LA-UR-. 98-3081

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CONF-980733--

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Submitted to: 39th. Annual Meeting of the Institute of Nuclear Materials Management (INMM) Naples, Florida. July 26-30, 1998

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SHUFFLER CALIBRATION AND MEASUREMENT OF MIXTURES OF URANIUM AND PLUTONIUM TRU-WASTE IN A PLANT ENVIRONMENT

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ABSTRACT

The active-passive shuffler installed and certified a few years ago in Los Alamos National Laboratory's plutonium facility has now been calibrated for different matrices to measure Waste Isolation Pilot Plant (WIPP)-destined transuranic (TRU)-waste. Little or no data presently exist for these types of measurements in plant environments where there may be sudden large changes in the neutron background radiation which causes distortions in the results. Measurements and analyses of twenty-two 55-gallon drums, consisting of mixtures of varying quantities of uranium and plutonium, have been recently completed at the plutonium facility. The calibration and measurement techniques, including the method used to separate out the plutonium component, will be presented and discussed. Particular attention will be directed to those problems identified as arising from the plant environment. The results of studies to quantify the distortion effects in the data will be presented. Various solution scenarios will be indicated, along with those adopted here.

INTRODUCTION

For the past ten years or so, 55-gallon drums containing TRU waste consisting of uranium-plutonium mixtures embedded in mostly noncombustible (dense, heavy) matrices have been gradually accumulating at the plutonium facility at Los Alamos. While the plutonium component could easily be measured using well known thermal neutron coincidence techniques[1], the uranium posed a much greater challenge. Conventional γ-ray and neutron counting techniques could not be employed because of the high density of the matrices and very low spontaneous fission cross section and heat output of the uranium[2]. Clearly, a different method was needed. This new capability was achieved with the recent acquisition[3] and certification[4] of an active-passive ^{252}Cf shuffler instrument. Because this instrument induces fissions in uranium and measures the resulting delayed neutrons[5], the aforementioned difficulties were surmounted and it became possible, in principle, to measure these WIPP-destined uranium-plutonium TRU-waste drums.

The intent here is to discuss the many challenges (and their resolution) encountered along the way to achieving the measurement results and analysis of these 22 drums. To this end, the paper is largely structured along chronological lines in order to describe the actual series of events as they occurred in time. Some of these challenges, such as distortions in the measured count rates caused by sudden shifts in the neutron background radiation, were clearly a result of the environment. Others, such as standards and matrix material not appropriately matching the measured items, were caused by a lack of adequate resources. The resolution of many of these difficulties are presented here.

MEASUREMENT CHALLENGES

The drums were measured in both passive and active modes[5] on the shuffler to obtain the plutonium and uranium masses, respectively. In addition, for comparison of the plutonium results, they were also measured on a thermal neutron drum counter[6] located near the shuffler in the nondestrucive assay laboratory. However, before the final mass values could be obtained, several initial challanges had to be met.

Drum Standard

The first obstacle was the lack of a drum standard[7] with which to calibrate the shuffler in the active (delayed-neutron counting) mode for uranium and plutonium. This challenge was met by simply acquiring corrected count rate data in the active mode and writing the data files to the hard disk of the acquisition computer. The data files were then written to a floppy disk and analyzed off line months later after the drum standard had become available and the instrument calibrated. The final uranium and plutonium masses were then extracted from that count-rate data.

Data Distortion

During acquisition of the count-rate data, outliers were occasionally (roughly 5% on average) observed. In fact, this phenomenon had first been noted while acquiring the initial calibration and certification data several years ago[4] using uranium oxide can standards. At that time, shifting neutron flux, caused by movement of nuclear material near the instrument (see Figure 1), was strongly suspected of being the cause but no definitive study had been conducted to prove it. Here, while acquiring the uranium calibration data with the drum standard, movement of nuclear material near the instrument during the replicate calibration runs was observed to clearly enhance the standard deviation. The decision was then made to undertake a study of this effect.

