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CHARACTERIZING AND IMPROVING PASSIVE-ACTIVE SHUFFLERS FOR ASSAYS OF 208-LITER WASTE DRUMS*

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Abstract

A passive and active neutron shuffler for 208-L waste drums has been used to perform over 1500 active and 500 passive measurements on uranium and plutonium samples in 28 different matrices. The shuffler is now better characterized and improvements have been implemented or suggested.

An improved correction for the effects of the matrix material was devised from flux-monitor responses. The most important cause of inaccuracies in assays is a localized instead of a uniform distribution of fissile material in a drum; a technique for deducing the distribution from the assay data and then applying a correction is suggested and will be developed further.

A technique is given to detect excessive amounts of moderator that could make hundreds of grams of ^{235}U assay as zero grams.

Sensitivities (minimum detectable masses) for ^{235}U with active assays and for $^{240}Pu_{eff}$ with passive assays are presented and the effects of moderators and absorbers on sensitivities noted.

1. Introduction

Passive and active neutron (PAN) shufflers at Los Alamos began by assaying cans of mixed-oxide fuels /1/ in 1974 and 208-L waste drums /2/ in 1982. Over 1500 active assays and 500 passive assays have recently been completed on 208-L drums with 28 matrices to characterize and improve the PAN shuffler assays of such drums.

The active neutron interrogation is used to assay uranium. Passive neutron counting gives more precise and accurate assays of plutonium than active interrogation, unless other emitters of neutrons (such as ^{244}Cm) are present. A combination of passive and active assays is needed when both uranium and plutonium are in a drum.

Four results of the present study are discussed in this paper. (a) The inaccuracies caused by localized distributions of fissile materials were quantified for a wide variety of matrices; a correction technique for this localization problem was explored. (b) The use of flux monitor detectors in correcting the matrix effects on neutron transport was improved. (c) A monitoring scheme was shown to detect a hydrogen density so high that an assay result would be severely inaccurate (possibly zero) even though the true mass might be hundreds of grams of ^{235}U localized near the center of a drum. (d) Sensitivities (minimum detectable masses) with the various matrices were determined; techniques to reduce background rates for improved sensitivities were either applied here or will be built into future shufflers.

Other aspects of this study and many more details on the topics in this paper are given in another report /3/.

2. Materials and Methods

The characteristics of the 28 matrices used in the study are presented in Table I. A range of moderating and absorbing abilities is included, plus a few matrices with combinations of moderating and absorbing properties. Many matrices are quite homogeneous, others are semi-homogeneous mixtures of large pieces, and others are deliberately inhomogeneous.

The matrices were well characterized except for two that would be difficult to reproduce with certainty. Peat moss was used to simulate earth, but the moisture content is not easy to control. The simulated junk was simply a loosely packed collection of common items that cannot be specified accurately.

The drums had 2.54-cm diam tubes running from top to bottom at distances of 12, 20, and 25 cm from the central axes of the drums. A capsule of uranium or plutonium was placed in these tubes at vertical heights of 8, 24, 40, 56, and [illegible] cm from the bottom. These radial and vertical coordinates are at the centroids of 15 equal-volume regions within a drum. The drums were rotated continuously during measurements.

The uranium was 94.5% enriched and had 4.67 g of ^{235}U distributed uniformly on alumina pellets in an aluminum cylinder 2.54 cm in diameter and 10 cm long. The plutonium sample was a 29.745-g cylinder of plutonium metal with 93.81% ^{239}Pu and 5.81% ^{240}Pu (1.75 g of $^{240}Pu_{eff}$ for passive coincidence counting); its multiplication was 1.10.

3. Results

Flux Monitor Corrections

Two ^3He tubes monitor the flux of neutrons escaping a drum being irradiated by the ^{252}Cf source during an active assay. A wrap of cadmium screens one tube from thermal neutrons while the other tube is bare; the ratio of their counts thus provides information on the moderating and absorbing properties of the matrix. These tubes are located almost one-quarter of the way around the assay chamber from the ^{252}Cf source, so they respond primarily to interrogating neutrons scattered from a drum's matrix.

