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Title: **IMPLEMENTING A TECHNIQUE TO IMPROVE THE ACCURACY OF SHUFFLER ASSAYS OF WASTE DRUMS**

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IMPLEMENTING A TECHNIQUE TO IMPROVE THE ACCURACY OF SHUFFLER ASSAYS OF WASTE DRUMS*

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ABSTRACT

The accuracy of shuffler assays for fissile materials is generally limited by the accuracy of the calibration standards, but when the matrix in a large drum has a sufficiently high hydrogen density (as exists in paper, for example) the accuracy in the active mode can be adversely affected by a nonuniform distribution of the fissile material within the matrix. This paper reports on a technique to determine the distribution nondestructively using delayed neutron signals generated by the shuffler itself. In assays employing this technique, correction factors are applied to the result of the conventional assay according to the distribution. Maximum inaccuracies in assays with a drum of paper, for example, are reduced by a factor of two or three.

INTRODUCTION

Shufflers in the active mode perform assays by irradiating fissile materials with neutrons from a ^{252}Cf source, then counting delayed neutrons after the source has been quickly removed into a shield, and finally deducing the mass of the fissile material from the count rate. The ^{252}Cf source is shuffled into and out of the shield a number of times during an assay to reach a desired detection limit or count-rate precision.[1] Shufflers in the passive mode perform assays on materials with spontaneous fission rates sufficiently high for coincidence counting.[2] Uranium is only assayed in the active mode; plutonium is generally assayed in the passive mode, but may be assayed in the active mode. If a container has both uranium and plutonium, both active and passive modes are needed to determine the masses of the individual elements. If the only fissile material is plutonium but another spontaneous fission material (e.g., ^{244}Cm) is present, the usual passive assay for plutonium (based on coincidence counting) is confused by the second material, but the active mode signal is from the fissile material only (with the spontaneous-fission neutrons contributing to the background).[3] Shufflers in these modes of operation have been applied to waste quantities (subgram to multigram) and production quantities (up to kilograms).[4]

This paper deals with data from a shuffler in the active mode, but the work can be applied to the passive mode as well. However, the changes in passive count rates with position within a drum are much less than with delayed-neutron count rates, so the need for a position correction is slight.[4] Waste quantities of uranium are emphasized, but the larger masses in process materials would only make it easier to apply the positioning technique.

Shufflers are generally precise instruments with an accuracy limited by the accuracies of the calibration standards. However, in one set of circumstances the accuracy is adversely affected by the matrix associated with the fissile material (generally uranium). Here are the conditions that lead to the problem; I will use a 55-gal. drum with 21.3 kg of paper[4] as an example throughout the following discussion and numerical examples.

(a) A problem is likely with a large container that has a large amount of hydrogenous matrix (such as the 55-gal. drum of paper). If the container is too small to have much matrix, or there is no matrix (e.g., production oxides), or the matrix is nonhydrogenous (e.g., scrap iron or steel), there is no problem and accuracy is limited by the accuracy of the standards. But if there is a hydrogenous matrix throughout a large container, the neutron flux and energy spectrum change with depth into the drum. This means that fission cross sections vary with position within the container, and the delayed neutron production rate varies with position. Therefore, the delayed neutron count rate depends on where the fissile material is located within the container. If the calibration for the paper drum assumed a uniform distribution of the fissile material, a localized distribution may be assayed with an inaccuracy between 0.5 and 2.0 times the true mass.

(b) If it is known that the fissile mass or density is too low for self-shielding to be a serious problem (e.g., a few grams of fissile material spread over a liter of matrix), the problem with a hydrogenous matrix can be mitigated by simply placing a polyethylene sleeve over the container. The moderator

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in the sleeve greatly reduces the effect of the moderator in the matrix and assay accuracy is again good.[4] The results from the drum of paper mentioned above would now be between 0.75 and 1.25 times the true mass. The fissile mass or density may be known from the generation of the material in the container or deduced from a provisional shuffler assay where an estimate of the fissile mass is produced and flux monitors indicate the degree of moderation by the matrix. However, if self-shielding is likely to be a problem or if the self-shielding is unknown, the sleeve should *not* be used because its moderation of neutron energies would further aggravate self-shielding.

(c) If neutron energy moderation by the matrix is too strong to ignore but self-shielding could be a serious problem with the polyethylene sleeve, an assay should be done without the sleeve and an appropriate correction factor applied for the position(s) of the fissile material within the container.

