

FINAL REPORT

Neutron Spectrometry for Identification of Filler Material in UXO

SERDP Project MM-1386

DECEMBER 2007

Mary Bliss
Pacific Northwest National Laboratory



Strategic Environmental Research and
Development Program

Report Documentation Page		Form Approved OMB No. 0704-0188
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1. REPORT DATE DEC 2007	2. REPORT TYPE	3. DATES COVERED 00-00-2007 to 00-00-2007
4. TITLE AND SUBTITLE Neutron Spectrometry for Identification of Filler Material in UXO		5a. CONTRACT NUMBER
		5b. GRANT NUMBER
		5c. PROGRAM ELEMENT NUMBER
6. AUTHOR(S)	5d. PROJECT NUMBER	
	5e. TASK NUMBER	
	5f. WORK UNIT NUMBER	
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) Pacific Northwest National Laboratory, 902 Battelle Boulevard, Richland, WA, 99352		8. PERFORMING ORGANIZATION REPORT NUMBER
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)		10. SPONSOR/MONITOR'S ACRONYM(S)
		11. SPONSOR/MONITOR'S REPORT NUMBER(S)
12. DISTRIBUTION/AVAILABILITY STATEMENT Approved for public release; distribution unlimited		
13. SUPPLEMENTARY NOTES		

14. ABSTRACT

Background: Unexploded ordnance (UXO)-contaminated sites often include ordnance filled with inert substances that were used in dummy rounds. During UXO surveys, it is difficult to determine whether ordnance is filled with explosives or inert material (e.g., concrete, Plaster of Paris, wax, etc.) or whether it is empty. Without verification of the filler material, handling procedures often necessitate that the object be destroyed in place, which has potential impacts to the environment, personnel, communities and survey costs. The Department of Defense (DoD) needs a reliable, timely, non-intrusive and cost-effective way to identify filler material before a removal action. A new technology that serves this purpose would minimize environmental impacts, personnel safety risks and removal costs; and, thus, would be especially beneficial to remediation activities.

Objective: Through this project, PNNL attempted demonstrate that a portable neutron spectrometer utilizing scintillating fiber detectors is capable of distinguishing between inert and explosive filler material in UXO. Data generated from the project is presented in this paper showing differentiation between inert fillers in controlled laboratory conditions using this technology.

Summary of Process/Technology: PNNL proposes a neutron spectrometry approach to the identification of filler material. The spectrometer utilizes a novel neutron detector developed at PNNL. The detector greatly improves on the portability and speed of current designs, offering much smaller portable packages with quicker analysis times. Neutron spectrometry can indicate the relative hydrogen carbon, nitrogen and oxygen ratios in dense containers remotely and non-destructively. The technology is similar to devices used commercially to determine soil moisture, characterize oil wells and measure asphalt quality. A neutron source interacts with the target object, some neutrons are backscattered into the detector, and their energies inferred. The differences in the incident and backscattered neutron energies are indicative of the relative hydrogen, carbon, nitrogen and oxygen contents of the target. These data can then be used to differentiate between inert filler materials and explosives. It is not necessary to fully determine the neutron energy spectra, or deconvolve the instrument response, in order to determine if given targets match certain criteria related to chemical ratios. A statistical analysis of the raw data is adequate to sort targets into inert and explosive categories.

Transition Plan: The filler identification system developed in this project will be broadly applicable to many installations and sites that are currently contaminated by UXO. PNNL recognizes the need to successfully develop, customize, deploy, and commercialize PNNL's neutron spectrometer technology to meet U.S. industry and government needs. Toward this end, PNNL already has licensed the core

15. SUBJECT TERMS

16. SECURITY CLASSIFICATION OF:

a. REPORT
unclassified

b. ABSTRACT
unclassified

c. THIS PAGE
unclassified

17. LIMITATION OF
ABSTRACT

**Same as
Report (SAR)**

18. NUMBER
OF PAGES

14

19a. NAME OF
RESPONSIBLE PERSON

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Contents

1. Abstract	3
2. Introduction	4
2.1 Background	5
3. Experimental	7
4. Results and Analysis	9
5. Summary	9
6. Conclusions and Future Work	12
7. References	12

1. Abstract

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2. Introduction

The goal of this work is to demonstrate the efficacy of using neutron spectrometry to determine the filler of UXO prior to excavation. A novel PNNL-developed detector is key to implementing this approach in a practical manner. The detector output is handled as a “fingerprint” of the environment combined with the target material. A training set of select target materials is required to develop a decision matrix. The fingerprinting approach was first demonstrated with neutrons in a blind test for classifying nuclear weapon “pits” (Ref 1). In this blind test, a statistical templating method was used to sort pits by design number. The intended application was warhead dismantlement. The spectroscopic fingerprinting approach provided clear discrimination between all types of pits examined, including pits that are difficult to distinguish by other means. Experiments with this instrument provided unequivocal discrimination between a variety of neutron and gamma ray sources (Ref 2). The extension of this methodology into the discrimination of UXO filler material is sound and supported by the work of others (Ref 3).

