ORNL/CP-98771

CONF-981003--

Physics of Enriched Uranyl Fluoride Deposit Characterizations Using Active Neutron andGamma Interrogation Techniques with ^{252}Cf

M. S. Wyatt
T. Uckan
J. T. Mihalczo
T. E. Valentine

Oak Ridge National Laboratory*
P.O. Box 2008
Oak Ridge, TN 37831-6004
(423) 574-0284

T. F. Hannon
East Tennessee Technology Park
P.O. Box 2009
Oak Ridge, Tennessee 37831

RECEIVED
JUL 29 1998
OSTI

Presented at the
International Conference on the Physics of Nuclear Science and Technology
October 5 - 8, 1998
Long Island, New York

"The submitted manuscript has been authored by a contractor of the U.S. Government under contract No. DE-AC05-96OR22464. Accordingly, the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or allow others to do so, for U.S. Government purposes."

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

MASTER

* Managed by Lockheed Martin Energy Research Corporation for the U.S. Department of Energy under contract DE-AC05-96OR22464.

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

PHYSICS OF ENRICHED URANYL FLUORIDE DEPOSIT CHARACTERIZATIONS USING ACTIVE NEUTRON ANDGAMMA INTERROGATION TECHNIQUES WITH ^{252}CF

Mark S. Wyatt
University of Tennessee
Department of Nuclear Engineering
Knoxville, TN 37996-2300
(423) 974 - 2525

Taner Uckan
J. T. Mihalczo
T. E. Valentine
Oak Ridge National Laboratory
P. O. Box 2008
Oak Ridge, TN 37831-6004
(423) 574 - 8008

Terry F. Hannon
East Tennessee Technology Park
P. O. Box 2009
Oak Ridge, TN 37831

ABSTRACT

A method was developed and successfully applied to characterize large uranyl fluoride (UO_2F_2) deposits at the former Oak Ridge Gaseous Diffusion Plant. These deposits were formed by a wet air in-leakage into the UF_6 process gas lines over a period of years. The resulting UO_2F_2 is hygroscopic, readily absorbing moisture from the air to form hydrates as UO_2F_2 - nH_2O. The ratio of hydrogen to uranium, denoted H/U, can vary from 0 - 16, and has significant nuclear criticality safety impacts for large deposits. In order to properly formulate the required course of action, a non-intrusive characterization of the distribution of the fissile material within the pipe, its total mass, and amount of hydration was needed. The Nuclear Weapons Identification System[1] (NWIS) previously developed at the Oak Ridge Y-12 Plant for identification of uranium weapons components in storage containers was used to successfully characterize the distribution, hydration, and total mass of these deposits.

I. BACKGROUND

Three separate deposits, all located in the same process building, were successfully characterized using the methods to be described. The first deposit existed in a 17-ft.-long, 24-in.-O.D. process gas line that because of its shape became to be known as the "Hockey Stick". The formation of this deposit was believed to be from a wet air in-leakage in the convoluted metal bellows of the block valve immediately upstream of the Hockey Stick; this block valve was the location of the second deposit that was characterized. The Hockey Stick and the block valve are shown in Figure 1.

The Hockey Stick is composed of a 6 ft. horizontal section and a 10 ft. vertical section that is at a 60 degree angle to the floor and terminates into a common header 13.5 feet above the floor. Previous passive Non-Destructive Assay (NDA) measurements using gamma ray spectrometry and neutron counting indicated that the Hockey Stick contained 1190 kg ± 595 kg of UO_2F_2 enriched to 3.3 wt% ± 0.66 wt% ^{235}U. The NDA measurements also indicated that the deposit was uniformly distributed around the circumference of the pipe as well as along the length of the pipe. These measurements are somewhat limited due the self-absorption of the 186 keV decay gamma in thick deposits and the low spontaneous fission rate of uranium for neutron counting.

