LA-UR-97-0692

Title:

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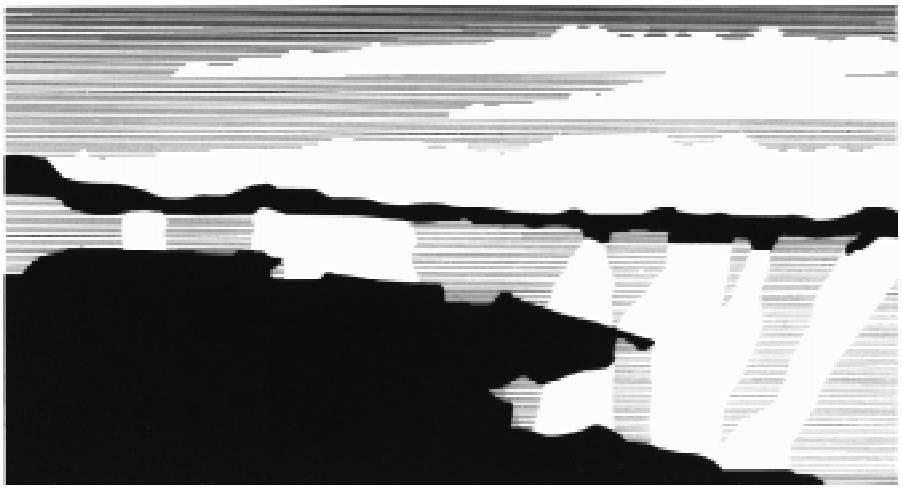

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Form No. 836 R5
ST 2629 10/91

MCNP™ Simulations for Identifying Environmental Contaminants Using Prompt Gamma Rays from Thermal Neutron Capture Reactions

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Abstract

The primary purpose of the Multispectral Neutron Logging Project (MSN Project) was to assess the effectiveness of existing neutron-induced spectral gamma-ray logging techniques for identifying environmental contaminants along boreholes, further improve the technology, and transfer that technology to industry (Conaway and Frankle, 1993). The logging instrument incorporated a pulsed 14-MeV neutron source and HPGe detector. Gamma-ray spectra from thermal neutron capture reactions were used to identify contaminants in the borehole environment. This type of technique complements physical sampling and is useful in environmental restoration projects where characterization of contaminated sites is required and long-term monitoring may be needed. We used an enhanced version of the Monte Carlo N-Particle computer code (MCNP™) to investigate many more potential contaminants than could be accomplished experimentally. MCNP was tested and benchmarked with experimental data for Cl provided by our collaborators (Myers, 1988 and Frankle, 1994) and experimental data for Cl, Cd, Sm, Gd and Hg obtained as part of this project (Conaway and Duray, 1994). A number of enhancements were developed for MCNP as a result of this project, some of which will be part of the next major code release. The results from the benchmarking effort, estimates of minimum detection thresholds for our instrument based on MCNP simulations, and some of the MCNP code enhancements will be discussed.

Introduction

The primary purposes of the Multispectral Neutron Logging Project, (MSN Project, funded by the U.S. Department of Energy), were to assess the effectiveness of existing neutron-induced spectral gamma-ray logging techniques for identifying environmental contaminants along boreholes, to further improve the technology, and to transfer that technology to industry (Conaway and Frankle, 1993). Using a pulsed neutron source with a high-resolution gamma-ray detector, spectra from thermal neutron capture reactions may be used to identify contaminants in the borehole environment. Direct borehole measurements such as this complement physical sampling and are useful in environmental restoration projects where characterization of contaminated sites is

required and long-term monitoring may be needed for many years following cleanup or stabilization.

In the MSN Project, a prototype logging instrument was designed which incorporated a pulsed 14-MeV neutron source and HPGe detector. Experimental measurements to determine minimum detection thresholds with the prototype instrument were conducted in the variable-contaminant test model for Cl, Cd, Sm, Gd, and Hg (Conaway and Duray, 1994). We benchmarked an enhanced version of the Monte Carlo N-Particle computer code MCNP™ (Breismeister, 1991) using experimental data for Cl provided by our collaborators (Myers, 1988; see also Frankle, 1994) and experimental data from the variable-contaminant test model. MCNP was then used to estimate detection thresholds for the other contaminants used in the variable-contaminant model with the goal of validating the use of MCNP to estimate detection thresholds for many other contaminants that were not measured.

