

Development of an instrument for non-destructive identification of Unexploded Ordnance using tagged neutrons - a proof of concept study

Sudeep Mitra and Istvan Dioszegi

Abstract— This work reports on the efficacy of using 14 MeV neutrons tagged by the associated particle neutron time-of-flight technique (APnTOF) to extract neutron induced characteristic γ -rays from an object-of-interest with high signal-to-noise ratio (SNR) and without interference from nearby clutter. A small portable API120 neutron generator was operated at a continuous output of $\sim 1 \times 10^7$ n/s while the γ -rays were detected using a 12.7x12.7 cm diameter NaI(Tl) detector. The α -particle detection system comprised a 5-cm diameter fast Amperex XP2020 photomultiplier tube which was mated with the 6.5 cm fiber-optic face-plate of the neutron generator's ZnO(Ga) α -detector. Standard NIM electronics was used for the α - γ coincidence spectroscopy. A graphite cube of dimension 15.2 cm (6 kg) was used as the object-of-interest while a half-gallon bottle of water and a 5-mm thick iron slab were used as clutter items in non-overlapping and overlapping modes. In all cases, carbon (C) signals from the graphite were successfully extracted without interference from the oxygen (O) signals from water. The SNR at the peak energies of 4.43 (C) and 6.13 MeV (O) were found to be 1.3 when spectra were acquired without the time coincidence mode. The SNRs were vastly improved; 22 and 10 for C and O respectively in the coincidence mode.

I. INTRODUCTION

RANGE clearance operations at munitions testing grounds must discriminate Unexploded Ordnance (UXO) from clutter items and distinguish UXO filled with High Explosives (HE) from those with inert fillers. Non-destructive technologies are thus necessary for the cost-effective disposal of UXO during remediation of such sites. The only technique showing promise so far for the non-destructive elemental

characterization of UXO fillers utilizes neutron interactions with the material to detect carbon (C), nitrogen (N) and oxygen (O) which have unique ratios in HE [1], [2]. Neutron – interrogation methods for fill material exploit either or both of two types of neutron interactions with nuclei, (a) inelastic scattering, and, (b) neutron capture, and then detects the induced element- specific high-energy prompt γ -rays. While these systems [3], [4] have some capability to characterize larger ammunitions (shells 81 mm diameter and above), several unresolved issues hinder the wide application of this potentially very suitable technique. The most important one is that neutrons interact with all surrounding matter in addition to the interrogated material, leading to a very high γ -ray background in the detector. Systems requiring bulky shielding and having poor signal-to-noise ratios (SNRs) for measuring elements are unsuitable for field deployment.

The inadequacies of conventional neutron interrogation methods are overcome by using the tagged-neutron approach, and the availability of compact sealed neutron generators exploiting this technique [5] - [8] offers field deployment of non-intrusive measurement systems for detecting threat materials, like explosives and drugs. By accelerating deuterium ions into a tritium target, the subsequent fusion reaction generates nearly back-to-back emissions of neutrons and α -particles of energy 14.1 and 3.5 MeV respectively. A position-sensitive detector recognizes the associated α -particle, thus furnishing the direction of the neutron. The tagged neutrons interact with the nuclei of the interrogated object, producing element-specific prompt γ -rays that the γ detectors recognize. Measuring the delay between the detections of the α -particle and the γ -ray determines the location where the reaction occurred along the axis of the neutron beam (14.1 MeV neutrons travel at 5 cm/ns, while γ -rays cover 30 cm/s). The main advantage of the technique is its ability to simultaneously provide 2D and 3D imaging of the elemental composition of the interrogated objects. This work reports on the efficacy of using 14 MeV neutrons tagged by the associated particle neutron time-of-flight technique to extract neutron induced characteristic γ -rays from an object-of-interest with high SNR and without interference from nearby clutter.