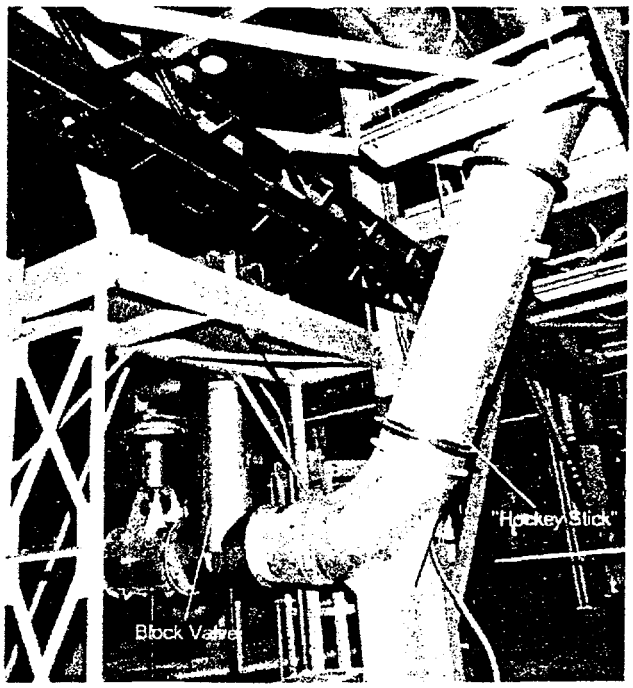

Figure 1. 24" O.D. Pipe (Hockey Stick) and Block Valve

The second deposit in the block valve immediately adjacent to the Hockey Stick was believed to exist in the

upper internals (around the valve stem and bellows). The valve is a 20-in.-dia. double disc gate valve and the valve bonnet surrounding the internals has an O.D. of 24 in. and a height of 36 in. Previous passive NDA measurements indicated that the block valve contained approximately 156 kg ± 78 kg of material enriched to 3.3 wt% ± 0.66 wt% ^{235}U. However, the large deposit in the Hockey Stick interfered with the gamma ray spectrometry measurements on the valve necessitating a second confirmatory measurement.

The third deposit existed in a pipe intersection consisting of a 10 ft long 30-in.-dia. horizontal pipe and a 12 ft long 24-in.-dia. vertical pipe joined via a custom fabricated transition piece (T-Pipe), and is shown in Figure 2. Previous passive NDA measurements indicated the T-Pipe contained approximately 240 kg ± 120 kg of material enriched to 3.4 wt% ± 0.68 wt% ^{235}U. The mechanism leading to the formation of this deposit was unknown.

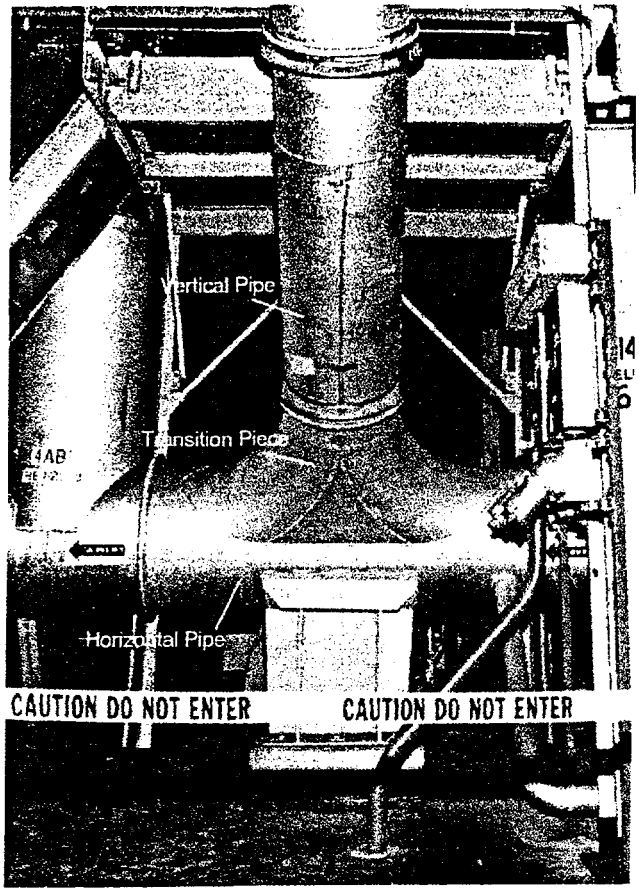

Figure 2. The "T-Pipe" is a 30" O.D. horizontal pipe conjoined to a 24" O.D. vertical pipe via a custom transition piece.

