

*Title:*

INSTALLATION OF PASSIVE-ACTIVE SHUFFLERS AT  
LOS ALAMOS PLANT ENVIRONMENTS

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## INSTALLATION OF PASSIVE-ACTIVE SHUFFLERS AT LOS ALAMOS PLANT ENVIRONMENTS

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### ABSTRACT

Two Canberra-built passive-active  $^{252}\text{Cf}$  shufflers of Los Alamos hardware and software design have been installed and are presently undergoing calibration and certification at Los Alamos National Laboratory. These instruments fulfill important safeguards and accountability measurement requirements for special nuclear material in matrices too dense or otherwise not appropriate for typical gamma-ray techniques. The ability of the shuffler to obtain precise assays under conditions of intense passive emissions of neutrons and gamma rays is a valuable asset in plant environments. This paper reports on the procurement process and the various steps involved in the installation of two shufflers at Los Alamos, one at the Chemical Metallurgical Research (CMR) Building Waste Assay Facility at TA-3 and the other at the PF-4 Plutonium Facility at TA-55. Details are given on the certification procedure including the development of standards, various expected matrices, and calibration. Some safety issues are addressed, and some preliminary performance characteristics are presented based on measured background rates in the plant environments.

### INTRODUCTION

A very important element of nuclear materials processing and safeguards at Los Alamos National Laboratory involves the use of nondestructive assay (NDA) techniques. In general, NDA techniques are able to provide the prompt, quality measurements needed to monitor and control the processing, movement, and storage of nuclear materials, including radioactive waste materials, and to detect their potential unauthorized use or diversion. This capability is even more important in today's dynamic, rapidly changing security climate due to the scaling down of the nuclear weapons program and concurrent reduction of the stockpile. A few years

ago a need was identified at both the CMR building and the Plutonium Facility for a versatile instrument capable of measuring both heterogeneous and homogenous distributions of plutonium and uranium in a wide variety of container sizes and matrices. In addition, the instrument had to be capable of performing the measurements within specified levels of precision, accuracy, and sensitivity within harsh plant environments having high levels of neutron and gamma-ray background radiation. The  $^{252}\text{Cf}$  shuffler was deemed to be the best available, well-proven technology to satisfy this wide range of requirements.

A shuffler is a passive-active neutron instrument for assaying uranium and plutonium. In the passive mode, the shuffler is a coincidence neutron counter for spontaneously fissioning isotopes and uses shift-register electronic circuitry to analyze the neutron signal; it functions in a manner nearly identical to the recently developed neutron barrel counter<sup>1</sup> (NBC) presently being used at the Plutonium Facility. In the active mode, the shuffler induces fissions in uranium by means of a  $^{252}\text{Cf}$  source that "shuffles" between an upper storage block and the assay chamber, producing alternating periods of irradiation in the chamber. The resulting delayed neutron precursors from the fission products are then counted during the storage periods between the irradiations. The theory behind the passive mode (coincidence counting) is described in Ref. 2 whereas that of the active mode (delayed neutron counting) is in Ref. 3.

### PROCUREMENT

Procurement of the two shufflers was by the Los Alamos Nuclear Materials Measurement and Accountability Group (NMT-4). The process included shuffler design, the bidding process, and fabrication.

### Design

The mechanical and electrical designs for the shuffler were created by the Los Alamos Safeguards

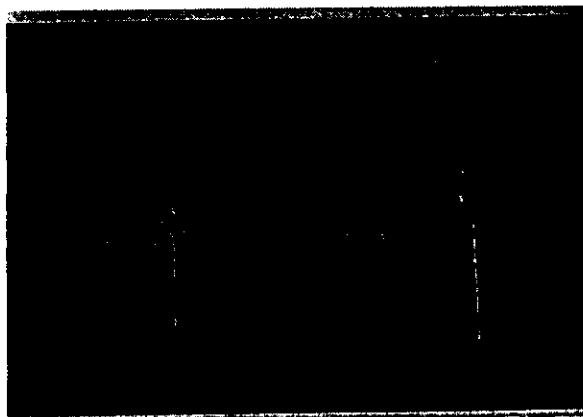
Assay Group (NIS-5). They are very similar to the designs described in Ref. 4, with a few upgrades of hardware components. The software was adapted, with some modification by NIS-5, from existing codes for other shufflers.