Simplistic versions of neutron backscatter spectroscopy are currently used in oil and gas reservoir assessment, soil moisture measurement, asphalt quality measurements, and measurements of other geotechnical properties. Typically the neutron energies are divided into only two categories: thermal and epithermal. The PNNL approach has up to six energy bins and could have more if needed. The two energy configurations PNNL developed are robust and accurate with a proven deployment record in the commercial sector.

The PNNL neutron spectrometer consists of six layers of scintillating fiber thermal neutron detector (Refs. 4,5,6) embedded in polyethylene. The detection depth in the polyethylene stack is related to the incident neutron momentum. Traditional Bonner Spheres use the same approach for neutron energy measurements one energy region at a time (Ref 7). In the PNNL detector, all six fiber layers count simultaneously, greatly reducing data collection time compared with that of Bonner Spheres. The hardware is also much more compact. The fiber layers are positioned to analyze 0.5 to 10 MeV neutrons with an effective energy resolution of 0.5 MeV. The present data output is a six-by-sixteen numerical matrix indicating the number and brightness of events detected in each fiber layer. Event brightness is used to distinguish neutron interactions from gamma rays (and/or beta particles). All spectral information is obtained from the fiber layer depth within the polyethylene. The data matrix collected by the spectrometer is the convolution of the radioactive (neutron) source, the environment, and the detector itself. Holding two of these variables constant yields the third variable.

In this particular application, the goal is to determine if the environment near the detector contains explosives above some minimal quantity. It is not necessary to deconvolve the neutron energy spectra or chemical information from the data if a strong statistical correlation can be made to a reference data set. This template-matching approach was used to sort “pits” and other known sources (Refs 1, 2). This approach worked extremely well when the innermost fiber layers were used to remove fluence (source strength) as a variable. The only differences remaining in the data set were related to the neutron (and to a much lesser extent, the gamma ray) energy distribution. Others have shown theoretically and experimentally that the perturbation of an incident neutron energy spectrum by a target is determined by the chemical composition of the target. This is especially true for light elements (Ref. 3, 8, 9, 10, 11, 12, 13, and 14). A source of high-energy neutrons (PuBe or AmBe for example) is required to ensure that an adequate number of high-energy neutrons impinge on the detector. These high-energy neutrons (0.1-10 MeV) will carry the most information about the target. Neutrons that lose a very large amount of energy



The PNNL neutron spectrometer consists of a detector module and laptop computer.

via hydrogen (moisture) interactions in the environment are detected in only the shallowest fiber layers and carry little target information. These can be filtered out easily via detector shielding, as necessary.

The reliable discrimination/template-matching of signatures from the PNNL neutron spectrometer requires a rich multi-channel representation of the neutron source spectrum. These measurements are subjected to a template-matching method to categorize the source.

2.1 Background

The proposed technique is **fundamentally different from** other neutron-based methods that have been explored for UXO detection or identification. These techniques (PELAN [Ref 15], PINS [Ref 16], neutron activation [Ref 17], PFNA [Ref 18], etc.) rely upon detection of the gamma rays that are produced when neutrons interact with nuclei in the test object. The *only* similarity between our approach and these other approaches is the use of a neutron source. Neutron spectrometry will provide analytical information similar to gamma ray-based methods, with the beneficial improvements summarized below:

- ❑ **Efficient source strength:** The source required for neutron spectroscopy can be three to five orders of magnitude weaker than that used for activation approaches (PINS, etc.) and is consequently of lesser operational and safety concern. Neutron scattering is very efficient compared to the reactions that induce gamma rays in a target or specimen.
- ❑ **Cost-effective detectors:** The proposed approach uses thermal neutron detectors, which are relatively inexpensive and efficient when compared to the high-resolution gamma ray detectors required for activation approaches. High-resolution gamma ray detectors typically require cryogenic cooling, a high degree of operator skill, and high maintenance.
- ❑ **Practical physics:** The production of gamma rays with a specific energy through neutron interaction is a very inefficient process. The moderation of neutrons is, by contrast, unavoidable and very efficient, requiring low power and simple detection. Gamma rays are also attenuated more rapidly than neutrons in soil, limiting the depth at which UXO can be analyzed relative to neutron spectroscopy.
- ❑ **Faster analysis:** Only about 100,000 neutrons need to be collected by the spectrometer to reach an acceptable confidence level for decision-making.

