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Design and expected performance of a fast neutron attenuation probe for light element density measurements

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ABSTRACT

We present the design and expected performance of a proof-of-concept 32 channel material identification system. Our system is based on the energy-dependent attenuation of fast neutrons for four elements: hydrogen, carbon, nitrogen and oxygen. We describe a new approach to obtaining a broad range of neutron energies to probe a sample, as well as our technique for reconstructing the molar densities within a sample. The system's performance as a function of time-of-flight energy resolution is explored using a Geant4-based Monte Carlo. Our results indicate that, with the expected detector response of our system, we will be able to determine the molar density of all four elements to within a 20–30% accuracy in a two hour scan time. In many cases this error is systematically low, thus the ratio between elements is more accurate. This degree of accuracy is enough to distinguish, for example, a sample of water from a sample of pure hydrogen peroxide: the ratio of oxygen to hydrogen is reconstructed to within $8\pm0.5\%$ of the true value. Finally, with future algorithm development that accounts for backgrounds caused by scattering within the sample itself, the accuracy of molar densities, not ratios, may improve to the 5–10% level for a two hour scan time.

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1. Introduction

1.1. Material identification techniques using neutrons

Since the late 1980s, renewed interest in screening capabilities has resulted in significant investment in Thermal Neutron Analysis (TNA) and Fast Neutron Analysis (FNA) techniques as a complement to x-ray techniques. Because x-ray devices are only sensitive to variations in density, they do not provide effective material identification for light elements. In TNA, gamma particles resulting from a handful of thermal neutron capture reactions in materials of interest are used as a means of identification. FNA measures either the gamma rays from nuclei activated by a neutron beam, or the energy-dependence of an attenuated neutron beam. Several groups have demonstrated elemental imaging capabilities, but differ in their method of obtaining a range of neutron probe energies (see [\[1\]](#page--1-0) for a review). Pulsed Fast Neutron Transmission Spectroscopy (PFNTS) employs the Time-of- Flight technique combined with sophisticated high-flux, pulsed accelerator technology to determine neutron energies [\[2,3\]](#page--1-0). Another method for obtaining a range of probe energies described in [\[4\]](#page--1-0) involves accelerating deuterium or protons onto a deuterium or lithium

* Corresponding author. E-mail address: msweany@sandia.gov (M. Sweany). target: neutrons are produced with energies dependent on the angle between the beam and detectors. This method requires multiple measurements at different orientations to obtain a range of energies, rather than a simultaneous measurement over all energies, increasing scan times. More recently, [\[5\]](#page--1-0) used deuterium on a boron target to obtain several discrete neutron energies as a sample probe. The transmitted flux at each of these energies, measured by the energy deposited in liquid scintillator detectors, was used to inform the hydrogen content in samples. Finally, the CSIRO/NUTECH detector $[6]$ uses 14.1 MeV neutrons from a $(d-T)$ source in combination with 6/3 MeV x-rays from a linear accelerator to determine both the average material density and composition of materials. The ratio of neutron and x-ray penetration, R, is indicative of the material present, and impressive 2D images have been obtained with conveyor belt speeds of 1–2 m/min. However for both illicit drugs and explosives, the discrimination parameter R has a significant overlap with common materials, causing a high false alarm rate.

1.2. Light element reconstruction with fast neutron attenuation

Our system is a neutron time-of-flight spectrometer consisting of two detector planes, designed to measure the unique attenuation spectrum of light elements. Specifically, the elements carbon, nitrogen and oxygen have many resonances in the total neutron cross section in the 1–10 MeV range, shown in [Fig. 1.](#page-1-0) Given

Fig. 1. Left: The total neutron interaction cross section spectra for hydrogen, carbon, nitrogen, and oxygen, from ENDF/B-VII.0 [\[7\].](#page--1-0)

sufficient energy resolution and a wide-range of energies to probe a sample, the attenuation spectrum can be used to uniquely determine the molar fractions of these elements contained in the sample. Although hydrogen does not have resonances, the energy dependance of its cross section does enable its molar fraction to be determined as well. The fraction of the aforementioned four elements can in many cases uniquely identify the material in question [\[1\]](#page--1-0).

