LA-UR-01-3539

c. 1

COSMIC RAY BACKGROUND ANALYSIS FOR A CARGO CONTAINER COUNTER

N. Ensslin, W. Geist, J. Lestone, D. R. Mayo, and H. O. Menlove
Los Alamos National Laboratory, MS E540
(505) 667-2444

Presented at the
Institute of Nuclear Material Management
42nd Annual Meeting
Indian Wells, CA
July 15-19, 2001

LOS ALAMOS NATIONAL LABORATORY
3 9338 00819 9217

COSMIC RAY BACKGROUND ANALYSIS FOR A CARGO CONTAINER COUNTER

N. Ensslin, W. Geist, J. Lestone, D. R. Mayo, and H. O. Menlove
Los Alamos National Laboratory, MS E540
Los Alamos, NM 87545
(505) 667-2444

ABSTRACT

We have developed a new model for calculating the expected yield of cosmic-ray spallation neutrons in a Cargo Container Counter, and we have benchmarked the model against measurements made with several existing large neutron counters. We also developed two versions of a new measurement uncertainty prediction code based on Microsoft Excel spreadsheets. The codes calculate the minimum detectability limit for the Cargo Container Counter for either neutron singles or doubles counting, and also propagate the uncertainties associated with efficiency normalization flux monitors and cosmic ray flux monitors. This paper will describe the physics basis for this analysis, and the results obtained for several different counter designs.

I. INTRODUCTION

Several of the major DOE/EM sites have strong economic incentives for characterizing large volumes of waste or demolition debris at very low levels, such as 1 to 10 nanocuries per gram of waste, or better still, the "free release" or "put-back" level at roughly 100 to 250 picocuries per gram of waste. To meet this need, it is technically feasible to build counters for 8'x8'x20' or 8'x8'x40' cargo containers based on existing high-efficiency neutron counter nondestructive assay (NDA) technology developed at Los Alamos. However, for very large volumes of waste and very low detection limits, the magnitude and variability of background neutrons produced by cosmic-ray spallations will be the limiting factor in defining the performance of the new counters. (Please see Ref. [1] for a preliminary review of NDA, matrix effects, and cosmic ray issues associated with cargo container counters).

We have carried out an engineering feasibility study [2] to demonstrate that a passive neutron coincidence counter large enough to measure cargo containers can be designed and built with very good detection sensitivity for screening of demolition debris for very low levels of plutonium and americium. The passive neutron counting options analyzed in this study were based on passive neutron totals or coincidence counting.

The passive neutron totals rate can be used to screen for the number of nanocuries per gram. The passive neutron count rate is the sum of (α, n) reactions and spontaneous fissions. The (α, n) reactions are due to alpha decay in plutonium and americium. If there are no spontaneous fissions and no americium, the totals count rate is a direct measure of the nanocuries per gram. If not, it is an upper limit, and can be high by as much as a factor of 2.

The passive neutron coincidence rate can quantify the amount of plutonium in the container, if other information on the plutonium isotopic composition is available. The coincidence signature is relatively insensitive to the (α, n) reactions that will cause neutron totals counting to overestimate the Pu content. Passive neutron coincidence counting can also determine the alpha disintegration rate to within a factor of 2, if necessary, even if the plutonium isotopic composition is not well known.

For put back or disposal of decommissioning debris, it is sufficient to prove that the amount of alpha-emitting radionuclides is below a certain threshold, such as 1 nCi/g or 10nCi/g, without quantifying the exact amount. For this application, total neutron counting is sufficient, and will always be correct or conservative on the high side. The total neutron count also detects ^{241}Am in the waste via the (α, n) reaction on the oxides.

As part of this study, we developed a new model for calculating the expected yield of cosmic-ray spallation neutrons, and incorporated this model into two new measurement uncertainty prediction codes based on Microsoft Excel spreadsheets. The codes calculate the minimum detectability limits for either neutron singles or doubles counting, and also propagate the uncertainties associated with efficiency and with cosmic ray flux monitors. This paper will describe the physics basis for this analysis, how it was benchmarked against measurements made with existing large neutron counters, and how it was utilized to develop conceptual designs for several passive neutron counter options.

