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Conf-940748--38

LA-UR- 94-2257

Title: TECHNIQUES FOR IMPROVING SHUFFLER ASSAY RESULTS FOR 55-GALLON WASTE DRUMS

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Submitted to: Institute of Nuclear Materials Management (INMM)
35th Annual Meeting
Naples, Florida
July 17-20, 1994
(FULL PAPER)

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TECHNIQUES FOR IMPROVING SHUFFLER ASSAY RESULTS FOR 55-GALLON WASTE DRUMS*

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ABSTRACT

Accurate assays of the fissile contents in waste drums are needed to ensure the most proper and economical handling and disposal of the waste. An improvement of accuracy will mean fewer drums disposed as transuranic waste when they really contain low-level waste, saving both money and burial sites.

Shufflers are used for assaying waste drums and are very accurate with nonmoderating matrices (such as iron). In the active mode they count delayed neutrons released after fissions are induced by irradiation neutrons from a ^{252}Cf source. However, as the hydrogen density from matrices such as paper or gloves increases, the accuracy can suffer without proper attention. The neutron transport and fission probabilities change with the hydrogen density, causing the neutron count rate to vary with the position of the fissile material within the drum. The magnitude of this variation grows with the hydrogen density.

For many common moderating matrices, a simple hardware addition to reduce the average energy of the irradiating neutrons eliminates this problem. But this has the potential of creating another loss of accuracy by increasing self-shielding. Three other techniques are being investigated that maintain the high average neutron energy. These are based on (a) the variance among detector bank counts, (b) a medium-resolution imaging technique, and (c) neural network analysis of detector bank counts. The present states of all four techniques are summarized and compared.

* This work is supported by the US Department of Energy, Office of Nonproliferation and National Security, and Office of Safeguards and Security.

** The research was performed under appointment to the Nuclear Engineering and Health Physics Fellowship program administered by Oak Ridge Institute for Science and Education for the US Department of Energy.

INTRODUCTION

Accurate assays for the fissile contents in waste drums are needed to ensure their most correct and economical handling and disposal. If waste must conservatively be considered transuranic instead of low-level simply because of a lack of accurate knowledge of the fissile mass loading, many drums will be treated as transuranic unnecessarily, burial costs will be much greater than necessary, and burial sites will fill faster than necessary. The passive-active shuffler is one of several instruments being used to assay waste drums, so improvements of its accuracy will also improve the handling and disposal process.

Assays for uranium with shufflers are based on delayed-neutron counting following irradiations by ^{252}Cf to induce fissions.[1] Measurements of large uranium masses in nonmoderating matrices are very precise and accurate.[2] With gram quantities of uranium in 55-gallon waste drums, the measurement can still be sufficiently precise (for example, 2%), but accuracy with moderating matrices (such as paper and rubber gloves) can suffer.[3] If the matrix is nonmoderating (for example, iron or any other metal) there is no loss of accuracy.

Moderators first impede the transport of irradiating neutrons from the ^{252}Cf source to the uranium, and then of delayed neutrons from the fission products to the detector banks. Moderators also lower the average energy of the neutrons, changing the neutrons' capture probabilities with distance traveled. Complete thermalization before capture by the uranium or a detector is unlikely, but even reducing energies from the MeV range to the keV range has important effects. This is still not a problem if the uranium is distributed homogeneously throughout a drum, but it is more likely (and must be assumed) that the distribution is very inhomogeneous and even localized within a small portion of a drum's volume. Therefore, the delayed-neutron count rate depends on the position of the localized uranium within a moderating matrix.[3] If an assay for mass does not correct for this position effect, the result can be in error by a significant amount.

For example, the delayed-neutron count rate from a localized mass of uranium in a drum of paper can be as small as half the rate found with a homogeneous distribution, so a prudent user who does not know the degree of localization or the position of the uranium should assume the worst and multiply the assay result by 1.5. This will usually cause an overestimate of the true mass, but this is more acceptable than an underestimate. A multiplier such as the above 1.5 is not a correction for position of a localized distribution of uranium but a factor applied to all assays out of ignorance of an estimate of the true distribution. It is the goal of this work to find one or more techniques that will provide a unique, reasoned correction factor for an individual drum.