The average of the 15 count rates measured throughout each matrix was used to study this correction process. This simulates a homogeneous distribution and separates the flux monitor correction from the fissile material localization problem.

Without any flux monitor correction, the relative standard deviation of count rates from 28 matrices was 80%. Corrections have often been made by dividing by the ratio

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TABLE I. Characteristics of the Matrices			
Matrix	Weight (kg)	Density (g/cm^3) Hydrogen	Boron
Neutral			
1. Empty	0.0	0.0	0.0
2. Iron Chunks	0.0	0.0	0.0
Moderators			
3. Vermiculite	34.0	0.0008	0.0
4. Vermiculite in Liner[a]	34.9	0.00082	0.0
5. Paper	21.3	unknown	0.0
6. Simulated Junk[b]	38.6	--------	0.0
7. Polyethylene Shavings	11.8	0.00857	0.0
8. Concrete Blocks[c]	218.4	0.013	0.0
9. Iron and Polyethylene Chunks	37.2	0.0135	0.0
10. Vermiculite and 29.5 kg of Polyethylene Beads	49.0	0.0212	0.0
11. Polyethylene Tubes	42.6	0.0310	0.0
12. Vermiculite and 59 kg of Polyethylene Beads	78.5	0.0423	0.0
13. Vermiculite and 68 kg of Polyethylene Beads	87.5	0.0488	0.0
14. Peat Moss[d]	50.8	--------	0.0
15. Polyethylene Chunks	91.2	0.0663	0.0
16. Polyethylene Beads	120.2	0.0863	0.0
Absorbers			
17. Raschig Rings[e]	142.0	0.0	>0.001?
18. Vermiculite and 0.3 kg of Borax	34.3	0.00083	0.000172
19. Vermiculite and 0.6 kg of Borax	34.6	0.00087	0.000343
20. Vermiculite and 0.9 kg of Borax	34.9	0.00091	0.000515
21. Vermiculite and 1.2 kg of Borax	35.2	0.00095	0.000687
22. Vermiculite and 1.8 kg of Borax	35.8	0.00103	0.00103
Moderators and Absorbers			
23. Alumina with 28% Water (by weight)[f]	237.7	0.0376	0.0
24. Vermiculite, 68 kg of Polyethylene Beads, 0.21 kg of Borax	88.1	0.0490	0.000120
25. Vermiculite, 68 kg of Polyethylene Beads, 0.42 kg of Borax	88.5	0.0490	0.000240
26. Vermiculite, 37 kg of Polyethylene Beads, 0.22 kg of Borax	61.7	0.0272	0.000133
27. Matrix 26, top; Vermiculite, bottom[g]	48.5	0.025	0.000133
28. Vermiculite, top; Matrix 26, bottom[h]	50.6	0.025	0.000133

[a] This liner was 5.4 kg of plastic.
[b] This matrix had scrap gloves, aluminum, and iron (loosely packed).
[c] The hydrogen density is calculated from a generic elemental composition of concrete.
[d] The moisture content of peat moss was poorly controlled and was probably different when used in the two instruments.
[e] The composition of these particular rings is poorly known. It is presumed that they are simply borated glass, but the response to active interrogation indicates the presence of some moderator also.
[f] This water content is unusually high. In routine use it is usually below 10% (by weight) and the ability to moderate neutrons is greatly reduced.
[g] The drum's bottom half was filled with vermiculite and then the top half was filled with matrix 26.
[h] The drum's bottom half was filled with matrix 26 and then the top half was filled with vermiculite.

of the cadmium-wrapped and bare detectors R; this procedure reduces the relative standard deviation to 53%.

A search for a more effective function of the flux monitor ratio led to the function $R^{p(R)}$, where $p(R)$ is a first-order polynomial with two parameters determined from all 28 matrices. Corrected count rates obtained by dividing $R^{p(R)}$ into the measured count rates had a relative standard deviation of 29%.

A facility can select matrices from Table I that best match its waste (or make new measurements on its own waste) to develop a more specialized $p(R)$ for use on drums whose matrices are poorly known.