The designs include several safety features. There is neutron and gamma-ray shielding to protect workers from the  $^{252}\text{Cf}$  source, an interlock mechanism to quickly retract the source into its storage block of borated polyethylene shielding should the doors be inadvertently opened when the source is being used, mechanical stops to prevent the source from leaving the shuffler's protective shielding, and status lamps to indicate deviations from normal operations.

#### Bidding Process and Fabrication

A specifications package was developed, and an extensive bidding process followed. A manufacturer was sought not just to build these two shufflers but to provide an opportunity for technology transfer, so that the company could provide similar shufflers to other facilities. Canberra Industries, Inc. (located in Meriden, CT) was awarded the contract. There were 6 or 7 bidders, and the entire process took about a year.

Canberra's Nuclear Product Group fabricated and assembled the shufflers in about ten months. The shufflers were mechanically and electrically tested at Canberra prior to shipment to Los Alamos. Figure 1 is a photograph of the two shufflers during the factory acceptance tests.



**Figure 1** This photograph of the two shufflers was taken during the acceptance test at Canberra; some of the test participants are included. The cover plates for the upper halves of the shufflers are not in place, revealing the motors and other accessories that drive the  $^{252}\text{Cf}$  sources. The shufflers are raised one foot above the floor to make room for a rotation motor and detector bank.

## INSTALLATION

#### Preparation

Site preparations were done by Los Alamos, and installation of the shufflers was a joint effort of Canberra and Los Alamos. A small pit was created in the floor in the CMR building to hold the turntable rotation motor and the bottom bank of detectors. The floor of the assay chamber is thus flush with the floor, and drums can be simply pushed into the assay chamber on a dolly. It was not possible to create a pit in the Plutonium Facility, so a one-foot-high platform was built. The rotation motor and detector bank, along with borated polyethylene shielding to prevent scattering of neutrons into the assay room, are below the platform whereas the body of the shuffler rests on the platform. Drums in the Plutonium Facility must be raised one foot to enter the assay chamber, but a mechanical assist already exists in the room so no new equipment was needed to handle the drums. Figure 2 shows the Plutonium Facility shuffler in its work environment.

#### CMR Building

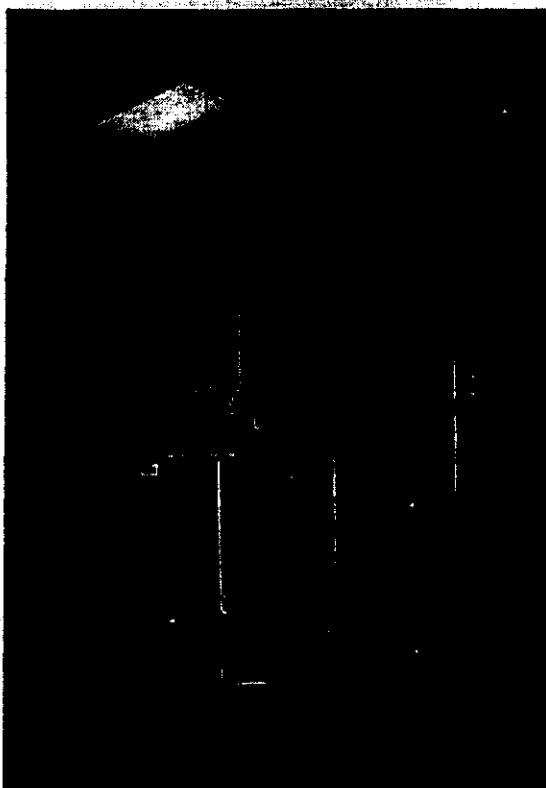
Installation of the shuffler in the CMR building took less than a week. The preparation was minimal other than opening the pit area and filling the adjacent area underneath with sand to strengthen the floor loading for transporting the shuffler. A support transportation team was on site to do the moving and assist with the installation. The base of the instrument was installed first, followed by the assay chamber, and then finally the upper-source storage compartment. The total weight was about seven tons, mainly due to the heavy polyethylene shielding for the neutron source.

#### Plutonium Facility

Two weeks were needed to install the Plutonium Facility shuffler because the working conditions were more restrictive and the platform had to be assembled. In addition, detailed floor loading and seismic calculations had to be done by a Los Alamos structural engineer before the shuffler could be placed on the platform. These calculations were not necessary at the CMR location because that instrument was located over solid ground. On-site acceptance tests were performed at both facilities.

