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CAPABILITIES OF NMIS FOR VERIFICATION OF HEU STORAGE CANS ENTERING AND IN THE HEUMF

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June 2003



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**CAPABILITIES OF NMIS FOR VERIFICATION OF HEU STORAGE CANS
ENTERING AND IN THE HEUMF**

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CAPABILITIES OF NMIS FOR VERIFICATION OF HEU STORAGE CANS ENTERING AND IN HEUMF

ABSTRACT

The Nuclear Materials Identification System (NMIS) properly designed to interrogate highly enriched uranium (HEU) storage cans going into and in the Highly Enriched Uranium Materials Facility (HEUMF) could determine: 1) shape, 2) fissile mass, 3) enrichment, 4) presence of oxide, 5) presence of fluoride, and 6) density and do it in less than five minutes of measurement time. If these quantities are desirable, the NMIS system could be modified for this application and be installed and operational to verify the initial movement of HEU storage cans into HEUMF.

1. INTRODUCTION

The Nuclear Materials Identification System (NMIS) has a variety of capabilities for verification of highly enriched uranium (HEU) storage cans at the Highly Enriched Uranium Materials Facility (HEUMF). These capabilities are as follows:

1. Image the contents of the can to determine the shape of the material,
2. Determine the mass to ~1% accuracy for castings,
3. Determine the enrichment to 1% accuracy,
4. Determine the presence of oxide,
5. Determine the presence of fluoride,
6. Determine the density.

All six verifications could be determined within five minutes of NMIS data accumulation. Only capability number five would not require the presence of a small ^{252}Cf spontaneous fission source. Capabilities two and three would require calibration standards. The Oak Ridge National Laboratory (ORNL) is not proposing what will be done but only what can be done with some modifications of the present NMIS hardware and software. These capabilities are based on previous experience at the Y-12 National Security Complex, simple physics considerations, or elaborate Monte Carlo neutron and gamma ray transport theory calculation using modifications of the MCNP code of the Los Alamos National Laboratory.

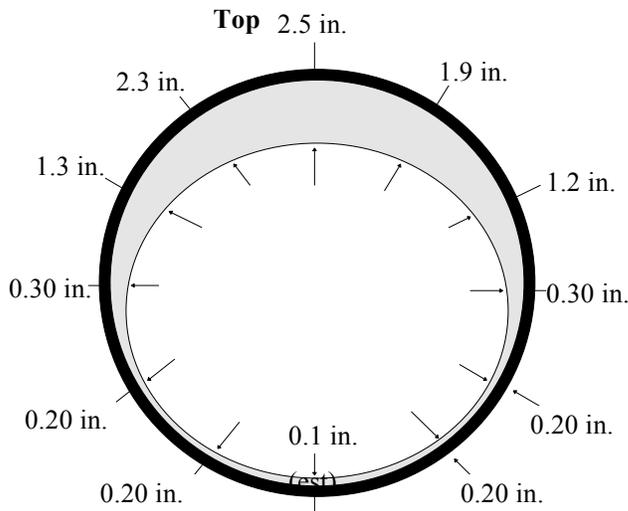
This report will briefly elaborate on these capabilities and reference previous Y-12, and in some cases, ORNL reports.

2. IMAGING

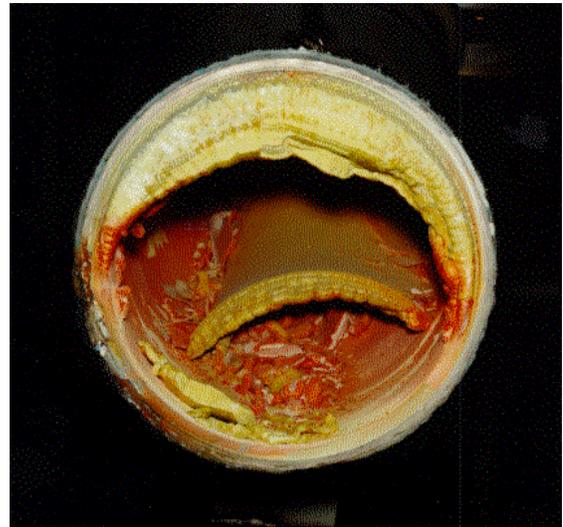
2.1 MEASUREMENTS

ORNL has successfully imaged the contents of a 24-in.-diam. pipe at the K-29 building of the shutdown Oak Ridge Gaseous Diffusion Plant that had a large deposit, estimated from gamma ray spectrometry measurements to be uniformly distributed and to contain

~1000 kgs.^{1,2} This deposit was called the hockey stick because of the shape of the pipe. In this application NMIS used scanning of the source with two detectors to produce a gamma and neutron radiograph of the pipe and its contents. NMIS measurements showed that the deposit was not uniformly distributed and contained approximately half the mass estimated from gamma ray spectrometry. Much to the consternation of engineering, the deposit was on the upper inner surface of the pipe at the lower portion of the hockey stick. A photograph of a cut section of the hockey stick during removal is shown in Fig. 1 where the deposit's shape is compared to the shape obtained by NMIS measurements.