Neutron spectrometry, as a general class of method, **is already proven to detect explosives** (Refs 3,7,9,10,11,12,13,14). However, the applications suggested in the past primarily have been **based on time-of-flight analysis**. In these approaches, an accelerator-based neutron source directs pulses of neutrons at an object. The neutrons interact as they **pass through the target material(s)**. The neutrons are scattered and slowed differently depending on which type of atomic nucleus they encounter (nitrogen, hydrogen, oxygen, carbon, etc...). As a result, neutrons that left the source at the same time arrive at the detector at different times, indicating which elements they have encountered along the way. In this manner, a chemical signature of the target is constructed by detecting the relative populations of neutrons arriving at the detector in given time frames. The National Academy of Sciences identified two major issues with time-of-flight spectroscopy for luggage interrogation (Ref. 14). The first was relative to Class A explosives (initiators/detonators) that could not be differentiated from other objects in luggage with any degree of accuracy. The second was time-of-flight spectroscopy that requires a large neutron accelerator and a long path length (ca. 4 m).

The PNNL method utilizes the principle of moderating neutron spectroscopy. The detector response is a function of the distribution of neutron energies. Neutrons pass into the object, interact with the contents, and are backscattered into the detector. The detector analyzes the energies of the returned neutrons and enables more accurate elemental determination.

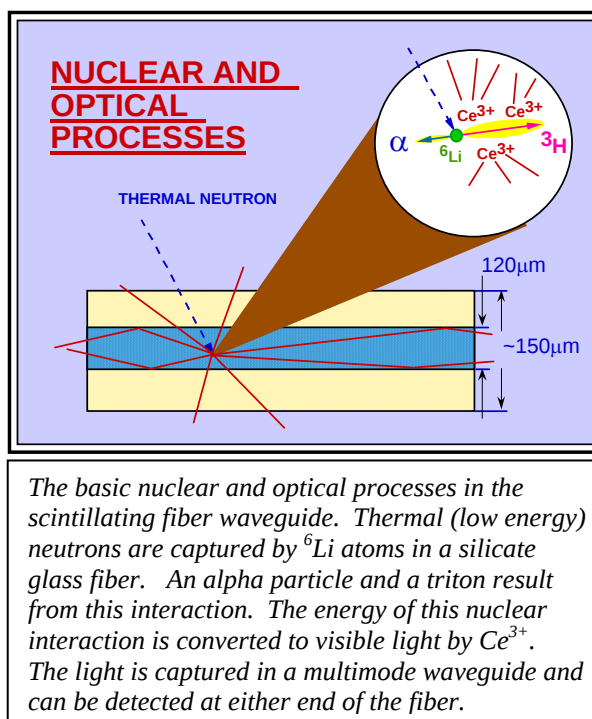
The detection of Class A explosives is a problem for luggage screening, however, for the UXO application it is not an issue because we are primarily differentiating between known inert fillers, propellants, and explosives. If a shell shows up as a false positive (unlikely given inert filler compositions), it is treated as any other explosive-packed shell. The analysis method will err on the side of caution, having a finite false positive rate and a negligible false negative rate.

