A 3-Dimensional Method for In-Situ Characterization of Buried Transuranic Waste using a Large Area Neutron Monitor

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ABSTRACT

A new passive neutron assay system has been designed for in-situ characterization of buried transuranic (TRU) waste. The system uses an array of high efficiency ^{3}He neutron detectors embedded in a polyethylene slab. In its standard mode of operation, the system will be deployed at a dig face prior to excavation as a means of controlling Material At Risk (MAR). Total neutron counting is used to quantify the plutonium content of the excavation zone. This technique offers considerable advantages over the traditional environmental survey methods that are based on detection of low energy gamma-rays and x-rays. Such techniques work well for characterization of surface-embedded particulates such as fall-out contamination. However, the short penetration depth of the photon radiation limits the investigation to the upper few centimeters of soil. Gamma based surveys can therefore only provide 2-dimensional information on the TRU contamination. Such an approach is often undesirable for historical waste burial sites where the distribution of spatial isotopes is highly heterogeneous. By detecting the far more penetrating neutrons emitted from (alpha,n) interactions and spontaneous fission, the total neutron counting method can provide an appropriate solution to this 3-dimensional problem. Monte Carlo N-Particle (MCNP) modeling has been performed to determine the optimum geometrical arrangement of polyethylene shielding / moderating materials and assess the performance limits of the neutron assay system. Various types of soils were modeled with a wide range of density and moisture content. This work has demonstrated that the counter is capable of accurately quantifying plutonium over an excavation zone of 3 m x 3 m and up to 1 meter below the surface. The typical lower limit of detection is less than 1 gram of Weapons Grade Pu in a 15 minute count.

INTRODUCTION

Table 1 gives the radiation penetration depth of plutonium and americium emissions for point sources embedded in dry soil at a density of 1.1 g/cc. The half-value layer is the thickness of soil required to reduce the intensity from a point source by a factor of 2. The final column gives the total effective penetration depth of the radiation (evaluated as the thickness of soil required to reduce the radiation to 95% of the original intensity). The final column is presented graphically in Figure 1. It can be seen that gamma rays will only penetrate a thin layer of top-soil, whereas fast neutrons can be measured at a depth of nearly 1 meter.

It is obvious that gamma-based techniques for the measurement of transuranic nuclides embedded in soil are only suitable for particles at or near the surface of a survey area. This effectively provides a survey team with only a 2-dimensional map of surface contamination distribution. By contrast, neutron-based assay systems can offer a 3-dimensional characterization solution enabling faster cleanup of buried waste by reducing the frequency of the survey measurement and allowing the retrieval operation to cut deeper into the dig face.

Table 1. Penetration depths of different radiation types in soil

Radiation	Energy (MeV)	Nuclide	Half Value Layer (cm)	95% Attenuation Thickness (cm)
Gamma	0.0596	^{241}Am	2.5	10.7
Gamma	0.129	^{239}Pu	4.3	18.3
Gamma	0.414	^{239}Pu	6.6	28.7
Neutron	~2	^{240}Pu	21.1	90.9

