

**Y-12
OAK RIDGE
Y-12
PLANT**

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FIELD USE OF NMIS AT OAK RIDGE

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ABSTRACT

The Nuclear Materials Identification System (NMIS), developed by the Oak Ridge National Laboratory and Oak Ridge Y-12 Plant (Y-12), has been successfully used at Y-12 for nuclear material control and accountability (NMC&A). It is particularly useful in the high gamma-ray background of storage arrays and for shielded HEU. With three systems in use at Y-12, NMIS has enhanced the NMC&A capability for verification and for confirmation of materials in storage and for HEU receipts by providing capability not available or practical by other NDA methods for safeguards. It has recently cost-effectively quantified the HEU mass and enrichment of hundreds of HEU metal items to within a total spread of $\pm 5\%$ (3 sigma) with mean deviations for all HEU verified of $+0.2\%$ for mass and -0.2% for enrichment. Three cart portable systems are easily moved around with minimal impact on facility operations since no permanent dedicated floor space is required. The positive impact of NMIS at the Oak Ridge Y-12 Plant is improved and more cost effective NMC&A as well as the resolution of NMC&A findings. Its operation at the Y-12 Plant is essential for compliance with the NMC&A requirements of the U. S. Department of Energy. NMIS portability has allowed one system to be moved temporarily to the former K-25 Gaseous Diffusion Plant for characterization of a large deposit of hydrated uranyl fluoride. The impact of this NMIS application was enhanced and verified nuclear criticality safety that led to the safe removal of a large deposit originally estimated by gamma-ray spectrometry and neutron counting to contain 1300 kg of 3.3 wt% ^{235}U material. NMIS has also been operational at Los Alamos National Laboratory and Pantex.

I. INTRODUCTION

The Nuclear Materials Identification System (NMIS), developed by the Oak Ridge National Laboratory (ORNL) and the Oak Ridge Y-12 Plant (Y-12), has been

used for a variety of NMC&A applications at Y-12 and at the East Tennessee Technology Park, formerly the Oak Ridge Gaseous Diffusion Plant at the K-25 site in Oak Ridge. The applications at Y-12 include: determination that training units did not contain fissile material, confirmation of highly enriched uranium (HEU) metal receipts, in-situ confirmation of weapons components in storage, and quantitative verification of declared uranium mass and enrichment of HEU metal in storage. At the former gaseous diffusion plant, NMIS was used to obtain the mass, hydration, and distribution of deposits of hydrated uranyl fluoride in shutdown gaseous diffusion plant piping.

For highly enriched uranium (HEU), NMIS operates as an active neutron interrogation system which uses a small ^{252}Cf source ($\sim 1\ \mu\text{g}$ or less) in an ionization chamber and scintillation detectors to acquire a variety of time correlation signatures.¹ In addition, NMIS measures multiplicities as defined by Hage and Cifarelli² with detection event triggered and randomly triggered time windows. Multiplicities with source event triggered time windows are also acquired during active measurements. Active neutron interrogation is useful in: (1) situations with significant background since detection events correlated with the source are uncorrelated with background, and (2) for shielded HEU. NMIS has also been applied successfully both actively and passively to Pu components.³

Three cart portable NMIS systems are in use at the Oak Ridge Y-12 Plant. One more portable system has been operated in testing at ORNL with battery power for up to three hours. This paper reviews the field uses of the NMIS technology at Oak Ridge where it has provided a capability not available and/or practical by other NDA methods. NMIS has greatly enhanced the NMC&A capability at Y-12, particularly for weapons components in containers and shielded HEU.

