**RESULTS OF THE REMOTE SENSING
FEASIBILITY STUDY FOR THE
URANIUM HEXAFLUORIDE STORAGE
CYLINDER YARD PROGRAM**

19980416 046

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RESULTS OF THE REMOTE SENSING FEASIBILITY STUDY FOR THE URANIUM HEXAFLUORIDE STORAGE CYLINDER YARD PROGRAM

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Executive Summary

The U.S. Department of Energy (DOE) manages the safe storage of approximately 650,000 tons of depleted uranium hexafluoride (UF₆) remaining from the cold war. This slightly radioactive, but chemically active, material is contained in more than 46,000 steel storage cylinders that are located at Oak Ridge, Tennessee; Paducah, Kentucky; and Portsmouth, Ohio.

Some of the cylinders are more than 40 years old, and approximately 17,500 are considered "problem cylinders" because their physical integrity is questionable. These cylinders require an annual visual inspection. The remainder of the 46,000-plus cylinders must be visually inspected every four years.

Currently, the cylinder inspection program is extremely labor intensive. Because these inspections are accomplished visually, they may not be effective in the early detection of leaking cylinders. The inspection program requires approximately 12–14 full-time-equivalent (FTE) employees. At the cost of approximately \$125K per FTE, this translates to \$1,500K per annum—just for cylinder inspection.

As part of the technology-development portion of the DOE Cylinder Management Program, the DOE Office of Facility Management (DOE/NE-40) requested the Remote Sensing Laboratory (RSL) to evaluate remote sensing techniques that have potential to increase the effectiveness of the inspection program and, at the same time, reduce inspection costs and personnel radiation exposure.

During two site visits (March and May of 1996) to the K-25 Site at Oak Ridge, Tennessee, RSL personnel tested and characterized seven different operating systems believed to detect leakage, surface contamination, thickness and corrosion of cylinder walls, and general area contamination resulting from breached cylinders. The following operating systems and techniques were used.

Laser-Induced Fluorescent Imaging (LIFI)

LIFI is a technique for analyzing the composition of a surface. Using spectral and temporal properties unique to the elements of a surface, it is often possible to detect contamination that may be invisible by other methods. Detection of uranium oxides with LIFI has been conducted for a number of years by personnel from the Bechtel Nevada Special Technologies Laboratory (STL). LIFI is advantageous because it is a chemical rather than a radiological probe, and measurements in the cylinder yards are unaffected by the background from the large amount of uranium stored nearby.

Measurements were conducted in the E, J, and K Yards at the K-25 Site on a series of valves, several breached cylinders, and a number of valve skirts and on concrete. To avoid limitations caused by solar background, operations were conducted at night. Several areas of suspected contamination were found. These areas fell roughly into three categories: (1) contaminated cement, (2) contaminated valves, and (3) contaminated cylinder scrapings. Samples of materials that showed evidence of contamination were collected and returned to the STL for further analysis. The results of these analyses indicated that the samples taken in the cylinder yards were contaminated with uranium.

It is estimated that one to five percent of all cylinders may be contaminated. Although the total amount of contamination on any given cylinder is small and virtually undetectable by means other than LIFI,

large-scale operations with many cylinders have the potential for producing substantial quantities of loose contamination. To optimize the LIFI system for routine use in the uranium-enrichment facilities, several developmental projects could be undertaken. First, incorporating commercially available technology may permit operation under daylight conditions. Second, the addition of a spark attachment to the LIFI system would provide detection sensitivities independent of the chemical nature of the element of interest. This technology may also have applications for detecting PCBs (polychlorinated biphenyls). Third, the LIFI could be repackaged to increase the system's portability and ease of use.

Long-Range Alpha Detection (LRAD)

The LRAD device detects ionization caused by alpha particles in the air rather than detecting the alpha particles directly. This device has been shown to be sensitive to the emission of a single alpha particle and to have a linear response up to very high activities that produce large inefficiencies in other types of detectors.

At the K-25 Site, the LRAD field team conducted searches for sources hidden in the cylinder yards and performed measurements on contaminated valves, contaminated pavement, and a leaking cylinder patch. In the source-search tests, the LRAD device was able to detect activities comparable to those expected from leaks. Within the high-background environment, the LRAD detected sources having activities twice its minimum detectable activity in favorable locations (a single isolated cylinder) and five times its minimum detectable activity in unfavorable locations. An actual leaking valve that produced an activity greater than the minimum detectable activity was easily observed during the field survey.

The LRAD "sniffer" offers several advantages for monitoring UF_6 storage cylinders for leaks: (1) it can be used by an operator in the narrow walkways between rows; (2) it can find leaks invisible to the naked eye; (3) it can monitor large areas more quickly than a normal alpha probe; and (4) it can monitor spaces not accessible to a normal probe. LRAD detectors may be useful in the uranium-enrichment facilities as surface monitors, airborne alpha monitors, and pipe monitors.

The field test revealed a need for further development to convert the proof-of-principle sniffer into a field-suitable device. This additional development should focus on increasing the practicality, reliability, and ease of use of the LRAD. The device should operate analogously to standard health physics instrumentation by incorporating an acoustic count-rate response and color-coded LEDs (light-emitting diodes) to simplify interpretation.

Neutron Activation Analysis

Neutron activation is a highly sensitive elemental-analysis technique. The basis of this method is that neutrons are captured in the material to be analyzed, and gamma rays are emitted. The rate of emitted gamma rays is directly proportional to the amount of material present. With a knowledge of the composition of the material (*i.e.*, the steel wall of a UF_6 storage cylinder) and the rate of emitted gamma rays, the thickness of the material can be determined.

The metallurgy of the UF₆ storage cylinders was investigated to determine the optimal isotopes to use in the analysis. Calculations were then made to design the measurements. Based on these calculations, a decision was made to proceed with bench testing using actual neutron sources. Subsequent laboratory measurements provided a correlation between the gamma-ray detection rate and the thickness of the cylinder-wall material. However, the results of the laboratory tests were not encouraging; the rate of gamma-ray emission was far too small for use in practical field work. Based on these results, it was decided not to proceed with field testing.

Differential Gamma-Ray Attenuation

High-energy gamma rays that pass through a given thickness of material are attenuated to a lesser degree than low-energy gamma rays. By measuring the ratio of the number of high-energy to low-energy gamma rays, the thickness of the intervening material (*i.e.*, the steel wall of a UF₆ storage cylinder) can be determined. Using this technique, cylinder-wall thicknesses were measured from the ratio of the number of two uranium gamma rays.

Six cylinders in the E and K Yards were analyzed using a collimated, high-resolution germanium detector; the wall thicknesses of these cylinders ranged from 250 to 625 mils. To obtain consistent results, it was found that the cylinders must be surveyed using similar detection geometries (*i.e.*, well below the UF₆ fill lines). The background count rates near the cylinders varied by nearly a factor of three and thus posed a complication to using this method. Therefore, it was necessary to perform additional measurements to correct for the background gamma rays emitted from neighboring cylinders. Within these constraints, cylinder-wall thicknesses could be measured to approximately five percent in 600 seconds. However, to measure the wall thickness to this precision, it was also necessary to have prior knowledge of the uranium isotopic fraction (the ratio of uranium-235 [²³⁵U] to uranium-238 [²³⁸U]) of each cylinder to a similar degree of precision.

A count time of 600 seconds is too long to be practical for a single measurement. To decrease the scanning time, the single high-resolution detector could be replaced by an array of low-cost, low-resolution detectors. While this developmental project shows promise, it should be given a lower priority than some of the other techniques that were investigated.

Compton Scatterometry

The Compton scatterometry technique relies on the detection of scattered gamma rays from an external source. A highly collimated detection system is employed to select a small volume of the material of interest for analysis. The material is scanned in depth by a motion of the detection system perpendicular to the surface of the material. Because the probability of scattering is determined by the density of the material, an interface between different materials, for example, between UF₆ and a steel cylinder, can be identified as the point where the number of scattered gamma rays changes significantly.

As a test of this technique, scattered gamma rays were detected using the system previously described in the differential gamma-ray attenuation studies. The scatter peak was identified, and the number of scattered gamma rays quantified to better than two percent in a measurement time of 600 seconds. Because the source used in this test was more than 10,000 times weaker than typical

sources employed in Compton scatterometry, much shorter measurement times can be expected with practical systems.

This technique holds promise as a simple, quick method to determine cylinder-wall thicknesses to accuracies approaching ten mils. A group from Los Alamos National Laboratory (LANL) with experience in using this technique is developing several follow-on ideas for hand-held wall-thickness gauges and computer-controlled robotic scanners.

Active Infrared (IR) Inspection

With this technique, the surface of the inspected material is heated using a brief pulse from a set of optical flash lamps. This flash heating generates a thermal pulse that propagates into the material. Discontinuities in the material cause a portion of the thermal energy to be reflected back to the surface. By monitoring the cooling rate at the surface, defects in the material can be identified.

Data were acquired on seven cylinders at the K-25 Site. While independent determinations of cylinder-wall thicknesses could not be obtained, the results were compared to quantitative ultrasound thickness measurements made at the site. In most cases, sufficient thermal contrast was obtained without prior preparation of the cylinders. Regions of corrosion were especially responsive to thermal pulses.

To perform quantitative measurements of cylinder-wall thicknesses using active IR imaging, a more detailed study of the temperature and time response of the cylinder walls is required. The use of a mobile, robotic-based system that allows access to the entire cylinder surface, including the underside, would minimize the time required to inspect a cylinder. For this and other future applications, augmenting the heating source would allow better resolution and an improved measuring capability.

Passive Thermal Infrared (IR) Imaging

Simply stated, passive thermal IR imaging detects the thermal energy emitted from a surface. Because the contents of the UF₆ storage cylinders produce heat and change temperature at a different rate than the cylinder and the environment, passive IR imaging is a non-contact method that provides information about both the cylinder material and the cylinder contents.

The commercial system with the highest degree of sensitivity available was leased for the field measurements at the K-25 Site. Thermal IR imagery was obtained on cylinders in the E, K, and J Yards. The results of the measurements indicated that quantitative information pertaining to the structural features of the cylinders was difficult to obtain; however, some information was obtained with this technology that could not be acquired with the other technologies. Passive thermal IR imaging was quite successful at quickly determining the fill level and the gross distribution of the UF₆ within the cylinders.

