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METHODS FOR NONDESTRUCTIVE ASSAY HOLDUP MEASUREMENTS IN SHUTDOWN URANIUM ENRICHMENT FACILITIES*

R. C. Hagenauer and R. L. Mayer, II
Martin Marietta Energy Systems, Inc.†
P. O. Box 2003
Oak Ridge, Tennessee 37830-7319
(615) 574-8835

ABSTRACT

Measurement surveys of uranium holdup using nondestructive assay (NDA) techniques are being conducted for shutdown gaseous diffusion facilities at the Oak Ridge K-25 Site (formerly the Oak Ridge Gaseous Diffusion Plant). When in operation, these facilities processed UF_6 with enrichments ranging from 0.2 to 93 wt % ^{235}U. Following final shutdown of all process facilities, NDA surveys were initiated to provide process holdup data for the planning and implementation of decontamination and decommissioning activities. A three-step process is used to locate and quantify deposits: (1) high-resolution gamma-ray measurements are performed to generally define the relative abundances of radioisotopes present, (2) sizable deposits are identified using gamma-ray scanning methods, and (3) the deposits are quantified using neutron measurement methods. Following initial quantitative measurements, deposit sizes are calculated; high-resolution gamma-ray measurements are then performed on the items containing large deposits. The quantitative estimates for the large deposits are refined on the basis of these measurements. Facility management is using the results of the survey to support a variety of activities including isolation and removal of large deposits; performing health, safety, and environmental analyses; and improving facility nuclear material control and accountability records.

I. INTRODUCTION

In 1987, a nondestructive assay (NDA) measurement survey program was established at the Oak Ridge Gaseous Diffusion Plant (ORGDP) to support decontamination and decommissioning (D&D) activities of the shutdown gaseous diffusion process buildings. The program's objective is to characterize residual uranium contained in diffusion process equipment and ancillary piping of the shutdown process buildings to provide information for D&D, material accountability, and safeguards purposes.

Uranium hexafluoride (UF_6) with enrichments ranging from 0.2 to 93 wt % ^{235}U was originally processed in five major process buildings located at the plant, which is now known as the Oak Ridge K-25 Site. When enriching operations were discontinued, the UF_6 gas was evacuated from the process systems; solid uranium deposits, however, have remained in the equipment and piping. The majority of these deposits were formed during normal operations when UF_6 process gas reacted with the metallic surfaces of the process equipment and with water vapor that entered the subatmospheric process systems with ambient air. The metal-UF_6 reactions produced a relatively diffuse deposit of uranium fluorides. The water vapor-UF_6 reactions tended to be more highly localized in the immediate vicinity of leaks and produced deposits of UO_2F_2. Additionally, localized deposits of UF_6 may be present in areas where process gas freezeout occurred prior to shutdown. Because of the presence of these uranium deposits, it was necessary to establish an NDA survey program to provide process holdup data for the planning and implementation of D&D activities.

Because standardized NDA methods for measuring holdup did not exist prior to this survey, new measurement methods were developed to quantify these deposits. The initial NDA measurement effort focused on surveying the two process buildings that processed uranium with enrichments above 15% ^{235}U.[1] Because equipment used to process low-enriched uranium (LEU) is significantly larger than the equipment used to process high-enriched material, different measurement methods were developed for each equipment size measured. The focus of this paper is to discuss NDA measurement methods used to locate and quantify large uranium deposits in the LEU gaseous diffusion process buildings.

II. MEASUREMENT APPROACH

A gaseous diffusion plant process building is comprised primarily of many repetitive stages of process equipment connected in series. Each stage consists of (1) a large cylindrical vessel called a diffuser, or converter, that contains the diffusion barrier; (2) a compressor used to compress the gas to the pressure needed for flow through the barrier tubes; and (3) process piping for stage and interstage connections. Stages are arranged into groups of 8 or 10 stages, called cells, which are the smallest unit of production that can be taken

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out of service for maintenance or other reasons. Associated auxiliary equipment includes chemical absorbent traps, purge equipment, surge tanks, feed and withdrawal stations, and cold traps. When ORGDP was operating to produce LEU, the cascade consisted of nearly 1600 stages contained in three separate process buildings. Because of the random distribution of the uranium deposits in the process equipment and piping, the process buildings now are being nondestructively surveyed in their entirety.