II. MEASUREMENTS FOR TRAINERS

Although the first active time correlation measurements with ^{252}Cf with fissile material were performed in 1970 at ORNL for HEU and by ORNL at Los Alamos National Laboratory (LANL) in 1971 for Pu, the first practical use of these methods at Y-12 occurred in 1984. NMIS type measurements with ^{252}Cf were used to distinguish war reserve (WR) units from mockups which had or alloy replaced by depleted uranium. The first field use of this method at Y-12 was to determine that B33 trainer parts returned from military bases to Y-12 in 1993 did not contain enriched uranium.⁴

The B33 was a gun-assembled weapon consisting of two components. Confirmation was accomplished by comparing signatures measured from the returned B33 trainer parts with those from calibration mock-ups (made with depleted uranium) packaged at Y-12 in M102 containers (Fig. 1) and signatures from one of the WR components packaged in a M102 container at Y-12. The calibration signature for the other WR component was estimated from Monte Carlo calculations. Some measured NMIS signatures were greater than a factor of 100 different for the WR unit. The low measurement uncertainty allowed the determination that the depleted uranium masses of 10 trainer components were low by 4.5%.

These confirmations were conducted in a timely, reliable manner and produced no false positives for the 512 confirmations (263 containers with component 1 and 249 containers with component 2). Measurement times were 10 min. each for both time- and frequency-analysis verifications. As many as 32 confirmations were performed in one 8-hour shift (more like 6 hours actual verification time) and 111 in one week which, of course, depended on operation schedules. A photograph of the containers and the measurement area is given in Fig. 1 showing arrays of five containers per pallet. Usually, ten units were within 10 ft. of the measurement station with ~100 units within 20 ft. This confirmation program occurred over a five month time period in which the temperature varied from 60 to 90°F.

Trainers for a variety of other weapons systems have also been confirmed to be free of fissile material. One trainer was an aluminum casting in a container. For this trainer, the time distribution of detection events after ^{252}Cf fission was used not only to determine that the trainer material was Al but also to determine the Al thickness to within 0.125 in.

The facility operations were minimally impacted since these confirmations were performed in unused space and performed so as to not impact other operations

significantly. The positive impact was significant because the confirmations were essential to the further handling and processing of this material at the plant both from nuclear criticality safety and safeguards viewpoints.

III. LARGE DEPOSIT IN DIFFUSION PLANT PIPING

NMIS was used (1997) to non-intrusively characterize a large hydrated uranyl fluoride ($\text{UO}_2\text{F}_2 \cdot n\text{H}_2\text{O}$) deposit at the former K-25 Oak Ridge Gaseous Diffusion Plant, now the East Tennessee Technology Park.⁵ This deposit existed in a 17-ft.-long, 24-in.-O.D. process gas line that, because of its shape, came to be known as the "Hockey Stick" (Fig. 2). The deposit was formed as a result of a steady moist air in-leakage over a number of years from the 20-in.-diam. double disc gate valve visible in Fig. 2. In order to assess criticality safety requirements and to provide information that would allow formulation of a safe method to remove the deposit, a characterization of the deposit mass distribution, and hydration (water content) was necessary.

Earlier passive neutron and gamma-ray spectrometry measurements had identified the largest deposits in the former diffusion plant. This largest deposit was estimated from passive gamma-ray spectrometry and neutron counting to be uniformly distributed in the pipe and to contain 1,300 kg ($\pm 50\%$ uncertainty) at ~3.3 wt % ^{235}U enrichment ($\pm 50\%$ uncertainty). If such a large deposit collapsed or had a high hydrogen to uranium (H/U) ratio, there would be significant nuclear critical safety concerns. The NMIS characterizations were performed by active neutron and gamma-ray time-of-flight transmission measurements through the pipe deposit.

The source was located on one side of the pipe and two plastic scintillation detectors were located on the opposite side of the pipe. The pipe was scanned vertically, horizontally (Fig. 2a), and where possible, rotationally at various positions along the pipe (Fig. 2b). The time-of-flight transmission measurements provided a gamma and neutron radiograph of the pipe deposit as a function of length. The effects of the pipe walls were removed utilizing measurements on an empty section of pipe.

High-energy neutron (> 8 MeV) transmission was used to measure the deposit thickness and to obtain a profile of the deposit distribution. This is possible because for hydrated uranyl fluoride, the neutron total interaction cross-section above 6 MeV is essentially independent of hydration for variations in H/U from 1 to 30. Consequently, high energy neutron transmission depends only on deposit thickness.

