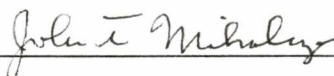


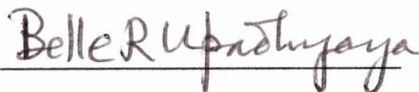
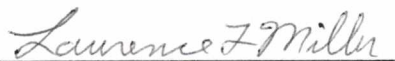
To the Graduate Council:

I am submitting herewith a thesis written by Mark Stephen Wyatt entitled "Characterization of an Enriched Uranyl Fluoride Deposit at the Former K-25 Enrichment Plant Using Active Neutron and Gamma Interrogation Techniques." I have examined the final copy of this thesis for form and content and recommend that it be accepted in partial fulfillment of the requirements for the degree of Master of Science, with a major in Nuclear Engineering.



John T. Mihalcz, Major Professor

We have read this dissertation
and recommend its acceptance:



Accepted for the Council:



Associate Vice Chancellor and
Dean of The Graduate School

**CHARACTERIZATION OF AN ENRICHED URANYL FLUORIDE DEPOSIT
AT THE FORMER K-25 ENRICHMENT PLANT USING ACTIVE NEUTRON
AND GAMMA INTERROGATION TECHNIQUES**

A Thesis
Presented for the
Master of Science
Degree
The University of Tennessee, Knoxville

Mark Stephen Wyatt
December 1997

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ABSTRACT

A method has been developed and successfully applied to measure uranium deposits in process gas piping at a former uranium enrichment plant. No *apriori* information other than material composition was required in order to accurately determine the deposit distribution or density. The density of uranium is directly related to its hydration which has significant nuclear criticality safety impacts for large deposits.

A ^{252}Cf source was used that provides two independent and simultaneous measurements in order to characterize the deposit. The first measurement was a transmission measurement using high energy fission neutrons, and can be shown to be independent of the deposit hydration and insensitive to geometrical effects of the deposit location with respect to the source and detectors. The second measurement was a gamma ray transmission measurement whose characteristics are a function of the deposit thickness and hydration. Using the deposit thickness from the neutron measurement, the second measurement was then used to determine the deposit hydration.

Measurements used to determine the spatial profile of the deposit indicated a volume of $195,000\text{ cm}^3$, yielding a deposit mass of $978 \pm 37\text{ kg}$ of uranium based on an average H/U of 3.4 ± 0.25 , where the uncertainty is the standard deviation of the mean. Measurements showed that the deposit had adhered to the top of the pipe in some locations contrary to expectation, and was later verified by an intrusive view into the pipe with a fiber optic camera. At the time of the intrusive look estimates of the H/U ratio based on the color of the deposit were 3.0 to 3.5, closely correlating the measured results.

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CHAPTER 1

INTRODUCTION

1.0 History of the K-25 Site

Construction of the K-25 Gaseous Diffusion Plant began in 1943, at the height of World War II, under the direction of the United States Army's Manhattan Engineer District. The Manhattan Engineer District, renamed the Manhattan Project after the war, was an enormous project directed at developing the materials and the technology to build an atomic bomb. After its completion in 1945, the K-25 site consisted of nearly 4000 diffusion stages used to isolate fissionable ^{235}U from its more abundant isotope ^{238}U . With the diffusion stages housed in a building having a perimeter of nearly two miles, the K-25 site was then estimated to consume one-seventh of the total electrical production of the United States.¹

After the end of World War II, the K-25 site was used to enrich uranium to be used for fuel in commercial, government, and military reactors. Enrichment operations were shut-down in the mid 1980's due to excess enrichment capacity and because of a reduction in the world market share of enriched uranium sales due to competition by foreign and domestic producers. In 1996 the Department of Energy (DOE) decided to transfer the K-25 site to a consortium of private firms in order to reduce decontamination and decommissioning costs, while simultaneously re-industrializing the area. In return for their remediation efforts the companies will take ownership of 300,000 tons of nickel and other valuable assets. The estimated impact of this agreement is the creation of 500 new

jobs and savings of 600 million dollars to taxpayers.² In early 1997, the K-25 site was renamed the East Tennessee Technology Park (ETTP) to reflect the focus of reducing the federal government's presence and leasing of floor space to outside companies.

1.1 The Gaseous Diffusion Process

Since isotopes of the same element are chemically identical, the gaseous diffusion process of separating the valuable ^{235}U from the other uranium isotopes, primarily ^{238}U , had to be developed. Natural uranium ore has isotopes of ^{234}U (0.0055%), ^{235}U (0.720%), and ^{238}U (99.2745%) that are hexavalent and can be converted to the compound uranium hexafluoride (UF_6). UF_6 is unique due to its low triple point that occurs at 22 psia (151.58 kPa) and 147.3 F (64.1 C). If the temperature or pressure is lower than the triple point then only a solid or gas can exist. This allows UF_6 to be easily converted to a gas for processing and back to a solid for storage and transportation, with each state occurring at relatively low pressures and temperatures. Additionally, fluorine consists of a single stable isotope so the separation involves only the species $^{234}\text{UF}_6$, $^{235}\text{UF}_6$ and $^{238}\text{UF}_6$.

The gaseous diffusion process is based on the difference in effusivity of isotopes of UF_6 . Each diffusion stage consists of a motor and compressor used to move the gaseous UF_6 , and a converter which houses a barrier material containing millions of microscopic holes per square inch. If the UF_6 molecules are maintained at a constant temperature T , then they have a kinetic energy given by

$$kT = \frac{1}{2} M v^2 \quad (1.1)$$

where k is the Boltzman constant, M is the molecular mass and v is the speed of the molecule. The ratio of the speeds between the two species of UF_6 is then given by

$$\frac{v_{235}}{v_{238}} = \sqrt{\frac{M_{238}}{M_{235}}} = s, \quad (1.2)$$

where s is defined as the separation factor. For a given temperature the $^{235}\text{UF}_6$ molecules will travel slightly faster than those of $^{238}\text{UF}_6$, and encounter the barrier material more frequently. This translates into the diffusion of more $^{235}\text{UF}_6$ across the barrier than $^{238}\text{UF}_6$, so that the process gas on the other side of the barrier is slightly enriched in $^{235}\text{UF}_6$. The theoretical value of the separation factor given in Eq. 1.2 can be found from the molecular masses of $^{235}\text{UF}_6$ and $^{238}\text{UF}_6$. Since the two masses differ by less than one percent, the separation factor is a relatively low 1.004289. This number is further reduced due to downstream backpressure across the barrier and non-separable flow, and has been found in practice to be closer to 1.003.³ ^{234}U is neglected in this analysis, since it is enriched to only about 1% if the enrichment of ^{235}U is carried up to 93%.

Because the separation value is so low, the process must be completed thousands of times to obtain the desired enrichment of ^{235}U . A motor, compressor, and converter are often referred to as a *stage*; placing several stages in a series forms a *cell*, cells are then connected in series to form a unit or *cascade*. Material that becomes enriched with the ^{235}U isotope is known as *product* and material depleted in ^{235}U is defined as *tails*.

As shown in Figure 1.1, UF_6 gas enters at the bottom of the cell at the stage 1 compressor. The gas is forced into the stage 1 converter, where the gas crossing the barrier becomes slightly enriched ^{235}U , and then moves into the stage 2 compressor via

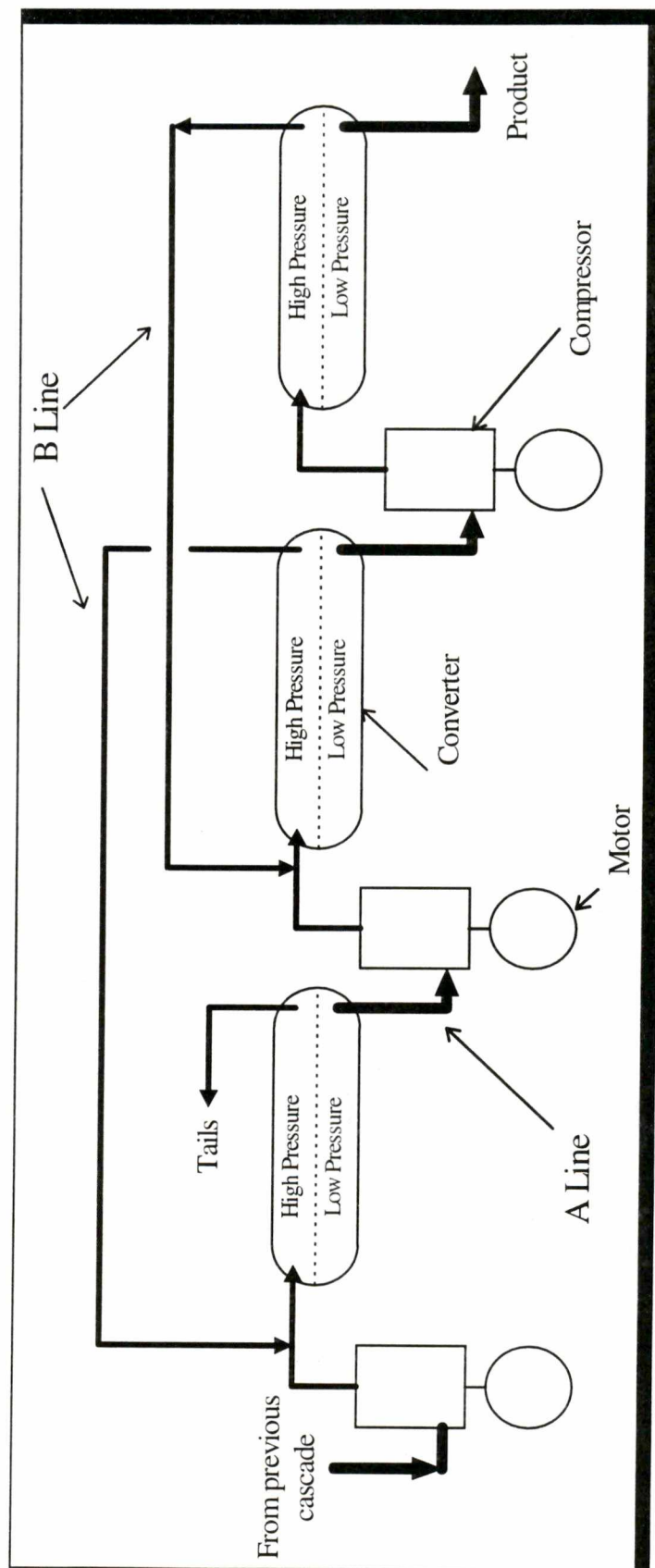
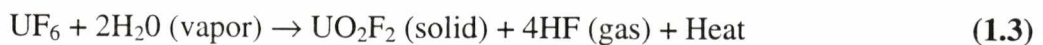


Figure 1.1 Gaseous Diffusion Cascade for the Enrichment of UF_6

the A-line or product line. This process continues through several more stages, enriching the gas stream at each location. Finally the gas stream leaves the last stage, where it continues on to the next cell. Another process is occurring on the high pressure side of the converter where gas that does not cross the barrier moves back down the cell via the B-line. This gas stream is depleted in ^{235}U , but at each stage any remaining $^{235}\text{UF}_6$ may cross the barrier. The tails continue to move down the cascade until they are eventually removed at some location and stored. The product continues to move up the cascade until the desired enrichment is reached, at which point it is also removed and stored.

In the solid state UF_6 is an almost white, dense crystalline solid. At atmospheric pressure (14.7 psia, 101 kPa) solid UF_6 sublimates at 56.6 C (133.8 F) to form a colorless gas. Normal gaseous diffusion plant process systems operate at around 140 °F, so occasionally the UF_6 gas will freeze out and create a localized deposit of solid UF_6 material. Gaseous and solid UF_6 do not react with oxygen, carbon dioxide, or dry air; but will react rapidly with water vapor as shown in Eq. 1.3. For this reason UF_6 is always handled in air tight containers and processing equipment. If UF_6 is exposed to water, even as trivial as in the form of elevated relative humidity, UO_2F_2 (uranyl fluoride) hydrates will be formed and a $\text{HF-H}_2\text{O}$ fog is produced which is visible as a white cloud.



UO_2F_2 is hygroscopic and readily absorbs moisture from the air to form hydrates until the stable compound $\text{UO}_2\text{F}_2 \cdot 2\text{H}_2\text{O}$ is reached.⁴ The ratio of hydrogen to uranium,

denoted H/U, can range from 0 to 4 while the UO_2F_2 still appears to be dry under visual inspection. After the stable compound $\text{UO}_2\text{F}_2 \cdot 2\text{H}_2\text{O}$ has been formed, the UO_2F_2 deposit can continue to absorb excess moisture in a volume-additive manner until a H/U of 16 is reached, at which point it begins to deliquesce. One indication of the amount of hydration present is given by the color of the deposit; at a very low H/U ratio the deposit is nearly white, as excess moisture is absorbed the deposit slowly changes color until it becomes a bright yellow at a H/U of 16.

1.2 Deposit Formation

In 1982, an event occurred at the Unit 2 Cell 7 Stage 6 Converter in the K-29 building. The investigation report of the 1982 event concluded that the Stage 6 compressor de-bladed, resulting in debris and extremely hot gases feeding into the Stage 6 converter. These hot gases initiated an exothermic reaction with the barrier material, forcing an emergency isolation and shutdown of the entire cell (Cell 7). Although the remainder of the ORGDP cascade continued to operate until 1985, Unit 2 Cell 7 in the K-29 building was never brought back into service and Cell 7 remained valved off from the rest of the unit by its four cell isolation block valves (B inlet, B outlet, A inlet, and A outlet).⁵

The Unit 2 Cell 7 B-outlet (7BB2) block valve is a 20-in.-dia. double disc gate valve used to isolate the Cell 7 Stage 1 B-stream from a 24 in. dia. B-bypass header that connects the various cells. The two are connected via a 24 in. dia. line that has a 20 in. horizontal run followed by a 60 degree elbow with a linear length of 38 in., and then

continues for 120 in. at a 60 degree angle until it meets the bypass line 13.5 feet above the floor. Figures 1.2 and 1.3 show the dimensions and physical attributes of this line. Due to its unique shape, the line connecting the block valve and B-bypass header has become to be known as the "Hockey Stick".

The 7BB2 block valve is of extreme importance because it is the location of, and perhaps the source of, the largest residual uranium deposit at the K-25 site. From March 1982 through August 1985, Unit 2 Cell 7 remained shutdown and isolated by four cell isolation block valves, while the rest of the enrichment cascade remained operational. The result was that an endless supply of UF_6 gas continued to rush past the upper end of the Cell 7 B-outlet line (the "Hockey Stick"), while the lower region of the line was isolated. The resultant stagnant region of the pipe is where the extremely large uranium deposit now exists. The exact cause of this deposit and time of deposition are unknown. The following description is how it is believed to be formed.⁵

Following the 1982 event in Cell 7, the cell was off line. Operators of the cascade noted perturbations in the cascade flows and pressures which led them to suspect a significant air in-leakage in the B-stream in the vicinity of Cell 7. Believing that the in-leakage must be coming from the damaged equipment in Cell 7, coupled with an incomplete closure of the B-outlet Block Valve (valve 7BB2), the Cell 7 B-outlet line was cut and a 1/2-inch metal plate was inserted into the line between the converter and 7BB2 block valve. This metal blank was welded in place on both sides, with the belief that this would positively block any further air in-leakage from the failed equipment in Cell 7.

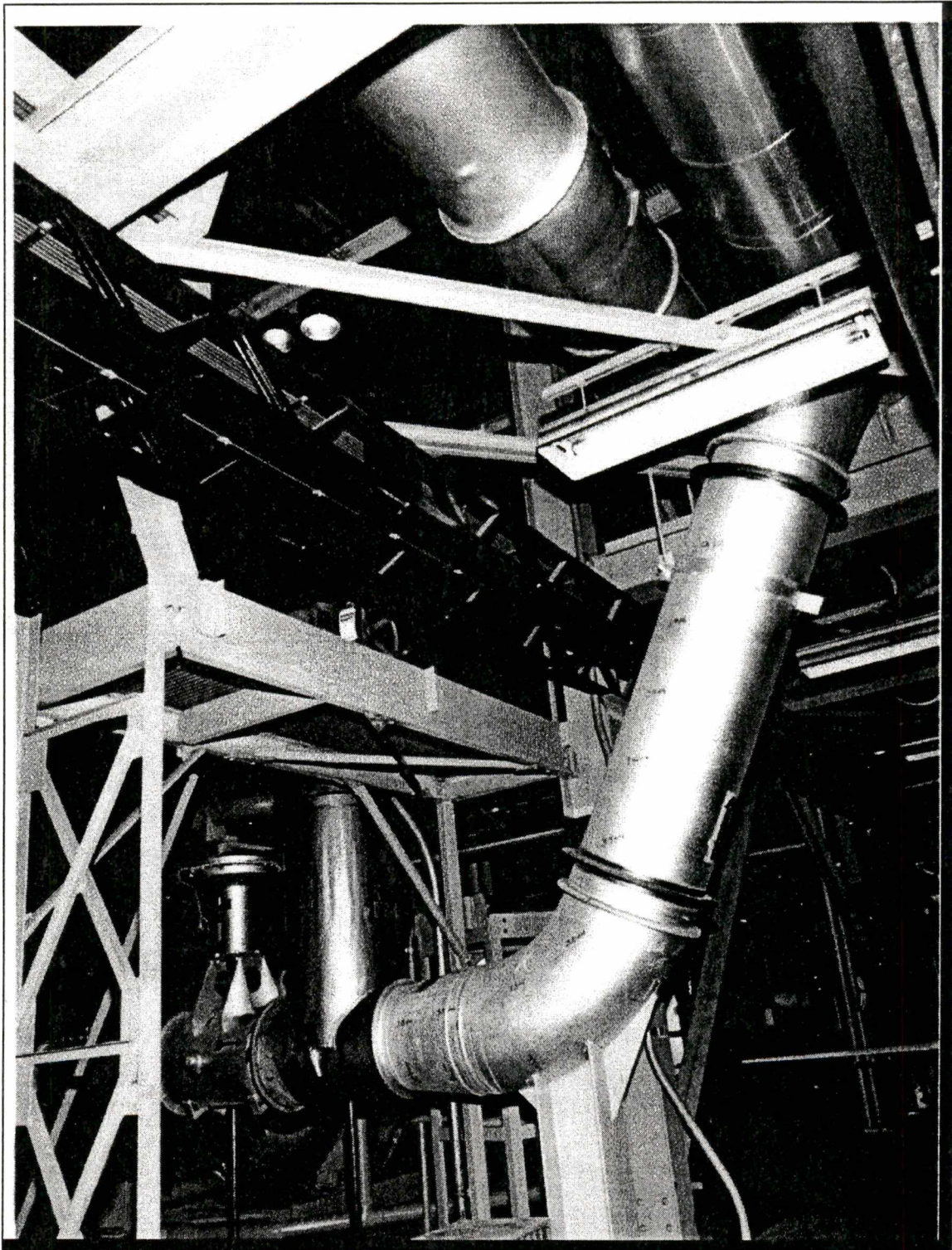


Figure 1.2 Bldg. K-29 Unit 2 Cell 7 Stage 1 Hockey Stick

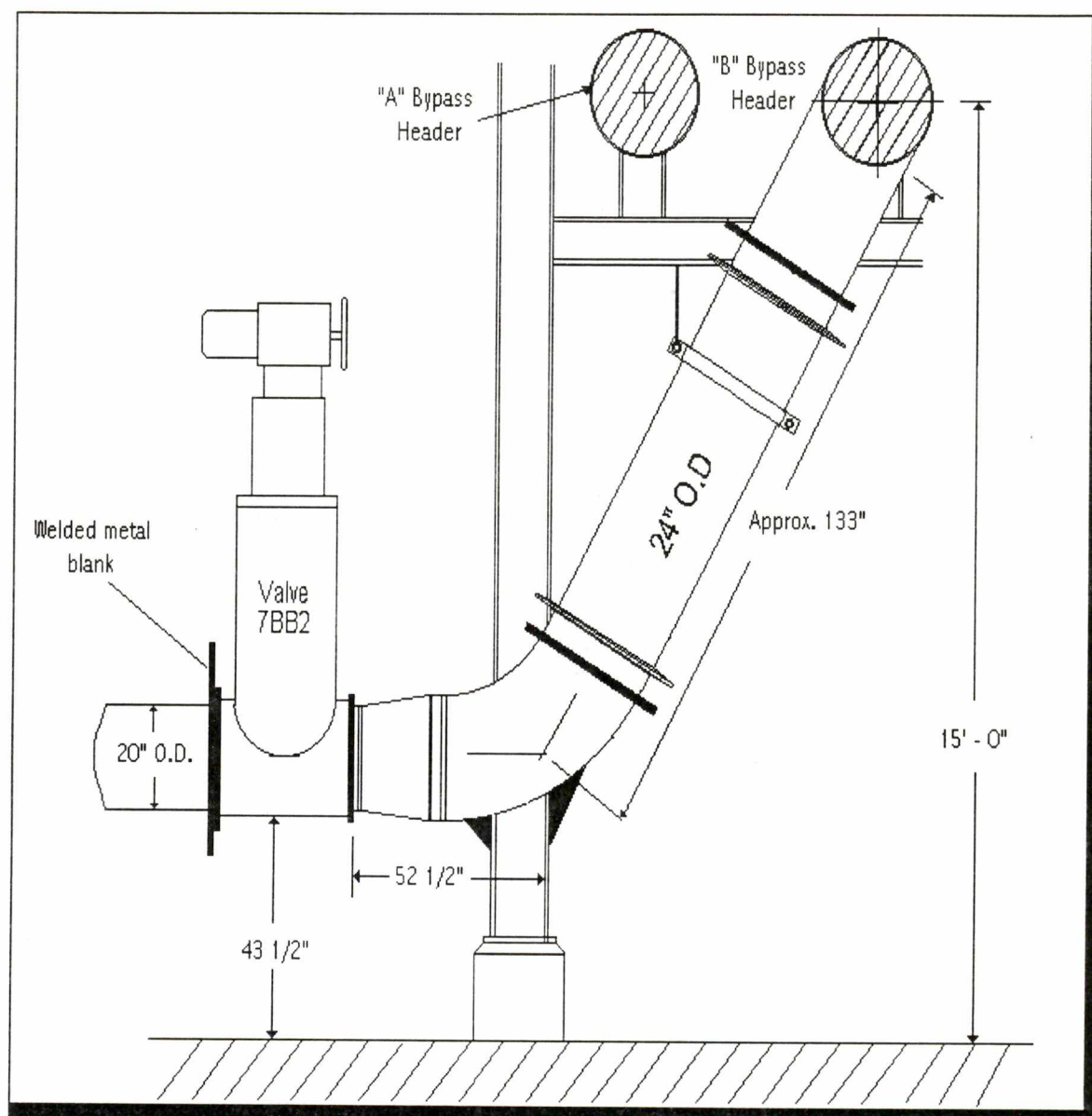


Figure 1.3 Dimensions of the Unit 2 Cell 7 Stage 1 (7BB2) Hockey Stick

The more likely reason for the observed in-leakage was probably a leak in the convoluted metal bellows or the evacuation line of the 7BB2 block valve. The possibility that a leak in the block valve was the source of the in-leakage is supported by the fact that the very large deposit begins at the valve body itself. There is no significant deposit upstream of the valve body. If the in-leakage was coming from the upper portion of the valve body it would explain why the valve body is reported to contain a significant amount of uranium, while no deposition exists anywhere upstream of the valve.

If the deposit at this location had already formed at the time of the 1982 event, it seems extremely unlikely that the deposit would not have been detected during the 1982 partial disassembly of Cell 7 during the extensive accident investigation. It is more believable that the deposit was somewhat smaller than its current size and was still growing as a result of continued air in-leakage. It seems logical that; (1) a leaking valve component was the source of in-leakage of humid ambient air, (2) that the operational B-stream header at the upper end of the pipe provided an endless supply of gaseous UF_6 , and (3) that the Hockey Stick pipe provided a stagnant zone for the mixing of gases and the deposition of the resultant UO_2F_2 . From 1982 to 1985, a deposit of extremely significant size formed, and is the largest such deposit identified at the site.

1.3 Passive NDA Estimates of Deposit Configuration

Non-Destructive Assay (NDA) measurements using gamma ray spectrometry performed in May of 1996 indicate that a deposit composed of 1190 kg of UO_2F_2 enriched to 3.3 wt% ^{235}U exists in the pipe. NDA uncertainties for the measured mass and

enrichment are 50% and 20% respectively.⁶ Gamma ray spectrometry indicates the deposit is uniformly distributed around the circumference of the pipe as well as along the length of the pipe. However, these measurements depend on the transmission of 186 keV gamma rays through a material of unknown density and thickness, which can mask significant changes of the material distribution inside the pipe. No information is known regarding the extent of hydration of the deposit. A deposit of this magnitude is a significant nuclear criticality safety concern. Under optimal conditions, the minimum critical mass is 102 kg for UO_2F_2 (3 %) and 55.5 kg for UO_2F_2 (4 %).⁷

The 60 degree incline and large diameter piping provide 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. Calculations using 3.3 wt% UO_2F_2 have indicated that a criticality could occur if the deposit were to occupy $\frac{1}{2}$ of the volume of the lower horizontal section of the Hockey Stick and if the H/U ratio is greater than 4. If the material were to occupy the full volume of the lower process piping the H/U ratio would only need to be 2 to cause a criticality event.⁶

1.4 Evaluation of Proposed Methods for Deposit Characterization

The Hockey Stick deposit in the K-29 building is a significant criticality safety concern. DOE and Lockheed Martin are committed to removing this deposit in order to minimize the personnel hazard that it poses, as well to allow the building to be eventually turned over for decontamination and decommissioning. Planning and performing the deposit removal is extremely difficult because very little information is known about the

characteristics of the material contained inside. A characterization of the distribution of fissile material within the pipe, its total mass, and amount of hydration was needed in order to help plan deposit removal activities. This information will allow planners to decide whether or not to implement mitigating actions such as dry buffer gas to prevent further hydration of the deposit, identify the best location to breach the piping so as to minimize disturbing the deposit, and evaluate all deposit removal work so as not to unduly expose personnel to the risk of a nuclear criticality.

Any characterization of the deposit must be non-intrusive for the reasons previously mentioned. Several techniques exist that could be applied to characterize the Hockey Stick deposit, with the best known and most widely used being passive Non-Destructive Assay (NDA).⁸ A common NDA technique is to analyze the spectrum of gamma rays emitted from a sample; identification of the gamma peak energies is then useful for identifying the radioactive material sampled. When applied to large or thick samples this technique in essence becomes a surface measurement due to self-absorption. This is particularly true when dealing with ^{235}U whose most energetic decay gamma is 186 KeV. A second NDA technique relies on measuring neutrons emitted from the sample, however this technique is difficult to apply due to the low spontaneous fission rate of uranium and would require very long counting times. NDA instrumentation is generally very portable and has no limiting physical constraints regarding its use, but is not particularly well suited for this application.

X-ray techniques can also be applied to assay the material inside the pipe and determine its distribution. By focusing a collimated beam of x-rays on one side of the

pipe and a detector on the other, the absorption spectra near the k-edge can be observed and correlated to the material composition. Sudden changes in the count rate can be used to make estimates of material edges as the beam and detector are scanned along the side of the pipe. X-rays are not as penetrating as high energy neutrons so this method could be limited in the case of thick deposits. Additionally this techniques requires a large x-ray generator and a high voltage AC power source in order to be deployed in the field.

