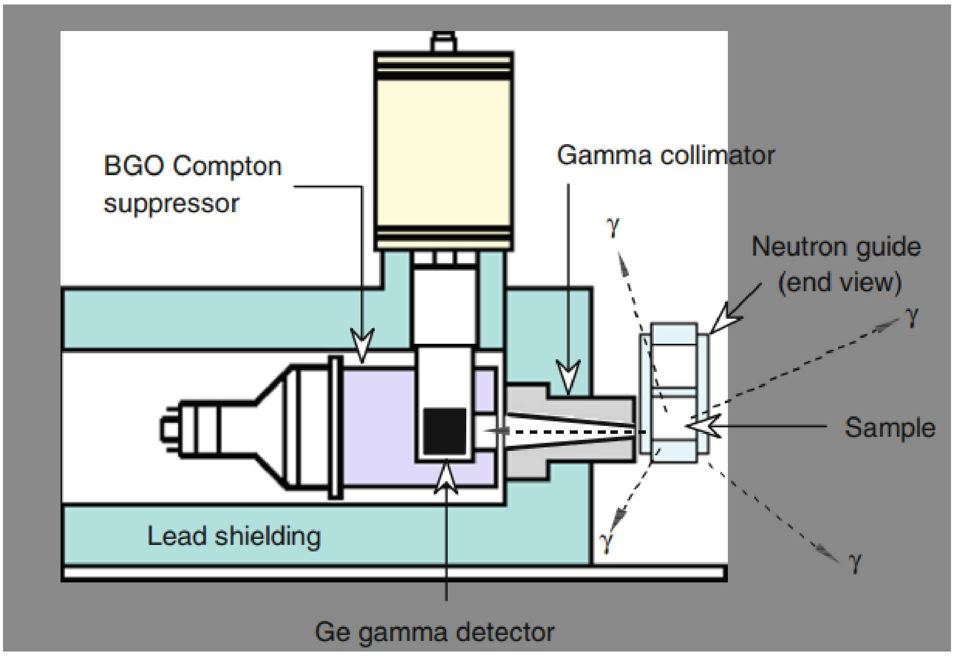

Figure 10: PGAA equipment for detecting hydrogen fractions in semiconductors and hydrogen uptake in fuel cells by cold neutron bombardment [26, 28]. Figure taken from [26].

Improvements to the cold neutron PGAA facility at NIST (USA) discussed in [26, 28] were presented by Paul et al. to improve the sensitivity of the detector and to alleviate the sample size limitations experienced in previous iterations [29]. With a neutron focusing lens, the instrumentation can also perform sample scanning by irradiating limited sections of the sample at a time. The sample chamber and detector assembly are shown in Figure 11a with a cut-away side profile of the detector provided in Figure 11b. The detector comprised a HPGe crystal inside of a BGO Compton suppressor. The detector shielding was 4” thick with lead and has a thin cadmium coating over the lead shield.

Neutrons travelling along the neutron guide are collimated (and can be focused if necessary) before striking the sample where γ-rays emitted toward the detector are collimated. The detector assembly can be repositioned manually to account for different count rates to minimise dead time in the detector. A vacuum pump is also attached to the sample chamber to allow samples to be studied at different pressures whilst limiting the background arising from neutron capture events in air.

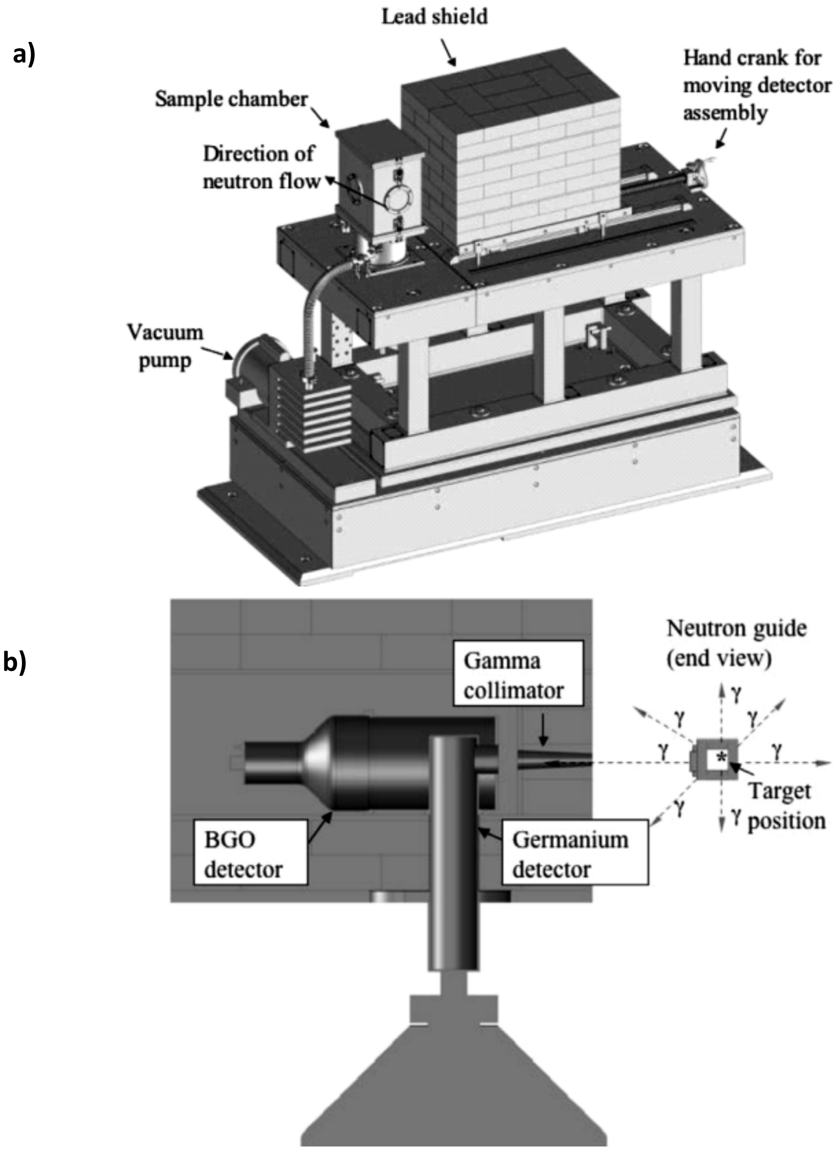

Figure 11: Computer generated models of the improved cold neutron PGAA system seen in Figure 10. Figure 11a) shows the detector shielding and sample chamber on a raised platform to align with the reactor neutron beam. The detector and lead shielding can be moved to compensate for different neutron fluxes. Figure 11b) shows the vertically mounted germanium detector with the BGO detector to eliminate background from Compton scattering [29].

