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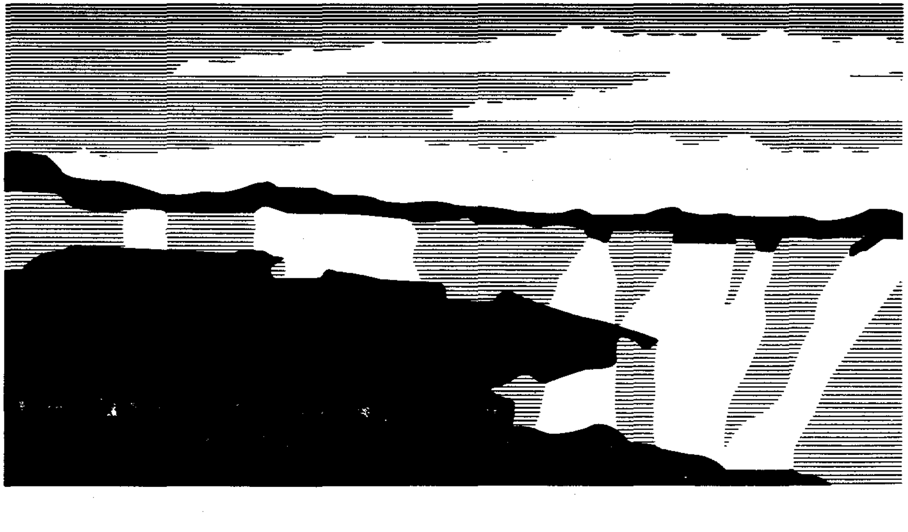

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IMPROVING SHUFFLER ASSAY ACCURACY*

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ABSTRACT

Drums of uranium waste should be disposed of in an economical and environmentally sound manner. The most accurate possible assays of the uranium masses in the drums are required for proper disposal. The accuracies of assays from a shuffler are affected by the type of matrix material in the drums. Non-hydrogenous matrices have little effect on neutron transport and accuracies are very good. If self-shielding is known to be a minor problem, good accuracies are also obtained with hydrogenous matrices when a polyethylene sleeve is placed around the drums. But for those cases where self-shielding may be a problem, matrices are hydrogenous, and uranium distributions are non-uniform throughout the drums, the accuracies are degraded. They can be greatly improved by determining the distributions of the uranium and then applying correction factors based on the distributions. This paper describes a technique for determining uranium distributions by using the neutron count rates in detector banks around the waste drum and solving a set of overdetermined linear equations. Other approaches were studied to determine the distributions and are described briefly. Implementation of this correction is anticipated on an existing shuffler next year.

INTRODUCTION

The uranium contents of waste drums must be known as accurately as possible to ensure their proper handling and disposal. A nondestructive assay for the uranium mass that is too low could result in a drum being buried as low-level waste when in fact it is not; an assay that is too high could force an unnecessary and expensive treatment of a drum.

Shufflers are one type of instrument being used to nondestructively assay for uranium in 55-gal. drums of waste. They induce fissions in the uranium with ^{252}Cf neutrons and then count the resulting delayed neutrons.[1-5] They also assay for plutonium through passive coincidence counting of neutrons from spontaneous fissions; the active mode described for uranium could also be used for plutonium, but the passive mode is more sensitive because the background rate is much lower.

The accuracy of an assay is always affected by the closeness of the match between the calibration standards and the drums. This is the most important source of inaccuracy for drums whose matrices have little or no moderating materials (primarily hydrogen). For such drums, the distribution of the uranium within the drum does not affect the assay accuracy.

But when the matrix has a hydrogen density of 0.002 g/cm^3 or more, the delayed neutron count rate will change with the uranium's position within the matrix.[1] Neutrons from the ^{252}Cf source that reach the interior of a drum have a lower average energy because of collisions with hydrogen and therefore have increased probabilities of inducing fissions. The delayed neutrons released from deep within the drum are less likely to be detected than those released near the surface, but the overall effect is an increase of count rates from the center of drums filled with paper, rubber gloves, or other such common wastes. Only when hydrogen densities are much higher is the count rate from a drum's center smaller than from the surface.

For matrices such as paper, the effects of the hydrogen are strong enough to cause a 75% error in an assay if the calibration was done for an average position but the uranium is actually far from the average position. This problem can be eliminated by placing a thin polyethylene moderating sleeve around the drum; the average energy of the neutrons entering the drum is reduced by the sleeve and the gradient of neutron energies throughout the drum is much smaller than without the sleeve. Inaccuracies are cut from 75% to 15% for the worst cases.