Each of the three deposits were a significant nuclear criticality safety concern. In the Hockey Stick, the 60-degree incline and the large diameter piping lead to extremely unfavorable conditions for preventing a nuclear criticality accident. A disturbance of the deposit could cause it to collapse and collect in the lower portion of the process piping. UO_2F_2 is hygroscopic and readily absorbs moisture from the air to form hydrates until the stable compound $UO_2F_2 \cdot 2H_2O$ is reached (H/U = 4). It can then continue to absorb excess moisture in a volume-additive manner until an H/U of 16 is reached, at which point it begins to deliquesce. Monte Carlo calculations using the SCALE Criticality Safety Analysis Sequence IX module (XSDRNPM-S) and the ENDF-B/IV 27-group cross section library concluded that a critical configuration could be reached with an H/U ratio of 2 if the lower section of pipe were completely filled; at an H/U of 4 only ½ of the lower pipe section needed to be filled.[2]

The primary concern in the T-pipe was the material distribution. If the majority of the deposit was located in the transition piece or the vertical pipe, any disturbance of the material could cause the deposit to fall and collect directly below in the horizontal pipe, possibly leading to a nuclear criticality accident. Under optimal conditions the minimum critical mass is 102 kg for UO_2F_2 (3%) and 55.5 kg for UO_2F_2 (4%),[3] compared to the 240 kg ± 120 kg thought to exist.

The 156 kg ± 78 kg of material thought to exist in the block valve also exceeded the minimum critical mass of material under optimal conditions of moderation and reflection. Criticality safety evaluations normally assume an H/U of 4, so in each case credit for the material distribution had to be taken in order to demonstrate subcriticality, thus necessitating deposit characterization followed by removal of the deposit if necessary.

II. THEORY OF MEASUREMENT

Briefly, NWIS consists of an external ^{252}Cf source in a parallel plate ionization chamber, two or more fast plastic scintillation detectors for measuring the arrival of neutrons and gamma's, a custom-built PC based 5 channel data acquisition and control board that has a sampling capability of up to 1 GHz, and a standard PC to process and display the data. The ^{252}Cf ionization chamber serves as a timed source of spontaneous fission neutrons and gamma's. Neutrons and gamma's emitted from the fission event either traverse the pipe and deposit with no interaction (transmission), are scattered within the material, or initiate the fission chain multiplication process. The detectors then measure the time distribution of counts that occur after the initiating event at the source. Since the time of fission is marked by the pulse from the

source ionization chamber, the energy of the neutron is determined from the measured time it takes to travel the known distance between the source and detector using the non-relativistic kinetic energy equation – a neutron time-of-flight (TOF) measurement. Gamma rays that are directly transmitted travel the same speed regardless of energy, thus their arrival at the detector after fission is based solely on the separation between the source and detector. Particles that are scattered arrive later in time than those that are directly transmitted, in essence providing a collimated source without having to heavily shield the detector, i.e., collimation by timing rather than lead. The method can be applied in high background radiation areas, since any background radiation is uncorrelated in time with the source. The time correlated signature is equivalent to the right half of the cross-correlation function between the source and detector and is essentially a randomly pulsed neutron measurement.[4]

As UO_2F_2 hydrates, the material density changes due to the displacement of the dense uranium component by the relatively light hydrogen. The uranium density in water moderated mixtures of UO_2F_2 - nH_2O can be estimated from Eq.'s (1) and (2)

$$\rho_u = 4.96 - 0.32(H/U), \qquad H/U < 4 \qquad (1)$$

$$\rho_u = \frac{m_u}{V_{UO_2F_2} + \left(\frac{H}{U} - 4\right)\frac{V_{H_2O}}{2}}, \qquad H/U > 4. \qquad (2)$$

where V is the molar volume of the compound and m is the molecular weight of uranium at the given enrichment. There is a discontinuity in the density at an H/U of 4 due to the change in water addition by the formation of hydrates to that of a volume-additive addition.[5]

The material density can change by about a factor of four over the full range of hydration, making a normal transmission measurement determination of the material properties impossible since neither the density or thickness of the deposit is known. The ^{252}Cf provides a timed source of neutrons and gamma's yielding two simultaneous measurements.