It may be possible to eventually determine fill volumes quantitatively using passive thermal IR imaging. To attain this goal, it will be necessary to model the heat-transfer processes more accurately and to perform a series of controlled measurements on filled and partially filled cylinders.

Conclusions

Several of the tested systems show great promise for use in routine monitoring operations in the cylinder yards: (1) LIFI and LRAD for the detection of surface contamination, (2) Compton scatterometry

for the measurement of cylinder-wall thicknesses, and (3) passive thermal IR imaging for the determination of fill levels and UF_6 distributions within the cylinders. In particular, the LRAD system is very near practical application and could be implemented with a small amount of further development. Differential gamma-ray attenuation and active IR inspection may prove useful following additional technical development. The neutron activation analysis technique shows little potential as an operational system.

Project Overview

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In early August 1995, representatives of the DOE/NE-40 contacted the DOE Nevada Operations Office (DOE/NV) to inquire whether problems relating to the storage and handling of depleted UF₆ might be characterized and monitored using the DOE/NV Remote Sensing Laboratory (RSL) assets. The DOE/NV site manager at the Eastern Measurements Office (EMO) of the RSL, located at Andrews Air Force Base in Washington, D.C., was instructed to host a meeting between DOE/NE-40 representatives and members of the Washington Aerial Measurements Operations (WAMO) scientific staff to determine whether such a collaboration was feasible. The WAMO staff operated the EMO for DOE/NV under EG&G Energy Measurements until their DOE contract expired on December 31, 1995, and has operated the EMO since then under Bechtel Nevada.

During the first part of the meeting held at WAMO on August 14, 1995, it was explained that the depleted UF₆ is collected as a result of the uranium-enrichment process that occurs in gaseous diffusion plants. Two gaseous diffusion plants are presently operating in Paducah, Kentucky, and Portsmouth, Ohio. A third plant was operational at the K-25 Site in Oak Ridge, Tennessee, until the mid-1980s. The depleted UF₆ is collected and stored in steel cylinders that are generally about 12 feet long and 4 feet in diameter. The cylinders are designed according to American National Standards Institute criteria (ANSI N14.1-1995) and Department of Transportation shipping regulations (Title 49 of the Code of Federal Regulations) to contain about 62 percent by volume of solid depleted UF₆—approximately 14 tons. Since the 1940s, approximately 47,000 of these cylinders have been accumulated in storage yards at the three sites. A portion of one Paducah, Kentucky, storage yard is shown in Figure A. The cylinders are stacked two-high in rows, generally on wooden or cement cradles supported by concrete foundations. In the past, the cylinders have been stacked directly on the ground.

On December 1, 1995, a more in-depth briefing and site visit was held in Paducah, Kentucky. Members of the RSL, WAMO, and STL gave presentations on potentially useful technologies. Based on the feedback from this meeting, a proposal was requested for a feasibility study. Funding for the proposal was approved for Bechtel Nevada by DOE/NE-40 in January 1996. In February 1996, work began in preparation for feasibility testing of five technologies:

- Laser-Induced Fluorescent Imaging (LIFI)
- Long-Range Alpha Detection (LRAD)
- Neutron Activation Analysis
- Active Infrared (IR) Inspection
- Passive Thermal Infrared (IR) Imaging

The feasibility study was conducted in the cylinder yards at the K-25 Site in Oak Ridge, Tennessee, during two site visits. The first occurred March 25–29, 1996. During this time, the LIFI technique was demonstrated successfully by STL as a tool used in the detection of surface contamination. Another group from RSL made passive thermal IR images of the cylinders and were able to semi-quantitatively

FIGURE A. CYLINDER STORAGE YARD AT PADUCAH, KENTUCKY

determine the fill levels of the UF_6 within the cylinders. Bechtel Nevada subcontracted Thermal Wave Imaging, Inc., (TWI) to use active thermal IR interrogation to determine structural integrity of the cylinder walls. The results from TWI were quite limited due to power constraints.

Prior to the second site visit, the WAMO staff investigating neutron activation analysis as a means to measure cylinder-wall thickness determined that this technique is not currently applicable for field measurements. Based on calculations and controlled laboratory measurements, it was determined that the neutron flux required to make a thickness measurement in a reasonable amount of time would lead to excessive radiation fields in the vicinity of the cylinder. The expense required to shield the high-radiation field or to enable a fast measurement using a smaller source is excessive compared to other techniques already in use. Therefore, the WAMO staff chose to abandon this effort. However, while investigating the neutron activation analysis technique, the WAMO staff determined that two other methods appeared promising:

- Differential Gamma-Ray Attenuation
- Compton Scatterometry

Both methods have been used to measure thickness, and they were easy to test. DOE/NE-40 gave permission for this change in the original scope of work.

Both the differential gamma-ray attenuation and the Compton scatterometry techniques were tested during the second site visit in May 1996 to the K-25 Site with existing equipment available to the WAMO

personnel. The differential gamma-ray attenuation technique was demonstrated to work within the required parameters, but there were issues concerning variability in background rates and the complexity and expense of a detector array required for such a system. The Compton scatterometry technique was tested using a method that members of the NIS-5 group at LANL had suggested. The WAMO group conducted the simple test and verified that the technique could be utilized in the ambient radiation fields of the cylinder yards in a stand-off mode (*i.e.*, the unit was not required to touch the cylinder). The positive results of this test have lead the NIS-5 group to begin prototype designs for both a hand-held thickness meter and a cylinder-imaging device.

The NIS-6 group from LANL was present during the May 1996 site visit to test LRAD as a means to detect the presence of leaks in the cylinder walls (breaches) or leaking cylinder valves through the alpha radiation signature of the exposed uranium compounds. The NIS-6 prototype was successfully demonstrated for locating sources that simulated cylinder breaches. In addition, LRAD was shown to outperform a standard alpha meter in locating alpha activity in the high backgrounds of the cylinder yards and in detecting contamination on irregular surfaces.

In this report, the scientific and technical details of each sensor technology and its application in the cylinder yards are described. While each technique that was demonstrated in the field was successful in some way, the following paragraphs describe the specific recommendations for further development. These recommendations are based on the performance of the sensors demonstrated in the field as well as the probability for a timely, successful, and cost-effective application of the technology in the UF₆ storage cylinder yard program. The priorities communicated by DOE/NE-40 have also been considered. The recommendations, with the highest priority listed first, are as follows:

1. The LRAD prototype should be developed into a fieldable unit suitable for use by cylinder yard technicians to search for breached cylinders. An additional LRAD model should be built specifically to detect leaking cylinder valves. LRAD is inexpensive, effective, and operates similarly to ordinary health physics meters that technical personnel are already trained on. This technology is mature enough that LRAD could be incorporated into the daily operations of the cylinder yards within a year following the commitment of funding.
2. Compton scatterometry should be prototyped and field tested for use as a thickness gauge. This technique shows the most promise for a consistent, cost-effective means for measuring the steel cylinder thickness averaged over a given area. The system can be designed to measure through direct contact with the cylinder or with some stand-off built-in. The presence of rust flakes or moisture on the surface has minimal effects on the thickness measured. No surface preparation or coupling agent is required as in the case of ultrasonic thickness gauges. The only requirement for the successful application of this technology is that the specifications of the measurement—particularly the resolution in thickness measured over unit area—must be defined before the final design of the system can be completed.
3. The LIFI system—optimized for detecting surface uranium contamination in the cylinder yards—should be tested once again. With the samples brought back from the cylinder yards and tested in the laboratory, the LIFI scientists and technical staff were able to obtain a much better understanding of the cylinder yard environment; hence, a better knowledge of the optimal settings of the LIFI system. Another field test would enable them to verify their findings and design a system that could come into routine use in the cylinder yards. Technicians could use LIFI to

certify that a cylinder is free from contamination for shipping, for painting, and for other movement and handling.

4. The passive thermal IR imaging technique should be studied further for its application in quickly determining cylinder fill levels. With the purchase of the appropriate commercially available thermal imaging camera and analysis software, a systematic study of cylinder fill levels could be carried out to determine how accurately and precisely the imaged fill line can be correlated to the actual fill volume of the cylinders. The same system could be used to image cylinders as they cool after they are filled or as the material redistributes itself after cooled cylinders are rotated to learn more about the properties and behavior of the UF_6 inside the cylinder.

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Chapter 1. Laser-Induced Fluorescence Imaging

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1.1 INTRODUCTION

Laser-induced fluorescence imaging (LIFI) is a technique used for imaging the fluorescent emissions from a surface. Using fluorescent spectral and temporal properties unique to the different chemicals that make up the surfaces that we can see, it is often possible to detect distributions or images that are invisible to the naked eye. This phenomena has been used for many years in the mining industry to differentiate minerals. Ultraviolet lamps are frequently referred to as "mineral lights."

LIFI detection of uranyl oxides has been studied by the Special Technologies Laboratory (STL) for a number of years. Support for these efforts has come primarily from the DOE Environmental Remediation and Waste Management program (DOE/EM-50) Office of Science and Technology (OST), Characterization Monitoring and Sensor Technology-Consulting Program (CMST-CP). Instrumentation development, field demonstration, and problem study have been the primary foci of this program. The LIFI system has previously been fielded at the K-27 Building at the Oak Ridge K-25 gaseous diffusion plant as part of a joint effort between CMST-CP and DOE/EM-40.

Based upon results obtained in the DOE/EM-50 program, additional support was made available by the DOE Office of Facility Management (DOE/NE-40) to test technology developed at several Oak Ridge uranium hexafluoride (UF_6) cylinder storage yards.

Fluorescence is a tool. When used in the proper environment with the proper background study, it can be very useful in locating contamination signatures that would otherwise not be visible. Several fundamental differences exist between LIFI detection of uranium and other, more conventional, radiological techniques. First, LIFI is strictly a surface probe. The absorption coefficient of uranyl salts is sufficiently large that laser-penetration depths are on the order of hundreds of microns. This means that, unlike radiological measurements, LIFI is unaffected by the background created by the large amount of uranium present in the storage yards. Conversely, if the contamination is obscured by corrosion or paint, it will not be detectable. Secondly, LIFI can produce a high-resolution image of surface contamination. The imagery allows a technician to determine the exact location of the contamination in real time.

Small shifts in the spectral content of uranium emission do occur with atomic mass. However, these changes are so slight that they are only detectable when uranium is in the gas phase, and vibrational broadening is greatly reduced. The fluorescence of uranium in the solid state is virtually unaffected by the isotopic content of the nucleus. LIFI is therefore not appropriate for measuring the degree of product enrichment.

