A new fast neutron facility for materials analysis at UCT

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Abstract. Fast neutron analysis is one of the techniques that can be used to perform qualitative and quantitative elemental analysis in a range of contexts, including the food, coal and minerals processing industries, and the detection of contraband. The technique makes use of mainly gamma rays as a characteristic signature, however, other signatures such as scattered and transmitted neutrons can be used for elemental characterisation. In 2017, the University of Cape Town (UCT) Department of Physics commissioned the n-lab, a fast neutron laboratory centred around a Thermo MP-320 deuterium-tritium Sealed Tube Neutron Generator (STNG) and a 220 GBq americium-beryllium (Am-Be) radioisotopic source. In this study, the aim is to characterise the n-lab and develop standardised analysis protocols for the elemental analysis of bulk materials. Experiments to characterise the facility have been undertaken, where the neutron yield produced by the STNG, was measured. The measured neutron yield was found to be $(1.22 \pm 0.10) \times 10^8$ neutrons s^{-1}, a value comparable to the specified yield in the STNG operation manual. Furthermore, proof-of-principle materials analysis measurements have also been made, where samples of graphite were characterised using prompt gamma ray neutron analysis. It was possible to positively identify ^{12}C using the 4.43 MeV gamma ray, which is induced by the inelastic scattering interaction between incident neutrons and the ^{12}C nuclei in the graphite sample.

1. Introduction

Fast neutrons can be used to perform elemental analyses of samples, both qualitatively and quantitatively through a range of techniques [1, 2]. A material of unknown composition is exposed to a field of neutrons for a predetermined period of time, resulting in the production of various signatures, such as gamma rays, scattered neutrons and transmitted neutrons [2, 3]. These signatures are highly characteristic of the individual constituent elements whose nuclei have interacted with the incident neutrons. In the case of gamma ray signatures, two techniques are commonly used: Prompt Gamma Neutron Activation Analysis (PGNAA) and Delayed Gamma Neutron Activation Analysis (DGNAA) [1]. Gamma rays are considered as prompt if their decay time is shorter than the resolving time (usually 10 ns to 10 μs) of the gamma ray detector system [4]. Delayed gamma-rays are usually due to activation of target nuclei, which is due to neutron capture or other reactions that lead to the production of radioactive isotopes [1].

A neutron facility for experimental neutron physics studies requires the availability of neutron sources, neutron and gamma-ray spectrometers. The neutron facility at UCT (n-lab) was commissioned in 2017 [5] and is centered around a Thermo MP-320 Sealed Tube Neutron Generator (STNG), which is an accelerator-based source that relies on the deuterium-tritium fusion reaction to produce 14.1 MeV

monoenergetic neutrons at a rate of approximately 1 x 10^8 neutrons s^{-1} as per manufacturer specification. Also available is a 220 GBq americium-beryllium (Am-Be) radioisotope neutron source that produces neutrons with a broad energy spectrum, with energies ranging from thermal to 11 MeV. Irradiation of samples can be performed either in the experimental area or close to the source inside the vault by positioning the sample through the variable collimator (see figure1).

We report on recent developments at the n-lab, particularly those focused on the development of techniques for the analysis of materials in bulk using the STNG.