The feasibility of using a cold neutron source outside of a dedicated facility is extremely limited. This is due to the need for liquid hydrogen or similar moderation techniques to sufficiently reduce the neutron energy. Transportation of a dewar and neutron source with a sufficient flux presents a challenge that may be unfeasible. Use of larger, deployable, detectors with high detection efficiencies in the region of 2.223 MeV may more appropriately compensate for the lack of cold neutrons. Thermalisation of neutrons from a passive internal neutron source also better suits a portable application for the detection of water in spent nuclear fuel.

3.3 Water and moisture detection

The detection and quantification of water by PGAA is largely absent from the literature when compared to the study of hydrogen. The focus instead lies on, for example, understanding the degree of water saturation in neutron-based petrochemical well logging [30] or the effect of water on cross-section measurements [31]. The motivation for the study of water is typically to better understand its effects on PGAA measurements when the focus is on attempting to detect other elements in a given material. PGAA studies towards the presence and effects of moisture typically utilise the 2.223 MeV γ-ray emitted by hydrogen following neutron capture. γ-ray emissions from hydrogen neutron capture are of interest due to the higher cross-section when compared to oxygen, as per Figures 12a and 12b.

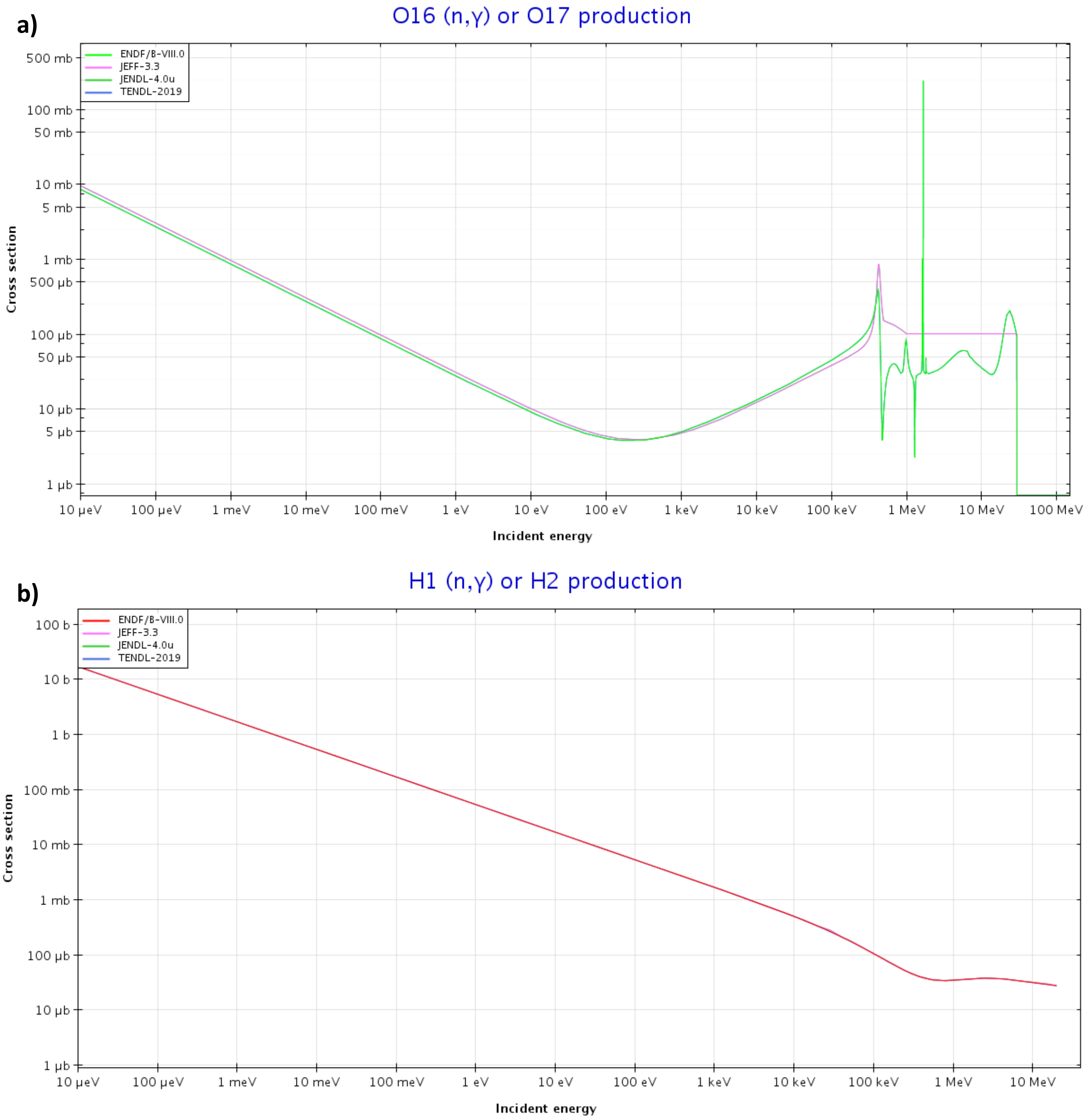

Figure 12: a) Cross-section of oxygen-16 as a function of incident neutron energy for the $^{16}O(n,\gamma)^{17}O$ reaction and b) of hydrogen as a function of incident neutron energy for the $^{1}H(n,\gamma)^{2}H$ reaction. Both graphs are from the composite JANIS database using the ENDF/B-VIII.0, JEFF-3.3, JENDL-4.0u, and TENDL-2019 datasets.

PGAA is a non-destructive method for providing precise and comprehensive material analysis in a wide range of industries and applications. Heurtebise and Lubkowitz [20, 32] demonstrated the measurement of multiple elements and moisture in catalysts used in the petroleum industry. A top-down view of the sample-detector arrangement can be seen in Figure 13 showing the detector shield and γ-ray collimator. It should be noted that the setup used in this experimental work is very similar to that of Hamermesh and Humel given in figure 5. Before striking the target, a neutron beam passes through paraffin and a compressed ^{6}LiF slab. γ-rays emitted toward the Ge detector also pass through a ^{6}LiF slab and are collimated.