Preliminary Studies with MCNP

Preliminary studies were conducted in 1993 using experimental data provided by our collaborators (Myers, 1988; see also Frankle, 1994). The physical model consisted of 1.27 cm thick glass plates stacked with 0.424 cm spacers surrounding a borehole (15.74 cm ID) cased with 2.07 cm thick steel casing and a 2.86 cm thick layer of cement. The borehole was filled with a sodium bromide solution, and the voids between the glass plates were filled with one of two saline solutions. The particular instrument used by our collaborators for these tests failed to detect Cl for either concentration, and only the experiment using the lower concentration of NaCl (9984 ppm by weight) was modeled using MCNP. The logging instrument used in these measurements consisted of a neutron generator and HPGe detector, and was sidewalled in the borehole (i.e. positioned against the borehole wall). The results of these simulations showed that MCNP could be used effectively to model this type of application. While Cl was not detected by the logging instrument, spectral peaks at discrete energies corresponding to thermal neutron capture reactions in Cl were evident in the simulated spectra. Since no Ge data are presently available for MCNP, the simulated photon spectra are equivalent to that for a 'perfect' detector, detecting all photons at their primary energy and including no contributions from (n,γ) reactions within the detector itself.

These studies indicated a number of improvements that could be made in the logging instrument as well as in the simulation code MCNP. Some of the improvements that were made for the prototype instrument design included using brass for the detector housing in place of stainless steel, increasing the detector size/efficiency substantially, placing the detector as close to the neutron source as possible, using ^{6}Li-loaded polyethylene for shielding in place of the more common steel or borated-polyethylene, and improving the electronics package. Enhancements to

the simulation code included incorporating a perturbation technique for assessing tally differences due to small changes in material composition (McKinney and Iverson, 1996), a special tally feature to separate photon spectra into bins based on the original source nuclide (Hughes, 1994), and improving the nuclear data used by MCNP for a number of nuclides. In particular, the photon production spectral data used by MCNP is often binned in 50-250 keV energy bins containing no discrete photon lines. This type of data is not adequate for use in applications using high resolution photon detectors. Additionally, we noted major differences in the discrete photon data for Cl between the MCNP data and compilations such as by Orphan (1970) or Lone (1981).

Experimental Measurements and Preliminary Contaminant Detection Thresholds

Experimental measurements were conducted using the variable-contaminant test model to determine detection thresholds of the prototype logging instrument for Cl, Cd, Sm, Gd, and Hg (Conaway and Duray, 1994). Sm and Gd were included to provide additional information about the instrument response, and not because they are contaminants of interest. The variable-contaminant test model consisted of stacked concrete cylinders with a 15.24 cm borehole down the middle. A central, removable 15.24 cm thick contaminated zone doped with selected contaminants was sandwiched between thicker layers of uncontaminated concrete. The concrete cylinders had outside diameters of 76.2 cm and were surrounded by a tank of gravel and water having an outside diameter of 2.07 m, providing an effectively infinite test medium. Six contaminated cylinders were manufactured for these experiments including two different concentrations of Cl and one each of the other contaminants listed above. Boron carbide was present in the uncontaminated and Cl-contaminated concrete cylinders to assure that all concrete cylinders had approximately the same total thermal neutron capture cross section. The 15.24 cm borehole was air-filled and the prototype logging instrument was centered in the borehole during the experiments.

The instrument used a pulsed neutron generator operating at 3 kHz with a burst duration of ~33 μs. The HPGe detector was an N-type crystal with an efficiency of 22.5% and a resolution of approximately 3.5 keV at FWHM at lower energies, and the center of the detector was located approximately 51 cm from the neutron source. Each photon spectrum was separated into inelastic (during pulse) and capture (between pulses) spectra. Spectra were acquired for 1000 s live time at 13 depth positions in the test model for each contaminated cylinder. The maximum detector response for the contaminants occurred when the center of the detector was located near the top of the contaminated zone and the neutron source was located about 36 cm below the bottom of the contaminated zone (Conaway and Duray, 1994).

In these experiments, comparisons between data taken at the maximum response position and at a location 60 cm away, along with published literature for the contaminants of interest,

positively identified the peaks used in the fitting process. Only the information contained in the primary spectral peak for a particular photon was used in the data analysis; the information in the first and second escape peaks was not utilized. The peak area was calculated by subtracting the average background continuum immediately to either side of the primary peak of interest.

