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SRS DELAYED NEUTRON INSTRUMENTS FOR SAFEGUARDS MEASUREMENTS (U)

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Background. The Savannah River Site (SRS) includes a variety of nuclear production facilities that, since 1953, have processed special nuclear materials (SNM) including highly-enriched uranium (>90% 235U), recycled enriched uranium (~50% 235U + 40% 239U), low burnup plutonium (> 90% 239Pu + < 6% 240Pu) and several other nuclear materials such as heat source plutonium (238Pu). DOE Orders, primarily 5633.3, require all nuclear materials to be safeguarded through accountability and material control. Accountability measurements determine the total amount of material in a facility, balancing inventory changes against receipts and shipments, to provide assurance (delayed) that all material was present. Material control immediately detects or deters theft or diversion by assuring materials remain in assigned locations or by impeding unplanned movement of materials within or from a material access area. Goals for accountability or material control, and, therefore, the design of measurement systems, are distinctly different. Accountability measurements are optimized for maximum precision and accuracy, usually for large amounts of special nuclear material. Material control measurements are oriented more toward security features and often must be optimized for sensitivity, to detect small amounts of materials where none should be.

Measurement Criteria. For safeguards applications, significant errors in measurement should not be caused by material form or matrix. However, significance of specific errors differs between measurements made for accountability or for material control. Overestimation is most serious for accountability but underestimation is more serious for material control (as well as for nuclear criticality safety, a frequent second use for measurement data). For accountability, errors must be avoided for probable variations in measured materials but for material control, improbable (but still possible) scenarios must also be considered.

Forms of materials to be measured in Savannah River Site (SRS) processes include finished product, process intermediates, scrap and waste. Most finished product items approach the ideal for high accuracy measurements, being very well characterized and uniform (both stoichiometric and physical form). Noninvasive, nondestructive assay (NDA) measurement techniques are required when finished items or sealed containers of such material must not be broached for sampling. Process intermediates include less pure and uniform materials that may not be suitable for sampling for destructive analysis because of form but must be measured for timely accountability. Scrap is often not uniform enough to permit representative sampling but must be measured before reuse, storage, or shipment. Waste forms may be any process effluent, solid, liquid, or gaseous, all of which have highly variable compositions, special nuclear material content, and distribution. Representative samples cannot be obtained and special nuclear material content must be determined in bulk form. Containers for any of these materials vary over a wide spectrum with size, shape, and material of construction determined by production, shipping, or safety personnel or, often arbitrarily, by some process designer and usually without consideration of measurement needs.

Measurement Design. Design of equipment used for safeguards measurements depends on: isotopic content, material form and matrix plus their range of variability, containers and their handling requirements plus the basic intent of the measurement, accountability or material control (security). Most NDA measurements use gamma or neutron counting techniques. Alpha and beta particles have little penetrating power and have not been useful. Passive techniques depend on measurements of natural radiation emitted by the material being measured. Active measurements usually employ external radiation and measure the attenuation (densitometry, etc.) or excite a reaction and measure the result. Natural gamma radiation from special nuclear material such as 235U and 239Pu is reduced by half or more for each one millimeter thickness of the special nuclear material metal and less but significantly (depending on accuracy required) for small amounts of low-density matrix. NDA systems measuring gamma radiation have traditionally underestimated the special nuclear material. No method to fully compensate this error has yet been demonstrated but significant progress has been made recently. Although high-energy gammas, such as from fission, do penetrate much better, no practical detector with adequate detection efficiency has yet been demonstrated.

Low-energy neutrons have the same poor penetration problems as do low-energy gammas in many materials. Also, no detector has been demonstrated for high-energy neutrons that
measurement period of up to 600 seconds. To achieve full precision, these instruments, called shufflers, usually provide delayed neutrons per gram special nuclear material rarely include less than 5%. 240Pu, measurements usually depend on these passive neutrons. However, materials containing 235U usually emit less than .001 neutron per second per gram special nuclear material, inadequate for most purposes. (Passive neutron counting for some uranium materials producing (α, n) reactions are usable but these materials are not common in SRS processes.)

However, 235U can be induced to fission by exposing it to a strong neutron flux. This irradiating flux is produced in several ways in a variety of active neutron assay instruments. Active coincidence and multiplicity counters use (α, n) sources such as AmBe to generate random (not fission pair) neutrons. The neutron coincidence counting electronics are designed to separate neutrons produced in pairs from induced fissions of the special nuclear material and the random single neutrons from the AmBe. About 2x10^5 source neutrons are required for each pair of fission neutrons measured. Measurement performance is limited, increasingly for smaller samples (<25 g) by the statistical probability of mistaking two random neutrons, which have a significant lifetime in common detector assemblies, for a coincident pair from fission. Size of measurable containers is also by limited source geometry to about 8 inches diameter and 12 inches high.