Past experience with the design of other large neutron counters has shown that a broad range of new detector design, data collection, and data analysis techniques must be utilized to reduce the background of spallation neutrons from cosmic rays. Otherwise it is not possible to achieve the low Minimum Detectable Limits (MDL) needed to make the counter useful and cost-effective for screening demolition debris. These techniques are not described in this report, but include the following:

1. Optimized design of the counter using Monte Carlo calculations,
2. Truncated multiplicity counting,
3. Spatial correlation veto,
4. Precision-based counting,
5. Archway detector design to allow the container to be scanned through,
6. Add-a-Source matrix correction technique, [3]
7. Substantial overhead shielding,
8. Neutron imaging information from the individual detector banks.

In addition, the spallation neutron yield from cosmic rays must be predicted in a realistic manner. These neutron yields must then be utilized by other prediction codes to predict the MDL as a function of counter design, shielding, measurement procedures, data analysis algorithms, and cosmic ray monitors. To design a counter with acceptable performance, the MDL prediction codes must incorporate or estimate the effects of all of the other techniques listed above. This paper will describe the tools that we developed to predict the spallation neutron yields and the expected MDL of a Cargo Container Counter.

II. SPALLATION NEUTRON YIELD CALCULATIONS

Spallation neutron production from cosmic rays is a complex process. The production rate changes with time, with altitude (for secondary cosmic rays), and with the target atomic number and these changes affect the neutron energy and multiplicity distribution. We developed a model that provides a simplified picture of this complex process, but still contains enough realistic physics to match the experimental data very well. The model develops a source term for the cosmic ray flux, which includes approximate information on the cosmic ray energy spectrum. An internuclear cascade model was used to calculate the number of high-energy neutrons generated by nucleon-nucleon collisions. An "evaporative model" was used to predict the sequential evaporation of neutrons from the compound nuclei remaining after the high-energy nucleon-nucleon collisions have finished. The average neutron multiplicity per spallation event is a power function of the target atomic mass. The multiplicity distributions were found to be relatively flat out to a maximum value. These properties are also obtained from experiments, which observe the neutron singles and doubles count rates from cosmic-ray spallation of high atomic number materials.

Finally, the cosmic-ray spallation neutrons are tracked to the point of interaction in the cargo container. If they drop to below 20 MeV, a Monte Carlo list mode neutron code, benchmarked to the standard MCNP code [4] is used. Secondary charged particles evaporated from the nucleus are not tracked because they do not contribute to the detected signal.

Other Monte Carlo calculations were done for an empty counter and for five matrix materials [1, 2]: a counter with containers of metal, concrete, asbestos, drywall, and concrete with 10% drywall by weight. For the case of a metal matrix, there is no significant absorption effect on the source neutrons. For concrete, the average escape fraction is about 70% for a container with the maximum allowable average loading of 0.673 g/cm^3. For asbestos, the low density reduces the effect of absorption by hydrogen to below what is expected from concrete. For drywall or gypsum, the high (20%, by weight) water fraction means that containers filled primarily with such materials will have neutron moderation and absorption effects too great to permit assay by neutron counting. For the case of 10% drywall in concrete, however, the average loss of neutron detection efficiency is not very great.

The Minimum Detectable Limit (MDL) calculations described below assume a uniform matrix throughout the cargo container. However, several Monte Carlo calculations indicate that if a concrete matrix is only located in the bottom half of the container, the MDL for concrete will increase by roughly a factor of 2. If the concrete is only in the bottom third of the container, the MDL for concrete will increase by roughly a factor of 3, etc. Note that neutron-imaging information from the individual detector banks can be used to flag such loading configurations and correct for them.