It is possible to have both false negative and false positive results with LIFI. Laser-induced fluorescence results from the decay of electrons promoted to an excited molecular or atomic state by the absorption of a laser photon. Uranium fluorescence is sensitive to the oxidation state of the material. Therefore, the chemical environment of the contamination has a major impact on the sensitivity of this technique. It is possible that the chemical environment of the target species completely quenches the uranium emission to the point that is not detectable above other background signatures. It is therefore possible to have false negative results.

Background interferences for LIFI signatures do exist. The technique that was used isolates the time-domain and spectral-domain fluorescence emission signatures of the target species, in this case, the uranyl (+6) cation (see Section 1.2.1 for details). There are, however, other materials in nature with fluorescent emissions that overlap the target species, producing possible interference. It is therefore possible to have false positives.

In spite of the limitations described in the preceding paragraphs and with proper understanding and application, LIFI can yield a data product unlike any other. High-resolution imagery of the location and distribution of contamination can prove invaluable to the cleanup, remediation, and documentation efforts of health physics professionals.

1.2 EQUIPMENT DESCRIPTIONS

1.2.1 Laser System

The primary tool used in this study was a portable laser-video system developed by STL for use in uranium decontamination and facility-decommissioning efforts (Figure 1). Developmental support was provided by DOE/EM-50 with supplemental support from the Oak Ridge K-25 gaseous diffusion plant under the DOE/EM-40 decontamination and decommissioning effort.

The laser system was portable, generally run on a single 24-volt, gelled-cell battery. The laser was a frequency-tripled YAG:Nd, solid-state laser with a nominal output power of 30 millijoules per pulse at 355 nanometers. The pulse repetition rate was ten hertz. The laser output beam was coupled to a special projection optic through a fiber-optic array. The projection optic was mounted to and boresighted with the video camera. The projection optic included a quartz mixing-rod element that matched the laser field of view to the field of view sensed by the video camera. The microchannel-plate-intensified camera was commercially available, manufactured by Xybion Corporation. Wavelength discrimination in the form of a 60-nanometer FWHM (full width at half maximum) band-pass filter enhanced the uranium signature by passing photons coincident with the primary uranium fluorescent band.

Figure 2 shows a timing diagram for the data acquisition of the uranium fluorescent imagery. The laser was running at ten hertz. The video was operated in the interlaced mode with a frame rate of 30 hertz.

FIGURE 1. PORTABLE LIFI SYSTEM

FIGURE 2. LASER AND VIDEO CAMERA TIMING

Therefore, three video frames occurred for each laser pulse. On the first frame of the series, a laser-illuminated video image was digitally acquired. On the second frame of the series, the camera gate was widened and an ambient light-illuminated frame was captured. On the third frame of the series, the laser data was overlaid onto the ambient light background image and a composite picture presented to the operator.

Uranium detection was further enhanced by changing the phase of the gate time when the video camera was exposed. The gate timing for the video camera was set so that the shutter opened several hundreds of nanoseconds after the laser pulse. This "phase delay" permitted the prompt fluorescence from any organic background in the scene to decay away (Figure 3). Prompt background signatures observed at the K-25 storage cylinder yards included materials such as mosses, lichens, oils, papers, and bird droppings and other animal refuse. These interfering signatures generally have lifetimes on the order of nanoseconds to tens of nanoseconds. By delaying the opening of the shutter, the prompt interfering signatures have decayed away and only long-lived, generally inorganic emissions are detected.

FIGURE 3. SHUTTER TIMING FOR "PHASE DELAY"

Other longer-lived, phosphorescent species are also discriminated against by limiting the shutter exposure to the decay time of the uranium contamination, generally on the order of tens of microseconds to one millisecond. Many inorganic phosphorescent minerals exist in nature with decay times varying from milliseconds to hours. Because the rate at which materials emit photons is inversely proportional to the decay time, one can reduce the overall interference from long decay-time materials by limiting the duration of the shutter.

An example of the data-processing algorithm is shown in Figure 4. These data were taken at a hydrodynamic test facility at LANL. Two images are shown (Figures 4a and 4b). The first image (Figure 4a) was taken with laser illumination only with the shutter delayed relative to the laser pulse up to several hundred nanoseconds and open for a time on the order of the decay time of the target material. The second image (Figure 4b) was taken an hour earlier with the shutter open sufficiently long to permit ambient light exposure without the laser. This image was for registration of the fluorescent emission in the ambient light background. These two images were then overlaid using the following algorithm.

A mask was constructed using the target image (Figure 4c). This mask zeroed out the ambient image everywhere that the target image exceeded a preset threshold. A second mask was applied to the target image, zeroing out all pixels that fell below the same preset threshold. The two images were then normalized such that the background fell in the bottom half of a preset range while the target data resided in the top half. A color pallet was then constructed that showed grey scale for the bottom half of the range and a blue-to-red rainbow for the top half. The result of this processing is shown in Figure 5. For this image, an intensity-modulated red overlay was used for the target image. In subsequent images, the target is represented by a rainbow palette.

Due to the lack of knowledge about field conditions *a priori*, the laser energy was set to a maximum of three millijoules per pulse for the feasibility study of the K-25 Site. This amount of power was found to be below optimal levels, necessitating changes in the projection optics. In order to increase the illumination intensity, the size of the illumination spot was decreased to approximately 20 percent of the viewed spot. This recovered the incident intensity but reduced the effectiveness of the instrument as a survey tool. Furthermore, it limited the stand-off distance where STL personnel could obtain effective data.

By reconfiguring the present equipment, a maximum laser energy on the order of 60 millijoules per pulse can be obtained. With equipment configured for this power level, many of the aforementioned difficulties can be avoided.

1.2.2 Ultraviolet Flash Photography

In addition to the laser illumination measurements, data were collected using an ultraviolet (UV) xenon flash lamp and modified photography equipment. The Pyrex envelope was cut away from a commercially available xenon strobe lamp (Norman 400B). An array of customized UG-11 ultraviolet filters was fitted to the front of the strobe reflector to isolate the flash lamp's UV emission from the fluorescence of the visible target. This strobe was then used in conjunction with a conventional 35-millimeter camera and color film. A 400-nanometer cut-on filter, fitted over the camera lens, was used to block the filtered UV strobe emission while passing the red-shifted fluorescence as shown in Figure 6 (see page 6).

4a Uranium Fluorescence at Sunset. Camera gate delayed 100 ns after laser.

4b Solar-Lit View of Scene at Sunset. Note sun in upper left corner.

4c LIFI Image is Used to Create a Mask.

4d Mask is Subtracted from Background Image. LIFI image will be placed in scene as red.

FIGURE 4. DATA-PROCESSING SEQUENCE FOR COLOR OVERLAYS

The use of color film allowed some limited degree of spectral identification in the scene. Uranium emission appeared green while organic emission frequently appeared blue and mosses or lichens were either blue or red.

The use of the camera did not permit the temporal analysis as described in the laser section, except in a very limited way. The Nikon F-3 camera that was used would not synchronize with a flash at shutter speeds greater than 1/125 of a second. Furthermore, mechanical shutters are not precise enough in

FIGURE 5. EXAMPLE OF REGISTERED DATA

their fire time to allow the "phase delay" technique to work. Uranium detection done in this way will therefore suffer from a greater false-positive rate, as the temporal analysis feature of laser-based fluorescence detection is not possible with current technology.

This was STL's first attempt to use photographic procedures in the field, and the results appeared to be of limited value. However, further investigation may be warranted (see Section 1.5.2).

FIGURE 6. FILTERED FLASH LAMP AND 35-mm CAMERA

1.3 LIFI PROCEDURE AT THE K-25 SITE

While uranium fluorescence is routinely detected in the presence of indoor ambient light background, the performance of the system at long gate times was degraded by intense solar background. Therefore, all field imagery was obtained after dark.

The general procedure carried out in the yards was a two-step process. First, an initial screening took place. "Scouts" surveyed the cylinder yards to locate likely sources of contamination. They wore ordinary plastic safety glasses so that UV photons from the lamp sources were effectively filtered out, leaving the eye sensitive only to fluorescent signatures. While laser-system measurements can be carried out with some success in the daylight, this "scouting" procedure required darkness because, when integrated over the multi-millisecond-response times of the eye, the fluorescent signatures were rapidly washed out by a strong ambient light background. Furthermore, visual sensitivity was greatly increased by dark adaption.

A list of representative fluorescent signatures was compiled by the scout team. The laser system was then brought in and LIFI images were recorded.

Samples were taken of the most relevant scenes for further analysis in the laboratory. While it was possible to process the images in real time, the on-site algorithm worked only on single frames. Averaged data were collected and returned to STL for further analysis. The images presented in this chapter are the processed results of 20 frames summed into 16-bit buffer files.

1.4 SPECTROSCOPIC MEASUREMENTS OF LABORATORY SAMPLES

1.4.1 Fluorescence Spectroscopy

Spectroscopy was performed on all samples that were collected and returned to STL. A commercial Spex Fluor Log instrument was used to make these measurements (Figure 7). This instrument uses a broad-band xenon arc lamp for an excitation source, a computer-controlled excitation monochrometer for spectral selection of the incident light, a computer-controlled emission monochrometer for analysis of the fluorescent emission, and a photon-counting multiplier phototube for detecting the fluorescent emission. The instrument had been calibrated to remove the instrument response function, both in emission and excitation (see Section 1.5.3.1 and Figure 10).

1.4.2 Time-Resolved Spectroscopy

Temporally resolved spectra were obtained for some of the samples recovered from the E Yard. These data were obtained using a pulsed nitrogen laser with a wavelength of 337 nanometers, a duration of 800 picoseconds, and a pulse energy of approximately one millijoule per pulse. The emission data were collected with a fiber optic and directed to a low-resolution spectrograph (Acton Research Spectra Pro-275, 300 lines per millimeter grating). The dispersed light was then detected with a microchannel-plate-intensified gated CCD^a detector. The gate was scanned in time, and spectra were collected as a function of time. Two-dimensional images of these data were then generated. Time was plotted along the ordinate; emission wavelength was plotted along the abscissa; and emission intensity was encoded in a color rainbow (see Section 1.5.3.2 and Figure 11).