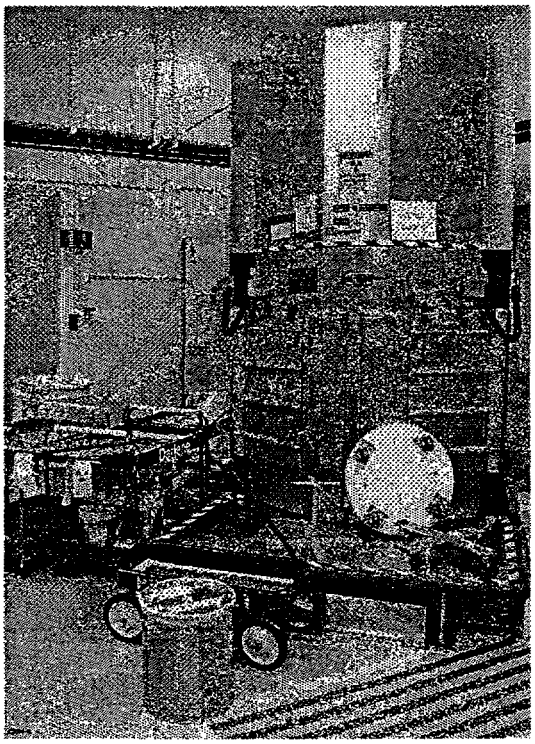

Figure 1. Shuffler in Plant Environment

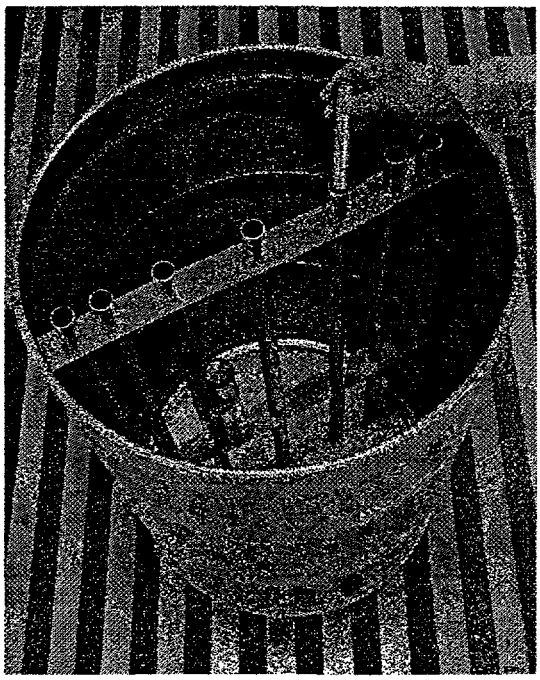

Figure 2. Drum Standard

STUDY The distortions arise not only because of inadequate exterior polyethylene shielding of the shuffler for a plant environment but also from the way the software handles the data acquisition. The instrument acquires a background count rate during the first part of each individual run. This background rate is then subtracted from the delayed neutron count rate obtained during the second part of the run. If neutron-emitting nuclear material is located near (within several meters) the shuffler during the background acquisition and then moved away during the second part of the run, the final result will be biased low because too much background will be subtracted from the

delayed neutron rate. The converse is also true. If a cart of neutron-emitting items is brought near the instrument at the midpoint of a run, the final result will be biased high. These situations, of course, represent the extremes of the distorting process. It isn't likely in reality that the timing would be so good. Clearly, any distortion between these extremes can occur depending on the timing and strength (activity) of the distorting source.

To test the magnitude of the effect in one specific case, a 4-gram uranium standard was placed near the center of the drum standard (see Figure 2) and three ^{238}Pu standards of known strength were obtained to provide the source of variable neutron flux (see Figure 3). (Of course, the relative magnitude of the effect will depend on the mass of the item being measured as well as the strength and timing of the distorting source.) The average distance of the nearest detector banks from most of the distorting items in the NDA Laboratory was crudely estimated at between 50 and 100 cm (see Figure 1). From Figure 3, this corresponds to a flux of about 5 to 6 mrem/hr for the ^{238}Pu standards. A series of five runs was then conducted using the ^{238}Pu standards as the variable flux source (shown plotted as the first run group in Figure 4). The first run was done as a control, i.e., there was no nuclear material movement in the vacinity of the shuffler. Next, the three ^{238}Pu standards were brought to the left front of the shuffler and placed on a cart in exactly the same manner that nuclear items are normally staged in the laboratory (see Figure 1). (The actual flux present at the nearest detector banks of the shuffler was about 7 to 8 mrem/hr because another cart was already present which had three items producing a total of about 2.5 mrem/hr at 50 cm.) For the second run, a background was acquired and then all the source nuclear material was moved away during the delayed neutron counting. As expected, the second run is clearly an outlier in the negative direction. The converse was done for the third run which generated an outlier of approximate equal magnitude in the positive direction, as expected. In the fourth run, the ^{238}Pu standards and extra cart of items were left in place for the duration of the entire run. Again, as expected, the results are the same as for the first run (control). For the fifth run, the standards were moved away after the background was acquired but the cart of less "hot" items was allowed to remain. This should drive the count rate down but not as much as the second run where both the standards and additional cart were allowed to remain. As Figure 4 shows, this was indeed the case.