The peak area for each characteristic photon energy as a function of detector location was used to obtain an equivalent thick-bed response using the linear superposition principle. In this procedure, the response to the 15.24 cm bed as a function of depth is offset in 15.24 cm increments and summed at each depth point to produce the "infinitely thick" bed response; in practice, infinitely thick is around one meter (Conaway and Duray, 1994). The thick-bed response was found to be approximately twice the response at the maximum response location for all photon energies and contaminants. The definitions for minimum detection threshold and quantitative thresholds are from Currie (1968). The quantitative determination threshold is approximately 6 times greater than the minimum detection threshold. Table 1 presents results for the minimum detection thresholds for the prototype logging instrument for the five contaminant materials.

MCNP Simulations of the Experimental Measurements

MCNP simulations were performed for both Cl concentrations along with Cd, and Hg, at the maximum detector response position determined during the experimental measurements. The contaminant concentrations for these simulations were 2700 and 13600 ppm of Cl, 595 ppm of Cd, and 7140 ppm of Hg by weight. A density of 2.1 g/cc was used for the contaminated and uncontaminated concrete cylinders and the boron carbide described above was also used in the simulations. Fairly detailed representations of the variable-contaminant test model and the prototype instrument were used in the simulations. Due to the lack of Ge cross section data, the detector volume was modeled as a void region. Standard variance reduction techniques were used such as weight windows as a function of position and energy for neutrons and photons. Additionally, source importance sampling was used in the final simulations (Lichtenstein, 1995). Standard ENDF/B-V based cross-section data were used for all nuclides other than the contaminant nuclides. Modified cross section data were used for Cl, Cd, and Hg, where the photon production data had been updated based on Orphan (1970).

The photon flux tally through the detector volume was separated into the inelastic and capture time bins, and was further separated into bins based on the source nuclide as well as the total. The photon flux spectra were binned in 5-keV wide energy bins. Simulations corresponding to approximately 1s of experimental data were performed (10^7-10^8 source neutrons). Figure 1 illustrates the total photon flux through the detector region for the capture time bin and the component of the spectrum associated with Cl over the photon energy range E_γ = 1.0-1.5 MeV.

Previous simulations using the perturbation technique in MCNP had shown that a linear extrapolation for estimating the minimum detection threshold for a contaminant was valid, subject to the limitation that the neutron transport is not significantly affected by the differences in material specifications.

Following procedures similar to those used for analyzing the experimental data, minimum detection thresholds for a thick-zone response for 1000s of live time were estimated from the MCNP results (Table 2). Recall that MCNP is simulating a perfect detector response and not the actual HPGe detector response of the prototype tool. Using information from the experimental measurements, the thick-bed response may be estimated by dividing the maximum response data by a factor of 2 (Conaway and Duray, 1994). A comparison between the MCNP estimates for a perfect detector and the experimental values as a function of photon energy gives a response function that can be applied to future MCNP results for other nuclides not experimentally measured. Figure 2 shows a comparison of experimental data to MCNP simulated data for Cd over the energy range E_γ = 0.4-0.8 MeV. The MCNP results are given as flux per source neutron and have been normalized to counts for this comparison. As one can see from figure 2, the MCNP simulated data lacks the Ge(n,γ) peaks from neutron interactions within the detector. The particular shape of the ^{10}B(n,α) peak at 0.478 keV is due to Doppler-broadening .

Summary

The preliminary MCNP simulations of experimental data provided by our collaborators (Myers, 1988; see also Frankle, 1994) indicated that MCNP could be used effectively to model measurements in the borehole environment. Additionally, valuable information was gained for further improving instrument design as well as the simulation code itself and the nuclear data used by MCNP. Information provided by MCNP was used in designing the prototype instrument used in the variable-contaminant test model for our latest measurements. Improvements in MCNP and its nuclear data allowed us to better model these measurements and estimate minimum detection thresholds for Cl, Cd, and Hg for the prototype instrument. A limitation still exists due to the lack of Ge data for modeling the HPGe detector in the prototype instrument. Therefore, MCNP is simulating a 'perfect' detector response, detecting all photons at their primary energy and including no photons from (n,γ) reactions within the detector itself. Comparing the experimentally-determined minimum detection thresholds to the MCNP results for the prototype instrument over a wide range of photon energies would give a detector response function that could be applied to future MCNP simulations for contaminants that have not been experimentally measured. Of course, each particular logging instrument would have its own detector response function. We

have shown that MCNP can be used effectively to determine minimum detection thresholds for contaminants in the environment using prompt gamma-rays from thermal neutron capture reactions.