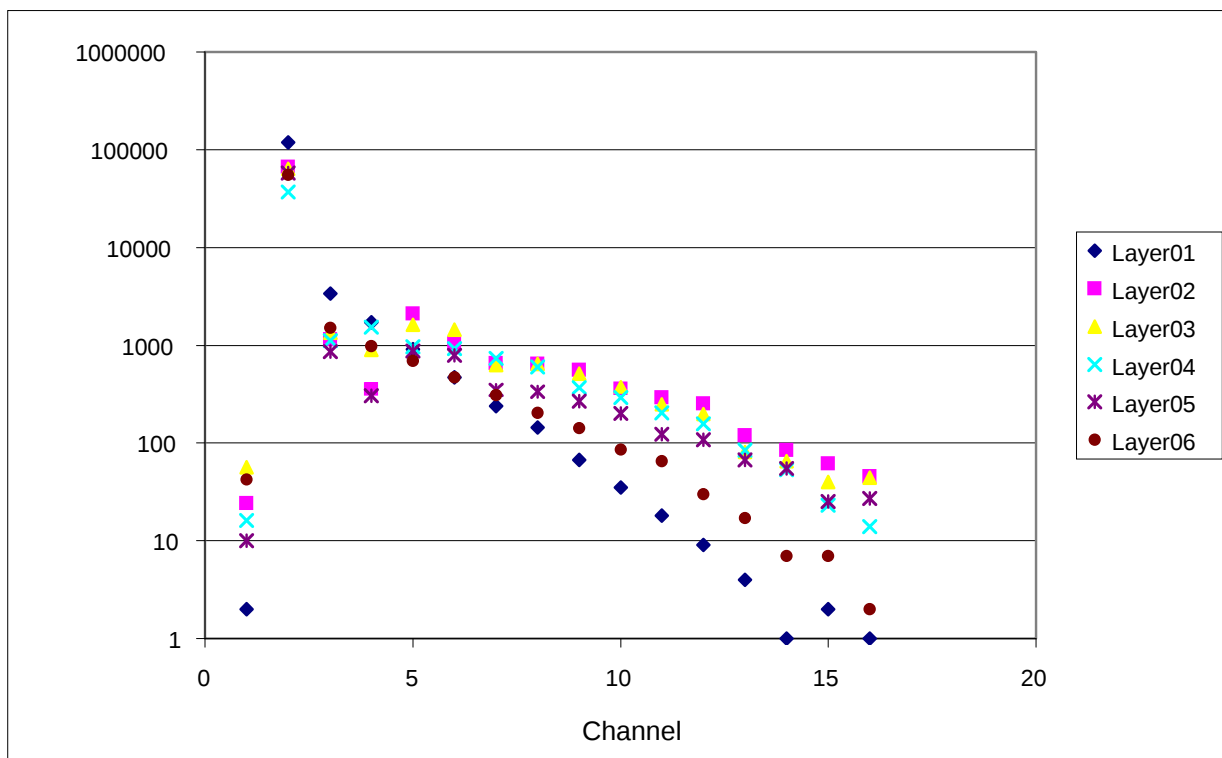
The second issue, as it relates to explosives detection, has to do with the sensitivity of the detection method. Time-of-flight analysis, as used in previous attempts, takes a huge number of neutrons and/or a very long analysis time to identify the contents of an object. In the case of the FAA tests (Ref 14) a very large accelerator was needed, which is prohibitive for available space at an airport. In the case of the University of Oregon tests (Ref 3) a smaller source was used but detection times were prohibitive.

PNNL's approach solves the second issue of sensitivity in two ways. First, the detector measures scattered neutron energy instead of time-of-flight. Second, detector efficiency is extremely high. The detector consists of multiple layers of lithium-rich scintillating fibers sandwiched between sheets of polyethylene that act as a neutron moderator. As scattered neutrons pass through the moderator they slow sufficiently to produce scintillation photons in the glass by a ${}^6\text{Li}(n,\alpha){}^3\text{H}$ reaction. This reaction is exothermic, releasing approximately 4.7 MeV,

of which the majority is carried away by the triton. The triton and alpha particle each interact with the glass matrix to produce an ionization trail. This ionization transfers energy by exciting Ce^{3+} ions, which scintillate with the emission of photons of wavelength ~ 400 nm. The light is transmitted out of the moderator block to the end of the fiber, where photomultiplier tubes produce an electrical signal. The fibers will emit 90% of the light created by a detection event in 200 ns. Depending on the count rate and background conditions the light collection window is followed by a dead time to allow the light signal to dissipate so it is not mistaken for a new detection event. Neutrons and gamma rays produce signals that can be differentiated by brightness such that only one in 10,000 neutron events are actually caused by bright gamma ray events. Photomultipliers are connected to both ends of the fibers. A noise filter or coincidence circuit requires light detection at both ends of the fiber to record a detection event.

The layer where the neutrons are detected provides information about the scattered neutrons' energy, which is analyzed to identify what interactions the neutron has seen and, hence, the elemental make up of the object from which the neutron was backscattered. This device is one of the few solid-state detectors in a world of pressurized gas detectors. Solid-state detectors are more rugged and transportable for field operations.





