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# NONDESTRUCTIVE ASSAY MEASUREMENTS OF GASEOUS DIFFUSION PROCESS EQUIPMENT*

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## Abstract

A nondestructive assay (NDA) measurement survey program has been established at Oak Ridge Gaseous Diffusion Plant in Oak Ridge, Tennessee, to support decontamination and decommissioning (D&D) activities at the shutdown facility. The program's objective is to develop and to implement NDA measurement techniques for estimating the residual uranium contained in the diffusion process equipment and ancillary piping of the shutdown process buildings on-site. Measurement techniques are described, support programs (e.g., technician training, hardware development, and quality assurance) are detailed, and results are summarized. The organization, development, and implementation of a large-scale NDA measurement survey program to provide reliable process holdup data for D&D activities is being successfully demonstrated.

## 1. Introduction

A nondestructive assay (NDA) survey program was established at Oak Ridge Gaseous Diffusion Plant (ORGDP) in June 1987 to provide support for decontamination and decommissioning (D&D) activities at ORGDP. The objective of the NDA survey program is to develop and to implement NDA measurement techniques for estimating the residual uranium contained in the diffusion process equipment and piping of the shutdown process buildings on-site. From 1945 through 1964, ORGDP produced highly enriched uranium (HEU) for U.S. national defense purposes, for nuclear-powered ships, and for research programs. Low-enriched uranium (LEU) for commercial nuclear reactors was subsequently produced until 1985 when site enrichment operations ceased. The decision was then made to shut down ORGDP's uranium enrichment facilities permanently and to D&D the process buildings.

When enriching operations were discontinued, the uranium hexafluoride ($UF_6$) process gas was evacuated from the process systems. Solid uranium deposits, however, remain in the process equipment and ancillary piping. The majority of these deposits were formed when the $UF_6$ gas reacted with the metallic surfaces of the process systems 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 $UO_2F_2$. 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.

## 2. Equipment and Techniques

### Process Equipment Measured

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) one or two compressors used to compress the gas to the pressure needed for flow through the barrier tubes, and (3) process piping for stage and interstage connections. Associated auxiliary equipment includes chemical absorbent traps, purge equipment, surge tanks, feed and withdrawal stations, and cold traps. When the entire plant was operating to produce HEU, the ORGDP cascade consisted of nearly 4800 stages contained in five separate process buildings. Because of the random distribution of the uranium deposits in the process equipment, a measurement survey of the gaseous diffusion process buildings in their entirety was recommended to locate and to quantify the residual uranium.

### Measurement Techniques

Because of the various process equipment configurations requiring measurement, two NDA techniques are used to acquire data for deposition estimates: gamma ray assay and neutron assay. The first involves the measurement of the 185.7-keV gamma ray signal emitted from $^{235}U$. The $^{235}U$ deposit size is proportional to the intensity of the measured signal. Corrections are made for process equipment or pipe wall thickness, self attenuation of the deposit, measurement configuration, and background radiation from nearby sources. Gamma ray measurements are used for equipment where attenuation of the gamma rays produced by the contained $^{235}U$ is not severe. Converters and piping are routinely measured by gamma ray assay.

Neutrons emanating from residual uranium in process equipment are the result of the $F(\alpha,n)$ reaction between the uranium isotopes and the fluorine contained in the deposit. Alpha particles emitted by the decaying uranium isotopes interact with the fluorine nuclei contained in the deposits to release neutrons. (For calculational purposes, it is assumed that the deposit is entirely $UO_2F_2$.) Because the high-energy alpha particle emitted from the $^{234}U$ has a high $F(\alpha,n)$ cross section and the dominant source of alpha activity in HEU is from $^{234}U$ decay, the measured neutron signal is assumed to be proportional to the quantity of $^{234}U$ present. The quantity of $^{235}U$ contained in the deposit is estimated by multiplying the $^{234}U$ estimate by the $^{235}U$:$^{234}U$ ratio. This ratio was estimated based on historical mass spectrometer data from process gas samples.