NMIS Measurement results at the L = 54 location



Photograph of actual deposit profile at L = 54. Note that some material has fallen during removal of the pipe.

Fig. 1. Comparison of NMIS Measurements of the Shape With the Actual Photograph of the Lower Section of the Hockey Stick During Dismantlement.

2.2 HEU CASTING IN A CAN

A series of Monte Carlo calculations were performed to simulate imaging a casting in a can.³ The Cf source was adjacent to the can with an arc of small (1-in.-square) plastic scintillation detectors on the opposite side so that the neutrons and gamma rays travel through the casting to the detector. The calculations had a large number of detectors whereas in actual application the can could be mounted on a small rotating lazy susan with a small number of detectors (~10 costing about \$15,000 for all 10). Standard imaging algorithms were applied to the NMIS transmission data to reconstruct the image of the casting. The results of these simulations are given in Fig. 2 for the equivalent of one minute of experimental data using a 1.5 microgram Cf source. To test the ability to detect additional material inside the annulus, a 1-cm-diam. polyethylene rod was located on the axis of the standard Y-12 annular storage casting. Clearly visible in the reconstructed image are the annular shaped casting, the polyethylene rod, and even the thin steel can.

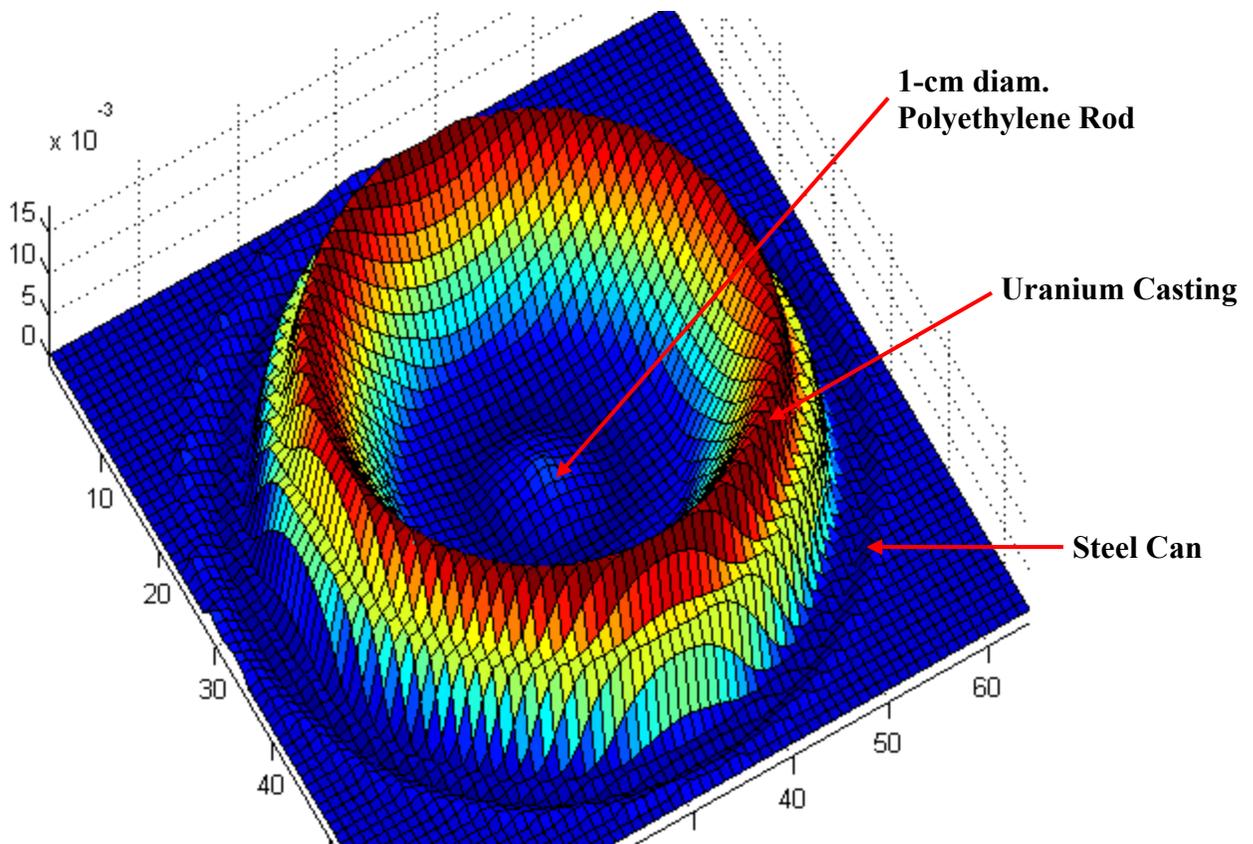


Fig 2. Reconstructed Image of a HEU Annular Casting With a 1-cm-diam. Polyethylene Rod on Axis in a Thin Steel Can From One Minute of NMIS Data.

3. FISSILE MASS AND ENRICHMENT

3.1 MEASUREMENTS WITH CASTINGS

NMIS measurements have been performed on cans with annular castings to investigate the practicality of NMIS measurements for castings.⁴ A previous measurement is shown in Fig. 3 where a Cf source is located on one side of a can with an annular HEU casting, in this case, 93 wt % enriched uranium.

NMIS measurements could determine the mass and enrichment to ± 1 % of casting in vault 16 without removing castings from the storage trays to satisfy International Atomic Energy Agency (IAEA) requirements and alleviate the safety and radiological concerns associated with the manual handling of the cans containing the castings in the present method of verification.