The Associated Particle Imaging (API) technique may be used to determine the shape of the deposits in pipes and their chemical composition. API uses a Deuterium-Tritium (DT) generator to produce a beam of high energy neutrons. These neutrons have a fixed angular correlation of 180 degrees with the deuterium particle due to conservation of momentum; by tracking where the deuterium particle strikes the target the trajectory of the resulting neutron can be determined. Neutrons then scatter inelastically with the medium, producing secondary neutrons and gamma's. By correlating the gamma emissions to the origin of the neutron scattering event and using an array of gamma sensitive detectors, a three-dimensional image of the object can be constructed. By using detectors with a high energy resolution, the material composition can be determined from the scattering cross-sections and measured gamma energies by comparison with the appropriate calibration measurements. The gamma rays may be attenuated if the deposit is thick, so that no information about the material distribution or composition would be available. More significant drawbacks of the API method include a requirement for a 28.8 kV AC power source, a refrigerated semi-truck trailer, and the scarcity, complexity, and limited life of portable DT generators, as well as long measurement times.

The ^{252}Cf -source-driven noise analysis (CSDNA) method⁹ previously developed by Mihalczko and Pare has several advantages that were explored and applied to this problem. This measurement technique consists of a Californium neutron source in an ionization chamber, two neutron/gamma detectors and associated data acquisition equipment. The source ionization chamber is a parallel plate ionization detector that marks the time of spontaneous fission of the ^{252}Cf source. The source is generally located at the geometric center or adjacent to the fissile material. Neutrons and gamma's emitted from the source enter the fissile material where they either traverse the material with no interaction, are scattered within the material, or initiate fission chain multiplication. This gives rise to statistical variations of the neutron and neutron induced gamma ray population that is measured by detectors placed adjacent to the system. Classical signal processing algorithms are then employed to obtain 19 different time and frequency domain signatures which are sensitive to the fissile mass, geometry, and moderation of the assembly.¹⁰ The shape and amplitude of the time domain signatures, which are essentially a randomly pulsed neutron measurement, are directly related to the amount of material between the source and detectors.

Recent technological breakthroughs have reduced the size of the CSDNA measurement equipment from its previous semi-truck trailer configuration to a portable system known as the Nuclear Weapons Identification System (NWIS) that fits on a laboratory cart. NWIS has been applied successfully to several other fissile systems, but has never been utilized for characterization of an unknown material. With its wide variety

of time and frequency domain signatures, fast on-board sampling and data processing, NWIS can characterize the Hockey Stick deposit.

NWIS was developed with the support of the Oak Ridge Y-12 plant for applications to identify highly enriched uranium in containers. This system was used at the K-29 building with the approval of the Y-12 plant.

1.5 Objective and Contributions

The purpose of this work is to characterize an enriched uranyl fluoride deposit that exists at a former uranium enrichment plant. The deposit was believed to consist of 1190 kg of UO_2F_2 enriched to 3.3 wt% ^{235}U based on gamma ray spectrometry. No viable information about the distribution of the deposit or its density is available. The density of the uranium deposit is directly related to its hydration which has significant nuclear criticality safety impacts for large deposits. The characterization entails determination of the deposit distribution or profile, the extent of deposit hydration, and the estimated total mass of the deposit.

The author was jointly responsible for setup and calibration of the NWIS equipment for this application and had primarily responsibility for all measurement activities at the location of the deposit (positioning and relocation of source and detectors, etc.). Because the deposit was located in a high contamination area the NWIS system was placed in a clean area approximately 400 ft. away and operated by a 2nd person. Additionally, the author was primarily responsible for the data analysis and interpretation,

with the advice of a number of colleagues. It should be noted that this project was still a team effort, requiring the coordination and expertise of a number of people.

1.6 Outline

Chapter 1 provides introductory material regarding the residual uranium deposit of interest and a description of various non-intrusive internal characterization techniques. In Chapter 2 the physical and chemical properties of uranyl fluoride are described and a more detailed analysis of the nuclear criticality safety limits and concerns for the deposit are given. Chapter 3 provides the theoretical framework and assumptions used to assess the deposit and the Monte Carlo modeling and calculated signatures used to verify the adequacy of the assumptions. Chapter 4 presents an overview of the measurement techniques and methods, describing the nuclear instrumentation and detectors used for the measurements, calibration efforts, and the source-detector positioning assembly used to scan the pipe. Chapter 5 presents the measurement results, deposit profiles, and calculated deposit characteristics such as mass and hydration. Chapter 6 summarizes and makes recommendations for future work.

CHAPTER 2

2. NUCLEAR CRITICALITY SAFETY CONCERNS

2.0 Physical and Chemical Properties of UO_2F_2

Uranyl Fluoride (UO_2F_2) is formed from the reaction of UF_6 , present as either a solid or gas, with water vapor. The UO_2F_2 produced is a salt, and at ambient conditions it can absorb water from the air to form hydrates until the stable compound $\text{UO}_2\text{F}_2 \cdot 2\text{H}_2\text{O}$ is reached. The ratio of H/U for this compound is 4. $\text{UO}_2\text{F}_2 \cdot 2\text{H}_2\text{O}$ is hygroscopic so that when excess moisture is present it is absorbed in a volume-additive manner up to a H/U of 16, at which point a solution begins to form. The density of UO_2F_2 changes with the absorption of water. A discontinuity in the density exists at a H/U of 4 due to the change from water addition by the formation of hydrates to that of a volume-additive addition.

E. J. Barber¹¹ suggested that the uranium density in water moderated mixtures of uranyl fluoride be estimated using Eqs. 2.1 and 2.2,

$$\rho_u = 4.96 - 0.32\left(\frac{H}{U}\right), \quad \text{H/U} \leq 4 \quad (2.1)$$

$$\rho_u = \frac{m_u}{V_{\text{UO}_2\text{F}_2 \cdot 2\text{H}_2\text{O}} + \left(\frac{H}{U} - 4\right) \frac{V_{\text{H}_2\text{O}}}{2}}, \quad \text{H/U} > 4 \quad (2.2)$$

where V is the molar volume of the compound and m is the molecular weight of uranium at the given enrichment.

Figure 2.1 is a plot of the UO_2F_2 density⁷ over the range of H/U from 0.152 to 15.2, indicating the material density changes significantly by about a factor of 4 as it becomes fully hydrated. The color of the deposit is also indicative of the amount of the hydration; at a very low H/U ratio the deposit is nearly white, and as excess moisture is absorbed the deposit slowly changes color until it becomes bright yellow at a H/U of 16.

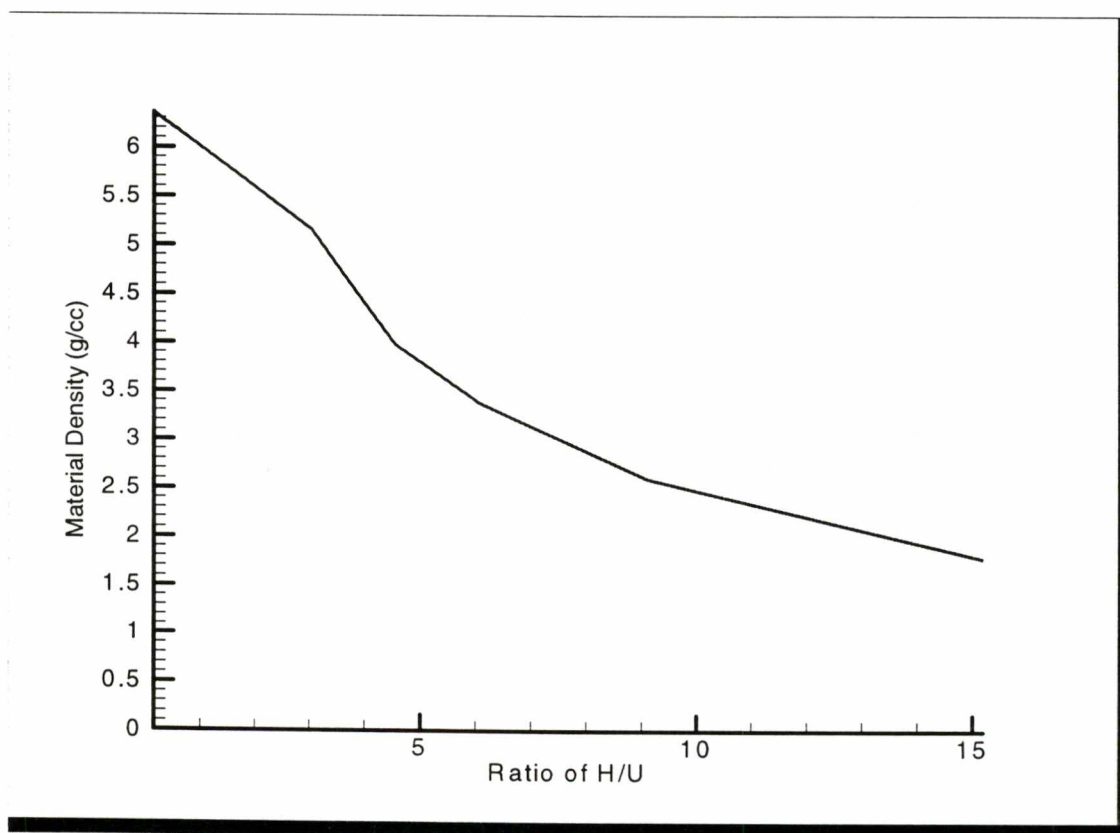


Figure 2.1 Material Density of Uranyl Fluoride as a Function of Hydration

2.1 Deposit Configuration From Gamma Ray Spectrometry

Gamma ray spectrometry measurements indicate that the deposit is composed of $1190 \text{ kg} \pm 595 \text{ kg}$ at an enrichment of $3.3 \text{ wt}\% \pm 0.66\%$ ^{235}U . Previous NDA measurements yielded a nominal assay of 2.8%. During the last four years of operation this section of the cascade operated at a maximum enrichment of 3.59%; therefore the depleted material in the tails line should be less than this value. The estimated enrichment based on the operating history and deposit location. is 3%.

By making passive gamma ray measurements around the circumference of the pipe and along the length of the pipe, NDA measurements suggested that the deposit is annular in shape and uniformly distributed along the length of the pipe. Assuming that the deposit exists at the nominal value of 1190 kg along the 186 in. active length of the pipe the deposit loading is calculated to be 81.6 kg/ft. Applying the 50% uncertainty of the NDA estimated mass in a conservative manner gives a deposit loading of 122 kg/ft.

2.2 Nuclear Criticality Safety Limits

Nuclear criticality safety standards¹² require that two independent parameters must change before a criticality can occur. In other words, the perturbation of any one parameter, no matter how unlikely, cannot lead to a criticality. Four independent parameters are present in the Hockey Stick deposit; (1) enrichment, (2) deposit geometry, (3) mass, and (4) moderation (i.e., H/U ratio).

2.2.1 Monte Carlo Criticality Safety Calculations

Monte Carlo calculations were performed by the Chemical Technology Division at Oak Ridge National Laboratory for a variety of deposit thickness' and H/U ratios.⁶ In this Monte Carlo analysis only symmetric annular deposits were considered, ranging from 2 - 16 cm in thickness and a separate case where the deposit fills the pipe. The hydrogen content was perturbed over the range of H/U of 0.81 to 8.1. The results of this study are shown in Table 2.1 and Figure 2.2.

Table 2.1 Calculated k-effective as a Function of Hydration and Symmetric Annular Deposit Thickness⁶				
H/U	Deposit Annular Thickness (cm) (in.)		Deposit Loading (kg/ft)	k-effective
0.81	2	0.79	51.5	0.162
0.81	4	1.57	99.5	0.260
0.81	8	3.15	184.6	0.418
0.81	16	6.30	311.6	0.608
0.81	full	full	395.4	0.705
2	2	0.79	47.3	0.202
2	4	1.57	91.3	0.364
2	8	3.15	169.4	0.609
2	16	6.30	285.9	0.845
2	full	full	362.8	0.947
4	2	0.79	35.9	0.225
4	4	1.57	69.2	0.436
4	8	3.15	128.5	0.726
4	16	6.30	216.9	0.976
4	full	full	275.2	1.079
8.1	2	0.79	23.9	0.231
8.1	4	1.57	46.1	0.474
8.1	8	3.15	85.5	0.787
8.1	16	6.30	144.3	1.04
8.1	full	full	183.2	1.14

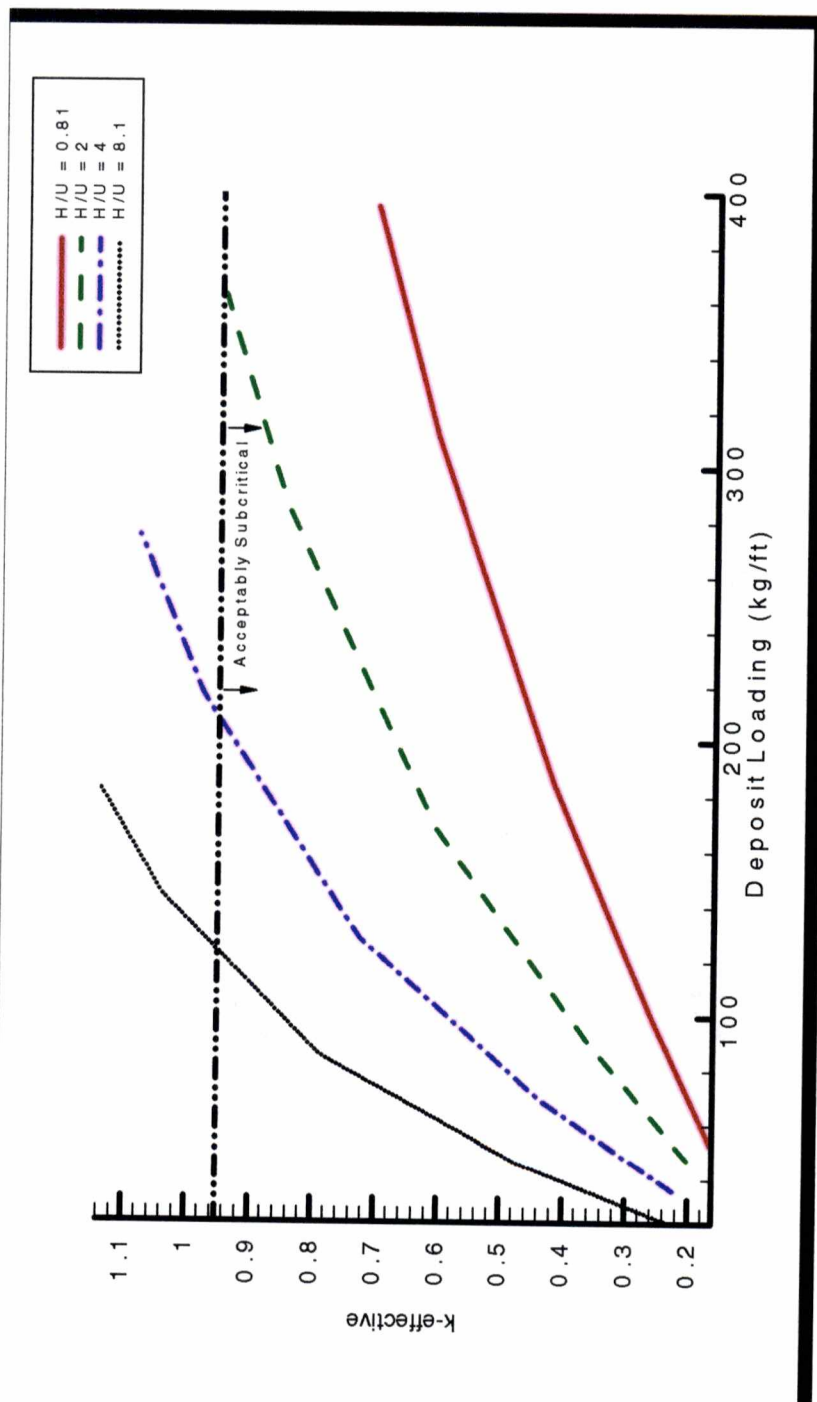


Figure 2.2 Calculated k-effective as a Function of Hydration and Annular Deposit Thickness ⁶

(The horizontal line at k-effective of 0.95 represents the acceptable subcritical margin for systems which are not well characterized. Estimated deposit loading from NDA is 70.7 kg/ft (+ 50%), and 47.1 kg/ft (- 50%).)

Calculations were performed using the SCALE Criticality Safety Analysis Sequence 1X module (XSNDRNPM-S) and ENDF-B/IV 27-group cross section library.¹³ Since the XSNDRNPM module is a one dimensional discrete ordinates code, some modeling simplifications were made for the geometry. The pipe was modeled as a 24-in.-O.D. cylinder with a length of 60 in.; a reflected boundary condition was applied at the ends of the pipe to achieve a infinite length. A P3 angular scattering expansion and a S-8 geometric quadrature was used for the mesh size. One inch of water reflection was applied outside the pipe to account for reflection from nearby structures and the floor.

From Figure 2.2 it is concluded that the deposit will remain subcritical if the hydrogen content is approximately less than a H/U of 1. If the deposit were collapse into the lower section, filling the pipe, a critical configuration can be reached if the H/U ratio were around 2. Note that a H/U ratio of 2 does not even correspond to a fully hydrated deposit. At a H/U ratio of 4, which corresponds to a fully hydrated deposit, a critical geometry is reached when the annular deposit thickness is 16 cm. This study did not consider chords, which could be formed from a deposit collapse where material collected in the lower section of the pipe. However, geometrically speaking a 16 cm annular shaped deposit is more conservative than a deposit formed as a 32 cm chord due to the increased material loading of the annulus over the chord.

2.2.2 HandBook Methods

A large amount of data from critical experiments is available and suitable for evaluation of the particular nuclear criticality safety hazards of the Hockey Stick deposit.

This data has been extensively peer reviewed, the experimental configurations repeated by numerous researchers, and is in fact the basis for quality assurance of the previously mentioned Monte Carlo codes. Analysis of the critical fissile mass, enrichment, and container shape and volume can be compared with known and estimated parameters of the system under study in order to prove or disprove double contingency.

Table 2.2 summarizes the known and estimated physical characteristics of the Hockey Stick deposit, and are the basis for comparison with the experimental data.

Table 2.2 Estimated Parameters of Hockey Stick Deposit for Comparison with Known Critical Dimensions		
Parameter	Value	Comments
Enrichment	2.64 - 3.96 wt%	NDA est. 3.3wt% $\pm$ 20%
Mass of Uranium	595 - 1785 kg	NDA est. 1190 kg $\pm$ 50%
Mass of ^{235}U	15.7 - 70.7 kg	595 at 2.64 %; 1785 at 3.96%
Geometry	23.25" I.D.	Infinite length (203")
Moderation (H/U ratio)	0.8 - 6	Estimate. Does not account for moderator addition by flooding.

2.2.2.1 Enrichment and Mass

Raffety and Mihalcz performed a series of clean critical experiments¹⁴ using homogenous mixtures of U(2)F_4 and U(3)F_4 dispersed in paraffin. The H/U ratio's ranged from 4 to 20 using the 2 wt.% material and 4 to 8.5 using the 3 wt.% material. The minimum critical mass of 2 wt.% reflected and unreflected spheres was 5.38 kg and 7.45 kg ^{235}U respectively, and occurred at an H/U ratio of 12.4. For the 3 wt.% material the minimum critical mass of the reflected and unreflected spheres was 3.83 kg and 6.40 kg

^{235}U respectively, and occurred at an H/U ratio of 8.4. There was not enough experimental data to determine the optimum moderation. From Table 2.2 there is a sufficient mass of ^{235}U (15.7 - 70.7 kg) estimated to be present in the pipe to achieve criticality at the given enrichment. Additionally, data from LA-10860-MS¹⁵ suggests that the limiting critical enrichment for homogeneous systems is about 1.03 wt.% ^{235}U and about 0.71 wt.% ^{235}U for heterogeneous systems. Based on the NDA parameters of Table 2.2, both the minimum critical mass and enrichment are exceeded for the Hockey Stick deposit in its present configuration.

2.2.2.2 Safe Geometry

The pipe containing the deposit is approximately 15.5 ft. long and as such can be considered an infinite cylinder. The minimum critical infinite cylinder diameter for 4 wt.% ^{235}U is approximately 9.8 in., compared to the 24 in. diameter Hockey Stick. Even for 3 wt.% material the minimum critical diameter changes to only 10.6 in. , indicating that the 24 in. O.D. hockey stick is not an acceptable means for preventing a criticality.

2.2.2.3 Moderation

Controls have been instituted to prevent the addition of moderator to the Hockey Stick although the present amount of actual moderation from interstitial hydrogen is unknown. UO_2F_2 is a salt and can absorb excess moisture from the air, increasing the moderation present. Generally, for these types of systems the risk of a criticality increases with increasing moderation. At an H/U of 4, corresponding to a condition of long term exposure of the deposit to ambient conditions, the minimum critical mass of ^{235}U is

approximately 8.5 kg. From Table 2.2, the minimum estimate of the ^{235}U mass in the Hockey Stick is 15.7 kg, implying that at least two critical masses are present. For most systems a minimum critical mass occurs at an H/X of 500, which for a 3 to 4 wt % deposit corresponds to a H/U of 15 to 20. Addition of a slight amount of external moderator would exceed the minimum critical mass of 2.2 kg. for the resulting condition.

2.3 Conclusions

The Hockey Stick deposit exceeds the minimum critical mass and is contained in an unsafe geometry. The water content of the deposit is unknown, but could conceivably exceed or equal an H/U of 4, again violating conditions for nuclear criticality safety. In its present condition, the double contingency principle for nuclear criticality safety is not met, and collapse of the deposit or the addition of a moderator could lead to a criticality excursion.

Chapter 3

Theoretical Development and Methodology

3.0 Introduction

The NWIS technology as used here is basically an active gamma ray and an active neutron radiograph to obtain an image of the deposit and its H/U ratio.

3.1 Transmission Measurements

For a beam incident on a thin slab having uniform composition and without geometrical anisotropy, the uncollided particle flux density emerging from opposite side is given by

$$I = I_o e^{-\alpha x} \quad (3.1)$$

where I_o is the incident particle flux density, x is the material thickness and α is the interaction probability per unit path length in the material.

For neutrons

$$\alpha = \sum_i \sigma_t^i(E) N^i, \quad (3.2)$$

where

σ_t^i = energy dependent microscopic cross section for the i^{th} nuclide (barns)

N^i = number density of the i^{th} nuclide (atoms/barn-cm),

and for gamma rays

$$\alpha = \sum_i \left(\frac{u(E)}{\rho} \right)_i \rho_i, \quad (3.3)$$

where

$[u(E)/\rho]_i$ = energy dependent mass attenuation coeff. of i^{th} nuclide (cm^2/g)

ρ_i = material density of the i^{th} nuclide (g/cm^3).

Two constraints apply when using Eq. 3.1. First, all particles that interact with the material must be effectively removed so that only the uncollided flux is present at the detector. Secondly, in-scattering is forbidden -- particles cannot scatter into the energy range of interest from higher energies. The first constraint can be satisfied by carefully constructing a measurement geometry as shown in Fig. 3.1. By providing sufficient separation between the source and detector, even particles that are scattered through small angles are guaranteed not to reach the detector. By carefully collimating the source and detector, only particles that are directly transmitted through the material without interaction are able to reach the detector.

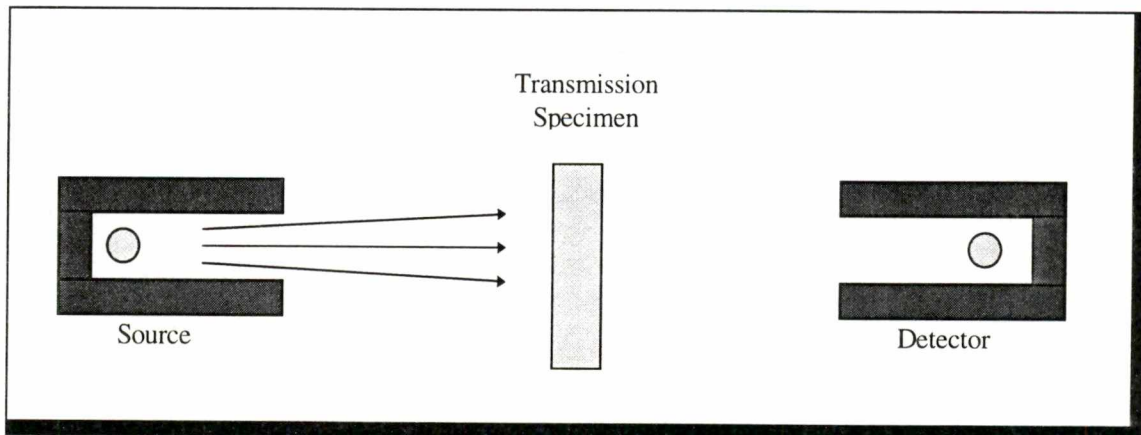


Figure 3.1 Idealized Geometry for Transmission Measurements

The second constraint can usually be satisfied by using a monoenergetic source. In the case where the incident particle flux density has an energy distribution, such as a fission source, the easiest way to satisfy the in-scattering constraint is to perform the measurement analysis with high energy neutrons. This assures that particles undergoing interactions will either be removed via scattering angle effects, or those that scatter into the detector will have an energy below the energy band under investigation.

Generally transmission measurements are conducted such that the thickness and material type are well characterized and known beforehand. Measurements may be performed to accurately determine one variable, such as the total cross-section or mass attenuation coefficient, given an idealized and known material sample of a given thickness. The uranyl fluoride deposit in the K-29 Hockey Stick deposit does not present an idealized measurement situation. Neither the exact composition, material density, or thickness at any location are known. The theoretical material composition may vary from a dry salt containing little or no hydrogen up to a damp deposit containing chemically bound and free hydrogen. Over this range of hydration, the material density can change by a factor of almost 4. No information is available regarding the deposit thickness or distribution with the exception that it is believed to be annular. Eq. 3.1 does not require that the exact incident or exiting flux be known, but only the ratio of the two. However, the remaining variables make unique solutions of Eq. 3.1 for the deposit thickness and density intractable without further information.

3.1.1 Neutron Interactions

The interaction cross-sections for neutrons with a material are only dependent on the incident neutron energy, the nuclide of interest, and its relative abundance. Neutrons, having a neutral charge, do not undergo Coulomb interactions and interact essentially only with the atomic nucleus. As UO_2F_2 hydrates the material density changes due to displacement of the dense uranium component by the relatively light hydrogen. Using Barber's Eq.'s 2.1 and 2.2, the constituencies of the elements as a function of H/U can be calculated and are shown in Figure 3.2.

ENDF/B-VI cross-section data¹⁶ was obtained from the National Nuclear Data Center (NNDC)¹⁷ at Brookhaven National Laboratory, evaluated at 300 K and averaged over energy intervals of 1/8 to 1/4 MeV at each energy point, depending on the cross-section sensitivity. Figure 3.3 shows the smoothed total neutron interaction cross-sections for hydrogen, oxygen, fluorine, and uranium. The non-uranium cross-sections decrease rapidly over the interval from 0.1 to 4 MeV, at which point they become almost constant with increasing energy. The ^{235}U and ^{238}U cross-sections behave similarly in the energy range of 0.1 to 10 MeV, decreasing rapidly for energies less than 1.5 MeV, and then becoming almost constant for neutron energies above 4 MeV. Note that this energy interval avoids the resonance structures for ^{235}U and ^{238}U which lie below 0.001 MeV. The uranium cross-sections dominate the hydrogen, oxygen, and fluorine cross-sections for neutron energies above 1 MeV, leading to a very important consequence when coupled to the changing material densities of the deposit with hydration.

