Characterisation of bulk materials using fast neutron transmission analysis

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Abstract. The use of neutrons for the purposes of analysing elemental composition or constructing an image of an object is one of the most common applications of neutrons in both research and industry. Neutrons interact with matter in several ways producing secondary radiation which is characteristic of the material being interrogated and their composite nuclei. In fast neutron transmission analysis (FNTA), measurement of the neutrons which are transmitted through a material can be used to infer the composition. We present the results from the proof-of-principle measurements with hydrogen and carbon taken at the n-lab, University of Cape Town. The results show the possibility to positively identify ^{12}C and ^{1}H in graphite and in high density polyethylene materials via cross section analysis.

1. Introduction

The interrogation of materials in order to determine their elemental composition is of interest to industry and in academic research. There are many well established techniques that are regularly used for this purpose [1,2], and in particular neutron-based techniques have seen development in the past decades [3, 4]. Neutron techniques typically interrogate a material by interacting with atomic nuclei, inducing secondary radiation which are characteristic of the composite nuclei of the material [5]. The types of radiation that can be induced, and measured, include prompt and delayed gamma rays, and scattered neutrons [4, 5]. Alternatively, investigation of neutrons which have not interacted within a material i.e. the transmitted neutrons, relative to knowledge of the incident neutron field, provides a characteristic signature which can be used to determine composition as in fast neutron transmission analysis (FNTA) [6].

The practical implementation of FNTA involves irradiating a sample with a well-characterized (energy, intensity and angle) neutron beam, and measuring the transmitted neutron energy spectrum. For a pencil beam of neutrons incident on a bulk sample with thickness t, the attenuation can be described by Eq. 1 [7], where $I(E)$ and $\Sigma_R(E)$ are the energy dependent neutron intensity and removal cross sections [8] respectively.

$$I(E,t) = I \exp(-\Sigma_R(E)\, t) \tag{1}$$

For a composite material the removal cross section is a linear combination of the constituents, weighted by their respective mass ratios. Removal cross sections are unique for each constituent and can be determined by measurement, simulation or from literature depending on the application. Previous work has demonstrated the use of FNTA to determine the water content in a concrete sample [6], or the elemental composition of sand [7]. In this work we present recent results from neutron transmission measurements through graphite (C) and polyethylene ($(C_2H_4)_n$) samples.

2. Experimental setup and procedure

The measurements in this work were taken using A D-T MP320 (Thermo Fisher) sealed tube neutron generator (STNG) at the n-lab, a fast neutron facility at UCT that was commissioned in 2017 [9]. The D-T STNG produces neutrons of energy 14.1 MeV which were collimated into a 0.8 cm diameter pencil beam (Figure 1). A 2" x 2" EJ301 organic liquid scintillator detector was positioned in the beam, and operated at a negative bias of 1100 V. Signals from the anode (fast) and dynode (slow) were acquired with a CAEN DT5370 digitiser coupled to the QtDAQ software [10]. As the detector was sensitive to both neutron and gamma ray radiation, pulse shape discrimination (PSD) was implemented through QtDAQ using the fast anode signal [11,12] in order to select only for neutron induced events. Energy information was obtained from the pulse height of the dynode signal after shaping and amplification with an Ortec 113 pre-amplifier and 427A amplifier. The resulting pulse height (L) spectra were calibrated using a series of gamma ray sources to produce an electron equivalent energy (MeV_{ee}) scaling. In organic scintillators, gamma rays interact primarily by Compton scatter, and the Compton edges [13] for ^{137}Cs, ^{60}Co, ^{22}Na and ^{241}Am-^{9}Be gamma rays were used for calibration.