Fig. 3. NMIS Measurements for an HEU Annular Casting in a Steel Can.

3.2 MEASUREMENTS TO RESOLVE A NMC&A FINDING AT Y-12

NMIS measurements were performed to obtain the mass and enrichment of HEU metal in bird cages in a vault at Y-12.⁵ This oxidized HEU could not practically be removed from the bird cages. A Cf source was located adjacent to the part in the bird cage and an array of four detectors was located at the outer perimeter of the bird cage as shown in Fig. 4.



Fig. 4. NMIS Measurement of HEU in Bird Cages.

These measurements were performed in the aisle ways of the vault and were not affected by metric tons of nearby materials since they were not significantly affected by background

gamma radiation. These measurements that used calibration standards, for which the mass was obtained by weighing and the enrichment from gamma ray spectrometry, obtained the mass and enrichment to a relative uncertainty of $\pm 1.5\%$ in a cost effective way with 4.5 minutes of NMIS data with as many as 55 verifications in one shift of operation in the vault.

These NMIS measurements were the first determination of enrichment of HEU at Y-12 by a method other than gamma ray spectrometry or mass spectrometry.

3.3 CALCULATIONS AND SIMULATIONS FOR MASS AND ENRICHMENTS

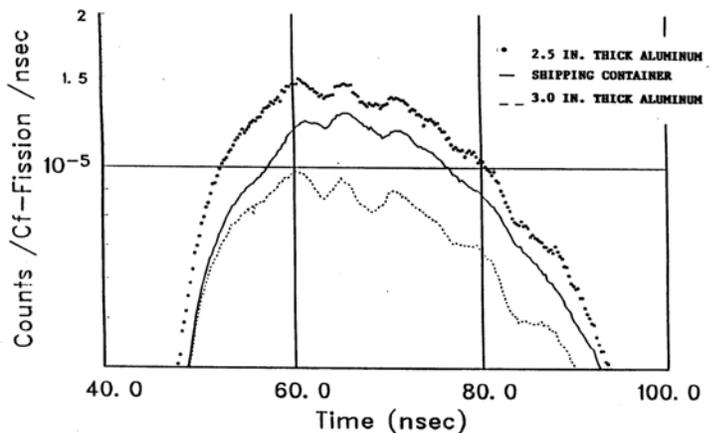
Monte Carlo calculations were used to provide NMIS signatures for various geometric shapes of varying enrichments and masses.⁶ These simulations then used to test the capability of NMIS to measure mass, enrichment, and shape and the results of this analysis showed that the mass and could be determined to $\pm 1\%$, with less accuracy for the enrichment.

4. PRESENCE OF OXIDE

Previous measurements with trainers at Y-12 showed that the neutron transmission data from NMIS could be used to verify the presence and thickness of material. In this particular case NMIS measurements were used to determine that the material was aluminum. The configuration of the source and detectors near the 1-meter-long box containing the trainer is shown in Fig. 5. This was done by comparing the signature for the trainer with those for small, cylindrical samples of aluminum. The data are shown in Fig. 5. The variations in the neutron transmission identified the material as aluminum. From this data the aluminum thickness was determined to be 2.75 in.