But the sleeve has the potential of introducing a new cause for inaccuracy: self-shielding. Lower-energy neutrons are less able to penetrate uranium; the surface of the uranium can shield the interior of the uranium from the interrogating neutrons and the assay result is proportional to the surface area instead of the mass. If the uranium is in the form of very small particles (about 1 mg or less) that are well dispersed, the self-shielding will not be a problem and the sleeve will still be very beneficial.

But there remains the problem of a drum with a moderating matrix and uranium that can be self-shielding with low-energy neutrons. Matrices with unknown hydrogen densities and unknown uranium self-shielding should be treated as part of this remaining problem. For

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these cases another approach is needed, and that is the subject of this paper.

Without a sleeve, we know that the assay value varies with the position of the uranium within a drum containing a moderator. From previous measurements we know how the results vary for a given matrix.[1] So if we can determine the uranium's distribution we can correct the count rate for the distribution.

Our first step toward this goal verified that positional information does exist in the delayed neutrons even after scattering by the matrix (e.g., paper) and the detector banks (polyethylene).[6] Additional data were then taken to generate response functions for use in the computational tests and simulations reported here.

Other instruments could be used to generate uranium distribution information for the shuffler. The most notable for low-density matrices is the tomographic gamma-ray scanner (TGS).[7] But when the matrix density is too large for gamma-ray transmission, the information from the shuffler's own neutrons must be used. Furthermore, the purchase of a second, somewhat expensive (perhaps $400K), instrument may not be practical for all users.

CANDIDATE ANALYSIS TECHNIQUES

All of the analysis techniques described here utilize delayed-neutron count rates from banks that surround the assay chamber. The typical geometry shown in Fig. 1 has six side banks plus a top and a bottom bank; each bank has 6 to 8 neutron detector tubes whose outputs are normally combined into a single output. The tubes' outputs need not be combined so completely and each bank could give two or three outputs for more spatial resolution. However, as resolution improves, the count rate per signal channel gets smaller and either counting precision suffers or count times lengthen.

The waste drum is divided into many cells of equal volume; such a division is indicated in Fig. 1. The delayed-neutron source strengths in these N_S cells are S_j; in practice, most of these are likely to be zero, but this cannot be assumed. These sources generate N_R measured count rates R_i from the N_B detector banks. The transport function from the cells to the banks has the element T_{ij} which gives the count rate in bank i that is caused by a source mass S_j in cell j.

$$R_i = \sum_{j=1}^{N_S} T_{ij} S_j, i = 1,2,...,N_R \qquad (1)$$

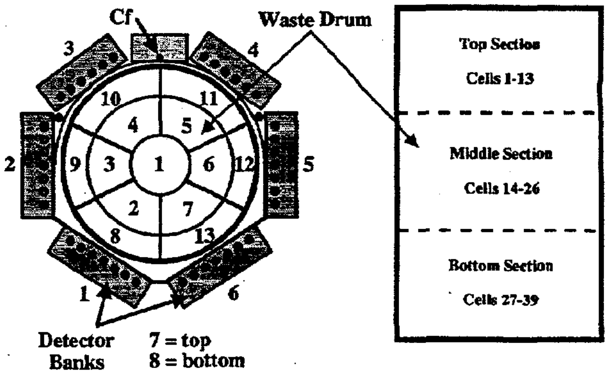

Fig. 1. Two cross sections of a 55-gal. drum are shown. The vertical section on the right has three sections of equal volume. The horizontal cross section on the left is divided into 13 cells of equal volume; the cell labels shown are for the top vertical section. Six lateral detector banks along with top and bottom banks surround the drum to count delayed neutrons. A correction factor for a ^{235}U *position is a function of the vertical section and the radius of the cell in which the* ^{235}U *is located; for example, the correction factors for cells 2 and 3 are the same, but those for cells 2 and 8 (or 2 and 21) are different.*

This is a set of N_R equations for the N_S unknown S_j. The transport function T_{ij} must be established through measurements with standards, as done in Ref. 1. The units of R_i are counts/s and of S_j are g of ^{235}U, so the units of T_{ij} are counts/s/g ^{235}U.

The usual assay adds all the counts from all sources to get an overall count rate, $R = \sum_i R_i$, and applies a calibration curve for a homogeneous distribution of the ^{235}U. This leads to inaccuracies when distributions are not homogenous.

A unique solution to Eq. (1) for the S_j is possible only if $N_R \geq N_S$. If there are only eight detector banks ($N_B = 8$) it might seem that N_R can be no larger than eight and each cell must be one-eighth of a drum. But the drum can be measured at different orientations relative to the ^{252}Cf source to increase N_R. In fact, different orientations are needed to get an R equal to the case of the continuously rotating drum. A set of six counts with the drum rotated 60° between them is equivalent to the more normal case of a continuously rotating drum.[1] The assay result based on R is then to be corrected from the relative values of S_j from Eq. (1).