This spallation neutron model was used to predict the rates of cosmic-ray spallation neutrons for both the cargo container and the smaller Standard Waste Box (SWB) geometry. The predicted rates for the SWBs were compared to measurements made recently in the SuperHENC neutron counter. Standard Waste Boxes have a volume of about 1.2 m^3, versus about 32.3 m^3 for cargo containers. However, the SuperHENC is the best available comparison, because its design includes several features intended to minimize cosmic ray backgrounds. For measurements of an empty chamber, an empty SWB, an SWB filled with iron, and lead stacked in the cavity, the predicted and measured neutron count rates agreed to within 10 to 20%. The excellent results of this benchmark exercise suggest that the predicted performance at the much larger volumes encountered in cargo containers will be realistic.

III. PREDICTION CODES FOR MEASUREMENT SENSITIVITY

Using the cosmic ray neutron spallation model described in the previous section, we developed two versions of a new measurement uncertainty prediction code. Both codes are based on Microsoft Excel spreadsheets, and both calculate the minimum detectability limit in milligrams of plutonium and in picocuries per gram of matrix material for the Cargo Container Counter for both neutron singles and neutron doubles counting. Input parameters that must be specified include count time, average neutron detection efficiency, detector composition, container composition, matrix composition, plutonium α value, and overhead shielding factors.

One code utilizes Visual Basic macrocodes and a quick approximate empirical estimate for the cosmic-ray spallation neutron yield. The empirical estimates are based on recent experimental studies (unpublished). This code focuses on the effects of matrix shield factors, spatial correlation veto factors, and external background count rates. It was used to compute the MDL for the archway, shielded archway, and 4π counter geometries.

The other code utilizes five Microsoft Excel spreadsheet pages that compute the volume, density, and atomic number distribution of the detector, container, and matrix materials. The code also includes the realistic algorithm, described above, used to predict the spallation neutron yield from cosmic rays.

Both codes propagate the uncertainties associated with efficiency normalization and with cosmic ray flux monitor(s). These uncertainties are very important for low-sensitivity measurements, and may dominate the MDL calculation even more than the available count time. To compute the final MDL, both codes use a conversion factor of 7.5×10^{-2} Ci/g Pu to relate plutonium mass and alpha emission.

IV. COSMIC RAY YIELDS FOR SEVERAL DEPLOYMENT OPTIONS

The spallation neutron yield calculations and sensitivity prediction codes described above were used to compute the MDL for several cargo container deployment options. For each deployment option, the MDL was computed for four fill materials: concrete and cinderblock rubble, metal, asbestos, and concrete with 10% drywall. The average specific density is assumed to be limited to roughly 0.67 gram / cm^3, corresponding to the maximum weight limit of about 22,000 kg in the cargo container. Previous work has shown that a container filled with just drywall, which has about 20% water, by weight, is too moderating to be assayed by neutron counting [1]. Table I provides a summary of the counter parameters used, the cosmic neutron spallation count rates, the Pu count rates, and the singles MDL for each of the four matrices.

Table I. Predicted Performance Estimates for Several Deployment Options.			
Deployment Option	**I**	**II**	**III**
	Unshielded	**Shielded**	**4-Pi**
Design and Engineering Data	**Archway**	**Archway**	**Counter**
Concrete shield thickness	None	4 meters	2 meters
Shielding factor	1	100	7
Efficiency of empty chamber	7.5%	8.0%	30.0%
Eff. For uniform concrete	4.9%	5.4%	20.0%
Eff. For uniform metal	8.0%	8.5%	31.5%
Eff. For uniform asbestos	4.9%	5.4%	19.0%
Eff. For 10% drywall in concrete	4.5%	4.9%	17.0%
Non-cosmic bkg singles rate (cps)	10.0	5.0	1.0
Non-cosmic bkg doubles rate (cps)	1.0	0.5	0.1
Counting Time	1 hour / quarter	1 hour / quarter	1 hour total
Primary activity analysis	Singles	Singles	Singles
Flag for (alpha, n) ratio	None	Doubles/Singles	Doubles/Singles
Matrix efficiency correction	Add-a-Source	Add-a-Source	Add-a-Source
Spallation neutron bkg correction	None	Shield	Shield
MDL for concrete matrix (pCi/g)	1786	345	225
MDL for metal matrix (pCi/g)	2684	235	370
MDL for asbestos matrix (pCi/g)	3269	911	381
MDL for 10% drywall in concrete (pCi/g)	1826	378	229
Pu-240 MDL for concrete (mg)	7.7	1.5	3.9
Pu-240 singles rate @ MDL (alpha=1,cps)	0.78	0.17	1.60
Pu-240 doubles rate @ MDL (cps)	0.010	0.002	0.087
Matrix mass for concrete (kg)	5450	5450	21800
Cosmic singles rate (cps)	40.0	0.4	96.7
Cosmic doubles rate (cps)	1.0	0.012	9.3
Final predicted MDL range	5–10 nCi/g	300–500 pCi/g	250–500 pCi/g