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Corrections are made for measurement configuration and background radiation from nearby sources. For low assay material, a correction is also included for the neutron contribution from the $^{238}U$ isotope. Neutron measurements are used for items where gamma ray attenuation is severe. Compressors are routinely measured by neutron assay. In addition, neutron measurements are conducted to verify estimates initially made by gamma ray assay on converters with deposition estimates large enough for self-absorption of gamma rays to be a concern. Neutron measurements are also made on other selected pieces of process equipment.

Measurement Equipment

Because the process equipment is measured in situ, the NDA instrumentation must be portable, rugged, and capable of operating accurately in adverse environmental conditions (e.g., temperatures ranging from -10° to 40°C and humidity levels approaching 100%). Commercially available NDA equipment has been modified to satisfy these requirements. Modifications have included tighter seals to prevent the influx of rust and dirt into the electronics, absorbent rubber padding around detectors to minimize jarring and bumping effects, Teflon covers over detector crystals to protect the surfaces, and the addition of desiccant in detectors to reduce humidity levels. Modifications have also been made to aid in physically conducting the measurements. Magnets have been attached to detectors for stabilizing the instrumentation on the process equipment being measured; jigs have been designed to ensure the reproducibility of detector positioning; calibrated extension rods have been attached to detectors for gauging detector-to-source distances, and detectors have been fitted to extension poles to accomodate measurements of overhead pipes.

Gamma ray measurement equipment consists of sodium iodide (NaI) detectors coupled to battery-operated scaler analyzers. The analyzer furnishes the operating voltage for the detector and displays the scaler readout of the gamma ray signal. Two sizes of NaI detectors are used: 2.5 x 1.3 cm and 5 x 1.3 cm. The NaI crystal thickness is optimized for the detection of the 185.7-keV gamma ray. The detector is enclosed in a lead shield collimator; the depth of collimation can be varied from 0 to 5 cm. The weight of the NaI detector and analyzer is approximately 5 kg.

The instrumentation used to measure the fast neutrons emitted by the residual uranium consists of portable shielded neutron assay probes (SNAPs) coupled to battery-operated scaler analyzers. The SNAP detector consists of two 2.5 x 23 cm $^{3}He$ tubes contained within a polyethylene cylinder surrounded by a cadmium shield. Fast neutrons are moderated inside the polyethylene cylinder and counted; thermal neutrons are captured by the cadmium shield and, therefore, do not contribute to the measured signal. The weight of the SNAP and analyzer is approximately 11 kg.

## 3. Measurement and Data Analysis Activities

The measurement philosophy for each process building is to have the measurement teams systematically pass through the building multiple times – measuring different process equipment in each campaign. This measurement approach simplifies scheduling, detection and support equipment requirements, and data reporting and analysis. Compressors and converters are measured during the first pass; process piping, during the second pass; and miscellaneous equipment not present in all areas of the building, during the third pass. Additional measurements are made as necessary to clarify abnormal readings and to verify larger-than-normal deposits. All routine measurements are conducted by operators who have been trained, tested, and certified on the use of standard operating procedures for these measurements. All nonroutine measurements are made by technicians or engineers who have been trained in the area of NDA measurements.

The procedure for measuring a compressor involves orienting the SNAP detector at a 45-degree angle looking into the pump (Fig. 1). Typically, a

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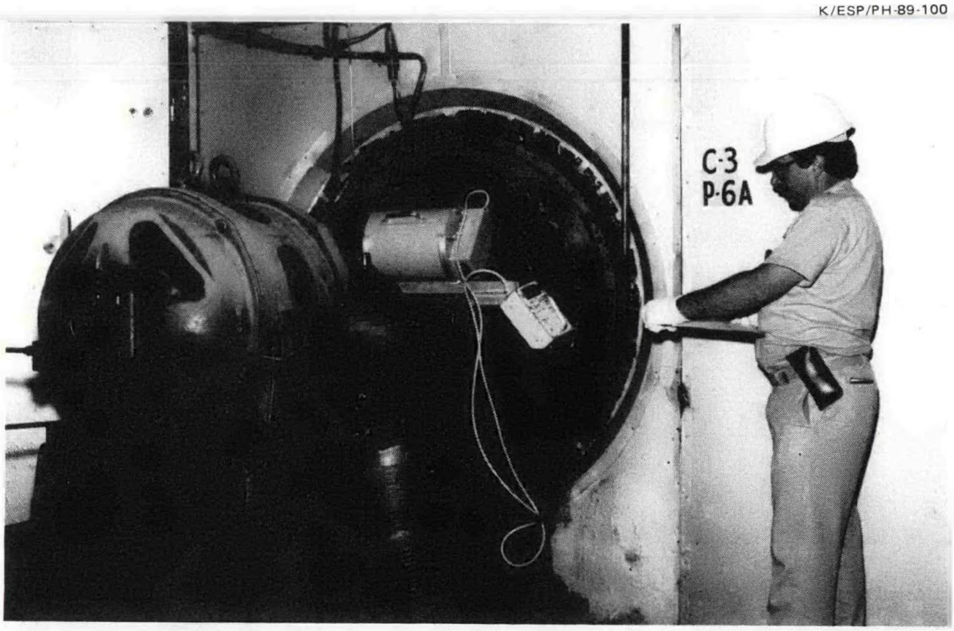


