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# OPERATING NEW 55-GALLON DRUM SHUFFLERS AT LOS ALAMOS

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## ABSTRACT

Two passive-active shufflers for the assay of uranium and plutonium have begun operation at Los Alamos National Laboratory. An extensive period of safety and technology assessments were made to meet Laboratory and DOE certification requirements. Many design features of the shufflers are in place to assist the operator in using the instruments efficiently, effectively, and safely. A calibration for uranium oxide has been completed and applied to a variety of uranium-bearing inventory materials. A new calibration for MOX materials is nearly complete and additional uranium and plutonium materials will be measured in the near future.

## I. INTRODUCTION

Two passive-active shufflers designed to assay 55-gal. drums of waste for $^{252}$Cf and plutonium have recently begun operation at Los Alamos National Laboratory (LANL): one at the Chemical Metallurgical Research (CMR) Building Waste Assay Facility at TA-3 and the other at the Plutonium Facility (PF-4) at TA-55. Their uses have been extended beyond waste drums to smaller containers with much larger fissile quantities and even to mixed oxide (MOX) materials. These shufflers were designed at Los Alamos and were fabricated by Canberra Industries, Inc. Similar shufflers are at Martin Marietta Utility Services Portsmouth Gaseous Diffusion Plant, Westinghouse Savannah River Site, and Lawrence Livermore National Laboratory.

Shufflers were selected for these applications at Los Alamos for several reasons. They can count passive neutrons from plutonium spontaneous fissions.[1] They can also actively interrogate uranium by counting delayed neutrons emitted after fissions induced by neutrons from a $^{252}$Cf source.[1,2] Shufflers can be applied to a wide variety of homogeneous and heterogeneous materials in containers ranging from small capsules to 55-gal. drums and for fissile masses from a fraction of a gram to multiple kilograms. Their mechanical and electrical reliability makes them suitable for a plant environment.

## II. REQUIREMENTS FOR ROUTINE OPERATION

Safety and technology assessments were required before either the CMR or PF-4 shuffler could be used for any programmatic purpose. A DOE-required certification process was also followed prior to any accountability measurements.

The installations of the two shufflers differ in that the floor of the room in the CMR building could have a 1-ft-deep pit to house a rotation motor and detector bank (Fig. 1) while the floor of the PF-4 room could not be altered. Therefore, the PF-4 shuffler (Fig. 2) is on a platform that required the following additional safety features: a bumper guard was added to the edge of the platform; anti-slip strips were placed around the perimeter of the platform; and a metal stop was added to the platform to prevent drum-loaded dollies from rolling off the platform. In addition, a stepping motor controller was covered with a clear-plastic plate to prevent accidental pressing of control keys. The gap between the platform and the PF-4 floor was filled with neutron shielding so that dose rates on contact with the shielding's surface are below 5 mrem/hr, although one position at the rear of the shuffler has slightly more than this dose rate requiring it to be posted and roped off. Dose rates at these and other operator positions are zero unless the $^{252}$Cf source is inside the assay chamber irradiating a container. Even if assays were done continuously the source is inside the assay chamber only one-third of the time, so time-averaged dose rates are less than 2 mrem/hr in contact with the shielding;

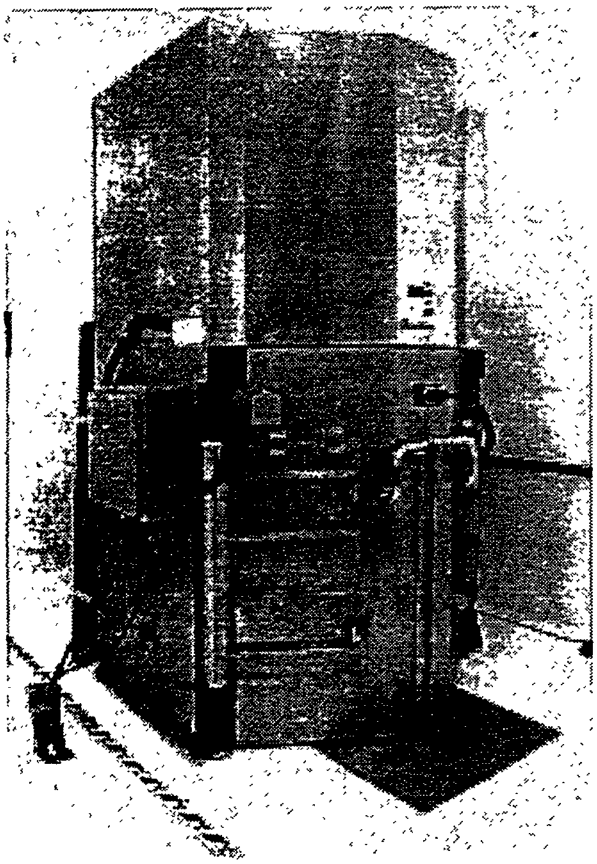

*Fig. 1. The shuffler in the CMR building is shown with the doors to the assay chamber closed. The shuffler's height is about 9 ft. A 1-ft-deep pit in the floor holds the turntable mechanism and a detector bank.*

dose rates drop quickly with distance and 3 ft from the shuffler's surface there is no dose rate requiring posting or regulation.