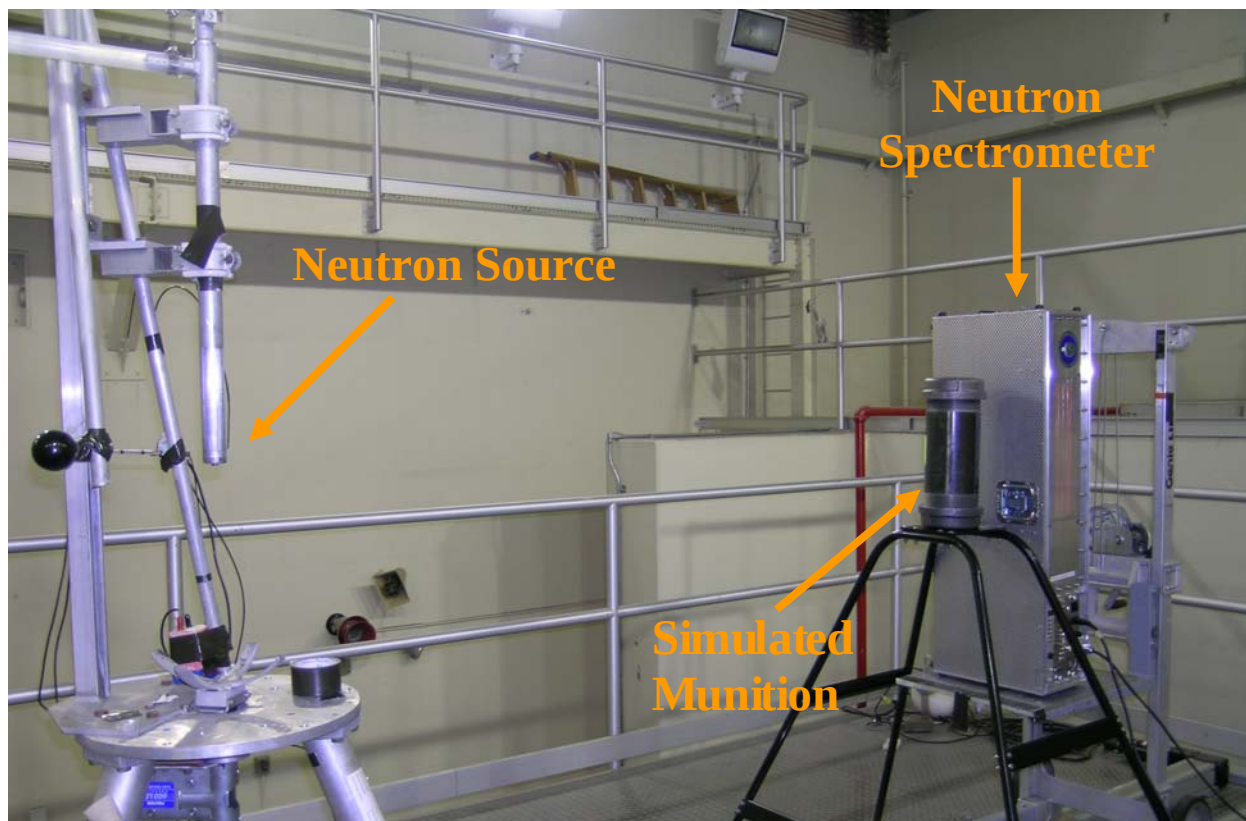
Graphical data set from the Neutron Spectrometer: coincidence is on and the source is ^{252}Cf . Events with brightness greater than channel 4 are generated by neutron capture.

PNNL's neutron detector has significant advantages over similar designs that use pressurized gas tubes (Ref. 19, 20). Using fiber dramatically reduces the size of detector needed to achieve equivalent results. ^3He detectors are ½- to 1-in.-diameter tubes filled with ^3He at two- to ten-atmosphere pressure, whereas, the fibers form a layer only 200 μm thick. A layer of tubes imbedded in an appropriate moderating material is much less effective at neutron detection than the fibers. Hence, the PNNL detector is much smaller and lighter for the same relative efficiency. Additionally, the fibers can be precisely located in space, which allows a corresponding precision in energy resolution calculations.