The prompt fission gamma-ray spectrum of ^{252}Cf was used in the analysis of the measured gamma-ray transmission through the deposit to determine the deposit hydration, which is related to the material density. The density dependent gamma-ray cross sections make the gamma-ray transmission measurement well suited for hydration or density determination applications. Due to the fast timing resolution of the NMIS system (1 nsec), gamma rays that interact are counted later in time and are separated in time from those that are directly transmitted, providing a well collimated source that was used to measure the material mass attenuation coefficient. Using the estimate of the deposit thickness obtained from neutron transmission, the mass attenuation coefficient was then determined from the gamma-ray transmission, which (knowing thickness), is uniquely related to deposit density or hydration. These initial estimates were then refined by a series of iterative calculations until values of the thickness and hydration were acceptably converged.

Contrary to expectations, measurement results indicated that in portions of the pipe the deposit had adhered to the top of the pipe with very little material existing on the bottom. The average NMIS measured H/U ratio was 3.5 ± 0.2 . A total uranium mass of 542 ± 92 kg was obtained from the deposit distribution measured along the process pipe. The adjacent detectors showed that the deposit thickness was very irregular, varying in some cases by more than a factor of 2 in. a distance of 4.5 in. along the pipe.

The measurement results were used to select the locations on the process pipes for the visual observations of the deposits. After completion of the measurements, a fiber-optic camera was inserted through a number of holes drilled into the pipe from the bottom rather than from the top as originally planned. At the same time, a special tool was inserted for measuring the deposit thickness. The fiber-optic camera observations and thickness measurements were performed at a number of different locations along the process pipe. In general, the intrusive camera observations at various locations were consistent with the results of the NMIS deposit profile measurements. The NMIS measurements were in exceptional agreement to the later intrusive findings as exemplified by the results at one location where the pipe was cut during removal as shown in Figures 3 and 4. The dominant orange and yellow-green colors observed with the fiber-optic camera indicated an H/U ~ 3 -4, which was consistent with the NMIS measurements. The Hockey Stick deposit was successfully removed, weighed, and the estimated total uranium mass was 479 kg (± 50 kg uncertainty) which was consistent with the NMIS measurements.

These measurements were performed in the summer in a contaminated abandoned building with the temperature at the measurement location increasing to over 90°F on hot days. Since the facility was abandoned, there was no negative impact on facility operation, but the results had a significant positive impact on the removal of the deposit through enhanced nuclear criticality safety. NMIS measurements were also performed using this same methodology on an intersection of three large pipes and for the gate valve at the Hockey Stick deposit thereby alleviating criticality safety concerns at those locations also.

IV. HEU METAL RECEIPTS

Various HEU metal is returned to the Oak Ridge Y-12 Plant. In 1998 in order to avoid an NMC&A finding from DOE-ORO or increased operational cost for opening containers, NMIS was utilized in an unairconditioned storage warehouse to obtain quantitative estimates of the mass of HEU metal in containers. For calibration standards, HEU metal with two different masses was utilized. The signatures of the remaining HEU metal receipts were compared to the calibrations in order to obtain the HEU masses which were then compared to the declared. Comparisons of the NMIS HEU mass estimates with those declared are presented in Fig. 5. The agreement was adequate for avoidance of an NMC&A finding. These confirmations were performed in a timely, cost-effective manner with as many as 64 items measured in one shift utilizing 4.5 minutes of data acquisition time per item. Facility operation was cost effectively enhanced by this use of NMIS.

V. WEAPONS COMPONENTS IN STORAGE

In large storage arrays of fissile material (Fig. 6), gamma-ray background is sufficiently high to preclude practical use of gamma-ray spectrometry even when the HEU is unshielded. The fact that some of the NMIS signatures are not sensitive to background (or changes in background) make it particularly suitable for storage configurations. In 1997, NMIS was used in a large storage array at the Oak Ridge Y-12 Plant⁶ to resolve a DOE Inspector General's (IG) NMC&A audit finding with regard to confirmation of items in storage arrays. Since then, it has been used semiannually for inventories of this particular storage array and in FY 2000 will be used for most storage arrays of weapons components at the Y-12 Plant.