Manuscript received November 3, 2011. This work was supported by the U.S. Department of Defense under its Strategic Environmental Research and Development Program (SERDP), Project# MR 1769. This manuscript has been co-authored by employees of Brookhaven Science Associates, LLC, under Contract no. DE-AC02-98CH10886 with the U.S. Department of Energy. The publisher, by accepting the manuscript for publication, acknowledges that the U.S. Government retains a nonexclusive, paid-up, irrevocable, worldwide license to publish or reproduce the published form of this manuscript, or allow others to do so, for U.S. Government purposes.

Sudeep Mitra is with the Environmental Sciences Department, Environmental Research and Technology Division, Brookhaven National Laboratory, Upton, NY 11973 USA (telephone: 631-344-6377, e-mail: smitra@bnl.gov).

Istvan Dioszegi is with the Nonproliferation and National Security Department, Brookhaven National Laboratory, Upton, NY 11973 USA (telephone: 631-344-7020, email dioszegi@bnl.gov).

II. METHODS

A. Instrumentation

The APnTOF system is a modular system, with independent functional modules. The main modules are:

- i) *The neutron generator*, ii) *The α -particle detector external read-out system*, iii) *The γ -ray detector*, and iv) *The data acquisition electronics (signal processor)*.
- i) *The neutron generator*. The sealed-tube neutron generator was a Thermo Electron Corporation's API 120 system [6], which is lightweight and portable. It produces 14 MeV neutrons via deuterium-tritium fusion reactions and the associated α -particles that are used for tagging the neutrons are detected by an internal sensor comprising a 6.5 cm diameter fiber-optic-face-plate (FOFP) coated with a fast light output ZnO(Ga) scintillator. The neutron generator was operated at an output of $\sim 1 \times 10^7$ n/s as determined using the Cu foil activation method. The reaction used is $^{63}\text{Cu}(n,2n)^{62}\text{Cu}$ which has a threshold energy of 11.5 MeV and is counted for 0.51 MeV γ -rays from positron annihilation. The neutron output fluctuations measured independently by counting the α -detector signal was less than 1%.
- ii) *The α -particle detector external read-out system*. A 5.1 cm diameter Amperex XP2020 fast photomultiplier tube (PMT) was mated with the 6.5 cm diameter fiber-optic-face-plate of the neutron generator's α detector.
- iii) *The γ -ray detector*. A 12.7 x 12.7 cm diameter NaI(Tl) γ -ray detector was refurbished with a fast 7.6 cm diameter Hamamatsu R1351 PMT. A light guide was used as an optical interface for uniform scintillation light collection from the larger diameter detector crystal. The γ -ray detector was shielded from direct neutrons by using a combination of iron, borated polyethylene composite and lead.
- iv) *The data acquisition electronics (signal processor)*. Conventional laboratory based NIM electronics were used for this study. The anode signals from the α - and γ -ray detectors were passed through a linear amplifier and fed to a Constant Fraction Discriminator (CFD) Unit. The delay of 63 ns in the α -channel served to compensate for the delay in triggering the discriminator of the γ -ray detector channel. The α -channel was fed to the STOP input while the γ -ray detector channel was fed to the START input of a Time-to-Amplitude Converter (TAC). The time difference between the START and STOP channels were converted to an amplitude pulse and recorded by a PC-based multi-channel analyzer (MCA) as the α - γ coincidence time spectrum. The information on the position of the interrogated object was contained in the time spectrum. A built in single channel analyzer (SCA) in the TAC unit allowed the setting of time windows to gate the MCA for recording the corresponding time correlated γ -ray energy spectrum. The energy spectra were shaped with 0.75 μ s shaping time. The schematic of the NIM electronics is shown in Fig. 1.

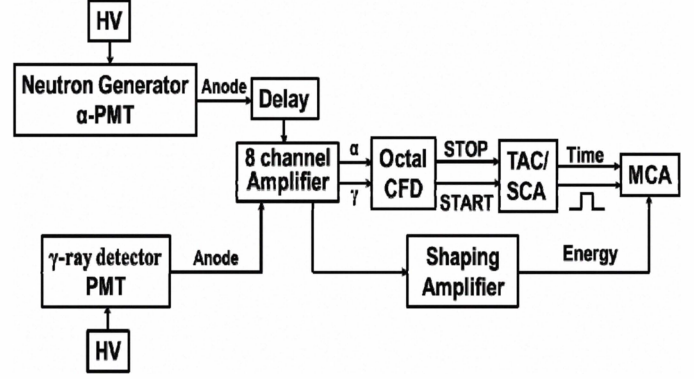


Fig.1. Schematic of the electronics for acquiring time and γ -ray energy spectra.