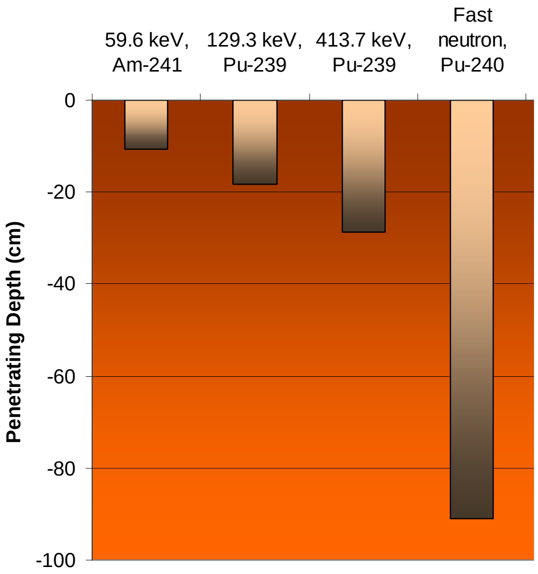


Figure 1. Radiation penetration depths in soil

DIG FACE MONITOR DESCRIPTION

A large area neutron monitor known as the “Dig Face Monitor (DFM)” has been developed to provide a high sensitivity platform for the detection of plutonium in soil using the total neutron counting technique. The system performance has been investigated with modeling and experimental measurements. A photograph of the system is shown in Figure 2. The system contains an array of 16 ^{3}He proportional counter tubes of 5.08 cm diameter and 182.8 cm length filled to a gas pressure of 2 atmospheres. The detectors are shielded by polythene, 10.16cm thick on top of each package, 12.07 cm thick on the exterior ends of the DFM. Detectors are protected against mechanical shock using neoprene gaskets evenly spaced along each neutron detector tube. The system has a total width of 234 cm and a length of 244 cm.

The DFM is designed to be presented close to the surface of soil and is primarily designed to alert the operator to potentially hazardous conditions by detection of plutonium within contaminated soil by measurement of neutron count rates. A calibration check stand is provided with the system. This allows the operators to verify the system performance on a daily basis, with a neutron emitting source located at a reproducible positions [1].

Figure 2. Photograph of the Dig Face Monitor

TECHNIQUE

The system relies on total neutron counting to detect and quantify the total plutonium present in the soil. This method assumes that the isotopics and chemical composition of the buried Pu are well known. At waste disposal sites where the isotopic distribution is highly variable, the standard approach is to calibrate the system based on the most likely isotopic / chemical form and allow for variations in the total measurement uncertainty term.

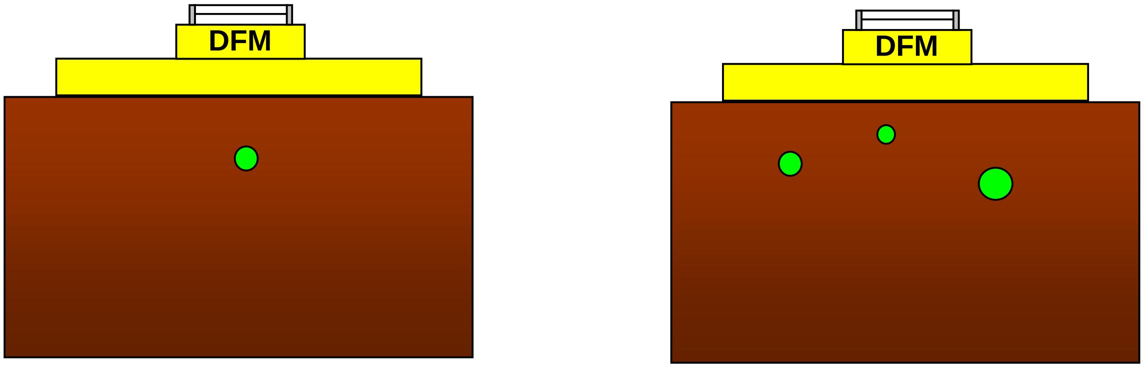
Buried plutonium would be mostly expected to be in the form of PuO_2 as the metal rapidly oxidizes in contact with air and large pieces of metallic plutonium would be highly unlikely to be have been consigned to a waste disposal site. However, fluorides, which emit up to an order of magnitude more (alpha,n) neutrons than oxides, could be present. In most cases the DFM would be calibrated with the assumption that the plutonium was in the oxide form, so that the presence of fluorides would result in an overestimate of plutonium content.

The system can be fitted with Geiger Müller detectors to provide a gross gamma alert for the operators. In addition, a NaI(Tl) detector may be used to provide low resolution spectroscopic capabilities. This allows the operators to screen for the presence of fission products such as ^{137}Cs or ^{60}Co and to detect the presence of plutonium fluoride from the characteristic 1274 keV peak emitted from ^{22}Na decay.