#### $^{252}\text{Cf}$ Sources

Two  $^{252}\text{Cf}$  sources were encapsulated by Frontier Technology Corporation located in Xenia, Ohio. Each source has a mass of about  $560\ \mu\text{g}$  ( $1.3 \times 10^9$  n per second or about 300 mCi); the shielded shipping



**Figure 2** This photograph was taken after installation of the Plutonium Facility shuffler in the non-destructive assay laboratory. The hoist to lift the heavy drums is seen along with the dolly to wheel them into the assay chamber. The electronics rack with the computer console is located behind the hoist. Bolts fastening the shuffler baseplate to the floor for seismic purposes are seen at the base of the front I-beam.

cask weighed 7400 lb. A procedure for handling and installing the sources was developed, and a "dry run" was performed at each location in the presence of facility safety and radiological protection personnel. The source transfer from the cask to the instrument took 1 to 2 minutes.

## CALIBRATION

Calibration in this paper is the process of relating instrument response to the physical attribute of mass. In order to accomplish this with maximum accuracy, well-characterized standards are required.

### Standards

Standards for the shufflers have been designed (with assistance from the Safeguards Assay Group) and are currently being fabricated at the Plutonium Facility at Los Alamos.<sup>5</sup> The design of the standards are similar to ones created for the aforementioned shufflers described in Ref. 4. Quantities of standard

material will be diluted with diatomaceous earth and placed in modular zircaloy capsules, such that each capsule is about 80 % full. These capsules can be loaded in a drum having fixtures to distribute them. The fixtures will consist of various aluminum tubes that run the length of the drums at fixed radii of 12, 20, and 25 cm from the drum axis. Various quantities of capsules containing uranium or plutonium can be placed inside these tubes at various heights during the measurements. It will be possible to obtain mass loadings from the lowest levels of sensitivity to well over 200 g.

### Matrices

Several drums with fixtures are already made and will be loaded with different materials to create various matrices. This is important because accuracy is dependent on the degree of similarity between the contents of the calibration drums and that of the measured items. Because the programmatic environment at Los Alamos is variable and dynamic, it is difficult to make matrix drums to cover all future requirements. However, we can expand the matrices as the need arises. In any case, a general high-density drum containing metal turnings, shavings, piping, tubing, filters, pumps, etc., along with a low-density drum containing paper products, plastic, rubber, etc., will be made initially.

### Procedures

*Active Assays* In each matrix, a set of  $^{235}\text{U}$  standards is measured with the active mode. The standards are arranged within a corresponding matrix drum to simulate a homogeneous distribution. A calibration curve is then fitted to these data. If the calibration is to include more than one enrichment, this process is followed for each enrichment. If a later assay is done for  $^{235}\text{U}$  with some other enrichment value, an interpolation is done among the calibration enrichments.

An additional general-purpose calibration is prepared for those cases when the matrix material in a drum is unknown or not in the list of matrices for which a calibration was done. The built-in flux monitors in the assay chamber are used in this case to characterize the moderating ability of the matrix.<sup>4</sup>

*Passive Assays* The nature of the matrix material has a smaller impact on passive assays<sup>4</sup> than active assays, but it cannot be ignored. The chemical form of the fissile material is also important because of the possible generation of neutrons from alpha-neutron reactions in light elements. Calibration curves are thus needed for plutonium with different chemical compositions in the various matrices.

As the mass of plutonium increases, the neutron multiplication also grows and eventually a calibration curve becomes nonlinear. The advantages of linear calibrations can be retained by a multiplication correction that can be applied in many cases.<sup>2</sup> Alternatively, nonlinear calibrations can be used if the standards are representative of the sample matrices and span the nuclear material mass range.

## OPERATIONAL REQUIREMENTS

### START

At the Plutonium Facility, before the instrument can be used, a "Safety and Technical Assessment for Readiness of Technology" (START) must be performed. Similar requirements are also in place at the CMR location. This procedure ensures that an independent and uniform safety and technical readiness assessment is performed. It also ensures that changes within the facility are independently and objectively reviewed and the results adequately documented. Implementation of START helps to reduce to acceptable low levels all identifiable risks associated with the handling or processing of radioactive and fissionable materials and to protect personnel, equipment, the environment, and essential operations from the effects of potential accidents.