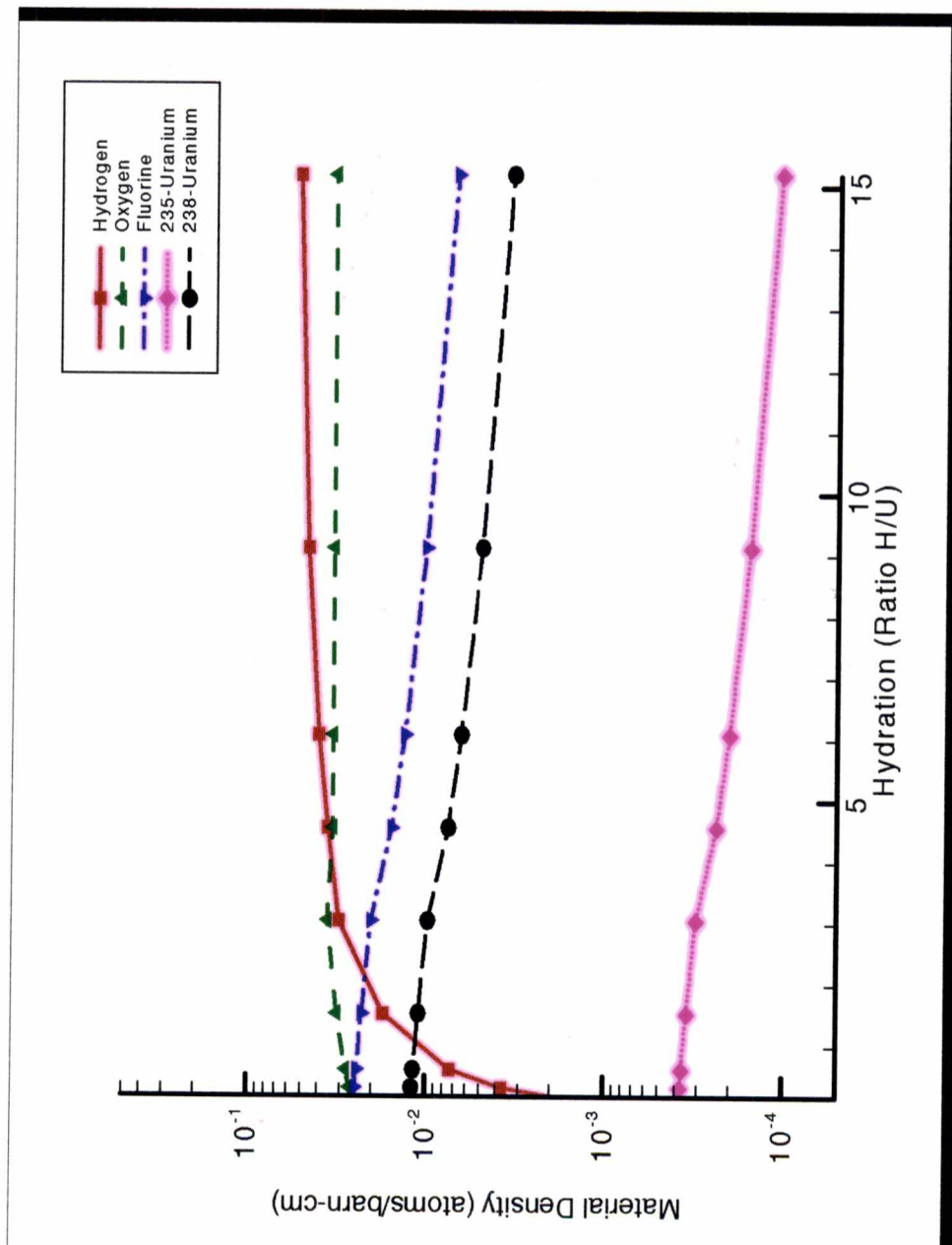


Figure 3.2 Number Densities of the Constituents of Uranyl Fluoride as a Function of Hydration

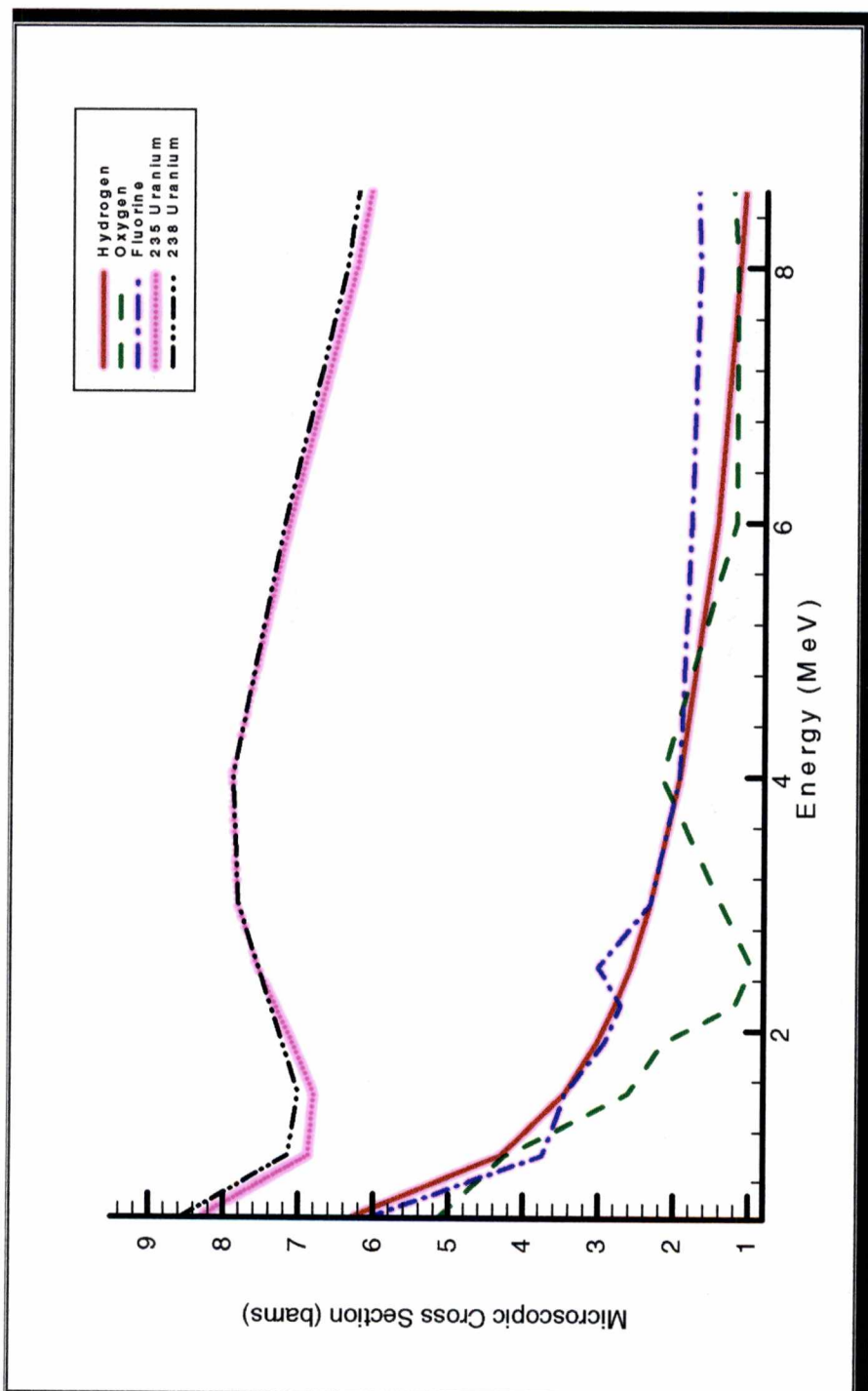


Figure 3.3 Microscopic Total Interaction Cross Sections of Uranyl Fluoride Constituents over the Energy Range of the ²⁵²Cf Neutron Fission Spectrum

Figure 3.4 shows the energy dependent total macroscopic cross-sections of a uranyl fluoride deposit over a range of assumed hydration's. At low energies the total macroscopic cross section is sensitive not only to changes in hydration, but to slight perturbations in the incident neutron energy, denoted by the large separation between the three cross-sections at low energy (0.5, 1.0, and 1.5 MeV). At high energies above 6 MeV the cross-sections are essentially flat with hydration, varying by less than 10 percent. Additionally, the cross-sections are only very weakly affected by slight perturbations of the incident neutron energy. In a measurement situation this becomes important since it demonstrates that timing uncertainties, which are related to the measured neutron energy, will not have a strong effect on the calculation of the material properties.

This effect immediately leads to a method for determining the thickness of a deposit using Eq 3.1, since we may assume that the total neutron cross-section is constant with hydration or energy for incident neutron energies greater than 6.0 MeV. Transmission measurements using high energy neutrons can be used to determine the deposit thickness, since the total macroscopic cross-section for UO_2F_2 at any density or hydration is essentially constant and can be calculated.

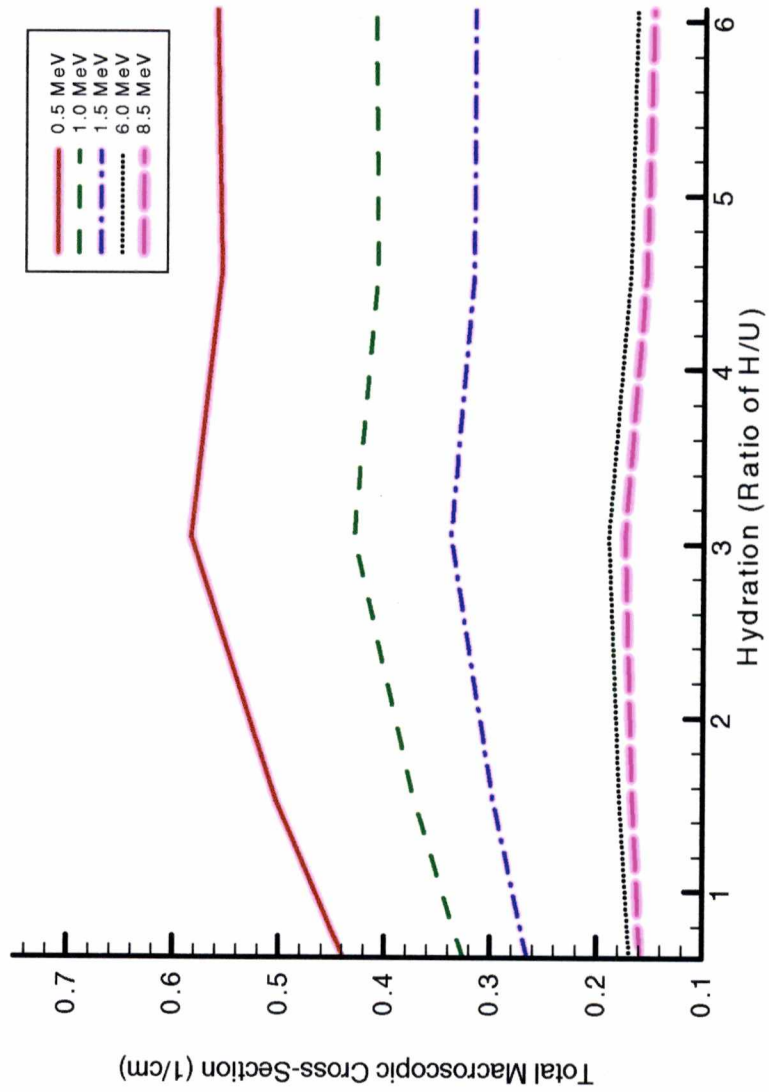


Figure 3.4 Total Macroscopic Neutron Cross-Sections at $E = 0.5, 1.0, 1.5, 6.0,$ and 8.5 MeV

3.1.2 Photon Interactions

Gamma rays interact with the atoms via mechanisms such as the photoelectric effect, Compton scattering, and pair production. In general, each of these mechanisms has a interaction probability that is dependent on both the incident gamma energy and the electron density of the absorbing medium.

The photoelectric effect is the dominant interaction for gamma energies less than 200 keV. The gamma interacts with an atom, ejecting a photoelectron electron from the k or l shell with a kinetic energy given by

$$E_e = h\nu - E_b \quad (3.4)$$

where

E_e = kinetic energy of photoelectron

E_b = binding energy of photoelectron

h = Planck's constant = 6.62×10^{-34} J-s

ν = frequency of incident gamma ray

The relative probability of the photoelectric effect per gram of absorber is governed by the empirically derived relationship¹⁸

$$\sigma_{ph}(E) \propto \frac{Z^m}{E^3}, \quad (3.5)$$

where Z is the atomic number and m varies from approximately 4 to 4.6 over the range of 100 keV to 3 MeV.

Between 200 keV and 5 MeV Compton scattering becomes the dominant method for gamma interaction. In the Compton scattering process the incident gamma scatters inelastically with an atomic electron, ejecting the electron from its orbit about the nucleus, and leaving the scattered gamma with energy $h\nu'$. Because all elements, with the exception of hydrogen, have approximately the same number of electrons per gram, the Compton scattering process, which occurs with an orbital electron, is independent of the atomic number.

The pair production process is characterized by the incident gamma disappearing in the vicinity of the nucleus, and the production of a electron-positron pair in its place. The minimum energy required for pair production is 1.02 MeV, but the process is usually not apparent until approximately 1.5 MeV, and only begins to dominate at gamma energies above 5 MeV. The pair production cross-section σ_{pp} varies approximately as Z^2 and increases asymptotically with energy.¹⁶

Two other interaction mechanisms bear mention, but are generally small enough to be negligible. Thomson scattering is the oscillation of a free electron perturbed by the electric field of the passing gamma ray. The electron then emits secondary gamma rays having a frequency the same as the incident wave. Raleigh scattering results from the combined, coherent action of the gamma ray on the electrons of an atom as a whole. For this reason, it is also referred to as coherent scattering. The effect of either of these mechanisms is generally negligible, since there is no appreciable loss of energy and a minimal redirection of the incident gamma.

The total interaction probability or cross-section is a linear combination of the cross-sections for each interaction type and therefore has a Z -dependence that varies roughly as

$$\sigma_t(E) = \frac{Z^4}{E^3} + Z^2. \quad (3.6)$$

As the uranyl fluoride hydrates from the addition of hydrogen, a low Z material, the high Z component of uranium is diluted and displaced. Equation 3.6 indicates that the gamma response of the detector will be extremely sensitive to slight changes in hydration due to the dependence of the total cross-section on 2nd and 4th order terms.

3.2 CSDNA and Pulsed Neutron Measurement Techniques

The ^{252}Cf -source-driven noise analysis (CSDNA) method evolved from Rossi- α ¹⁹ and pulsed neutron measurements.²⁰ The Rossi- α measurement relates the time distribution of counts in a detector after an initiating count occurs in the same or another detector. Rossi- α measurements typically rely on the inherent neutron source strength of the material or the use of an external neutron source. The pulsed neutron measurement utilizes an external neutron source that injects a pulse of neutrons into the assembly, detectors in or near the assembly then measure the time distribution of counts arriving after the initial pulse.

The CSDNA technique utilizes ^{252}Cf in a small parallel-plate ionization chamber (detector 1) that serves as a timed source of spontaneous fission neutrons. Neutrons and gamma's emitted from the ^{252}Cf source enter the fissile array where they either traverse

the material with no interaction (transmission), are scattered within the material, or initiate the fission chain multiplication process. One or more detectors are then used to measure the time distribution of counts occurring after the initiating event at the source. Figure 3.5 shows a schematic of the source and detectors for a measurement. Essentially a neutron time-of-flight (TOF) measurement through the pipe, the energy of the neutron is determined from the measured time it takes to travel the known distance between the source and the detector using the non-relativistic kinetic energy equation. 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 source and detector.

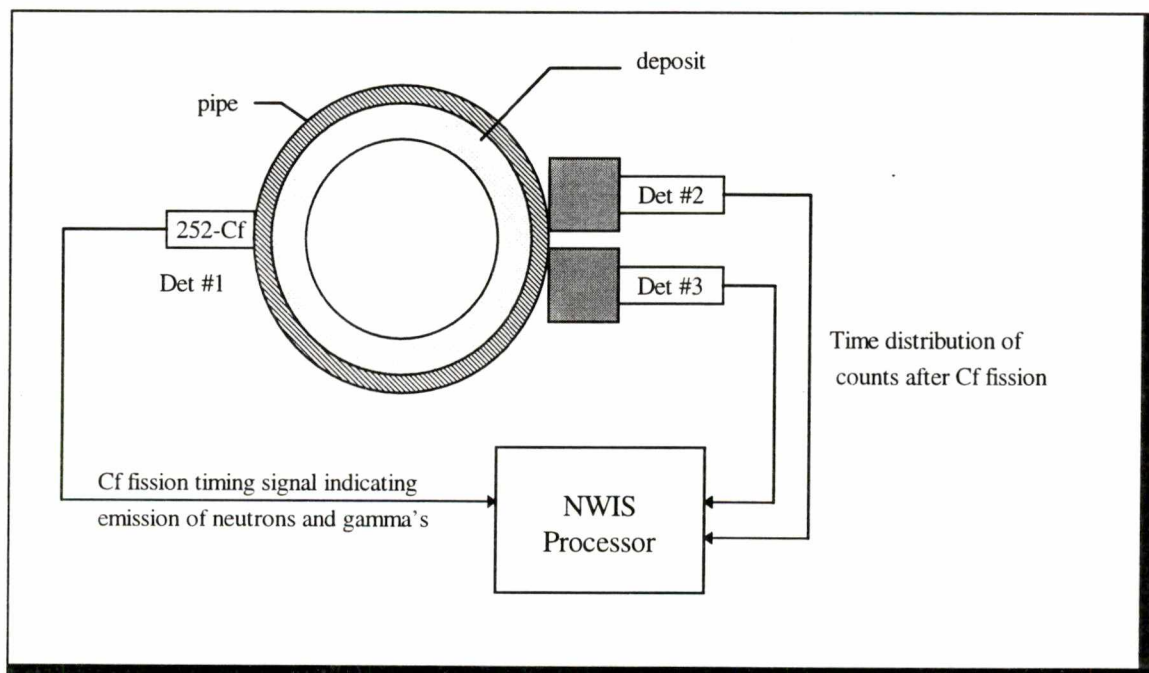


Figure 3.5 Schematic of Measurement Configuration at K-29

Since the counts in the detectors (2 and 3) are correlated in time to the source ionization chamber (detector 1), gamma rays that are scattered or produced from nuclear reactions arrive later in time than those that are directly transmitted. In essence this provides a collimated source of gamma rays that can be used to measure gamma ray attenuation coefficients without having to heavily shield the detector, i.e., collimation by timing rather than lead. The method can also be applied in high background radiation areas, since any background radiation is uncorrelated in time with the source. This time correlated signature is equivalent to the right half of the cross correlation function between the ^{252}Cf source and the detector

$$R_{xy}(\tau) = \lim_{T \rightarrow \infty} \frac{1}{T} \int_0^T x(t)y(t+\tau)dt, \quad (3.7)$$

and is a measure of the correlated information in two signals, one signal at time t and the other signal at time $t + \tau$. A typical right half of the cross-correlation function between the ^{252}Cf fission source with a single detector is shown in Figure 3.6. The figure has been arbitrarily adjusted on the time axis so that the gamma peak is placed at 25 nsec for clarity of the figure and labeling.

One of the primary advantages of ^{252}Cf is its availability and high specific neutron activity ($2.3 \text{ E}+012 \text{ n/s/g}$), as well as its simultaneous gamma emissions which can be used as a second independent measurement technique utilizing information from the gamma response of the detector. ^{252}Cf has a spontaneous fission half life of 87.5 years, and an effective half life of 2.64 years, which is governed by its α -decay rate.²¹ Details of the ^{252}Cf fission spectrum are given in Appendix A.

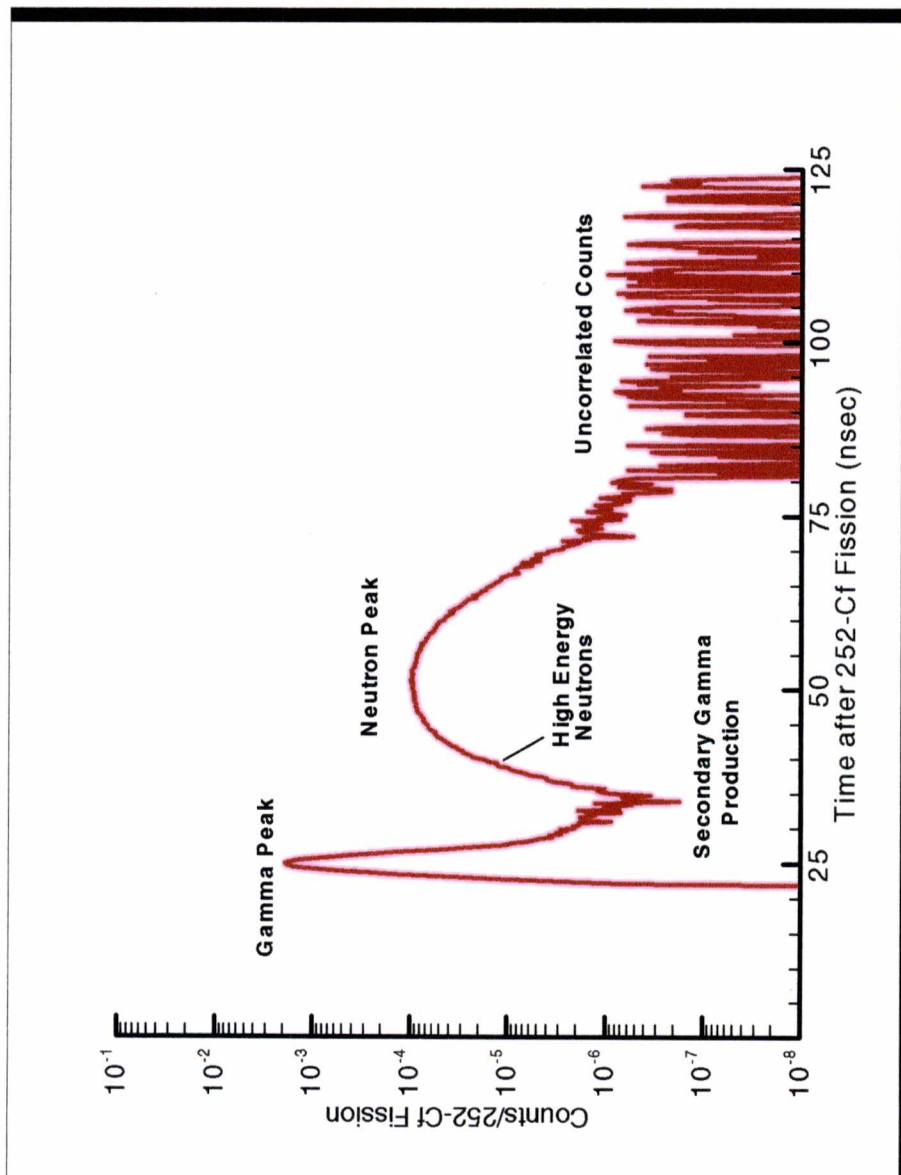


Figure 3.6 Measured Cross-Correlation Between ^{252}Cf Source and Detector

3.3 Applying NWIS Signatures to Deposit Measurements

The thickness of the deposit is determined from the neutron signatures by assuming that the high energy neutron cross-sections are essentially flat with hydration and insensitive to slight perturbations in the incident neutron energy (Fig. 3.4). Additionally, because it is necessary to limit particle in-scattering from higher energies into the energy range being measured, it is necessary to use the highest incident neutron energy available, approximately 8.5 MeV. Once given the deposit thickness, the gamma signatures can be used to determine the deposit hydration. However, several additional steps are required in order to have a meaningful interpretation of the data.

3.3.1 Removal of Pipe Contribution from Measurement

Calculation of the deposit thickness and hydration using Eq. 3.1 requires two measurements (I and I_o), one containing the material being measured (I) and a second reference measurement with the source and detector in air (I_o). The first measurement consists of a contribution from the deposit and the pipe that contains the deposit, adding additional variables to the analysis, as shown in Eq. 3.8

$$I^p = I^{air} e^{-(\Sigma_p X + \Sigma_d S)} \quad (3.8)$$

where

Σ_p = macroscopic cross-section of pipe

X = pipe thickness

Σ_d = macroscopic cross-section of deposit

S = deposit thickness.

By making a reference measurement with an empty pipe containing no deposit and dividing the two measurements as shown in Eq. 3.9

$$\frac{I^p}{I^{ref}} = \frac{I^{air} e^{-\Sigma_p X} e^{-\Sigma_d S}}{I^{air} e^{-\Sigma_p X}} = e^{-\Sigma_d S}, \quad (3.9)$$

it is possible to interpret the data so that the results only depend on the thickness and hydration of the deposit.

3.3.2 Deposit Hydration

Because all the transmission gamma rays arrive at the detector at the same time regardless of energy, an energy spectrum-weighted approach must be taken to determine the deposit hydration. This is required because the mass attenuation coefficient given in Eq. 3.3 is a function of incident ^{252}Cf gamma ray spectrum, detailed in Appendix B, as well as deposit composition. Again relating the deposit measurement and the reference measurement as in the previous section

$$\frac{A_\gamma}{A_{ref}} = \frac{\int_0^\infty dE G_o(E) e^{-\mu(E)X} e^{-\sum_i \left(\frac{\mu(E)}{\rho} \right)_i \rho_i S}}{\int_0^\infty dE G_o(E) e^{-\mu(E)X}}, \quad (3.10)$$

where

A_γ = detector response due to pipe and deposit

A_{ref} = detector response to empty pipe

$G_o(E)$ = ^{252}Cf gamma ray emission spectrum

S = deposit thickness

X = pipe thickness.

Eq. 3.10 is a Volterra equation of the first kind. 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 pipe are well known, it is possible to solve this equation numerically.

Energy dependent mass attenuation coefficients from NNDC 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 for different hydrations and material thicknesses. Figure 3.7 shows the results of the integration and evaluation of Eq. 3.10 for a variety of H/U ratios and material thicknesses.

For thin deposits approximately 2 cm or less, the ratio of the gamma peaks are closely grouped and the slope of the curves are quite steep, as shown in Fig. 3.7. This indicates that estimation of the H/U ratio should be obtained from data in locations where the deposit is sufficiently thick. For a thin deposit, an error of 0.5 cm can correspond to 3 different values of H/U. Similarly, a 10 percent error in the ratio of the two gamma ray transmissions can also correspond to 3 different values of H/U.

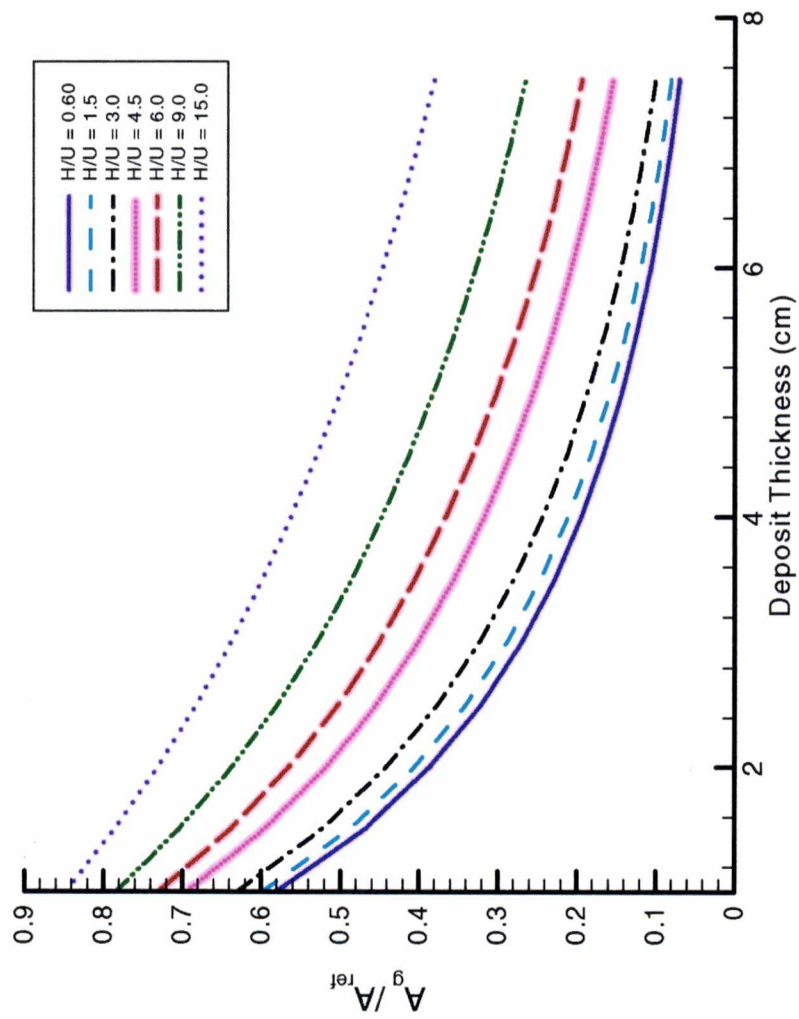


Figure 3.7 Calculated Ratio's of the Gamma Response as a Function of Deposit Thickness and Hydration

A least-squares polynomial fit, described in Appendix C, was performed to functionalize the results of each integrand of Eq. 3.10. The deposit density or H/U ratio is then determined by finding

$$\left(\frac{A_g}{A_{ref}} \right)_{meas} - \left(\frac{A_g}{A_{ref}}(\rho, S) \right)_{calc} = 0. \quad (3.11)$$

The deposit thickness S is substituted into the functional expression for each H/U until Eq. 3.11 is satisfied.