3. Experimental

Schedule 40 steel pipe was selected to mimic the munitions shell casings. After consulting several experts in munitions identification (A. Caffrey and G. Vourvopoulos), it was realized that separating inert fillers from each other is the more challenging problem for neutron based techniques. If the neutron spectroscopy approach could be used to separate inert filler signatures then determining explosive from inert fillers would be simple. The inert fillers selected were: Plaster of Paris, polyethylene beads, dry sand and air. These materials represent widely varying hydrogen contents and overall chemistry. Because neutrons and hydrogen atoms have essentially the same mass their interaction is key to testing the sensitivity of this technique. A statistical testing matrix was designed. Measurements were started in the Low Scatter Room in the PNNL NVLAP Accredited Calibration Laboratory for Ionizing Radiation. NIST traceable ^{252}Cf sources were used. Measurements were made in transmission mode. The simulated munitions were placed between the source and the detector. The Low Scatter Room is designed to

minimize neutron scattering by the environment. The source and detector are on an aluminum platform in the center of a large, empty room with neutron absorbing walls (concrete).



The hardware was moved in and out of the Low Scatter Room for one-day or half-day data collection runs. Early in the data collection process it became apparent that the detector was not functioning well. The distance between the detector and source was maximized to lower the count rate and runs were compared day to day. The detector was stable for a given run but changed between each set-up. The cooling fan was replaced and the hardware was allowed to warm up overnight. This helped somewhat but the age of the electronics (between 5 and 10 years) was suspect. Since revamping the electronics was not part of the work scope or achievable within the budget, detector stability was added to the statistical analysis model. After several data collection campaigns it became apparent that entire layers of the detector were disabled when coincidence was required between the photomultiplier tubes at either end of the fiber layers. Generally, coincidence is used to reduce noise without significantly reducing sensitivity. This could be caused by failure of individual photomultiplier tubes, broken wires or the coincidence logic circuit itself. The coincidence requirement could be turned off via the data collection software. This increases the electrical noise content of the data especially in the lower channel numbers where gamma ray events dominate. Data could be collected and the statistical analysis applied even if the absolute count numbers from a given layer were affected by the electronics problem, since it was a systematic error. Turning off the coincidence requirement yielded detection events in every layer of the detector but it is not clear which layers had only one operable photomultiplier tube. The statistical analysis focused on the higher channel numbers (greater than channel four) to ensure that neutron events were dominant.

The primary data collection configuration was to have the distance between the source and detector set at 2 m and the centerline of the simulated munitions approximately 15 cm from the detector face. The

largest pipe was 13 cm diameter and 30.5 cm high. The filler materials occupied the entire inner volume of the pipe. The filler mass was as follows: Plaster of Paris - 5 kg, sand – 5.7 kg, and polyethylene beads – 2.3 kg. The top and bottom end caps screwed on and held the contents in place. The filler was changed for each sequential run to ensure statistical independence. The largest data set was seven measurements on each of the filler types. Data collection consisted of a background measurement (no source), then source measurement, followed by another background measurement. A second pipe 15 cm tall was also used to determine filler mass effects. Data sets were collected for two minutes each. The detector settings were without coincidence between the photomultiplier tubes for each layer (electronic stability problems were less severe without coincidence), with 50 ns allowed for event detection followed by 600 ns of dead time to separate detection events.

4. Results and Analysis

The measurement for each combination of layer and munition configuration was the total energy in bins 5 through 16, e.g., Channel 5 Channel 6+...+ Channel 16. A two-way analysis of variance (ANOVA) was performed on these data to determine if a significant difference was observed between the munition configurations. If the polyethylene beads configuration is statistically different than all other configurations, then the neutron spectrometer has the potential as a detection system for separating explosive from dummy rounds. The data set was comprised of seven independent measurements on the three inert fillers, the empty 30 cm tall pipe and no object in front of the detector (5 configurations). Electronics problems prevented the reporting of the actual number of neutrons detected in each layer - some layers had two working photomultiplier tubes and some only one. This affects the amount of noise filtering in the raw data and the sensitivity to gamma rays in the lower channels (1-4). However, the system was stable across all experimental runs. The sum of channels 5-16 were assigned to “neutrons”.

Table 1 shows the results of the statistical analysis using two-way ANOVA. The significant interaction between Layer and Treatment (configuration) indicates that degrees of differences between Treatments are dependent on Layer. A multiple comparison test was applied to determine which Treatments are different from others across the Layers. A Turkey’s studentized range test was used to develop Table 2. Treatments that are not statistically different appear grouped under the same color. Fiber layers 1 and 2 are not useful in distinguishing between the inert fillers (grouped under the same color in Table 2). For Layers 3 through 6, the data has very little scatter (plots after Table 2) and the spectrometer is able to distinguish between Treatments when the deeper fiber layers (3-6) are used. The Treatment differences as a function of Layer are provided as box plots in the next section.