Although implementation of START consists of many elements, the primary one is the establishment of an independent review team whose members are comprised of experts in ES&H, radiation protection, waste management, and technical areas. All concerns must be adequately addressed before the instrument can operate in a routine manner. For example, referring to Figure 2, rubber gloves can be seen taped to the sharp corners of the base plate as a temporary measure. Another concern is that the dolly holding the drum could roll off the platform. These and many more safety issues will have to be resolved.

### Certification

In accordance with DOE Order 5633.3A, before an NDA instrument is permitted to be used for nuclear material accountability measurements it must undergo a process known as "certification." At Los Alamos, the primary component of certification consists of a plan comprised of about 16 requirements. Of these, the most important are the establishment of a measurement control program and failure response plan; a calibration method with certified standards that support the desired operating range and a procedure to establish the calibration uncertainty; and a data collection and assessment

plan to include a set of measurements to establish the instrument stability, measurement control limits over the range of operation, and a comparison to an existing qualified superior technique, if possible. Also, operator training with documentation has to be established and an "uncontrolled" copy of the safe operating procedure has to be provided.

After the certification data are acquired, the analysis results are reviewed by a Laboratory statistician, the technical support section of the Material Control and Accountability Group, and the technical support section of the Nuclear Materials Measurement and Accountability Group. This certification process typically takes 3 to 6 months to complete.

## INITIAL INSTRUMENT PERFORMANCE

### Precision

Precision, expressed as a fraction or percent, is the relative standard deviation of a set of repeated measurements on the same sample of fissile material. The precision requirements were specified for the shufflers to be less than or equal to 5% (1 sigma) for 15 consecutive measurements on standards containing about 10 g of <sup>240</sup>Pu effective or 100 g of <sup>235</sup>U for assay times of about 1000s.

*Active Mode* The precision of an active assay depends on the mass of <sup>235</sup>U present, the strength of the <sup>252</sup>Cf source, the time allowed to perform an assay, and the background count rate. Precision can be calculated for various combinations of these parameters once the delayed neutron and background count rates have been measured under a specified set of conditions [Ref. 4, Eq. (6)].

Calculated precisions for these shufflers are shown in Figure 3 for various <sup>235</sup>U masses and background rates. The background count time is 270 s, the delayed-neutron count time is 238 s, and the delayed-neutron count rate per gram of <sup>235</sup>U is 12 counts per second per gram. The delayed-neutron count rate varies with the <sup>252</sup>Cf source strength and the amount of moderator in the drum, in addition to the <sup>235</sup>U mass. The relative precisions in Figure 3 are thus only crudely representative; the actual precisions will be determined from measurements in the near future.

The high background rate for the Plutonium Facility shuffler limits the precision more severely than usual, but the expected precisions will still meet the requirements.

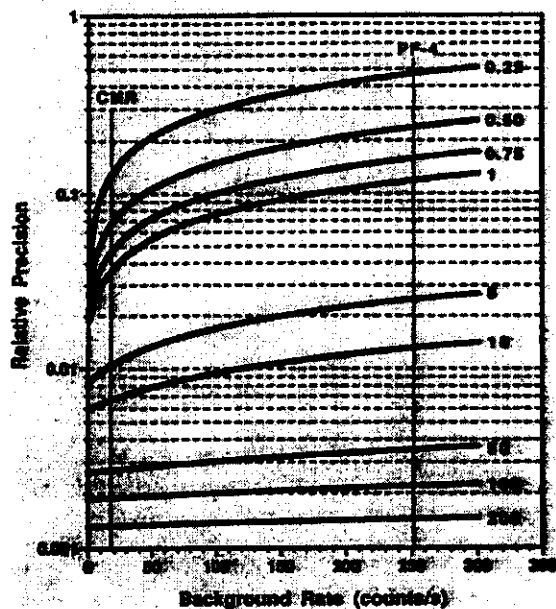


Figure 3 The relative precision (expressed as a fraction) for this type of shuffler in the active-assay mode is shown for a wide range of background rates and various masses of  $^{235}\text{U}$  (shown to the right of the curves). These values were calculated for a mid-sized  $^{252}\text{Cf}$  source of  $400\text{ }\mu\text{g}$  and a specific response rate of 12 delayed neutrons per second per gram of  $^{235}\text{U}$ . A larger source or a larger specific response rate would improve the precision (i.e., produce a smaller precision). The background rate in the CMR is much lower than in the Plutonium Facility where many sources of neutrons are nearby. Background rates are indicated by the labeled vertical lines.