Special nuclear material, when induced to fission, continues to produce neutrons (delayed neutrons) after the exciting flux is removed. As unstable fission fragments decay, many release additional neutrons for several seconds. The limitation in minimum detectable quantity of coincidence counters thus can be avoided by using a neutron source that can be stopped or removed from the detector area to allow these delayed neutrons to be counted without interference from the source. Small electronic accelerators called D-T guns ("Zetatrons") are available but have had limited life, are only marginally stable, and generate neutrons that are energetic enough to cause interfering fissions in isotopes other than the special nuclear material. Use of these electronic sources has been limited to less accurate measurements; although recent improvements and the availability of D-D guns producing lower neutron energies may increase applicability of the electronic sources.

The usual (α, n) sources are physically large, massive, and difficult to manipulate. However, small 252Cf sources emit 2.34x10^8 fission neutrons per 100 micrograms (typical useful size). A container of special nuclear material is irradiated long enough to induce a reasonably stable delayed neutron-generation rate, 5-10 seconds. This source is then quickly withdrawn to a fully shielded storage location. Detectors surrounding the special nuclear material container then count the rapidly decaying delayed neutrons, about 10 seconds. Since only a few delayed neutrons per gram special nuclear material can be counted, this irradiation and counting cycle must be repeated to accumulate enough counts for acceptable measurement precision. These instruments, called shufflers, usually provide 15 to 30 irradiation-counting cycles per assay, resulting in a measurement period of up to 600 seconds. To achieve full sensitivity, background is usually counted before shuffling for about 300 seconds.

Delayed Neutron Measurement Systems. At the Savannah River Technology Center at SRS, the 252Cf Neutron Activation Analysis Facility (Figure 1) was designed and installed in the early 1970s. An intense 100 mg (~50 Ci) 2x10^7 n/s 252Cf source in a D_2O moderator is surrounded by a water shield. A pneumatic transfer system shuttles small plastic capsules containing samples through one of several plastic tubes into the vicinity of the source. After a short irradiation period, capsules are quickly shuttled to remote neutron and gamma counting chambers. This facility has been used to measure fission and other constituents of ore, environmental, and general research samples, and continues in operation.

Since the mid 1970s when shufflers were developed into a viable measurement system by the (now) N-1 Safeguards Assay Group at the Los Alamos National Laboratory (LANL), six shufflers have been installed at SRS (plus another being fabricated and one in the proposal stage). These shufflers have been employed to measure a wide variety of 235U bearing materials ranging from one instrument for high accuracy accountability assay of finished product, one for precise assays of product intermediate within another process, two for assays of packages of scrap for each of these processes, two for material control evaluation of waste from the two previous processes, plus one (in fabrication) for waste in drums from a third process.

One of the scrap shufflers was installed at SRS in 1978, the first shiffer outside the LANL development laboratory. This instrument, with subsequent electronic and program upgrades has continued in operation since installation. (A photograph and a simplified diagram are shown in Figures 2 and 3.) This shuffler is about 4 ft x 4 ft x 9 ft long with the 8-inch diameter

![Figure 1. 252Cf Neutron Activation Analysis Facility](image-url)
by 14-inch high assay chamber below the sample handling crane, centered in one 4 ft cube, the shielded source storage position centered in the other 4 ft cube. Most of the volume of the shuffler is occupied by polyethylene to shield personnel from the 0.3 Ci source (a lead liner is also included to reduce exposure from neutron capture gammas). The source is moved between storage and irradiation positions quickly (< 1 second) and reproducibly using a flexible drive cable housed in a stainless steel tube, driven by a stepping motor. The irradiation position, radial to the scrap can axis is behind a nickel and iron flux scattering, moderating and spreading assembly next to the assay cavity. The assay cavity is surrounded by 16 3He proportional counters in polyethylene moderator banks. An additional detector bank, with a rotator for the scrap can, is located beneath the chamber. The cavity is lined with cadmium to maintain an energetic neutron flux in the sample by blocking reflection of low-energy neutrons from the detector moderator. Irradiation times of slightly more than ten seconds are followed by restoring the source in less than 1 second and a delayed neutron counting time of about 10 seconds. These 20-second cycles are repeated up to 30 times to obtain a precision of 0.5% for 2 kg 235U. Background counting for 300 seconds allows a minimum detectable quantity of a few grams to be achieved in a total assay time less than 17 minutes. This shuffler was designed for a 235Cf source of 800 μg (~500 mCi) emitting about 2x10^8 neutrons per second. About two feet of polyethylene shielding plus lead are required to reduce personnel exposure (ALARA, now 0.25 mRem) for uncontrolled continuous exposure at 30 cm from any normally accessible surface. This is the main reason that shufflers are large and typically weigh 5 tons.