General Position Effects

If the fissile material is localized in only a portion of a drum's volume, the assay result will generally depend on the location. As the fissile material becomes more uniformly spread throughout a drum, this position effect disappears.

Calibration is often done with drums of homogeneous contents, but the actual distribution of fissile material within a drum is generally unknown. Ratios of average responses to minimum and maximum responses are then the multipliers of an assay result that give the possible range of the true mass. As the position effect worsens, these ratios depart farther from unity, and the accuracy of an assay result is poorer.

The shuffler currently makes no position correction, but data taken as part of this study point to a possible technique /3/. Assays were done in four stages; during each stage the drum was stationary but rotated 90 degrees from the previous stage. Ratios of counts in opposing detector banks, in combination with the flux monitor responses, are used to estimate the distribution. The average delayed-neutron count rate from four stationary drum orientations was found to be the same as the count rate with a continuously rotating drum, regardless of the initial orientation angle of the drum. This topic will be pursued further, although its accuracy with the smallest waste quantities may be poor.

The position effects measured with the small uranium and plutonium samples in the various matrices will now be described for active and passive assays. These effects are with very localized sources, the most difficult assay situation.

Active Assay Position Effects

Active assays were made with both the uranium and plutonium samples. The results given here are based on the assays of uranium for which active assays are most generally applied.

Moderating Matrices. Figure 1 shows the ratio of average responses to extreme responses plotted against the hydrogen density for moderating matrices (and the empty and iron-filled drums).

With no moderator in a drum, the shuffler's responses are nearly independent of the position of the sample and the average-to-minimum and average-to-maximum ratios are both nearly equal to unity. This shows that there is no bias caused by the shuffler's design.

The hydrogen in a matrix generates responses that can often differ from the average response by a factor of two. The energy spectrum of interrogating neutrons changes with depth of penetration, making the fission rate dependent on the position of the fissile material. The moderator also hinders the transport of delayed neutrons to the detectors.

A drum's assay result (after calibration for uniform fissile distributions) can be multiplied by the average-to-minimum ratio to calculate the largest possible true fissile mass (from a point distribution). Figure 1 shows that this ratio is as large as 2.5 for matrix number 13 with 68 kg of polyethylene beads. The hydrogen density in matrix 13 is 87 kg of water; such a high hydrogen density is unlikely to be encountered in practice.

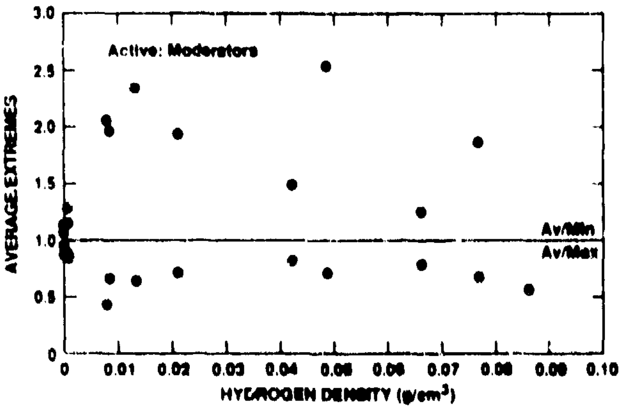

Fig. 1 Ratios of average active responses to the minimum or maximum active responses are shown here for localized fissile materials in moderating matrices (1 through 16 in Table I). The ideal value for these ratios is unity. The values of the two ratios for an instrument indicate the range of possible assay results for a particular matrix.

If a drum's contents are known to be homogeneous, no such multiplier of the assay value should be applied. But the flux monitors lose sensitivity to hydrogen densities above about 0.042 g/cm^3, so it is recommended that the contents of a drum exceeding this limit be repackaged before assaying. This should be a rare event, because this hydrogen density requires 58.8 kg of polyethylene or 75.6 kg of water; it is unlikely that waste drums will have this much hydrogen.