The signatures for the unknown containers were compared to those for a calibration container. Signatures were obtained with NMIS for the calibration container in a variety of conditions. The calibration container was opened and the HEU content of the component verified

by measurements out of the container with the source and detector adjacent to the weapons component. Other measurements or MCNP-DSP calculations⁷ of the measurement were used to determine the sensitivity of the signatures to the HEU content of the component. Measurements were then performed with the calibration component in its storage container and again MCNP-DSP calculations were used to obtain the sensitivity of the signatures to the presence of HEU. This process was repeated for all types of storage containers. For the particular component measured here, it was stored in three different containers. Therefore, three sets of calibration signatures were obtained.

A step in this process is of course obtaining the statistical precision of the calibration signatures and deciding on a measurement time. Before comparisons can be made between unknowns and the calibration signatures, the variation in the signatures for the calibration must be established. These variations are: (1) random, (2) variation from source-detector locations, (3) variations in the components themselves, and (4) variations resulting from location of the component in the container and packaging. The source-detector placement variations were obtained from repeated measurements with the three-calibration components in their containers where the source and detectors were removed and relocated several times around the container and the variations measured. The composite of variations (3) and (4) was obtained by measurements with several containers and included rotation of containers. Measurements were performed with the calibration container located in a variety of storage configurations one of which is shown in Fig. 7. With these data, if all items match the calibration, an average calibration signature and its acceptable variation were established and used for all subsequent confirmations. To display the signatures without revealing design information, the signatures for the unknown components in containers were divided by those for the calibration and the results subsequently displayed.

The signature that was used for the resolution of the IG's finding was the cross power spectral density (CPSD12) between detector 2 and the Cf source ionization chamber 1. This signature is a measurement of the correlated information in both signals and was used because for frequencies >0 Hz, it is not affected by the background.

These confirmations had to be performed without moving any of the containers in the storage array (Fig. 6) in which the containers were stacked four to a pallet, up to three pallets high. The confirmatory measurements proceeded by placing the source on one side of the container and the detectors on the other side. The source was located near the center of the pallet of four containers

with the detectors in the aisles between pallets. A typical result for this weapons component in one type of container is shown in Fig. 8 where the principal signature for this confirmation for a particular container divided by that of the calibration is plotted. The solid line, which is the ratio for the particular container shown, is within the acceptable variation (shaded band) of the calibration signature for the component in this container. This type of comparison was used to identify the weapons component in each container. Measurement times were 4.5 minutes with over 25 units measured in one shift (5 hours of access time) with considerable time being used to locate the source and detectors on opposite sides of a container in these large arrays without moving containers.

Recent (1999) measurements have shown that NMIS signatures can be used to determine attributes such as mass, shape, etc. of major constituents of weapons components in containers. This capability will be used to enhance future confirmations at Y-12.

The impact on facility operation is negligible since this is a storage array and confirmations were performed in-situ. These confirmations resolved NMC&A findings and the use of NMIS for this vault keeps the Y-12 Plant in compliance with the DOE orders for NMC&A.

VI. NMIS VERIFICATION OF MASS AND ENRICHMENT OF HEU METAL IN STORAGE

A series of nuclear materials identification system (NMIS) verifications has recently (1999) been completed for hundreds of birdcages containing highly enriched uranium (HEU) metal in storage at the Oak Ridge Y-12 Plant. These verifications of both HEU mass and enrichment were performed in the aisles between storage arrays with the HEU in birdcages as shown in Fig. 9. An array of scintillators was located adjacent to each birdcage containing the HEU metal and the ²⁵²Cf source was located adjacent to the HEU metal inside the birdcage.

This is the first time NMIS has been used to obtain enrichment. Since calibration standards of varying mass and enrichment were not available, 10 of the items themselves (with independent verification) were used for calibration standards. The sensitivities to HEU mass and enrichment were obtained from Monte Carlo neutron transport calculations using MCNP-DSP which simulates the NMIS measurement. The total correlated counts and first moment of the correlated detector counts after Cf fission (one NMIS signature) were the features used obtain a calibration "surface" versus the HEU mass and enrichment. Deviations between the measured and declared mass and enrichment are shown in Fig. 10 for some of these verifications.