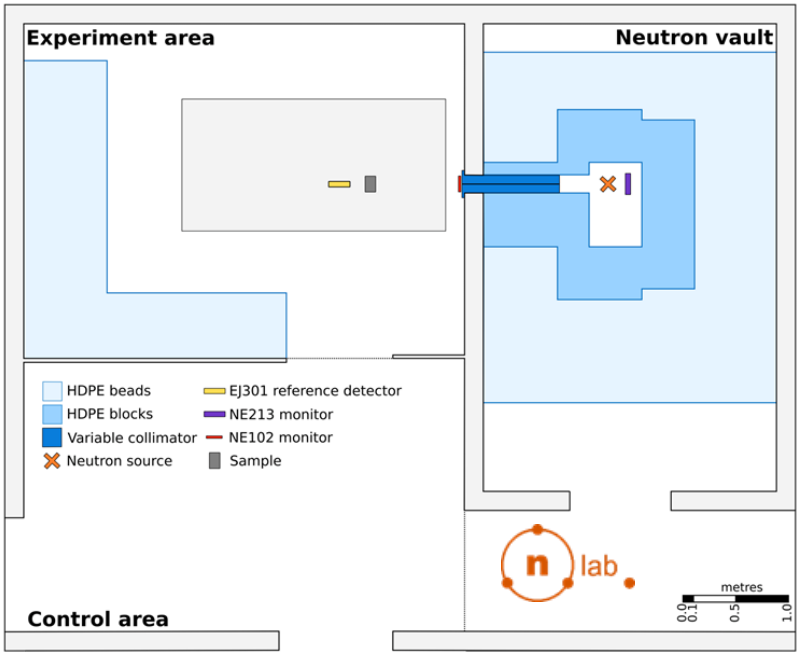

Figure 1. A schematic of the n-lab facility showing the neutron vault, where the STNG is well-shielded with High Density Polyethylene (HDPE), the control area and the experimental area.

To explore the use of FNTA for elemental analysis two samples were chosen: graphite; and high density polyethylene (HDPE). Graphite is an easily obtainable form of pure carbon, and is primarily comprised of ^{12}C. The samples used in this work consisted of solid blocks with a density of 1.8 g cm^{-3}, cross sectional area of approximately 5.0 x 5.0 cm^2 and thicknesses up to 14.3 cm. High density

polyethylene is a simple combination of hydrogen and carbon with the chemical formula $(C_2H_4)_n$. Sheets of HDPE were used, with a density of 0.9 g cm^{-3}, cross sectional area of 50.0 x 50.0 cm^2 and thicknesses up to 5.0 cm. An example of the experimental set up can be seen in Figure 2.

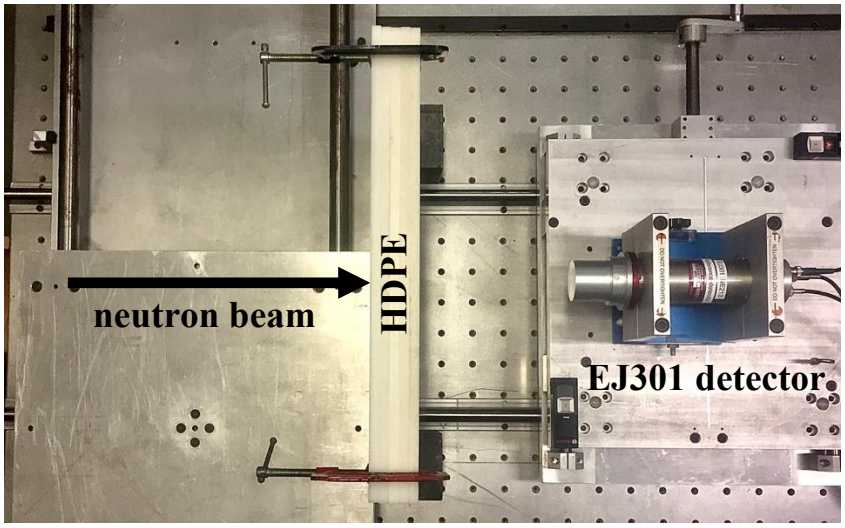

Figure 2. Experimental setup for a 0.8 cm diameter pencil beam of STNG neutrons incident on 5 x 1.0 cm thick HDPE samples. Neutrons transmitted through the sample are detected with an EJ301 organic liquid scintillator.