The CMR shuffler is 40 ft from a portal monitor and as the $^{252}$Cf source moves rapidly between its shielded storage cube and the assay chamber there is a fraction of a second when the gamma-ray background rate in the monitor is increased. If a person happens to pass through the monitor during this time there is an increased probability of an accidental alarm. This effect was greatly mitigated with an additional iron shielding plate near the shuffler.

Both instruments have recently passed the DOE-required certification procedure for active assays of $^{235}$U. The major components of the procedure were the establishment of a measurement control program and failure response plan, a calibration method with certified standards, and a data collection and assessment plan to establish an instrument's performance and initial measurement

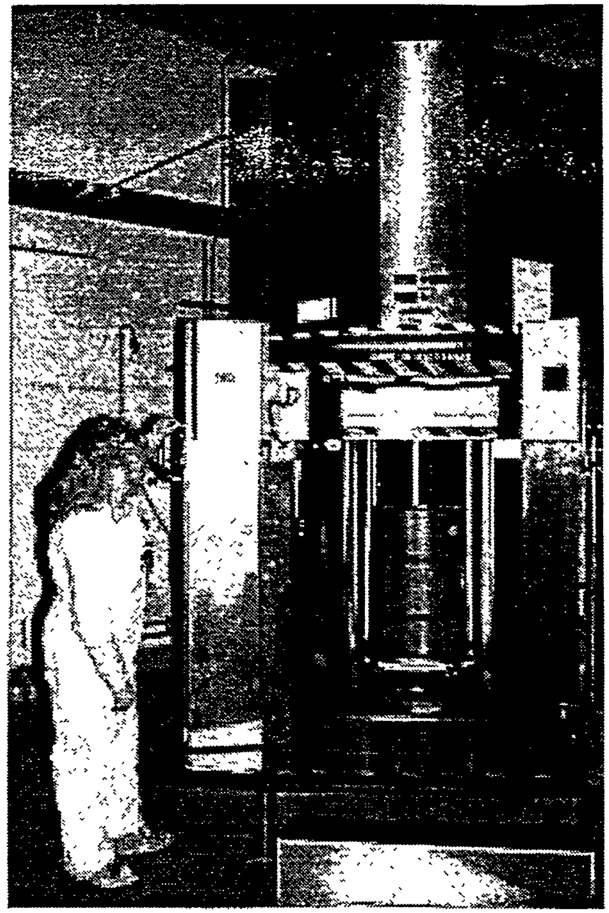

*Fig. 2. The shuffler in the PF-4 is identical to the shuffler in the CMR building, but a pit could not be placed in the floor so it sits on a one-foot-high platform. A 55-gal. drum can be seen inside the assay chamber.*

control limits. Assays on standards were taken repeatedly over an extended time and short-term and long-term variations were analyzed. The only problem was caused by variations in the neutron background rates in PF-4 where no controls were applied to the movement of plutonium in the room with the shuffler. The severity of this problem will be reduced through administrative controls, additional shielding, and software modifications.

## III. SHUFFLER OPERATIONS INTERFACE FEATURES

The shuffler's hardware and software were designed with several features to assist the operators in using it effectively and safely. The major features are described in the following paragraphs.

(a) The $^{252}$Cf source is kept in a shielding storage cube 4 ft on a side (the top halves in Figs. 1 and 2) except when the source is driven into the assay chamber (the botom halves in Figs. 1 and 2) by a stepping motor system to irradiate a container. Interlocks prevent the source from leaving the storage cube if the doors to the assay chamber are not completely closed. Should a door be opened when the source is not stored, the stepping motor system automatically and quickly returns the source to the storage cube; an audible alarm sounds until the source is stored. The stepping motor system's controller senses these interlocks and will respond even if the shuffler's computer is not running. When electrical power to the electronics rack is turned on, the stepping motor controller boots quickly and checks that the source is stored; if it is not, the controller moves the source to the storage position. The computer finishes its booting a little later and automatically runs the shuffler software code; the first computer action redundantly checks the source position and stores the source if necessary.