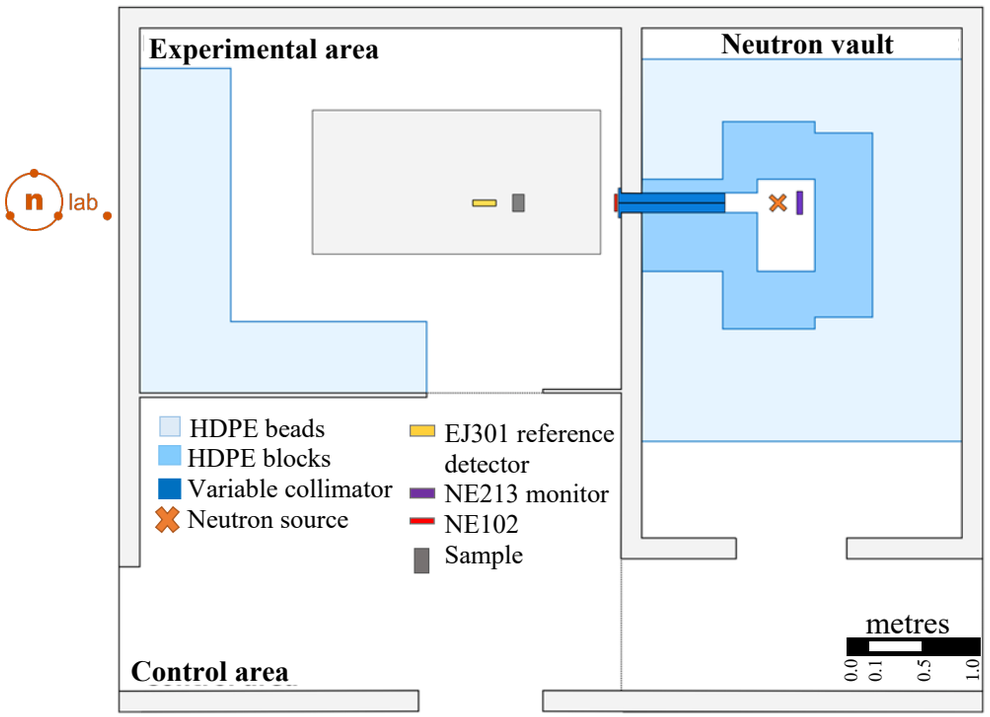

Figure 1. A schematic of the n-lab facility showing the neutron vault, where the STNG is well shielded with High Density Polyethylene (HDPE), the control area and experimental area.

2. Neutron yield measurements of the STNG

The analysis of materials using neutron techniques requires knowledge of the yield and energy distribution of the neutron field. A neutron yield and energy distribution can be measured by active methods such as using neutron spectrometers, or by passive methods, such as foil activation. At the n-lab, the foil activation method was used to measure the fast neutron yield produced by the STNG. Since this requires a material with a known cross section for specific incident neutron energy, a natural copper foil with dimensions of 2.0 x 2.0 x 0.1 cm^3 was used. The foil was placed at 12 ± 1 cm from the centre of the STNG and was irradiated at maximum operating capacity for approximately 2 hours.

Natural copper has two isotopes, ^{63}Cu and ^{65}Cu, with abundances of 69.15% and 30.85%, respectively. Both these isotopes have well-known and relatively high cross sections for (n,2n) reactions at 14.1 MeV. The ^{63}Cu(n,2n)^{62}Cu and ^{65}Cu(n,2n)^{64}Cu reactions have threshold energies of 11.03 MeV and 10.06 MeV [6], respectively, thus, 14.1 MeV neutron energies are sufficient to induce these reactions. The resulting radioactive products, ^{62}Cu and ^{64}Cu, have half-lives of 9 min and 12 h, respectively, and both decay by positron emission followed by two 0.511 MeV annihilation gamma rays.

The gamma rays were measured using a calibrated spectrometer consisting of a cerium-doped lanthanum bromide ($LaBr_3(Ce)$) detector and electronics for signal processing. The measurements were taken at 30 s intervals, for a total acquisition time of 2 h. The 0.511 MeV gamma ray peak in the spectrum shown in figure 2(a) has contributions from both ^{62}Cu and ^{64}Cu. The 0.511 MeV gamma ray counts from ^{64}Cu can be obtained by summing the count rates from t = 3030 s to t = 7200 s in figure 2 (b), since the radioactivity of ^{62}Cu is low in this region. Generally, the activity of an isotope diminishes to negligible levels after a period of time that is equal or more than five times its half-life [7].