**Passive Mode** The precision of a passive assay depends on the masses of spontaneously fissioning isotopes of plutonium, the assay count time, the counting efficiency of the shuffler, and the background count rate. This precision can also be calculated for combinations of these parameters that are inconvenient to measure [Ref. 6, Eq. (A-8)].

Some representative precisions are shown in Figure 4 for the wide range of total-neutron background rates expected for these two shufflers. The count time is 400 s, with 18 coincidence counts per second per gram of  $^{240}\text{Pu}$  effective<sup>2</sup> and 186 total neutron counts per second per gram of  $^{240}\text{Pu}$  effective; the background coincidence rate was 0.25 counts per second and the coincidence gate width was 128  $\mu\text{s}$ .

The precision for small plutonium masses is strongly limited by the background rate, so the high background at the Plutonium Facility shuffler is especially important for less than 5 g of  $^{240}\text{Pu}$  effective. The precision requirements will be met even with the background in the Plutonium Facility room.

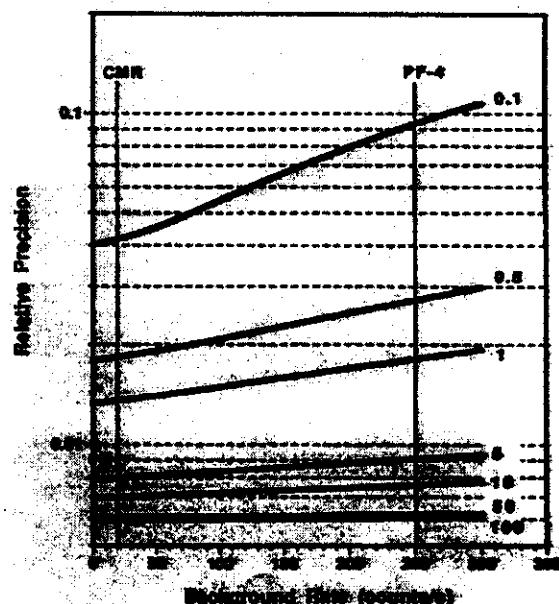


Figure 4 The relative precision (expressed as a fraction) for this type of shuffler in the passive-assay mode is shown for a wide range of total neutron background rates and various masses of  $^{240}\text{Pu}$  effective masses (shown to the right of the curves). For masses greater than 100 g the changes are too small to attempt to indicate on the plot. The background rates in the CMR and Plutonium Facility buildings are indicated by the vertical lines.

### Sensitivity

Sensitivity in this paper means the smallest mass that produces a signal three times as large as its uncertainty (including the contribution to the uncertainty from the background). This corresponds to a relative precision of 0.333. The minimum detectable mass (or sensitivity) is closely related to precision; as precision improves (becomes a smaller percentage), the minimum detectable mass gets smaller. Sensitivities can be estimated from curves like those in Figures 3 and 4 by finding where a curve crosses the relative precision value of 0.333. For example, for a background rate of 150 counts per second, the sensitivity is 0.25 g of  $^{235}\text{U}$ . However, it is more accurate to solve the precision equation for the minimum mass directly.<sup>4</sup>

### Accuracy

Accuracy is usually limited by the calibration standards, although matrix effects can also be very important. The standards for these shufflers are well characterized and documented, so that variations with positions of the fissile materials within a moderating matrix will be important for accuracy.

The degree of inaccuracies with the active and passive modes has been described in Ref. 4. A

scheme for improving the accuracy through a position correction technique has been explored and will be applied to these shufflers. Effects for representative matrix categories will be assessed in order to improve measurement accuracy at the two facilities. It is expected that the inaccuracies will be reduced by at least a factor of two, and hopefully more.

## FUTURE ACTIVITIES

Immediate future work will involve completing the fabrication of the standards, completing the calibration, addressing the issues and findings from the START review, and finishing the certification process in order to be able to support the material control and accountability (MC&A) program. These activities will probably require at least six months and perhaps a year or more.

In addition to routine MC&A measurements, support will be provided to other programs, such as weapons dismantlement and the Laboratory Inventory Verification Program. Studies will also be done to broaden the measurement capability (e.g., measure additional nuclides) as well as improve the overall accuracy by continuing the aforementioned position studies and matrix studies involving various geometries. The intent is to present these results at future meetings.

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