A flowchart of the NDA measurement approach used in each LEU process building is shown in Fig. 1. The measurement survey consists of three major components: (1) ***isotopic mapping*** of selected items to identify radionuclides present in the deposits and to determine the isotopic composition of those nuclides, (2) ***scanning*** of the entire building to locate sizable deposits of uranium within process equipment and piping, and (3) ***quantitative measurement*** of the large deposits. Isotopic measurements are made at several selected locations and averaged to provide enrichment values for specific sections of the building. Based on the quantitative measurement data and average enrichment values, the ^{235}U and uranium quantities are estimated for each item identified as containing a sizable deposit. The items that contain the largest deposits are identified, and isotopic measurements are taken of each of these items. Quantitative measurement results for these items are then refined based on the results of the isotopic measurements.

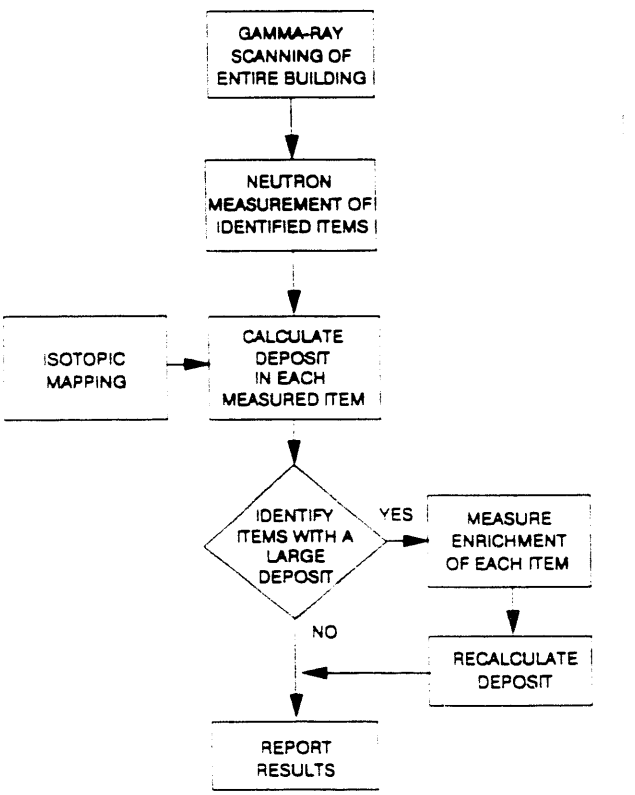

Fig. 1. LEU building measurement approach.

A. Isotopic Mapping

Isotopic mapping involves conducting measurements using high-resolution gamma-ray (HRG) measurement instrumentation and then analyzing the spectra to determine (1) ^{235}U concentration of the uranium contained in the measured process equipment and (2) if unexpected isotopes are present that can interfere with NDA quantitative measurements. Because low- and medium-energy gamma rays are severely attenuated by the walls of process equipment, as well as by thick uranium deposits, these HRG enrichment mapping measurements are typically performed on piping, process openings, or thin-walled items such as expansion joints and vacuum balances. Results from isotopic mapping are used to establish the average ^{235}U concentration in a given area of the process building and to determine the relative abundance of unexpected isotopes. HRG measurements are also performed on individual items initially identified as containing a large deposit to obtain item-specific enrichment values.

Gamma-ray spectra are obtained using planar and coaxial hyperpure germanium (HPGe) detectors connected to portable multichannel analyzers. Planar detectors are used to measure thin-walled items because they have better resolution and better efficiency for low- and medium-energy gamma rays that can penetrate thin-walls. Coaxial detectors are used to measure deposits in thick-walled containers, such as converters and compressors, because they have high efficiency for the high-energy gamma rays that are capable of penetrating the container walls.

To conduct an HRG measurement, a low-resolution gamma-ray detection system is used to locate a position on a piece of equipment from which sufficient gamma rays are being emitted to permit acquisition of a high-quality spectrum within a reasonable timeframe (e.g., less than one hour). The HPGe detector is positioned to measure the gamma rays from the item of interest while minimizing the detection of gamma rays from nearby sources (shielding or collimation is frequently required to screen out unwanted activity). The spectrum is collected and recorded on tape or disk. Spectra from the analyzer are subsequently transferred to an IBM-compatible personal computer and stored on diskettes for further analysis.