^a CCD – Charge Couple Device

FIGURE 7. SPEX FLUOR LOG

1.4.3 Laser Spark Spectroscopy

Laser spark is a technique whereby a sample having sufficient intensity to create a plasma or spark on the surface of interest is illuminated. If one analyzes the spectral content of the spark with a high-resolution grating, it is possible to observe characteristic atomic lines of the sample material. This technique is very different from fluorescence as the lines are atomic and the result is from ion recombination and relaxation of atoms in a plasma. Illumination intensities required for this measurement were on the order of 10^9 watts per square centimeter. These intensities are achievable with relatively modest pulse laser systems using focusing optics (see Section 1.5.3.3 and Figure 12).

1.5 RESULTS

1.5.1 LIFI Image Data Taken at the K Yard

Figures 8a through 8g show representative samples of the LIFI data collected in March 1996 at the K Yard. The size of the displayed images ranges from 25–50 centimeters on a side.

Figures 8a and 8b are LIFI images of valve hoods on two different cylinders. Fluorescence was noted on what appeared to be a layer of paint on some fraction of the valve hoods. This fraction, while not actually measured, was on the order of one–ten percent of the cylinders in the yard. Because the fluorescence was sufficiently prevalent, the emission was strongly considered to be originating from a primer layer in the paint. Subsequent laboratory analysis lead to the conclusion that this signature resulted from a uranium residue (see Sections 1.5.3.2 and 1.5.3.3). The fluorescent material was observed as a glossy dark coating over bare brown iron scale.

Figures 8c and 8d are images of valve stems and valve caps. In Figure 8c, a glowing deposit was found around the base of the valve nipple (with further weaker emission from a coating on the cylinder) spread in sporadic deposits from roughly 9 to 11 o'clock on a four-inch radius around the valve. The source of the emission was a hard, blister-like material, tan to brown in color, and spread around the intersection of the cylinder and the valve nipple. Again, samples were scraped and brought back to STL for analysis. Based just on current work, the source of these signatures appears to be uranium contamination.

8a Valve Hood on Cylinder 116707

8b Valve Hood on Cylinder 11669

8c Valve Stem on Cylinder 123046

8d Valve Cap on Cylinder 116754

FIGURE 8. LIF IMAGES AT E AND K YARDS

Figure 8d shows emissions observed from a valve cap with some fluorescence also observed from the surface of the cylinder. This emission was considerably weaker than the others in this series, as is seen by the poor quality of the image. The glowing substance in this instance was a thin coat on the hex-nut valve cap. The coat was unique to this valve. The presence of a slab of Krytox grease on the tank as well as the appearance of the valve led to the belief that this tank most likely had a new valve. Unfortunately, samples or scrapings were not obtained from this cylinder.

8e Concrete Approximately 6 ft in Front of
Cylinder 114959

8f Concrete Approximately 6 ft in Front of
Cylinders 116712 and 116707

8g Breached Cylinder at K-25

FIGURE 8. LIFI IMAGES AT E AND K YARDS (continued)

Figures 8e and 8f are images of contaminated concrete. This concrete was located below a breached cylinder. The contamination was concentrated along cracks and low spots in the slab. It was well fixed to the concrete. The extent of contamination was relatively wide spread in this area, dispersed over an area on the order of several square meters.

Figure 8g is an image of a breached cylinder that was patched. At the time that these data were taken, the cylinder was located in the K Yard. The breach had been patched and was reported to still be seeping, with widespread surface contamination in the general area. This image was taken without the several micro-second-phase delay. While it was visually observed to be a green color, the decay time was such that the emission was not detectable when the gate was delayed. Presumably, this emission was the result of the epoxy patch material that was used to repair the breach. Because LIFI is a surface technique, the uranium would not be detectable if it were bound in an opaque epoxy as the UV laser radiation would be absorbed by the colored epoxy substrate, not the uranium.

1.5.2 Color Photography

Figures 9a through 9e show digitized color results of the photographic efforts. Two different techniques were tried. On the first night, 400 ASA film was used with no ambient lighting. Figures 9a and 9c were taken in this configuration. On the second night, 3200 ASA color film was used with a red-filtered flash-light to provide an ambient light background. Figures 9b, 9d, and 9e were taken on the second night in that configuration.

Comparison of Figures 9a and 9b show the difference in these two techniques. The slower-speed film resulted in higher quality with less grain and sharper images; however, the sensitivity was clearly quite a bit less. In support of this conclusion, the valve stem, shown in Figure 9d, was photographed on the first night with the slower 400 ASA film, the results being no detectable image. Figures 9a and 9b compare with Figure 8f, which was taken with LIFI. The prompt blue fluorescence present in all of the concrete cracks is clearly visible in Figures 9a and 9b. The yellow-colored material corresponds with the persistent image measured in Figure 8f and is presumably uranium fluorescence. Laboratory work on cement scrapings supports this conclusion (see Section 1.5.3.2).

Figure 9c shows an image of the valve covers. This emission is quite detectable. However, the blue color of the prompt emission leads one to question if the fluorescence observed in this image may not be the same material observed in the delayed LIFI images shown in Figures 8a and 8b. Figure 9e is a typical image of organic background material, common throughout the yards. In this case, the object was identified as a bird dropping.

Following this field trip, a more intense, UV-rich, relatively high-speed flash lamp has been obtained. This flash has nearly ten times the energy in the ultraviolet range when compared to the flash that was previously used. Furthermore, it was discovered that the cut-on filter that was used to remove the flash lamp illumination had a long-lived phosphorescent emission of its own. Presumably the blue haze, evident in the high-speed film data (Figures 9b, 9d, and 9e), was due to the filter emission. Thus relatively minor improvements in technique may yield substantial improvement in results, yielding a low-cost yet very useful data product.

1.5.3 Spectroscopy

1.5.3.1 Fluorescence Spectroscopy

Figure 10 is a compilation of emission/excitation spectra taken on the samples from the E Yard. These spectra are two-dimensional images with the excitation wavelength plotted in the x direction and emission wavelength plotted in the y direction. The laser system that was fielded emitted at 355 nanometers. Thus the emission spectra observed in the field would be represented by vertical-line slices

9a Concrete Approximately 6 Feet in Front of Cylinders 11671 and 116707

9b Concrete Approximately 6 Feet in Front of Cylinders 11671 and 116707

9c Valve Skirt

9d Valve on Cylinder 123046

9e Organic Deposits

FIGURE 9. FLASH-LAMP PHOTOGRAPHY OF E YARD

through these images taken at 355 nanometers. It is important to note that these spectra were not gated. Thus they represent the integrated yields from each of the samples. This is analogous to the data taken with the photographic techniques and is much more sensitive to interference than the laser technique. Time-resolved spectra, excited at 337 nanometers with a pulsed nitrogen laser, were taken of the more interesting samples. For more information on these results, see Section 1.5.3.2.

Figures 10a through 10m show emission/excitation spectra of the samples that were obtained in E Yard. Figures 10n, 10o, and 10p show the spectra of three samples known to contain uranium.

Figure 10n is a laboratory-grade sample of uranyl fluoride (UO_2F_2). The vibrational bands are clearly visible. The excitation is nearly independent of wavelength with the exception of a dip in the 380-nanometer region and the general decrease in excitation efficiency as the excitation wavelength approached the fluorescent band.

Figure 10o is a laboratory-grade sample of uranium tetrafluoride (UF_4). This material, when compared to UO_2F_2 , is nearly nonfluorescent. Based upon these data, it is not possible to determine if the observed emission was actually due to a minute impurity in the sample or the emission was indeed from UF_4 .

Figure 10p is a spectra of a highly contaminated soil sample from the Sandia National Laboratory (SNL). This spectra was included as an example of weathered uranium. Notice that the vibrational structure seen in 10n is absent. Presumably, this broadening was due to hydrolysis and/or mixed crystalline sites in the weathered samples. Furthermore, there was a noticeable drop in efficiency in the region from 250–300 nanometers. This was most likely caused by another substance in the soil with optical opacity in this spectral region. The primary, unique feature in these uranium spectra appears to be a uniformity in the excitation spectra between 300–500 nanometers, with emission energy in the 500–550 nanometers region.

Figures 10a and 10b are cement scrapings taken in the same region as LIFI image Figure 8f. These spectra show some fluorescence in the 500–600-nanometer region; however, they are dominated by a peak in the region of 430 nanometers.

Figures 10c and 10m are cylinder-head scrapings. These time-integrated peaks are large and bright. Figure 10m was taken from the valve stem shot shown in Figure 8c. If this is a single band with a uniform response time, the emission extends further into the red than one would expect for a uranium band. There are, however, some uranium minerals that do exhibit emission in the yellow-orange region of the visible spectrum.

Figures 10e through 10j show various paint samples, including the light blue surface paint (Figures 10e and 10h), the rusty scale (Figures 10f and 10i), a glassy coat (Figure 10g), and a yellow streak that runs across the rusty corrosion (Figure 10j). The emission maximum at approximately 460 nanometers is at a wavelength that is too short to be uranium. It is interesting, however, to compare the emission spectrum of the yellow streak on the rust with rust itself (Figures 10j and 10i). The yellow streak does show increased emission coincident with the uranium emission band.

1.5.3.2 Time-Resolved Spectroscopy

Figure 11 shows the results of time-resolved spectral measurements on a selection of materials taken at the E Yard, as well as spectra of pure samples of UO_2F_2 and UF_4 . These spectra are presented as

10a Cement Scrapings Between Cylinders 116712 and 116707

10b Cement Scrapings on Cylinder 116712

10c Valve Skirt Scraping on Cylinder 116712

10d Krytox Grease Approximately 6 Inches Below Valve on Cylinder 116712

10e Light Blue Paint Chips from Valve Skirt on Cylinder 116707

10f Primer Rust Region on Cylinder 116707

10g Rust with Thin Glassy Coat on Primer on Cylinder 116707

10h Light Blue Paint Chips from Valve Skirt on Cylinder 116707

10i Paint Chips from Rust Side on Cylinder 116707

FIGURE 10. EMISSION EXCITATION MATRIX OF K-25 SAMPLES

10j Paint Chips from Yellow Streak Across Rusty Side on Cylinder 116707

10k Drip/Paint Scrapings Approximately 2 Inches from Valve on Cylinder 11559

10l Scrapings from Bottom of Cylinder 116712

10m Cylinder Bead scrapings on Cylinder 123046

10n Uranyl Fluoride

10o Uranium Tetrafluoride

10p Uranium-Contaminated Soil from Sandia National Laboratories

FIGURE 10. EMISSION EXCITATION MATRIX OF K-25 SAMPLES (continued)

two-dimensional images with wavelength presented on the y axis, time presented on the x axis, and normalized intensity encoded as a z-axis color.