Fig. 1 - Neutron measurement of a compressor using a shielded neutron assay probe

single 5-min measurement is performed on each compressor. A 5-min background measurement is conducted for a group of six compressors by placing the detector face down between the middle two compressors. After every six compressor measurements, a 1-min measurement is performed on a $^{252}$Cf calibration source to verify that the measurement equipment is operating properly. Each converter is measured with a 5-cm NaI detector. Generally, two 2-min gamma ray measurements are conducted on small converters: in the first, the detector is placed flush against the top; in the second, flush against the bottom. Two measurements are taken on the top and bottom for large converters. A 5-min background measurement is taken between each pair of converters. Approximately every 30 min a 2-min count on a uranium calibration foil is conducted to verify that the gamma instrumentation is operating properly. When neutron measurements are required for converters, the SNAP detector is positioned in the same manner as that described for the gamma measurements on the small converters; counting times are increased to 10 min. Each process pipe is scanned with a 2.5-cm NaI detector nearly flush to the pipe at a rate of approximately 1 m/12 s. Incremental measurements are made at pipe locations with measureable deposits (Fig. 2). For these measurements the detector is positioned 7 to 20 cm from the pipe depending upon the pipe diameter; counting times vary from 60 to 100 s. Readings on uranium calibration foils are conducted every 30 min. Measurement techniques using a combination of methods are developed for miscellaneous equipment.

All measurement information from the process equipment, background, and calibration source readings is recorded on data sheets. Also recorded are the measurement locations, the names of the measurement teams, detection equipment identification numbers, and calibration source identification numbers. All measurement information is entered into a computer data base. Software has been developed to transcribe raw measurement data to the data base and to manipulate the data to provide deposition estimates for each piece of process equipment measured. Accuracy of the techniques is checked by hand calculations. Status and summary reports are periodically generated.

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Fig. 2 - Scanning and incremental measurements of process piping with sodium iodide detectors

## 4. 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, equipment, or analyses as improved techniques are developed. Quality assurance activities focus on four principal areas: training and qualification, procedures, inspection, and measurement control.

### Training and Qualification

A training course is conducted for all operators prior to the start of any measurement campaign. All operators who will make measurements in support of the NDA survey are required to participate in the training and qualification annually. The principal emphasis of the training course is to instruct the operators how to perform quality NDA measurements and to identify what parameters are important to each measurement. Basic physical principles used in making measurements are taught, and laboratory sessions are held to familiarize the operators with the measurement instrumentation and to help them gain a better understanding of the physical principles involved. General information (e.g., safety and criticality concerns) is also provided. Measurement certification testing is performed by having each participant make measurements on selected process equipment using the techniques taught in the course. To retain qualification during the measurement campaign, each operator must make measurements of process equipment a minimum of once per month. Technicians and members of the engineering staff who make nonroutine NDA measurements receive advanced training in NDA techniques by attending appropriate training sessions (e.g., Los Alamos National Laboratory NDA courses).

### Procedures

Standard Operating Procedures (SOPs) are maintained for all routine operations. Measurement procedures are printed on the back of all data entry forms for reference by the operators in the field. SOPs also exist for daily inspections of measurement instrumentation, process equipment calibrations (used for conversion of counts to grams of material), instrument calibrations, calibration source measurements, data handling, and quality assurance activities. These procedures are followed to ensure that a consistent approach is taken by the various personnel who perform these activities.

