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IN-PLANT EXPERIENCE WITH PASSIVE-ACTIVE SHUFFLERS AT LOS ALAMOS

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ABSTRACT

Two Canberra-built passive-active ^{252}Cf shufflers of Los Alamos hardware and software design have been installed at Los Alamos National Laboratory, one at the Chemistry and Metallurgy Research (CMR) Facility at TA-3 and the other at the Plutonium Facility (PF-4) at TA-55. These instruments fulfill important safeguards and accountability measurement requirements for special nuclear material (SNM) in matrices too dense or otherwise not appropriate for typical gamma-ray or other neutron counting techniques. They support many programmatic requirements including measurements of transuranic (TRU) waste and inventory verification. This paper describes the instrument performance under plant conditions with various background radiations on well-characterized standards to determine long-term stability and establish a calibration. Results are also reported on verification measurements of previously unmeasured inventory items in various matrices and geometric distributions. Preliminary investigative measurements are presented on standards of mixed uranium and plutonium oxide (MOX).

INTRODUCTION

During the past two years two passive-active ^{252}Cf shuffler instruments were procured, fabricated, and installed at Los Alamos plant environments[1], one at the CMR Building Waste Assay Facility (WAF) and the other at the Plutonium Facility (PF-4) Nondestructive Assay (NDA) Laboratory. These instruments were acquired to enhance the capability of measuring up to 55-gallon drum-sized containers of plutonium and uranium distributed both heterogeneously or homogeneously in a wide variety of matrices. The shufflers had to meet specified levels of precision, sensitivity, and accuracy within the severe plant environments including high levels of fluctuating gamma-ray and neutron background radiation in the PF-4 case.

Although the hardware and software installation were completed a little over a year ago, the instruments are just now beginning to make full-scale accountability measurements. The intervening time was spent addressing and meeting the two major operational requirements: the PF-4 Safety and Technical Assessment for Readiness of Technology (START) review and the DOE-mandated (Order 5633.3A) certification process. Several additional factors contributed to the time span: (1) PF-4 was in a "standdown" operational mode from April to July, 1994, (2) Additional shielding was required in both instruments to correct radiation safety deficiencies, (3) A National Environmental Policy Act (NEPA) review had to be completed by DOE, (4) A design defect was identified requiring the addition of cadmium in both instruments, and (5) The standards discussed in the next section had to undergo additional statistical analyses in order to establish their precise fabrication uncertainties.

This paper reports on the calibration procedures as well as results of the START review and certification process. Some interesting anamolies, caused by a high, fluctuating neutron background flux, were discovered at the PF-4 NDA Laboratory. Results of measurements made at the CMR WAF on unmeasured uranium inventory items are discussed and data are presented on measurements made at PF-4 on mixed uranium and plutonium oxide (MOX) standards. Finally, future uses and plans for these instruments are identified and discussed.

CALIBRATION

Standards

The two shufflers were calibrated with National Institute of Standards Technology (NIST)-traceable standard items consisting of well-characterized uranium oxide (U_3O_8) powder enriched to 92.41% in ^{235}U. The standards were packaged in thin-walled, tin-plated steel containers 5 inches in diameter. The different masses gave different fill heights and, consequently, different ratios of height to diameter for the oxide. Calibration data were collected with the CMR shuffler using cans with ^{235}U masses of 50, 125, 250, 500, and 1000 g. Cans with 10, 75, 150, 500, and 750 g of ^{235}U were used at the Plutonium Facility (PF-4). In both cases a 0 g result was achieved by assaying with the chamber empty.

Another can with 2000 g was also measured at PF-4, but its diameter and containment were different than the others (4.5 in. inner diameter, doubly contained); nevertheless, its data appear to give a reasonable extension of the calibration curve to its higher mass since the inner container diameter reasonably matched those of the smaller-mass singly-contained standards.

Data Integration

The ^{252}Cf sources in the two shufflers differ in strength by about 10%, so an adjustment was made to the PF-4 shuffler's data analysis to remove this difference mathematically as part of every measurement. The magnitude of the adjustment was determined by measuring the responses of the two shufflers to the can with 500 g of ^{235}U and then normalizing the PF-4 data to match that of the CMR. The two sets of data are integrated in Fig. 1. This cross calibration was feasible because the two instruments were built identically at Canberra Industries, Inc., using the same design and software[1]. Except for the neutron background flux to be addressed in the next section, the environments in which the shufflers exist are quite similar. No other aspect of the two environments could influence the performance characteristics; e.g., gamma rays will never affect the count rate, temperature variations are normal and have no impact, and electrical noise is kept from each instrument by a line filter and voltage regulator. The only reason the two shufflers could produce different count rates from the same item would be the neutron emission rates from their respective ^{252}Cf sources.