B. Proof-of-concept studies

Controlled bench-top experiments were conducted to simulate conditions that might be encountered in the field. In particular, it was intended to demonstrate the efficacy of the APnTOF technique in isolating γ -rays from a material of interest which are produced in the region defined by the neutron cone and in eliminating signals from clutter items that lie close to the object. A graphite cube of dimension 15.2 cm (mass 6 kg) was used as the object-of-interest ("UXO") while a half-gallon bottle of water and a 5 mm thick iron slab were used as clutter items. It was also intended that the iron slab would simulate shell thickness. Three cases were investigated to extract carbon signals from the graphite without interference from the oxygen signals from water: In case 1 (no overlapping clutter), the bottle of water was outside the defined neutron cone; cases 2 and 3 (overlapping clutter) progressively introduced the bottle of water and iron slab between the neutron beam and the graphite sample. For all cases the graphite block was at a distance of 47 cm along the axis of the neutron beam and the bottle of water was at a radial distance of 27 cm from the neutron generator target. In a separate series of experiments, time correlated γ -ray spectra of the graphite block and a one gallon bottle of water were independently determined when located in the neutron cone at a distance of 27 cm from the target. The SNR are compared with the respective γ -ray spectra when no coincidence time-gate was applied.

III. RESULTS AND DISCUSSION

A. Signal-to-noise

The time dependence of α - γ coincidences for an object (graphite or a one-gallon bottle of water) positioned at a distance of 27 cm from the neutron generator target is shown in Fig. 2. The peak at approximately channel number 455, to the right of the main peak is due to time correlated neutrons being scattered into the γ -ray detector. The γ -ray spectra of carbon and oxygen from the graphite and water bottle respectively, were obtained with a 9 ns wide time slice and are shown in Figs. 3 and 4 respectively. To compare the APnTOF technique with the conventional neutron interrogation technique, the γ -ray spectra of the same objects were also measured with no time restrictions on the data

acquisition. The spectra are presented in Fig. 5 for a 600s acquisition time. This corresponds to a continuous neutron output, typical for a one hour operation of a 14 MeV pulsed neutron generator operating at a 10 kHz repetition rate and 15 μ s wide neutron pulse. The spectra with no time restrictions show the high γ -ray background from surrounding material and from neutron reactions in the detector. The peaks due to the shielding materials, lead (2.62 MeV) and carbon (4.43 MeV) from the borated polyethylene clearly dominate the spectra. As a comparison, the random background recorded with a 9 ns window which was set from the region shown in Fig. 2 was almost a factor 10^4 lower than the background obtained with no time window. The basic difference between the two background determinations is that the spectrum with no time window can only be determined without an object in the beam, whereas the time gated background determined using the APnTOF technique is obtained with the object in the beam. The latter background more accurately reflects the neutron scattering effects related to the object. The SNR at the peak energy of 4.43 (C) and 6.13 MeV (O) was found to be 22 and 10 for C and O respectively in the coincidence mode. These values were vastly inferior; 1.3 when spectra were acquired without the time coincidence mode.