B. Scanning

Deposits are initially identified by systematically scanning the process equipment and piping using gamma-ray measurement techniques. In general, the size of the uranium deposit is proportional to the intensity of the measured gamma-ray signal. Gamma-ray measurement methods are usually more sensitive than neutron methods for detection of ^{235}U deposits. Because uranium and its daughter products emit gamma rays over a wide range of energies, gamma-ray scans are conducted using detectors and analyzers programmed to detect gamma rays from 25 to 2700 keV. For thin-walled (e.g., less than 1 cm) pieces of process equipment, detection of the primary gamma rays from ^{235}U (i.e., 144, 164, and 186 keV) is the key indicator of a uranium deposit. For thicker-walled items, the higher-energy (1001 keV) gamma rays emitted by daughter products of ^{238}U are not severely attenuated and are used as a deposit indicator. Thus, sizable

LEU deposits can be detected in virtually all types of process equipment using gamma-ray scanning methods.

Two types of instruments are used to conduct scanning operations in the LEU process buildings: (1) hand-held sodium iodide (NaI) gamma-ray detectors (typically 7.6-cm-diam. 7.6-cm-thick crystals surrounded by 3-mm-thick lead shielding) and (2) a cart-mounted NaI gamma-ray detector (12.7-cm-diam. 7.6-cm-thick crystal surrounded by a 6.3-cm-thick lead shield weighing approximately 1320 kg). The cart-mounted detector, referred to as an elephant gun, is more sensitive than the hand-held NaI detectors; however, because of its weight, it cannot be positioned close to many of the items. Initial scans are conducted using the elephant gun. Subsequent scanning using the hand-held NaI detectors determines exact locations of deposits within process equipment and verifies the results of elephant gun scans. Detection threshold limits are established at a level so that: (1) all sizable deposits are identified and (2) items are excluded that do not contain a sufficient quantity of uranium to emit a measurable neutron flux. Items that emit gamma-ray radiation above threshold limits for either detector are identified for later quantitative measurement.

Initially, the cascade equipment is scanned with the elephant gun by pushing the cart through the alleyways adjacent to the process equipment. The location of each item emitting a gamma-ray signal greater than the threshold value is recorded on a data sheet. Following the elephant gun scan, each cell is scanned with the hand-held NaI detector by walking slowly around each piece of process equipment and aiming the detector at the equipment. The detector is placed as close as possible to the equipment and is moved slowly (at a rate of about 0.5 m/s) across the equipment surface. For most pieces of process equipment, the detector is placed adjacent to the equipment surface. Ladders are used to enable measurement personnel to scan the items that cannot be reached from ground level. Each piece of process equipment emitting a gamma-ray signal greater than the threshold limit is marked on a map for later neutron measurement. A data sheet is generated for each process cell in the building. As the cell is scanned, each item exceeding the measurement threshold is numbered sequentially and the general location of the deposit is marked on the schematic. (An example data sheet is shown in Fig. 2.) For each deposit, the corresponding stage number, maximum gamma-ray reading, and a description of the process equipment is recorded. Similar data sheets are generated to record the data for the overhead piping, booster stages, chemical traps, and other pieces of miscellaneous equipment.

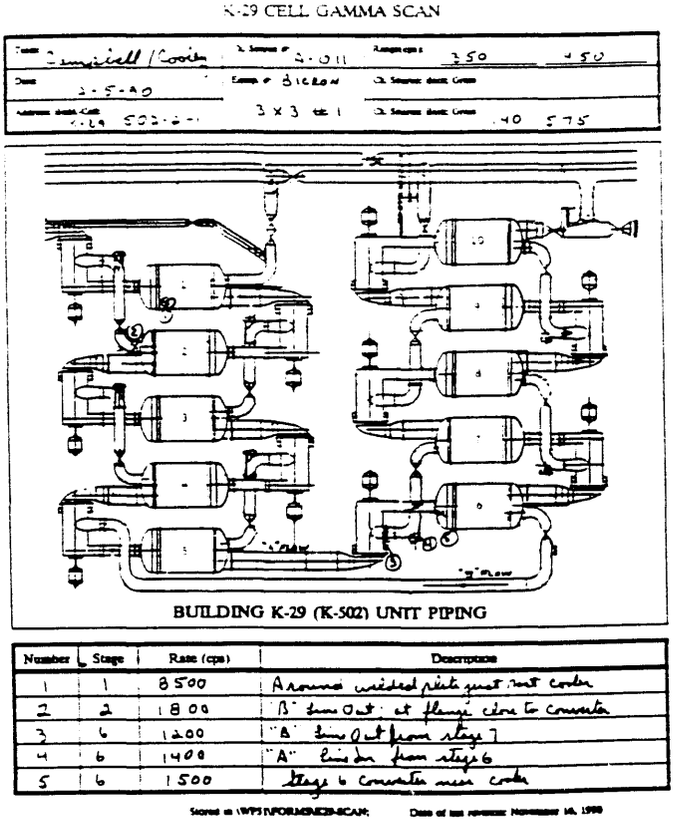

Fig. 2. A cell map data sheet used for scanning process equipment.