This paper is primarily concerned with the most complex case (c), but also offers an alternative to the sleeve of case (b). By measuring the position(s) of the fissile material, appropriate correction factors to the measured count rate can be applied. The sleeve can be avoided for case (c) and may be superseded in case (b).

It is not difficult to use standard drums to determine the correction factors for fissile materials at known positions,[4] but it is quite another task to find the position(s) of the fissile material within a waste or production container. The relative count rates in detectors surrounding the container are used here to determine the position(s) of the fissile material.[5,6]

An assay process with the position-correction option is outlined in Fig. 1. It assumes that the sleeve is not used because the position-correction technique is always better (it has about the same measurement time and there is no increase in self-shielding). In this figure it is further assumed that the time needed to do the position correction is longer than a conventional assay, so the position-correction measurements are done only if necessary. (If it is known that a drum will need the position correction, then the conventional assay can be skipped and the position correction data used for both purposes.)[4] If the position correction measurements take about the same time as a conventional assay (as is indicated by the results later in this paper), then a simpler procedure is to skip the conventional assay step for all drums and do only position-correction measurements. The data can still be

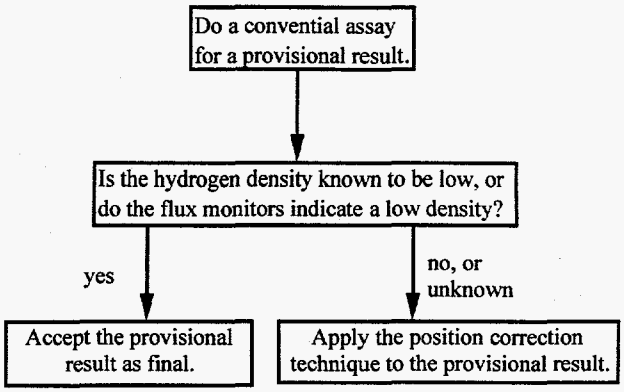

Fig. 1. The shuffler itself can be used to determine if the position-correction technique is needed, but any prior knowledge would save time. This decision tree guides the user or the shuffler's computer through the issues and results in the best assay technique to use on the drum in question. If the time needed for the position correction measurements is about the same as a conventional assay time, the conventional assay can be skipped and its results gathered from the position correction measurements.

analyzed as if this was a conventional assay (with no loss of precision or accuracy)[4] and then the position correction can be applied if warranted by the flux monitor data or packaging information.

POSITION DETERMINATION SCHEME

In the passive mode, the banks nearest the fissile material (plutonium) will have the highest count rates. This is also true in the active mode but there is the additional complication that the source strength depends on the position of the fissile material (uranium) relative to the ^{252}Cf source. The volume in the container is divided into cells of equal volume; the specific division scheme used in the tests described below is shown in Fig. 2, but others could be used. The 39 cells in Fig. 2 are the smallest number that will still improve assay accuracies by at least a factor of two.

The count rate R_i in bank i depends on the source strength S_j of the material in cell j; a transport function T_{ij} shows the count rate a unit of the fissile material in cell j produces in bank i, for a particular ^{252}Cf emission rate.

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

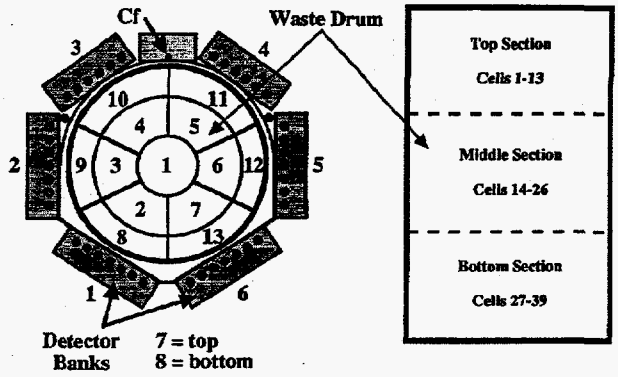

Fig. 2. A 55-gal. drum has been divided into 39 equal-volume cells by forming 13 regions in a horizontal plane for each of 3 vertical sections. Delayed neutrons are counted with six lateral detector banks plus banks above and below the drum.

N_S is the number of cells and N_R is the number of count rates from the detector banks. This equation is solved using the conjugate gradient technique,[7] which finds the S_j through an iterative approach of minimizing the chi-squared function.[6] The correction factors to be applied to each cell are determined through measurements (as in Ref. 4) or calculations.