3.3.3 Refinement of Deposit Thickness and Hydration

The measurement results can be further improved by making a number of iterative calculations as shown in Fig. 3.8. The process begins by assuming a constant total macroscopic cross-section for neutrons (Fig. 3.4), which in fact varies by about fifteen percent over the range of assumed H/U. Once the thickness is calculated, the hydration is determined from the gamma transmission portion of the signature and Eq.s 3.10 and 3.11. This hydration is then used to refine the estimate of the neutron cross-sections and obtain a new deposit 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 iterative process continues until the values of the deposit thickness or hydration converges.

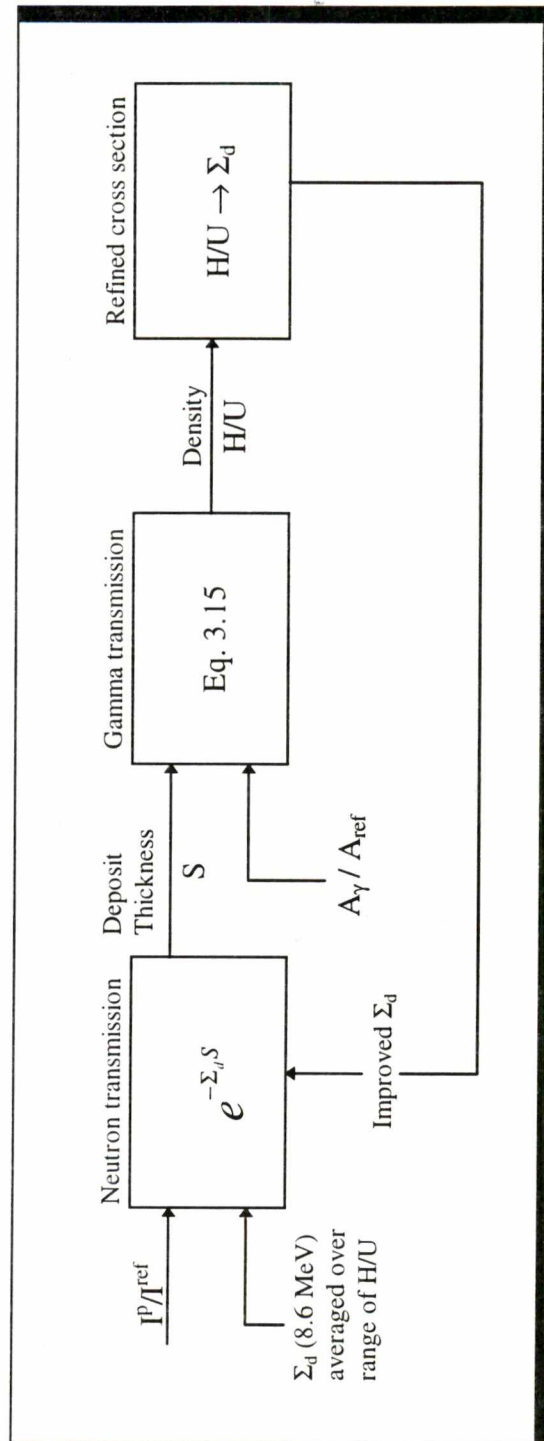


Figure 3.8 Iterative Refinement of the Deposit Thickness and Hydration

3.3.4 Geometry and Inscattering Effects

MCNP-DSP calculations were used to verify the assumptions that measurements of the deposit thickness from high energy neutrons is not sensitive to geometry or inscattering effects. MCNP-DSP is a continuous energy Monte Carlo code capable of tracking both neutrons and photons in a manner that simulates the CSDNA measurement.^{22 23} MCNP-DSP (a modification of at Los Alamos National Laboratory's MCNP 4A²⁴) models the stochastic nature of particle interactions in a medium in which space, angle, and energy are continuous variables. MCNP-DSP replaces average values like $\bar{\nu}$, the average number of prompt neutrons produced per fission, with a probability distribution in order to achieve the statistical fluctuations found in nature. Additional modifications include a dual-particle source that produces neutrons and photons having multiplicities and energy distributions of ²⁵²Cf. One of the most important modifications for the Hockey Stick characterization is the proper treatment of scattering detectors which is not obtainable using standard MCNP. MCNP-DSP has been widely benchmarked^{25, 26} and shown to be an excellent tool for planning experiments in criticality safety, safeguards, nondestructive assay, and arms control verification.²⁷

Figure 3.9 shows two potential measurement configurations for a postulated distribution of material in the Hockey Stick. Because the deposit distribution is unknown, it must be determined whether the measurement results will depend on the position of the source and detector with respect to the deposit. These distributions were generalized to three separate MCNP-DSP calculations, shown in Figure 3.10, in order to quantify

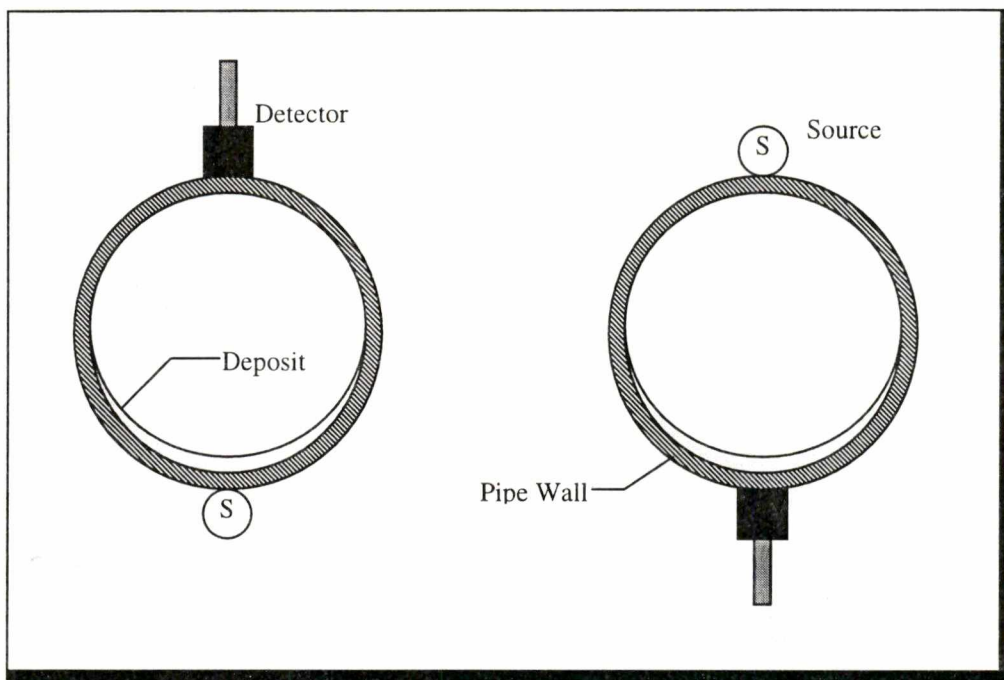


Figure 3.9 Potential Measurement Configurations for a Postulated Material Distribution in the Hockey Stick

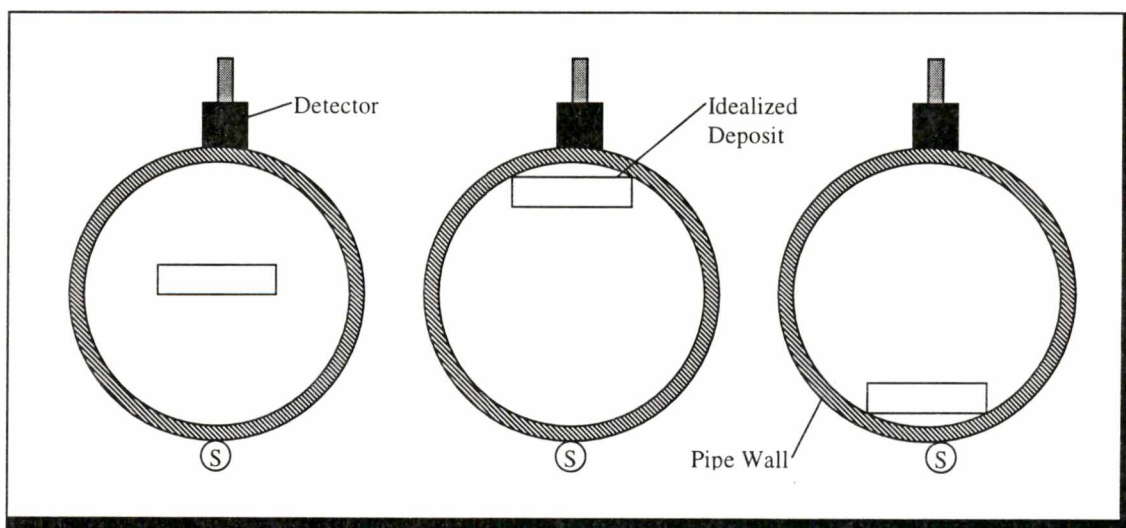


Figure 3.10 Cross-Sectional View of MCNP-DSP Models to Investigate Geometric Effects of Source-Detector Location with Respect to Deposit

potential sensitivities to geometry effects. All three calculations assumed a constant pipe wall thickness and separation between source and detector, as well as a constant deposit thickness. The only parameter modified in the calculation is the placement of the deposit, either close to the source, close to the detector, or splitting the distance between the two.

Figure 3.11 shows the MCNP-DSP calculated time distribution of counts in the detector after the spontaneous fission of ^{252}Cf for each geometry. Note that the data has been arbitrarily transformed on the time axis so that the gamma peak is placed at 25 nsec for clarity of the figure. Based on a separation distance of 24 inches between the source and detector, the rising portion of the neutron response between 38 and 42 nsec corresponds to the arrival of neutrons 13 to 17 nsec after the source fission.

This arrival time corresponds to neutron energies between 6.7 and 11.5 MeV with the midpoint of 15 nsec corresponding to neutron energies of 8.5 MeV. The portion of the neutron response is essentially invariant to the deposit position, indicating that the assumptions are correct and the measured data can be interpreted without having to absolutely determine the deposit location with respect to the source or detector. Conversely, a series of measurements at a single location should be able to adequately determine the deposit thickness without having to account for geometric effects.

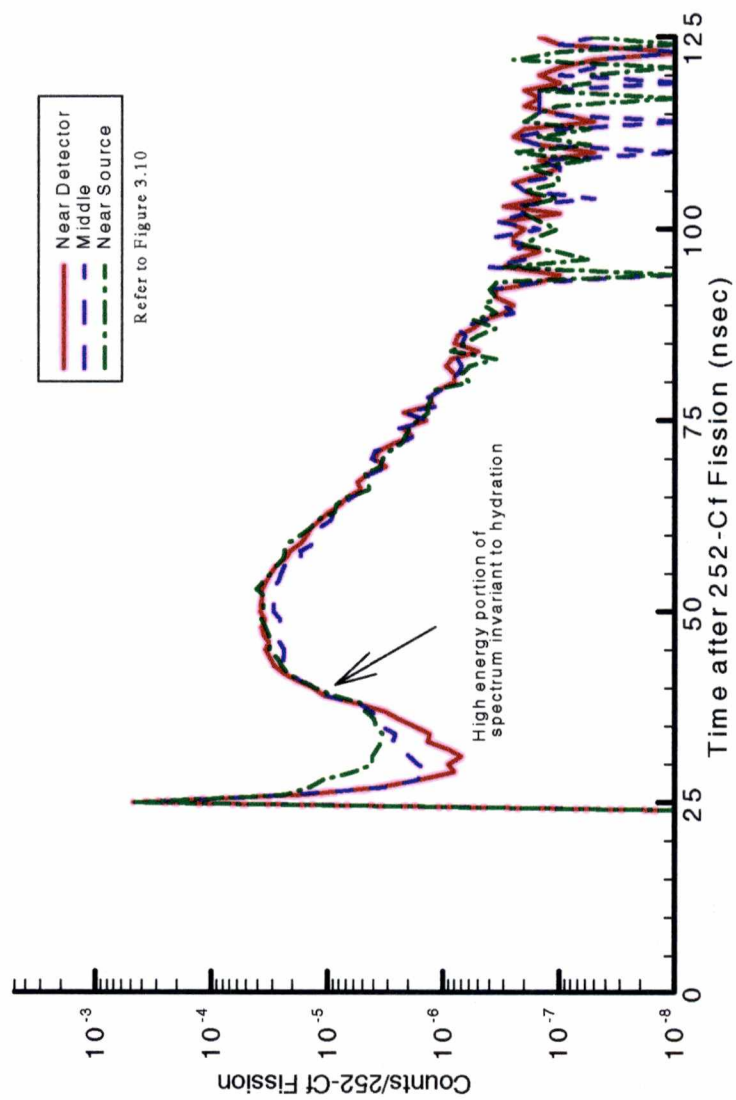


Figure 3.11 Calculated Signatures of Three Deposit Locations with Respect to Source and Detector, Indicating that the Rising Portion of the Neutron Spectrum is Invariant to Position

One subtle effect, although not obvious in Figure 3.11, is predicted from the perturbation of the deposit with respect to the source and detectors. Comparing the data when the source is close to the deposit and when the detector is close to the deposit shows a 1.01% difference between the two signatures in the 6.7 and 11.5 MeV energy range. When the detector is close to the deposit, ^{252}Cf neutrons that are scattered within the deposit still have a chance to reach the detector and be detected. When the source and detector position are reversed, scattering occurs in the material far from the detector and these neutrons are subsequently not detected. Figure 3.12 illustrates this effect more clearly, indicating that it may be useful for confirming the relative material location.

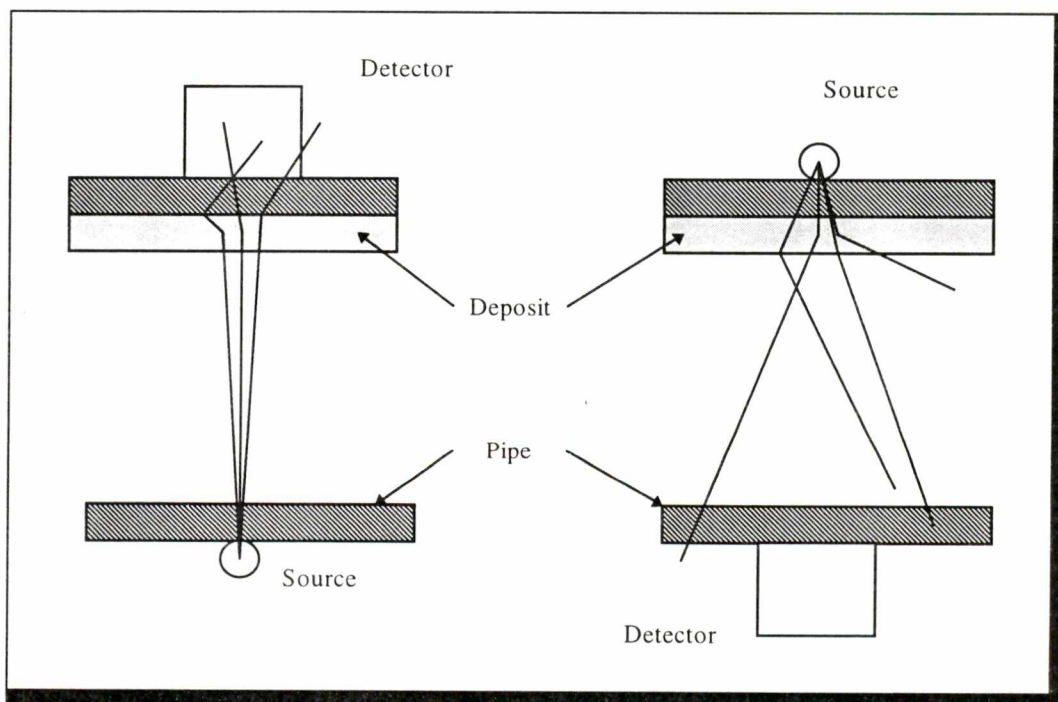


Figure 3.12 Scattering Effects of Source and Detector Placement

Additional calculations were performed where the hydrogen content of the deposit was varied over the range of 0.6 to 6.1 to verify that the neutron transmission at 8.5 MeV is invariant to hydrogen content and thus material density. Figure 3.13 shows the calculated responses for two of these models, indicating that the results for high energy neutrons are essentially invariant to hydration. The MCNP-DSP models for the geometry and hydration perturbations are included in Appendix D.

In order to verify that MCNP-DSP could accurately calculate the cross-correlation between the source and detector, two simple measurements and calculations were performed. In the first, the source and detector were placed in air with a 24 in. separation between them. The second measurement and calculation placed the source and detector on opposite sides of an empty pipe section identical to that installed in the process gas system. The results of these measurements and calculations are shown in Figures 3.14 and 3.15. Note again the arbitrary shift of the gamma peak to 25 nsec for figure clarity. The calculated and measured photon peaks disagree because of timing resolution and jitter in the physical system which cause the counts to be somewhat spread into adjacent time bins. These uncertainties are not accounted for in MCNP-DSP so that all the gamma direct transmission counts fall into a single bin. Otherwise the measured and calculated signatures show excellent agreement.

The MCNP-DSP calculations verify (1) the high energy portion of the neutron response is invariant to source-detector placement or deposit hydration for a given thickness, and (2) the upper portion of the energy spectrum around 8.5 MeV is sufficient for conducting the analysis without the detrimental effects of inscattering occurring.

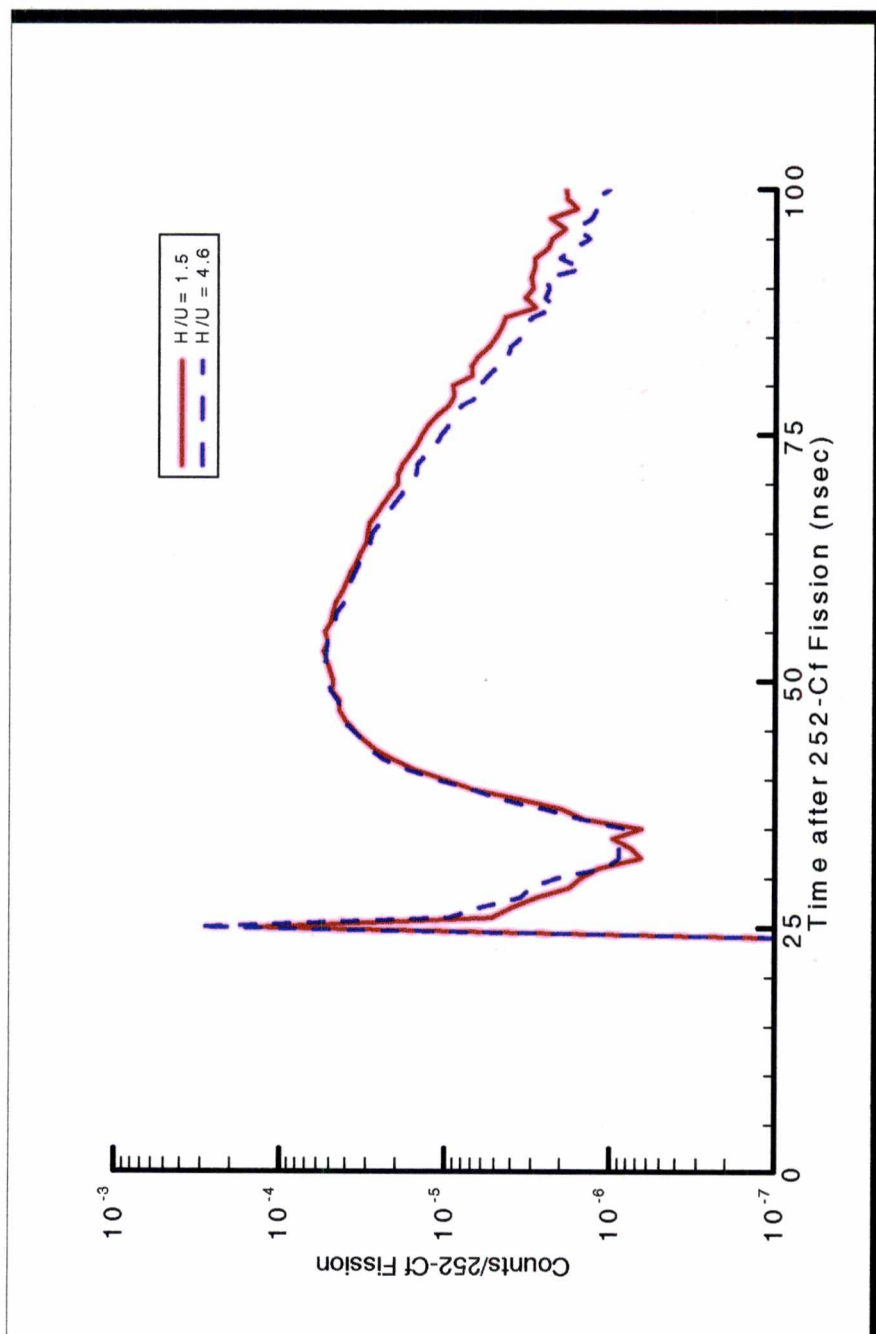


Figure 3.13 Calculated Signatures for Two Different Material Hydrations, Indicating that the Rise Portion of the Neutron Spectrum is Invariant to Hydration

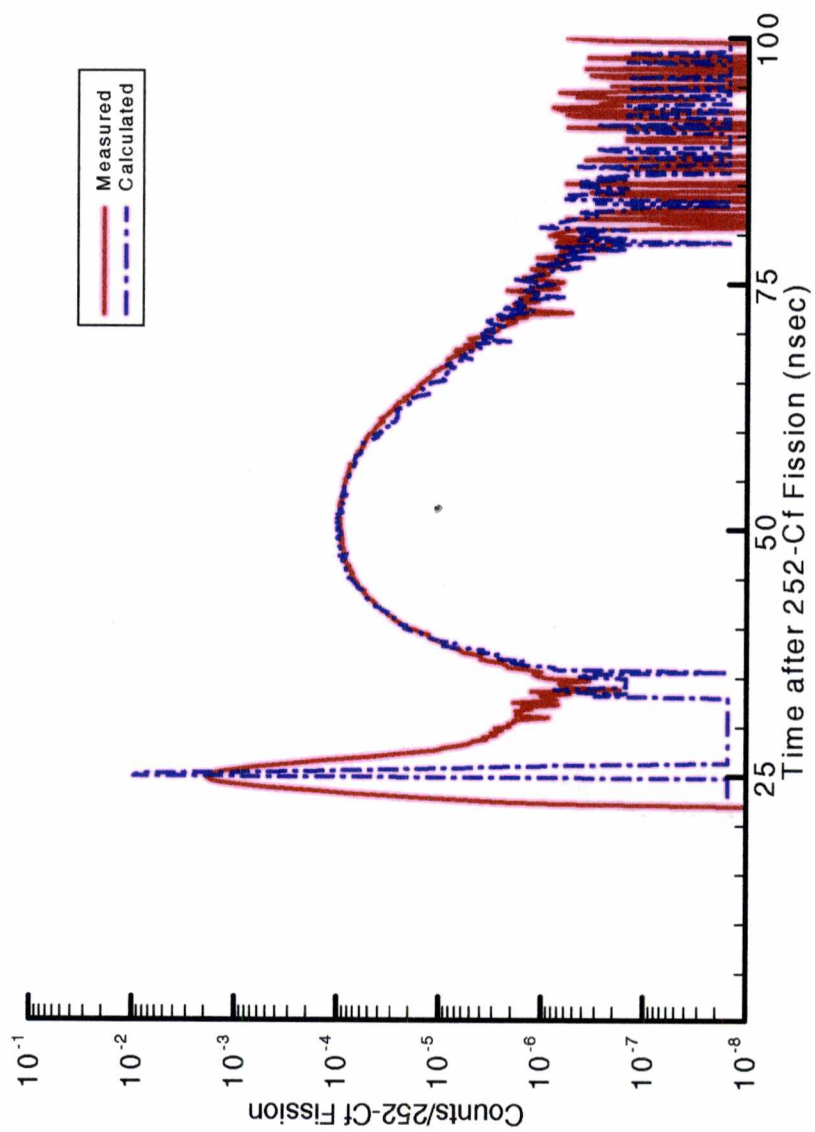


Figure 3.14 Calculated MCNP-DSP and Measured Signatures for Source and Detector in Air

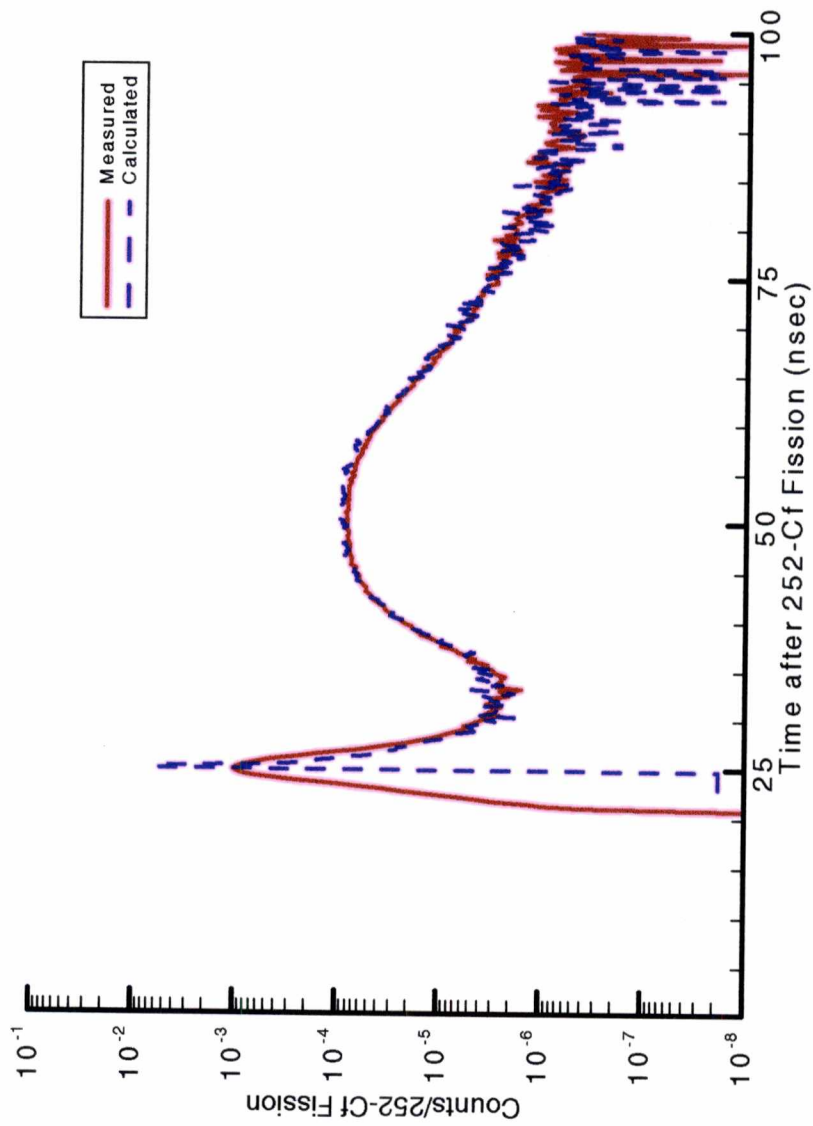


Figure 3.15 Calculated MCNP-DSP and Measured Signatures for Transmission Measurement Through Empty Pipe

Chapter 4

Measurement Methods and Procedures

4.0 NWIS Processor

Figure 4.1 shows a simplified block diagram of the measurement system. The ^{252}Cf source is approximately 1.0 in. O.D. and 1.25 in. long, with 0.6 μg electroplated onto one plate for a spontaneous fission rate of 1.38×10^6 fissions/s. The detectors are organic plastic scintillators (BICRON-420) suitable for fast timing. Each detector is 3.75 x 3.75 in. square and enclosed in an aluminum can 0.125 in. thick mounted on a photomultiplier tube. The detectors are wrapped with $\frac{1}{4}$ in. of lead to reduce background count rates in the detector except on the front face of the detector. BICRON-420 has a hydrogen carbon ratio of 1.1/1.0 and a bulk density of 1.032 g/cm^3 , providing high efficiency neutron detection via elastic scattering with protons (proton-recoil scintillation). Gamma detection occurs via the usual process of interactions with electrons as the dominant energy loss mechanism.