C. Quantitative Measurement

Quantitative measurements in the LEU process buildings are performed using passive neutron measurement methods that detect neutrons emitted from three sources: (1) the $^{19}F(\alpha,n)^{22}Na$ reaction (the major source); (2) the $^{18}O(\alpha,n)^{21}Ne$ reaction [contributes approximately 1.6% of the (α,n) activity[2]]; and (3) the spontaneous fission of ^{238}U (0.014 n/s/g ^{238}U). Because the isotope of interest is ^{235}U and the dominant source of neutrons for LEU is from the $^{19}F(\alpha,n)^{22}Na$ reaction between α-particles emitted from ^{234}U and bonded F atoms, neutron measurement results are highly dependent on knowledge of the isotopic and chemical composition of the measured material. Isotopic composition is estimated using gamma-ray isotopic measurement methods developed in-house (Sect. III). Except for suspected deposits of UF_6, all deposits are assumed to consist of UO_2F_2.

Two sizes of neutron detectors are used to perform quantitative measurements: (1) shielded neutron assay probes (SNAPs), which consist of two ^3He tubes encased in polyethylene and surrounded by a cadmium shield, and (2) slab detectors, which consist of 11 ^3He tubes encased in polyethylene and surrounded by a cadmium shield. Additional layers of polyethylene shielding were added to the back and sides of both detectors to reduce the effects of ambient background. SNAP detectors and added shielding weigh approximately 154 kg, while slab detectors, for which detection efficiency is approximately seven times better than SNAP detectors, and added shielding weigh approximately 230 kg. Because of their greater efficiency, slab detectors are used for the majority of the quantitative neutron measurements; SNAP detectors are used only in locations where positioning the slab detector is not practical.

Neutron-detector positioning is influenced by three factors: (1) net count rate achievable at a given distance, (2) the size of the item being measured, and (3) the presence of other deposits in the area. To obtain sufficient counting statistics in a reasonable time frame, the detectors need to be placed as close to the deposits as is practical. To minimize the effects of possible inhomogeneous distribution of uranium