These measurements of mass and enrichment were performed in a timely manner with as many as 55 measurements completed in one shift (~6 hrs of actual operation). They demonstrated a field use of NMIS for quantitative determination of HEU mass and enrichment. The data accumulation time was 4.5 minutes with change out of birdcages occurring in as little as 1.5 minutes. These verifications met the requirements to determine HEU mass and enrichment within a spread of $\pm 5\%$ (3 sigma). For the total number of verifications, the mean uranium mass was 0.2% above declared and the mean enrichment was 0.2% below declared.

This NMIS verification provides the capability to verify the mass and enrichment of HEU metal in storage at the Oak Ridge Y-12 Plant and in this case, eliminated the need to obtain enrichment from contact gamma-ray spectrometry using NaI detectors. This is the first use of NMIS to accurately measure both enrichment and uranium mass and resolved a DOE finding for NMC&A deficiency in this vault. Since these verifications could be performed with the HEU metals still in birdcages in the storage vault, airborne contamination was minimized. These verifications were a timely cost effective way of meeting the requirements of DOE order for NMC&A for HEU metal in this storage array without significantly impacting facility operations.

VII. OPERATIONAL CONSIDERATIONS

Three complete NMIS cart portable systems are now operational at the Y-12 Plant with a fourth to be added in FY 2000. This portability alleviates the need for permanent installation requiring dedicated floor space that could be used for other facility operation. NMIS is brought to the fissile areas as needed and stored in relatively unused facility space when not in use. NMIS has also been operational at LANL for measurements with plutonium in 1997 and at the Pantex Plant for measurements with fully assembled weapons systems and weapons components in 1998.

For active measurements with HEU, the total radiation dose from an unshielded Cf source^a of 1 μg of ²⁵²Cf is ~ 2.5 mr/hr at one meter. For the measurements described in this paper, the ²⁵²Cf source contained between 0.1 and 1.0 μg ²⁵²Cf. Utilization of NMIS at the Oak Ridge requires a radiation work permit with an area of ~ 16 \times 16 ft square for the larger sources, within which neutron dosimetry is required.

^aAlternate source such as an associated particle neutron generator which can be turned off when not in use could also be used. The advantages and disadvantages of sources are discussed in Ref. 8.

Each day before a measurement with fissile material is performed, a time-of-flight measurement with the source and detectors separated in air ~ 40 in., about 40 in. above the floor is performed to obtain the detection efficiency and make small adjustments if necessary. This verifies proper operation of the system and assures reproducibility of the detection system to within $\pm 1\%$ in detection efficiency. Stability of the total NMIS has been demonstrated over long periods of time for a variety of applications.

VIII. CONCLUSIONS

NMIS has been successfully applied for extensive field use at the Oak Ridge Y-12 plant for NMC&A and is particularly useful in the high gamma-ray backgrounds of storage arrays or for shielded HEU. NMIS active neutron interrogation has been used as a practical NMC&A capability for configurations not practical by passive measurements. In some cases, it has been more cost effective to measure in-situ than to remove items from a full vault to another area where gamma-ray background is low. For some vaults, this is nearly impossible, certainly impractical, and would be very costly. The use of a small ²⁵²Cf source (~1 μg with 2.5 mr/hr dose rate at 1 meter) and a cart portable system makes it very practical to set up NMIS almost anywhere in Oak Ridge with minimal impact on facility operations. Because of the cost-effective operation of NMIS, it has been utilized where other methods that would have significantly more facility impact could have been used. Also, it is important to recognize that NMIS was successfully used to obtain the enrichment as well as the uranium mass for HEU metal in storage at the Oak Ridge Y-12 Plant.

The impact of NMIS at the Oak Ridge Y-12 Plant is improved and more cost effective NMC&A as well as the resolution of NMC&A findings. NMIS operation at Y-12 is necessary for compliance with NMC&A requirements of the U.S. Department of Energy. NMIS use for physically characterizing the pipe deposit at the diffusion plant at the K-25 site enhanced nuclear criticality safety and was essential to the safe removal of the fissile material.