Absorbing Matrices. Vermiculite with different concentrations of borax (matrices 18 through 22 in Table I) produced no position effect. The energies of the interrogation neutrons and the delayed neutrons are too high for significant capture in boron and vermiculite does not have enough hydrogen to thermalize a significant fraction of the neutrons. A drum with this type of matrix is little different from an empty drum.

Moderating and Absorbing Matrices. An absorber becomes effective when mixed with a moderator that lowers the energies of the neutrons. Matrices 23 through 28 are of this type.

A moderator-absorber combination can produce large average-to-minimum ratios because count rates from the drum's interior can be greatly reduced.

The boron density of 0.000133 g/cm^3 has a large effect in combination with the hydrogen density of 0.0272 g/cm^3 (matrix 26). Not enough matrices with different combinations of moderator and absorber were studied to define acceptable limits for the concentrations.

Passive Assay Position Effects

Only passive assays based on real coincidence counting will be discussed here because they have more applications in actual facilities than assays with total neutron counting.

Moderating Matrices. The ratios of average to extreme passive count rates in Fig. 2 are from a localized plutonium sample moved throughout the drums. There are no interrogation neutrons in this case, but the matrix affects the transport of fission neutrons from within a drum into a detector. The fission neutrons have higher average energies than delayed neutrons, so the transport to the detectors is somewhat different than during active assays.

The position effect is smaller than with the active assays for hydrogen densities below about 0.04 g/cm^3; beyond that density the position effect is generally much worse. The average-to-minimum ratios become very large with the larger amounts of moderator because neutrons are less likely to escape from near the drum's center.

The average-to-maximum ratios are less affected because the maximum responses are from plutonium positions near a drum's surface and outside most of the matrix.

It is again recommended that the contents of a drum with an unusually high hydrogen density beyond 0.042 g/cm^3 be repackaged to lower the density before the assay.

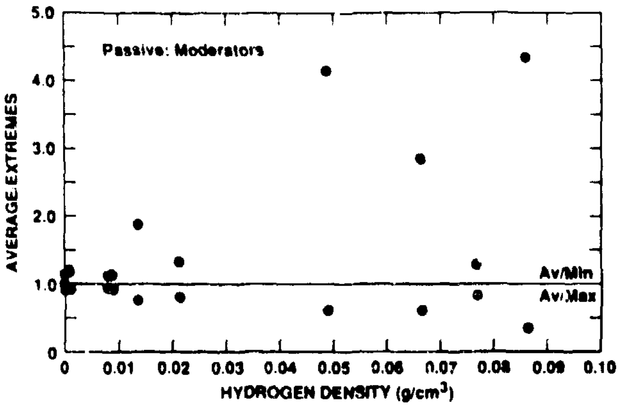

Fig. 2. The ratios of Fig. 1 are shown here for passive responses (real coincidence counting) from localized waste quantities (tens of milligrams) of plutonium. The shuffler's average-to-minimum ratios are large for the matrices of vermiculite with 68 kg of polyethylene beads, the 91 kg of polyethylene chunks, and the 120 kg of polyethylene beads.

Absorbing Matrices. Absorbers have no effect on passive coincidence counting. A neutron that is nearly thermalized (and thus has a good possibility of being captured) will not contribute to the real coincidence count rate because it cannot be detected within the electronic time gate with other neutrons from the same fission. Therefore, it does not matter if this neutron is absorbed or not.

Moderating and Absorbing Matrices. A moderator increases the capture rate in the absorber, but any impact on a passive coincidence assay is caused by the moderator alone. The absorber still has no effect for the reasons just discussed.

Excessive Moderator

An upper limit of 0.042 g/cm^3 for hydrogen in a drum has been suggested above for both the active and passive assays. As the hydrogen density grows beyond this limit, inaccuracies in assay results increase. With a hydrogen density approaching 0.1 g/cm^3, the fissile material is shielded to the point where the assay result is zero even though hundreds of grams of ^{235}U may be localized near the center of a drum.

The flux monitors used to correct for transport effects of the matrix are inadequate to detect excessive amounts of moderator because their count rates saturate at the limit of 0.042 g/cm^3 of hydrogen.