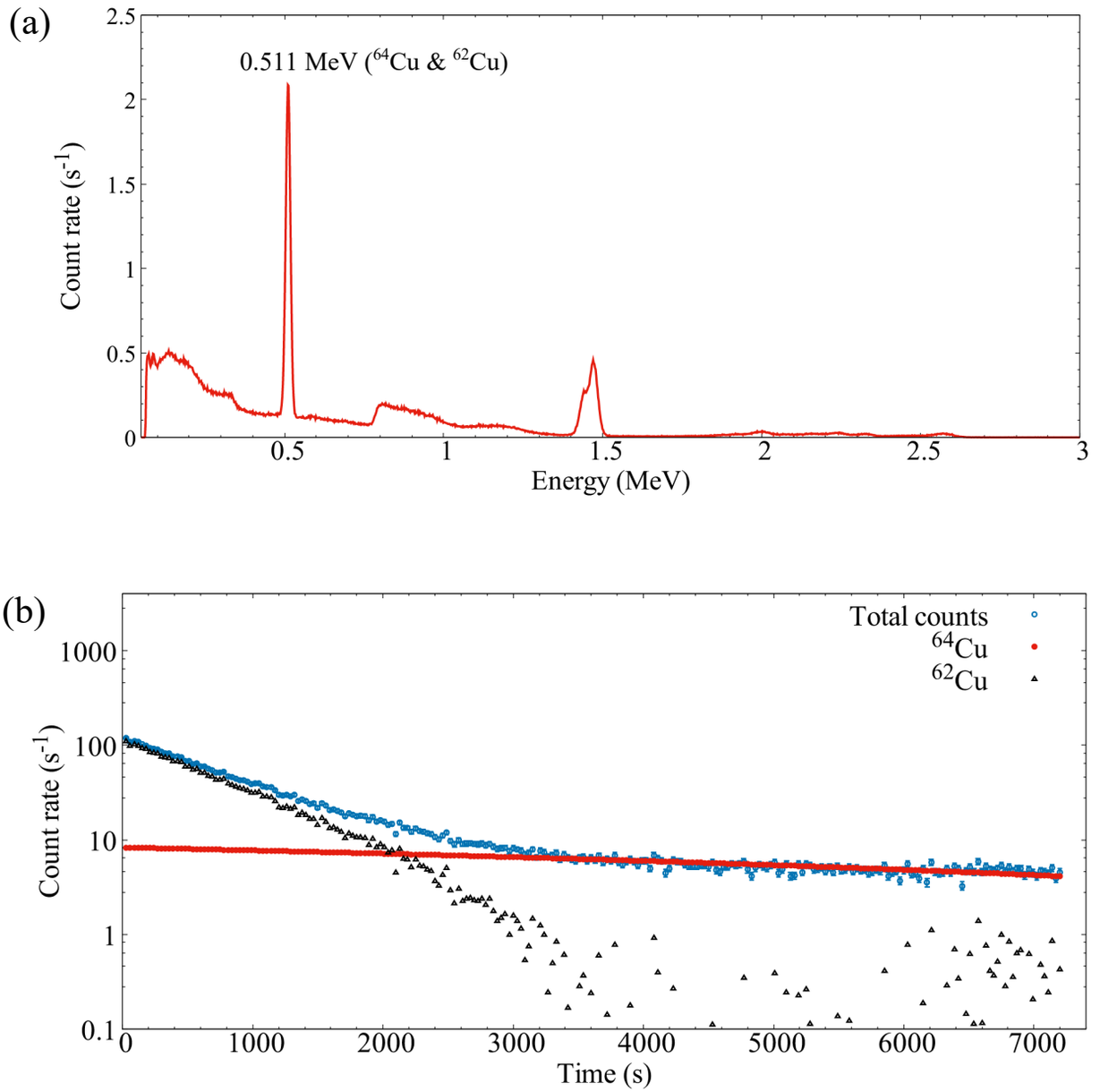

Figure 2. The figure shows (a) the gamma ray spectrum of the copper sample after irradiation with the STNG and (b) the 0.511 MeV gamma ray decay curves. The blue data points are 0.511 MeV gamma ray decays of the STNG irradiated copper. The red and black curves correspond to the decay of ^{64}Cu and ^{62}Cu, respectively.