### Inspection

Inspections are carried out to ensure that the quality of measurements performed is maintained. Three types of inspections are used: compliance reviews, anomaly inspection, and item remeasurement.

*Compliance reviews* consist of both random and systematic review of all facets of the measurement program for compliance with SOPs. The intent of this review is to verify that SOPs are being followed, to assess their appropriateness, and to identify areas for improvement. Anomaly inspections are conducted on a daily basis to ensure that anomalous data are not entered into the data base. Original data sheets generated by the measurement teams are inspected for apparent errors. Once the data is entered into the data base, software is used to check for abnormal entries such as high background, low background, nonrandom data fluctuations, abnormally high equipment readings, and multiple teams using the same equipment on the same date. An item remeasurement program is implemented to verify that valid measurement values are being obtained. Items are randomly remeasured both by operators and by technicians without knowledge of prior measurement results.

### Measurement Control

Measurement control activities are conducted to ensure that instruments are working properly and that performance (i.e., precision and accuracy) is maintained throughout the measurement campaign. 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. A calibration source measurement is conducted prior to beginning survey measurements and is repeated several times throughout the day. Instruments are removed from service when calibration source readings are outside the allowable limits. Response modeling of instruments is also performed. The response of instruments to $^{235}$U is characterized for each measurement type and for each type of process equipment. The effects of material distribution, position, density, equipment configuration, and attenuation factors on NDA measurements are modeled using empirical data.

## 5. Measurement Results

To date, approximately 250 chemical traps, 6,200 compressors, 3,400 converters, 11,000 exhaust pipes, 150 km of process piping, and 200 pieces of miscellaneous equipment have been measured in the gaseous diffusion process buildings at ORGDP. Over 100,000 individual measurements were required to accomplish this task. Quantities of $^{235}$U deposits have been estimated for each item measured, and results have been reported to the engineering teams involved in D&D planning activities for the site.

The current NDA deposition estimates are being reported with a measurement uncertainty of ± 50%. While the intensity of the neutrons or gamma rays emitted from a deposit is proportional to the deposit size, quantifying the deposit by measuring a gamma ray or neutron signal is extremely complex. Measurement uncertainty is composed of two parts: precision and accuracy. Variation in the measurement precision is a result of random fluctuations in the rate of gamma ray and neutron production (counting statistics), slight variations in positioning the NDA instrumentation on the process equipment, and electronic fluctuations in the detection equipment. Uncertainty in the measurement accuracy is governed by (1) difficult measurement geometries due to unusual process equipment configurations and accessibility of the process equipment, (2) the limited knowledge regarding the form and composition of the deposits, (3) variation in the location of deposits within each piece of process equipment, (4) variation in the thickness of the deposits, (5) background measurement techniques, (6) different NDA equipment brands, and (7) variations in environmental conditions (e.g., temperature and humidity).

A destructive analysis plan has been developed that will validate and potentially reduce the estimated measurement uncertainty. Destructive analysis involves the removal and analysis of deposits from select pieces of process equipment to determine the isotopic content, chemical composition, and uranium quantity. The results are then compared with the corresonding deposition estimates based on NDA measurements. Because ORGDP has been shut down for some years, decontamination facilities are currently not functional. Limited destructive analyses conducted over the past 2 years have substantiated the reported measurement uncertainty; recovered $^{235}$U quantities were well within ± 50% of the estimated values. Plans are being made to restart the decontamination facilities so that a complete destructive analysis program can be implemented.

## 6. 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 is being successfully demonstrated. State-of-the-art detection equipment and measurement techniques are being developed by NDA engineering specialists. On-site training classes and certification programs are being conducted to verify that measurement teams can perform the NDA measurements and can recognize equipment malfunctions or unusual measurement readings. Standard operating procedures exist for process equipment measurements, instrument response modeling, daily instrumentation checks, calibration source measurements, data handling, and inspection activities. Specialized computer data bases have been established for collecting, handling, analyzing, reporting, and storing the measurement data. Quality assurance programs are being implemented to ensure measurement consistency and reliability. As program activities progress, the measurement results, experience, techniques, and methodolgy will continue to be shared with the safeguards community.