Upon installation, leakage radiation did exceed acceptable levels. Lead sheets were added to the sides to reduce the gamma component and occupancy in the vicinity was restricted. However, operators need to attend to a shuffler only for short periods during sample changes between assays and such restriction presents no problems. This is the only shuffler known where the 235Cf source capsule became disconnected from the drive cable (during initial acceptance testing at 3RS) leaving the source near the irradiation position. After much conferring and developing special procedures, the drive cable was simply pushed into contact with the source capsule, twisted to rethread it, and the capsule was easily removed. Since then, better set screw design and Locitite have prevented further occurrences. The primary problem with this first production shuffler was the extreme (> 30%) vertical nonuniformity of the irradiating neutron flux in the sample cavity because of the radial presentation of the source to the sample. Measurement response is a strong function of scrap can height, only partially compensated by using counting ratios of the side to the bottom detector banks. Additional measurement errors also result from neutron energy changes caused by variable moderation in sample (water or low-density matrix, a problem with all shufflers), from sample density variability (small) and from isotopic variations (small response to even isotopes).

Neutron flux monitors installed in the sample cavity were used in this shuffler to measure the irradiation flux because the half life of 235Cf was not well known. Later shufflers usually use such detectors to evaluate neutron flux energy shifts due to sample moderation. Primary limit to sensitivity (minimum detectable quantity = MDQ) of about 1.6g 235U is the uncertainty in background caused by neutron leakage from the stored source, cosmic particle interactions, and neutrons generated in measured materials (small). Performance for most reasonable materials (and the most desirable), however, is not degraded worse than 1%, 1σ precision and standards are of sufficient accuracy not to degrade this significantly.

The next three SRS shufflers, begun in 1982, included two of similar design, both installed below gloveboxes to handle potentially contaminated containers “in-line” (Figures 4 and 5). One of these was used to assay sealed cans of finished product before transfer to storage before shipping. These shufflers achieved a measurement precision of 0.12%, 1σ at delivery and better than 0.25% at the end of five years (~two half-lives) source life. A set of representative calibration standards developed for these shufflers achieved a traceable accuracy better than 0.04% ± 1σ. Total measurement accuracy of 0.13% was therefore achieved. Design of these shufflers provided a significantly improved uniformity in the vertical sensitivity of the measurement cavity by placing the source guide tube vertically to parallel the axis of the sample. During each irradiation cycle, the source is scanned repeatedly along the axis of the rotating item. Material containers were 5 inches high and 3 inches in diameter. Very close positioning of the source guide tube to the sample increased coupling efficiency so that a 150 μ
Figure 4. In-line Shufflers With Gloveboxes

The 252Cf source could induce over 10^6 counts during a 1000 second assay. Minimum detectable quantity for these shufflers was about 4 grams, primarily limited by neutron leakage from the stored source, a result of the short distance between the storage and measurement cavity positions required by the installation. The very close positioning of the source to the measured material during irradiation accentuates sensitivity to changes in this distance and required tight positioning controls for design and use, which were achieved.

The second of these shufflers was used to assay a wide variety of scrap materials from throughout the process, which were contained in similar small cans. Significant variations in moderator content of this scrap, mostly moisture, caused wide deviation in neutrons detected per gram 252Cf, an inherent problem with any neutron measurement system. An attempt can be made to derive compensation for these errors in response caused by shifts in the neutron energy spectrum. Neutron flux levels in the cavity may be measured with additional detectors (flux monitors) or the shift in neutron energy may be evaluated using the ratio of counts from two rings of detector tubes placed at different depths in the detector moderator surrounding the cavity. Design of these shufflers provided both possibilities.

The most difficult part of any such error evaluation is development and acquisition of suitable standards truly and accurately representing only a single parameter variation in measured materials. Determination of suitable corrections was underway when operation of this production facility was terminated. Both of these shufflers have since been transferred to another user outside SRS. Radiation leakage from these two shufflers (with new sources) exceeded allowable personnel exposure rates, this time neutron levels. Polyethylene slabs of 4 inches had to be added at the rear and sides plus a one foot area was roped off at the rear.