Figures 11a and 11b show spectra of pure +6 and +4 oxidation states of uranium. The vibrational spacing, evident in the two-dimensional, emission excitation matrix (EEM) spectra of Figures 10a and 10b, are also very apparent in these spectra. For all data with the exception of the UF_4 data shown in Figure 11b, the shutter duration was held constant, and the shutter initiation time was delayed relative to the laser pulse. The images were formed by interpolating a series of spectra and plotting the delay time verses the emission wavelength.

The UF_4 results are shown in Figure 11b. These data were taken using a slightly different technique because the UF_4 fluorescent efficiency is highly quenched. For the UF_4 data only, the opening time of the spectrometer was fixed at the pulsar minimum, 43 nanoseconds. The width of the gate was then varied from 300–10 microseconds. By plotting the spectra-versus-gate width, as shown in Figure 11b, one is actually representing the integral of the fluorescent decay and thus increasing the sensitivity. One can obtain an estimate of the decay time using this technique. In this instance, UF_4 has a decay time on the order of one microsecond.

Figure 11C shows the emission spectra of a uranium-contaminated soil sample from SNL. It represents a weathered uranium sample. The decay is highly quenched with a decay time on the order of 500 nanoseconds. The vibronic structure, present in Figures 11a and 11b, is absent. This is presumably due to the noncrystalline state of the uranium fragments in Figure 11c.

1.5.3.3 Laser Spark Spectroscopy

Figure 12 (see page 18) shows plots of the laser-induced spark emission from four different experiments. Laser spark spectra were obtained by focusing a pulse laser on the sample such that the incident spot had sufficient intensity to create a small plasma on the surface resulting in a flash of light, hence the name spark spectra. The spectral nature of the spark is generally a broad-band plasma emission with narrow atomic lines riding on top. The atomic emission is characteristic of the elemental makeup of the ablated layer.

The top graph, Spectrum 1, shows the spark spectra of the iron rust side of the cylinder scale, Sample E-5 from Cylinder 116707. The lines are presumably the result of an iron emission. Spectra 2 and 3 are from the same sample, illuminated on the glossy fluorescent side. The difference between Spectra 2 and 3 originates from the technique used to test the sample. In Spectrum 2, the sample was subjected to multiple laser (30) pulses without moving the sample. The data in Spectrum 3 were taken by moving the sample relative to the laser beam between each of the 30 laser pulses, thereby exposing a fresh region of the sample with every pulse. When the sample was not moved, the fluorescent layer was obliterated, exposing the iron substrate to the laser. Hence Spectrum 2 has lines common with both the iron rust scale and with the fluorescent material. Spectrum 3 exhibits lines only from the fluorescent surface. Spectrum 4 in Figure 12 shows the spark spectrum of a sample of uranium-contaminated soil taken from a SNL sled-track site. Spectra 2, 3, and 4 show common emission at a wavelength coincident with a uranium atomic emission line. This peak was not observed on the uncontaminated side of the cylinder scale.

11a Uranyl Fluoride, 100-ns Shutter Width
Delay Range from 30 ns to 3 ms

11b Uranium Tetrafluoride, 43-ns Shutter Width
Delay Range from 0.3 to 10 μ s

11c Uranium Contaminated Soil from Sandia
Sled Track

11d Sample E-1, Cement Scrapings 5 ft East of
Cylinder 116712 and 116707, West Side
of Crack

11e Sample E-3, Paint and Primer Scrapings
from Valve Skirt on Cylinder 116712

11f Sample E-5, Paint and Primer Scrapings
from Valve Skirt on Cylinder 116707

FIGURE 11. TIME-RESOLVED SPECTRA OF URANIUM-CONTAMINATED SAMPLES

FIGURE 12. LASER SPARK SPECTRA OF VARIOUS SAMPLES

Figure 13 shows a micrograph of the surface of the contaminated cylinder scale. These photographs were taken with a magnification of $50\times$. The width of the laser-drilled crater was on the order of 100 microns. The track, evident in Figure 13, is the result of scanning the laser across the surface while taking spark measurements. The drilled crater is where the beam was held stationary while the measurements were being made. Under the microscope, a clean metallic surface was visible at the bottom of the crater. The thickness of the layer was measured using the depth of field of the microscope and was found to be approximately 30 microns. The surface quality of the contamination layer appeared fairly smooth and continuous, even under microscopic observation.

1.6 CONCLUSIONS

The remote detection and imaging of persistent fluorescent signatures were demonstrated at the UF_6 cylinder yards at the K-25 Site. A variety of spectroscopic techniques were applied to these samples, and it was determined, to the greatest extent, that the samples were contaminated with uranium.

The signatures fell roughly into three classes:

- Cement contamination
- Valve contamination
- Cylinder-scale contamination

The cement contamination observed in the E Yard appeared to be well fixed in the concrete matrix. Likewise, the observed valve contamination was minor and difficult to remove. However, the conditions of the valves in the other cylinder yards may be different.

FIGURE 13. MICROSCOPIC IMAGE OF LASER SPARK TRACK

Of the three classes of contamination that were observed, the cylinder-scale problem is potentially the most interesting. Sufficient time was not spent in the yards to get an accurate statistical sampling of the number of contaminated cylinders; however, the best estimate would be one–five percent. In general, the contamination seemed to be concentrated on the upper surfaces of the valve hoods. This may be an artifact of different filling or purging procedures that had been used over time. The uniformity of the coating on the scale samples leads one to believe that the contamination is due to condensation or deposition of fine particulates from the atmosphere.

The cylinder-scale problem is most interesting in that it is currently uncontrolled. Although the total amount of uranium on any given piece of scale was small and virtually undetectable by conventional means, large-scale operations with the cylinders have the potential for producing substantial quantities of loose contamination.

In conclusion, a variety of fluorescent signatures from the E Yard were characterized. LIFI was shown to be a useful technique in identifying previously unknown and potentially troublesome sources of contamination.

1.7 FOLLOW-ON WORK

Further work in the following areas would be useful to expand the knowledge base and provide instrumentation to aid in managing and decommissioning DOE production facilities.

1.7.1 Facilities Study

The efforts that have been made to date have provided some unexpected results. In the time available, STL personnel were not able to conduct a comprehensive survey at the K-25 Site. Furthermore, the status of the yards is potentially different from those at other operational facilities. STL personnel propose that follow-on field work be performed at other cylinder yards, inside operational buildings, and inside areas that have been shut down. The purpose of this work would be three-fold.

- A. First, a better estimate of the extent of the scale-contamination problem is needed. To do this, a systematic survey of a large number of cylinders from a variety of different sites must be undertaken.
- B. Second, applying STL techniques to the interiors of operational facilities could be useful. The fluorescent signatures are persistent, yet there are questions pertaining to the age of the contamination, for example, differences in historic-versus-modern-day techniques.
- C. Third, there are a number of issues pertaining to the decommissioning of these facilities that should be addressed. As the buildings are disassembled and equipment is exposed, the technology could be used to locate areas of contamination.

1.7.2 Equipment Development

STL personnel used a variety of techniques throughout this investigation. All of the techniques have application to the identification of uranium. Furthermore, these techniques could be developed to provide on-site diagnostics of potential contamination.

Flash photography provides a low-cost, useful screening tool for the detection of uranium contamination. Portable battery-powered flash-lamp sources, two-ten times brighter than those used in the field for this study, exist commercially. Better synchronization of cameras and flash lamps may permit operation in areas of elevated ambient lighting.

Laser spark spectroscopy provides a reasonably unambiguous means to identify the elemental constituents of materials. The laser requirements for this type of measurement are very similar to those required for the portable LIFI system. The differences lie in the beam-delivery optics and the detection requirements. The addition of a spark attachment to the LIFI system would greatly enhance the power of the measurement and the ability to state unequivocally the chemical nature of the target. Furthermore, this measurement would potentially expand the suite of materials to items not detectable in a fluorescent mode. This technology may have application to the detection and analysis of materials such as technetium (Tc) and PCBs, both of which are issues in decommissioning and decontaminating buildings.

Finally, the LIFI system requires further development to be optimized for the cylinder-yard project. The current configuration was designed to create the lightest possible package using the existing technology at the time of development (1994). Laser and camera technology have progressed to the point

where the laser head and camera can be designed in a single package, thus eliminating the use of the fiber-optic transmitter. This development is currently underway as a part of the OST CMST-CP. Much of the difficulty experienced in the field has originated with the alignment and maintenance of the fiber transmitter. It is believed, based on this work, that the delivery of a LIFI system or the delivery of survey services to the K-25 Site would be useful in managing this facility.

Chapter 2. Long-Range Alpha Detection

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2.1 INTRODUCTION

Researchers from LANL spent a week at the K-25 gaseous diffusion plant to test a new "sniffer" type of radiation detector that was intended to be more effective in the cylinder yards than conventional alpha/beta probes. The new device detects alpha radiation using a technique that LANL has developed called long-range alpha detection (LRAD). This technology relies on detecting the air ionization caused by alpha particles rather than detecting the alpha particles themselves. It has been shown to be sensitive down to the single-disintegration level and to have a linear response up to activities that cause large dead-time losses in other detectors.

LRAD detectors have been used to measure activities in enclosed areas in various ways. In contrast, the LRAD "sniffer" was designed to measure open areas, in particular, the UF_6 storage cylinder yards.

A cross-section view of the LRAD sniffer is shown in Figure 14. Air, pulled by the fan, flows from right to left. This air carries positive and negative ions produced by ionizing radiation in the sample volume. Ions of one polarity are collected on the signal plane. The collected ions form a current that is measured by a sensitive electrometer. The electrometer signal is digitized and read by a palmtop computer (not shown).

Optional hoses, having an outside diameter of two inches, can be attached to the input to extend the reach of the detector to otherwise inaccessible areas. To prevent the monitor from behaving like a vacuum cleaner, the fan speed is limited to provide an airflow equivalent to a five-mile-per-hour breeze.