The correction factor to be applied after the position of localized uranium is known is deduced from data given in Ref. 3 that show the variation of count rate with position for a given hydrogen density.

Other instruments might be used to determine the position, but this paper will discuss in detail only techniques that use the shuffler itself. A segmented gamma-scanner[4] can provide some position information for low-density matrices, but a tomographic gamma-ray scanner[5] can provide accurate 3-D information on the location and distribution of uranium or plutonium in heterogeneous matrices. Measurements by a gamma-ray scanner and a shuffler on the same drums complement each other and are being done in some facilities.

When the matrix is nonmoderating, the delayed-neutron count rate is independent of the position of the uranium within the drum[3] and therefore accuracy is not affected. Corrections discussed in this paper are needed only with moderating matrices.

Shuffler assays for plutonium use passive counting of neutrons emitted by spontaneous fissions, so the effects of moderators are less important. For matrices with homogeneous hydrogen densities below 0.02 g/cm^3 the position of localized sources is unimportant.[3] Matrices such as paper and gloves will have hydrogen densities well below this value, so the present paper concentrates on the active mode of the shuffler.

Seven shufflers for 55-gallon drums are now in service with an eighth being fabricated. Three are at the Westinghouse Savannah River Site; two are at the Martin Marietta Energy Systems, Inc., plant at Piketon, OH; two more are at Los Alamos National Laboratory; and the eighth is being made for Lawrence Livermore National Laboratory. Five of these are from a Los Alamos design that has been transferred to Canberra Industries, Inc.,* which has made three of these five units. The other two shufflers are Los Alamos designs with some special features.

A CORRECTION THROUGH HARDWARE

A simple hardware addition can eliminate the inaccuracy caused by the position of localized uranium when the matrix moderation is not too severe and self-shielding can be assumed to be slight. The shuffler's normal configuration has the assay chamber lined with cadmium to absorb thermal and nearly thermal neutrons attempting to enter the assay chamber from the polyethylene detector banks; this minimizes the effects of self-shielding. The cadmium is not needed if it is known that self-shielding cannot be important.

The cadmium in the present shuffler design can be removed in a few minutes, but we have found it simpler to place a polyethylene sleeve around a drum to lower the average energy of interrogating neutrons.[3] This is done routinely with the shufflers already installed. An alternative is to not install the cadmium liner of the assay chamber and to use a cadmium sleeve over a drum. (The cadmium would be sandwiched between metal sheets to protect handlers.) The choice between these two techniques might depend on which sleeve would be used the least. (A secondary inaccuracy found during development with a sleeve[3] has been eliminated through a simple modification in the irradiation scheme.**)

Our measurements[3] over a wide range of moderation show that the polyethylene sleeve eliminates the inaccuracy caused by position when the hydrogen density is 0.01 g/cm^3 or less. To reach this seemingly low density in 55-gallon drums requires large amounts of ordinary materials: 30 kg (66 lbs) of paper or 13.6 kg (30 lbs) of polyethylene shavings, chips, or gloves, for two examples. Our test drum of paper (compressed by hand) held 21.3 kg (47 lbs), or about two-thirds of the hardware-correction limit. In these cases the largest drop in neutron energy occurs in the sleeve rather than in the outermost layer of the matrix, flattening the fission rate throughout the drum.

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A hardware correction can thus eliminate the inaccuracy from localized uranium within a moderating matrix for many of the moderating wastes. However, if the hydrogen density is > 0.01 g/cm^3 or self-shielding concerns require the highest-energy irradiating neutrons, another approach is needed; three options are given in the next sections of this paper.

DETECTOR BANKS' COUNT VARIANCE

The delayed-neutron counts in the detector banks around the side of the assay chamber are not equal and thus have a nonzero variance. The random elements in the emission and transport of delayed neutrons cause fluctuations in the banks' counts, but so does the position of a localized source within a drum. A drum is normally rotated continuously during an assay and makes less than a revolution during a single irradiation and count cycle. If a localized source is near the end of a drum, those detector banks that happen to be passed by the source during the count time will have larger numbers of counts than the other banks. This increases the variance in the counts beyond the variance from counting statistics alone. The amount of increase drops as the source is moved toward the rotation axis of the drum, so the variance beyond that expected from counting statistics is an indicator of the radial coordinate of the source.