(b) The storage cube was designed with the aid of Monte Carlo calculations so that 500 micrograms of $^{252}$Cf would pose no radiation hazard to operators. Dose rates in contact with the cube are less than 5 mrem/hr at positions 6 or 7 ft above the ground. But operators are not in contact with the storage cube; their routine closest position is farther away at the door level where the dose rates are essentially zero. During the irradiation phases of assays, the dose rates near the shuffler rise slightly because the assay chamber walls are not as thick as the storage cube; time-averaged dose rates 3 ft from the shuffler are under 1 mrem/hr and, by being more than 3 ft away, an operator's dose rate during an assay is essentially zero. An assay is performed automatically once an operator has started the process, so there is no need for an operator to be near the shuffler except to load and unload containers.

(c) Several status lamps inform operators of (1) the position of the $^{252}$Cf source, (2) if an assay is in progress, (3) if the doors are open, (4) if the detector banks are operating normally, (4) if neutron fluxes in the assay chamber are normal, and (5) if the turntable at the base of the assay chamber is rotating. If an operator wants to check on the position of the $^{252}$Cf source, a quick glance at the lamps will show if the source is stored; if it is not stored, other lamps will show if it is near the assay chamber and causing pulses in the neutron detection system. In any case, should an operator inadvertently open the door, the source will be stored automatically and quickly.

(d) Access to operating the shuffler is controlled by a user name and password feature of the software. Three levels of access are available. "Operators" can perform only routine measurements (assays, calibration measurements, and performance and safety checks). "Supervisors" have all the options of an operator and can over-ride failed performance checks that prevent an operator from proceeding further; a supervisor can also modify the parameters that control the various measurement protocols. "Managers" have all the options of a supervisor but also control valid user names and passwords.

(e) The software can require a safety check on a scheduled basis; if a check is overdue or is failed, measurements cannot be performed by an operator until the check is passed. The sensors for the $^{252}$Cf source and the doors are checked for proper operation, and the status lamps under control of the computer are checked. The time interval between required checks can be set by a supervisor or a manager.

(f) Another check required by the software on a preset time interval consists of two assays: one of an empty assay chamber and a second on a standard item. The two assay results expected here have been placed in a parameter file by a supervisor or manager along with their acceptable deviation limits. The empty-chamber assay is a check that no contamination or holdup is present that would incorrectly increase the assay results of items with low masses of uranium. This option thus checks the instrument for proper operation.

(g) Bias and precision checks for measurement control purposes are automated through the shuffler's software. Repeated assays can be made on a reference standard, as preset by a supervisor or manager.

(h) A calibration measurement option follows the same stages of an assay but the result is a corrected count rate rather than an assay value in grams. A set of such count rates is needed from standards for calibration.

(i) Assay times can be set by a supervisor or manager. The nominal active-assay time is 1000 s, which gives a minimum detectable mass under half a gram when the $^{252}$Cf source has a mass of more than 400 $\mu$g. If the source is much smaller, or an enhanced sensitivity is needed, the assay time can easily be extended. If assays are being done on hundreds or thousands of grams of $^{235}$U the assay time can be reduced without significantly sacrificing count-rate precision.

## IV. SHUFFLER APPLICATIONS

The shufflers were ordered a few years ago for the primary purpose of assaying waste drums, although other containers would be assayed also. Because of programmatic changes other uses have initially been dominant.

A calibration campaign was first conducted for both shufflers using well-characterized, pure standards of uranium oxide powder and the shuffler in the active interrogation mode. Masses from 10 to 1000 g were used in identical containers, with a 2000-g standard in a similar container. Data from the two shufflers were combined to get a single calibration curve for both shufflers. Delayed neutron count rates are a nearly linear function of the $^{235}U$ mass, with about 1.2 counts/s for each gram; a more accurate nonlinear relation is used in the shuffler software. This calibration is appropriate for use on assays of process materials.

The first phase of an inventory verification program was then completed at the CMR building on a wide assortment of $^{235}U$ items that could not be measured before because of the lack of a suitable instrument. Many of the inventory items differed from the standards in important ways involving geometry, impurities, and matrices. Most of the $^{235}U$ masses were 200 g or less, but two were as high as 1000 g. Of the measurements for 46 items, about two-thirds were accepted as verifications; the assays were within 30% of the book values. A few cans had shapes similar to those of the calibration standards and were known to contain pure uranium oxides; the shuffler assays for these cans differed from the book values by only 1% to 15%. The other one-third of the measurements were considered good enough for confirmations; these items were generally impure oxides in containers with shapes greatly different from the pure calibration standards.