The first deployment option in Table I is a pass-through archway detector large enough for cargo containers to be scanned through on a conveyor system or driven through on a truck. Fig. 1 shows a pictorial of this option. The primary method of data analysis will be neutron singles counting, and there is no correction or flag for plutonium with high α values. The Add-a-Source matrix correction can be implemented to correct for neutron absorption in matrix materials. However, it is important to note that—for a concrete matrix—the unshielded archway has a cosmic ray-induced spallation neutron singles rate that is roughly 50 times the plutonium neutron singles rate if the minimum detectable amount of plutonium is in the container. The final predicted MDL is in the range of 5 to 10 nCi/g.

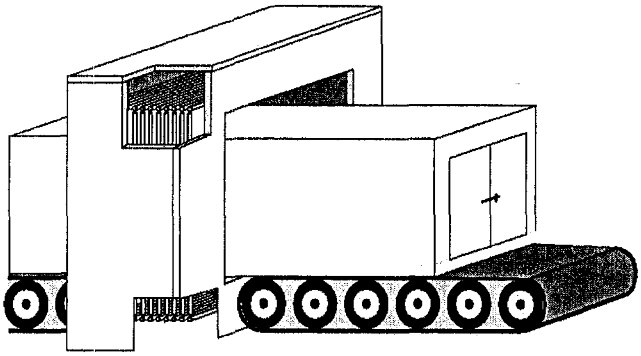

Fig. 1. A conceptual drawing of an "arch" based passive neutron counter (from Ref. 1).

The second option is a pass-through archway detector with a four-meter thick overhead cosmic ray shield consisting of concrete, dirt, or other materials of similar composition. The cosmic ray-induced spallation neutron singles rate will be roughly 2 or 3 times the plutonium neutron singles rate if the minimum detectable amount of plutonium is in the container. The final predicted MDL is in the range of 300 to 500 pCi/g.

The third option is a 4π-geometry counter large enough to accommodate the entire cargo container without scanning, as depicted in Fig. 2. This option is assumed to have two meters of concrete on top of the entire counter, or the equivalent amount of other materials, to provide a factor of 7 reduction in cosmic ray-induced backgrounds. The primary method of data analysis is again neutron singles counting. A 4π-geometry counter with a shield factor of 7 will have a cosmic ray-induced spallation neutron singles rate that is roughly 60 times the plutonium neutron singles rate if the minimum detectable amount of plutonium is in the container. The final predicted MDL is in the range of 250 to 500 pCi/g. Depending on the effective shielding factor that is used, and the availability of representative matrix containers, this deployment option could have a limit of detection of 200 pCi/g or better, so low that the rubble could be put back in place after measurement (the "put back" or free release level).