Solving Eq. (1) for the S_j gives estimates of the masses of fissile material in the cells. However, the T_{ij} have unavoidable inaccuracies from experimental determination, as do the measured count rates R_i. Only a certain level of inaccuracy will still give usefully accurate solutions for the S_i. The measurement imprecisions can always be reduced by using more shuffles (irradiations and delayed-neutron count cycles), but at some point practicalities limit the precision that can be reached.

In any case, the masses in the cells S_j will have uncertainties from counting statistics and from the process of solving Eq. (1). For relatively high fissile masses, the S_i will be good estimates of the masses. Below a certain fissile mass, however, the S_j may not be accurate enough to use as assay results because of measurement imprecisions, but they can still indicate the positions of the fissile material and be used to correct the result of the conventional assay. The cutoff point between these two modes depends on the moderating ability of the matrix (generally the hydrogen density) and the fissile isotope.

The application of the position-determination scheme described here has been done with a shuffler for 55-gal. drums[4,7] holding uranium, but it is sufficiently general to apply to any shuffler and also to the passive mode. These 55-gal. drum shufflers have six detector banks around the side of a drum, plus a bank above and below the drum. It is not useful to use a number of cells equal to the small number of detector banks, so the number of measurements is extended by giving a drum six orientations relative to the ^{252}Cf source by successive rotations of 60°. This allows as many as 48 cells, but a reasonable division of a drum uses only 39 cells.[6] Therefore, Eq. (1) in this case is a set of 48 equations for 39 unknowns.

By using more detector banks, with fewer tubes in each bank, the number of measurements is increased at the expense of measurement precision in each bank. A compromise must be selected, and Fig. 2 shows the compromise selected for this study. It matches the waste-drum shufflers installed in DOE facilities and therefore requires only simple modifications to implement. The spatial resolution needed for the situation shown in Fig. 2 is on the order of 10 cm.

A QUESTION OF SYMMETRY

A drum has a vertical axis of rotational symmetry and the matrix in a drum may have the same symmetry. If the distribution of uranium within such a matrix shares this symmetry, can this positioning scheme distinguish among these axially symmetric cases: a "line" of uranium along the axis, a "hollow cylinder" of uranium around the axis, and a uniform distribution throughout the drum volume? If not, appropriate correction factors for the conventional assays cannot be determined.

However, the answer is "yes" because the ^{252}Cf source position is asymmetric and the drum does not rotate during a position measurement. With the "axial line" of uranium (the combination of cells 1, 14, and 27 in Fig. 2), count rates in the lateral banks will always be nearly equal. With the "hollow cylinder" (such as cells 8-13, 21-26, and 34-39) or a uniform distribution, count rates in banks near the ^{252}Cf source will be larger than in the others; these two cases are distinguished by the quantitative differences in count rates from banks at different distances from the ^{252}Cf source. The solutions to Eq. (1) do identify these cases and give the correct distribution of fissile material.

This feature does not apply to a passive instrument because there is no asymmetric ^{252}Cf source involved; the three distributions in the preceding paragraph are then indistinguishable. A matrix-correction factor would have to be conservatively large and an assay would then give an upper limit to the mass of spontaneously fissioning material in the drum. Fortunately, passive coincidence

count rates are much less dependent on position within matrices than are the delayed-neutron count rates,[4] so the issue is much less important.

IMPLEMENTATION

The 55-gal. drum shuffler at the Los Alamos National Laboratory (LANL) Chemistry and Metallurgy Research (CMR) facility was available for this study.[8] The shuffler's design is hardly optimal for determining the distribution of uranium in a drum, but it is adequate. Furthermore, there are a total of six such shufflers in the DOE complex and it is important to determine the performance improvement after only a minimal upgrade. There are a few others with different designs that might profit from this work, and any new design should incorporate positioning features from the beginning.

The following modifications were made to the CMR shuffler. (a) A drum rests on a turntable which has traditionally been turned continually during an assay by an analog motor; such a motor cannot make an accurate rotation of only 60°. The analog motor was replaced by a stepping motor that does allow such control; the rest of the rotation mechanism was unchanged. The stepping-motor controller used to move the ^{252}Cf source has the capability of driving four motors, so a new controller was not needed. (b) Some new signal cables for the motor system were routed through the shuffler body without removing more than some shrouds lining the assay chamber. The signals from the eight detector banks were already brought to a 12-channel scaler and counted individually, so no change was needed to the detection electronics. (c) The software was adapted to control the stepping motor instead of the analog motor, but the code for this had already been developed for another shuffler and the change was easily made. At this point, the shuffler was in the same operational state as before any modifications and it was put back into routine service for the CMR facility. A prototype shuffler code was written to rotate a drum in 60° steps while six irradiations and counts are performed with the drum stationary.