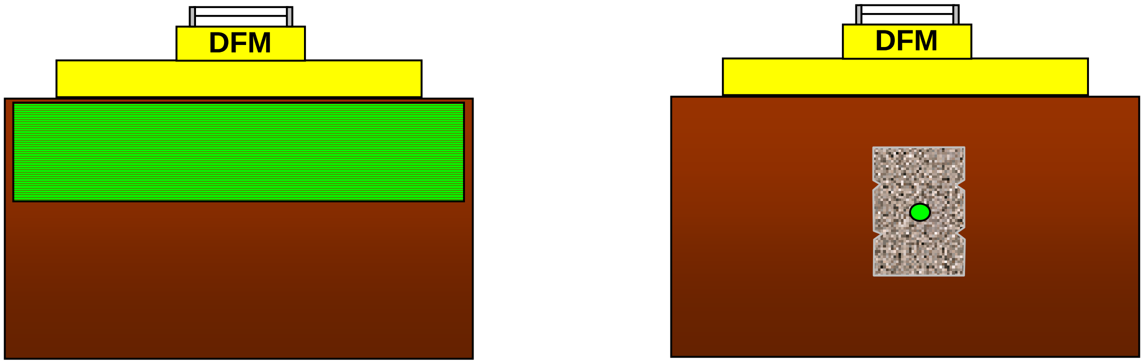
APPLICATIONS

The DFM is primarily designed for detection of buried plutonium. Various scenarios of soil / waste / plutonium distribution commonly encountered at waste burial sites are depicted in Figure 3.

The DFM may also be deployed as a moisture meter. In this mode, a standard neutron emitting source is inserted into the soil at a fixed depth in a guide tube. A simple quadratic function is used to relate moisture content to total neutron count rate. The moisture content is the primary neutron absorber due to hydrogen’s dual role in moderation and absorption of neutrons.



Scenario 1 : Single Pu particle in soil ***Scenario 2 : Multiple Pu particles in soil***



Scenario 3 : Homogeneous Pu distribution in soil ***Scenario 4 : Waste drum with Pu buried in soil***

Figure 3. Various measurement and detection scenarios for buried plutonium in soil

MCNP MODEL

The sensitivity of the DFM for the detection of plutonium at sub-surface positions in soil has been investigated using Monte Carlo N-Particle (MCNP) Modeling [2]. The following scenarios were modelled:

i. Plutonium point sources at various vertical depths from 10 to 400 cm located in soil with a water content of 6% wt., 10% wt. and 14.3% wt.. The sources were located below the DFM at a slight offset from the geometrical center (underneath detector #9).

ii. Plutonium point sources located at a vertical depth of 50 cm and offset from the center-line of the x-axis (defined as orthogonal to the cylindrical axis of the detector tubes) at various distances between 22 cm and 300 cm for water content of 10% wt.

iii. As (ii), but with the sources offset from the y-axis (defined as parallel to the cylindrical axis of the detector tubes).

The neutron detection efficiency was tallied for individual neutron detector at each source depth as well as a tally for the sum of all 16 neutron detectors. The efficiency tally provides the number of ^{3}He (n,p) reactions per source neutron. The plutonium was assumed to be in the form of a pure ^{240}Pu point spontaneous fission source, with the neutron emission characteristics of the Watt fission spectrum given in Equation (1),

$$P(E) = \exp\left(-\frac{E}{a}\right)\sinh(\sqrt{bE}) \qquad (1)$$

where E is the neutron energy in MeV, a = 0.799 MeV, b = 4.903 MeV^{-1}.

Notably, the results show that as the point source depth increases, there is an approximately exponential decrease in neutron detection efficiency. This is not particularly surprising given that the neutrons are subject to a greater probability of scatter as the depth of soil increases. It is also apparent that the rate of efficiency decrease varies with the detector in question, which would be expected given the longer effective path lengths that the neutrons require to reach the outer detectors.

No results have been presented for depths greater than 2.0 m. All models for depths of 3.0 m and 4.0 m (including application of variance reduction techniques) gave a null result indicating that either the models were computed for an insufficient period of time, or reflecting the fact that point sources at these depths are beyond the limit of detection for the dig face monitor.

Figure 4 clearly shows that with increasing water content and increasing depth, the neutron detection efficiency decreases. This behaviour is as would be expected, given that with increasing water content, the neutrons will tend to suffer from higher levels of neutron capture and thermalization resulting in a smaller flux reaching the neutron detectors from lower depths.

Figure 5 shows the neutron detection efficiency as a function of source offset position from the geometrical centers of the x- and y- axes respectively. The response is as would be expected intuitively, i.e. as the source position is moved longitudinally along the detectors, the overall efficiency profile will remain similar, but decrease in magnitude as a smaller fraction of the detectors solid angle is available to detect neutrons emitted from the source.