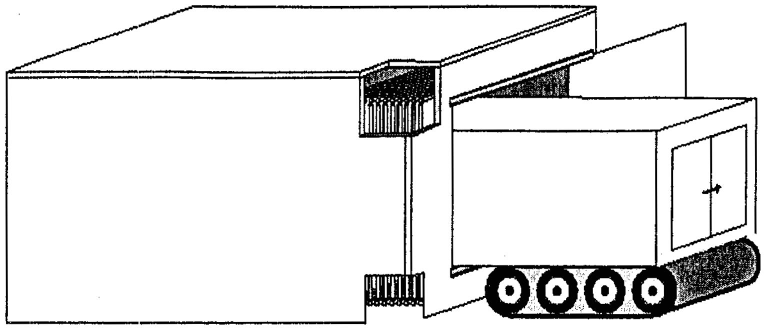

Fig. 2. A conceptual drawing of a full 4π-geometry cargo container counter (from Ref. 1).

For these three deployment options, there are two important considerations that affect the actual MDL that can be obtained in practice.

1. The site will need to prepare a cargo container with a representative matrix fill for each major debris category, so that the background neutron spallation rate can be carefully determined. Then, for containers with actual debris, any net signal above the expected background rate—adjusted for container net weight and changes in cosmic ray activity—can be attributed to plutonium or americium activity. For deployment options with substantial overhead shielding, the range of matrix variability that can be tolerated will be much greater. The MDL values quoted in Table I take this effect into consideration via the propagated measurement uncertainties.

2. Even for cargo containers with similar matrix materials, there will be variations in cosmic ray-induced spallation neutrons from one container to another due to small variations in the matrix. We have carried out a calculation for a concrete matrix with 10% steel and 1% lead added and have determined the incremental increase in the MDL due to these materials for all three options. Clearly, the options with an overhead shield show substantially less effect. Assuming that this is a realistic example of what might be encountered in actual debris, we have scaled-up the final MDL's quoted in Table I to include this effect.

V. CONCLUSION

We have developed algorithms to calculate the cosmic-ray spallation neutron yield in a Cargo Container Counter and to predict the minimum detectability limit (MDL) in milligrams of plutonium and in picocuries per gram of matrix material, as a function of count time, shielding, detector efficiency, detector composition, and container matrix material. The large volume of a Cargo Container Counter means that very low MDL's can be obtained, in principle, but that the background signal of cosmic ray-induced spallation neutrons will be much larger than the plutonium signal. The container-to-container variability in this neutron spallation background will require representative container matrix standards and will limit the performance in the field, unless a heavily shielded archway is used. Low MDL's can only be obtained by compensating for cosmic ray backgrounds using new data analysis and correction

Using our new prediction codes, we have computed the MDL for several different co materials for several deployment options. For fill materials consisting of concrete rubb asbestos, and 10% drywall in concrete, the predicted detectability limits are in the follow ranges:

(1) Unshielded archway counter, 5 to 10 nCi/g,

(2) Shielded archway counter, 300 to 500 picoCi/g,

(3) 4π counter, 250 to 500 picoCi/g.

Depending on the effective shielding factor that is used, and the availability of representative matrix containers, the 4π counter could have an MDL of 200 pCi/g or better, meeting the put-back or free release requirement for some sites.

ACKNOWLEDGMENT

The DOE Environmental Management and Restoration Program sponsored this work.

REFERENCES

[1] D. R. Mayo, H. O. Menlove, and M. M. Pickrell, "Evaluation of SeaLand Cargo Container Nondestructive Assay," Los Alamos National Laboratory document LA-UR-98-5471 (November 5, 1998).

[2] N. Ensslin, W. Geist, E. C. Horley, J. Lestone, D. R. Mayo, and H. O. Menlove, "Engineering Feasibility Study for a Cargo Container Counter," Los Alamos National Laboratory document LA-UR-00-4713 (December, 2000).

[3] H. O. Menlove, "Passive Neutron Waste Drum Assay with Improved Accuracy and Sensitivity for Pu Using the Add-a-Source Method," *J. Inst. of Nucl. Mater. Manage.*, **17**, pp. 17-26 (July 1992).

[4] J. F. Briesmeister, Ed., "MCNP - A General Purpose Monte Carlo Code for Neutron and Photon Transport," Los Alamos National Laboratory report LA-12625-M, Ver. 4A (November 1993).