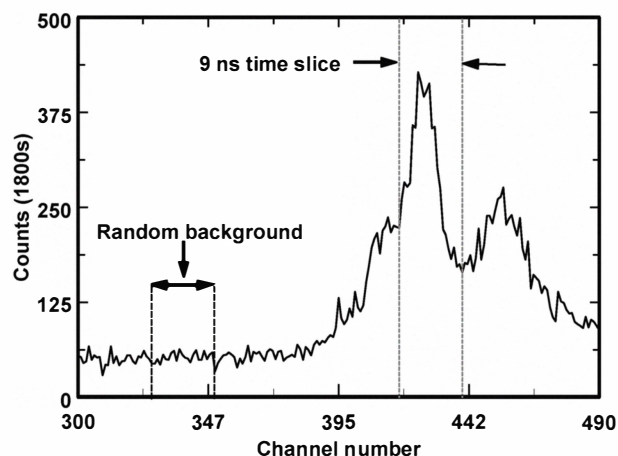


Fig. 2. Time distribution of α - γ coincidences recorded for an object, 27 cm from the neutron generator target. The peak at \sim channel number 455 to the right of the main peak is due to time correlated neutrons being scattered into the γ -ray detector.

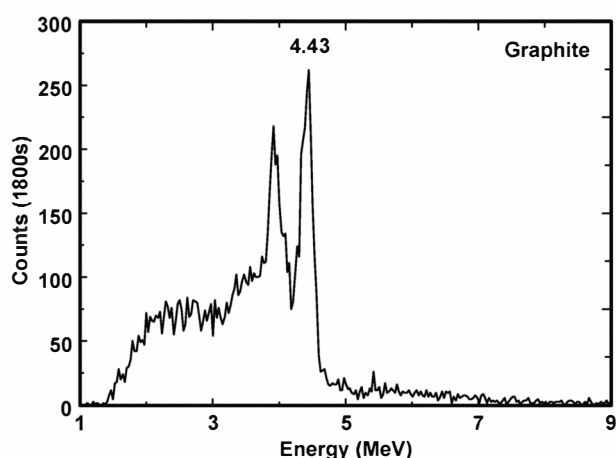


Fig. 3. Time correlated γ -ray spectrum of carbon, from the graphite located 27 cm from the neutron generator target.

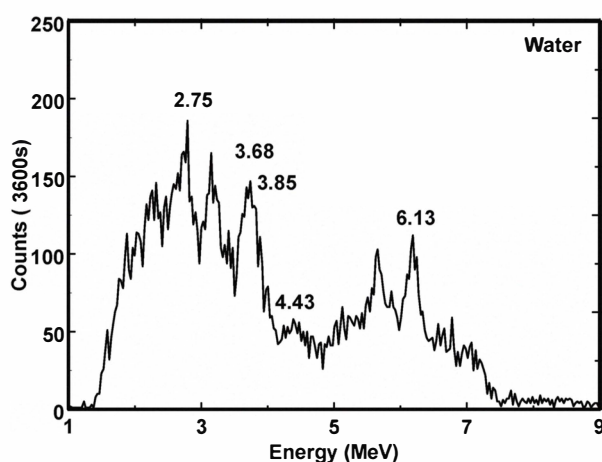


Fig. 4. Time correlated γ -ray spectrum of oxygen from a 1 gallon water bottle located 27 cm from the neutron generator target.

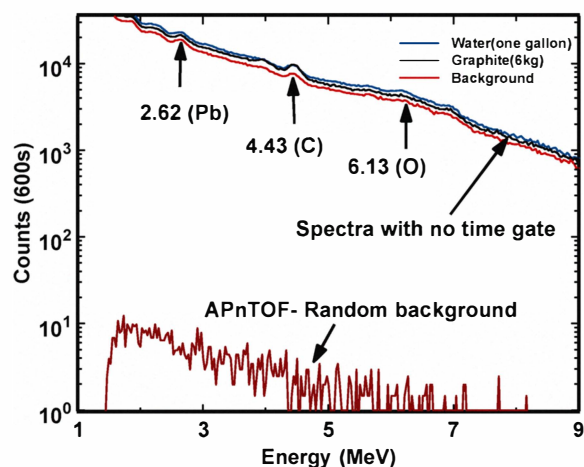


Fig. 5. γ -ray spectra of graphite, water and background (with no object in the beam) recorded without time restrictions on the data acquisition. The APnTOF-Random background was obtained with a 9 ns time slice from the region of the time spectrum shown in Fig. 2.