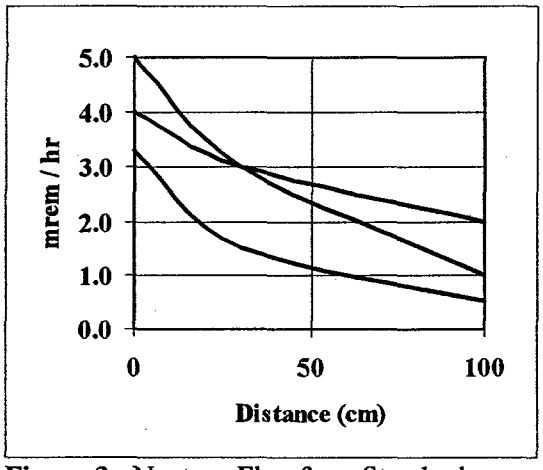

Figure 3. Neutron Flux from Standards

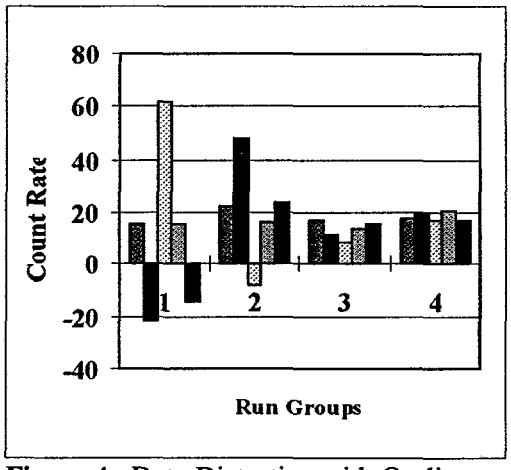

Figure 4. Data Distortion with Outliers

The second group of five runs was conducted in a manner similar to the first except that the ^{238}Pu standards were on a cart with about 1.25 cm of special shielding material designed to shield both neutrons and γ rays. Here, although the magnitude is less than in the first grouping, outliers are still generated and 1.25 cm is not adequate. The third and fourth run groups were done without the

^{238}Pu standards. In the third group, the five runs were acquired in a very routine manner with normal movement of nuclear material. The runs in the fourth group were acquired with no nuclear material movement at all. Although no outliers are generated in the third group, the standard deviation is increased by a factor of 2 to 3 over that of the fourth (control) group. In fact, the relative standard deviations for the four groups are the following: 287%, 98%, 25%, and 9%, respectively. The third group represents the most realistic situation because normally the flux changes partway through the background and/or delayed neutron counting and the standard deviation is accordingly increased.

SOLUTION The best long-term solution is either to redesign the acquisition software such that the background is acquired throughout the run or add additional neutron shielding. Other nonengineered fixes such as controlling the movement and location of the potential source nuclear material would not be viable because of space constraints and the large volume of activity in the NDA Laboratory. For the 22 drums measured here, the challenge was met by simply acquiring a series of replicate runs, anywhere from 3 to 15, on each drum. By comparing these runs with similar runs acquired overnight and on weekends when nuclear material movement was very minimal, it was easy to identify the outliers and badly distorted data.

Matrices

The primary requirement for obtaining mass values from the corrected count-rate data was, of course, to calibrate the shuffler. The challenge was to locate representative matrix material for the drum standard. This was a daunting task because little was known about the TRU-waste drum matrices. Only general catagories were given such as "Glass", "Nonactinide Metal", and "Nonspecific" which could be anything noncombustible. A number of questions naturally arose. For example, if the drum standard were to be loaded with a nonactinide metal matrix, could that matrix also be used for the glass matrices? Could it be used for the nonspecific category? How much bias would be introduced? For that matter, how much effect could slight matrix variations within a given matrix category have on the final result?

