MCNP Modeling Results for Location of Buried TRU Waste Drums

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Abstract In the 1960’s, fifty-five gallon drums of TRU waste were buried in shallow pits on remote U.S. Government facilities such as the Idaho National Engineering Laboratory (now split into the Idaho National Laboratory and the Idaho Completion Project [ICP]). Subsequently, it was decided to remove the drums and the material that was in them from the burial pits and send the material to the Waste Isolation Pilot Plant in New Mexico. Several technologies have been tried to locate the drums non-intrusively with enough precision to minimize the chance for material to be spread into the environment. One of these technologies is the placement of steel probe holes in the pits into which wireline logging probes can be lowered to measure properties and concentrations of material surrounding the probe holes for evidence of TRU material. There is also a concern that large quantities of volatile organic compounds (VOC) are also present that would contaminate the environment during removal. In 2001, the Idaho National Engineering and Environmental Laboratory (INEEL) built two pulsed neutron wireline logging tools to measure TRU and VOC around the probe holes. The tools are the Prompt Fission Neutron (PFN) and the Pulsed Neutron Gamma (PNG), respectively. They were tested experimentally in surrogate test holes in 2003. The work reported here estimates the performance of the tools using Monte-Carlo modelling prior to field deployment. A MCNP\(^2\) model was constructed by INEEL personnel\(^3\). It was modified by the authors to assess the ability of the tools to predict quantitatively the position and concentration of TRU and VOC materials disposed around the probe holes. The model was used to simulate the tools scanning the probe holes vertically in five centimetre increments. A drum was included in the model that could be placed near the probe hole and at other locations out to forty-five centimetres from the probe-hole in five centimetre increments. Scans were performed with no chlorine in the earth around the probe hole and with three concentrations of chlorine. The PFN is configured with two detectors, and therefore data from the scans can be used to gauge the approximate distance of a drum from the probe-hole. The PNG uses ratios of gamma-ray intensities from a specific element to estimate the quantitative distance to the drum. Moreover, the PNG can distinguish VOC materials in the formation from those in the drum if there is sufficient contrast in the concentrations. The PNG and PFN can be used together to assist in planning the safe removal of contaminated materials, both TRU and VOC.

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\(^3\) McGrath, C., and Van Ausdeln, L., private communication
1. Introduction

The goal of this work is to use the Monte-Carlo code, MCNP, to demonstrate that both the Prompt Fission Neutron (PFN) and Pulsed Neutron Gamma (PNG) tools have a capability to provide information about the distance of an object from the probe-hole wall while also quantifying the concentration of TRU and volatile organic compounds (VOC) in the formation surrounding the probe-holes.

The research was undertaken to demonstrate the feasibility of two technical objectives:

1. Show how data from these tools can be used for accurate, in situ, buried TRU and MLLW waste location and characterization.
2. Demonstrate that heterogeneities encountered at waste disposal sites can be identified from signals in the tools and that accurate mass distributions in inhomogeneous waste disposal regions can be determined from in situ logging measurements.

A modeling effort was undertaken to determine the expected performance of the two logging tools for satisfying these criteria. The PFN tool produces a sharply defined, fifteen microsecond-wide, monoenergetic neutron pulse and contains a set of both epithermal and thermal neutron detectors. The detectors are used to determine neutron count rates as a function of time. For detecting TRU (and MLLW containing fissile elements), fission neutrons are detected following the end of the neutron burst and the fast decay of epithermal neutrons from the burst. Epithermal neutrons are then produced by thermal fission of Pu (or uranium) during the time decay of the thermal neutron flux. The PNG acquires time-dependent gamma-ray spectra that can be used for general elemental analysis for characterizing the properties of the environment and to detect components of waste that are hazards in the environment such as carbon tetrachloride. The model uses the MCNP program developed at Los Alamos (Briesmeister 2000) which is a Monte Carlo program designed to calculate radiation transport for neutrons, gamma rays, and coupled neutron-gamma ray through extended media. The use of this modeling approach allows us to estimate the performance of actual tools under a variety of different environments and conditions.

We have constructed a model that is intended to represent the subsurface area where TRU and MLLW have been buried. A probe hole for lowering the PFN and PNG tools into the subsurface without coming in contact with the environment has been included. A drum containing a representation of what might be typically contained in such a burial site has been included in the model. During the set of calculations, the location of the drum is moved to evaluate the sensitivity for detecting the drum and its contents as a function of location relative to the probe hole.