B. Clutter conditions

The time spectra of the bottle of water and the graphite when positioned in isolation at their respective locations are shown in Fig. 6. The time difference between the two peaks corresponds to the neutron flight time between the centers of the two objects. However, there is a considerable overlap of the two time spectra because of the large dimensions of the two objects. Fig. 7 shows the composite time spectrum when the bottle of water was placed between the neutron generator and the graphite sample in an overlapping mode. The spectrum shows the two peaks due to the objects at the same respective channel numbers as in Fig. 6. However, the intensity of the graphite peak is significantly diminished as expected, because the water bottle removes useful neutrons from the beam mainly via elastic scattering reactions with the hydrogen of water. Based on the time spectra of the two objects in isolation, a suitable time slice from a region that had minimum overlap between the objects, was set for the graphite ("UXO"), (see Figs. 6 and 7) and its time correlated γ -ray spectrum was extracted for the three cases. Fig. 8 shows the "inelastic" γ -rays of energy 4.43 MeV and its escape peak due to carbon from the graphite ("UXO") for the different clutter conditions. It can be seen that the oxygen signals from water are eliminated in all the three cases: Case 1(non-overlapping mode), the water bottle was outside the defined cone, Cases 2 and 3(overlapping mode), introduced the water bottle and iron slab progressively to the clutter in the neutron cone. There was a significant drop ($\sim 50\%$), in the 4.43 MeV peak intensity because the water attenuated the tagged neutron beam. When the iron slab was additionally introduced in the beam, a further reduction of $\sim 10\%$ in the intensity resulted. The lower energy signals below 3 MeV for cases 2 and 3 are a result of additional time correlated neutron scattering effects because of the near-by overlapping clutter objects.

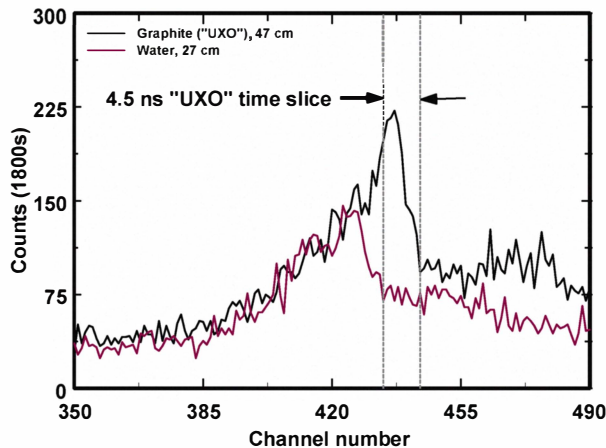


Fig. 6. Time spectra of objects present in isolation in the defined neutron cone. The water bottle and graphite were at a distance of 27 and 47 cm respectively, from the neutron generator target.

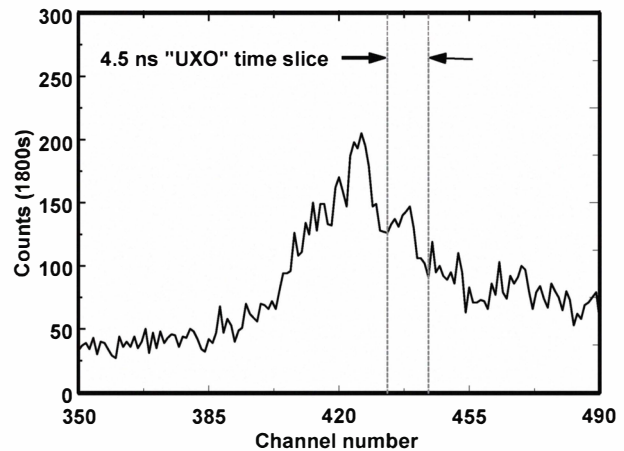


Fig. 7. Time spectrum acquired for the overlapping-clutter case where the bottle of water was interposed between the neutron generator and the graphite sample in the neutron cone.