Source-Trainer -Detector Geometry



Time-distribution of neutrons after ²⁵²Cf-fission (889 mm between source and detector)

Fig. 5. Neutron Transmission Measurements for Trainer Consisting of an Aluminum Casting (Photograph and Data)

Several years ago we were asked by Savannah River Laboratory if NMIS could determine if Pu buttons had oxidized in their sealed containers. Monte Carlo calculations were performed to evaluate this possibility.⁷ These evaluations have shown that NMIS transmission measurements could determine if as little as 10% of the Pu metal had oxidized. This same transmission measurement could be used to verify the presence of oxide.

In addition, the cross correlation between detectors obtained by NMIS for oxide is significantly different from metal because the presence of oxygen broadens the time distributions so that it is characteristically different from metal. The presence of oxygen changes the prompt neutron decay of the fission chains so that the fission chain multiplication process occurs on a longer time scale. Previous Monte Carlo simulations have demonstrated this effect.⁸ This longer time scale for oxide would also be evident in the time correlations between the source and a detector.

5. PRESENCE OF FLUORIDE

The presence of fluoride can easily be determined without the Cf source since the alpha neutron (α,n) reaction will produce detectable neutrons.

6. DENSITY

Gamma ray transmission is dependent on the density of materials. If the shape of the material from imaging and the mass is known the density can be estimated as well as determined from gamma ray transmission.

The hockey stick deposit at the K-29 building is an example of using NMIS gamma ray transmission to determine the density. In the hockey stick application the neutron transmission was used to determine the shape. Knowing the shape, the gamma transmission was used to obtain the density, which is a strong function of hydrogen to uranium (H/U) ratio that drastically impacts criticality safety. From the color (H/U effects color of the deposit) engineers observing the deposit during removal estimated that the H/U ratio was about 3.5, whereas the NMIS measurements predicted 3.4 before the pipe was examined internally.

7. CONCLUSIONS

A NMIS system properly designed to interrogate HEU storage cans going into and in the HEUMF could determine: 1) shape, 2) fissile mass, 3) enrichment, 4) presence of oxide, 5) presence of fluoride, and 6) density and do it in less than five minutes of measurement time. If these quantities are desirable, the NMIS system could be modified for this application and be installed and operational to verify the initial movement of HEU storage cans into HEUMF.

REFERENCES

1. T. Uckan, M. S. Wyatt, J. T. Mihalczo, T. E. Valentine, J. A. Mullens, and T. F. Hannon, “ ^{252}Cf -Source-Correlated Transmission Measurements for Uranyl Fluoride Deposit in a 24-in.-OD Process Pipe,” *Nuclear Instruments and Methods in Physics Research, A* **422**, 26-34 (1999).
2. T. Uckan, M. S. Wyatt, J. T. Mihalczo, T. E. Valentine, J. A. Mullens, T. F. Hannon, ORNL/TM-13642, *Fissile Deposit Characterization at the Former Oak Ridge K-25 Gaseous Diffusion Plant by ^{252}Cf -Source-Driven Measurements*, Oak Ridge National Laboratory, May 1998.
3. J. A. Mullens, Y/LB-16,160, “Addition of Tomographic Capabilities To NMIS,” the Y-12 National Security Complex, March 11, 2003.
4. T. E. Valentine, J. K. Mattingly, J. T. Mihalczo, “ ^{252}Cf -Source-Driven Noise Analysis Measurements with Annular High-Enriched Uranium Metal Castings,” paper presented to the American Nuclear Society International Conference on Physics of Nuclear Science and Technology, Vol. 2, 1129-1137, Long Island, New York, October 5-8, 1998.
5. L. G. Chiang, J. K. Mattingly, J. A. Ramsey, and J. T. Mihalczo, “Verification of Uranium Mass and Enrichments of Highly Enriched Uranium (HEU) Using the Nuclear Materials Identification System (NMIS),” paper presented at the Institute of Nuclear Materials Management Conference, New Orleans, Louisiana, July 16-20, 2000.
6. S. A. Pozzi and F. J. Segovia, ORNL/P00-107760, “Application of stochastic and Artificial Intelligence Methods for Nuclear Material Identification,” Oak Ridge National Laboratory, July 2000.
7. T. E. Valentine and J. T. Mihalczo, “Evaluation of Plutonium Oxidation Analysis Using Pulsed Neutron Measurements With ^{252}Cf ,” Institute of Nuclear Materials Management Meeting, Phoenix, Arizona, July 20-24, 1997.
8. S. A. Pozzi, Enrico Padovani, and J. T. Mihalczo, “MCNP-POLIMI Evaluation of Time-Dependent Coincidences Between Detectors for Fissile Metal vs. Oxide Determinations,” Y/LB-16,133, May 2002.

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