FIGURE 14. SCHEMATIC SHOWING A CROSS-SECTION VIEW OF THE BODY OF THE IONIZATION SNIFFER TESTED AT THE K-25 SITE

The whole detector is powered by a rechargeable camcorder battery carried in a fanny pack. Measured battery life is ten hours on a full charge.

The sniffer includes several innovations that were considered but never implemented in an LRAD detector. To minimize weight and maximize practicality, a new miniature serial port analog-to-digital converter and an HP 200 palmtop computer were used. The transition from more powerful laptop computers and GPIB (general purpose instrument bus) devices required writing a new data-acquisition program. The electrometer board was the third and smallest revision of an in-house design, and the K-25 Site visit was that revision's first use in the field. In spite of the relatively short preparation time prior to the field demonstration, the many bugs involved in building such a new monitor were resolved, and as described in the following section, the monitor performed well.

2.2 CHARACTERIZATION OF THE SNIFFER BEFORE THE K-25 VISIT

The first proof-of-principle sniffer was a modified LRAD object monitor suitable only for laboratory tests. It filled the top of a roll cart, was powered by a car battery with an alternating-current adapter, and was held together with yellow tape. Once it was determined that a sniffer could detect external alpha sources and that it could work outdoors, a unit was designed that would be practical to use. The new unit was tested in the laboratory to ensure that it gave reproducible results and to determine some of its basic characteristics. A number of tests were conducted using the bench-top setup depicted in Figure 15.

Tests to determine the sniffer's linearity and detection volume were among the experiments that were performed with this configuration. NIST-traceable,^b plutonium-239 (^{239}Pu) check sources were used in both the linearity and the volume tests. The response to other sources was proportional to the ratio of the alpha energies.

Figure 16 shows the data from the linearity study. A two-foot hose was attached to the sniffer and the sources were placed two inches from its opening, the nearly optimal position. The detector's response to ^{239}Pu under these laboratory conditions was 12.6 dpm/fA, where dpm is disintegrations per minute and fA (femtoampere) is equal to 10^{-15} amperes.

FIGURE 15. *DIAGRAM OF EXPERIMENTAL SETUP (PLAN VIEW IS NOT TO SCALE)*

^b NIST – National Institute of Science and Technology

FIGURE 16. RESULTS OF A BENCH-TOP LINEARITY STUDY

Figure 17 shows the data from measuring the detection volume. The axes are positions in inches and the gray-scale values represent the current in femtoamperes. The source was placed at various positions on the grid. The detection volume was arbitrarily defined to be that in which at least ten percent of the generated ions were collected by the detector. For instance, Figure 17 shows that, with a two-foot hose, the detection volume is approximately one cubic foot. Outside the laboratory, the shape of the detection volume changes when it is redefined by solid objects, but it is assumed the volume stays the same. A similar set of data exists with four-inch-diameter hose. The two-inch hose was found to be more efficient and had lower background than the four-inch hose. For the K-25 demonstration, two-inch-diameter hoses of zero-, two-, three-, and five-foot lengths were used. These hoses could be joined together to create lengths to up to ten feet, the length of the UF_6 storage cylinders.

From the known behavior of LRAD detectors and the measured responses shown in Figure 17, the minimum detectable activities of uranium were calculated. Samples of both enriched (93 percent) and depleted bare uranium were obtained, and comparisons were made with a Ludlum 139 alpha meter, as described in the following paragraphs.

With its high-specific activity, 1/2-inch diameter 93 percent enriched uranium foils were easily observed (30,000 femtoamperes above a background of 1000 femtoamperes). No difference was measured for foils of 1-mil and 5-mil thicknesses. No change was expected for different thicknesses because alpha particles travel only a few tens of microns in solids.

Depleted uranium has about 150 times less alpha activity than 93 percent enriched uranium per given surface area. This was verified with the sniffer, obtaining a 600-femtoampere signal for a 7/8-inch-diameter depleted uranium alpha check source (three times the area of the 1/2-inch foils). The sniffer and Ludlum 139 measurements of alpha activity agreed to within 20 percent (8.4 and 7 thousand disintegrations per minute, respectively, for the depleted uranium).

Fig. 4a. 2"Dia/No Hose

Fig. 4b. 2"Dia/2' Long Hose

Fig. 4c. 2"Dia/5' Long Hose

Fig. 4d. 2"Dia/10' Long Hose

FIGURE 17. DATA FROM TESTS TO DETERMINE THE DETECTION VOLUME WITH VARIOUS LENGTHS OF HOSE. Axes are in inches; gray-scale values are in femtoamperes

2.3 SNIFFER ADVANTAGES

The sniffer was observed to have several advantages over the conventional alpha probe. The LRAD sniffer has a faster response time. The time from initial detection to maximum response was 2 seconds for the sniffer versus 18 seconds for the Ludlum 139. The source was detectable up to one foot away with the sniffer, but the alpha probe had to be within one inch. At 1/2-inch separation, the traditional probe registered only 300 counts per minute versus 3500 on contact. As shown in Figure 17b, the sniffer had a nearly flat maximum signal over about four inches of travel and at up to 2 1/2 inches from the hose. The difference means the sniffer is much less demanding about positioning than a traditional probe.

The depleted uranium produced only one click of the traditional alpha probe at a scan speed of two inches per second and at a 1/4-inch height, the generally accepted scan rate. Through experience at a nuclear facility, a single click is usually ignored. At one inch per second, the traditional alpha probe registered several clicks over the source, and a surveyor would have noticed the activity. In contrast, the LRAD sniffer could "see" the source at a scan rate of four inches per second, and the total surface area covered in the sniffer scan was much greater.

To be fair, the traditional probe was observed to have two advantages of its own. The traditional probe had a background of nearly zero counts per minute. Any signal of more than a single click raises suspicion. The sniffer had a background that was proportional to the background radiation level. The higher the background, the higher the minimum detectable activity.

Insulating materials, especially plastic, tend to carry a static charge that pulls ions from the air, making it more difficult to detect small sources with the sniffer. The traditional probes do not have problems with insulators. Fortunately, the cylinders to be measured were metal, and even the paint on a new cylinder was conductive enough that this phenomenon was not a problem at the K-25 Site.

These comparisons bode well for the sniffer as a scanning tool. The faster scan speed and the ability to reach places inaccessible to other probes are considered the best features of the sniffer.

It should be stressed that the sniffer is not intended for remotely quantifying amounts of nuclear material. To do so would require frequent and impractical recalibration for the given measurement geometry. Rather, the sniffer is intended specifically to indicate whether or not there is radioactive contamination in spaces that are not accessible to traditional alpha probes. Given that these cylinders are large, a leak would appear as a localized increase in signal in a scan of the exterior surface. The expected gamma background, up to a few milliroentgens per hour (mR/h) in the cylinder yards, merely raises the background in the detector but is relatively constant around each cylinder.

2.4 RESULTS OF FIELD TEST AND DEMONSTRATION

There were three goals for the demonstration of the sniffer at the K-25 Site. Because the LRAD sniffer was the first fielded unit of its kind, the LANL staff needed to determine (a) if it would work in the humidity and high-gamma backgrounds of the yards, (b) if it would be a practical and useful tool for workers looking for leaks in the cylinder yards, and (c) what its minimum detectable activity was in the high-background environment.

Data was gathered during all five days at K-25. The sniffer performed well every day. As a first of its kind, it was not surprising that the sniffer "broke" three times during the week. It was incredible that all three times were in Building 33 rather than in the yards. At the end of the first day, the background level in the sniffer went high and stayed high. The next morning, the cause was isolated and fixed: a shorted insulator. The second night the electrometer board failed. It was replaced the next morning. Finally, when setting up the sniffer in the room for the fourth night, a power connection worked loose and had to be reattached. All three of these breakdowns are considered to be part of the learning curve for a new detector, and all three problems were easily and permanently corrected.

Rather than present the measurements and results from each day in chronological order, the most important results of the sniffer demonstration are presented in the following sections.

2.4.1 K Yard Cylinder Rows

The K Yard has the oldest and most leaky cylinders at K-25. The cylinders are stacked two-high in long rows, with narrow walkways between rows. A survey was conducted in part of the yard to determine how well the sniffer worked and to establish the minimum detectable activity under actual conditions.

It was decided not to spend time and effort finding possible leaks of unknown sizes in the yard. Rather, leaks were simulated by planting an alpha source of known strength among the cylinders. One LANL person hid the source, and the other person tried to find it. Because no one was allowed to climb on the cylinders, the source was placed within arm's reach of the cylinder ends. The stiffening rings on the cylinders provided a reachable location; it was easy to hide the source out of sight behind these rings.

Each searcher started at the end of the row and worked down 41 cylinders. In the interest of saving time and performing the worse-case scenario, only the surfaces around the lower gap formed by three touching cylinders were surveyed (the black areas in Figure 18). Here visibility was worse and the gamma background was the highest. The source was planted at three locations on each row. This procedure provided sufficient background measurements to get a good understanding of the minimum detectable activity as well as providing several examples to show an operator how a leak would appear. Each of the two LANL people searched for and found the hidden source.

Figure 19 shows data from one of the searches. Individual data points in Figure 19 are two-second averages of the ten points per second that the searcher saw. Black vertical lines delineate searching a new cylinder location. As evidenced by the data, it took about one minute to survey around each location. The planted source was found and showed up clearly above background at 10:08:30 a.m.

Several small spikes in the data correspond to banging the detector on the ground or against one of the cylinders. On the operator's real-time display, such spikes appeared instantly, and the signal quickly returned to background. In contrast, the signal from a source gradually built up and fell off as the sniffer passed over it. The sniffer operator could easily distinguish noise spikes from a true signal. The distinction was less evident in the two-second averages displayed in Figure 19.

Two important points to get out of Figure 19 are background levels and activity levels. The background varies between 6000 and 8000 femtoamperes with a standard deviation about the mean somewhat less than the difference. The signal from the source was about 7000 femtoamperes.

FIGURE 18. REGIONS BETWEEN THE K YARD CYLINDERS SURVEYED BY SNIFFER ARE INDICATED IN BLACK

FIGURE 19. DATA TAKEN ALONG A ROW OF STACKED CYLINDERS IN K YARD (all readings were collected in the morning). *Black vertical lines correspond to different cylinder locations. The high reading at the end is from the source.*

The source was a commercial antistatic brush refill. It contained 500 microcuries of polonium-210 (^{210}Po) when new, but it had decayed for eight half-lives by the time of the demonstration. Even so, the activity was still so high that tape was put over three-fourths of the active area of the source. A K-25 health physicist measured the taped source to be 301.8 thousand disintegrations per minute, using an alpha meter. Upon returning to LANL, the same source was measured to be 407.5 thousand disintegrations per minute of ^{210}Po . This produces ionization equivalent to 495.8 thousand disintegrations per minute of depleted uranium. Given the previous measurements of depleted uranium, this corresponds to an amazing 35 square inches of activity.