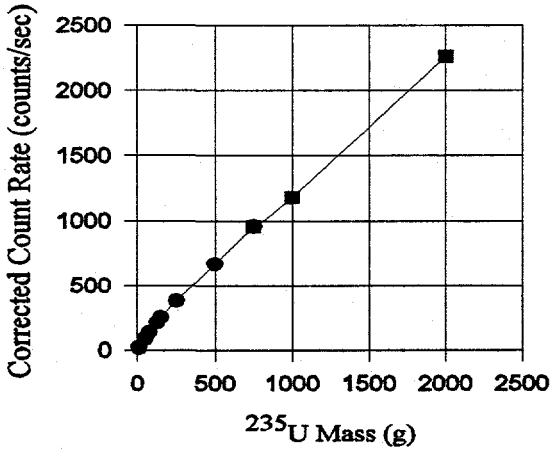

Figure 1. Ten cans of uranium oxide (U_3O_8) were divided between the shufflers for separate calibrations and the results combined into this single curve. The 500 g can was measured on both shufflers for normalization. Because the U_3O_8 density varies with handling, the very small scatter about the fitted curve attests to the value of the sample handling technique used.

Results

The combined calibration curve in Fig. 1 is a combination of a third-order polynomial from 0 to 750 g of ^{235}U and a straight line from 750 to 2000 g. A single third-order polynomial from 0 to 1000 g is nearly as accurate, but a single curve from 0 to 2000 g cannot give the linear response between 1000 and 2000 g of ^{235}U. The differences between the declared masses and the masses interpolated from this curve are shown in Table 1; the largest relative differences are with the smallest masses.

Table 1. Differences Between Declared and Interpolated Masses in Fig. 1

Calibrat. Mass (g)	Uncert. (g)	Interpol. Mass (g)	% Err
9.98	0.42	10.9	9.4
49.95	0.56	49.2	-1.7
75.00	0.65	77.0	2.7
125.00	0.82	127.2	1.7
149.99	0.91	151.0	0.64
250.00	1.25	248.4	-0.62
500.03	2.17	500.0	0.01
750.01	3.03	750.1	0.01
750.01	3.03	750.1	0.1
1000.01	3.89	999.9	0.00
2000.05	7.60	2002.0	-0.12

Average Absolute Error: 1.1g
Average Absolute Percent Error: 1.5%

Preliminary calibration measurements revealed variations in the delayed neutron count rates as large as 20% according to how the standard cans were handled prior to the measurement. Subsequent measurements[2] indicated that the oxide powder's density can change by as much as 10% if the standards are shaken or tamped to either loosen or compact the powder. We have now chosen to always tamp the standards (e.g., by tapping them several times on the assay chamber floor) before a measurement to minimize this problem; the very small scatter about the fitted curve shows its effectiveness.

Future

Calibration for drums of waste will be done when the fabrication of appropriate standards[3] is

completed. Small capsules of ^{235}U, diluted in diatomacious earth, are being prepared and will be placed in assorted configurations within drums containing various matrices (such as paper and scrap iron). This approach is similar to that described in Ref. 4. The uranium matrix densities of these drum standards should not be subject to the variations caused by the handling described above.