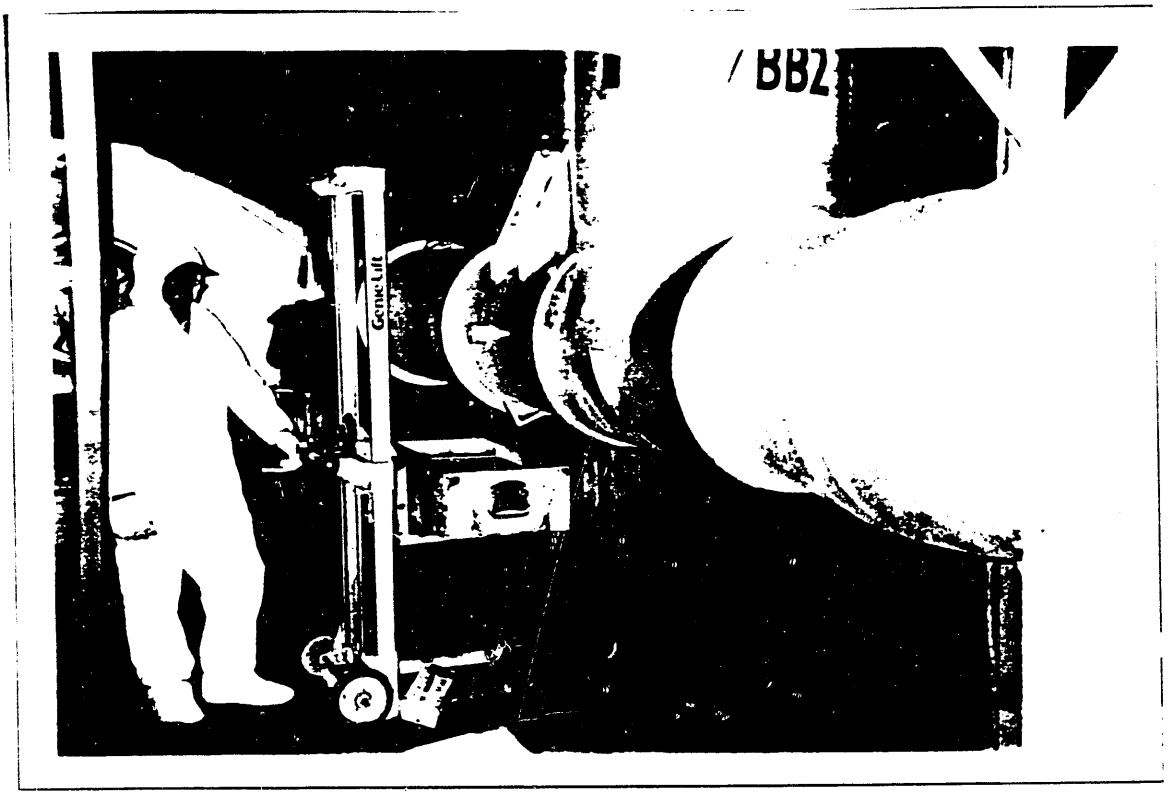

Fig. 3. The measurement of a compressor using a slab detector.

within the items, however, the detectors need to be placed as far away from the deposits as is practical. In addition, the detectors need to be positioned in such a way that influence from other deposits in the area is minimized. The best balance between the considerations of these three factors is determined for each deposit and the detection instrumentation is positioned accordingly. Positioning for background measurements is also determined by achieving the best balance between reducing the influence from the measured item and maintaining positioning such that the contribution to the background from nearby sources was similar for both the background and item measurements.

For localized deposits, a single measurement is performed with the detector placed at a fixed distance from the center of activity. For continuous deposits, such as those found in a pipe, incremental measurements are made along the entire length of the deposit. For most measurement configurations, the neutron detectors are positioned with small portable hand-lifts to maneuver the detector close to the deposit and to hold the detector in place during the measurements. In Fig. 3, a quantification measurement is being conducted on a compressor using a slab detector. In addition, a heavy-duty camera tripod with an attached base plate is used to position a SNAP in tight locations in which a slab detector cannot be safely positioned. An overhead crane is used to position slab detectors atop equipment or in areas where the hand-lift cannot be maneuvered. All measurement information about the deposits and detector positions is written on data sheets. This information includes a drawing of the deposit located in the process equipment, distances from the deposit to the detector, and the gross and background neutron counts of the deposit. An example data sheet is shown in Fig. 4.

III. DATA ANALYSIS

Because a suitable method was not available to nondestructively determine uranium enrichment of a deposit in process equipment, a new technique was developed.[3] This technique uses HRG detection instrumentation and data analysis using commercially available spectral analysis software and an in-house developed software program (ISOTOPIC) (1) to determine the ^{235}U concentration and ^{234}U abundance, (2) to detect the presence of other gamma-ray-emitting isotopes (e.g., ^{232}U and ^{237}Np), and (3) to compute the neutron specific activity of a deposit. Gamma- and X-ray peaks from all energies of the spectrum are used to estimate container attenuation and sample-absorption factors; once estimated, these factors are used to compute corrected peak areas. Estimates of ^{235}U concentration can then be made using ^{235}U to ^{238}U peak ratios from three energy regions: (1) low energy (58 and 63 keV), (2) medium energy (90 and 92 keV), and (3) high energy (186 and 1001 keV). Because of severe container attenuation and self-attenuation of low-energy gamma rays, region 1 is primarily used for analysis of spectra taken of deposits in thin-walled (less than 3 mm) containers. Region 2 contains gamma rays from ^{238}U daughters and X rays from ^{235}U that are close in energy. Useful estimates, therefore, can be obtained from this region without severe biases from attenuation correction estimates. Gamma rays from this region, however, are not available for measurements through thick container walls (e.g., greater than 1 cm). Region 3 contains the most intense gamma rays, which are distributed in energy bands sufficiently wide to estimate corrections for container attenuation and sample absorption. When used to perform analyses, program ISOTOPIC provides initial estimates of container and deposit thicknesses. Using