The samples were positioned at 74 cm from the collimator exit, while the reference detector was positioned at 120 cm from the collimator exit, at an angle of 0 degrees with respect to the beam axis. The samples were irradiated until a minimum of 10^5 neutron events were recorded. The measured light output spectra were calibrated, and normalized with respect to the acquisition time.

3. Results and analysis

Measurements of the transmitted neutrons were made for multiple thicknesses of each sample. Figure 3 shows the normalized and calibrated neutron light output spectra measured for graphite and HDPE. As expected from Eq. 1, increasing the amount of material results in a decrease in the transmitted neutron intensity. The no sample case is taken to be representative of the incident neutron spectrum, $I(0)$.

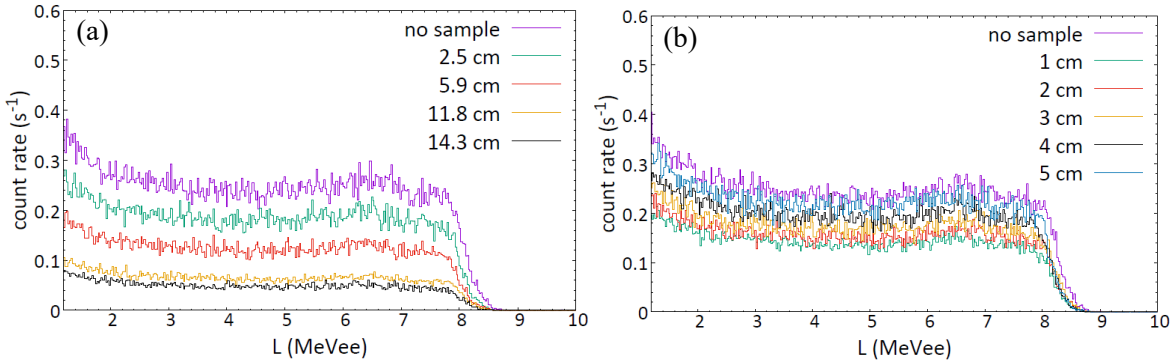

Figure 3. Plots showing the calibrated pulse height spectra of transmitted 14.1 MeV neutrons through (a) graphite and (b) HDPE.

The effective removal cross section at 14.1 MeV for each sample was obtained from the fit parameters of the transmitted neutron intensity (obtained by integrating count rates within the 1.2 MeV_{ee} to 8.4 MeV_{ee} range of L in the pulse height spectra) with respect to the material thickness. The correlation between neutron intensity and material thickness is exponential for both graphite and HDPE, as seen in Figure 4.

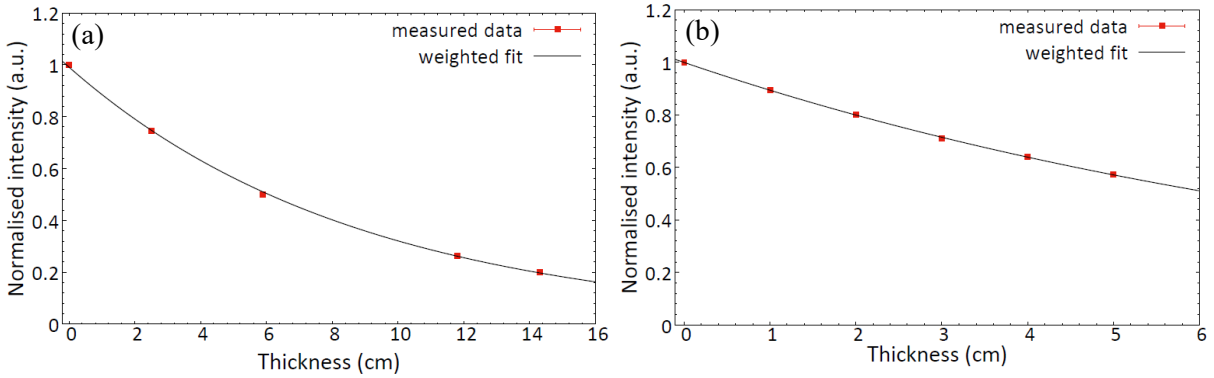

Figure 4. Transmitted neutron intensity as a function of sample thickness at 14.1 MeV for (a) graphite and (b) HDPE. The uncertainties of transmitted neutron intensities for both the materials ranged between 0.3% to 0.5%.