The complete environment surrounding the probe for measurement is approximately 1.3 meters high and 1.5 meters in diameter. The region is divided into cells. For the calculation there are twenty-six vertical divisions and nine sections in the radial direction plus the probe hole containing the measurement tool. A 55 gallon drum is created as an independent object and is moved around the region as needed to simulate different locations relative to the probe. Figure 1 shows a vertical cross section of the formation, the PFN tool in the probe hole, and the 55 gallon drum entering the lower right of the formation. Figure 2 shows the horizontal cross section of the same region through the probe hole and with the 55 gallon drum containing TRU and MLLW entering on the right. Approximately three hundred calculations were performed for the PFN tool and for the PNG tool to obtain the basic response of the two tools in the environment that is expected for the buried waste.
Figure 1 Vertical cross section view of model. The white area entering the formation on the lower right is the 55 gallon drum.

Figure 2 Horizontal cross section of the model showing the probe hole with the PFN tool in the center and the 55 gallon drum entering on the right.

2. PFN-Tool Results
The first, and most significant, question is whether TRU can be observed by one of the two instruments. The drum in the model has concentrations of weapons grade plutonium (WGPu) ranging from 0.1
weight% to 0.01 weight% representing the TRU. Because one of the objectives of the calculation was to have sufficient statistical precision to map the spatial response, the higher concentrations were used far from the probe hole and the lower concentrations were used near the probe hole. However, the data reported here have been normalized by the concentration of Pu. The response profiles of the PFN tool to the plutonium in the drum can be seen in Figure 3. This figure shows three calculations of the PFN near epithermal detector count-rate response to the drum containing plutonium at 1.6, 4.6, and 9.6-inches form the probe hole, respectively. It is clear that there is good sensitivity to the presence of plutonium at substantial distances from the probe hole.

If we consider the re-normalized data of Figure 3 to the response at 1.6-inches, shown in Figure 4, we see that there is not only an attenuation of the counts with distance from the probe hole, but that the width of the distributions change as a function of the distance of the source from the probe hole. Thus the combination of the count rates and the width of the measured distributions permit both an estimate of the activity of the source and the radial position it has relative to the probe hole.

A similar response to the presence of plutonium is obtained for the far epithermal detector of the PFN tool. The two sets of data taken together permit an accurate determination of the amount of plutonium present and also its radial and vertical position relative to the probe-hole and the measure point of the PFN tool. Our research shows that the probe is capable of measuring plutonium at a concentration of 10 nano-Curies/gram (nCi/gm) of WGPu. To appreciate the sensitivity of this measurement, Figure 5 shows the time-decay spectrum of epithermal neutrons following the burst of neutrons from the PFN tool.

Figure 3 PFN response of the near epithermal detector to plutonium-containing drum at four distances from the probe hole. The blue diamonds are for the drum 25 centimeters from the probe hole, the purple squares are for the drum 14 centimeters from the probe hole, the triangles are for the drum at 9 centimeters from the probe hole, and the brown circles are with the drum at 4 centimeters.
Figure 4 Normalized data from Figure 3 showing the narrowing of the width of the distribution of the signals from plutonium as the location of the drum is moved away from the probe hole. Legend is the same as Figure 3 except with the 9-cm data set absent for clarity.

**Epithermal Neutron Decay Curves**

Figure 5 Epithermal neutron decay curves with no plutonium present and with four Nuclear Accident Dosimeter (NAD) sources at different distances from the probe hole.

When there is no plutonium in the formation, the epithermal neutron counts go to zero by about 400 microseconds after the neutron burst as shown by the magenta squares in Figure 5. Thus by only counting the epithermal neutrons that are present later than 400 microseconds after the burst a nearly background-free signal is obtained of the presence of TRU.
While it is generally accepted that MCNP calculations can reproduce the response observed in actual calculations, it is important to ensure that the model has been built accurately and in sufficient detail. For the PFN, calculations and experimental measurements are compared with the shape of the Near and Far epithermal detector count rates as the tool logs past NAD sources. Figure 6 compares the near and far detector response shapes observed by the PFN tool logging a laboratory formation containing NAD sources at a distance of 3.75 inches from the borehole with the comparable response calculated from MCNP with the source located at three inches from the borehole. Note that both figures are actually on the same horizontal scale. The shapes of both experimental curves are well reproduced by the calculations with regard to the general shapes of the curves, the widths of the curves, and the spatial separation between the responses of the near and the far detectors. In addition, the response of the far detector is asymmetric in both the experiment and the calculation while the near detector has a symmetric response. Thus, this comparison confirms the calculations are sufficiently accurate to reproduce the experimental tool response.