Water content measurements in these catalysts are performed typically by measuring the weight lost after heating. However, Mo-based catalysts cannot withstand the necessary temperature for water removal, and hence PGAA provides an appealing alternative. In addition to determining Co, Mo, Ni, and W content of the catalysts simultaneously, 9.2% more of the water content in these highly hygroscopic materials was also detected.

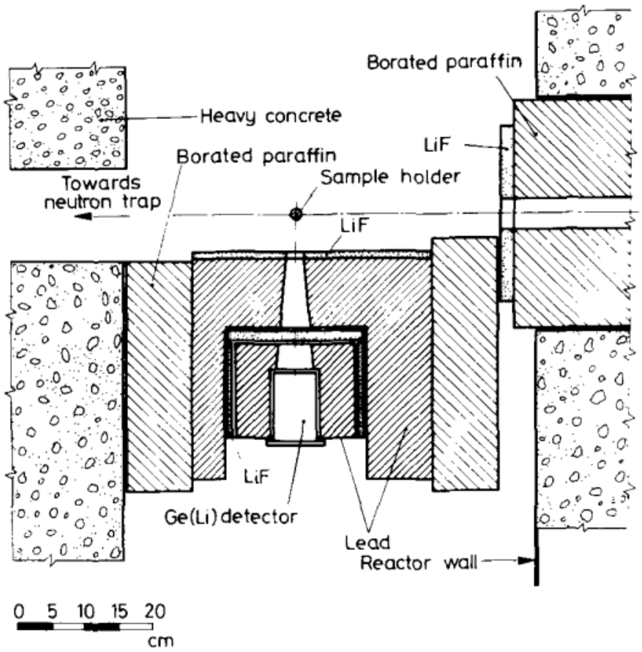

Figure 13: Top-down view of the apparatus used by Heurtebise and Lubkowitz [20] for the measurement of moisture content in hydrodesulfurisation catalysts used in the petroleum industry. Figure taken from [20].

There is an interest in moisture present in boreholes as both an indicator of potential groundwater contamination and as an effective neutron moderator when performing neutron analysis techniques. Moisture was logged by Wilson et al. [33] in a cased borehole using the probe seen in Figure 14, measuring the $^{1}H(n,\gamma)^{2}H$ response at different volume percentages of water and comparing the experimental results to an MCNP model. The probe contained a cooled HPGe crystal with a ^{252}Cf neutron source that was separated from the detector by a tungsten shield. In the absence of other hydrogen-containing materials, the hydrogen spectrum provides a direct moisture measurement. If delayed γ-ray emitters are present in the borehole, the recorded spectrum will suffer from interference. However, it is thought that the HPGe crystal provides sufficient energy resolution to identify and remove unwanted γ-ray peaks.

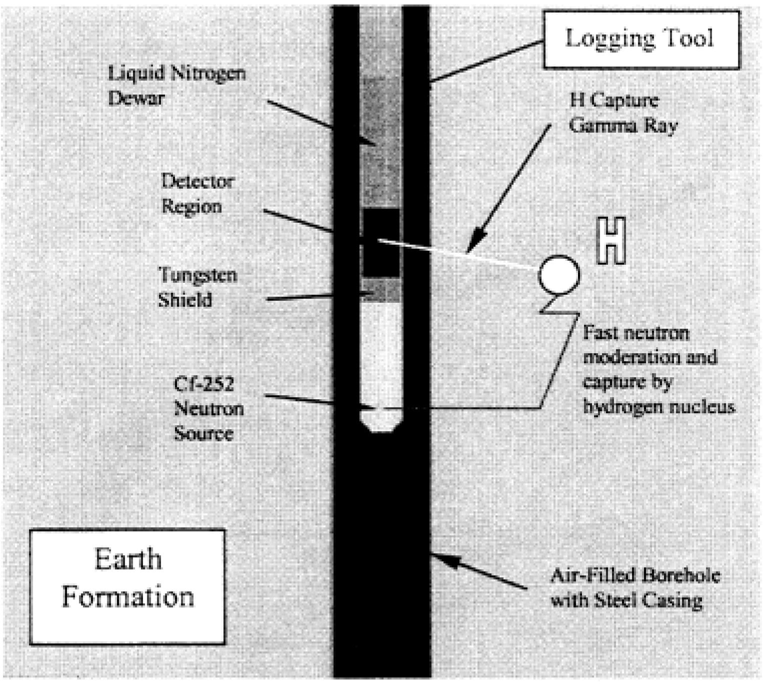

Figure 14: Probe designed for moisture detection down boreholes containing a ^{252}Cf- neutron source and HPGe γ-ray detector [33]. Figure taken from [33].

When performing material analysis by PGAA, water increases neutron moderation and hence the rate of neutron capture by other isotopes present. In work by Moxham et al. [34], a borehole in rock containing notable quantities of nickel and copper was examined with a ^{252}Cf source to obtain both delayed and capture γ-ray spectra. When water was present in the borehole, an increase in the delayed 1.039 MeV full-energy peak associated with the γ-ray emission from ^{66}Cu was observed, with further increase in the full-energy peak observed as the ore became wet. The Ge(Li) detector was able to detect the 2.223 MeV prompt γ-ray from neutron capture on hydrogen.

Understanding the relationship between the 2.223 MeV emission and increase in the full-energy peak for other isotopes is essential to quantify materials in the borehole accurately. Probes used for borehole measurements must fit in the borehole being surveyed limiting the size of components compatible with use in the probe. Neither the probe used by Wilson et al. nor that used by Moxham et al. had on-board Compton suppressors. A possible effect of this is increased low-energy noise from the incomplete detection of scattered γ-rays and the detection of γ-rays generated by neutron capture in the germanium crystals.

The effects of soil moisture on PGAA measurements are also of relevance to devices used for the detection of buried explosives [35]. Since common explosives are composed predominately of carbon, hydrogen, and nitrogen, melamine ($C_3H_6N_6$) can be used. Melamine is placed in a container and buried at a variety of depths and soil moisture levels to represent a common explosive. The detector system used in this research consisted of a ^{252}Cf neutron source inside a Pb sphere surrounded by a HDPE cylinder for neutron moderation, as per figure 15. The distance d1 in figure 15 is 20 cm. The γ-ray detector used comprised a 7.5 cm × 7.5 cm NaI(Tl) crystal placed at an angle to the soil and shielded from the moderator by a 2-cm thick Pb block. Soil moisture levels at 0 wt%, 13.7 wt%, 16.8 wt% and 17.1 wt% were assessed, and the impact of the moisture content considered.