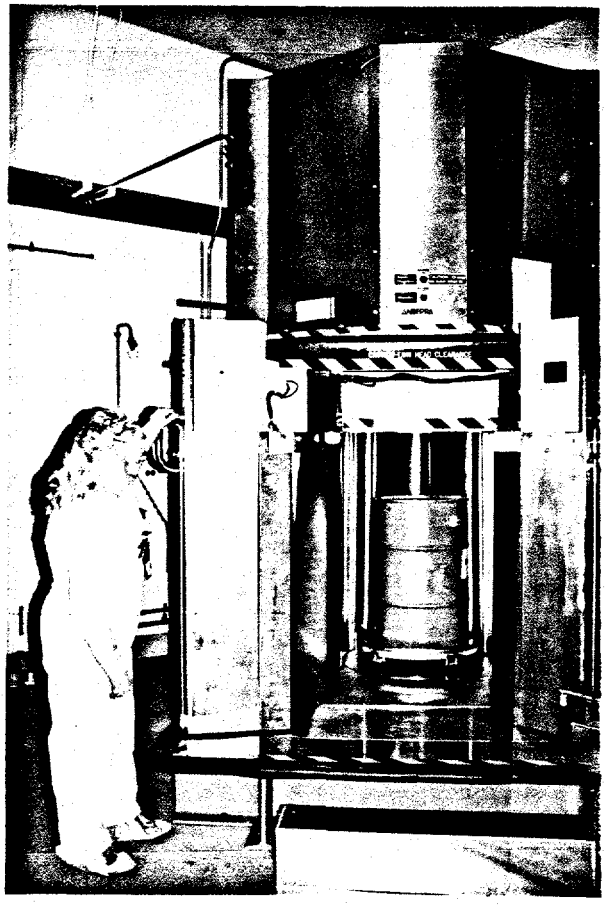

Figure 2. This photograph shows an operator looking into the PF-4 shuffler in the NDA Laboratory. A number of safety enhancements are visible including a rubber bump guard with striping and anti-slip strips around the perimeter of the baseplate, striping and stenciling inside the assay chamber, and conspicuous labeling by the status lamps above the assay chamber. Note also the portable step in front of the baseplate.

OPERATIONAL MANDATES

Before the instrument could be used for any programmatic purpose, a Safety and Technical Assessment for Readiness of Technology (START) review had to be completed at the Plutonium Facility. Similar requirements of lesser degree had to be met at the CMR facility. In addition, both instruments had to undergo the DOE-required certification process. Since these operational requirements were reoprted in last year's conference proceedings[1], only the results are discussed here.

START Review

An on-site committee composed of experts in ES&H, radiation protection, waste management, and technical areas was convened late last year to identify and outline corrective action for any inadequacies found in the instrument within the defined safety envelope at the Plutonium Facility. The examination of the shuffler in the NDA Laboratory setting revealed a number of mostly minor deficiencies which have been addressed already. From Fig. 2 we can immediately identify a number of safety upgrades when compared with the similar picture in Ref. 1 taken a year earlier. For example, all the sharp edges and rough areas have been ground down or smoothed. A rubber bump guard along with striping and anti-slip strips were placed around the outer perimeter of the baseplate. Striping and stenciling were added inside the assay chamber along with enlarged and better defined warning labels by the status lamps just above the chamber. Small areas of exposed lead, visible at the top of the assay chamber doors, were covered with a clear sealant. A small portable step can be seen in front of the platform. Not visible in the figure is a long rectangular aluminum piece to be positioned behind the drum-dolly wheels to minimize rolling on the platform. Also, the motion controller, located just above the computer monitor in the electronics rack, was covered with a clear plastic shield to prevent any inadvertent driving of the ^{252}Cf source into the assay chamber.

Shortly after the NDA Laboratory shuffler was installed, a radiation shielding deficiency was discovered in the baseplate allowing neutrons to scatter into the laboratory. Additional borated polyethylene was placed inside the baseplate. Even with that, there is one spot in back that is slightly above the acceptable level of 5 mR/hr at contact which requires the radiation posting barely visible in the figure at the left of the instrument.

In contrast, the shuffler at the CMR WAF (Fig.3) required fewer safety upgrades since that instrument was positioned over a pit in the floor thereby obviating the requirement of the base platform for elevation[1]. Obviously, all the safety issues associated with the platform are nonexistent here. One interesting phenomenon did occur, however. The gamma radiation from the ^{252}Cf source during an assay was sufficient to raise the background in a

portal monitor at a guard station about 40 ft away. The steel plate seen at the left rear of the instrument was added to attenuate the gamma rays.

One final radiation safety issue deserving mention concerns the potential danger during a power outage. If a loss of power were to occur during an assay, the ^{252}Cf source could be in the assay chamber. If the doors were opened, the interlock would be disabled since there would be no power to drive the source into the storage cube. At present we are administratively controlling this. In the near term we plan to implement some form of engineered control such as connecting to an uninterruptable power source.