The PF-4 shuffler has been used in the active mode to study the assaying of cans of MOX in shipping containers. A gamma-ray spectrum of a container is needed to assign the mixture of plutonium isotopes present, then a passive neutron count yields the plutonium mass, and data from the shuffler's active mode reveals the uranium mass (after subtracting the delayed neutron count rate for plutonium from the measured count rate).

The uranium and plutonium in a container interact by both shielding neutrons from each other through absorption and by irradiating each other with fission neutrons. A simple but accurate expression has been developed for the seven MOX standards to describe these competing interactions. The delayed neutron count rate $r$ from the mixed uranium and plutonium is

$$r = c_U m_U + c_{Pu} m_{Pu} - k_1 m_U m_{Pu} / (m_U + k_2 m_{Pu}) , \qquad (1)$$

where $m_U$ and $m_{Pu}$ are the masses of the two elements (either assuming a constant percentage of $^{239}Pu$ or using only the mass of $^{239}Pu$), $c_U$ and $c_{Pu}$ are the count rates expected from 1 g of uranium and plutonium (as deduced from standards with only one of these elements each), and $k_1$ and $k_2$ are parameters calculated from a best-fit process. Table I shows the data and the accuracy with which Eq. (1) reproduces the measured count rates. The values of $c_U$ and $c_{Pu}$ were computed from STD 1 and STD 7 as 2.2509 counts/s•g-U and 0.84345 counts/s•g-Pu. The best-fit values of $k_1$ and $k_2$ (using STD 1 through STD 7) are 1.99812 and 4.80438, respectively.

TABLE I
SHUFFLER ACTIVE-INTERROGATION MEASUREMENTS
ON MOX STANDARDS

| | Elemental Masses (g) | | Count Rates (counts/s) | | |
|---|---|---|---|---|---|
| Standard | Pu | U | Measured | Calculated | Ratio |
| STD 1 | 0.08 | 1691 | 3806.22 | 3806.13 | 0.99998 |
| STD 2 | 43.6 | 1647 | 3718.00 | 3666.67 | 0.98619 |
| STD 3 | 87.1 | 1606 | 3492.62 | 3550.30 | 1.01651 |
| STD 4 | 175 | 1523 | 3426.42 | 3350.38 | 0.97781 |
| STD 5 | 349 | 1352 | 2964.14 | 3026.25 | 1.02095 |
| STD 6 | 1308 | 422 | 1926.32 | 1888.63 | 0.98043 |
| STD 7 | 1747 | 0.15 | 1473.50 | 1473.78 | 1.00019 |
| STD 2&6 | 1351.6 | 2069 | 5241.22 | 5144.49 | 0.98154 |
| STD 3&5 | 436.1 | 2958 | 6332.54 | 6515.82 | 1.02894 |

Count rates from two containers holding two of the standards are also shown at the bottom of Table I. These data were not used in the fit to find $k_1$ and $k_2$, but their count rates are still accurately described by Eq. (1). Monte Carlo calculations show that the multiplication of such a combination is only slightly different from that of a single can. If a shipping container has three of these cans (with no spacers), it can be expected that calculated count rates from Eq. (1), with the above values of the parameters, will not be as accurate as those in Table I.

In the near future additional calibrations for $^{235}U$ will be possible because a set of capsules of $^{235}U$ will be ready. They will be placed in different numbers and geometrical combinations inside cans and drums with different matrices, extending the range of materials for which the shufflers are calibrated. This will be useful in the continuing effort of assaying unmeasured inventory items at both the CMR and PF-4 locations. There are also many 55-gal. drums of uranium and transuranic wastes at both facilities awaiting measurements by the shufflers.

Cans of plutonium oxide will be routinely assayed in the passive mode with the PF-4 shuffler after it is certified to replace the existing neutron barrel counter. Weapon components may also be measured to study the feasibility of assaying such materials with a shuffler. The assay of fissionable nuclides other than uranium and plutonium may also be studied in the long term.

We expect to participate in the DOE Performance Demonstration Program for Nondestructive Assay this fall to demonstrate the capability and performance of the Los Alamos shufflers in characterizing wastes. A comparison of shuffler and segmented gamma scanner results will help define the range of application of each instrument and perhaps how to use them together to yield results better than either could give alone.

## V. SUMMARY

It took about a year to meet the DOE and Los Alamos requirements for routine operation of the two new shufflers at Los Alamos. But the shufflers have already provided a new nondestructive measurement capability for both the CMR and PF-4 facilities that has been applied to previously unmeasured uranium inventory items and to cans of MOX. The shufflers have a wide range of applications and they will be busy and important instruments at Los Alamos.

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