Using N_O orientations increases the number of measurements to $N_R = N_O N_B \geq N_S$. The example in Fig. 1 implies $N_O = 6$ and $N_B = 8$, so N_S can be as large as 48. But this example only has $N_S = 39$ because the drum is divided in three layers, each with 13 cells, so Eq. (1) is over-determined and solvable. This geometry is a compromise among spatial resolution, count time, and analysis time. More resolution could be obtained with more cells and more orientations, but count and analysis times would have to grow. Any less resolution would not improve assay accuracy enough to be worthwhile.

Analysis Principles

It might appear that Eq. (1) could be simply treated as N_S equations with N_S unknowns, after ignoring the "extra," $N_O N_B$ - N_S, measured count rates. However, the statistical fluctuations inherent in nuclear decay emissions arise in these count rates and are not negligible for waste quantities of uranium. The solution to Eq. (1) as an exact set of equations is very sensitive to these fluctuations; while the solutions are mathematically correct, they are physically wrong by such large amounts as to be useless.

The full set of over-determined $N_O N_B$ equations can be retained and a best fit for the N_S unknowns found. Three techniques for following this path have been studied.

- Neural networks can mimic Eq. (1) by developing a general, but very complex relationship, among the S_j and R_i that is not based on any physically determined transfer function T_{ij}.

- The Linear Programming and Simplex algorithm[8] can be effective when it is known that only a small number ($N_O N_B$ - N_S) of cells might hold uranium.

- The Conjugate Gradient (CG) technique[8] does not limit the number of cells that may contain uranium. This is a variation on the method of steepest descent but is more computationally efficient.

A very large number of parameters are involved in a neural network for this problem, so the data used to determine their values (to "train" the network) must be very extensive. After testing networks for uranium present in only one or two cells and finding very limited success, we believe it is impractical to attempt a network for more general cases. This problem's number of inputs and outputs is apparently too large for the neural network approach. The potential advantage of a neural network is the speed with which the correction factor would be calculated, but the accuracy of such a correction factor seems to be too poor to be useful.

The Linear Programming and Simplex method is not general enough for this problem because a limited distribution of uranium cannot be assumed. This approach would not even be correct for the important case of a homogenous distribution of uranium.

The CG algorithm seems to be the most practical way to solve Eq. (1). The values of S_j are found that minimize the standard chi-squared function:

$$\chi^2 = \sum_{i=1}^{N_R}\left[\left(R_i - \sum_{j=1}^{N_S} T_{ij}S_j\right)/\sigma_{Ri}\right]^2 . \qquad (2)$$

The solution is an iterative process and convergence may require a large number of iterations, but with the speed of today's computers and some techniques to accelerate convergence the calculations are practical. The rest of this paper will concern the CG technique only.

Tests of the Conjugate Gradient Method

Measurements have been made with the multiple-orientation procedure to determine the T_{ij} for a paper-filled drum and the geometry in Fig. 1. Six orientations of the drum gave 48 responses from the 8 detector banks and 39 source cells divided equally among three vertical layers. Measurements could then be simulated on a computer to develop and test the CG procedure.

The following is one example of testing the CG procedure. All the S_j were set to zero except S_{16} = 100 g of ^{235}U. Precise R_i were calculated from Eq. (1) and then made realistic by adding statistical terms from normal distributions appropriate for the count rates. Uranium masses S_j for these imprecise R_i were calculated from Eqs. (1) and (2) using the CG procedure with the results shown in Table I. The initial values of all the S_j were zero and about 300 iterations were required for convergence.

The solution in Table I is mathematically precise; the values of R_i calculated by Eq. (1) are all within 1% of the given detector bank responses. But the solution cannot be physically exact because the statistical fluctuations made the given responses inexact. The small S_j values are ignored as not indicating any uranium and a correction factor is to be based on the dominating value for cell 16; its 72 g of ^{235}U is 3.6 times larger than for any other cell.

Cell 16 is near the r = 20 cm and z = 40 cm position of Ref. 1. At that location the measured count rate is known to be 1.77 times the count rate for the average position, so a normal assay based solely on $\Sigma_i R_i$ and a calibration curve for a homogenous distribution would give about 177 g of ^{235}U instead of 100 g. But the data in Table I indicates that the only strong source is in cell 16, so a correction factor of 1/1.77 would be applied and the final assay result would be near the correct value of 100 g of ^{235}U. Even though the value of S_{16} is inaccurate (72 g is far from 100 g), its large magnitude

TABLE I: Example Conjugate Gradient Solution for 100 g of ^{235}U in a Paper-Filled Drum