The ^{252}Cf source has +200 volts placed across its parallel plates in order to collect charged particles produced by ionization of the counting gas. These pulses are then amplified by a fast high gain amplifier and the analog signal passed through a constant fraction discriminator for pulse discrimination and generation of an output signal suitable for transmission over long distances. The discriminator also provides a constant timing mark that is independent of the pulse amplitude.

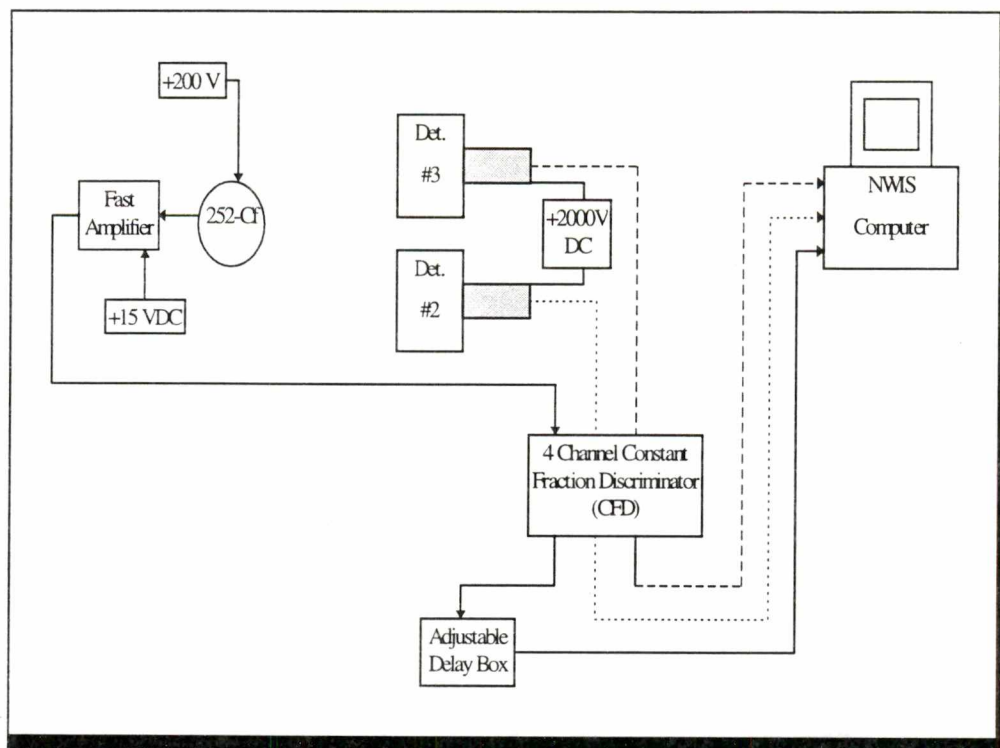


Figure 4.1 Simplified Diagram of NWIS

The ^{252}Cf pulse rise time is on the order of 1 nsec, compared to the approximate 10 nsec time constant of the detector photomultiplier tube. The ^{252}Cf pulse is delayed in order to account for the pulse propagation time through the photomultiplier tubes. Pulses from the ^{252}Cf and detector channels are then sampled at 1 GHz (1 ns timing resolution) by the NWIS processor. Data is sampled continuously over 256, 512, or 1024 time bins, with one group of time bins forming a block of data. The NWIS processor provides almost real-time data processing of the collected blocks of data until the signature is sufficiently converged. Measurement times on the Hockey Stick were a nominal 14 minutes per location in order to have a well converged high energy neutron signal. Figure

4.2 shows the NWIS processor deployed in the lower level of the K-29 building for the deposit measurements. Because the Hockey Stick is in a high contamination area, the NWIS processor is located in a clean area approximately 400 ft. from the deposit

4.1 Deposit Profile

The spatial distribution of the deposit in the process gas piping is obtained by performing synchronized horizontal and vertical scans of the source and detector along the pipe as shown in Figure 4.3. An additional set of source-detector rotations around the circumference of the pipe in 30 degree increments are performed in order to supplement the information obtained from the vertical and horizontal scans. The source and detector are moved in 2 in. increments during the horizontal and vertical scans in order to provide 2 in. of overlap from the 4 in. detector at each position. For a vertical scan, positions above the midplane are denoted by a positive number corresponding to the distance, measured from the midplane to the center of the detector. Locations below the midplane are given by a corresponding negative number. For instance, +10 places the source and detector midline 10 in. above the midplane of the pipe, with the upper edge of the detector in a plane tangent to the top of the pipe. In a similar manner for horizontal scans, positions to the left are given a positive number and those to the right a negative number. Rotations begin with the source on top of the pipe (zero degrees rotation) and the detectors on bottom. The fixture is rotated in 30 degree increments until the source reaches the 330 degree position. A fixture, shown in Figure 4.4, was fabricated in order to

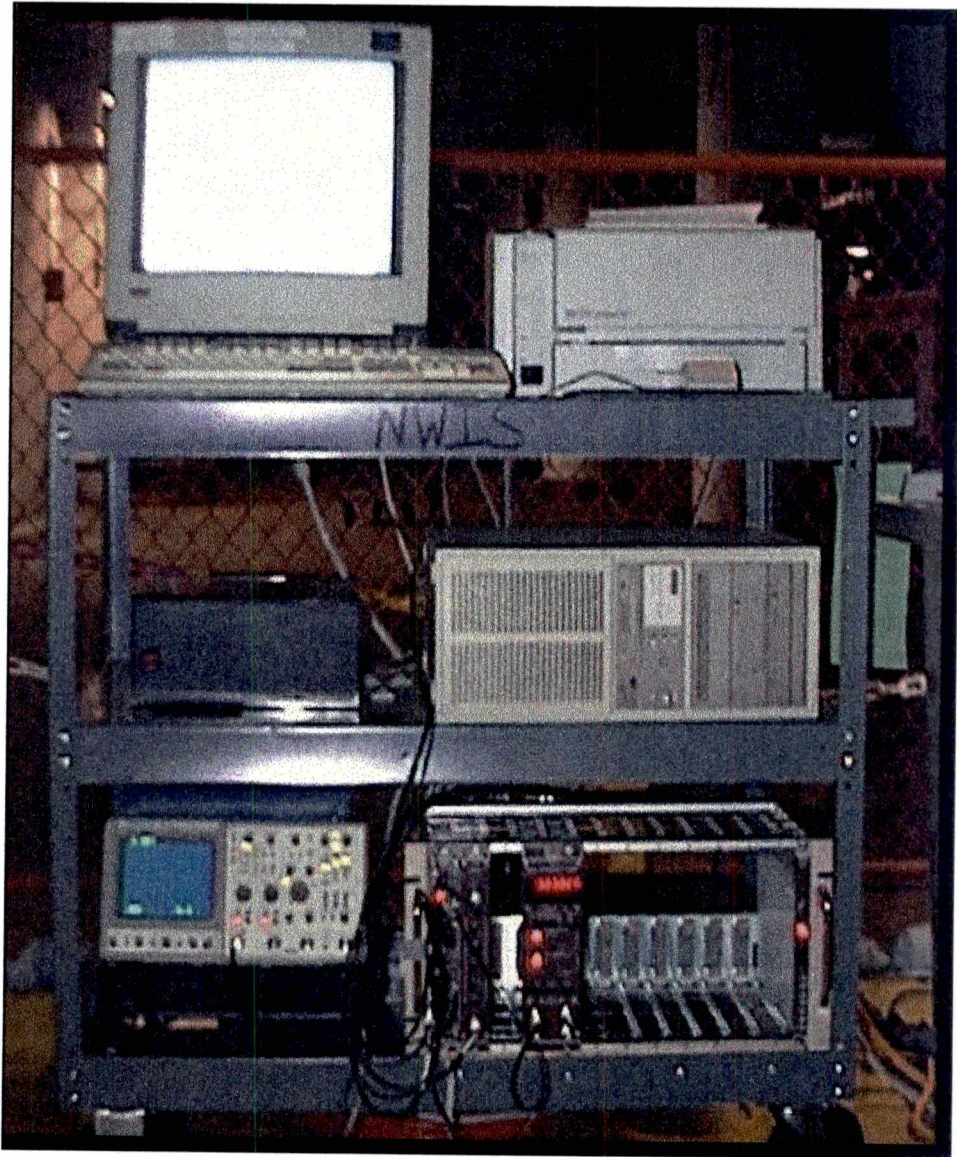


Figure 4.2 NWIS Field Deployed in Lower Level of K-29 Bldg.

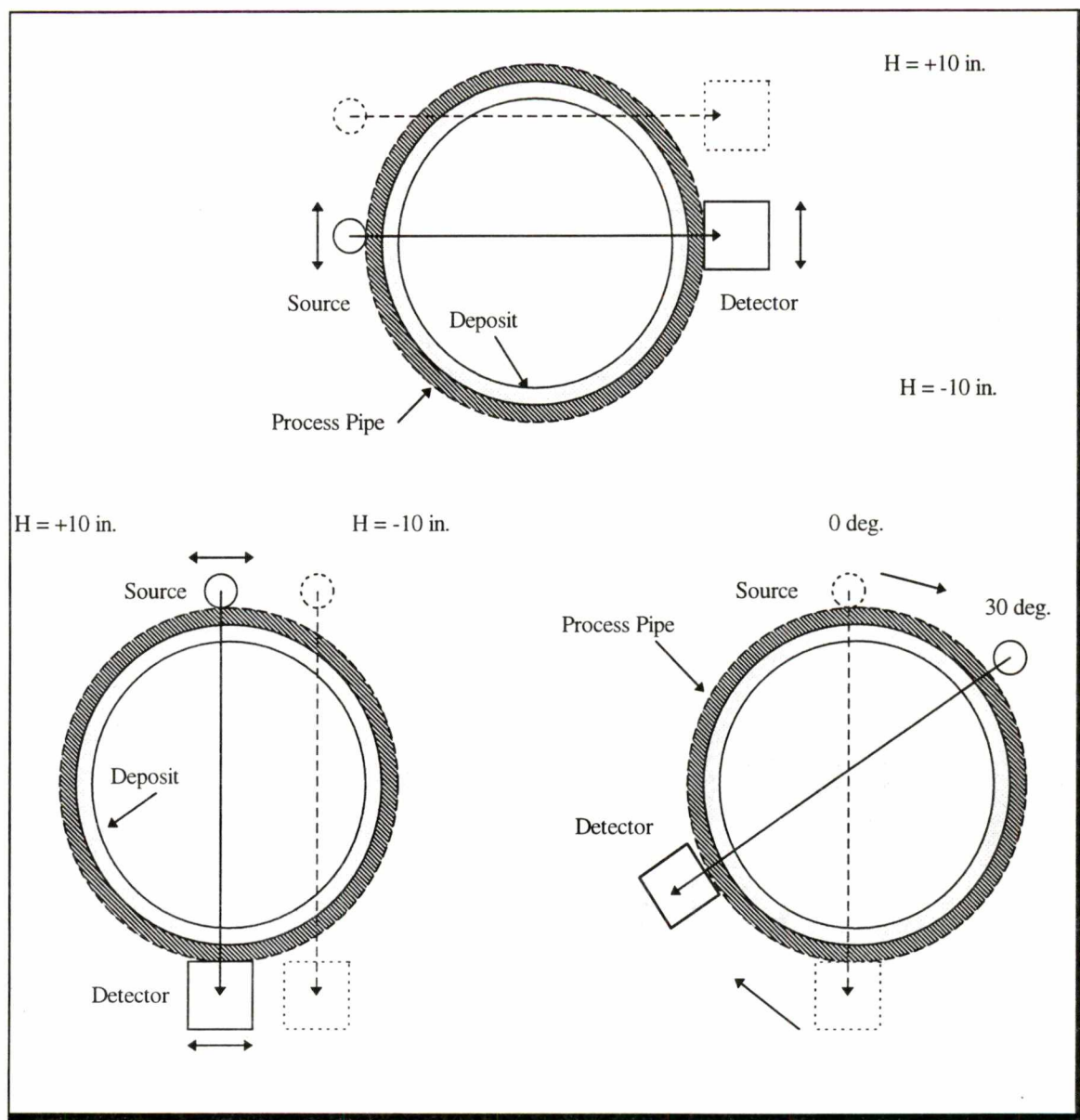


Figure 4.3 Vertical Source and Detector Scan (Top) , Horizontal Source and Detector Scan (Bottom Left), and Source-Detector Rotation (Bottom Right) Used for Mapping the Distribution of Material in the Process Gas Piping

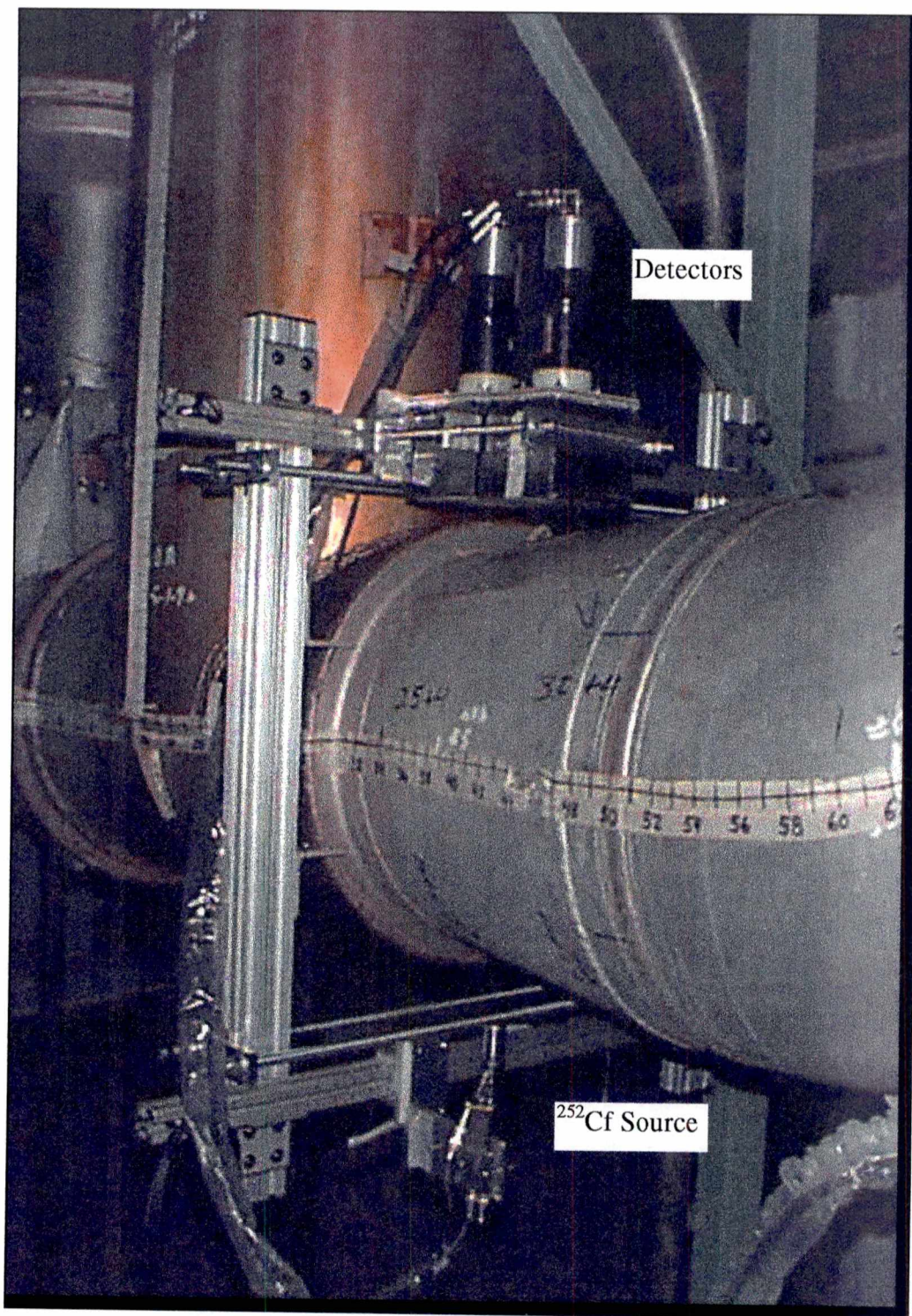


Figure 4.4 Source and Detectors on Positioning Fixture Performing a Horizontal Scan

allow quick positioning of the source and detectors in order to minimize time spent near the source, and to ensure proper alignment and spacing.

4.2 Measurement Calibration

In order to remove the contribution of the process gas piping in the measured signatures as described in Sect. 3.2.1, a set of calibration measurements were completed on a 2 ft. section of clean pipe that had been previously removed from the cascade system. Calibrations were performed at 2 in. increments in a manner consistent with measurements on the Hockey Stick. Fig. 4.5 shows the calibration section in place, at the same height above the floor as the Hockey Stick, to account for the effects of reflection from the floor.

A neutron time-of-flight measurement with the source and detector spaced 24 in. in air were performed at the beginning, middle, and end of each shift in order to verify proper equipment operation.

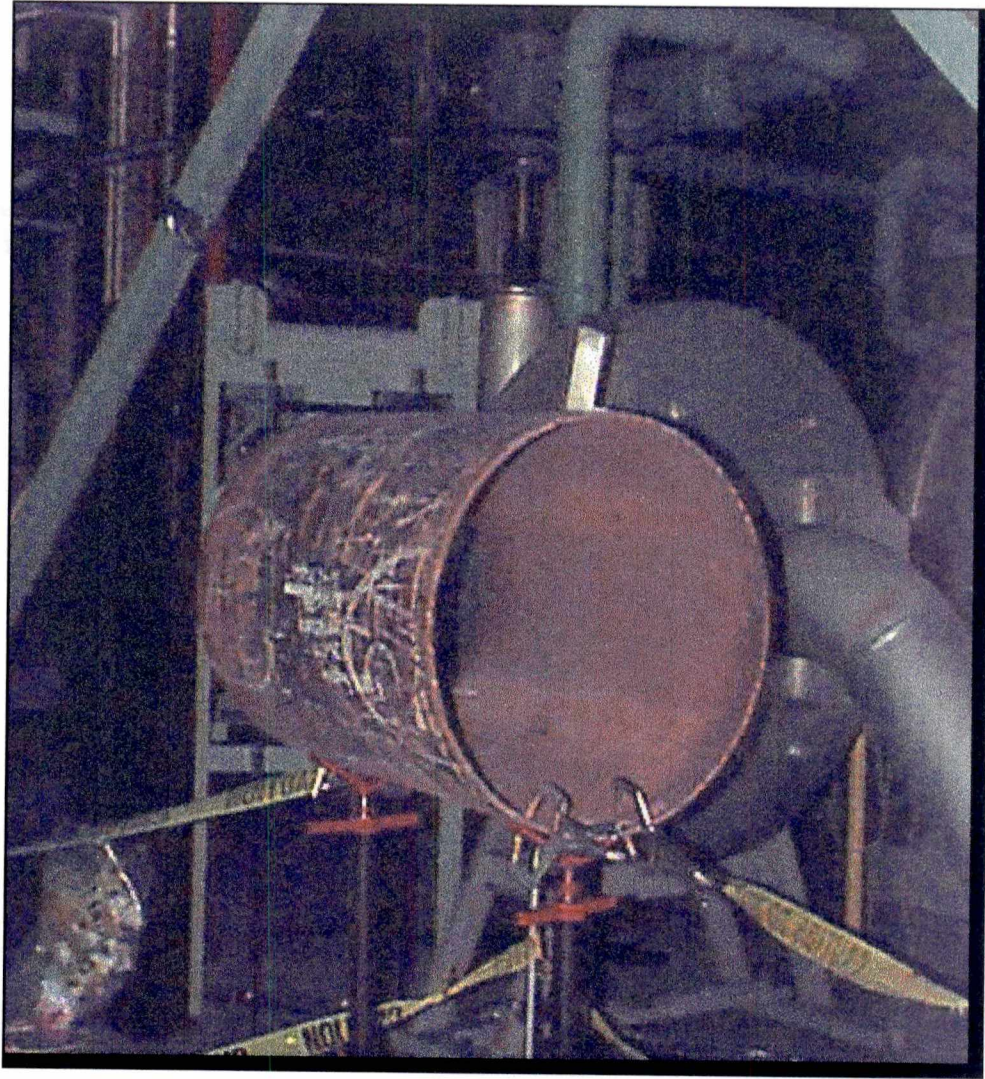


Figure 4.5 Clean Pipe Section for Calibration Measurements

CHAPTER 5

Measurement Results of the Hockey Stick Deposit

5.0 Introduction

The Hockey Stick deposit was characterized by determining the cross-sectional distribution of material at various locations along the length of the pipe. Once the deposit profile was determined, the hydrogen content, or ratio of H/U, was obtained using the techniques outlined in the previous sections. Not every location was equally accessible due to obstructions or safety concerns, so some measured profiles are not as well characterized as others. In order to provide reference points for future deposit removal activities, the pipe was marked in 1 in. increments on both sides at the midplane, beginning from the welded metal blank behind the 7BB2 block valve and continuing to just below the bypass header at the top of the Hockey Stick. The locations where the deposit profiles are measured were identified by a numeric, corresponding to the measured distance marked on the pipe as measured from the welded metal blank. For instance, $L = 80$ in. refers to a profile taken at 80 in. along the pipe with respect to the metal blank. Figure 5.1 shows the locations which were characterized for this project. Appendix E contains additional photographs of the Hockey Stick characterization during various phases of the measurements.

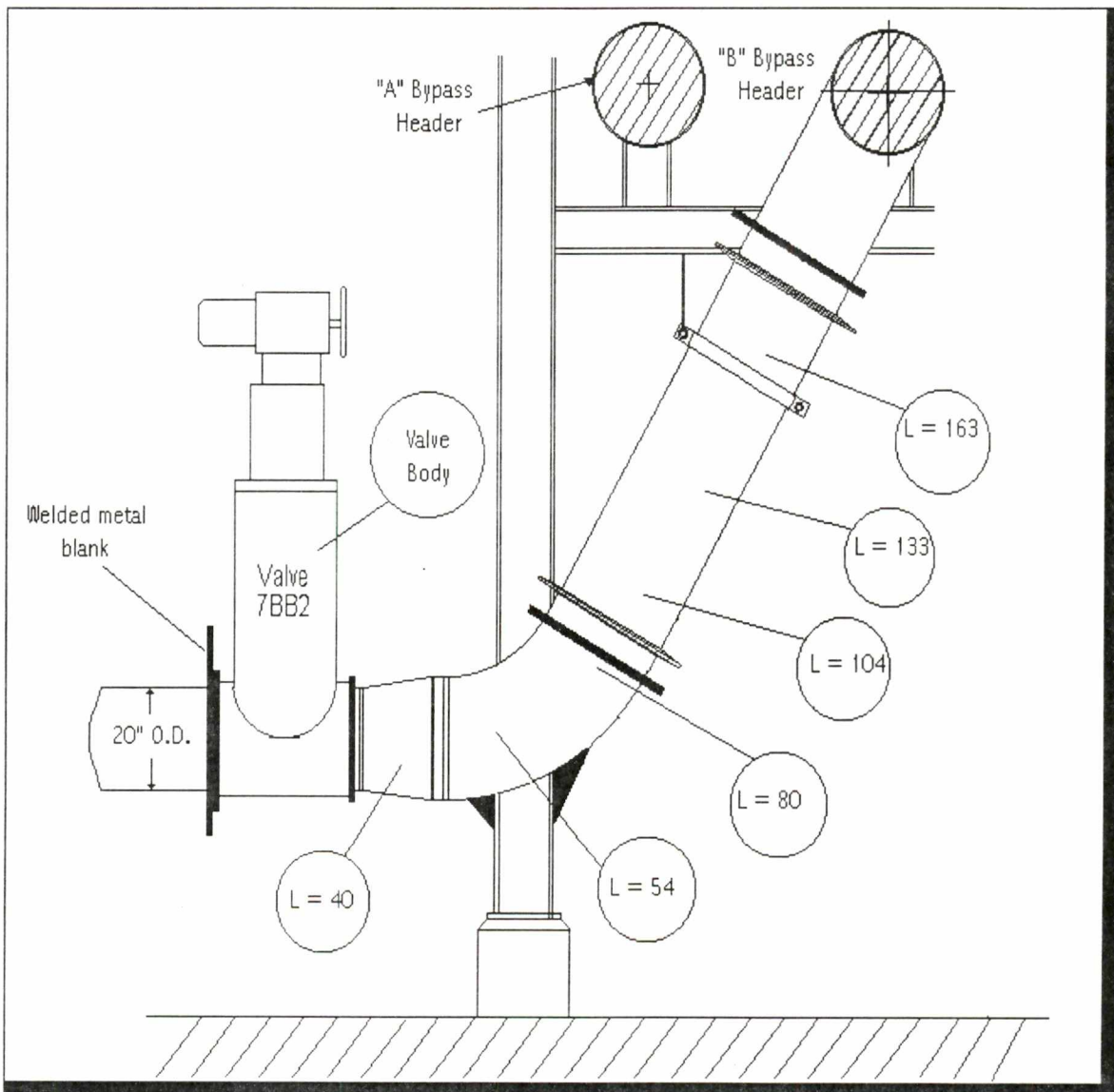


Figure 5.1 Measured Locations for the Hockey Stick Deposit Characterization

5.1 Measurement Results at L = 54

The location L = 54 is a 24-in.-O.D. pipe location near the bottom of the Hockey Stick, near the 7BB2 inlet valve where the wet air in-leakage was thought to have occurred. Vertical and horizontal scans were performed, as well as a source-detector rotation with obstructions preventing measurements at 0 and 270 degrees rotation. The neutron data was analyzed at 8.6 MeV, which corresponds to a time of 15 ns after fission for a source-detector separation of 24 in. Initially, an average value of the total neutron interaction cross-section over the possible range of H/U, corresponding to a H/U of 4.8, was used to calculate the deposit thickness from the transmission measurements. The first set of results of the measured thickness, without iterative improvement, are presented in Tables 5.1 - 5.3.