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Table 1. Estimated detection thresholds for several contaminants from data acquired with the experimental prototype in the variable-contaminant test model. The values are based on data acquired during 1000s of live time and are corrected to show a thick-bed response (Conaway and Duray, 1994).

Element	Thermal Capture Cross Section (barns)	Photon Energy (keV) and Yield [a]	Minimum Detectable Concentration (ppm) [b] with Experimental Prototype
Mercury (Hg)	3.76×10^{2}	368; 81.35	33
Cadmium (Cd)	2.45×10^{3}	559; 72.73	1.4
Chlorine (Cl)	3.32×10^{1}	1165; 14.16	86
Gadolinium (Gd)	4.90×10^{4}	1186; 10.83 [c]	1.2
Samarium (Sm)	5.80×10^{3}	334; 98.90	0.9

[a] The yield is given as number of photons per 100 neutron captures (Orphan, 1970).

[b] ppm = parts per million by weight

[c] There are contributions to this peak from the following unresolved photon lines (Tuli, 1991); ^{156}Gd at 1187 keV, ^{158}Gd at 1184, 1186, and 1187 keV. For natural Gd, the overall strength of this line is given as 10.83 photons per 100 neutron capture reactions (Lone, 1981).

Table 2. Estimated detection thresholds for several contaminants from simulated data using MCNP. The values are estimated for data acquired during 1000s of live time for the prototype instrument having a perfect detector, and are corrected to show a thick-bed response.

Element	Thermal Capture Cross Section (barns)	Photon Energy (keV) and Yield [a]	Minimum Detectable Concentration (ppm) [b]
Mercury (Hg)	3.76×10^{2}	368; 81.35	0.8
Cadmium (Cd)	2.45×10^{3}	559; 72.73	0.05
Chlorine (Cl)	3.32×10^{1}	1165; 14.16	3

[a] The yield is given as number of photons per 100 neutron captures (Orphan, 1970).