Ideal Solution: S(16) = 100, all others zero.
$\chi^2 = 14.1$

Cell Index (*j*)	Source Mass *S* (*j*) (g ^{235}U)	Cell Index (*j*)	Source Mass *S* (*j*) (g ^{235}U)
1	4.8	20	0.0
2	2.6	21	-1.2
3	0.6	22	12.6
4	-8.9	23	6.0
5	-2.9	24	5.1
6	6.0	25	1.1
7	0.1	26	4.3
8	-1.7	27	-4.1
9	3.1	28	-9.3
10	1.0	29	20.9
11	2.1	30	14.0
12	-4.1	31	-5.4
13	-5.0	32	-0.4
14	1.7	33	-7.1
15	1.0	34	6.5
16	**72.2**	35	-11.9
17	13.8	36	-9.2
18	-10.0	37	-5.6
19	-1.4	38	6.0
		39	4.1

relative to the other values indicates the uranium position and this leads to a useful correction factor.

IMPLEMENTATION

To implement the scheme outlined above requires changes to the hardware and software of an existing shuffler, but they are not very complicated. It is expected that a shuffler will be modified next year and serve as a testbed for demonstration and further development.

Hardware Changes

A stepping motor is needed to produce accurate orientations of a drum during an assay. Most previous shufflers used an analog motor, which does not provide the control needed. Stepping motors have already been used in two existing shufflers so this presents no new problems. The stepping motor controller already used to position the ^{252}Cf source can control as many as four motors, so no new controller is needed.

If greater resolution were desired, additional signal cables would be needed from the detector banks to the electronics rack. Another multichannel scaler would also be needed to receive the signals from the new cables. But the impact of this higher resolution on count rates and count times should be evaluated relative to the assay requirements. Counts rates from individual banks will be lowered by the increased resolution, so either the count time has to be increased or sensitivity sacrificed. Count times may have to be increased in any case to ensure sufficiently accurate solutions to Eq. (1).

Software Changes

Controlling the orientation stepping motor presents no software problems; the number of steps between orientations is easy to calculate and command. If more signals are used, a second multiple-channel scaler must be controlled through another serial port, but this is not difficult either.

The data analysis based on Eqs. (1) and (2) and the CG technique is the most extensive addition to existing software. After we determine the uranium distribution in a drum, calculating a correction factor for the delayed neutron count rate requires calibration data, but this is simple and quick to perform.

Correction Procedure

Experience with this technique has been limited during this development phase, but an initial procedure for applying the correction can be stated and then modified with further experience.

An uncorrected assay value is calculated in the usual way from the total delayed neutron count rate and a calibration curve for a homogeneous distribution of uranium. There are two cases when a correction need not be even considered:

- If the uncorrected assay value is statistically zero, no correction will change it so none should be calculated.
- If a plant is only interested in screening drums for more than 150 g of ^{235}U (as an example), any value less than 75 g of ^{235}U may be certain to represent less than 150 g of ^{235}U for the matrix and a correction is not needed. If the uncorrected value is 200 g of ^{235}U, it is likely that the actual mass is more than 150 g of ^{235}U and a correction need not be applied unless it would be useful to know how far the drum is over the 150 g limit.

When a corrected assay value is needed, this procedure could be followed by a computer program.

Step 1. Calculate the S_j from Eqs. (1) and (2).

Step 2. If only one of the S_j is much larger than the others (as in Table I), the correction factor for that one location is applied.

Step 3. If more than one of the S_j are larger than the others, the average of the correction factors (weighted by the S_j) for the corresponding locations is applied.

Step 4. If all or most of the S_j are large, a homogeneous distribution is indicated and no correction is made.

SUMMARY

The largest source of inaccuracy in assays from shufflers is generally a poor match between calibration standards and the drums being assayed. Self-shielding can also be a source of inaccuracy for the larger waste quantities of uranium. Shuffler assay accuracy is already very good for non-moderating materials (metals) and moderating materials (paper, gloves) with insignificant self-shielding in waste drums.

The use of a moderating sleeve around a drum with a moderating matrix virtually eliminates the variation of the assay result with the distribution of uranium within the drum.

But the sleeve should not be used when the matrix is moderating and self-shielding is a concern. The delayed neutron count rate now varies with the position of the uranium within the drum and a correction must be based on determining that position (or positions). The neutron count rates in the detector banks surrounding the drums can be used for that purpose.

The solution to Eqs. (1) and (2) by the conjugate gradient technique can reduce the inaccuracy by about a factor of three. The solution will be more accurate when the count rates in the detector banks are more precise, so there is a lower limit to the amount of uranium needed before a correction is possible. This limit for a drum of paper seems to be about 10 g of ^{235}U, based on computer simulations. Further optimization of this correction technique will be done through simulations and measurements with a shuffler modified to perform the new assay procedure.

ACKNOWLEDGMENTS

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