Table 5.1 Vertical Source and Detector Scan at L = 54 with Cross-Section Data			
Averaged Over the Range of H/U			
Run #	Position (in)	Ln (I / I_o)	Thick (in)
df04r13	10	2.13	5.38
df05r01	8	1.29	3.27
df05r02	6	0.870	2.20
df05r03	4	0.391	0.99
df05r04	2	0.259	0.65
df05r05	0	0.065	0.16
df05r06	-2	0.287	0.73
df05r07	-4	0.261	0.66
df05r08	-6	0.200	0.51
df05r09	-8	0.225	0.57
df05r10	-10	0.070	0.18

Table 5.2 Horizontal Source and Detector Scan at L = 54 with Cross-Section Data Averaged Over the Range of H/U

Run #	Position (in)	Ln (I / I _o)	Thick (in)
df05r16	10	0.506	1.28
df05r15	8	0.713	1.80
df05r14	6	0.858	2.17
df05r13	4	0.978	2.47
df05r12	2	0.987	2.50
df05r11	0	0.769	1.94
df05r17	-2	0.909	2.30
df05r18	-4	0.819	2.07
df05r19	-6	0.623	1.58
df05r20	-8	0.649	1.64
df05r21	-10	0.445	1.13

Table 5.3 Rotation of Source and Detector at L = 54 with Cross-Section Data Averaged Over the Range of H/U

Run #	Source Rotation (Deg.)	Ln (I / I _o)	Thick (in)
df17r24	30	0.782	1.98
df17r25	60	0.409	1.03
df17r26	90	0.360	0.91
df17r27	120	0.585	1.48
df17r28	150	0.929	2.35
df17r29	180	1.19	3.00
df17r30	210	0.907	2.29
df17r31	240	0.573	1.45
df17r32	300	0.494	1.25
df17r33	330	0.823	2.08

Table 5.1 immediately suggests that, contrary to expectation, at $L = 54$ the deposit exists on the top of the pipe and that the bottom contains very little material. The data also suggests that the deposit has an annular shape because increasing displacement from the midplane yields an increasing measured thickness. This phenomena, termed edge effects, is more clearly explained by Figure 5.2.

Table 5.2 indicates that the deposit profile is fairly symmetric from left-to-right by comparing the values at ± 10 in., ± 8 in., etc. Since the vertical scans reveals that very little material exists in the bottom of the pipe, the information from the horizontal scan can be used to calculate the deposit thickness at the top of the pipe. Care must be taken to avoid extrapolating far from the midplane because of edge effects.

Information about the profile is further enhanced by the source-detector rotation data from Table 5.3. Obstructions prevented measurements at the 0 and 270 degree positions. Ideally measurements locations that are 180 degrees apart should yield the same results, but slight differences are noted. Measurements with the detector located on the top of the pipe indicate slightly more material than the complementary measurements with the source and detector position reversed. The slight increase when the detector is on the top of the pipe is attributed to its proximity to the deposit, confirming that the deposit resides at that location.

Table 5.4 summarizes the results, without iterative improvement, of the gamma portion of the signature at $L=54$ used to calculate the hydration of the deposit. In order to avoid edge effects, the deposit hydration is only measured near the horizontal and vertical midplanes of the pipe, corresponding to 0, ± 2 , and ± 4 inches.

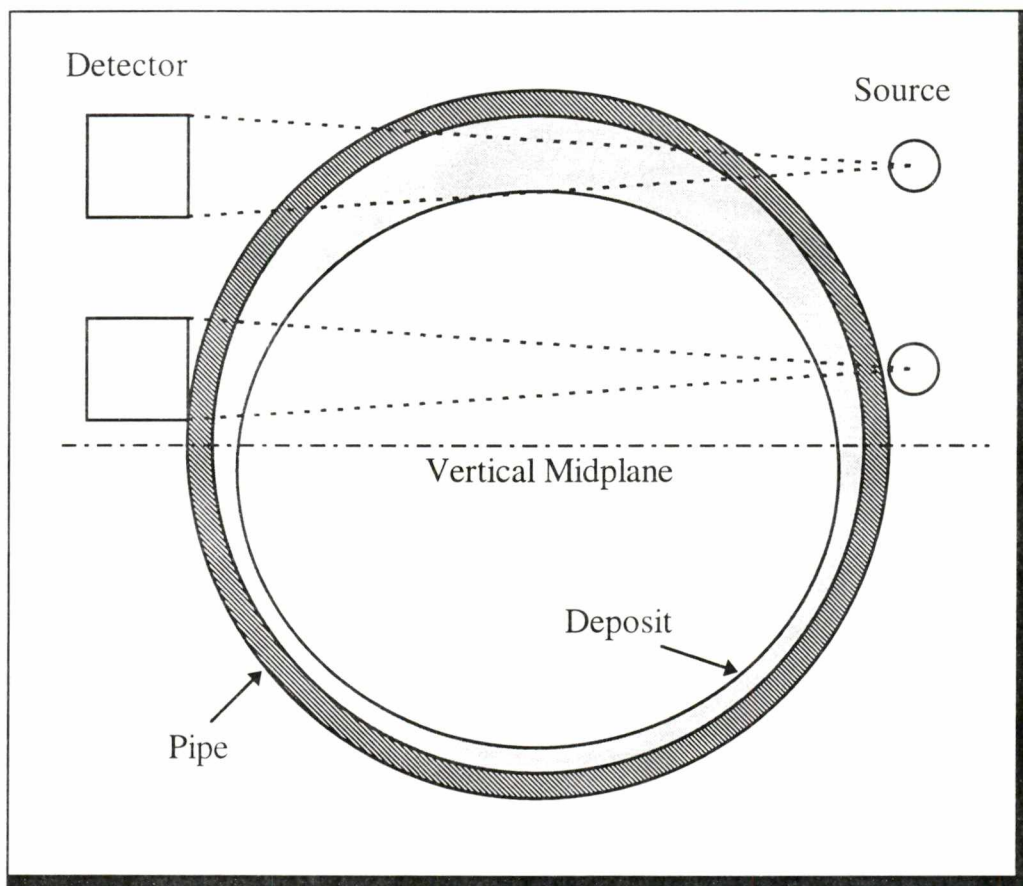


Figure 5.2 Increasing Thickness for an Annular Deposit Due to Edge Effects

Table 5.4 First Estimate of Deposit Hydration at L = 54 from Vertical and Horizontal Scan				
Vertical Run #	Thick (in.)	Position	Ln (A_g / A_{ref})	H/U
df05r03	0.99	4	0.70	> 15
df05r04	0.65	2	0.69	11.5
df05r05	0.16	0	0.73	> 15
df05r06	0.73	-2	0.74	3.5
df05r07	0.66	-4	0.73	14.7
Horizontal Run #	Thick (in.)	Position	Ln (A_g / A_{ref})	H/U
df05r13	2.47	4	0.21	5.5
df05r12	2.50	2	0.21	5.5
df05r11	1.94	0	0.18	2.0
df05r17	2.30	-2	0.18	3.5
df05r18	2.07	-4	0.19	3.0

These results clearly reiterate the problem identified in Section 3.2.2 for thin deposits. Comparing the data from the -2 and -4 position for the vertical scan, a difference of 0.01 is present in the ratio of the gamma peaks. Compare this to the difference in the estimated thickness at the two locations of 0.07 in. However, due to the extreme sensitivity of the method in thin materials, there is a large difference in the estimation of the value for H/U. For this reason, only data with deposit thickness greater than 1.25 in were used in determination of the deposit hydration.

5.2 Iterative Improvement of Measurement Results at $L = 54$

Using the method outlined in Sect. 3.2.3, an iterative refinement of the deposit thickness and hydration is presented in Table 5.5 for the deposit profile at $L = 54$. Acceptable convergence was reached in three iterations of the cross-sections. The second iteration has an average H/U value of 3.7, and the third iteration has an average value of 3.8. These small changes in H/U do not significantly effect the cross-sections, so that the estimated thickness will not change and the solution has converged. Table 5.5 suggests that the deposit thickness is not well characterized by the measurement due to the variations in the deposit thickness from 1.83 in. to 2.35 in. This problem is addressed in Sect. 5.6 by comparing the measured data to the findings of an intrusive look into the Hockey Stick using a fiber optic camera which shows the non-uniformity of the deposit.

The average value of H/U at this location, using results only from the horizontal scan, is 3.8. This number is somewhat premature due to the small sample size of 5 data points and the noted variations in the deposit thickness which strongly effect the calculated material density. In order to better quantify the value of H/U for the Hockey Stick, the remainder of the profiles should be included in the analysis. Figure 5.3 presents the estimated profile of the deposit at $L=54$. These results incorporate data from the vertical, horizontal, and rotational scans, and assume that the deposit profile is smoothly varying. It should also be noted that all deposit thicknesses and hydrations reported are *average* results, based on the region covered by the 4 in. x 4 in. detectors.

Table 5.5 Iterative Results at L = 54 Using Measurement Data from Source and Detector Horizontal Scan									
Location		Initial Results		Iteration #1		Iteration #2		Iteration #3	
(Horiz. Scan)		Thick (in.) ^a	H/U ^a	Thick (in.) ^a	H/U ^a	Thick (in.) ^a	H/U ^a	Thick (in.) ^a	H/U ^a
HS +4		2.47	5.5	2.21	4.5	2.30	4.8	2.33	4.9
HS +2		2.50	5.5	2.23	4.5	2.30	4.8	2.35	4.9
HS 0		1.94	2.0	1.74	1.5	1.80	2.1	1.83	2.3
HS -2		2.30	3.5	2.05	3.5	2.12	3.6	2.17	3.7
HS -4		2.07	3.0	1.85	3.0	1.92	3.2	1.96	3.3

^a Average thickness and hydration of region seen by 4 in. x 4in. detectors

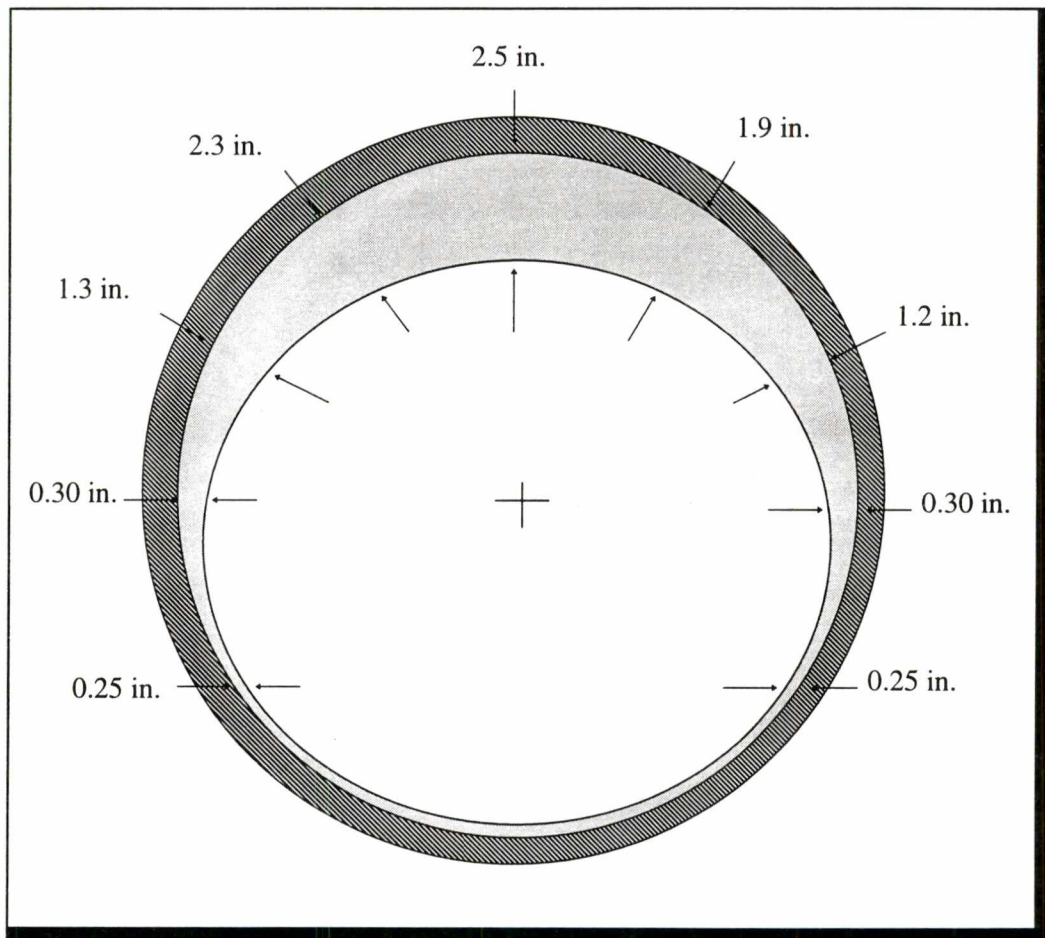


Figure 5.3 Deposit Profile at $L = 54$

5.3 Measured Deposit Profiles for the Remainder of the Pipe

This section presents the measurement results, with iterative improvement, for locations $L = 40, 80, 104, 133,$ and 163 . Overall, the deposit moves from a very asymmetric distribution at the bottom of the Hockey Stick near the 7BB2 block valve to an increasingly symmetric deposit at the top of the riser. The deposit loading (kg/ft) or thickness also decreases moving up the pipe, and the material distribution appears to become more smoothly varying.

5.3.1 Measurement Results at $L = 40$

This location is a transition piece connecting the 20 in. O.D. valve body and the 24 in. O.D. process gas pipe. Due to its smaller diameter, no measurement was possible at the ± 10 in. source-detector locations. Calibration measurements were taken from a nearby identical transition piece already determined to contain no deposits. Data from the vertical and horizontal scans are presented in Table 5.6 and 5.7. The profile is similar to $L = 54$, with the majority of the deposit on the top of the pipe. Figure 5.4 presents the estimated deposit profile, which is again assumed to be smoothly varying.

Table 5.6 Results of Vertical Source and Detector Scan at $L = 40$			
Run #	Position (in.)	$\ln(I/I_o)$	Thick (in)
df19r13	8	1.79	4.10
df19r14	6	1.18	2.71
df19r15	4	0.51	1.18
df19r16	2	0.51	1.17
df19r17	0	0.20	0.47
df19r18	-2	0.28	0.64
df19r19	-4	0.17	0.39
df19r20	-6	0.14	0.31
df19r21	-8	0.12	0.28

Table 5.7 Results of Horizontal Source and Detector Scan at L = 40			
Run #	Position (in.)	$\text{Ln} (I / I_0)$	Thick (in)
df19r22	8	0.20	0.46
df19r23	6	0.14	0.31
df19r24	4	0.32	0.72
df19r25	2	0.41	0.93
df19r26	0	0.41	0.93
df19r27	-2	0.43	0.99
df19r28	-4	0.25	0.57
df19r29	-6	0.24	0.55
df19r30	-8	0.19	0.43

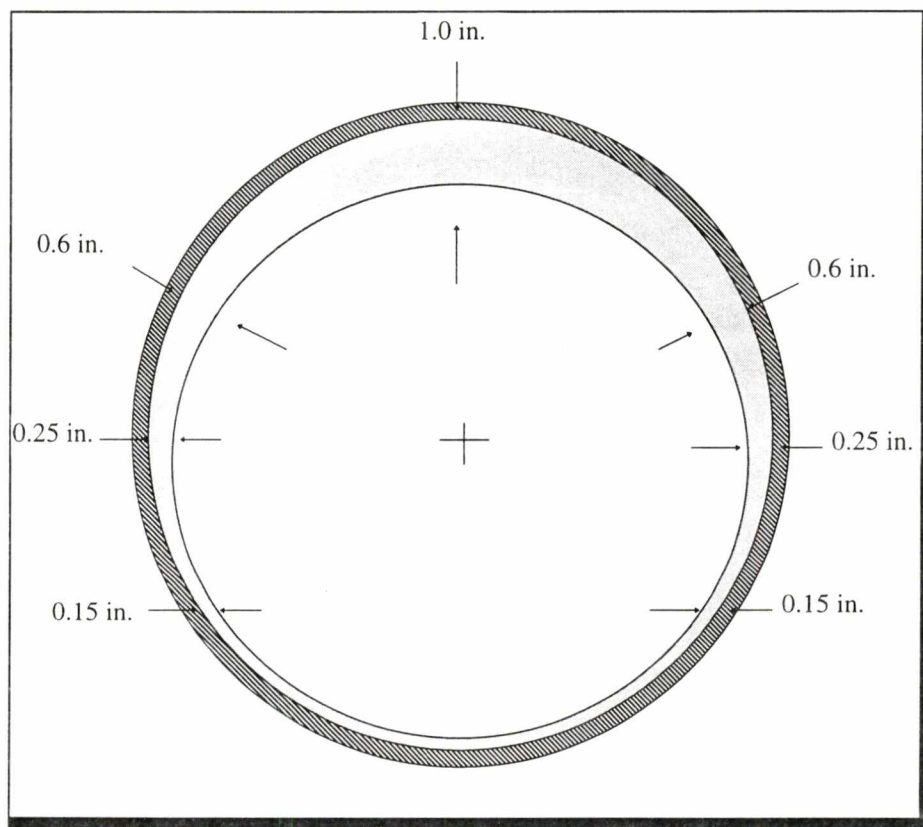


Figure 5.4 Deposit Profile at L = 40

5.3.2 Measurement Results at L = 80

This position corresponds to the curved section of the pipe directly below the expansion joint. This location was extremely difficult to measure due to interference of the expansion joint with the fixture that hold the source and detector, and consequently only a single vertical scan was performed at this location. Results from the vertical scan alone indicate that the deposit still exists primarily on the top section of the pipe. By comparing the thickness of the deposit near the edges to a similar location that is well characterized, such as L = 54, an estimate for the remainder of the profile can be inferred. The measurement results from the vertical scan are presented in Table 5.8 and the estimated deposit profile is shown in Figure 5.5.

Table 5.8 Results of Vertical Source and Detector Scan at L = 80			
Run #	Position	Ln (I / I _o)	Thick (in)
df05r22	10	1.97	5.07
df05r24	8	1.13	2.90
df05r25	6	0.79	2.02
df05r26	4	0.45	1.15
df05r27	2	0.63	1.62
df05r28	0	0.35	0.91
df05r29	-2	0.45	1.16
df05r30	-4	0.45	1.16
df05r31	-6	0.52	1.33
df05r32	-8	0.45	1.16
df20r23	10	0.29	0.76

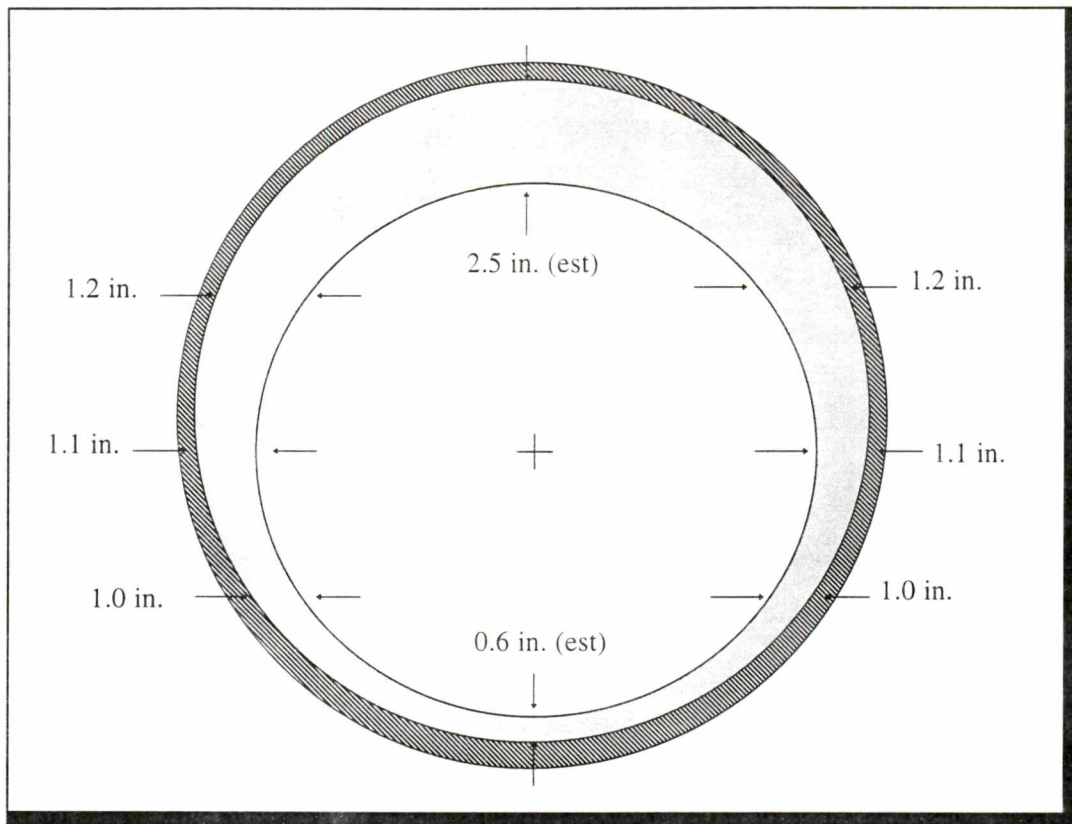


Figure 5.5 Deposit Profile at L = 80

5.3.3 Measurement Results at L = 104

This location is on the vertical riser, immediately above the expansion joint (Fig. 5.1). As evidenced from Table 5.9 and 5.10, the deposit is more symmetric in this location, with material appearing in the bottom half of the pipe where none had been seen in previous locations. Data from the source-detector rotation in Table 5.11 suggests that a hole exists along the periphery of the deposit, as indicated by sudden dips in the estimated thickness at 60 and 240 degrees. This seems to be confirmed by corresponding dips at the -4 and -6 positions on the horizontal scan. The estimated deposit profile for this location is given by Figure 5.6.

Table 5.9 Results of Vertical Source and Detector Scan at L = 104			
Run #	Position (in.)	Ln (I / I _o)	Thick (in)
df03r13	10	1.17	2.87
df03r12	8	0.68	1.68
df03r11	6	0.39	0.95
df03r10	4	0.33	0.81
df03r09	2	0.42	1.02
df03r08	0	0.42	1.02
df03r14	-2	0.23	0.57
df03r15	-4	0.19	0.47
df03r16	-6	0.29	0.71
df03r17	-8	0.26	0.65
df03r18	10	0.35	0.87

Table 5.10 Results of Horizontal Source and Detector Scan at L = 104			
Run #	Position (in.)	Ln (I / I _o)	Thick (in)
df04r06	10	0.69	1.70
df04r05	8	0.71	1.75
df04r04	6	0.63	1.54
df04r03	4	0.51	1.24
df04r02	2	0.64	1.57
df04r01	0	0.52	1.27
df04r07	-2	0.49	1.21
df04r08	-4	0.38	0.92
df04r09	-6	0.39	0.95
df04r10	-8	0.60	1.47
df04r11	10	0.68	1.67

Table 5.11 Results of Source and Detector Rotation at L = 104

Run #	Rotation (deg.)	$\text{Ln} (I / I_0)$	Thick (in)
df13r01	0	0.45	1.11
df13r02	30	0.44	1.08
df13r03	60	0.21	0.51
df13r04	90	0.52	1.27
df13r05	120	0.45	1.11
df13r06	150	0.68	1.67
df13r07	180	0.63	1.55
df13r08	210	0.40	0.98
df13r09	240	0.28	0.68
df13r10	270	0.64	1.58
df13r11	300	0.77	1.88
df13r12	330	0.63	1.55

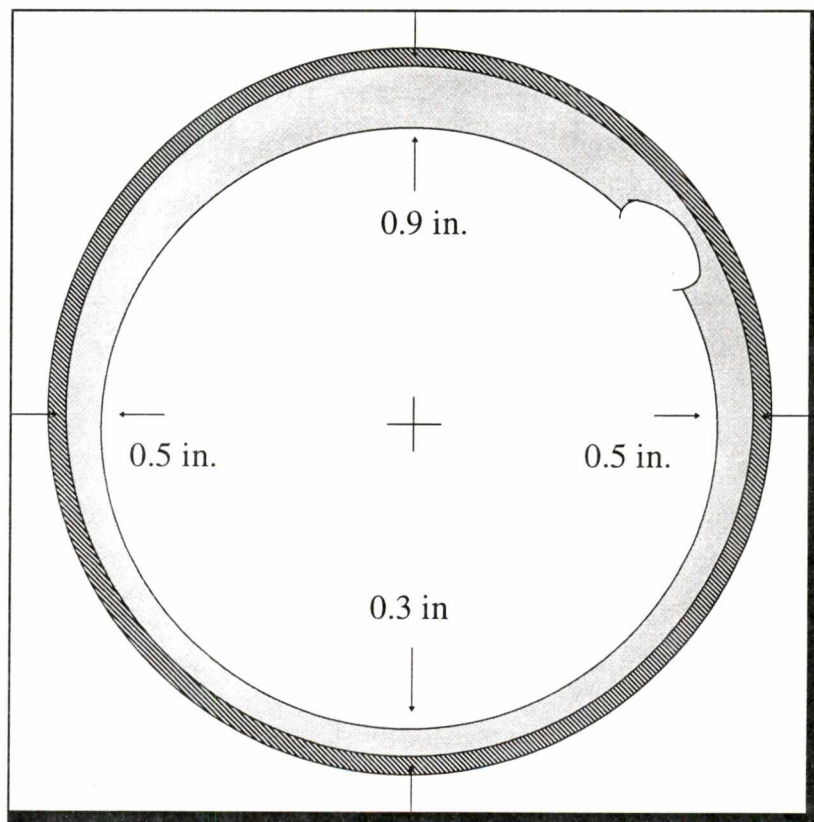


Figure 5.6 Deposit Profile at L = 104

5.3.4 Measurement Results at L =133

Moving further up the vertical riser, the deposit profile is becoming increasing symmetric although there is still a slight tendency for more material to exist on the upper half of the pipe as compared to the lower. The data is varying more smoothly from location to location, in contrast to the data at L = 54. Results from the vertical and horizontal scans are presented in Tables 5.12 and 5.13, and the estimated deposit profile is given in Figure 5.7.

Table 5.12 Results of Vertical Source and Detector Scan at L = 133

Run #	Position (in.)	Ln (I / I _o)	Thick (in)
df16r02	10	1.06	2.94
df16r03	8	0.91	2.51
df16r04	6	0.62	1.73
df16r05	4	0.51	1.42
df16r06	2	0.60	1.65
df16r07	0	0.41	1.12
df16r08	-2	0.63	1.74
df16r09	-4	0.44	1.21
df16r10	-6	0.58	1.60
df16r11	-8	0.61	1.68
df16r12	10	0.73	2.01

Table 5.13 Results of Horizontal Source and Detector Scan at L = 133

Run #	Position (in.)	$\text{Ln} (I / I_0)$	Thick (in)
df14r12	10	0.63	1.62
df14r13	8	0.88	2.29
df14r14	6	0.76	1.97
df14r15	4	0.93	2.42
df14r16	2	1.21	3.14
df14r17	0	0.63	1.62
df14r18	-2	0.83	2.15
df14r19	-4	0.90	2.34
df14r20	-6	0.77	2.00
df14r21	-8	0.65	1.68
df14r22	10	0.61	1.59

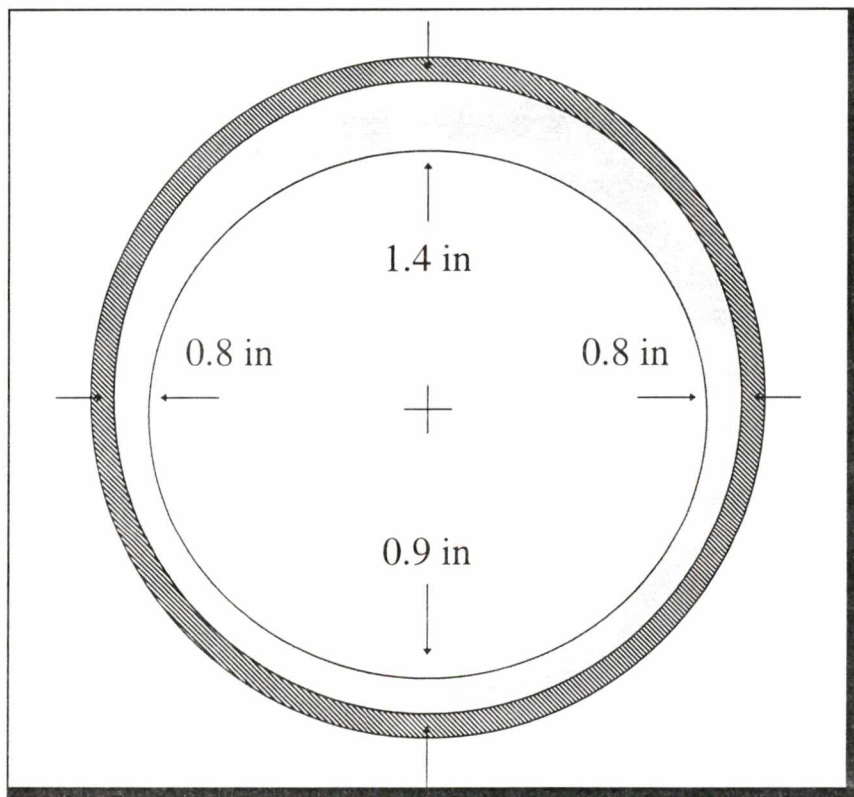


Figure 5.7 Deposit Profile at L = 133

5.3.5 Measurement Results at L =163

This is the last location measured and corresponds to the highest position up the vertical riser, approximately 10 ft. from the floor. Here the deposit is very symmetric compared to other locations, and is somewhat thinner than the previous location at L = 133. Again, the measured data appears to varying much more smoothly than the measured results at L =54 and L = 80. Tables 5.14 and 5.15 present the measured data for this location, and the estimated deposit profile is given in Figure 5.7.