The more recent shufflers built at Los Alamos already use stepping motors for the turntable, so they would need no hardware changes at all to use a position-correction scheme. Any of the other 55-gal. drum shufflers of the same general design as the CMR shuffler (of which there are five) could have its hardware modified as easily as at the CMR facility.

TESTING

A paper-filled 55-gal. drum is an excellent case for development and testing. Our drum[4] has 21.3 kg of office scrap paper, giving a hydrogen density of about 0.007 g/cm^3. This matrix is commonly encountered and has a high hydrogen density relative to most other waste matrices.

In last year's work[6] the T_{ij} were measured with the same paper drum but the R_i were simulations based on statistically varied rates adapted from the T_{ij}. These were used to initially test the process of solving Eq. (1). In the present work the R_i were measured independently of the T_{ij} using various different samples of uranium.

The ^{252}Cf source in the LANL CMR shuffler had a mass of 350 μg and therefore a neutron emission rate of 8.19×10^8 n/s. A new source for such a shuffler is 500 to 550 μg, so the source used here happened to have about an average emission rate (over the useful life of a source). Better results will be obtained with a larger source and worse results with a smaller source, unless measurement times are extended in compensation.

The transport function T_{ij} was determined by placing a known mass of ^{235}U in each of the 39 cells and rotating the drum into six positions separated by 60°. The ^{235}U was in the form of a fluoride absorbed on alumina beads. A can of seven capsules had 32.69 g of ^{235}U (at 94% enrichment). The uranium was dispersed through a volume of about 400 cm^3, so the ^{235}U density was about 0.082 g/cm^3. This dispersed mass of ^{235}U was used to minimize self-shielding effects while still allowing practical count rates from the detector banks. Previous experience with these capsules has indicated that the self-shielding effect on the count rate is about 10%.

At each orientation of the drum, the ^{252}Cf source was shuffled into the assay chamber 20 times for a total irradiation time of about 235 s. The 20 count times at each orientation summed to about 180 s. When the ^{235}U can was near the ^{252}Cf source, the counts in the nearby detector banks were about 10^4. The lowest counts came with the ^{235}U far from the ^{252}Cf and also far from a detector bank–they were about 2×10^3. Better precision in the T_{ij} values would be obtained by using more shuffles at each drum orientation or more ^{235}U (although self-shielding effects limit the ^{235}U mass). The completed set of measurements was done in less than eight hours.

Combinations of cans and individual capsules were then placed in various cells in the drum and the measured R_i were used to test the ability of Eq. (1) to locate their positions (singly and in combinations). Seven shuffles at each orientation were generally used, which took 16 min. to complete. (A conventional assay generally uses about 32 shuffles and is also done in 16 min., including a 4.5-min. background count. Such parameters are readily controlled by the shuffler's authorized user.) Some exploration was done with fewer and greater numbers of shuffles.

Even with the modest precision in the T_{ij} (from only 20 shuffles) and the R_i values (from only 7 shuffles), the results from Eq. (1) were good enough to greatly improve the accuracy of the conventional assay. Table I summarizes some examples of the test combinations, with the ^{235}U masses rounded to the nearest gram. Remember that the goal is not to obtain accurate uranium masses, but accurate positions of the uranium from which correction factors to conventional assays are deduced.

TABLE I
EXAMPLES OF TEST RESULTS

21.3 kg of paper in a 55-gal. drum
350 μg of ^{252}Cf

Example Number	Nonzero ^{235}U Masses in Specified Cells	Number of Shuffles per Orientation	Total Measurement Time (min)	Solution to Eq. (1)
1	5 g in 26	7	16	10 g in 26; < 3 g elsewhere.
2	33 g in 26	7	16	23 g in 26; < 10 elsewhere.
3	12 g in 2	5	11	Inaccurate. Too few shuffles.
4	12 g in 2	7	16	9 g in 12; < 1 g elsewhere.
5	33 g in 12 and 28	10	25	26 and 32 g in 12 and 28; < 4.5 g elsewhere.
6	33 g in 19 and 28	7	16	23 and 32 g in 19 and 28; < 8 g elsewhere.
7	33 g in 33 and 28	7	16	27 and 34 g in 33 and 28; < 4 g elsewhere.
8	33 g in 12, 22, and 28	7	16	27, 30, and 35 g in 12, 22, 28; < 5.5 g elsewhere, except 10 g in 1.
9	1000 g in 6; 12 g in 2	7	16	390 g in 6, 0 in 12; < 100 elsewhere.