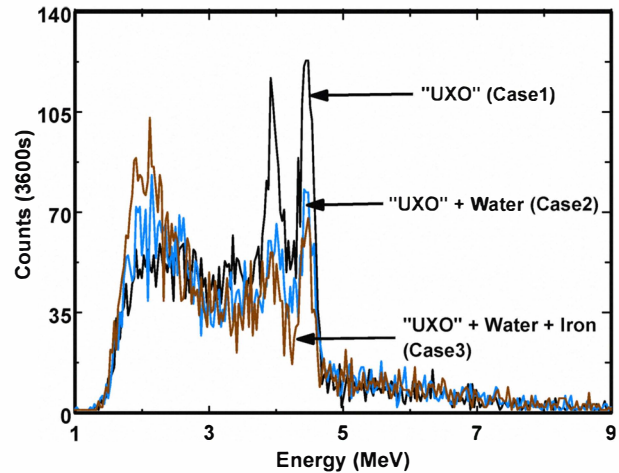


Fig. 8. Time correlated γ -ray spectra of graphite "UXO" under non-overlapping (Case1) and overlapping (Case2 and 3) clutter conditions using a 4.5 ns time window.

IV. CONCLUSIONS

In contrast to conventional neutron interrogation methods, the results from this study clearly demonstrated the efficacy of the neutron time-of flight technique for obtaining γ -ray signals from an object-of-interest with (1) high SNR and (2) without interference from signals of near-by clutter. This makes it ideal for the technique to be deployed in the field.

REFERENCES

- [1] P.C.Womble, F.J. Schultz and G. Vourvopoulos, "Non-destructive characterization using pulsed fast-thermal neutrons," *Nucl. Instr. and Methods in Phys. Research, B*, vol. 99, no.1- 4, pp.757-760, May 1995.
- [2] A. Buffler, F.D. Brooks, M.S. Allie, K. Bharuth-Ram and M.R. Nchodu, "Materials classification by fast neutron scattering," *Nucl. Instr. and Methods in Phys. Research, B*, vol. 173, no.4, pp 483-502, Feb. 2001.
- [3] P.C. Womble, Phase 1 demonstration report summary for: PELAN-A transportable neutron-based UXO Identification probe, "Environmental Security Technology Certification Program (ESTCP: 200106), Feb. 13, 2004.

- [4] S. Steward and D. Forsht, "Use of nuclear techniques to determine the fill of found unexploded ordnance," *Appl. Radiat. Isot.*, vol. 63, no. 5/6, pp. 795-797, Nov-Dec. 2005.
- [5] S. Mitra, J.E. Wolff and R. Garrett, "Calibration of a prototype in vivo total body composition analyzer using 14 MeV neutron activation and the associated particle technique," *Appl. Radiat. Isot.*, vol. 49, no. 5/6, pp. 537-539, May-June, 1998.
- [6] D.L. Chichester, M. Lemchak, J.D. Simpson, "The API 120: A portable neutron generator for the associated particle technique," *Nucl. Instr. and Methods in Phys. Research, B*, vol. 241, no. 1/4, pp. 753-758, Dec. 2005.
- [7] S. Pesente, G. Nebbia, M. Lunardon, G. Viesti, S. Blagus, K. Nad, D. Sudac, V. Valkovic, I. Lefesvre, M.J. Lopez-Jimenez, "Tagged neutron inspection system (TNIS) based on portable sealed generators," *Nucl. Instr. and Methods in Phys. Research, B*, vol. 241, no. 1/4, pp. 743-747, Dec. 2005.
- [8] C. Carasco, B. Perot, S. Bernard, A. Mariani, J.-L. Szabo, G. Sannie, Th. Roll, V. Valkovic, D. Sudac, G. Viesti, M. Lunardon, C. Bottosso, D. Fabris, G. Nebbia, S. Pesente, S. Moretto, A. Zenoni, A. Donzella, M. Moszynski, M. Gierlik, T. Batsch, D. Wolski, W. Klamra, P. Le Tourneur, M. Lhuissier, A. Colonna, C. Tintori, P. Peerani, V. Sequeira and M. Salvato, "In-field tests of the EURITRACK tagged neutron inspection system," *Nucl. Instr. and Methods in Phys. Research, B*, vol. 588, no. 3, pp. 397-405, April 2008.