As expected, the presence of moisture over dry soil increased the moderation of neutrons thereby increasing the recorded counts due to an increase in the likelihood of the $^{14}N(n,\gamma)^{15}N$ reaction. A $1/r^2$ relationship between count rate and sample depth is observed as fewer γ-rays strike the detector. For depths over approx. 7 cm, the highest moisture content of 17.1 wt% did not produce a measurable count rate. MCNP modelling of the thermal neutron fluence induced by a 14 MeV neutron source 15 cm above wet dense soil shows a thermal neutron peak fluence at 10 cm deep into the soil [36]. Considering the average neutron energy of approximately 2 MeV from a ^{252}Cf source, the depth limitation of 7 cm at 17.1 wt% water content appears to be in close agreement with the simulations using higher energy neutrons. This suggests that, at this water content, the hydrogen neutron capture reaction becomes more prominent than its moderating effect, leading to very few neutrons being captured by ^{14}N.

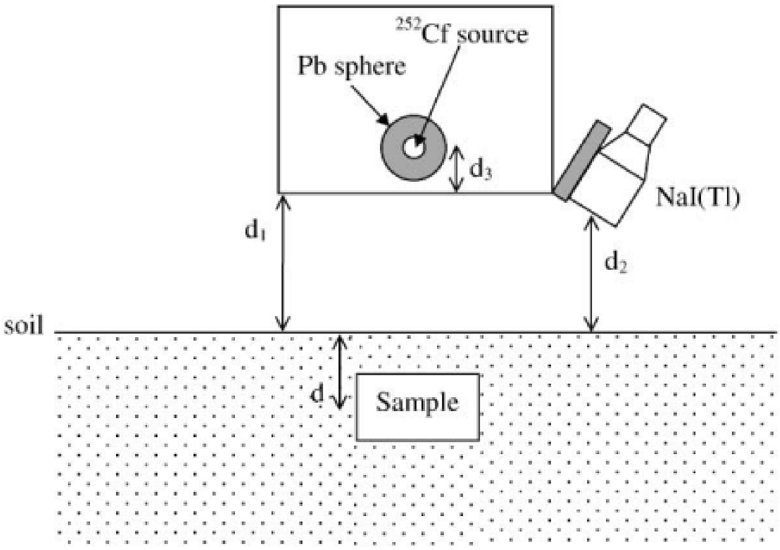

Figure 15: Apparatus for determining the effect of moisture on the detection of buried explosives [35] with melamine used as a simulant for buried explosives due to its similar chemical composition buried in a container. Figure taken from [35].

Acoustic probing is used widely to identify manufacturing defects in lightweight composite materials [37]. However, this often requires the sample to be immersed in water to couple the probe and sample to ensure an accurate evaluation. Carbon fibre reinforced epoxy matrix materials exhibit a tendency to absorb water which may compromise the structure of the material. PGAA provides a means to determine the extent of water uptake in this regard by measuring the 2.223 MeV γ-rays from neutron capture on hydrogen. The detector used was an Ortec GMX series n-type HPGe detector system shielded by 6” of lead on all sides with a 2” diameter collimation hole covered by a polymer window containing ^{6}Li. The lead is covered by borated rubber to reduce radiative capture reactions.

Figure 16 shows the PGAA system used with the sample placed in its holder. The reactor used in this work output a cold neutron flux of around 6.5×10^6 n cm^{-2}s^{-1}. Whilst several issues arose concerning the composite due to long-term exposure to water at 70°C, calculations of the relative change in hydrogen content following absorption and desorption were in close agreement with the results obtained by PGAA. The calculated and measured hydrogen mass during absorption and desorption are reproduced in Figures 17a and 17b, respectively. Despite the high hydrogen content of the epoxy material, accurate determination of the water content by means of PGAA of hydrogen is possible and was shown to be more sensitive than gravimetric measurements.

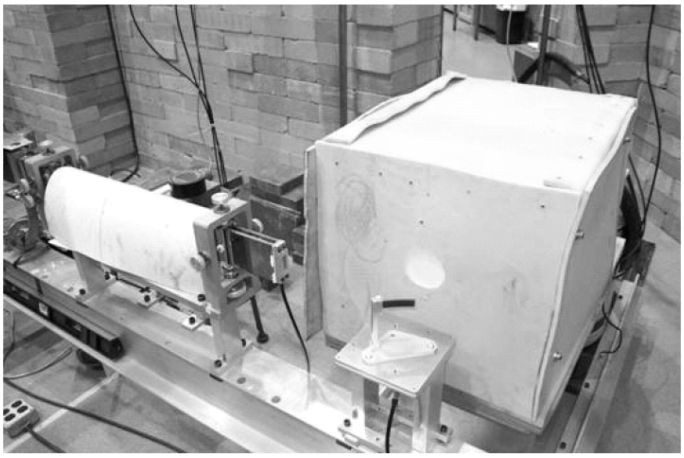

Figure 16: The HPGe detector system used for the measurement of water in composite materials. The HPGe crystal was placed within a lead box with 6”-thick walls and covered by borated rubber to prevent radiative neutron capture in lead. Cold neutrons were collimated towards the sample placed outside the 2” aperture for γ collimation [37]. Figure taken from [37].