The introduction of this concept[6] was part of a general matrix effect deduced with the alternating conditional expectation (ACE) algorithm. The importance of the variance in side banks relative to the overall correction was not easy to identify. A separate study on this question has shown that the range of values of the radial correction term from the side-bank variance is smaller than the positional variations seen in the data. This individual correction was thus muted by being combined with other corrections.

More effort can be put into the development of this technique and it might profit from an increase in the number of detector banks. But as the number of detector banks increases, the banks' count variance from normal counting fluctuations will increase and make it more difficult to calculate and apply the variance from position. An optimum number of banks should be determined not only for this technique but for those described in the next two sections.

MEDIUM-RESOLUTION IMAGING

New opportunities to determine the position of a localized source arise if the assay procedure uses four static orientations of a drum (relative to the ^{252}Cf source) instead of continuous rotation. It has been shown[3] that the total delayed-neutron count is the same for these two procedures, so neither has an advantage in precision. If some irradiation and count cycles are done with a stationary drum, the differences among the detector banks' counts will be maximized. If this is done at more than one orientation, the uranium's position can be estimated. Four orientations satisfy both goals of maintaining the precision while improving the accuracy.

Various image reconstruction algorithms have been applied to data specially taken for this study. A strong neutron source was used to test the concept and the data could be modified to simulate a weaker source. The maximum resolution was allowed by collecting counts from individual detector tubes; lower resolutions were obtained by summing counts from different numbers of adjacent tubes. An empty drum was first used to see if the detector banks would allow imaging under the most favorable circumstances. After obtaining encouraging results, a drum of polyethylene shavings with a hydrogen density of 0.00857 g/cm^3 was used as a highly moderating matrix; this is a somewhat higher density of moderator than expected from combustibles.

The algebraic reconstruction technique[7] (ART) was applied to the cases just described. An intense point source in an empty drum was readily identified; a weak source in a moderating matrix was still located. A pattern of point sources was clearly located in an empty drum, but the points near the center of the pattern were not clearly located in the drum containing the dense hydrogen moderator. The SIMPLEX[8] and maximum likelihood-expectation maximization (ML-EM)[9] algorithms were also applied, but did not improve upon the results with ART for this simple case. The effort concentrated on the radial coordinate, but the height could be estimated from counts in top and bottom detectors (as mentioned in the previous section).

A limitation of this technique is the dependence of the instrument's response function on the moderator density and neutron absorbers. These can be estimated with flux monitors[3] but still introduce inaccuracies.

NEURAL NETWORKS

The same data gathered for medium-resolution imaging were also used to generate neural networks to determine positions of localized uranium. The goal was to assign the radial coordinate correctly to one of four radial zones and to assign the height coordinate correctly to one of five vertical zones. Separate neural networks were developed for the radial and height coordinates after we found that a single network did not give satisfactory results.

Back-propagation networks worked well in assigning the radial coordinate, even with counts from tubes summed so that the number of banks was eight. (The imaging algorithms could not have functioned well under this condition.) The small number of outputs had the advantage of reducing the size of the network and therefore the amount of data needed to "train" the network. A single "hidden" layer of only three processing elements was needed. The number of outputs was four, one for each radial zone. Success rates of 99% were obtained; the rare failures differed from the correct radial zones by only one zone.

Counts from top and bottom detector banks were again used to determine the height coordinate. The back-propagation network was ineffective in this case, but the radial-basis function was successfully applied. The ratio of top-to-bottom banks' counts (T/B) can vary widely for extreme positions of a source (or a weak source) within a moderating matrix, so an average-difference ratio (T-B)/(T+B) was also used that is better behaved.

The iterations needed to train a neural network are done in advance of any assays of unknown drums and the resulting network is added to the shuffler's software. The time needed to train does not affect the time to complete an assay. In practice, the training time has been only a few minutes and the application of a network to new data is a straight forward, quick calculation.

SUMMARY

When the matrix in a 55-gallon waste drum is nonmoderating (for example, hydrogen density < 0.003 g/cm^3), the shuffler's active-assay result is unaffected by the position of the uranium within the drum. The assay accuracy is limited by the calibration standards.