In the first measurement high energy neutrons (> 6 MeV) are used to measure the deposit thickness and construct a profile of the deposit distribution. This is possible because the neutron total interaction cross-section above 6 MeV is essentially flat with hydration, varying by less than 10 percent. Above 1.5 MeV the hydrogen cross-section rolls off sharply leaving the ^{235}U and ^{238}U cross-sections to dominate. However, the relative abundance of the hydrogen is much greater than the uranium and these two competing effects offset each other, leading to the relatively flat total interaction cross-section. As shown in Figure 3, the high energy cross-sections are only weakly affected by perturbations of the incident neutron energy which is ideal in a TOF measurement. Since the total interaction cross-section is approximately constant and can be calculated for any hydration (density), the deposit thickness can be determined from the high energy neutron transmission data.

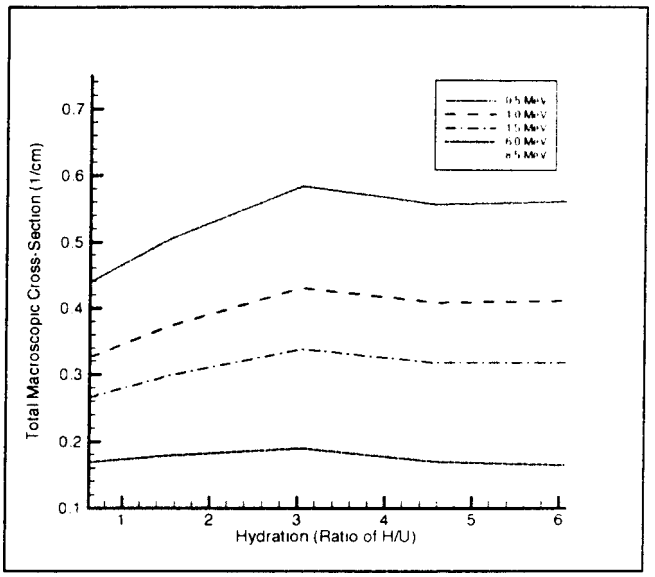

Figure 3. Total macroscopic neutron cross-sections as a function of hydration for varying energies.

The second measurement uses the gamma ray emission spectrum from ^{252}Cf to determine the deposit hydration, which is related to the material density. The gamma ray cross-sections are highly Z-dependent, making the measurement well suited for this application. Due to the fast timing resolution of the NWIS system (1 nsec), gamma's that are scattered or interact arrive later in time than those that are directly transmitted, in essence providing a perfectly collimated source used to measure the material mass attenuation coefficient. However, because all transmission gamma rays arrive at the detector at the same time regardless of energy it is necessary to use a spectrum-weighted approach to determine the material properties as shown in Eq. 3

$$\frac{A\gamma}{A_{ref}} = \frac{\int_0^\infty dE\, G_o(E)\, e^{-\mu(E)X}\, e^{-\sum_i \left(\frac{\mu(E)}{\rho}\right)_i \rho_i S}}{\int_0^\infty dE\, G_o(E)\, e^{-\mu(E)X}}. \qquad (3)$$

where A_γ is the detector response due to the pipe and deposit, A_{ref} is the detector response to the clean (empty) pipe, $G_o(E)$ is the ^{252}Cf gamma ray emission spectrum, S is the deposit thickness, and X is the pipe thickness. Because the weight fractions of each nuclide and the material density can be calculated for any hydration; and because the ^{252}Cf gamma ray emission spectrum and the material properties of the clean pipe are well known, it is possible to solve this equation numerically. Energy dependent mass attenuation coefficients from the National Nuclear Data Center [6] were obtained over the range of 0.1 to 8 MeV. These results were folded with the known ^{252}Cf gamma ray emission spectrum and the integral evaluated using an 8 point Gaussian quadrature scheme using the estimate of the deposit thickness obtained from the neutron data. The term for the deposit density is then adjusted until a best estimate is obtained from Eq. 4

$$\left(\frac{A_\gamma}{A_{ref}}\right)_{meas.} - \left(\frac{A_\gamma}{A_{ref}}(\rho, S)\right)_{calc.} \approx 0. \qquad (4)$$

The deposit density is distinctly related to the hydration, and thus the first estimate of H/U is obtained.