The fitting of ^{64}Cu data in the 3030-7200 s region makes it possible to also estimate the number of counts from ^{64}Cu in the 0-3000 s region; the 0.511 MeV gamma ray counts from ^{62}Cu can be estimated by obtaining the difference between the abscissas of the ^{64}Cu and the total counts curves. The number of gamma rays attributed to ^{64}Cu, N_p, can then be used to calculate the flux using [7]

$$\Phi(E) = \frac{N_p M \lambda}{N_A m \sigma \theta I_\gamma \varepsilon} \frac{1}{(1 - e^{-\lambda t_i}) e^{-\lambda t_d} (1 - e^{-\lambda t_c})}. \quad (1)$$

The parameter definitions, as well as their values used to calculate the flux are summarised in table 1.

Table 1. The summary of the parameters used in equation (1) with their definitions. The values that were used to calculate the flux are also given, with their uncertainties where possible.

Symbol	Definition	Value
N_p	Net counts in the photopeak	44720 ± 210 counts
M	Isotopic mass	64.93 g mol^{-1}
λ	Decay constant of ^{64}Cu	1.52 x 10^{-5} decays s^{-1} [a]
N_A	Avogadro's constant	6.02 x 10^{23} atoms mol^{-1}
m	Mass of copper foil	5.34 ± 0.02 g
σ	Cross section at 14.1 MeV	(9.61 ± 0.14) x 10^{-25} cm^2 [a]
θ	Isotopic abundance of ^{65}Cu	0.31
I_γ	Gamma ray intensity	0.352 ± 0.004 [b]
ε	Detector efficiency at 0.511 MeV	0.189 ± 0.004
t_i	Irradiation time	7260 s
t_d	Delay time before counting	3144 s
t_c	Counting time	7200 s
Φ(E)	Neutron flux	(6.75 ± 0.13) x 10^4 cm^{-2} s^{-1}

[a]Adapted from [8]
[b]Adapted from [9]

Using the source-target distance of 12 ± 1 cm, it was then possible to calculate the neutron yield, Y, of the STNG in 4π sr using equation (2) given as

$$Y = 4\pi r^2 \Phi(E). \quad (2)$$

By substituting the neutron flux into equation (2) the neutron yield of the STNG was calculated to be (1.22 ± 0.10) x 10^8 neutrons s^{-1} in 4π sr, which is comparable to the expected value of 10^8 neutrons s^{-1}, as documented in the user manual.

3. Materials analysis using 14 MeV neutrons

We report on first proof-of-principle experiments to characterise graphite samples, irradiated in the experimental area with a 14.1 MeV neutron beam shaped by a 0.8 cm Ø x 100.0 cm HDPE collimator. At the n-lab, a graphite block with dimensions 13.8 x 5.9 x 5.9 cm^3 was irradiated for a period of 1 hour. The gamma ray spectra were acquired during irradiation with a calibrated 3″ NaI (Tl) detector positioned 30 cm from the center of the sample and 90˚ with respect to the beam direction (see figure 3), with and without the sample present.

The results of the experiment show promise that it is possible to positively identify carbon using the 4.43 MeV gamma rays from the $^{12}C(n,n')^{12}C$ reaction, though the rates are low. Figure 4 shows gamma ray spectra acquired during the irradiation of the graphite sample (red) and when the STNG was running without the graphite sample in the sample position (blue). While the 4.43 MeV peak and the single-escape peak at 3.92 MeV are present in both the spectra, it can be seen that the count rates are higher in the red spectrum due to the presence of graphite in the sample position. The 2.20 MeV and 4.43 MeV gamma ray photopeaks in the no sample spectrum are produced from the radiative capture on hydrogen, and inelastic scattering on carbon, respectively, in the HDPE shielding around the STNG.