With hydrogen densities as high as 0.01 g/cm^3, as would be found in a drum with 30 kg (66 lbs) of paper (a rather high value), active assays are practical, but accuracies would be greatly improved with a reduction in the average energy of irradiating neutrons from the ^{252}Cf source. But this approach is acceptable only if it is known that an increase in self-shielding will be insignificant or at least can be tolerated.

The minimal self-shielding property of the shuffler can be retained if the position of localized uranium within a drum is determined from either the shuffler data or from an imaging instrument (such as a tomographic gamma-ray scanner). This approach is not limited to hydrogen densities of 0.01 g/cm^3, but rather by the value of 0.05 g/cm^3 where neutron transport becomes too severe to obtain reliable signals. Three different techniques have been investigated to deduce positions from shuffler data.

A comparison of techniques for making position corrections includes these topics:

- **Accuracy**: how good is the correction? The hardware solution is known to work well where it can be applied, but the effect on self-shielding has not been fully explored.
- **Sensitivity**: how low can the count rate be and still allow a useful correction? All of the techniques except the hardware sleeve perform better with high count rates, but waste will generate low count rates; when the fissile mass might be significant, is the signal sufficiently strong to make a correction? This important issue has been only partially studied for some of the techniques described here so a detailed comparison will be given on a later occasion.
- **Distributed Sources**: how well can more than one source be identified? The medium-resolution imaging technique worked well under ideal conditions (strong signals from each detector tube and a nonmoderating matrix) but not so well under more difficult conditions. This topic has not yet been explored with the other techniques.
- **Implementation**: how easily can a technique be implemented in existing and new shufflers? The hardware addition of a moderating sleeve around a drum is the simplest to implement and in fact is already being done, although self-shielding increases must always be kept in mind. The detector bank variance technique has been applied to a current shuffler, but it would probably be more effective with the modest hardware change of doubling the number of detector banks and signals. The medium-resolution imaging needs a compromise between the number of detector banks and the signal strength from each bank; if only a few banks can be used to have sufficient precision in the counts, the resolution of the images will degrade. Neural networks have been shown to give low-resolution positions with the modest hardware change of doubling the number of detector banks and signals.

More work on these techniques and issues are in progress and a more informed consideration of them will be done in the future.

REFERENCES

1 P. M. Rinard, "Shuffler Instruments for the Nondestructive Assay of Fissile Materials," Los

Alamos National Laboratory report LA-12105 (May 1991).

2 P. M. Rinard, E. T. Sadowski, and F. G. Armstrong, "Billet Shuffler Calibration Data and Interpolation Scheme," Los Alamos National Laboratory report LA-12351-MS (July 1992).

3 P. M. Rinard, E. L. Adams, H. O. Menlove, and J. K. Sprinkle, Jr., "The Nondestructive Assay of 55-Gallon Drums Containing Uranium and Transuranic Waste Using Passive-Active Shufflers," Los Alamos National Laboratory report LA-12446-MS (November 1992).

4 T. D. Reilly, N. Ensslin, H. Smith, Jr., and S. Kreiner, Eds., *Passive Nondestructive Assay of Nuclear Materials*, NUREG/CR-5550 (U.S. Nuclear Regulatory Commission, Washington, D. C., 1991).

5 T. H. Prettyman, R. J. Estep, and G. A. Sheppard, "Development of a Tomographic Instrument for Gamma-Ray Nondestructive Assay," Los Alamos National Laboratory document LA-UR-93-2580, Rev. 1 (1993).

6 M. M. Pickrell and P. M. Rinard, "Matrix and Position Correction of Shuffler Assays by Application of the Alternating Conditional Expectation Algorithm to Shuffler Data," *Nucl. Mater Manage.* **XXI** (Proc. Issue) 235-242 (1992).

7 A. C. Kak and M. Slaney, *Principles of Computerized Tomographic Imaging* (IEEE Press, The Institute of Electrical and Electronics Engineers, Inc., New York, 1987), pp. 283-284.

8 W. H. Press, B. P. Flannery, S. A. Teukolsky, and W. T. Vetterling, *Numerical Recipes, The Art of Scientific Computing* (Cambridge University Press, Cambridge, U.K., 1992), pp. 312-326.

9 L. A. Shepp and Y. Vardi, "Maximum Likelihood Reconstruction for Emission Tomography," *IEEE Transactions on Medical Imaging*, **MI-1**(2), 113-122 (October 1982).