5. Summary

Although the electrical problems with the detector made the absolute detector response unreliable (the data could not be used to deconvolve the source neutron energy spectra with a fixed instrument function); these did not prevent the differentiation of the various inert fillers from each other. This supports the existing physics literature on the validity of the technique in transmission. Efforts are underway with another funding source to upgrade the electronics and develop an energy deconvolution algorithm for the prototype hardware.

Table 1. Two-way ANOVA Table

Table shows significant interaction between Treatment and Layer. This indicates spectrometer ability to differentiate between Treatments is dependent on Layer

Change Dependant Variable: PE05+... to Dependant Variable: Channel 5+...

ANOVA Table					
Dependent Variable: PE05+PE06+É+PE16					
Source	df	Sum of Squares	Mean Square	F Value	Pr>F
Model	29	27807389879	958875513	43298	<0.0001
Error	180	3986275	22146		
Corrected Total	209	27811376154			
	R-Square	Coeff. Var.	Root MSE	Count Mean	
	0.999857	1.16916	148.8152	12728.39	
Source	df	ANOVA SS	Mean Square	F Value	Pr>F
Treatment	4	932455987	233113997	10526.2	<0.0001
Layer	5	26083187330	5216637466	235557	<0.0001
Treatment*Layer	20	791746562	39587328	1787.56	<0.0001

Table 2. Summary Statistics by Layer and Treatment

Colors are used to show statistically equivalent Treatments across all fiber layers and within a fiber layer.

Layer 1								
Treatment	N	Mean	StdDev	Minimum	25th Pctl.	Median	75th Pctl.	Maximum
Air	7	85.43	14.89	55	81	88	93	102
None	7	83.00	20.86	56	62	90	99	108
Plaster	7	55.29	14.84	39	40	51	72	77
Sand	7	54.00	18.37	27	42	54	65	84
Poly	7	48.86	14.21	28	39	48	62	69

Layer 4								
Treatment	N	Mean	StdDev	Minimum	25th Pctl.	Median	75th Pctl.	Maximum
None	7	42531.86	287.8225	41938	42459	42586	42762	42782
Air	7	36187.43	181.3255	35954	36009	36161	36345	36474
Sand	7	30168.57	189.1735	29816	30068	30203	30304	30409
Plaster	7	29228	141.1477	29032	29109	29265	29363	29408
Poly	7	27536.57	258.2498	27142	27379	27530	27687	27974

Layer 2								
Treatment	N	Mean	StdDev	Minimum	25th Pctl.	Median	75th Pctl.	Maximum
None	7	3290	106.19	3180	3194	3267	3365	3488
Air	7	2871	124.964	2720	2803	2844	2887	3124
Sand	7	2271	81.4409	2160	2182	2270	2358	2380
Plaster	7	2240	34.4211	2193	2211	2239	2282	2283
Poly	7	2133	88.0792	1943	2122	2175	2186	2194