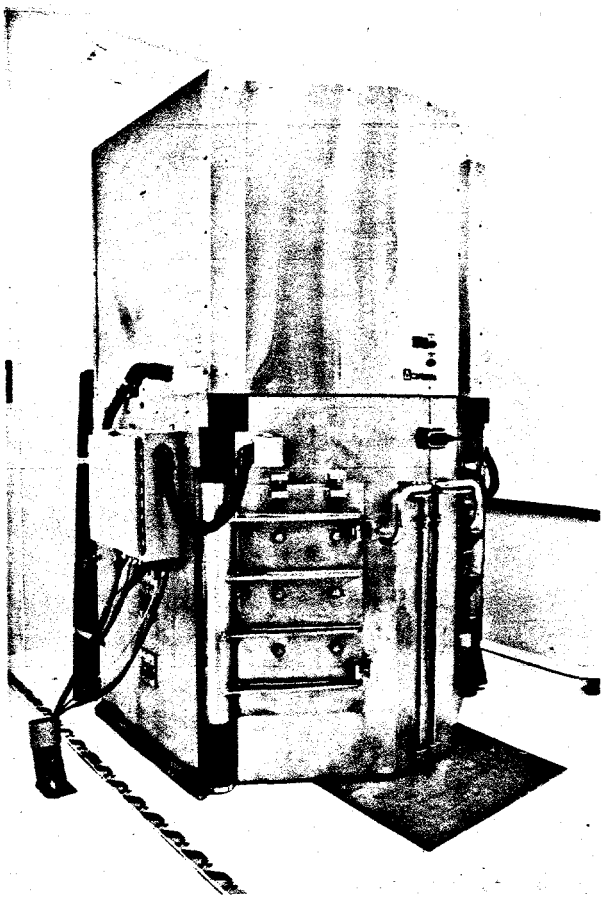

Figure 3. This photograph shows the CMR shuffler at the WAF. Since it is positioned over a pit in the floor, the baseplate platform is unnecessary which eliminates many safety concerns. Note the large piece of steel at the left rear of the instrument to attenuate the gamma rays produced by the ^{252}Cf source.

Certification

Both instruments have recently successfully completed the certification process. Safe operating procedures, along with operator training requirements, have been written and are in place. Measurement control programs, failure response plans, a calibration method with certified standards, and a procedure to determine the calibration uncertainty have all been established. To establish statistical control limits and demonstrate the instruments' stability for the measurement control program, data collection and assessment plans had to be successfully completed. The data were collected in the plant environments over a four-week period with a random selection of the standards.

During the data assessment, a small percentage (3 to 5%) of "outliers" were observed in the NDA Laboratory shuffler data, mostly involving the smaller-mass standards. The environments in which the shufflers exist are quite similar except for their neutron backgrounds. There are almost no neutron sources (such as Pu) in the CMR WAF and the background rate is low and steady. AT PF-4 the background is normally much higher because of plutonium in and near the room with the shuffler, and the background rate can change as the plutonium is moved. The higher rate by itself is not a serious problem because a new background count is taken as part of every assay and the background rate is subtracted as part of the assay process. But the background rate is assumed to stay constant during the rest of the assay; if this is not the case, the accuracy will suffer in some degree. For large masses of uranium, the signal will be much larger than the background and variations in the background will have little effect. For small masses, a large change in the background rate can make the assay result very inaccurate.

Studies are being planned to better quantify the magnitude of the assay error for a given uranium mass and a given change in the background rate. Procedures for handling materials in the NDA Laboratory will be developed to minimize the changes in background rates in the shuffler and shielding or assay protocols may be implemented to further alleviate the problem.

INITIAL ASSAY RESULTS

Inventory Verification at CMR

A wide variety of unmeasured inventory items containing enriched uranium were examined with the CMR shuffler. The calibration from the uranium oxide standards discussed in the previous section was used for all items. Many of the inventory items differed from the standards in important ways such as geometry, impurities, and matrices. Most of the ^{235}U masses were 200 g or less, but two went as high as 1000 g. Of the 46 items measured by the CMR shuffler, about two-thirds passed the verifications

measurements. The assay values of these items were within 30% agreement with the inventory values. The other one-third were considered good enough for confirmations. These items were generally impure oxides in odd-shaped containers and were determined difficult to measure. The shuffler's assays were in excellent agreement (from 1% to 15%) with the inventory values for three items identified as uranium mixed with calcium in containers about twice as large as the standards in all dimensions (8 in. diameter, 10 in. high); the ^{235}U masses of the three items ranged from 200 to 1000 g. Many other cans had diameters of 6 in. and heights of 7 in. with less than 100 g of ^{235}U. The assay results were again in good agreement with the inventory values.