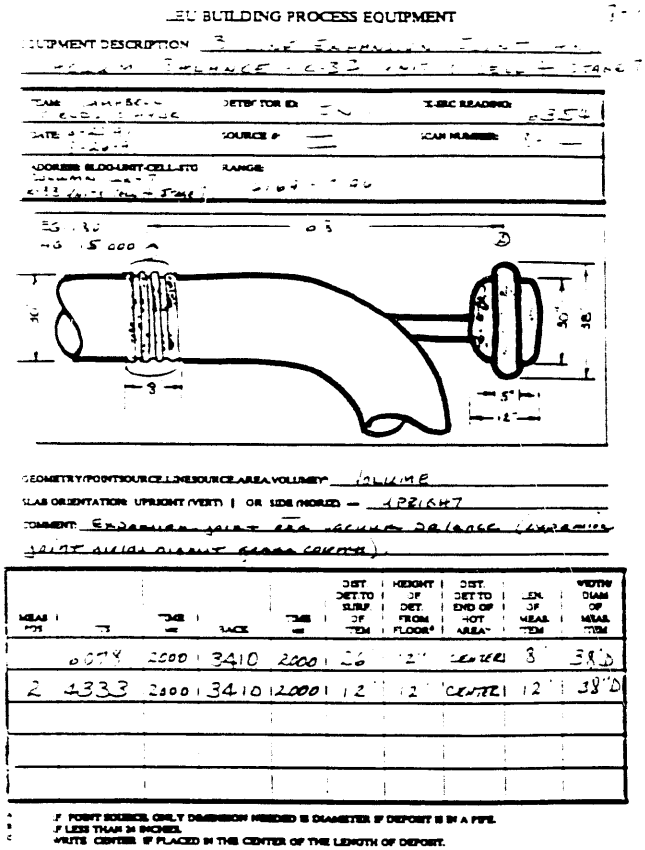

Fig. 4. An example of an uranium deposit data sheet used for quantifying measurements.

these thicknesses, the program computes attenuation and self-attenuation corrected gamma-ray activities for each gamma ray observed for each isotope present. The analyst then adjusts the container and deposit thicknesses until all of the corrected gamma-ray activities for a given isotope are as close as can be achieved. ISOTOPIC then computes enrichment for each of the energy regions. The relative quantities of other gamma-ray-emitting isotopes and the neutron specific activity of the measured item are also computed.

Measurement uncertainty for individual enrichment measurements is: ±20% relative. Because the process buildings were operated at a variety of enrichments and because process equipment was occasionally moved from one area to another during maintenance activities, the enrichment of a deposit in an individual piece of process equipment may be different from the average enrichment used for the area.

Analysis methods for neutron measurement data were also developed in-house. Detection efficiency for each detector used was modeled utilizing ^{252}Cf sources (traceable to the National Institute of Standards and Testing). The sources were placed at various distances from angles of incidence to the detector. Mathematical models of the point-source efficiency of each detector were then developed. Software programs were developed which, using the mathematical models and analyst-input parameters (i.e., distance of the detector from the deposit, orientation of the detector, and size of the deposit), compute the average detection efficiency of the neutron detectors for neutrons emitted from the deposit. A description of this method is shown in Fig. 5.

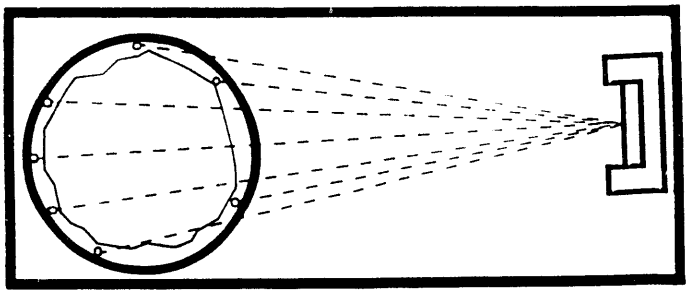

Fig. 5. Neutron calibration technique utilizing point-source efficiencies.

Neutron-specific activity is computed by the software using the following equation:

$$SA_U = \sum \left(\frac{SA_i}{wf_i} \right),$$

where

SA_U = uranium specific activity (n/s/gU),
SA_i = specific activity of each isotope present (n/s/g_i) (includes non-uranium isotopes, if present),
wf_i = weight fraction of the isotope (g_i/g_U).