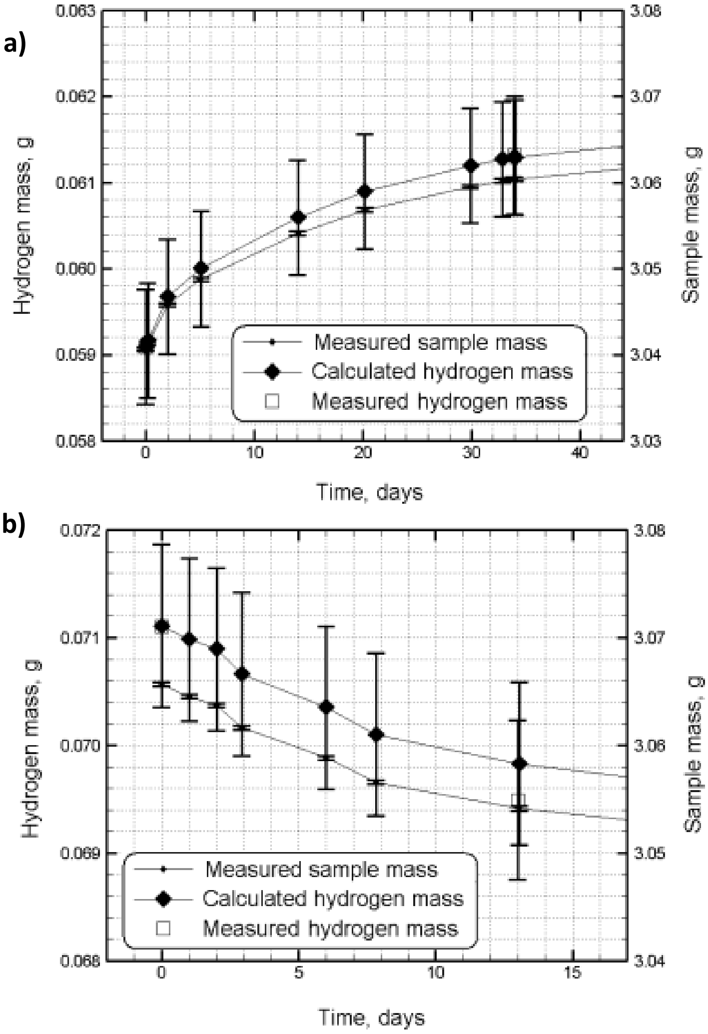

Figure 17: The calculated and measured hydrogen mass during: a) absorption and b) desorption of water. In (a) and (b), the top trace shows the calculated and measured hydrogen mass and the bottom trace shows the sample mass. The composite sample was submerged in a water bath at 70°C and the samples were irradiated for 4 hours [37]. Figure taken from [37].

Reference	Neutron source	Detector
[20]	4.75 x 10^7 n cm^{-2} s neutron beam generated by the 3 MW RV-1 nuclear reactor	Ge(Li) detector
[33]	^{252}Cf spontaneous fission neutrons	HPGe detector
[34]	^{252}Cf spontaneous fission neutrons	40 cm^3 Ge(Li) detector
[35]	^{252}Cf spontaneous fission neutrons	7.5 x 7.5 cm cylindrical NaI(Tl) crystal
[37]	6.54±0.15 x 10^6 n cm^{-2} s^{-1} cold neutron beam generated by a 1 MW Triga Mark 2 reactor	Ortec GMX Series HPGe

Table 1: Summary of neutron sources and detectors used for water or moisture detection discussed in this review.

As seen in table 1, a wide range of detectors were used for detection of water or moisture with a ^{252}Cf source preferentially chosen for portable applications. Sensitivities and hydrogen detection efficiencies of the system were determined differently in each of the listed works making comparisons challenging. In general, close agreement was found between theoretical and experimental values with the exception of work by Wilson et al. who saw the simulated count rates at 15%-45% higher than the measured count rates [33]. Germanium based crystals were favoured for detection due to a better resolution at higher energies than NaI(Tl) scintillators. Regardless of the detector, increased moisture content saw an increase in the count rate of capture gammas, both the 2.223 MeV hydrogen gamma and other gammas due to additional neutron moderation by hydrogen. The portable ^{252}Cf detectors show the most potential for use as a deployable water detection system for spent nuclear fuel. Improvements on the detection systems are possible as the constraints of fitting into a borehole [33, 34] or being portable for explosives detection [35] are not in place. This allows for larger detectors, improved moderation and shielding, and further refinement to the system geometry.

4. Further applications of PGAA and modern relevance

In most of the studies discussed by Gladney [13], the facilities used were not designed for analytical measurements of materials. A major drawback of this is that neutron and γ-ray emissions resulting from other materials may be picked up by the detection apparatus, degrading the quality of results. A dedicated, reactor-based PGAA system at the University of Missouri Research Reactor (USA) is shown in Figure 18 [16]. The beam's thermal neutron purity is enhanced by additional filtration and collimation to the neutron beam. Although this reactor is also used for research applications aside from PGAA, its dedication to PGAA removes all geometrical constraints experienced by Paul [26, 28]. Many other facilities around the world have also been developed using both thermal and cold neutrons designed to reduce background, improve sensitivity and hence enable a wider avenue of research studies [38].

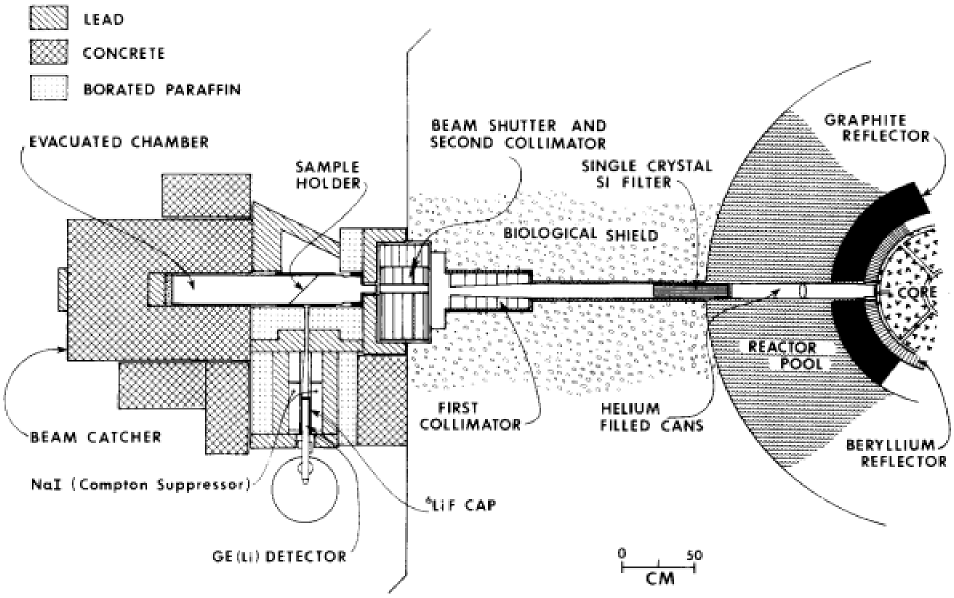

Figure 18: A schematic plan diagram of the PGAA facility at the University of Missouri (USA). The Si filter reduces the fast neutron and γ-ray flux incident at the sample. A 10^{14} n cm^{-2}s^{-1} flux is observed at the reactor face and the sample is subject to 5×10^{8} n cm^{-2}s^{-1} [16]. Figure taken from [16].