The measurement results are then refined by a series of iterative calculations. The process begins by assuming a constant total macroscopic cross-section for neutrons (which in fact varies by about ten percent over the range of hydration). Once the thickness is determined from the neutron portion of the measurement, the hydration is calculated from the gamma portion of the measurement. This hydration is then used to refine the neutron cross-sections and obtain a new thickness from the neutron data. This new estimate of the thickness is then re-applied to the gamma transmission measurement for a new estimate of H/U. This process continues until values of the thickness and hydration are acceptably converged. One caveat to the iterative procedure is that is does not work for thin deposits. The exponential behavior of the transmission gamma rays yield large variations in the mass attenuation coefficient for small uncertainties in the measured deposit thickness. Because the deposit hydration is derived from the mass attenuation coefficient the procedure breaks down.

III. MEASUREMENT RESULTS

Reference measurements were performed on clean pipe sections and a block valve identical to those being characterized in order to remove the contribution of the pipe walls and valve internals from the measurement. The material distribution in each location was then mapped by performing a series of horizontal, vertical, and rotational scans with the source and detectors. A typical measurement setup is shown in Figure 4.

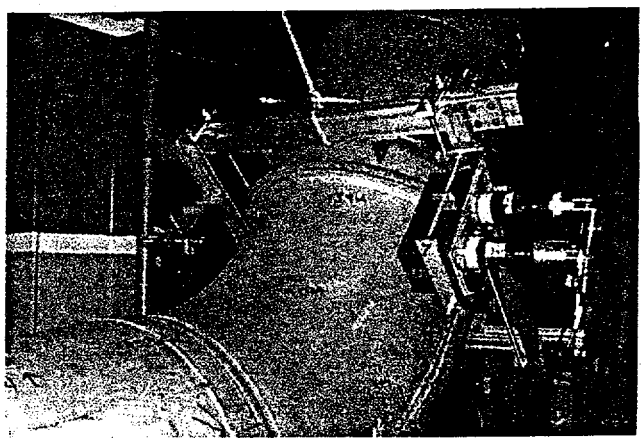

Figure 4. ^{252}Cf source on left; detectors on right.

A. Material Distribution in the Hockey Stick

The Hockey Stick deposit was the most significant safety concern and therefore was the first deposit characterized. Cross sectional profiles of the material distribution were determined from measurements taken at the six locations shown in Figure 5.

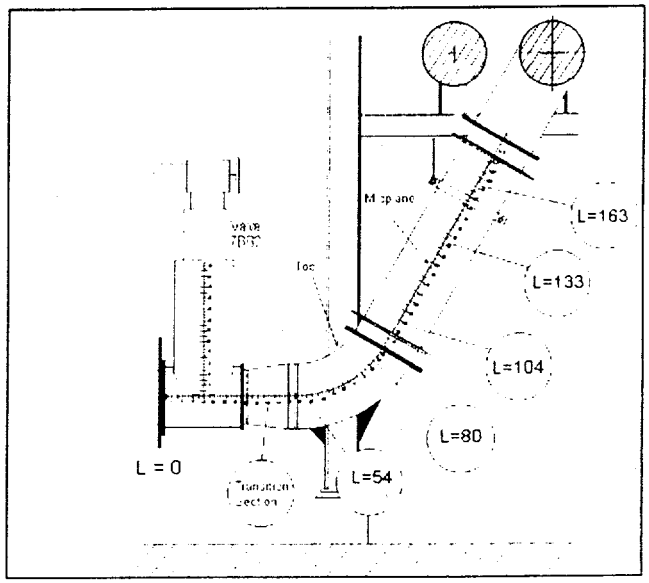

Figure 5. Locations characterized for Hockey Stick

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 measurement findings were in exceptional agreement to the later intrusive findings as

shown by the results at one location along the pipe in Figures 6 and 7.