Because of the geometry, the source that should have registered 35,000 femtoamperes on a flat surface showed up as only 6000 to 8000 femtoamperes. This reduction can be attributed to two effects. First, the source placements were behind the cylinder stiffening rings so that they would not be visible. Placement along an edge cut down the solid angle and halved the available signal. Additionally, by not knowing where the source was located and approaching it from only one side, the geometry and airflow were not optimal and could easily account for another factor of three in detection efficiency. Together, these measurements represent the worst case: a leak located in a corner and also in a high-gamma field.

From Figure 19, it would appear that the minimum detectable source would be one-quarter to one-eighth the size of the source that was used or 60 thousand disintegrations per minute of depleted uranium (four square inches). In fact, the graph does not relay the real-time nature of the readings where small variations in signal can be double- or triple-checked for repeatability. To determine the minimum detectable activity, the variation of background is the driving factor. The background level between the cylinders anywhere in the rows resembled the background in Figure 19. As an absolute lower limit, it was deduced that a signal of 1500 femtoamperes should be noticeable during these readings because it repeated with position during a scan. This 1500 femtoamperes corresponded to 21 thousand disintegrations per minute or 1 1/2 square inches of exposed, depleted uranium if it was in the

most favorable geometry. Therefore, 21 thousand disintegrations per minute on an unobstructed surface was the minimum detectable activity for the highest gamma fields that were observed.

2.4.2 Leaking Bleed Valve on Patched Cylinder

An actual leaking cylinder was measured in a low-background area of the K Yard (Cylinder 7953). The cylinder was well removed from all other cylinders. The leaking cylinder had been patched, but surface contamination was easily detected. Figure 20 shows repeated horizontal scans along the length of the cylinder. The scan rate was about four inches per second. The data was saved at ten points per second—the same rate that the sniffer operator observed in the field. Background increased with dose rate from the ends to the middle of the cylinder, varying slowly from 1200 femtoamperes at the ends to 1800 femtoamperes in the middle of the cylinder. Noticeable spikes occurred each time the sniffer passed the bleed valve on the cylinder's patch. The large signal after 10:08 a.m. occurred when the operator searched the area immediately surrounding the valve looking for the maximum signal. There was some discoloration but no obvious sign of uranium on this patch.

The health physicist monitored the cylinder. A reading of 916 counts per minute was taken in the area around the bleed valve; elsewhere, a reading of less than 20 counts per minute was obtained. The sniffer obtained a signal of 2000 to 4000 femtoamperes, implying an activity of 28–56 thousand disintegrations per minute. Thus the sniffer read 10–20 times greater activity than measured with the smaller-area traditional alpha probe.

Part of the difference in readings occurred because the bleed valve protrudes from the patch and the alpha probe could not be placed in contact with the surface closest to the valve. Furthermore, the activity likely extended beyond the footprint of the traditional probe. Reiterating, there was some discoloration but no obvious sign of uranium on this patch. Nevertheless, it generated a signal equivalent to

FIGURE 20. *SCANS OF PATCHED CYLINDER 7953. Horizontal northward and southward scans of the cylinder are indicated. Spikes during the scans and the high activity after 10:08 correspond to readings near the bleed valve on the patch (all readings were collected in the morning).*

two–four square inches of depleted uranium. The contaminated area on the patch was probably closer to one square foot with a much lower uranium quantity per unit area than the uranium metal on which the comparisons were based. If a similar leak existed in the K Yard cylinder rows, the sniffer should have been able to find it. These comparison measurements illustrate the ability of the sniffer to detect activity better than an alpha probe in some situations.

For such an ideal case as this isolated cylinder, an absolute minimum detectable activity would be 750 femtoamperes or 10.5 thousand disintegrations per minute (0.75 square inches) of depleted uranium.

2.4.3 B Yard Valves

The sniffer was used in the B Yard to measure contaminated valves. The B Yard cylinders were skirted and most were empty. They all had brass-colored valves and many had green or yellow-green stains on them. The sniffer was used to measure the contamination on all of a given valve at once, something a traditional alpha probe cannot do.

A data set from eleven cylinders is shown in Figure 21. Cylinder identification numbers are available but not shown. Several of the valves are shown to have significant contamination on them.

Workers at K-25 were observed to base guesses on contamination levels by the quantity of discoloration. All but two of the valves covered by Figure 21 had significant amounts of discoloration, but activity did not necessarily track the estimated areas of the stains. Spot checks with a traditional alpha probe gave agreement with the sniffer's readings.

The sniffer was judged to be a faster and more efficient way to monitor the valves as compared to using regular alpha meters. It is believed that a dedicated LRAD detector would be even more effective.

FIGURE 21. MEASUREMENTS OF VALVES IN B YARD. Arrows indicate readings of different valves, with the width of the horizontal bar being time near the valve (all readings were collected in the morning). Areas are estimates of green or yellow-green discoloration on valves.

2.4.4 Using the E Yard Sniffer as a Road Monitor

In addition to testing the sniffer on cylinders in the E Yard, an area of concrete pavement where some uranium had washed off equipment years ago was also monitored. Background levels were slightly less than 1000 femtoamperes and most of the marked areas produced readings in the sniffer. The correlation between the sniffer reading and the activity marked on the concrete was about 40 disintegrations per minute per femtoampere. It is not well understood why the reading was only one-third as much as what was expected.

Excessive time was not spent in this area because there was an existing, well-developed LRAD monitor—the soil-surface monitor—that is ideal for monitoring flat areas. The soil-surface monitor has a lower background (300 femtoamperes) and higher sensitivity (six disintegrations per minute per femtoampere) than the sniffer. The sniffer compromised on both to be able to measure inaccessible areas between tanks.

2.5 OBSERVATIONS RESULTING FROM SNIFFER DEMONSTRATION

The field tests revealed some need for further development. Some of the noise in the signal could be reduced by filtering out sharp spikes and eliminating microphonics. The sniffer needs to be more robust against rust flakes. The sniffer is hand carried but should probably be made less unwieldy. The hose also could be lighter and more manageable. A better method of holding the computer definitely needs to be devised. All of these improvements would contribute to making the sniffer a more practical device. Additionally, feedback from the probable users would be very beneficial.

Besides further development of the sniffer, other LRAD monitors of interest are suggested by this demonstration. The well-developed surface monitor was mentioned in previous sections as a better solution to measuring large, flat areas. It has higher sensitivity and lower background than the sniffer for such applications.

When monitoring the cylinder valves, the sniffer was more effective than the traditional alpha meter. However, it was apparent to the operators that, for the job of valve monitoring, a smaller, more rugged, and simpler detector with no airflow would give excellent results.

2.6 CONCLUSIONS

The demonstration was remarkably successful considering the short time that was available for development. Three experiences in particular lead to the belief that the ionization sniffer is an appropriate tool for surveying the cylinder yards: (a) the hidden source searches (b) the measurements of the leaking bleed valve on the patched cylinder in the K Yard, and (c) the data collected on the contaminated valves in the B Yard.

The demonstration revealed that the sniffer is well capable of detecting activities on the order of what might be expected from a leak. Even in the yard's highest gamma environment and in the least favorable areas of the cylinder stacks (*i.e.*, in the spaces between three cylinders), 60 thousand disintegrations per minute could be detected with a high degree of confidence. In more favorable locations but

still in the high-gamma background, it is predicted that as little as 21 thousand disintegrations per minute could be detected, the expected signature of a 1 1/2-square-inch leak. In the ideal case of an isolated cylinder, an absolute minimum detectable activity would be 10.5 thousand disintegrations per minute.

The sniffer offers the following advantages for monitoring UF_6 storage cylinders for leaks: (a) the sniffer is able to find leaks between cylinders from the walkways between rows; (b) the sniffer can find leaks not visible to the naked eye; (c) the sniffer monitors a larger area faster than any normal alpha probe; and (d) the sniffer can monitor spaces not accessible to a normal alpha probe. It is believed that the sniffer could be a valuable tool for monitoring cylinders.

2.7 FOLLOW-ON POSSIBILITIES

Considerable work has been done on pipe monitors where a sniffer-like detector attached to one end of the pipe measures electrostatically filtered air admitted from the far end of the pipe. Measurements from each end of the pipe combine to give the total activity in the pipe regardless of the unknown position of activity along the pipe. Pipe monitors have been used on 20-foot sections, limited only by the laboratory space for the demonstration. Bends and open valves do not prevent measurements.

LANL has conceptual designs for barrel and 55-gallon drum monitors, both external and internal. Gaseous diffusion plants are excellent candidates where such monitors can be used.

The LANL staff has a preliminary design for an LRAD-based conveyor belt monitor. Scrap or processed material may be analyzed for alpha, beta, and gamma activity simultaneously in such a device.

Recent work has concentrated on airborne alpha monitors. Such real-time monitors could be useful in several areas at the K-25 Site and elsewhere, either as complements to continuous air monitors or as monitors for stack emissions.

Chapter 3. Neutron Activation Analysis

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3.1 INTRODUCTION

The purpose of this project was to determine whether neutron activation analysis (NAA) techniques could be used to measure the thickness of UF₆ storage cylinder walls with five percent uncertainty and within a reasonable time. Assuming that the chemical composition of the cylinder material is known, the neutron activation cross sections and the gamma-ray measurement parameters would determine how well and how rapidly the thickness could be measured. The neutron cross sections determine the neutron flux required to make the gamma-ray measurement in a given time. The variable parameters in the experiments include the neutron flux, detector thickness, area measured, and scanning time. The gross thickness of the steel cylinder walls (less than 5/16 inch) is thin enough that the corrections for scattering losses and gamma-ray attenuation should be either small or negligible. The relatively high energy of the prompt neutron-capture gamma rays (generally between 5 and 10 MeV [megaelectronvolts]) is a great benefit in these measurements because nearly all of the background radiation from the UF₆ is below 3 MeV. This means that the signal-to-noise ratio of the gamma measurement would be quite favorable in this environment of high-gamma-ray flux.