Table 5.14 Results of Vertical Source and Detector Scan at L = 163

Run #	Position (in.)	$\text{Ln} (I / I_o)$	Thick (in)
df20r24	10	0.27	0.74
df20r25	8	0.42	1.13
df20r26	6	0.38	1.05
df20r27	4	0.46	1.24
df20r28	2	0.57	1.56
df20r29	0	0.45	1.22
df20r30	-2	0.57	1.56
df20r31	-4	0.60	1.62
df20r32	-6	0.48	1.29
df20r33	-8	0.49	1.32
df20r34	10	0.41	1.12

Table 5.15 Results of Horizontal Source and Detector Scan at L = 163

Run #	Position (in.)	$\text{Ln} (I / I_0)$	Thick (in)
df20r35	10	0.34	0.93
df20r36	8	0.52	1.41
df20r37	6	0.68	1.84
df20r38	4	0.60	1.63
df20r39	2	0.67	1.83
df20r40	0	0.59	1.59
df20r41	-2	0.73	1.98
df20r42	-4	0.74	2.02
df20r43	-6	0.70	1.89
df20r44	-8	0.66	1.80
df20r45	10	0.74	2.02

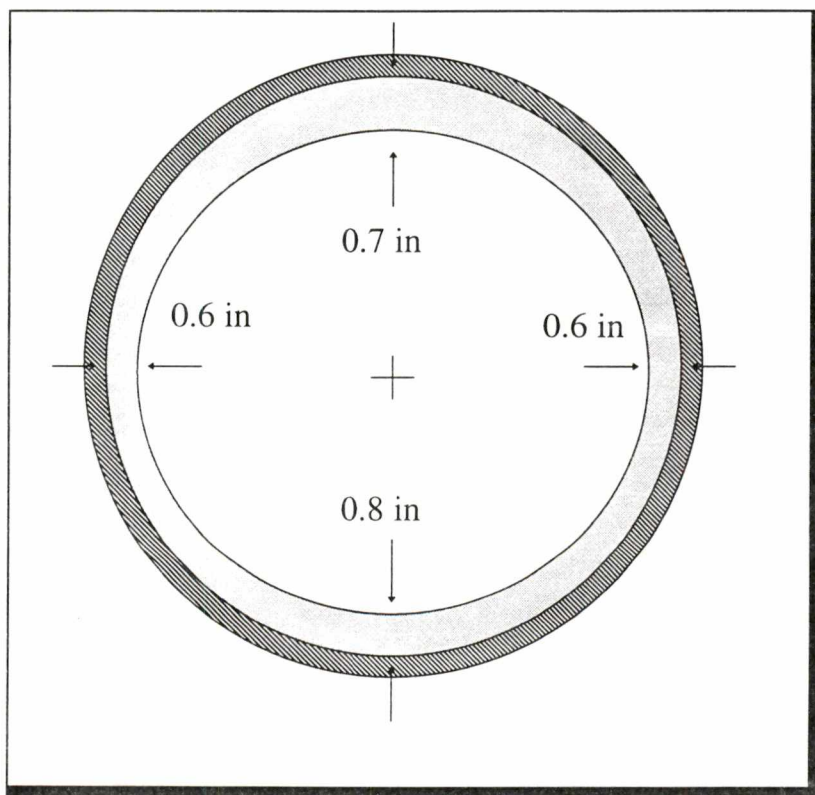


Figure 5.8 Deposit Profile at L = 163

5.4 H/U Ratio

As previously explained, a minimum of 1.25 in. of material is needed in order to obtain an accurate estimate of the H/U ratio. Using the data from source-detector horizontal and vertical scans near the midplane, 32 locations were identified that fit this criteria. H/U values as low as 0.5 and as high as 6.5 were calculated from these deposit profiles and gamma ray signatures. Averaging these data points, the average H/U ratio is determined to be 3.4 ± 0.25 where the uncertainty is the standard deviation of the mean. The measured values of H/U are given in Table 5.16, and their distribution is shown in Fig. 5.9.

5.5 Mass of Deposit

Because the total deposit mass strongly depends on the measured hydration and its relationship to the material density, the deposit volume is first calculated from the deposit profile. Using the estimated deposit profile at the six locations and linearly interpolating the shape between each point, the deposit volume is $195,000 \text{ cm}^3$. The average H/U value of 3.4 ± 0.25 corresponds to a material density of $5.02 \pm 0.19 \text{ g/cm}^3$, which yields a total deposit mass of $978 \pm 37 \text{ kg}$ of uranium. This value compares favorably with the results of the NDA gamma spectrometry, which estimated a deposit mass of $1190 \text{ kg} \pm 595 \text{ kg}$.

Table 5.16 Values of H/U from Measurements				
0.5	2.3	3.5	4.2	4.9
0.5	2.8	3.7	4.2	5.1
0.5	2.9	3.7	4.5	5.1
1.5	3	3.8	4.5	5.7
1.5	3	3.9	4.7	
2	3.3	4	4.8	
2.2	3.3	4	4.9	

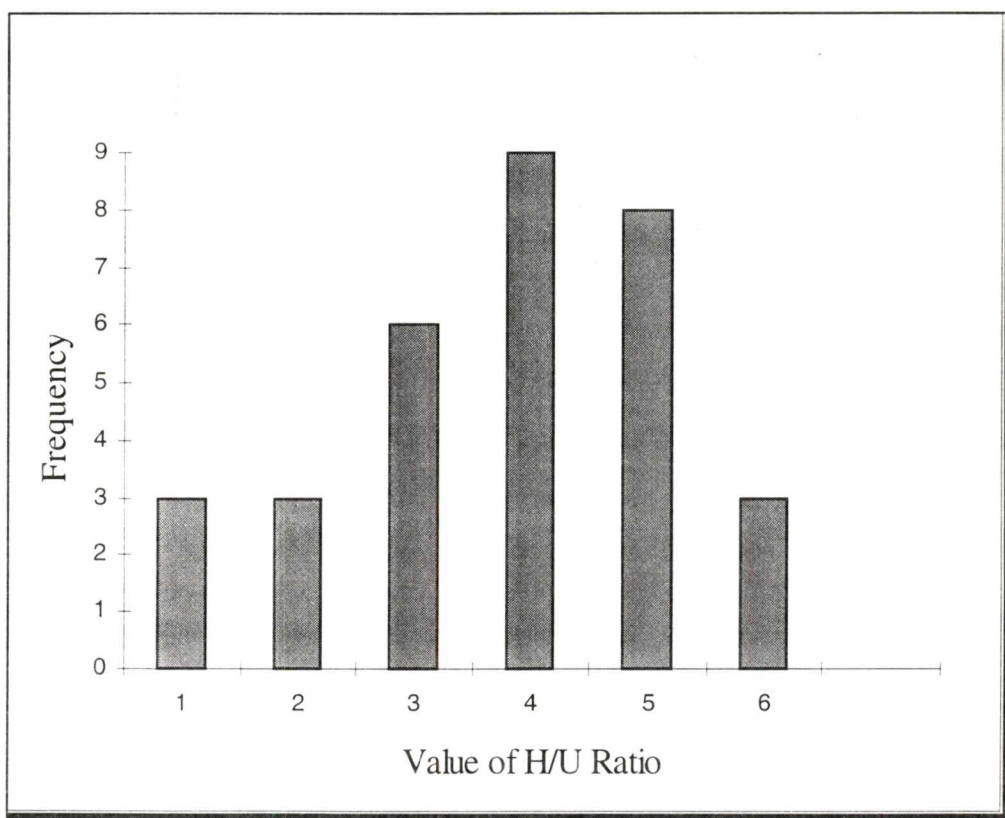


Figure 5.9 Distribution of H/U Ratio's from Measurement Data

5.6 Preliminary Results from Intrusive Look

In October 1997, an intrusive look into the K-29 Hockey Stick using a fiber optic camera was performed by the K-29 Deposit Removal Project Team, which has overall responsibility for mitigating and removing the Hockey Stick deposit. The location selected for the intrusive look was based on the results of this work. Preliminary results from the intrusive look indicate that (1) the deposit has formed on the top of the pipe in the $L = 40$ to $L = 80$ region, (2) the deposit between $L = 40$ and $L = 80$ is not smooth but consists of large closely grouped nodules, (3) the apparent material distribution throughout the pipe closely agrees with the measurements, and (4) based on the color of the deposit, which are green to green-yellow and brown, estimates of the H/U ratio were between 3.0 and 3.5. Figures 5.10 - 5.12 are video stills taken from inside the Hockey Stick, all in the lower horizontal region between $L = 40$ and $L = 80$. Note that the orientation of the picture in Figure 5.10 is rotated approximately 45 degrees to the left.

The nodules that exist in the region between $L = 40$ and $L = 80$ are responsible for the somewhat erratic behavior of the measured thicknesses in this region. Slight changes in the source-detector position can dramatically alter the amount of material between the source and detector, causing the noted perturbations. The fiber optic camera did show that the deposit profile becomes more smooth and symmetric up the riser, correlating the measured data. Based on results from the intrusive look, the Deposit Removal Project Team has concluded that our measurements closely correlate the actual deposit thickness, although no absolute measurements are yet available. It is hoped that there will be a possibility to acquire this data once actual removal of the deposit begins.

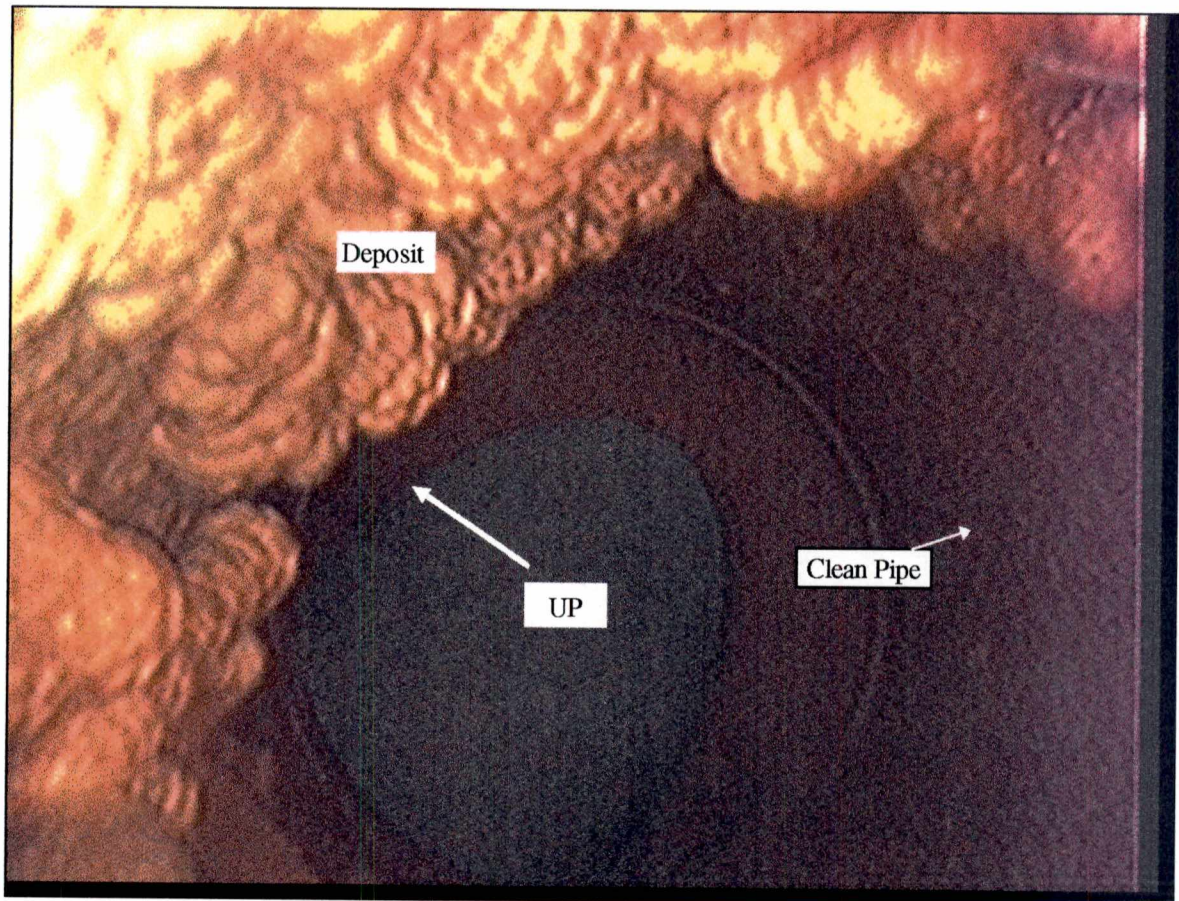


Figure 5.10 Intrusive Look Into the Hockey Stick in the Region at $L = 40$ to $L = 80$

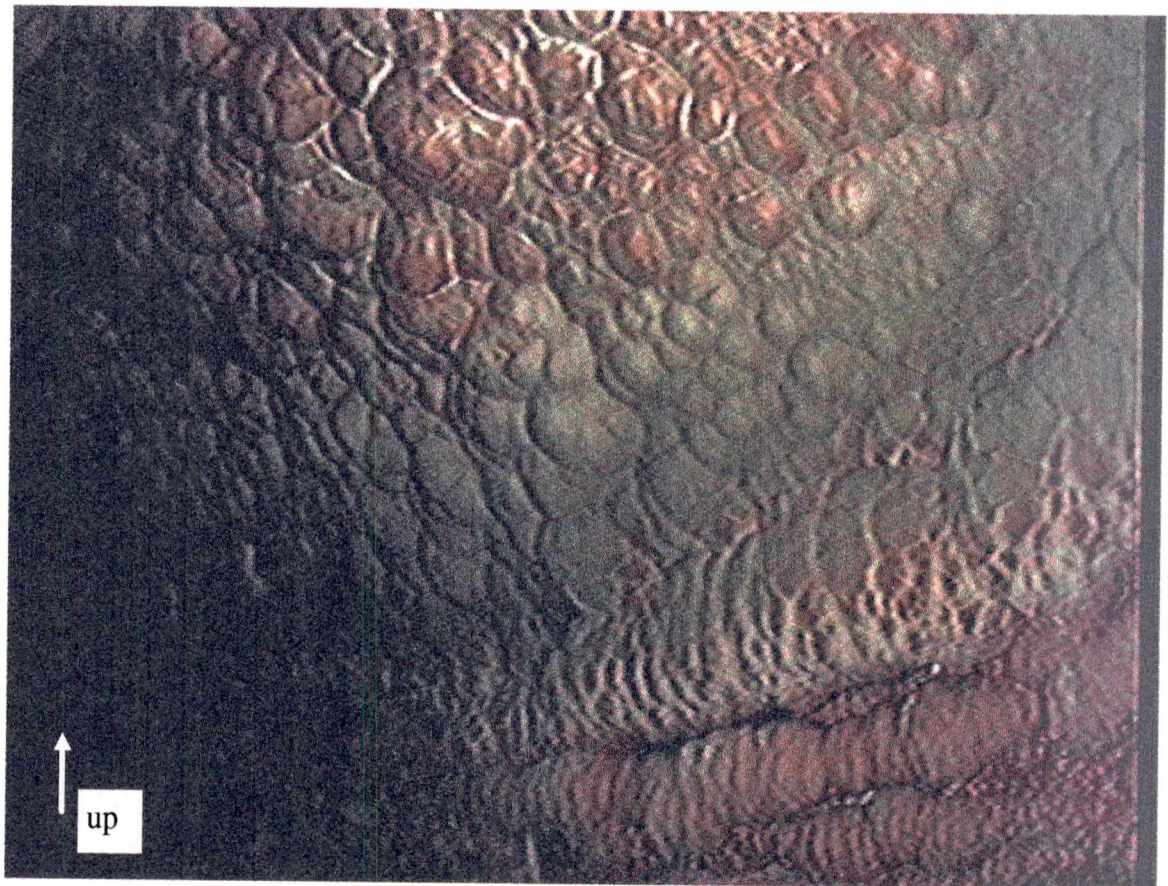


Figure 5.11 Deposit Nodules Formed on the Upper Hemisphere of the Pipe

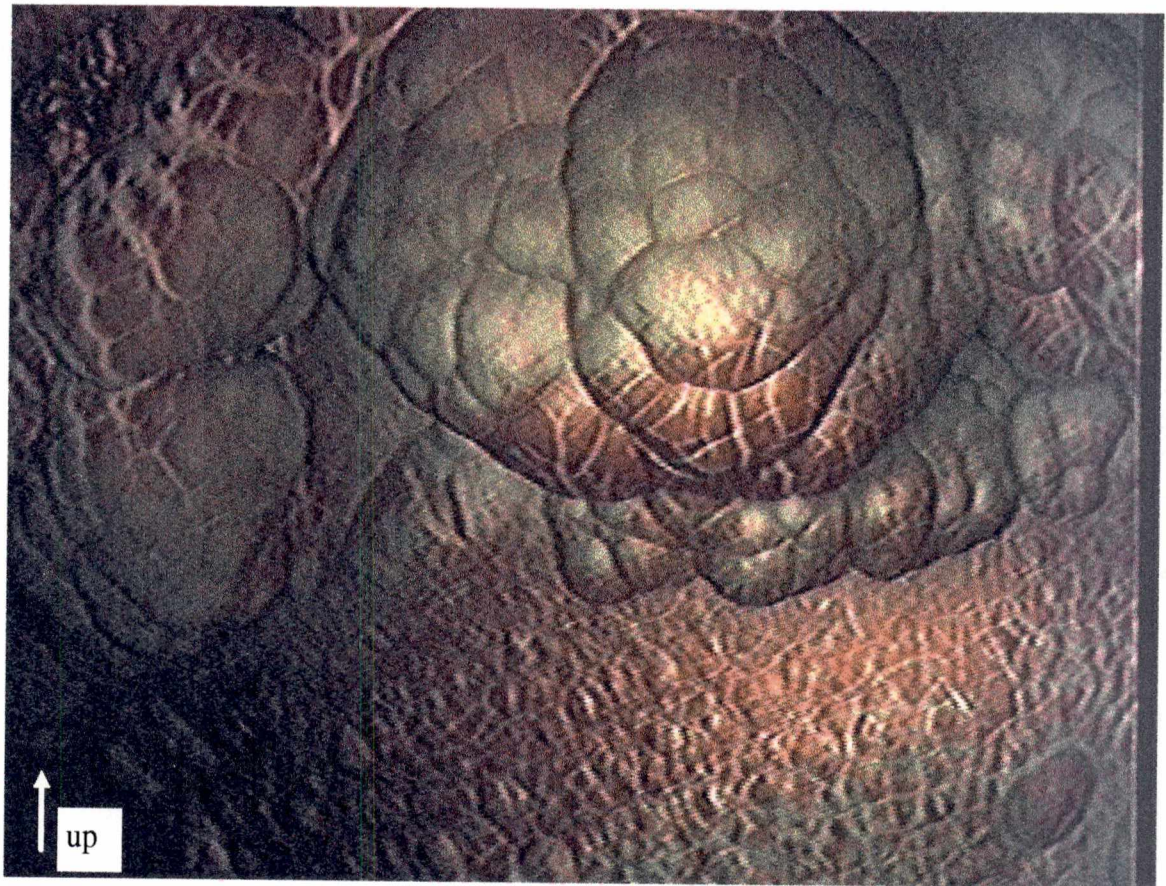


Figure 5.12 A Large Nodule Believed to Exist Close to $L = 70$

CHAPTER 6

CONCLUSIONS AND RECOMMENDATIONS FOR FUTURE WORK

6.0 Summary

The K-29 Hockey Stick deposit is a significant criticality safety concern because perturbation of any single parameter, coupled with the pipe geometry and deposit mass, enrichment, or hydration could lead to a criticality event. The Nuclear Weapons Identification System (NWIS) that is based on the ^{252}Cf -source-driven noise analysis techniques of Mihalcz and Pare, provides a portable platform that was used to assess and accurately determine the deposit characteristics.

The dual particle ^{252}Cf source essentially yields two independent and simultaneous measurements used to characterize the deposit. The first measurement is a transmission measurement using high energy fission neutrons (essentially neutron imaging), which can be shown to be independent of the deposit hydration and insensitive to geometrical effects of the deposit location with respect to the source and detectors. The second measurement is a gamma ray transmission measurement (gamma ray imaging), whose characteristics are dependent on the deposit hydration and thickness. Using the deposit thickness from the first measurement, the second measurement is then used to obtain the deposit hydration.

By performing synchronized source and detector scans along and around the pipe axis, the spatial distribution of the deposit was determined. Once the distribution, total mass, and hydration of the deposit was known, planners were able to decide on the best course of action for removing the deposit and any required mitigating actions that may be needed.

Measurements and their analysis predicted a deposit volume of $195,000 \text{ cm}^3$, corresponding to a total deposit mass of $978 \pm 37 \text{ kg}$ of uranium based on an averaged H/U value of 3.4 ± 0.25 . Data analysis also predicted that the deposit had adhered to the top of the pipe in the $L = 40$ to $L = 80$ region, and was verified by an intrusive look into the pipe with a fiber optic camera. Additionally, at the time of the intrusive look, estimates of the H/U ratio based on the color of the deposit were 3.0 to 3.5 which closely correlates the measured results.

These measurements have shown that this method is applicable to a variety of other uranium deposits that might exist at an enrichment facility, operating or not, such as deposits existing in freezer-sublimers, process gas coolers, or UF_6 cylinders.

6.1 Recommendations for Future Work

This work only used a small fraction of the available data from these measurements. More information could possibly be extracted by looking at other information in the neutron signatures besides that from the high energy neutron data; analyzing information from secondary gamma production; or even trying to extract resonance information from the neutron signatures.

The methods developed here were previously unrealized, and may be applicable to a wide variety of other systems. Investigation into the potential applicability of NWIS methodology for use in other non-intrusive material characterizations should be evaluated. Additionally, a method to quickly reconstruct a 2 or 3 dimensional image would be a very effective tool in any future characterization work, and should be evaluated.

References

1. Gosling, F. G., "The Manhattan Project: Science in the Second World War," U. S. Department of Energy, Washington D. C., June 1992.
2. Munger, F., DOE says plan to clean up K-25 to save money, create 500 jobs. The Knoxville News-Sentinel, (1996, October 31), p. A5.
3. Cochran, R. G. and Tsoulfanidis, N., The Nuclear Fuel Cycle: Analysis and Management, American Nuclear Society, La Grange Park, IL., 1990.
4. Russell, R. G., "Investigation of Hygroscopic Properties of Uranyl Fluoride," Paducah Gaseous Diffusion Plant Document No. KY/L-1699 (November 1994), as given in "Relative Risk of Nuclear Criticality Occuring from LEU and HEU Deposits at the K-25 Site," K/ER-215, by Haire, M. J., January 1995.
5. Hannon, Terry F. K-29 Deposit Removal Team Engineer, personal communication, May 1997.
6. Haire, M. J. and Jordan, W. C., "Nuclear Criticality Evaluations of a Uranium Deposit in the K-29 Building," Martin-Marietta Energy Systems Inc., Oak Ridge National Laboratory, internal memo.
7. Jordan, W. C. and Turner, J. C., "Minimum Mass of Moderator Required for Criticality of Homogeneous Low-Enriched Uranium Systems", ORNL/CSD/TM-284, Martin-Marietta Energy Systems, Inc., Oak Ridge National Laboratory, December 1992.
8. Reilly, Ensslin, and Smith, Passive Nondestructive Assay of Nuclear Materials, NUREG/CR-5550, United States Nuclear Regulatory Commission, Washington, DC, 1991.
9. Pare, V. K., and Mihalcz, J. T., "Reactivity from Power Spectral Density Measurements with Californium-252," *Nucl. Sci. and Eng.*, **56**, 213, (1975).

10. Mihalcz, J. T., Blakeman, E. D., Reagan, G. E., Johnson, E. B., and Hachiya, Y., "Dynamic Subcriticality Measurements Using the ^{252}Cf -Source-Driven Noise Analysis Method," *Nucl. Sci. Eng.*, **104**, 314 (1990).
11. 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.
12. "American National Standard for Nuclear Criticality Safety in Operations with Fissionable Materials Outside Reactors," ANSI/ANS-8.1-1983, American Nuclear Society, La Grange Park, Illinois (1983).
13. "SCALE: A Modular Code System for Performing Standardized Computer Analysis for Licensing Evaluation," NUREG/CR-0200, Vol. II, Oak Ridge National Lab., U. S. Nuclear Regulatory Commission (Jan. 1990).
14. Raffety, S. J. and Mihalcz, J.T., "Homogeneous Critical Assemblies of 2 and 3% Enriched Uranium in Paraffin," Y-DR-14, Union Carbide Corp., Nucl. Div., Y-12 Plant, 1969.
15. Paxton, H. C. and Pruvost, N. L., "Critical Dimensions of Systems Containing ^{235}U , ^{239}Pu , and ^{233}U (1986 Rev)," LA-10860-MS, Los Alamos National Laboratory, 1987.
16. Rose, P. F., *ENDF - 201, ENDF/B-VI Summary Documentation*, BNL-17541, Brookhaven National Laboratory, 1991.
17. National Nuclear Data Center at Brookhaven National Laboratory, Upton, N.Y., <http://www.nndc.bnl.gov>, maintained by Thomas W. Burrows (nndctb@bnl.gov).
18. Chilton, A., Shultis, J., and Faw, R., Principles of Radiation Shielding, Prentice-Hall, 1984.
19. Ordnoff, J. D., "Prompt Neutron Periods of Metal Critical Assemblies," *Nucl. Sci. Eng.*, **2**, 450 (1957).