![Shape Comparison MCNP VS Experiment](image)

Figure 6 Near and Far detector scans and MCNP scans past NAD sources located 3.75 inches from the borehole by the PFN tool in a laboratory formation. Counts are normalized to compare shapes only.

3. PNG-Tool Results

The PNG tool is based on a large-volume, high energy resolution intrinsic n-type germanium detector, a digital signal processor (DSP) for high-throughput with good energy resolution processing of the signals from the germanium detector, and a pulsed neutron generator to measure time-dependent spectra of the gamma-rays. The neutron generator has an output on the order of 10^8 neutrons/sec. This type of detector permits the most sensitive measurement of the energies of gamma rays that are incident on the detector, thus providing the cleanest, least ambiguous result to be obtained from the spectral data. Essentially all of the elements in the formation can be detected by gamma rays produced either during the burst of
neutrons, “inelastic,” or those produced after the burst of neutrons through the capture of thermal neutrons, or “capture.” In addition to these types of gamma rays, the PNG can also measure natural radioactivity or that from man-made radioisotopes such as plutonium, and can also measure delayed, neutron-induced radioactivity following “inelastic” or “capture” events. Both “inelastic” and “capture” gamma rays are efficiently calculated with MCNP, and the other types will not be discussed here. An example of the spectrum that is measured by the detector in the PNG is shown in Figure 7. While many of the gamma rays in this spectrum are due to thermal capture by chlorine, other gamma rays can be used to identify the concentration of essentially all other formation components. The following list shows a number of the prominent gamma rays that can be used to identify the element that is present and its concentration in the environment. Elements that can be seen in the inelastic spectrum include: C – 4.4395 MeV, O – 6.131 and 7.12 MeV, Fe – .847, 1.039, 1.813, and 2.113 MeV, Cu - .963 MeV and Si -1.273 and 1.779 MeV. Elements that can be seen in the capture spectrum include: H – 2.225 MeV, Cl – 6.109, 1.957, and 1.164 MeV, (and many others), Si – 3.539, 4.936, 6.383, 1.273, and 2.094 MeV, Fe – 7.631 and 7.645 MeV, and Al – 7.727 MeV.

![Ge Spectrum in Chlorinated Surrogate](image)

**Figure 7** Spectrum from germanium detector obtained in a chlorinated environment. Horizontal scale energy range is approximately 8 MeV.

While the MCNP calculations provide information on many elements that allow us to determine the presence of metals and the soil environment of the measurement region, we focus here on two main aspects, the measurement of carbon and chlorine content and the use of multiple gamma rays from a particular element to estimate the distance of that element from the detector. The purpose of measuring carbon and chlorine is to identify the presence of chlorinated compounds such as carbon tetrachloride and to separate them from inorganic chlorides such as NaCl. The purpose of identifying the location of the element relative to the detector is to see if a chlorinated compound is located in a drum and where the drum is located or if it has leaked and its contents have permeated the environment.

Approximately three weight percent of carbon was placed in the drum in our Monte Carlo model. The relative sensitivity of carbon counts as a function of distance from the borehole is shown in Figure 8. The flat response seen from about 10 inches outwards from the borehole wall is because there is carbon present in the tool itself. There is a clear decrease in the response as a function of the radial distance from
the borehole. It is worth noting that this response is that expected from $10^8$ neutrons, or about one seconds’ worth of data. Of course, the detector is not 100% efficient at these energies, but a typical measurement of 100 seconds will likely provide better precision (estimated uncertainties on the points shown on the figure are about +/- 10%). Thus, it is likely that if carbon is present in a drum, especially at expected concentrations, it would be seen out to a foot or more from the borehole. This distance could be increased further by reducing some of the carbon content in the tool.