The shuffler's assays were much higher than the inventory values for cans with exaggerated shapes and impure oxides (the impurities were not specified). Results were also high for rods inside cardboard tubes (3 in. diameter, 30 in. long). Neutron moderating impurities and extended shapes of the uranium are expected to give high assay results when the calibration standards are pure oxides in squat containers. There are large differences in penetrability of the irradiating neutrons and in the neutron multiplication within the items.

The shuffler's usefulness in this activity is clearly related to the effort placed on its calibration. If a variety of materials and containers are to be assayed, a variety of matching calibration standards are needed. The fabrication of standards is costly and time consuming, when it can be done at all. A program of modeling the shuffler for a Monte Carlo code could be attempted. If the model can successfully match the results with the existing standards, it could be extended to other materials for which standards do not exist.

MOX Standards at PF-4

A program is underway to develop techniques for assaying cans of mixed uranium and plutonium oxides (MOX). The shuffler's passive mode and gamma-ray isotopics measurements are used to determine the plutonium contents, which are then combined with the shuffler's active mode to determine the uranium contents. Measurements have been completed on seven well-characterized, pure standards. Table 2 shows the standards' loadings and the active-mode measured count rates. The plutonium has about 5.86% ^{240}Pu and the uranium is 93% enriched in ^{235}U.

The net mass of each can is about 1700 g, but the proportions of the two elements range widely. The standards STD 1 and STD 7 are almost entirely plutonium and uranium, respectively, so they indicate the response per gram of each element: 2.25 counts/s/g of U and 0.843 counts/s/g of Pu. If the uranium and plutonium had no neutronic interactions, the count rates from their mixtures could be calculated from the masses in Table 2. But the two elements do interact (self-shielding and induced fissions) and the degree of interactions are indicated by the errors in the simply-calculated count rates shown in the last column of Table 2. Work is in progress to understand the interactions and to correctly calculate the count rates. Then the process can be reversed and count rates used to find the uranium masses from active-mode shuffler assays, after using the shuffler's passive mode and gamma-ray isotopics to find the plutonium masses.

Table 2. Shuffler Measurements on MOX Standards

Standard	g Pu	g U	Measured Count Rates	Sigma of Count Rates	"Extra" cps from U-Pu
STD1	0.08	1691	3806.2	11.1	-0.07
STD2	436.	1647	3718.0	43.8	-25.96
STD3	87.1	1606	3492.6	6.4	-195.74
STD4	175	1523	3426.4	14.3	-149.26
STD5	349	1352	2964.1	10.7	-373.4
STD6	1308	422	1926.3	6.0	-126.77
STD7	1747	0.15	1473.5	18.2	-0.34

cps/g U = 2.250869

cps/g Pu = 0.843446

FUTURE ACTIVITIES

With the certifications of both shufflers already in place, we can now plan to make accountability measurements on uranium and plutonium. At CMR, the verification measurement activities for the uranium inventory will be continued. Approximately 200 unmeasured items are still awaiting verification or confirmation. A similar program has just begun at PF-4 with about 100 uranium items. We have also accumulated a considerable number of 55-gallon drums containing TRU and uranium waste at both facilities. When the fabrication of the drum standards[3] is completed, we will be able to calibrate the shufflers to assay these waste drums. In addition, we need to complete the investigation already underway on heterogeneous matrices with varied geometries and put in measurement efforts at PF-4 to support the weapon dismantlement program. All these will necessitate completing the studies addressed in this paper such as Monte Carlo calculations of standards, resolving neutron background problems at PF-4, solving the power-

outage safety issue, and completing the "interaction" studies on MOX items.

In the long term, we have a number of important tasks planned for the shufflers. First, we will participate in the DOE Performance Demonstration Program (PDP CYCLE 1) for nondestructive assay this fall to demonstrate the capability and performance of the Los Alamos shufflers in waste characterization. Second, a matrix evaluation and comparison between the shufflers and segmented gamma scanners will provide valuable information to improve measurement accuracy. Accuracy may also be improved by employing neutron activation techniques. Finally, studies will be initiated to explore the feasibility of measuring additional nuclides.

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