The detector bank across the assay chamber from the ^{252}Cf source that normally counts delayed or prompt fission neutrons is able to also count neutrons transmitted through drums while the ^{252}Cf irradiates the drums. This bank's count rate did not saturate even with no drum present. A drum with an excessive amount of moderator gave about half the transmission count rate of an acceptable drum, so the presence of such moderation is readily detected.

The shuffler can inform an operator of the excessive amount of moderator so that the drum's contents can be repackaged. The facility may wish to investigate how and why so much moderator got into the drum in the first place.

Sensitivities

Sensitivity here means the smallest fissile mass that produces a signal three times greater than its precision; its precision is the standard deviation of a large set of repeat assays on the same drum.

The shuffler sensitivities given below are for drums with *uniform* distributions of the fissile material. Sensitivity is the same function of position discussed as assay responses if the fissile material is localized.

The background rate is an important limiting factor on sensitivity. Cosmic-ray interactions in the shuffler's body are the source of coincident neutrons; the background rate is weaker at lower elevations. A small fraction of the neutrons from the ^{252}Cf source leak through the shielding and increase the total background rate. Intense neutron sources at a facility have the potential of adding to the total background if they are near the shuffler, but the shuffler's thick walls of shielding and facility controls can eliminate this potential problem. The total neutron background affects the accidental coincidence rate and thus a passive assay's sensitivity, although this is a minor effect compared to the cosmic-ray background.

New hardware and software techniques are discussed below to reduce the background rates and thus improve the sensitivities.

Active Sensitivities. Table II shows the sensitivities from active assays of ^{235}U in grams of that isotope. These are computed for a 1000-s assay consisting of a 270-s background count preceding 730 s of active interrogation and delayed-neutron counting. The background count rate used in the calculations consists of 2 counts/s from cosmic-ray interactions (at 170 m above sea level) and 12 counts/s from a 500-μg ^{252}Cf interrogation source.

Neutrons from the ^{252}Cf dominate the background, so the sensitivity would be improved by better isolation of the ^{252}Cf from the assay chamber. If the total background rate were 4 counts/s instead of 14 counts/s, the sensitivities would be approximately half those shown here. Improved shielding materials have become available since this shuffler was built and are being studied for possible use in future shufflers.

Two-thirds of the sensitivities are under 0.25 g of ^{235}U and about half are under 0.1 g of ^{235}U. Sensitivity improves as the hydrogen density increases until 0.025 g/cm^3 is reached: the fission rate is enhanced by the moderated energies of the interrogation neutrons, but interrogation and delayed neutrons are able to still pass through the matrices without excessive capture rates. Even at the highest hydrogen densities, the sensitivities are well under 0.1 g of ^{235}U.

The sensitivities to plutonium for active assays are much worse than with uranium because the yield of delayed neutrons per fission in plutonium is about a third of that in uranium. The sensitivities range from 1 to 4 g of low-burnup plutonium (0.058 to 0.233 g of $^{240}Pu_{eff}$). The passive assay sensitivities are much better than the active sensitivities, so it is recommended that the passive assay option of the PAN shuffler be used on plutonium.

Sensitivities are greatly improved by reducing the energy of the interrogating neutrons when it is known that the hydrogen density is below 0.01 g/cm^3 (corresponding to 14.6 kg of polyethylene or 18.7 kg of water). This was demonstrated in this study by surrounding drums with about 2 cm of polyethylene. However, none of the results given in this paper are from data taken with this technique.