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Fig. 1. Verification Area Showing the Measurement Tables on the Left and Nearby Unverified Units Stacked on Pallets (Inset Shows M102 Container Out of Shipping Container).

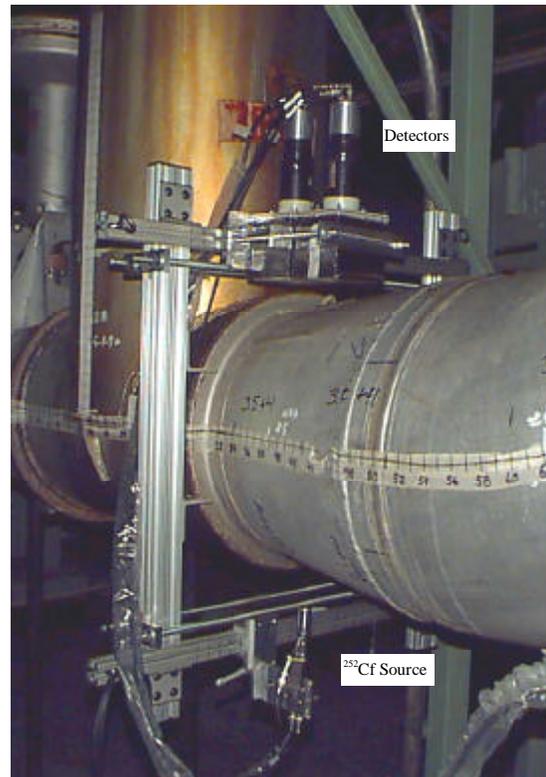


Fig. 2a. Source-Detector Configuration for a Horizontal Scan of Hockey Stick Deposit at Former Gaseous Diffusion Plant.



Fig. 2b. Overall View of the Hockey Stick Deposit at the Former Gaseous Diffusion Plant.

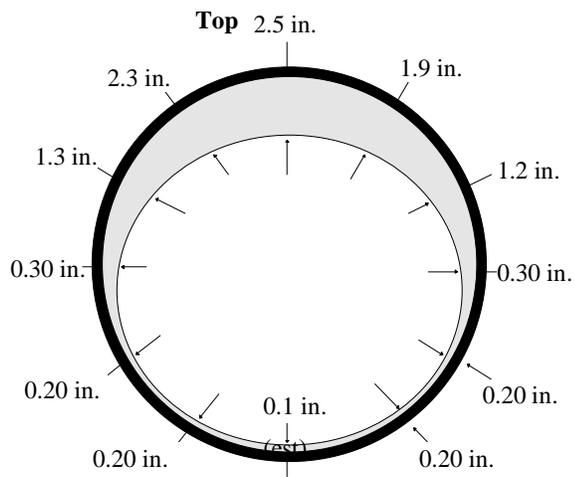


Fig. 3. NMIS Measurement Results at One of the Locations on the Hockey Stick Deposit.

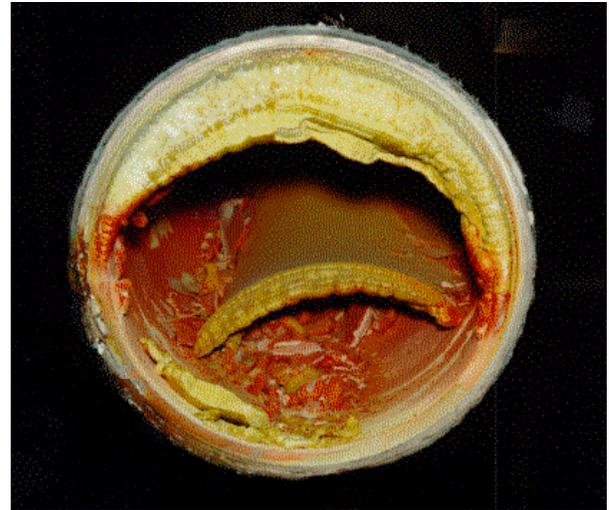


Fig. 4. The Actual Deposit Profile at the Same Location as Measured in Fig. 3. Note That Some Material Has Fallen During Removal of the Pipe.