This challenge was met by utilizing the flux monitor results[5,8]. The flux monitor ratio provides a measure of the hydrogeneousness of the measured item. Because the hydrogeneous content is the primary bias driver, the flux monitor ratio is a very important indicator for determining the effect of the matrix on the count rate. Fortunately, flux monitor results were available for all the drum measurements because they were a part of the data package that accompanied every run written to the computer disk. It was a simple matter to extract that information off line and tabulate the results.

FLUX MONITOR RESULTS The flux monitor ratios for the TRU-waste drums and standards are shown plotted in Figure 5. Most of the 22 drums reside in matrix categories 3 and 4: Category 3 is nonactinide metal with 13 drums and category 4 is nonspecific with 5 drums. Category 5, glass, has only 2 drums and categories 1 and 2, combustible matrices, have 1 drum each. Only categories 3 and 4 have enough drums to determine a meaningful standard deviation of the flux ratio within the category. This range of the flux ratios is shown plotted like error bars.

These results were very encouraging because all the noncombustible categories ranged around a flux ratio of 1.4. This implied that one set of matrix material could be used in the drum standard to represent the entirety of the noncombustible matrices with a resulting enhancement in the uncertainty that would be well within the uncertainty generated by any attempt to correct for the matrix differences. The two combustible drums could be corrected separately[8].

Category 6 contained the empty drum standard and category 7 the uranium oxide can standards and the empty assay chamber. One very surprising result was that the empty drum standard exhibited the same flux monitor ratio as the TRU-waste drums. If this were indeed true, then perhaps no matrix material would be needed in the drum standard. But why should the empty drum standard have essentially the same flux monitor ratio as the TRU-waste drums? Why should the empty metal drum standard have a flux ratio considerably larger than the ratios of the small uranium oxide can standards or the empty chamber? To answer these perplexing questions, a separate study was undertaken.

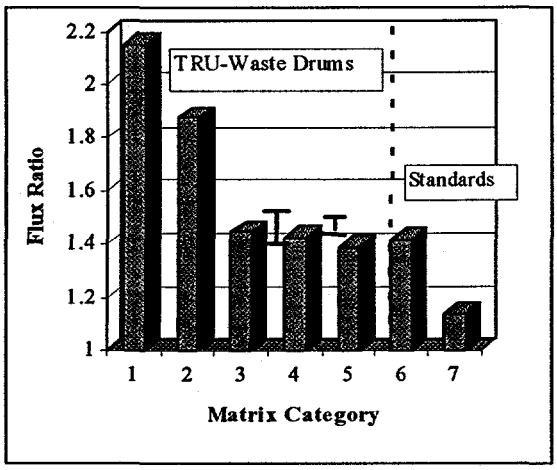

Figure 5. Flux Monitor Results

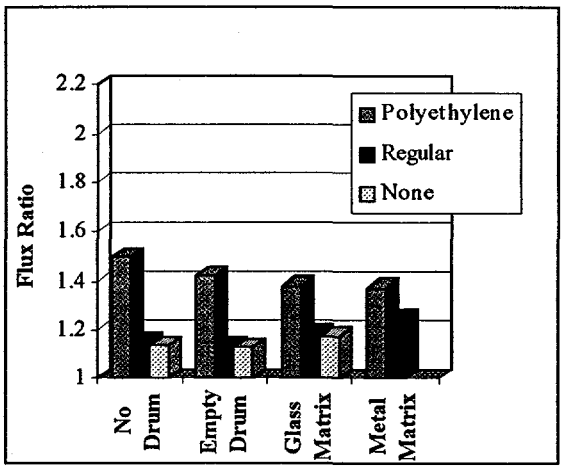

Figure 6. Drum Dolly Study

DRUM DOLLY STUDY Because the drum standard is all metal, the only factar which could possibly have affected the flux ratio was the dolly holding the drum. That dolly, shown in Figure 1, is seen leaning against the closed shuffler doors. It is a specially designed polyethylene-bottomed dolly which came with the instrument. In contrast,.a normal drum dolly is shown just in front of the special dolly. The normal dolly is all metal except for the plastic wheels. The all-metal stand used for the U_3O_8 can standards is shown just to the left.

To test the effect of the two dollies on the flux ratio, each was measured separately along with the empty assay chamber as a control. The results are shown in the first grouping, "No Drum," in Figure 6. Clearly, the special polyethylene-bottomed dolly is producing all the flux ratio. The regular drum dolly and the empty chamber are essentially equivalent. Just to be sure, the measurements were repeated with the empty standard drum on each of the dollies as well as by itself, i.e., no dolly. The results, shown in the next grouping, "Empty Drum," are the same as the first case with only the dollies. Clearly, the special dolly is producing all the flux ratio.