[b] ppm = parts per million by weight

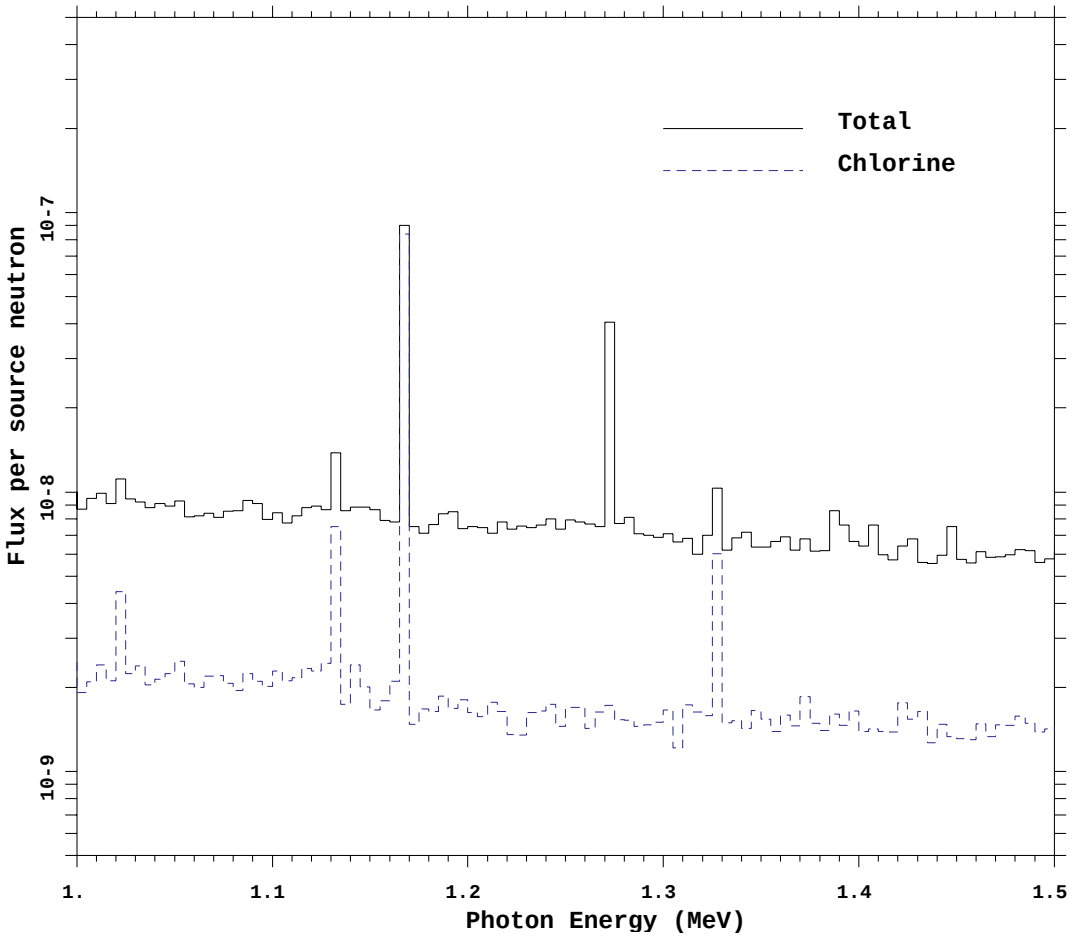

Figure 1. MCNP simulated photon spectra over the energy range E_γ=1.0-1.5 MeV. The total photon spectra is indicated by the solid line and the contribution from thermal neutron capture reactions in Cl (13600 ppm) is given by the dashed line.

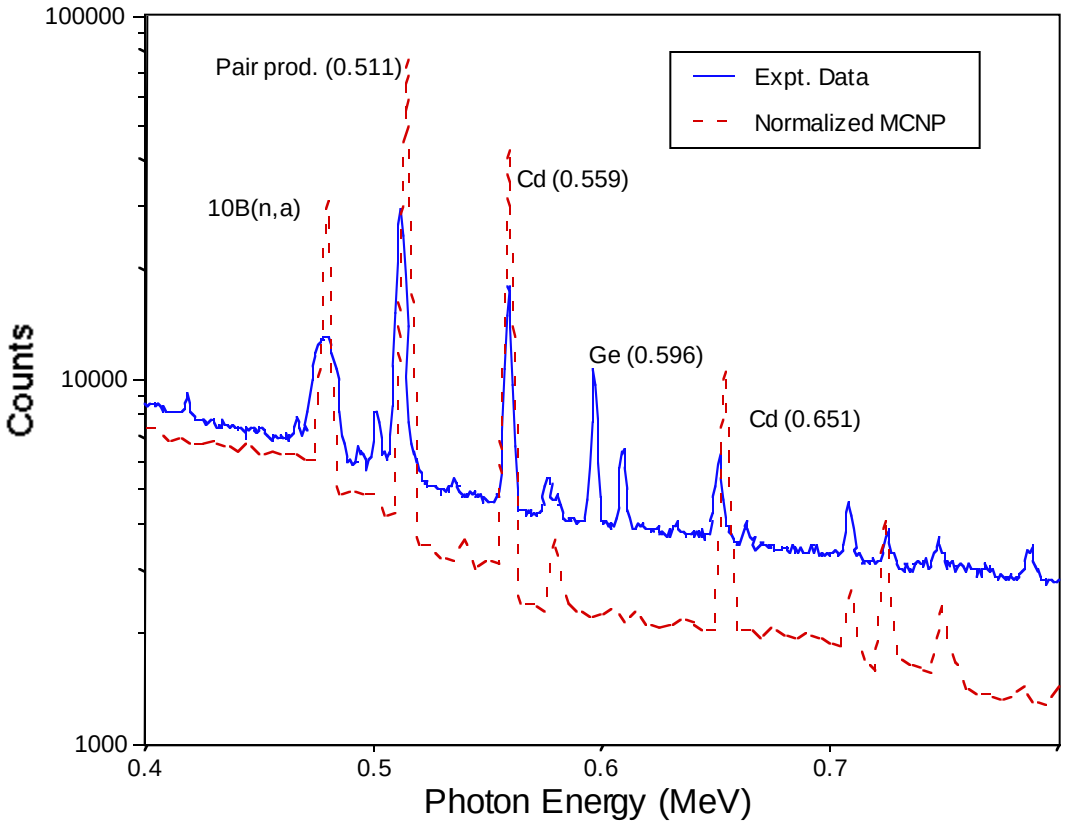

Figure 2. A comparison of experimental data (solid line) and normalized MCNP simulated photon spectra (dashed line) for Cd over the energy range E_γ= 0.4-0.8 MeV. The MCNP data is given in flux per source neutron and has been normalized to counts for comparison purposes.