^{235}U quantity is then determined as follows:

$$g^{235}U = \frac{CR}{SA_U \times e} \times wf_{^{235}U},$$

where

$g\ ^{235}U$ = quantity of ^{235}U in the deposit,
CR = net count rate (c/s),
SA_U = uranium specific activity (n/s/gU),
e = detection efficiency (c/n),
$wf_{^{235}U}$ = weight fraction of ^{235}U.

The importance of obtaining accurate isotopic composition estimates is illustrated in Table 1. The table shows four gram quantity estimates obtained from a single measurement simply by using different isotopic composition assumptions. Because (1) ^{234}U concentration at a given enrichment was found to vary by as much as a factor of 2 for different process operating conditions and (2) enrichment in a given process area was found to vary by as much as a factor of 3, estimates of isotopic composition made using gamma-ray spectra were used whenever possible.

Measurement uncertainty for neutron quantitative measurements is ±50%. This uncertainty includes both random effects and biases. Random effects are dominated by

Table 1. Gram quantity estimates obtained from a single measurement using different isotopic composition assumptions

Condition	^{235}U (wt %)	^{234}U (wt %)	Specific Activity (n/s/g)	Detector Efficiency (c/n)	Net Rate (c/s)	Quantity (g ^{235}U)
1	3	.03	.096	.001	2	627
2	3	.015	.058	.001	2	1041
3	6	.06	.172	.001	2	696
4	6	.03	.096	.001	2	1245

fluctuations in the response of electronic instrumentation and by counting statistics. Biases are dominated by calibration uncertainty, detector placement, inhomogeneous material distribution, background determination, and uncertainty in the material composition and chemical form.

IV. QUALITY ASSURANCE ACTIVITIES

Quality assurance activities for the NDA survey program are implemented to provide consistent, reliable results for all measurements while allowing the flexibility to upgrade methods, instrumentation, or analyses as improved techniques are developed. Quality assurance activities are structured around the areas defined by the American National Standards Institute and focus on three principal areas: procedures, measurement control, and inspection.

Standard operating procedures (SOPs) are maintained for all measurement operations. Measurement procedures that were developed for gamma-ray scanning, quantitative neutron measurements, and enrichment measurements apply to virtually all types of process equipment. These procedures are reproduced on the back of all data sheets for reference by measurement personnel in the field. SOPs also exist for daily instrumentation checks, process equipment calibrations, instrument calibrations, calibration standard measurements, data handling, and quality assurance activities. These procedures are maintained and followed to ensure that a consistent approach is taken by the various personnel who perform these activities.

Measurement-control activities are conducted to ensure that instruments are working properly and that performance (i.e., precision and accuracy) is maintained throughout the program. Control charts are maintained for calibration source and reference equipment measurements for each instrument. These charts are used to detect control loss, instrument problems, and shifts in instrument performance that require corrective measures. Response modeling of instruments is also performed. The response of instrument to incident radiation is characterized for each piece of measurement instrumentation. The effects of material distribution, position, density, equipment configuration, and attenuation factors on NDA measurements are modeled using empirical data. Mathematical models of the anticipated response of each detector to material contained in process equipment have been developed.

Periodic inspections (i.e., compliance reviews) are conducted to ensure that the quality of measurement techniques is maintained. Compliance reviews consist of examination of all facets of the measurement program for compliance with SOPs. The intent of these reviews is to verify that SOPs are being followed, to assess their appropriateness, and to identify areas for improvement. Anomaly inspections of data records are conducted as data sheets are received to ensure that anomalous data are not entered into the data base.

V. RESULTS AND CONCLUSIONS

The organization, development, and implementation of a large-scale NDA measurement survey program to provide quality uranium deposition data in support of gaseous diffusion plant D&D activities continues to be successfully demonstrated. NDA measurement surveys to locate and quantify sizable uranium deposits have been completed for the three LEU process buildings at the K-25 Site. Approximately 5 work-years of effort over a 10-month period have been involved in scanning, quantifying, analyzing, and documenting the results for the three LEU buildings. More efficient detection equipment combined with new analysis techniques are meeting the challenge of characterizing LEU holdup in a variety of process configurations without extreme measurement times. Facility management is using the results of the survey to support a variety of activities including isolation and removal of large deposits; performing health, safety, and environmental analyses; and improving facility nuclear material control and accountability records. Future development challenges include improving detector efficiency while reducing the contribution from background and determining and quantifying measurement biases. The NDA program at the K-25 Site will continue to meet the needs of site D&D activities while identifying applications for the new measurement and analysis techniques to be used at other nuclear fuel cycle facilities.

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