We create a range of probe energies with a mono-energetic neutron source by down-scattering in the forward detector plane of the system. A neutron from a d-T generator elastically interact in the forward detector plane, depositing energy to start a time-offlight clock. For a single interaction, the energy of the neutron after this interaction (the probe energy) is dependent on the scattering angle, θ :

$$
E_i = \frac{14.1 \text{MeV}}{1 + \tan^2 \theta}.
$$
\n⁽¹⁾

The neutron then passes through the sample region to the back detector plane, depositing energy and stopping the time-of-flight clock. The energy probing the sample is then calculated through time-of-flight with the distance between detector centers and measured time (r and t, respectively) between the two detector hits:

$$
E = \frac{m}{2} \left(\frac{r}{t}\right)^2. \tag{2}
$$

While the energy deposited in the forward detector plane is also a handle on the energy as it passes through the sample, the resolution is not as good as that determined by time-of-flight. Because we are using time-of-flight to directly calculate the scattered neutron's energy, we are able to remove the assumption that a single interaction occurred in the forward detector plane. After many events over a range of scattering angles a spectrum is obtained. By comparing spectra with and without a sample present, an energy dependent attenuation is obtained. A two-detector version of this system is described in $[8]$: the goal of the work described here is to scale to a larger system for faster scan times, and to develop a model which will be validated with data in the future. This model will be used to estimate the limits of this method given variables such as acquisition time, and timing and position resolution. Future studies will focus on room return backgrounds and how the number of detectors in the system effects reconstruction errors and background levels.

In Section 2 the Geant4-based Monte Carlo model is described, including geometry, event generation, and detector response. An evaluation of the detector system's performance is discussed in [Section 3:](#page--1-0) although there is the potential for imaging capabilities with future algorithm development, in this work we evaluate material reconstruction alone. Finally, future work is discussed in [Section 4.](#page--1-0)

2. Geant4-based detector model

Below we describe the geometry, event generation, and processing for these simulations. In the following, an event is denoted as any interaction resulting from one call to the primary generator class of Geant4: in this case, a call to the primary generator class results in one primary particle released into the world volume. To avoid confusion, what is commonly described as a detector event, resulting from e.g. energy deposited in scintillator, is referred to as a deposition. For all simulations in this work, Geant4 [\[9\]](#page--1-0) version 10.01 is used with the QGSP_BERT_HP physics list. The goal of these simulations is to understand the effects of position and timing resolution of the individual detectors on the reconstructed elemental molar densities for a range of materials, as well as to understand and account for effects such as scattering within the sample.

Our system is 32 channels, each consisting of a 3×3 inch right cylindrical aluminum cell, 0.1 inches thick on the top and 0.045 inches thick on the rounded side, containing liquid scintillator (EI-309) and painted with a highly reflective coating (EJ 520). Coupled to each cell is a 3-inch PMT (Electron Tubes model 9821 KB) shielded from the earth's magnetic field with mu-metal casing: according to the manufacturer [\[10\]](#page--1-0), the typical electron transit time spread is 2.2 ns FWHM, and the single electron width is 3.2 ns (FWHM). The timing of our system is therefore limited to a standard deviation of approximately 1 ns. It is feasible that we could reach this limit using a 500 MHz digitizer, so for these studies we explore performance with timing resolutions of 2.5, 1, 0.5, and 0 ns in order to span the range from worst case scenario, likely scenario, likely improvement with upgraded PMTs, and ideal. For the position, the system is limited by the size of the cells and the probability of neutron interactions within each of them. Because the interaction position is geometry dependent, the interaction position for each cell is approximated by convolving the average simulated true interaction position distribution. The specifics of this characterization are described in [Section 2.3](#page--1-0).

We calculate the error on time-of-flight energy, E_i , as:

$$
\sigma_E = \sqrt{\left(\frac{\partial E}{\partial x}\right)^2 \sigma_x^2 + \left(\frac{\partial E}{\partial z}\right)^2 \sigma_z^2 + \left(\frac{\partial E}{\partial t}\right)^2 \sigma_t^2}
$$
\n(3)

$$
\frac{\sigma_E}{E} = 2\sqrt{\left(\frac{1}{r}\right)^2 \sigma_x^2 + \left(\frac{1}{t}\right)^2 \sigma_t^2}
$$
\n(4)

where r and t are the distance and time separation of two neutron depositions, and σ_x and σ_t are the standard deviation of the position and time, as discussed above. The position resolution is illdefined in this case, as we have characterized the overall resulting energy resolution, $f(E)$, due to the average position response for each individual detector. Eq. (4) becomes:

$$
\frac{\sigma_E}{E} = \sqrt{\left(\frac{f(E)}{E}\right)^2 + 4\left(\frac{1}{t}\right)^2 \sigma_t^2}
$$
\n(5)

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