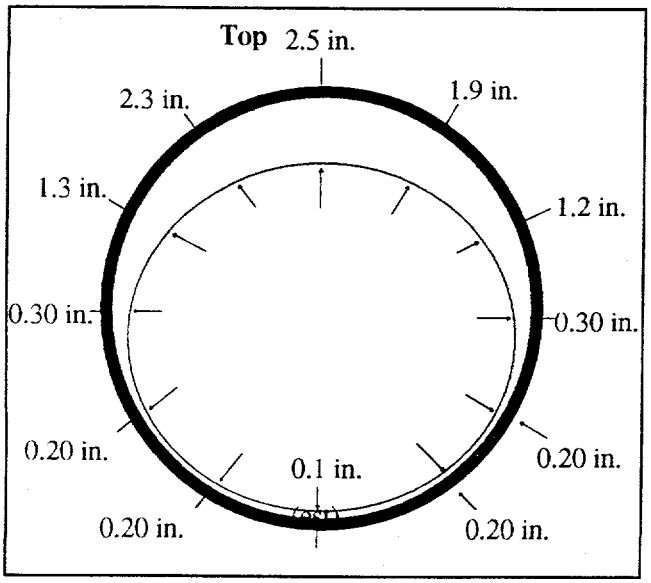

Figure 6. Measurement results at the L = 54 location

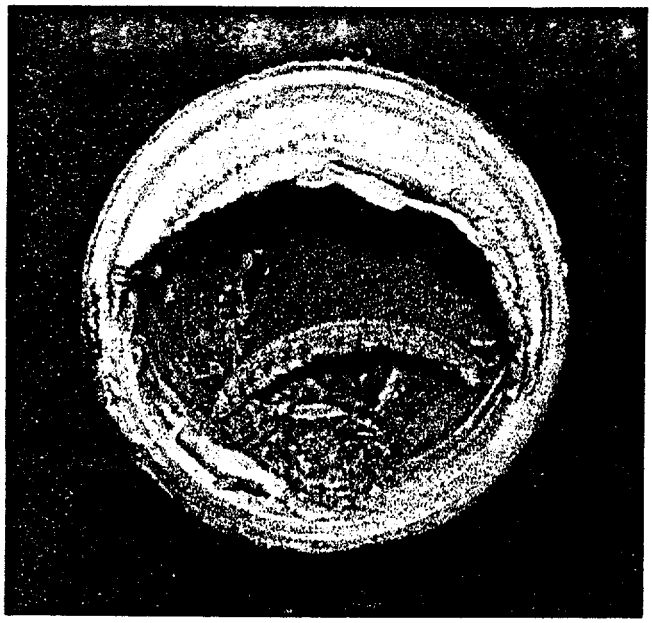

Figure 7. Actual deposit profile at L=54. Note that some material has fallen during removal of the pipe.

The measured deposit profile were used with the gamma ray transmission measurements to obtain the H/U values. To eliminate the effects of the extreme sensitivity of thin deposits in resolving the H/U values, deposit locations less than 1" thick were not used in the analysis. Approximately 32 measurement locations were used in the analysis, which had an average H/U value of 3.4 ± 0.25, where the uncertainty is the standard deviation of the mean. An estimated total deposit volume of 0.142 m^3 was made based on the 6 measured profiles. Using the average value of H/U and its associated uncertainty, a uranium density of 3.87 ± 0.08 g/cm^3 is obtained from Eq. 1 for H/U < 4. The total uranium mass from the measurements was estimated to be 552 ± 93 kg, which compared favorably to the 478.64 kg removed. The dominant orange and yellow-green colors of the deposit, which are related to the H/U, indicate an H/U of ~ 3 to 4, also consistent with the measurement findings.

B. Material Distribution in the T-Pipe

In both the valve and the T-Pipe the deposits were very thin, necessitating that the value of H/U be estimated by using the results from previous measurements on the Hockey Stick. Referring to Figure 2, measurements found no significant deposition in the extreme left or middle portion of the horizontal pipe, with exception of a thin coating around the perimeter approximately 0.1" thick. On the inlet (right) side of the horizontal pipe a roughly annular deposit of 0.25" to 0.34" thick was measured. The large support structure and irregular surfaces prevented a thorough investigation of the bottom of the horizontal pipe. In the transition piece no material was noted on the front or rear faces but a deposit approximately 0.2" thick was found on each of the curved shoulders. The vertical riser contained a annular deposit that tapered from 0.2" thick near the bottom to 0.1" thick approximately 75" up the riser, which was as far as could be measured for safety concerns. Based on the measured material distribution and the H/U value assumed from the Hockey Stick measurements, the estimated material mass was 93 kg. The deposit mass removed from the T-pipe, based on gross scale measurements, was 126 kg which compared favorably to the measured estimate of 93 kg and was significantly less than the previous estimates of 240 kg ± 120 kg from gamma ray spectrometry and neutron counting with the T-Pipe assembled.