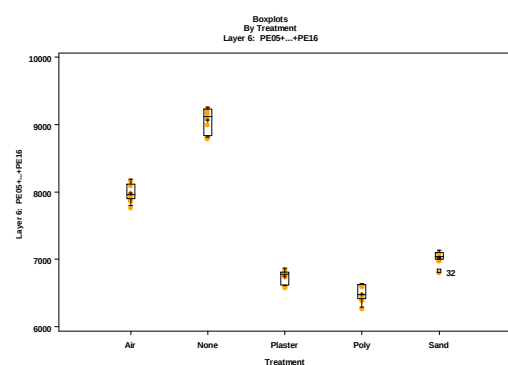
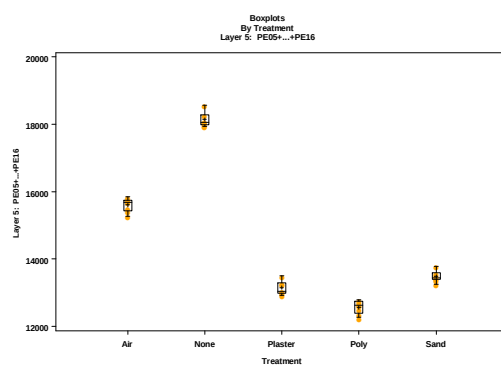
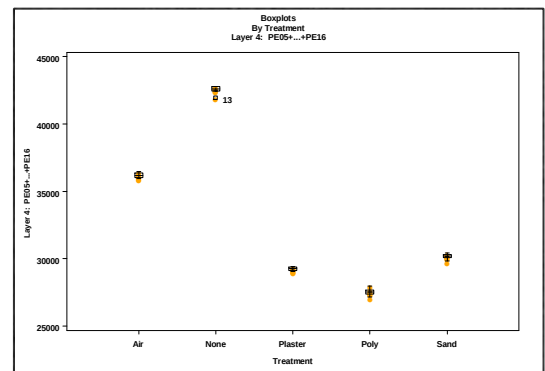
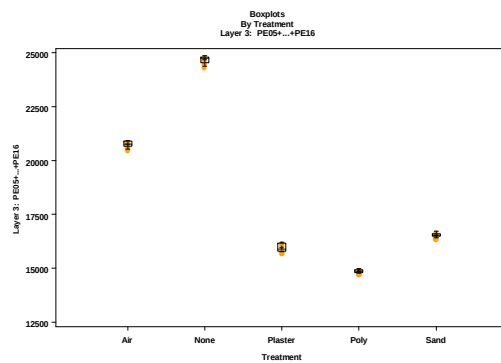
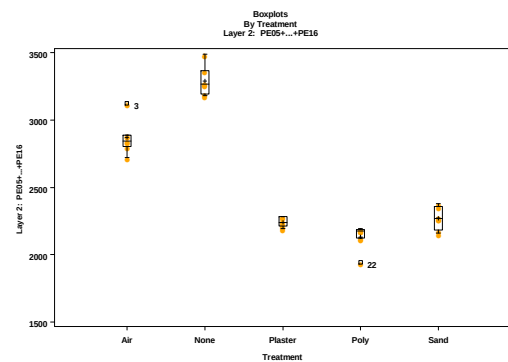
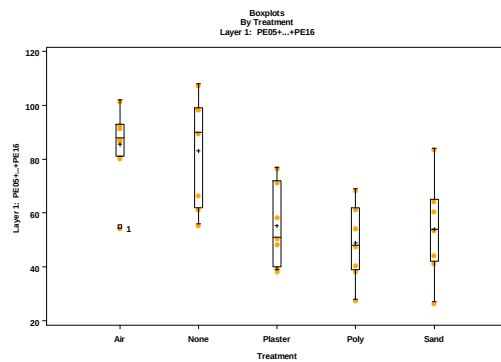
Layer 5								
Treatment	N	Mean	StdDev	Minimum	25th Pctl.	Median	75th Pctl.	Maximum
None	7	18142.86	215.0794	17938	17983	18063	18286	18561
Air	7	15601	206.0801	15269	15425	15683	15747	15840
Sand	7	13486	166.1134	13253	13391	13443	13584	13781
Plaster	7	13147.14	207.2803	12922	12981	13045	13293	13502
Poly	7	12561.71	190.5428	12260	12379	12628	12737	12788

Layer 3								
Treatment	N	Mean	StdDev	Minimum	25th Pctl.	Median	75th Pctl.	Maximum
None	7	24679	167.146	24382	24541	24752	24796	24852
Air	7	20766	148.658	20534	20661	20754	20900	20928
Sand	7	16545	100.343	16417	16480	16527	16613	16729
Plaster	7	15964	190.031	15767	15796	15880	16169	16215
Poly	7	14869	72.7249	14777	14788	14857	14937	14973

Layer 6								
Treatment	N	Mean	StdDev	Minimum	25th Pctl.	Median	75th Pctl.	Maximum
None	7	9066.714	181.5	8815	8839	9117	9227	9258
Air	7	7982.571	135.4398	7799	7897	7960	8115	8193
Sand	7	7024	96.33968	6831	7004	7042	7095	7132
Plaster	7	6749.857	99.67519	6607	6617	6776	6809	6868
Poly	7	6482.571	121.5303	6291	6416	6474	6621	6640

BOX PLOTS OF REPICATE DATA FOR TREATMENTS BY FIBER LAYER

CHANGE PE TO CHANNEL IN PLOTS



6. Conclusions and Future Work

This effort supports the work of others showing that alteration of neutron momentum spectra includes information about the chemical content of the intervening materials. A high-efficiency moderating detector reduces data collection times to a few minutes, or less, and enables utilization of smaller neutron sources than previous efforts. Three inert fillers (plus an empty container) were separated by analysis of the detector response without regard to container effects. (Separating inert fillers from each other is more challenging than separating ordinance from inert fillers.) Technical difficulties prevented measurements to determine the minimum mass of ordinance filler and measurements in other geometries. Currently another U.S. government client is supporting fabrication of a new electronics package and development of a physics model of this type of detector for use in nuclear safeguards.

7. References

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