TABLE II. Assay Sensitivities		
	Sensitivity	
Matrix	Active ($g\ ^{235}U$)	Passive ($mg\ ^{240}Pu_{eff}$)
Neutral		
1. Empty	0.384	1.35
2. Iron Chunks	0.402	1.22
Moderators		
3. Vermiculite	0.292	1.34
4. Vermiculite in Liner	0.141	1.39
5. Paper	0.111	1.42
6. Simulated Junk	0.120	1.46
7. Polyethylene Shavings	0.076	1.44
8. Concrete Blocks	0.147	-------
9. Iron and Polyethylene Chunks	0.075	2.33
10. Vermiculite and 29.5 kg of Polyethylene Beads	0.033	2.21
11. Polyethylene Tubes	0.029	-------
12. Vermiculite and 59 kg of Polyethylene Beads	0.038	-------
13. Vermiculite and 68 kg of Polyethylene Beads	0.047	3.87
14. Peat Moss	0.027	2.12
15. Polyethylene Chunks	0.074	4.27
16. Polyethylene Beads	0.077	12.06
Absorbers		
17. Raschig Rings	0.753	-------
18. Vermiculite and 0.3 kg of Borax	0.315	1.35
19. Vermiculite and 0.6 kg of Borax	0.337	1.34
20. Vermiculite and 0.9 kg of Borax	0.335	1.31
21. Vermiculite and 1.2 kg of Borax	0.345	1.39
22. Vermiculite and 1.8 kg of Borax	0.345	1.38
Moderators and Absorbers		
23. Alumina with 28% Water (by weight)	0.063	4.74
25. Vermiculite, 68 kg of Polyethylene Beads, 0.21 kg of Borax	0.061	-------
25. Vermiculite, 68 kg of Polyethylene Beads, 0.42 kg of Borax	0.072	5.22
26. Vermiculite, 37 kg of Polyethylene Beads, 0.22 kg of Borax	0.053	2.49
27. Matrix 26, top; Vermiculite, bottom	0.086	1.74
28. Vermiculite, top; Matrix 26, bottom	0.073	1.86

Passive Sensitivities. Sensitivities with passive assays of plutonium are given in Table II for a real coincidence background rate of 0.17 counts/s and an assay time of 1000 s.

This background rate is again for an elevation of 170 m above sea level but includes a cosmic-ray rejection scheme that detects and eliminates coincidence counts with high multiplicity that could only be created by cosmic rays in the shuffler's body. At the Los Alamos elevation of 2225 m, this technique reduces the real-coincidence background rate by a factor of 1.3 and significantly improves the sensitivity.

The accidental coincidence counts create another component of the background, but for waste quantities of plutonium they are nearly negligible.

Matrices numbered 2 through 10 represent a range of materials that best represent facility waste. The sensitivities for these materials range from 1.2 to 2.3 mg of $^{240}Pu_{eff}$ (or 20 to 39 mg of plutonium, for the low-burnup isotopics used in this study).

Sensitivities worsen to 4 or 5 mg of $^{240}Pu_{eff}$ as the hydrogen density goes beyond 0.042 g/cm^3. Matrix 16 with 120 kg of polyethylene beads creates an effective shield for plutonium near the drum's center, so the sensitivity is 12 mg of $^{240}Pu_{eff}$.

Absorbers have no effect on passive sensitivity, even in combination with moderators, for the same reasons discussed earlier for position effects.

4. Conclusions

The PAN shuffler performs precise assays of uranium (active mode) and plutonium (passive mode) on 208-L waste drums. The most important cause of inaccuracies in assays is ignorance of the distribution of fissile material within a drum. Calibration is likely to be done with a uniform distribution, while highly localized distributions are possible in practice. A technique for determining the distribution from the assay data and correcting for the assay result accordingly is suggested here and will be explored further.

The hydrogen density in a drum should be limited to 0.042 g/cm^3 for both active and passive assays, which corresponds to very large amounts of water (75.6 kg) or polyethylene (58.8 kg) in a 208-L drum of waste. Flux monitor corrections have been devised to correct the measured count rates for the matrices.

In drums holding more than this density of hydrogen, the variation of assay results with position can lead to very large inaccuracies and the flux monitor responses do not reflect the proper amount of hydrogen on which a correction would be based. Detectors on the opposite side of the assay chamber from the ^{252}Cf source can monitor the transmission of interrogation neutrons through the drums and give a warning when the hydrogen density is excessive.

Sensitivity (minimum detectable mass) from an active or passive assay depends on the nature of the matrix, the background rate, and count times. A software technique has been used to reduce the cosmic-ray background rate and new shielding materials are being studied to reduce the background component from the ^{252}Cf source.

5. References

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