C. Block Valve

Time constraints associated with project deadlines did not allow for an extensive characterization of the block valve. Only three set of vertical scans were performed by moving the ^{252}Cf source and detectors in conjunction up the valve bonnet in 6" vertical increments. The vertical scans were spaced 6" apart on the horizontal; one on the vertical midline of the valve and two on either side spaced 6" apart. The measurements found that the majority of the valve bonnet contained no deposit whatsoever, with the exception of a slight deposition on one edge. Due to the small number of measurements taken from only a single

direction it is impossible to reconstruct even a rough image of the material distribution in this area. Assuming a slab-like geometry viewed orthogonally, the measurements indicated 0.1" to 1.0" of material was present with an estimated total mass of 13 kg. A normalized representation of the deposit distribution is presented in Figure 8.

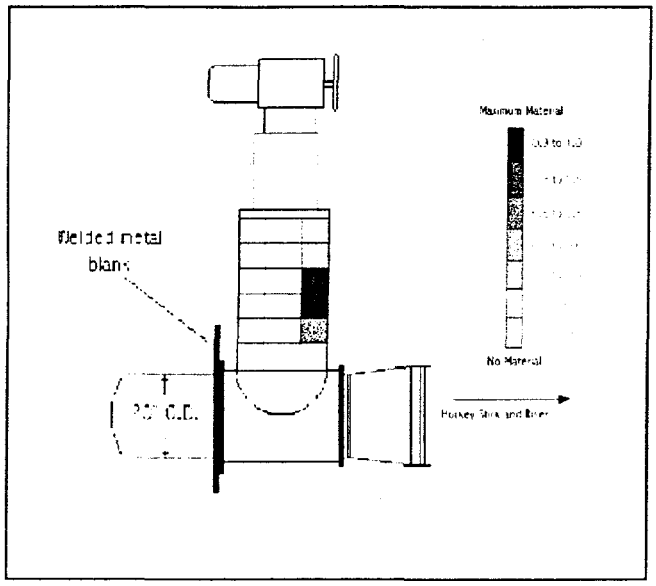

Figure 8. A representation of the deposit distribution in the valve. Note that the distribution has been normalized.

Based on these measurement results it would be impossible to have the 156 kg ± 78 kg of deposit previously estimated and that most likely even much less material existed. Therefore, the decision was made to forego any deposit removal activities on the valve. At a later date, after the Hockey Stick deposit had been removed which interfered with the gamma spectrometry measurements, NDA personnel re-measured and concluded that it contained less than 14.8 kg of material.

IV. CONCLUSIONS

It was successfully demonstrated that the Nuclear Weapons Identification System (NWIS) provides a reliable method for non-intrusive characterization of hydrated uranyl fluoride deposits. The characterization can be performed when neither the material mass or density is known, and can be successfully applied in environments where other methods fail due to high background radiation conditions or self-shielding effects. The method can be used to determine the material distribution, the level of hydration, and the total mass of the deposit. This technique may be applied to other UF_6 gaseous diffusion plants for deposit measurements, for other types of holdup measurements such as in freezer-sublimers and process gas coolers, or for other processes with nuclear material.

REFERENCES

1. Mihalczo, J. T., Valentine, T. E., Mullens, J. A., and Mattingly, J. K., "Physical and Mathematical Description of the Nuclear Weapons Identification System NWIS Signatures," Oak Ridge Y-12 Plant report No. Y/LB-15, 946 R3, September 1997.

2. Haire, M. J., Elam, K. R., and Jordan, W. C., "Nuclear Criticality Analyses of a Uranium Deposit in the K-29 Building, K-25 Site, Oak Ridge, Tennessee," K/ER-310, Oak Ridge National Lab. (June 1996).

3. Jordan, W. C. and Turner, J. C., "Minimum Mass of Moderator Required for Criticality of Homogenous Low-Enriched Uranium Systems," ORNL/CSD/TM-284, Oak Ridge National Lab. (Dec. 1992).

4. Mihalczo, J. T., "Use of ^{252}Cf as a Randomly Pulsed Neutron Source for Prompt Neutron Decay Measurements," *Nucl. Sci. Eng.*, **41**, 296 (1970).

5. Barber, E. J., "Estimation of Pressures Developed by retrained Solid UF_6," K/ET-307, Union Carbide Corp., Nucl. Div., Oak Ridge Gaseous Diffusion Plant, 1979.

6. National Nuclear Data Center at Brookhaven National Laboratory, Upton, N.Y., http://www.nndc.bnl.gov, maintained by Thomas W. Burrows (nndcth@bnl.gov).