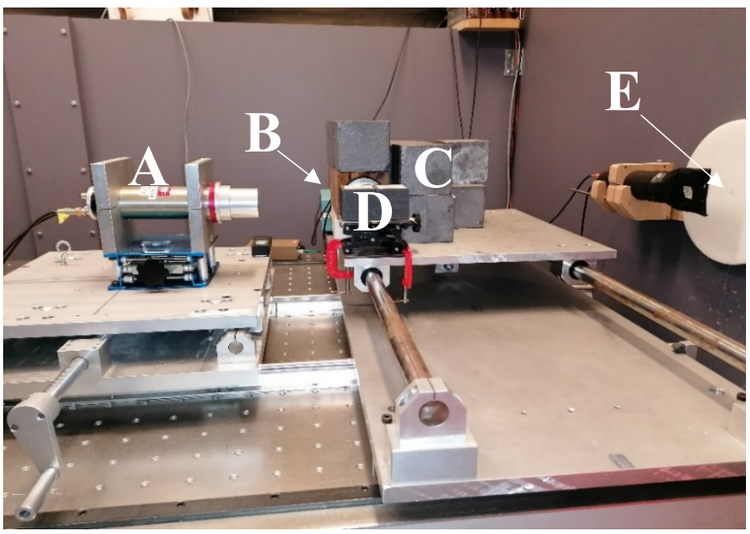

Figure 3. The experimental setup used for neutron analysis of graphite samples consisting (A) neutron detector, which was not in use, (B) 3″ NaI(Tl) detector, (C) lead blocks for shielding the NaI detector from gamma rays produced in the HDPE shielding and (D) the graphite block. The detector is positioned at a 90˚ angle with respect to the beam direction. Neutrons exit of the collimator at (E).

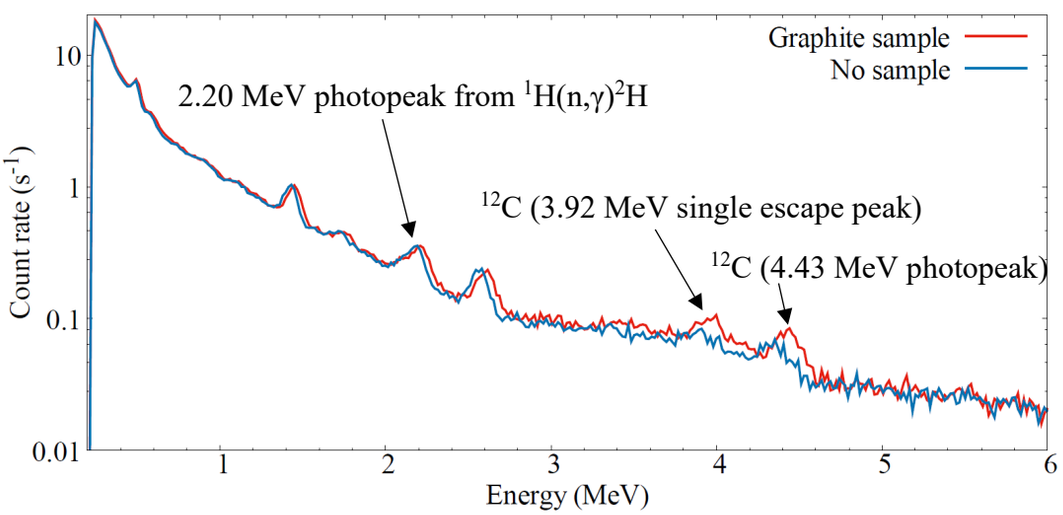

Figure 4. The spectrum acquired during the irradiation of a graphite sample (red) superimposed on the spectrum acquired during the run of the STNG without the graphite sample (blue) in the sample position.

4. Conclusion

The analysis of materials using fast neutrons requires a well characterised neutron field. At the n-lab, the maximum neutron yield from the 14.1 MeV STNG was measured to be $(1.22 \pm 0.10) \times 10^8$ neutrons s^{-1} and is in agreement with the value of 1×10^8 neutrons s^{-1} specified by the manufacturer. Furthermore, the prompt gamma neutron analysis of graphite samples shows that it is possible to qualitatively characterise carbon using the 4.43 MeV gamma rays from the $^{12}C(n,n')^{12}C$ reaction.

In the next phase of the study the experimental setup will be redesigned to improve gamma ray count rates by increasing the neutron flux and the geometric detector efficiency. This will be achieved by increasing the diameter of the collimator and by using multiple gamma ray detectors. The measurement of other signatures (scattered and transmitted neutrons) will be explored in parallel. Ultimately, the goal is to develop and demonstrate multi-modal standardised measurement techniques for elemental characterisation of samples in bulk. It is envisaged that this will improve the sensitivity and reduce or eliminate ambiguity where there are suspected interferences when analysing a multi-elemental sample.

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