The DFM design is optimized for detection of thermal neutrons. The front face of the detector packages is open to the dig face with only an aluminium cover (to protect the detectors from shock and intrusion of contamination). Polyethylene is located at the sides and on top of the detector in order to act as a reflector to improve neutron counting efficiency. As illustrated in Figure 7, the MCNP model has demonstrated that, for neutron sources buried 30cm or more in soil, the emerging neutron flux at ground level is dominated by the low energy component.

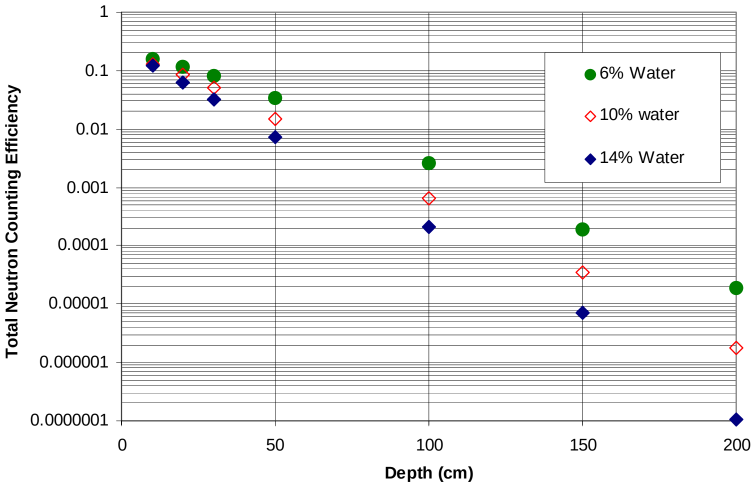


Figure 4. Total Neutron Counting Efficiency as a function of plutonium depth

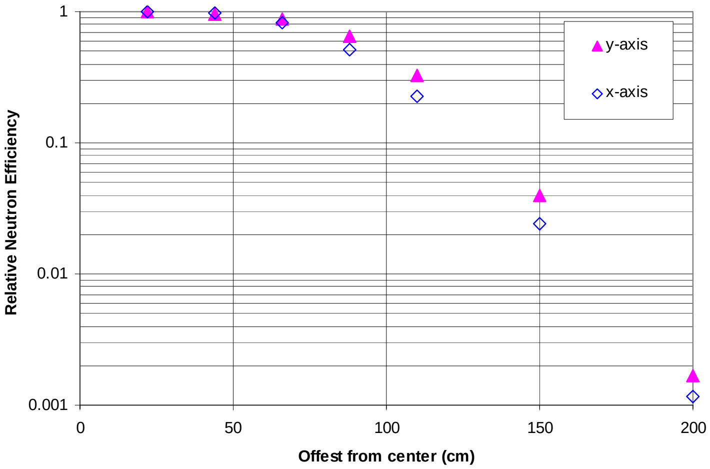


Figure 5. Neutron Efficiency as a function of offset from center along orthogonal axes

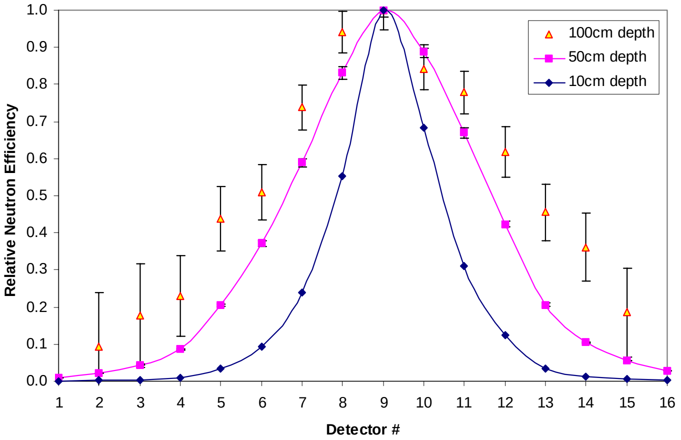


Figure 6. Neutron Efficiency for each detector with plutonium at various depths in soil