The third shuffler installed in this facility was designed to provide material security screening of solid process waste packaged in 55 gallon steel drums (or any other smaller package). This was the first barrel shuffler (Figures 6 and 7) other than a prototype tested previously at LANL. This large shuffler provides a sensitivity better than 2 g 252Cf and a precision of about 1%, 1% for 100 g 235U. Significant assay problems caused by presence of neutron energy moderators in waste frequently resulted in errors of two in measured values.
amount of lubricant on the flexible drive cable formed a thick grease and overloaded the motors. Cleaning and adding a small pore air filter to the source drive tube vent eliminated the problem.

The fifth shuffler installed at SRS (Figures 8 and 9) was designed to provide high precision accountability measurements of in-process solid metal billets prior to their extraction into SRS reactor fuel tubes. This shuffler has nearly optimum coupling between the source and measure item because the source travels through the center of the hollow, cylindrical UA1 billet. Only 50g $^{252}$Cf is needed to achieve a measurement precision of 0.3% in a 500 second assay of 1 kg $^{235}$U. A new program package in the "C" language developed for this application is the base for all current shufflers. The small hole through the billet provides no room for neutron scattering assemblies, which would slightly reduce the average energies of the $^{252}$Cf neutrons. These higher energy neutrons therefore result in some small response from the $^{238}$U present in the recycled U used for these fuel tubes. Methods to compensate for this error in accounting for the $^{235}$U special nuclear material have been provided and suitable corrections are being developed. In the meantime, a full set of standards, including sets with differing isotopic mixes, allows calibration of a family of curves. Entry of the known $^{236}$U enrichment of each billet allows interpolation of an accurate result. Sensitivity of this shuffler is better than 0.5g $^{235}$U and exposure at any accessible location is below 0.5 mRem.

The most recent shuffler delivered to SRS and a similar one currently being fabricated are designed to provide materials security screening of drums of waste (or smaller items) as they are removed from two SRS uranium processing facilities. Both of these shufflers (Figures 10 and 11) incorporate a unique,
"pass-through" design using two doors, plus suitable security features, to allow them to be installed across the boundary of the material access area. Waste loaded through the input door is assayed and, if below limits, is ejected through the exit door without need for additional security checks or administrative controls normally required for exiting material. If the package is over special nuclear material limits or overweight, etc., the exit door remains closed and the package is automatically rejected through the input into the material access area. The shuffler, which has been delivered, includes a large, "smart" floor-level conveyer to assist in package handling. Sensitivity (per LANL measurements) is better than 3 g 235U, worst case in an assay time of 1000 seconds and better than 0.6 g 235U for a solid source in an average box of waste paper (moderating). Also unique to this design, the 232Th source storage location is below ground to save shielding weight, space and expense and to achieve minimum personnel exposure. This shuffler also meets personnel exposure limits of less than 0.5 mRem at any accessible location.

The shuffler being built is similar but lacks a conveyer. Source storage will be in a hole in an adjacent 51-inch thick concrete wall since this shuffler will be installed on the second level of a building. A unique requirement for this shuffler is the need to screen waste from both U and Pu processes in that building. A segmented gamma scanner will also be installed in conjunction with this shuffler and each item of waste must be measured by both instruments. The segmented gamma scanner, which is not a "pass-through" design will reject packages, especially those with excess Pu, for reprocessing in the material access area. The segmented gamma scanner will also communicate isotopic data to the shuffler so that the proper calibration curve may be applied. This shuffler will provide a new passive-active measurement analysis to acquire more information on Pu-bearing materials. Ideally, for optimum safeguards as well as maximum operating efficiency, both segmented gamma scanner and shuffler measurements would be made simultaneously in a single instrument. This is not feasible with currently available gamma detectors and equipment.

A replacement for the original scrap shuffler has been proposed (Figure 12) and discussed with LANL investigators and designers. Goals would be to: employ the design now used to achieve highly linear vertical irradiation in the measurement cavity, revise the detector array to increase vertical linearity of detection and lower detector assembly cost, greatly increase coupling efficiency between the source and measured item to allow a smaller source and less personnel shielding with improved performance, simplify accessibility for sample handling without excessive lifting, and to simplify mechanical design and assembly for ease of maintenance and decreased cost. However, uncertainty about probable continued operation of the production facility or the reactors which use this type fuel has halted this project. Instead, since an operative scrap assay system is mandatory for safeguards, especially during facility shutdown and restoration, the current shuffler is being upgraded with a new computer, programs (version of the Billet Shuffler program), electronics and source drive. These should significantly improve operation of this 16-year-old instrument.

REFERENCES

Figure 12. Replacement Scrap Shuffler

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