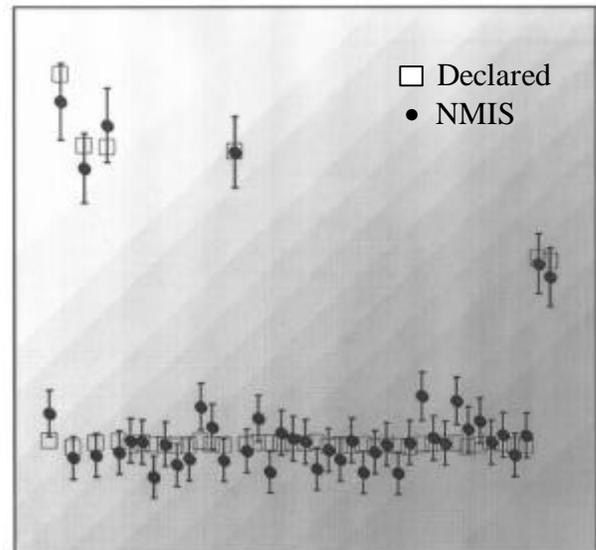


Fig. 5. Comparison of NMIS HEU Masses With Declared For Receipts.



Fig. 6. Storage Array Configuration.



Fig. 9. Equipment Set Up for Verification of HEU Metal.



Fig. 7. Configuration of NMIS in Use at Y-12 Storage Facility.

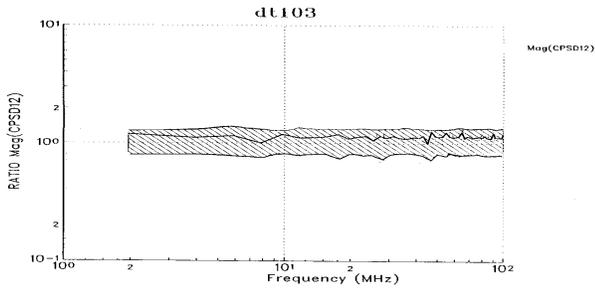


Fig. 8. Comparison of CPSD12 with the Calibration Signature and Its Variation.

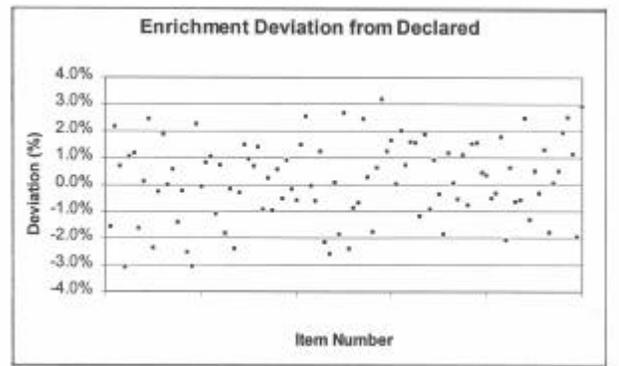


Fig. 10a. Percent Deviation of NMIS Enrichment from Declared.

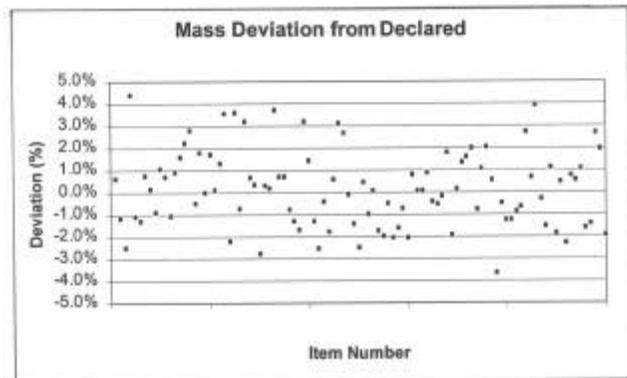


Fig. 10b. Percent Deviation of NMIS Mass from Declared.