Because the TRU-waste drums were all measured with the special dolly, the approximate flux ratio of 1.4 could undoubtedly be attributed to that dolly. To be sure of this, a couple of the drums, one with a glass matrix and one with metal, were picked at random and remeasured. The results, again shown in Figure 6, prove that the dolly was the cause of the flux ratio. Unfortunately, because of the range of flux ratios from category 3 shown in Figure 5, the randomly-chosen metal-matrix drum was not one of the more illustrative drums that could have been picked but, nevertheless, the point is clearly proven. The good news was that noncombustible matrix material didn't have to be loaded into the standard drum; in fact, no matrix material was needed (see next section). The bad news was that there may have been a positional flux dependence introduced by the dolly which

would also cause a positional dependence in the count rate. The effect of this will be discussed in the next section.

CALIBRATION

Since the TRU-waste drums contain both uranium and plutonium, the shuffler had to be calibrated in the active mode for both nuclides because both have fission cross sections and emit delayed neutrons[5]. However, the plutonium mass is actually determined by measuring in the passive mode because it can be done more accurately and with better precision and because uranium has an extremely small spontaneous fission cross section which permits a natural separation of the plutonium count rate from that of the uranium. The active mode calibration for plutonium then provides the link to separate the plutonium from the uranium in the active-mode measurement.

The active-mode calibration was done using the uranium capsules and drum standard described in Reference 7, along with the Performance Demonstration Program (PDP) plutonium capsules[9]. Because the PDP capsules weren't designed for the drum standard used here, they wouldn't fit into the tubes and had to be taped into position along the tubes (Figure 2 shows a taped PDP standard along with a uranium capsule being loaded into a tube). In order to determine if there was any positional count-rate dependence, the calibration was established as a function of height and radius by varying the positions of the capsules throughout the drum in a manner very similar to that described in Reference 8. From the results of the drum dolly study of the previous section, no matrix material was needed or used in the drum standard. This is also consistent with the flux monitor and count-rate results shown for an empty drum and a metal (noncombustible) matrix in Reference 8.

Because the special polyethylene-bottomed drum dolly (discussed in the drum dolly study of the previous section and shown in Figure 1) had been used for all the TRU-waste drum measurements and because there was strong reason to suspect that a count rate dependence on position would exist, the special dolly was used for the calibration. As expected, a positional dependence was discovered. The count rate in the bottom third of the drum was roughly a factor of two larger than in the upper section. This, of course, increased the uncertainties considerably and this information was used in the error propagation. The uranium and plutonium calibration curves are shown in Figures 7 and 8, respectively. The data were fitted linearly using the curve-fitting routine based on the Deming method[10].

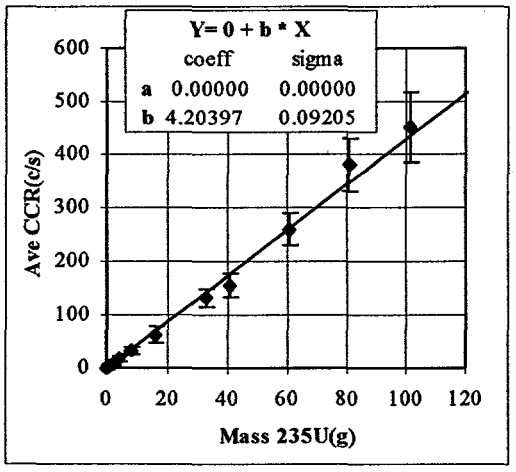

Figure 7. Uranium Calibration

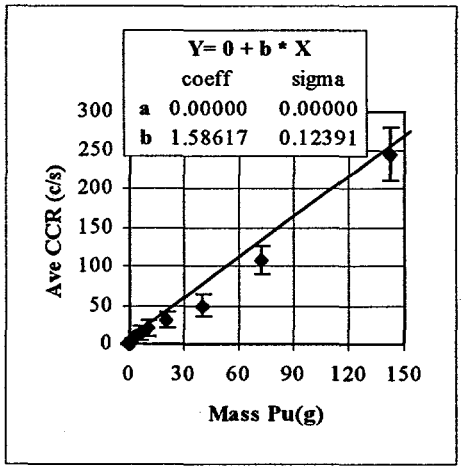

Figure 8. Plutonium Calibration

DISCUSSION

The first step in obtaining the final uranium mass results was simply to apply the calibrations to the corrected count rate data. The plutonium masses were determined directly from thermal neutron coincidence counting (passive mode of the shuffler and the NBC thermal neutron counter[6] for corroboration). These plutonium masses were converted to count rates using the active-mode plutonium calibration. These count rates were then subtracted from the measured rates. The resulting rates were converted to effective ^{235}U masses using the uranium calibration. The measured effective ^{235}U masses were then decoupled into $^{Total}U$ and ^{235}U masses.