Examples 1 and 2 show 5 or 33 grams of ^{235}U in a single cell being located in only a 16-min. measurement. Each solution to Eq. (1) has a dominant value in cell 26 (where the uranium was placed); values for many other cells are close to zero, and values in a few cells are 0.3 to 0.5 times the mass in cell 26 (instead of nearly zero). The magnitudes of these latter values can be reduced with longer count times, but probably not eliminated completely with the rather low resolution with this set of eight detector banks. Nevertheless, the accuracy of conventional assays will be greatly improved by applying an overall correction factor from the largest calculated masses as weighting factors for localized-correction factors. All cells with less than half the calculated mass of the largest calculated mass can be assumed empty for purposes of calculating the overall correction factor.

Examples 3 and 4 involve a can with only 12 g of ^{235}U for different measurement times. In example 3 the measurement time was reduced by using only five shuffles per orientation; the solution did not usefully indicate even the position of the can, so the imprecisions in the count rates were clearly too large for a reliable solution to Eq. (1). A remeasurement with seven shuffles gave an excellent result, with the position of the can clearly shown.

The next three examples (5 through 7) involve two cans, each with 33 g of ^{235}U. One can was moved successively

closer to the other can in cell 28, starting in cell 12 almost diagonally opposite cell 28 and ending in cell 33 adjacent to cell 28. The calculated mass in cell 28 was unaffected by the second mass (and reasonably accurate at 32 and 34 g) regardless of the proximity of the second can. The calculated masses in the second can were also fairly accurate at 23 to 27 g, although the scheme proposed here does not rely on results this accurate. Calculated masses in other cells were nearly always less than 5 g, which are considered to be zero for the purpose of calculating a correction factor.

A third can was added to the previous pair (example 8) but it did not introduce any problem for the solution process. Masses in the three occupied cells were 27 to 34 g while calculated masses elsewhere were much smaller.

Example 9 is an extreme case with both a 1000-g can and a 12-g can in the drum. The 1000-g can was easily positioned; its erroneously low mass is at least partly caused by the much greater self-shielding than was present in the 33-g can used to generate the transport function. Not surprisingly, the smaller 12-g mass was not detected because the count rates were dominated by the emissions from the 1000-g mass.

Results at these masses would be improved by more precise values of the T_{ij} and the R_i measured by using as many shuffles as practical time limitations allow. But it is encouraging that the solutions to Eq. (1) are sufficiently robust to be as accurate as shown in Table I even when the T_{ij} and R_i have relative precisions in the 1% to 3% range from quite practical 16-min measurements.

The accuracies in assaying drums with hydrogenous matrices can be limited to nearly the accuracies of the calibration standards, with smaller contributions from adjustments for hydrogen density differences. The sleeve in case (b) of the introduction would not be needed, but could still be used if its established inaccuracies are nevertheless acceptable and assay time needs to be minimized. Case (c), with the potential of self-shielding, now can be handled with an accuracy as high as in the simpler cases.

FUTURE WORK

The position-determination scheme is ready for inclusion in shuffler software for the active mode and should be put to work on uranium-bearing waste drums and process-material containers where hydrogenous matrices may adversely affect conventional assays. It can also be applied to passive assays based on coincidence counting to help get better accuracies for spontaneous-fissioning elements (typically plutonium). Although the shuffler model used here was designed long before position determination was even considered and its large detector banks are hardly optimal for the task, a quite useful indication of the distribution of the fissile material can be deduced.

The boundaries within which this position determination scheme will work satisfactorily need to be better defined. Computer simulations, benchmarked with the help of the existing shuffler, would allow more flexibility and better definition than measurements on existing materials. The clear difference in the results for examples 3 and 4 in Table I is an indication of how sharp some of the boundary may be. More combinations of a facility's matrices, fissile masses, and measurement times need to be studied through measurements and simulations.

Extensions of this scheme can be done with a new prototype instrument at Los Alamos where the pulses from each detector tube can be counted individually or summed into banks of various sizes. The trade-off between an increased resolution and a lower count-rate precision per bank needs to be investigated further.

ACKNOWLEDGMENTS

Faye Hsue gave us the opportunity to perform the hardware modification to the CMR shuffler and to use it for these test measurements. We greatly appreciate this invaluable cooperation.

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