Hydrogen has been the focus of PGAA in this review due to the effectiveness of this technique when compared to others for determining the presence of trace quantities [38]. For example, PGAA of hydrogen has been used to monitor the chemistry of lithium-ion batteries and to measure water content in composites and building materials. Hydrogen in metals is of interest to various industries due to hydrogen embrittlement in materials for hydrogen mass fractions as small as 10 mg/kg [39]. In jet engine compressor blades, hydrogen was measured as low as 50 mg H/kg of Ti where concentrations of several hundred mg/kg were thought to cause embrittlement. Hydrogen is also thought to have considerable influences on the pore size and structure in nanocrystalline metals, thus affecting the mechanical properties of the material. PGAA was used successfully to confirm the presence of excess hydrogen down to 20 ± 4 µg/g, leading to improvements in material preparation methods.

Fusion power presents an environment much richer in neutrons than nuclear fission. Fission of ^{235}U produces 2.5 neutrons per fission event and a 205 MeV release [40] whilst DT fusion produces 1 neutron and a 17.6 MeV release [41]. Therefore, for an equivalent power output, prompt and delayed gammas resulting from neutron capture and activation of reactor materials lead to a need for specialist considerations for shielding in fusion nuclear environments. Shielding must attenuate the high energy neutrons, born at 14.1 MeV in the case of DT fusion, prompt gammas, and delayed gammas from neutron activation, minimising heating to sensitive components such as superconducting magnets [42]. Largely due to the extreme environments in which fusion takes place, hydrogen detection in materials during repair and decommissioning can be challenging. PGAA may provide a suitable solution to study the behaviour of hydrogen, and its isotopes, deuterium and tritium, in various materials and reactor components. Plasma-facing components may suffer from ion or energetic neutral DT atom implantation leading to retention in these components or migration through reactor walls [43]. Measuring the degree of hydrogen contamination in these elements by intrinsic PGAA could reduce the risk of component damage and failure due to hydrogen-induced embrittlement and blistering [44].

Experimental fusion devices for neutron diagnostics often use hydrogenous shielding to protect the detector material which is often a fast scintillator [45]. Whilst it provides some analytical purpose, in this instance the 2.223 MeV gamma produced during hydrogen-neutron capture is detrimental to these devices. This is because the scintillator is sensitive to both gammas and neutrons. To limit deadtime induced by gamma detection in the scintillator, additional shielding must be implemented so that only neutrons are detected.

An in depth and reliable understanding of the neutron fluence allows the power output and plasma properties to be determined. Measuring the neutron fluence far from the reactor plasma is also critical to the effective radiation protection of personnel during maintenance and waste material handling. Neutrons produced in the DT fusion reaction have an energy of 14 MeV and are highly penetrative with a mean free path of 14.1 cm in concrete [46]. This makes fluence monitoring about the reactor challenging. Radiative neutron capture provides an avenue for fluence determination, independent of environmental interferences [47]. Using a variety of metal foils, measured neutron fluences were in agreement with previous experimental values and have enabled benchmarking of other experimental techniques and computational tools. In addition to the necessary dose considerations from energetic neutrons, plasma-facing components may undergo structural and functional modifications from neutron impinging upon them [48]. High energy neutrons (10.5 MeV and above) also have the chance to activate reactor cooling water to a lesser extent in fission reactors but more prominently in fusion reactors [49]. Neutron activation of water is achieved through the production of ^{16}N, ^{17}N and ^{19}O leading to delayed gamma emissions from radioactive decay with energies of 6 – 7 MeV. High energy gammas emanating from the cooling loop necessitate additional shielding for detectors, instrumentation, and personnel as it stretches beyond the primary shielding area [50]. Similarly to high energy neutrons, activated water may also cause nuclear heating to sensitive elements such as superconducting magnets. Work to understand the activation of cooling water will improve the reliability of sensitive reactor components and reduce dose to personnel.

The neutron flux of the fuel containing material (FCM) at the Fukushima Daiichi Nuclear Power Station (1F NPS, Japan) has been measured by PGAA of water [51] and neutron capture by hydrogen in water has also been suggested as a marker for submerged fuel debris. However, the intensity of the 2.223 MeV full-energy peak is small when compared to other, competing γ-ray emissions. FCM arising as a result of nuclear accidents, such as those at the Chernobyl Nuclear Power Plant Unit 4 (ChNPP4, Ukraine) and 1F NPS, present complex and hazardous decommissioning challenges [52].

Before the introduction of the New Safe Containment (NSC) over ChNPP4 in 2016, rainwater had accumulated in the facility, as illustrated in Figure 19, providing additional avenues by which FCM can age and by which water-soluble uranium minerals can be redistributed as it moves and evaporates [52]. Zirconium oxide is also of concern as it is a notable component of the lava-like materials containing nuclear fuel (LMNF) and has been observed to weaken structurally by a phase transition in moist environments. Such structural changes may cause unanticipated movement of solid materials, raising additional safety concerns in the ruined facility.

Similarly, seawater pumped into the reactor core at 1F to stabilise the facilities following the accident at Fukushima in 2011 and the addition of a reducing reagent to prevent reactor pressure vessel (RPV) corrosion has the potential to alter the redox chemistry of the FCM. The FCM in 1F NPS is expected to undergo similar aging processes to those observed at ChNPP4. The use of PGAA to interrogate FCM to quantify the presence of water will improve predictions of the potential effects of water on fuel ageing and the consequences of fuel drying on the distribution and behaviour of FCM in ChNPP4 and 1F NPS.