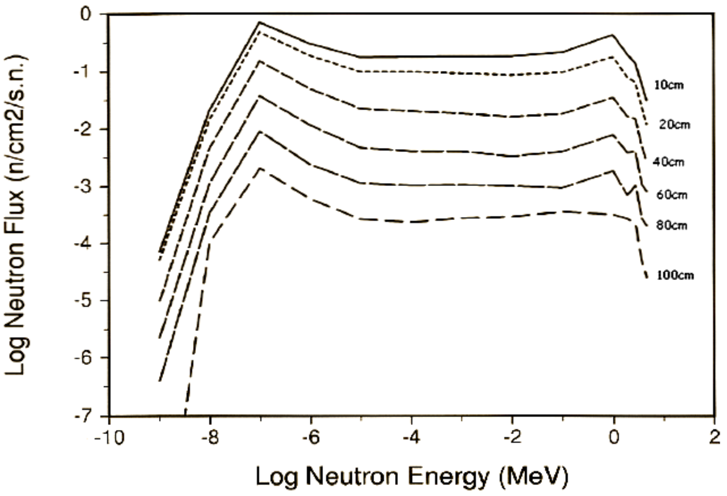


Figure 7. Energy distribution of neutron flux measured at surface for Pu at various depths in soil

PERFORMANCE

It has been demonstrated that the DFM's lower limit of detection is less than 20 g Weapons Grade (WG) Pu for a source in a homogeneuous matrix at a depth of 1 m for a 500 second count time [3]. With a source uniformly distributed in the soil over this depth, and standard operating mode count time of 15 minutes, the detection limit is less than 1g WG Pu.

Uncertainties in the measurement are relatively large because (i) the specific neutron emission rate is a function of chemical and isotopic composition (ii) source distribution is unknown and response varies by an order of magnitude for every 30 cm of soil depth (iii) response is a strong function of soil moisture content, which is often unknown. For these reasons the DFM should not be considered an assay instrument, rather a field survey meter.

Demonstration tests [3] have been performed with the DFM that demonstrate that an absorbing or highly scattering layer of material in a drum does not significantly perturb the response function of the system. Measurements were performed with sources loaded into simulated 200 liter waste drums containing mixtures of polyethylene, vermiculite, wood, graphite, borax (a neutron absorber), concrete rubble, light metals and lead. The total count rate varies by less than 30% about the mean value.

IMAGING

The layout of the dig face monitor neutron detectors within the polyethylene moderator has been designed to optimize the imaging capabilities of the system [4]. Where sufficient signal is detected from a buried source, the differential in the detector signal vector can be used to provide a x-y location of single or multiple underground sources. This requires that the system is deployed with two orthogonal measurements over a hot-spot. Furthermore, the depth of the source may be determined by analysis of the shape of the peak in the detector response vector. A peak that falls of rapidly indicates a source near the surface, whereas a shallow peak indicates a deep source.

Figure 6 shows how the neutron detection efficiency (normalized to maximum efficiency) varies as a function of detector channel for sources at various depths under detector #9. This function may be modeled using an asymmetric double sigmoidal function such as given in Equation (2),

$$y = \frac{a}{1+\exp\left(\dfrac{-x-b+c/2}{d}\right)}\left(1-\frac{1}{1+\exp\left(\dfrac{-x-b-c/2}{e}\right)}\right) \qquad (2)$$

The *d* and *e* parameters determine how quickly the response function, *y*, falls off with detector channel number, *x*, on the left and right sides, respectively, of the peak. The slope of the data function depends on the depth of the source, therefore these parameters can be used as a measure of depth. The parameter *a* is related to the maximum of the function and the *b* and *c* parameters relate to the centroid of the function.

CONCLUSIONS

Application of the total neutron counting technique is a powerful analytical tool for investigation of buried plutonium. The path length of the emitted fast neutrons allow characterization to be performed at significantly greater depth than gamma or x-ray based methods which are limited to surveying of surface contamination.

Modeling and experimental work has demonstrated that the Dig Face Monitor is capable of detecting plutonium over an excavation zone of 3 m x 3 m and up to 1 meter below the surface. The lower limit of detection is less than 1g of Weapons Grade Pu in a 15 minute count. The system can also be deployed in imaging mode by taking orthogonal measurements over a neutron hot-spot. This will allow the end user to create a 3-dimensional image of underground plutonium at a burial site.

REFERENCES

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