20. Mihalcz, J. T., "The Use of ^{252}Cf as a Randomly Pulsed Neutron Source for Prompt Neutron Decay Measurements," *Nucl. Sci. Eng.*, **41**, 296 (1970).
21. Gozani, T., Active Nondestructive Assay of Nuclear Materials - Principles and Applications, NUREG/CR-0602, U.S. Nuclear Regulatory Commission, 1981.
22. T. E. Valentine and J. T. Mihalcz, "MCNP-DSP: A Neutron and Gamma Ray Monte Carlo Calculation of Source-Driven Noise-Measured Parameters," *Annals of Nuclear Energy* Vol. 23, No. 16, 1271-1287 (1996).
23. Valentine, T. E., "MCNP-DSP: A Neutron and Gamma Ray Monte Carlo Calculation of Source-Driven Noise Measured Parameters," Ph.D. Dissertation, Univ. of Tennessee, 1995.
24. Briesmeister, J. F., Ed., "MCNP-4a : A General Monte Carlo N-Particle Transport Code," LA-12625-M, Los Alamos National Laboratory, 1993.
25. T. E. Valentine and J. T. Mihalcz, "Validation of the Monte Carlo Code MCNP-DSP," *Annals of Nuclear Energy* 24, No. 2, 79-98 (1997).
26. Valentine, T. E., Mihalcz, J. T., and Koehler, P. E., "Calculated NWIS Signatures for Enriched Uranium Metal," *Proceedings of the 36th Annual Meeting of Institute of Nuclear Materials Management*, Palm Desert, Ca., Vol. XXIV, 88-393 (1995).
27. Mihalcz, J. T., Valentine, T. E., and Mattingly, J. K., "Feasibility of Subcriticality and NDA Measurements for Spent Fuel by Frequency Analysis Techniques with ^{252}Cf ," *Proceedings of the Topical Meeting on Nuclear Plant Instrumentation, Control, and Human-Machine Interface Technologies*, University Park, Pennsylvania, American Nuclear Society, Vol. II, 883-891 (1996).
28. Manhart, W., "Evaluation of Cf-252 Fission Neutron Spectrum Between 0 MeV and 20 MeV," *Proc. Advisory Group Mtg. Neutron Sources*, Leningrad, USSR, 1986 (IAEA-TECDOC-410), Vienna, 1987.
29. Maienschein, F. C., Peelle, R. W. and Love, T. A., *Neutron Phys. Ann. Prog. Rep. Sep. 1*, 1958, ORNL-2609, Oak Ridge National Laboratory.

Appendices

Appendix A: Energy Distribution of Neutrons from ^{252}Cf Fission

Only about 3 percent of the californium nuclei undergo spontaneous fission, producing on average 3.773 prompt neutrons per fission that have a spectrum similar to ^{235}U but slightly harder, with the energy spectrum given by a modified Maxwellian

$$F(E) = \frac{2}{\sqrt{\pi}} \frac{\sqrt{E}}{T^{3/2}} e^{-E/T} R(E) \quad (\text{A.1})$$

where E is the neutron energy and T is the neutron temperature 1.42 MeV.^{21, 23} The energy dependent correction factor $R(E)$ was proposed by Manhart²⁸ in order for the ^{252}Cf fission spectrum to agree with experimental data. Valentine²³ developed a functional form for this correction factor using least squares regression analysis; for energies less than 5.0 MeV the correction factor takes the form

$$R(E) = 0.955 + 0.0707 E - 0.0444 E^2 + 0.0199 E^3 - 0.00457 E^4 + 0.000368 E^5 \quad (\text{A.2})$$

and for energies greater than 5.0 MeV the correction factor is given by

$$R(E) = 1.16 - 0.0432 E + 0.00185 E^2 - 3.16 \cdot 10^{-5} E^3. \quad (\text{A.3})$$

This corrected Maxwellian distribution results in an average fission neutron energy of 2.13 MeV, which agrees with the measurement.

Appendix B: Energy Distribution of Gamma Rays from ^{252}Cf Fission

The gamma ray multiplicity for ^{252}Cf has an average value of 7.79, which corresponds approximately to two photons per californium neutron released from fission.^{21, 23} The measured ^{252}Cf gamma ray spectrum from fission is nearly identical to the gamma ray spectra from the thermal neutron fission of ^{235}U . Valentine fitted²³ the prompt fission gamma ray spectrum below 0.3 MeV by

$$G(E) = 38.13(E-0.085) e^{1.648E}. \quad (\text{B.1})$$

Based on Maienschein's measurements²⁹ of the ^{235}U spectra, the rest of the ^{252}Cf gamma ray spectrum is the spectrum is given by

$$G(E)=26.8e^{-2.30E} \quad (\text{B.2})$$

in the interval 0.3 to 1.0 MeV, and

$$G(E)=8.0e^{-1.10E} \quad (\text{B.3})$$

from 1.0 to 8.0 MeV The average gamma ray energy per fission from the distribution is 0.898 ± 0.01 MeV, agreeing with the accepted value of 0.88 ± 0.01 MeV.²⁹

Appendix C: Least Squares Polynomial Fit for Calculated Hydration

The measured gamma response is a function of both thickness and density, which is closely related to hydration. The calculated gamma response of Eq.3.10 was determined for 14 thicknesses, ranging from 1 cm to 7.5 cm in 0.5 cm increments, for nine levels of hydration ($H/U = 0.15, 0.30, 0.61, 1.5, 3.0, 4.6, 6.1, 9.1, \text{ and } 15.2$). In order to speed up and better refine the spatial portion of the calculation, a least squares polynomial fit was calculated for each hydration level over the range of 1 to 7.5 cm. The coefficients for each expression are given in Table A.1, and correspond to

$$A_0 + A_1 X + A_2 X^2. \quad (\text{C.1})$$

Table C.1 Coefficients of Least Squares Polynomial Fit			
H/U	A_0	A_1	A_2
0.151	0.71139	-0.18602	0.01366
0.303	0.71461	-0.18607	0.01364
0.607	0.72099	-0.18613	0.01358
1.51	0.74039	-0.18591	0.01338
3.03	0.77340	-0.18436	0.01292
4.55	0.83506	-0.17598	0.01160
6.07	0.86852	-0.16815	0.01063
9.11	0.91293	-0.15219	0.00893
15.2	0.95741	-0.12426	0.00639

Appendix D: MCNP Modeling

K-25 Hockey Stick 1" Symmetric 3wt% U235 and H/X=20 (H/U=0.6077)

c One Time Domain Detector, Lithium glass removed

c

c =====Cell Cards=====

c

1	0	-1 +5 -6	imp:n,p=1 \$ inner pipe void
2	1 0.071289	+1 -2 +5 -6	imp:n,p=1 \$ DEPOSIT
3	2 -8.90	+2 -3 +5 -6	imp:n,p=1 \$ Ni lining
4	3 -7.82	+3 -4 +5 -6	imp:n,p=1 \$ Carbon Steel
5	4 -2.70	-7 +8 +13 -16 +19 -22	imp:n,p=1 \$ Al front
6	4 -2.70	+13 -14 -8 +10 +19 -22	imp:n,p=1 \$ Al L side
7	4 -2.70	-10 +11 +13 -16 +19 -22	imp:n,p=1 \$ Al back
8	4 -2.70	+15 -16 +10 -8 +19 -22	imp:n,p=1 \$ Al R side
9	4 -2.70	+14 -15 -8 +10 +19 -20	imp:n,p=1 \$ Al bottom
10	4 -2.70	+14 -15 -8 +10 +21 -22	imp:n,p=1 \$ Al top
11	5 -11.4	+12 -13 -7 +11 +19 -22	imp:n,p=1 \$ Pb left
12	5 -11.4	+16 -17 -7 +11 +19 -22	imp:n,p=1 \$ Pb right
13	5 -11.4	-7 +11 +12 -17 +22 -23	imp:n,p=1 \$ Pb top
14	5 -11.4	-7 +11 +12 -17 +18 -19	imp:n,p=1 \$ Pb bottom
15	6 -0.020	-8 +9 +14 -15 +20 -21	imp:n,p=1 \$ Foam Filler
16	7 -1.032	-9 +10 +14 -15 +20 -21	imp:n,p=1 \$ Det. Bicron 420
17	0	+4 +7 +5 -6 -24 +27 -26	imp:n,p=1 \$ Void arnd pipe
18	0	-7 +25 +5 -12 +27 -26	imp:n,p=1 \$ Void left
19	0	-7 +25 +17 -6 +27 -26	imp:n,p=1 \$ Void right
20	0	-11 +25 +12 -17 +27 -26	imp:n,p=1 \$ Void behind
21	0	+12 -17 -7 +11 +23 -26	imp:n,p=1 \$ Void above
22	0	+12 -17 -7 +11 -18 +27	imp:n,p=1 \$ Void below
23	0	-5:+6:+24:-25:+26:-27	imp:n,p=0 \$ Cosmos

c

c =====Surface Cards=====

```

1 cx 26.984
2 cx 29.52369
3 cx 29.52750
4 cx 30.48
5 px 0
6 px 337.185
7 py -30.5
8 py -30.625
9 py -31.325

```

10 py -40.485
 11 py -40.61
 12 px 162.4775
 13 px 163.1125
 14 px 163.2375
 15 px 172.7625
 16 px 172.8875
 17 px 173.5225
 18 pz -5.5225
 19 pz -4.8875
 20 pz -4.7625
 21 pz 4.7625
 22 pz 4.8875
 23 pz 5.5225
 24 py 50
 25 py -50
 26 pz 50
 27 pz -50

c

c =====Material Cards=====

c

m1	1001.50c	0.007326	8016.50c	0.027783	9019.50c	0.02412 &
	92235.50c	0.000366	92238.50c	0.011694		\$ UO2F2 - H2O
m2	28000.50c	1.0				\$ Nickel
m3	6000.50c	-0.005	26000.50c	-0.995		\$ Carbon Steel
m4	13027.50c	1.0				\$ Al
m5	82000.50c	1.0				\$ lead
m6	1001.50c	-0.041	6012.50c	-0.544	&	
	7014.50c	-0.121	8016.50c	-0.294		\$ Foam filler
m7	1001.50c	0.5238	6012.50c	0.4762		\$ Scintillation media

c

c

c =====MCNP-DSP Control Cards=====

mode n p
 idum 1
 dbcn 19333486399999 11j 1500000
 prdmp 1j -900
 files 21 stick.d s f 0
 22 d1320.t1 s f 0
 c ctme 2880

K-25 Hockey Stick 3wt% U235 and H/X=100 (c) (H/U=3.038)

c One Time Domain Detector, Disk of material at pipe wall

c

c =====Cell Cards=====

c

1	0		+1	-2	+5	-6		imp:n,p=1	\$ inner pipe void		
2	1	0.071289	-1	+28	-29			imp:n,p=1	\$ DEPOSIT		
3	2	-8.90	+2	-3	+5	-6		imp:n,p=1	\$ Ni lining		
4	3	-7.82	+3	-4	+5	-6		imp:n,p=1	\$ Carbon Steel		
5	4	-2.70	-7	+8	+13	-16	+19	-22	imp:n,p=1	\$ Al front	
• 6	4	-2.70	+13	-14	-8	+10	+19	-22	imp:n,p=1	\$ Al L side	
7	4	-2.70	-10	+11	+13	-16	+19	-22	imp:n,p=1	\$ Al back	
8	4	-2.70	+15	-16	+10	-8	+19	-22	imp:n,p=1	\$ Al R side	
9	4	-2.70	+14	-15	-8	+10	+19	-20	imp:n,p=1	\$ Al bottom	
10	4	-2.70	+14	-15	-8	+10	+21	-22	imp:n,p=1	\$ Al top	
11	5	-11.4	+12	-13	-7	+11	+19	-22	imp:n,p=1	\$ Pb left	
12	5	-11.4	+16	-17	-7	+11	+19	-22	imp:n,p=1	\$ Pb right	
13	5	-11.4	-7	+11	+12	-17	+22	-23	imp:n,p=1	\$ Pb top	
14	5	-11.4	-7	+11	+12	-17	+18	-19	imp:n,p=1	\$ Pb bottom	
15	6	0.081338	-8	+9	+14	-15	+20	-21	imp:n,p=1	\$ Foam Filler	
16	7	0.088	-9	+10	+14	-15	+20	-21	imp:n,p=1	\$ Det Bicron 420	
17	0		+4	+7	+5	-6	-24	+27	-26	imp:n,p=1	\$ Void arnd pipe
18	0		-7	+25	+5	-12	+27	-26		imp:n,p=1	\$ Void left
19	0		-7	+25	+17	-6	+27	-26		imp:n,p=1	\$ Void right
20	0		-11	+25	+12	-17	+27	-26		imp:n,p=1	\$ Void behind
21	0		+12	-17	-7	+11	+23	-26		imp:n,p=1	\$ Void above
22	0		+12	-17	-7	+11	-18	+27		imp:n,p=1	\$ Void below
23	0		-1	-2	-28					imp:n,p=1	\$ Void matl
24	0		-1	-2	+29					imp:n,p=1	\$ Void matl
25	0		-5	+6	+24	-25	+26	-27		imp:n,p=0	\$ Cosmos

c

c =====Surface Cards=====

```

1 c/y 168 0 5.5225
2 cx 29.52369
3 cx 29.52750
4 cx 30.48
5 px 0
6 px 337.185
7 py -30.5
8 py -30.625
9 py -31.325
10 py -40.15
11 py -40.275

```

12 px 162.4775
 13 px 163.1125
 14 px 163.2375
 15 px 172.7625
 16 px 172.8875
 17 px 173.5225
 18 pz -5.5225
 19 pz -4.8875
 20 pz -4.7625
 21 pz 4.7625
 22 pz 4.8875
 23 pz 5.5225
 24 py 50
 25 py -50
 26 pz 50
 27 pz -50
 28 py -29.0
 29 py -22.65

c

c =====Material Cards=====

c

m1	1001.50c	0.030651	8016.50c	0.035509	9019.50c	0.020183 &
	92235.50c	0.000307	92238.50c	0.009785		\$ UO2F2 - H2O
m2	28000.50c	1.0				\$ Nickel
m3	6000.50c	-0.005	26000.50c	-0.995		\$ Carbon Steel
m4	13027.50c	1.0				\$ Al
m5	82000.50c	1.0				\$ lead
m6	1001.50c	-0.041	6012.50c	-0.544	&	
	7014.50c	-0.121				\$ Foam filler
	8016.50c	-0.294				
m7	1001.50c	0.5238	6012.50c	0.4762		\$ Scintillation media

c

c

c =====MCNP-DSP Control Cards=====

mode n p
 idum 1
 dbcn 19333486399997 11j 1500000
 prdmp 1j -900
 files 21 stick.x s f 0
 22 corr3.t2 s f 0
 c ctme 2880

13. Appendix E: Additional Photographs of Hockey Stick Measurements

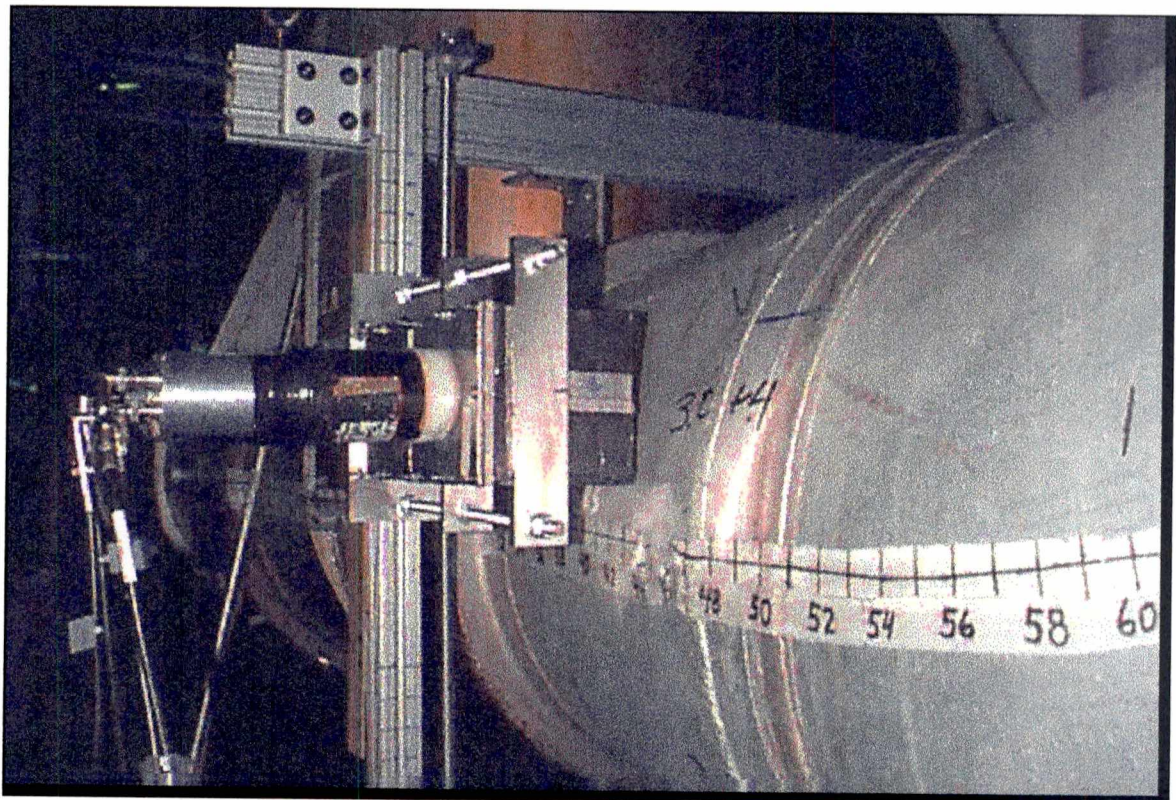


Figure E.1 Source and Detector Vertical Scan at $L = 40$

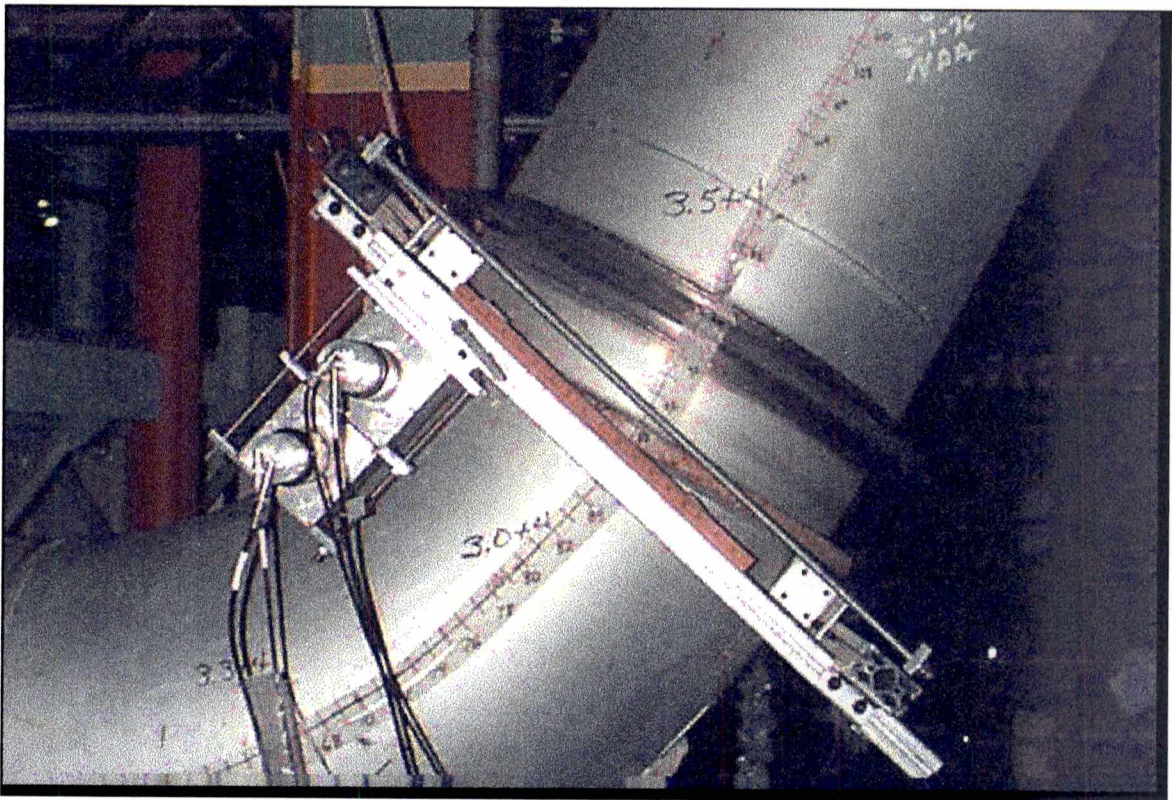


Figure E.2 Vertical Scan at $L = 80$, Complicated by Nearby Expansion Joint

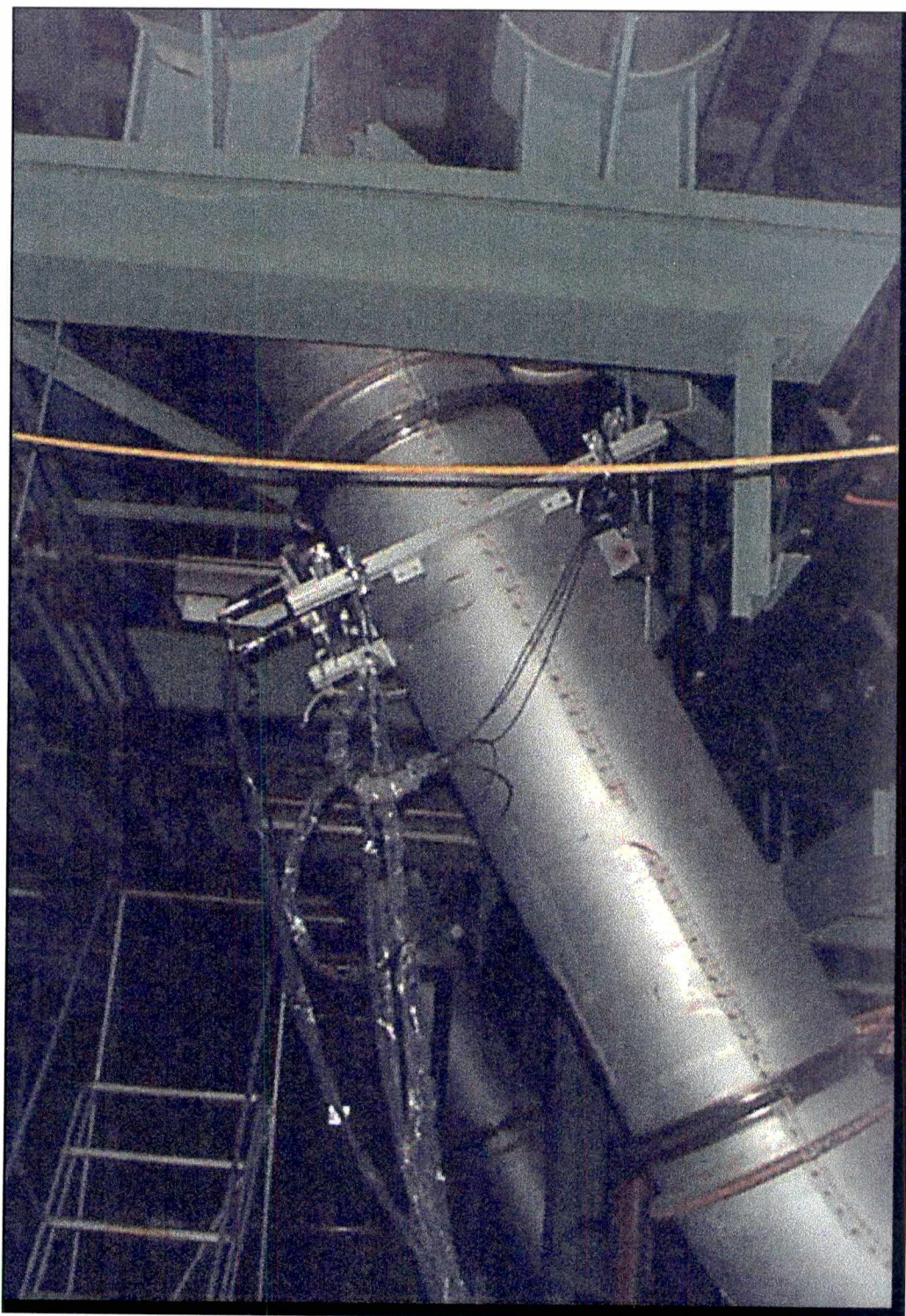


Figure E.3 Setup at L =163

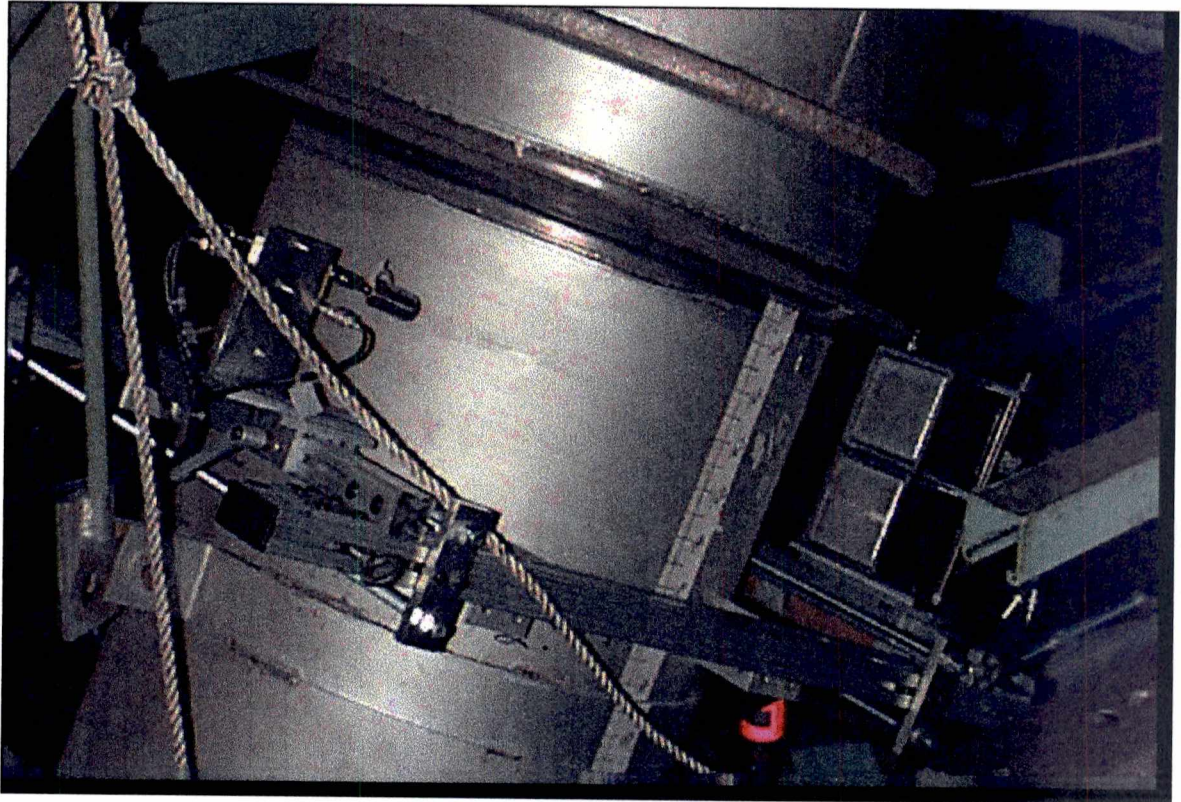


Figure E.4 Scan at $L = 163$, with Source and Detector in the -10 Position

VITA

Mark Stephen Wyatt was born in Falls Church, Virginia, on July 11, 1964. He graduated from Marshal County High School in Benton, Kentucky in 1982 and then joined the U. S. Navy. He served as a reactor mechanical operator onboard the USS Carl Vinson and received a honorable discharge in September 1988. He enrolled in Paducah Community College in January 1992 and transferred to The University of Tennessee, Knoxville in January 1994. In June of 1994 he married Kerry Elizabeth Evans. In May of 1996 he graduated *Magna Cum Laude* from The University of Tennessee, Knoxville with a Bachelor of Science in Nuclear Engineering. He expects to receive a Master of Science degree in Nuclear Engineering in December 1997 from The University of Tennessee, Knoxville and then begin work towards a Doctor of Philosophy degree in Nuclear Engineering.