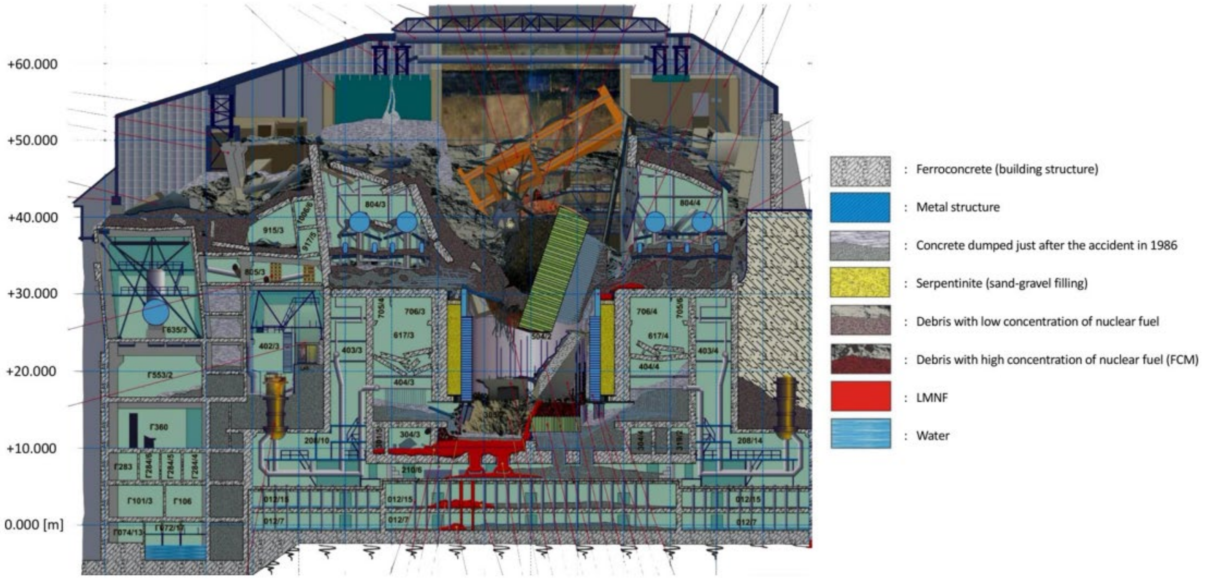

Figure 19: Cross section of ChNPP4 showing the collapsed sections of the building following the accident and the sarcophagus placed over the top. FCM and LMNF have seeped throughout the facility and into the basement levels [52]. Figure taken from [52].

In addition to assessing water content in the FCM in both ChNPP4 and 1F NPS, the measurement of the 2.223 MeV photopeak originating from neutron capture in hydrogen may be used for the characterisation of fuel debris [53]. Nauchi et al [53]have demonstrated an experimental setup, shown in Figure 20, that measures 2.223 MeV capture γ-rays from hydrogen, induced by the neutron yield of a spent BWR fuel sample. Neutron and gamma emissions from the fuel sample pass through a carbon steel collimator where they strike a polyethylene block and are captured or scattered. To further reduce the intensity of gamma rays from fission products (FPs), the HPGe detector is placed off the collimator axis thereby reducing the dead time. Moreover, gammas from FPs must be scattered through a large angle to be detected, significantly reducing their energy, limiting interference around the 2.223 MeV region of interest. Providing there is adequate shielding from FP γ-rays, PGAA of the 2.223 MeV γ grants an accurate method of quantifying the neutron yield from spent fuel. Subsequently, it will be possible to analyse the composition of FCM. With further refinements, this method will allow safe handling of fuel debris at both ChNPP4 and 1F NPS during retrieval and storage.

Oxidation of PuO_2 and the subsequent production of hydrogen and oxygen gas is a known hazard associated with improper drying of SNF containing plutonium prior to storage [54, 55]. PuO_2 oxidation by water is strongly endothermic and may be driven by radiolysis due to strong oxidants such as hydrogen peroxide [54]. This reaction demonstrates a significant risk in the event of a fire where hydrogen or even steam build-up could lead to an explosion. Sellafield in the UK hosts a large PuO_2 stockpile from the historic Magnox reactors and Thorp reprocessing site [55]. Extreme water adsorption in some very limited cases has led to pressurisation of the storage cans. Radiolysis was thought to be the cause of H_2 production, occurring under high relative humidity (75%-95%) or with a high number of water monolayers. A portable PGAA system could provide monitoring capabilities to determine the presence of water or a build-up of hydrogen gas.

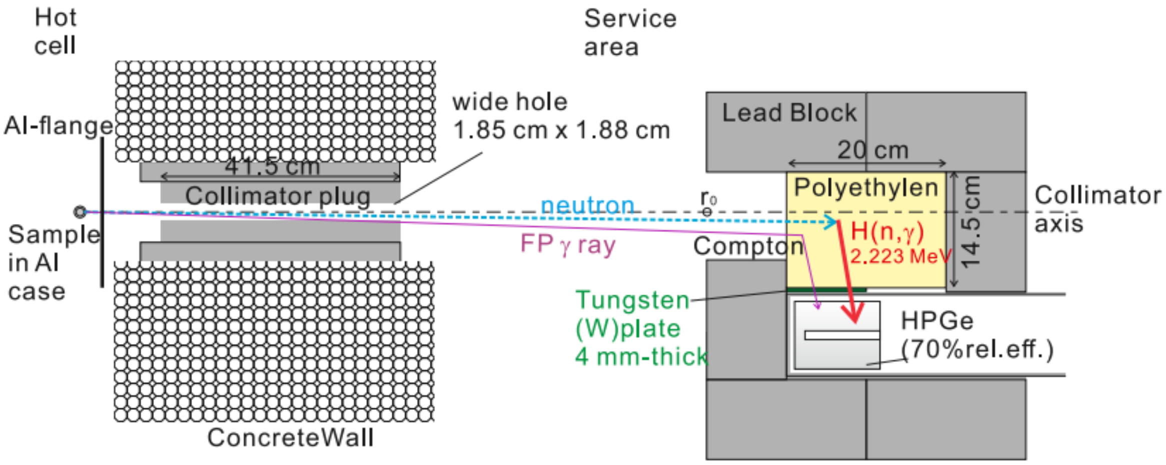

Figure 20: Side profile of a system for measuring 2.223 MeV gamma rays from the ^{1}H(n,γ)^{2}H reaction [53]. Figure taken from [53].

As well as material analysis applications, neutron capture has been explored for power monitoring in nuclear reactors with metal samples and hydrogen has been considered [56, 57]. Okada et al. [56] used neutron capture in Ti, V, Ni and Cu targets which could withstand temperatures in a boiling water reactor under normal operation exploiting a limited number of high-energy γ-ray emissions. The conceptual arrangement for this is reproduced in Figure 21 with the target metals in the reactor pressure vessel and an HPGe detector placed externally. Since prompt γ-ray emission intensity is proportional to the thermal neutron flux, assuming the material is not significantly transmuted, a combination of metal samples placed in selected locations enables the thermal neutron flux to be monitored throughout the plant based on at least one identifiable full-energy peak for each metal.