DECOUPLING CALCULATION The concept of an effective ^{235}U mass emerged because the ^{235}U enrichments of the TRU-waste drums differed in every case from that of the calibration standards (92.11%). The enrichments of the drums, taken from the material accountability system and not measured separately, varied from 0.711% (normal uranium) to over 97%. In order to use the calibration curve for these multiple enrichments, the delayed neutron count-rate contribution from the ^{238}U had to be taken into account. One of the final challenges was to determine the best way to do this. The key was to understand that a calibration developed at a particular enrichment can only be used to convert a count rate at a different enrichment to an effective ^{235}U mass where the effective mass contains both ^{238}U as well as ^{235}U contributions. The ^{235}U effective mass can then be expressed as a function of the enrichment of the total uranium mass in the following manner:

$$^{Total}U * [ENR + (1\text{-}ENR)*(0.365)*(ABI)] = {}^{235}U_{eff}$$

where $^{Total}U$ = sum of the ^{238}U and ^{235}U components
ENR = ^{235}U enrichment as a fractional percentage
1-ENR = ^{238}U fractional percentage
0.365 = (^{238}U / ^{235}U) conversion factor
ABI = ^{238}U fractional adjustment
$^{235}U_{eff}$ = the effective ^{235}U mass.

The $^{235}U_{eff}$ is, of course, the mass value obtained from applying the uranium calibration to the delayed neutron count rate. From the above equation the total uranium is easily obtained and then the total, multiplied by the fractional ^{235}U enrichment, gives the ^{235}U. The conversion factor, obtained from known fission cross sections and delayed neutron rates[11], relates the ^{238}U mass to its equivalent ^{235}U value as measured on the shuffler. The most difficult factor to calculate was the ^{238}U fractional adjustment. The adjustment is necessary because the calibration already has some ^{238}U built into it from the standards and this needs to be taken into account to achieve maximum accuracy. For example, it can easily be seen that ABI should be close to zero for enrichment values close to those of the standards (92.11%). For enrichments much less, ABI should be close to one. The challenge was in finding the dependence as a function of enrichment. The final best result is the following:

$$ABI = (0.9211 - ENR) / (1 - ENR).$$

This is the equation for a hyperbola and makes physical sense because, in order to keep the ^{235}U / $^{Total}U$ linear, more ^{238}U has to be added back per incremental enrichment change in the high enrichment range than in the low. Also, the functional dependence is correct at both enrichment bounds, i.e., 0.00 and 92.11%. It can easily be shown that an error much less than 1% is introduced by setting ABI equal to zero for enrichments greater than 92.11%.

CONCLUSIONS

Twenty-two TRU-waste drums containing mixtures of uranium and plutonium embedded in dense, noncombustible matrices were measured on a ^{252}Cf shuffler at the plutonium facility at Los Alamos. Results with reasonable uncertainties were achievable as long as environmental and resource limitations, such as variable nuetron backgrounds causing data distortions and lack of sufficient calibration standards, were properly taken into account. The scope of this paper didn't permit showing the measurement results including treatment of two drums in combustible matrices, two drums with mixtures of uranium enrichment, and one drum with a plutonium enrichment mixture. These, as well as the details of the theory development and the propagation of the various components of uncertainty, including both bias and precision, will be presented in a forthcoming report.

Perhaps one of the more important results from this effort was the knowledge gained to point the way to making future improvements in the measurement capability of the shuffler. For example, plans are already underway to add to the polyethylene shielding around the base perimeter of the instrument. This should eliminate all future data outliers caused by varying neutron background flux and, in general, greatly reduce the data distortion experienced here. In progress also is the acquisition of a set of uranium drum standards with enrichments spanning the entire range. These standards will not only provide the means for spanning the entire calibration enrichment range but will also permit checking the theory and results here.

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