Equation 1 was linearized (Eq. 2) and a weighted linear least squares minimization used to determine $\Sigma_R(14.1\ \text{MeV})$ from the fit parameters. The data were well-fitted, with a Chi-squared per degrees of freedom being 3.4. The effective removal cross sections for graphite and HDPE are presented in table 1.

$$\ln\left(\frac{I(E,0)}{I(E,t)}\right) = \Sigma_R(E)\, t \tag{2}$$

Table 1. Effective removal cross sections for 14.1 MeV neutrons.

Material	**Σ_R(cm^{-1})**
Graphite	0.1150 ± 0.0012
HDPE	0.1118 ± 0.0007

The effective removal cross section Σ_R is regarded as a bulk property and depends on the physical preparation of the sample. In order to develop elemental signatures, it is necessary to define a microscopic effective removal cross section σ_R, which removes the dependence on the physical form of the sample. The microscopic removal cross section is related to Σ_R by:

$$\Sigma_R = \frac{\rho N_A}{A}\,\sigma_R \tag{3}$$

where ρ is the sample density, N_A is Avogadro's number, and A is the molar mass of the isotope, element, or compound as required. As graphite is comprised solely of C, the elemental microscopic removal cross section $\sigma_{R,C}$ was directly obtained from the measured value for Σ_R. High density polyethylene is comprised of a known ratio of H and C, so the elemental microscopic removal cross section for H can be extracted according to Eq. 4:

$$\sigma_{HDPE} = 2\sigma_{R,C} + 4\sigma_{R,H} \quad (4)$$

The microscopic removal cross sections σ_R for C and H are presented in table 2 together with the total microscopic cross sections σ_t as defined in the ENDF/B-VIII.0 data library [14].

Table 2. Microscopic cross sections of C and H for 14.1 MeV neutrons.

Element	**σ (b)**	
	σ_R	σ_t
C	1.265 ± 0.037	1.298 ± 0.055
H	0.750 ± 0.002	0.676 ± 0.023

4. Discussion and Conclusion

In this work we have demonstrated the use of fast neutron transmission analysis, and effective removal cross sections to investigate the composition of materials in bulk. In order to deconvolve the composition it is necessary to produce a series of unique responses, from transmitted neutron measurements, and measurements of other signatures, for each element of interest.

The macroscopic removal cross sections at 14.1 MeV measured for graphite and HDPE, were used to extract the microscopic removal cross sections of C and H, which were found to be 1.265 ± 0.037 barn and 0.750 ± 0.002 barn respectively. There is agreement to within experimental uncertainty between the measured (removal) cross section and total microscopic cross section (literature) of C [14], but this is not the case for H, which requires further consideration. Based on the empirical rule that removal cross sections are approximately equal to two thirds of the total cross sections for neutron energies in the 6 MeV to 8 MeV range, we expect our measured values to be lesser than the total cross sections since this has been shown to also apply to 14 MeV neutron energies [15]. We also expect the ratios of the removal cross sections to total cross sections to be constant for both H and C [15].

While there are still outstanding issues to be resolved, the results presented here show promise with respect to using FNTA for materials analysis. Additional measurements are ongoing to investigate the effective removal cross section over a broader range of neutron energies using an ^{241}Am-^{9}Be radioisotopic source, in combination with gamma ray measurements with the STNG to generate multi-modal elemental response functions.

Acknowledgements
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