Chernikova et al. [57] demonstrated thermal neutron flux measurement in spent fuel storage ponds, using a ^{252}Cf source and an ^{241}Am-Be neutron source in a water container to simulate a source of neutrons in a fuel pond. A NaI(Tl) scintillator was used as the γ-ray detector, positioned externally to the neutron source and water tank, as shown in Figure 22. The measurement of prompt γ-ray intensity from hydrogen neutron capture demonstrated a close correlation with the thermal neutron flux. This approach provides a potential alternative to direct thermal neutron detection with ^{3}He detectors or similar instrumentation.

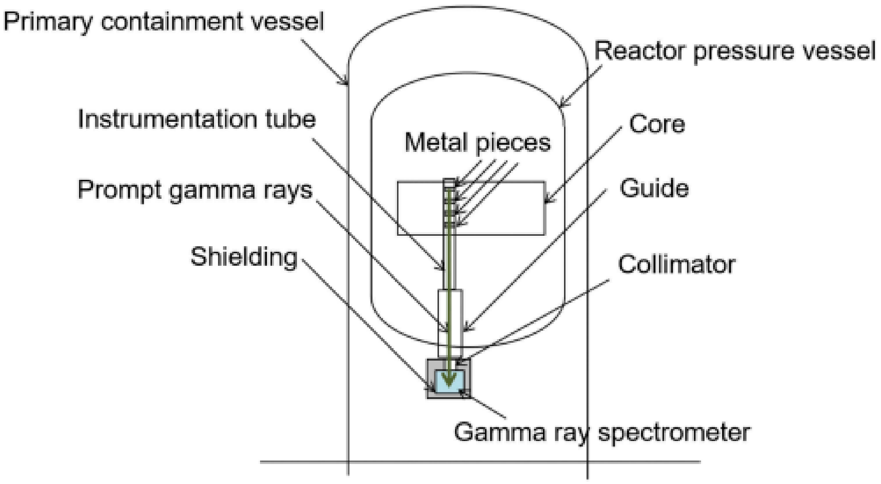

Figure 21: A schematic diagram of a conceptual reactor power monitoring system with the target metals for radiative neutron capture located inside the RPV and a γ-ray spectrometer placed externally [56]. Figure taken from [56].

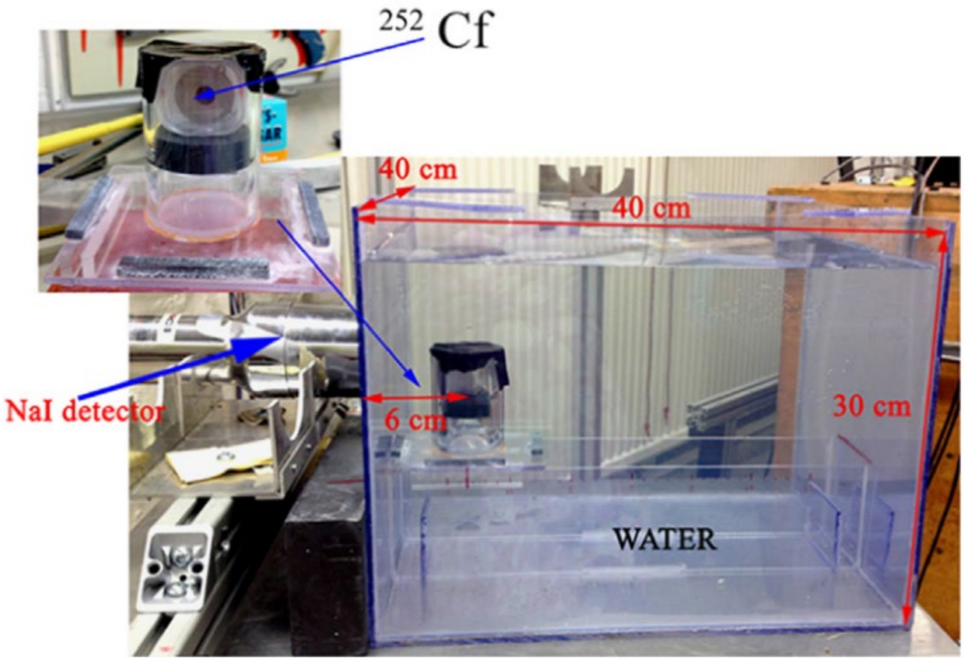

Figure 22: Photographs of the set-up for the measurement of thermal neutron flux in spent fuel storage ponds [57]. Figure taken from [57].

5. Conclusion

PGAA provides an effective method for the measurement of stable isotopes by inducing artificial γ radioactivity with neutrons. By separating γ-rays emitted promptly from delayed decay products, trace quantities of elements can be identified due via characteristic γ-ray emissions of their constituent isotopes. Hydrogen has been the subject of many PGAA research efforts due to its 1-to-1 neutron capture-γ emission relationship and the characteristic 2.223 MeV γ-ray. The detection of hydrogen allows for water measurement via the 2.223 MeV γ-ray, highlighting another advantage of PGAA, that is, independence from the chemical form of the target element. Reducing the incident neutron energy enables the increased radiative neutron capture cross-section to be exploited at lower neutron energies resulting ultimately in the use of cold neutron sources for very high-sensitivity applications. When unable to further moderate neutrons to a sub-thermal state, increasing the neutron flux is a complementary way to reduce the detection limit of PGAA systems.

Radiative neutron capture is relevant to analyses in many industries and a wide range of applications including the identification of trace elements, the structural analysis of engineering materials, and the detection of explosives and concealed items. PGAA of hydrogen in these areas is of interest due to, for example, the embrittlement effects of hydrogen in metals and other engineering materials. Absorption and desorption of water may also have negative effects on the structural and mechanical properties of materials.

Measurement of 2.223 MeV γ-rays from neutron capture on hydrogen provides an effective way to monitor moisture content, even in materials with a high, bound hydrogen content. Indirect neutron flux measurements by neutron capture of hydrogen have also been proposed as a method of locating FCM in 1F NPS. This approach may also be used to measure neutron flux in spent fuel storage ponds or inside the FCM in Chernobyl to predict changes in chemical properties and radiative emissions due to drying.

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