![Carbon Response](image)

**Figure 8** Carbon response as a function of distance from the borehole.

Similar calculations were performed to evaluate the response to chlorine in the drum as a function of distance from the borehole. The sensitivity to chlorine is very high. When the only chlorine was in the drum at a concentration of three weight percent, the signal obtained in the 6.109 MeV peak of chlorine is shown in Figure 9. It can be clearly seen that there is sensitivity to the presence of chlorine even when the drum is more than fourteen inches from the probe hole. Calculations also were performed with no chlorine in the drum and low levels of chlorine present in the formation. This corresponds to, for example, carbon tetrachloride leaking out of a drum and being in the vapor phase in the formation. Extrapolating from the levels used in the model, we conclude that we can see chlorine levels to approximately 20 ppm in the formation in reasonable measurement times. Thus, by using multiple peaks in the spectrum from chlorine, we should be able to have very good sensitivity to chlorine in both low concentrations in the formation and in drums a substantial distance from the probe hole.

To enhance our sensitivity for determining the distance of a chlorine source, and to demonstrate the fundamental measurement ability, we examined the relative counts of different energy peaks in the capture chlorine spectrum. The simple idea is that the mean free path of a gamma ray increases with increasing energy. Thus, low energy peaks are scattered to a greater extent traveling the same distance to the detector than does a higher energy peak. Therefore, the ratio of a low energy peak to a high energy peak will decrease with increasing distance of the source from the borehole. We have examined the case for the 1.96 and 1.16 MeV peaks in chlorine as compared with the count rates for the 6.109 MeV peak. The results are shown in Figure 10. It is seen that when the drum is about two-inches from the probe hole, the initial ratios are above 0.2 and 0.1, respectively. As the drum is moved out about 8 inches more, these ratios are reduced by over a factor of two. Thus, the combination of the intensity from the 6.109 MeV peak and the ratios of lower energy peaks to the 6.109 MeV peak provide an accurate method to
extract both the chlorine concentration in the drum and the location of the drum relative to the probe hole. We have also performed calculations with chlorine in the formation and not in the drum. In those cases, because the chlorine is located closer to the probe hole, the ratios are increased by about 20%. Thus, we believe that it is also possible to separate chlorine in a drum from chlorine in the formation based only on the ratio of the low energy chlorine peaks to the high energy peak. This technique can then be applied to other elements which produce both high energy and low energy peaks through either a capture reaction or an inelastic reaction.

A few other observations about the chlorine response and the measurement capabilities of PFN and PNG tools are worth noting. As the chlorine concentration in the formation is increased the response is not linear. This is because of the well known “SIGMA Effect” which reduces the additional counts that are produced as more chlorine (or other high capture cross section element) is added to the environment. Unlike a thin target measurement where the counts are proportional to the content in the target, a measurement in extended media will have all the neutrons absorbed regardless of the contents of the formation. The number captured by a particular element will be proportional to the partial macroscopic absorption cross section for the element relative to the total absorption cross section for the medium. In the case where any element is a small portion of the total cross section, then increasing the amount of that element will lead to a proportionate increase in the count rate observed. But when a particular element dominates the total cross section, an increase in its macroscopic cross section does not result in a proportional increase in the count rate. Problems with non-linear chlorine responses were observed in the earlier work at the SDA, and the logging researchers were not able to characterize the amount of chlorine properly. However, two important developments in our research resolve this problem. First, both the PFN and the PNG have the capability for directly measuring the total macroscopic absorption cross section. Second, by being able to measure the concentrations of essentially all elements in the formation environment, a geochemical model for the absolute concentrations can be constructed. Work in oilfield logging has shown this technique allows one to determine the concentration of all measured elements in the environment, independent of the SIGMA Effect.

4. Conclusions
By combining the results of MCNP calculations from the PFN and the PNG tools we can now conclude that
a) We will be able to use data from these tools for accurate, \textit{in situ}, buried TRU and MLLW waste location and characterization and
b) Heterogeneities encountered at waste disposal sites can be identified from signals in the tools and
c) Accurate mass distributions in inhomogeneous waste disposal regions can be determined from \textit{in situ} logging measurements.

![Gamma Intensity Ratios vs. Radial Position](image.png)

Figure 10 Ratio of intensity of low energy chlorine capture peaks to the 6.109 MeV peak for chlorine in drums, as a function of distance from the probe hole.