3.2 OPERATIONAL AREAS

The entire project was initially divided into four phases. Following each phase, a determination was made regarding the reasonableness to continue with the proof of concept. After the second phase, it was decided that proceeding beyond Phase 2 would not be useful. The first two phases are described in the following sections.

3.2.1 Phase 1 – Calculations and Computer Modeling

In Phase 1, the metallurgy of the cylinders was investigated to determine the isotope(s) with the most unique prompt gamma-ray signature(s) from the neutron capture. Based on the cross sections and content in the steel, an appropriate neutron flux was determined for the measurement. Calculations were made to model the measurement, including the neutron-capture rate of the uranium inside the cylinder. Based on these simulations, a decision was made to proceed to Phase 2 and use strong neutron sources.

3.2.2 Phase 2 – Laboratory Measurements of Steel Samples

The test equipment was assembled to perform the proof-of-concept measurements under controlled laboratory conditions. Laboratory measurements were made in an attempt to determine different known thicknesses of steel. Samples of corroded steel were measured. The transmitted neutron flux was known by using NIST-traceable sources. The results of these tests were not encouraging. It was decided not to proceed with Phase 3.

3.3 NEUTRON ACTIVATION ANALYSIS

Neutron activation measurement is a highly sensitive, elemental-analysis technique. It relies on the process by which a neutron, usually thermal (approximately 0.025 eV [electronvolts]), can collide with a stable target nucleus and be captured. The process of neutron capture results in an energetic gamma ray (between 5 to 12 MeV) being emitted from the nucleus. The gamma ray is called a prompt gamma ray because it is emitted simultaneously as the neutron capture takes place. The intensity of the gamma ray that is produced is directly proportional to the concentration of the target element in the sample. This relationship is true for both the prompt gamma rays and the decay gamma rays. Both can be used in NAA for absolute and comparative methods of calculation. Most laboratories use the decay gamma rays for analysis because the prompt gamma rays require measurement near the neutron source while the sample is being irradiated. The decay gamma rays can be conveniently measured after the irradiation is complete in a laboratory far from the neutron source. However, prompt gamma-ray measurement was chosen for this purpose.

Comparative NAA is the simplest and most accurate approach to the problem. "Standards" of known composition are irradiated at the same time as the samples so that both the "known" standard and the unknown samples are exposed to the same neutron flux. By knowing (a) the concentration of a particular element in the standard and (b) the ratio of the standard's counting rate and weight for a particular activation product to that of the sample, the concentrations of the elements in the sample can then be determined. No knowledge of the neutron flux, the activation cross section, or even the detector efficiency is required to calculate the concentrations of many elements in the sample. Comparative NAA is elegant because of its simple method of calculation. As can be seen in Equation 1, the accuracy of the method approaches the counting precision because all of the uncertainties are from the counting uncertainty, the weighing uncertainty, and the uncertainty of the standard's elemental concentration:

$$\text{Conc}_{\text{smp}} = \text{Conc}_{\text{std}} (C/S_{\text{smp}} \cdot Wt_{\text{std}}) / (C/S_{\text{std}} \cdot Wt_{\text{smp}}) \quad (1)$$

where Conc is the concentration, C/S is the count rate, and Wt is the weight of the sample under consideration. In absolute NAA, the actual concentration can be determined only if one knows the neutron flux, the neutron-capture cross section of the element, the detector efficiency, and the isotopic abundance in addition to the parameters required for a comparative analysis. While the absolute method avoids the need for a standard, it requires a knowledge of the parameters that increase the overall uncertainty of the measurement. A list of typical NAA applications are given in Appendix A.

3.4 PHASE I CALCULATIONS – QUANTITATIVE NEUTRON ACTIVATION TO MEASURE CYLINDER-WALL THICKNESS

The chemical compositional requirements (per ANSI N 14.1, 1990) for the ferrous materials used to construct the 14-ton cylinders are given in Table 1.

Table 1. Chemical Compositional Requirements for the Ferrous Materials Used to Construct the 14-Ton Cylinders

Elements	Grade 380 (%)	Grade 415 (%)	Grade 450 (%)	Grade 485 (%)
Carbon (max)	0.18	0.21	0.24	0.27
Manganese	0.8 – 0.9	0.8 – 0.9	0.85 – 1.20	0.85 – 1.20
Phosphorous (max)	0.035	0.035	0.035	0.035
Sulfur (max)	0.035	0.035	0.035	0.035
Silicon	0.15 – 0.40	0.15 – 0.40	0.15 – 0.40	0.15 – 0.40

The iron content of the steel is above 95 percent. From Table 1, it is apparent that the other components of the alloy do not contribute significantly in the neutron activation process. A list of elements producing prompt gamma rays after thermal neutron capture is shown in Appendix B. From Appendix B, it is seen that iron produces a gamma-ray doublet at 7631.1 keV (kiloelectronvolts) and 7645.4 keV after absorbing a thermal neutron. The relative strength of this doublet is directly proportional to the thickness of the steel.

The activity in decays per second can be related to the net count of a specific photopeak using Equation 2:

$$A_{\text{DPS}} = P/\omega \times \varepsilon \times T_c \times \Gamma \quad (2)$$

where A is the activity, P is the net area counts under the photopeak, ω is the solid angle, ε is the absolute efficiency, T_c is the counting time, and Γ is the branching ratio.

3.4.1 Efficiency of High-Purity Germanium

Relative to sodium iodide detectors (NaI(Tl)), the high-purity germanium (HPGe) detector used in this study is 40 percent efficient for high-energy gamma detection. The theoretical value for the efficiency of a cylindrical NaI(Tl) crystal measuring 3×3 inches and located 25 centimeters from the source is 1.2×10^{-3} . Thus a 40 percent HPGe detector has an absolute efficiency of 4.8×10^{-4} at the gamma-ray energy of 1332 keV and at a distance of 25 centimeters from the source.

3.4.2 Solid Angle and Geometric Correction

Assuming (a) the thermal neutron beam is tight and is spread over a circle with a one-centimeter diameter on the steel surface and (b) there is a distance of 25 centimeters between the activated surface and the HPGe detector, which has a surface area of approximately 20 square centimeters, the solid angle will be 5.0×10^{-4} str.

3.4.3 Branching Ratio for the Doublet

The branching ratio Γ for the 7631.1 keV line is 27.7 percent and the branching ratio Γ for the 7645.5 keV line is 24.4 percent. Assuming an average branching ratio of 26 percent, a net peak count of 1000 counts per minute (allowing three percent statistical uncertainty in a minute) would require an activity as calculated in Equation 3.

$$A_{\text{DPS}} = 2.7 \times 10^8 \quad (3)$$

3.4.4 Cross Section for Thermal Neutron Capture

The cross section for thermal neutron capture in iron and instantaneous release of a prompt gamma (σ) is 2.55 barns or 2.55×10^{-24} square centimeters.

3.4.5 Number of Iron Atoms per Square Centimeter

It is assumed that steel contains 95 percent iron by weight and has an average density of 7.67 grams per cubic centimeter. The average thickness of the steel wall is assumed to be 5/16 inch or .794 centimeters. Over a circular area having a one-centimeter diameter, the number of iron atoms (N_c) is 4.9×10^{22} .

3.4.6 Required Neutron Flux

To achieve the activity, A_{DPS} of 2.7×10^8 , one needs a neutron flux Π_n given by

$$A_{\text{DPS}} = \Pi_n \sigma N_c \quad (4)$$

or, $\Pi_n = 2.16 \times 10^9$ thermal neutrons per second per square centimeter.

This thermal neutron flux is too high, even for the larger-size neutron generator, to produce (see Table C-1 in Appendix C).

3.5 PHASE II EXPERIMENTS

The experimental setup is shown in Figure 22. Instead of using a bismuth screen, a cadmium sheet backed up by a lead sheet was used to avoid direct thermal neutrons and low-energy gammas from entering the HPGe detector. A highly moderated californium-252 (^{252}Cf) (moderated up to 85 percent) source of known strength of 3.56×10^6 neutrons per second was used to irradiate two samples of the steel actually used to construct the cylinders containing UF_6 . The surface area of the polyethylene cylinder is 21.4 square centimeters. The source strength is therefore 1.66×10^5 neutrons per second per square centimeter. The thickness of the samples was 5/16 inch. Each piece of the steel element measured approximately 2.5×3 inches. The pulse-height spectra of the resultant prompt gamma

FIGURE 22. THE EXPERIMENTAL SETUP FOR THE NEUTRON ACTIVATION STUDY

rays were analyzed with a Canberra Inspector MCA. An effective gamma rate of 3.4 counts per minute was obtained. This translates to 0.07 counts per minute per square centimeter. To obtain a gamma-ray count rate of 1000 counts per minute, the thermal neutron flux must be 2.37×10^9 neutrons per second per square centimeter, which agrees fairly well with the Π_n obtained in Phase 1. The net peak area under the doublet peaks is as follows:

For a peak at 7631.1 keV, net count equals $10,070 \pm 1.77$ percent

For a peak at 7645.5 keV, net count equals 9862 ± 1.60 percent.

Again, this rate has been proven to be too slow for this technique to be a useful field application.

3.6 REFERENCES

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Chapter 4. Differential Gamma-Ray Attenuation

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4.1 INTRODUCTION

Results of a feasibility study on a technique used to gauge the thickness of cylinder walls are presented in this chapter. The goals of this study were to determine (a) the accuracy with which the wall thickness of UF₆ storage cylinders could be measured using differential gamma-ray attenuation and (b) the time necessary to make such measurements.

The count rate from an extended gamma-ray source viewed through a passive absorber is dependent upon many factors.¹ Some of these factors are difficult to measure accurately, such as the absolute detector efficiency for each source-detector geometry and the distribution of activity within the source. By a simultaneous measurement of the count rates of two gamma rays, the thicknesses of cylinder walls can be determined independent of the absolute detector efficiency or the source geometry.

The thickness of a passive absorber, the steel cylinder wall in this case, can be determined from the following equation:

$$t_a = \frac{\ln \left[\frac{R(\gamma_1) I_w(\gamma_2) \varepsilon(\gamma_2) S(\gamma_2) \mu_s(\gamma_1)}{R(\gamma_2) I_w(\gamma_1) \varepsilon(\gamma_1) S(\gamma_1) \mu_s(\gamma_2)} \right]}{\rho_a [\mu_a(\gamma_2) - \mu_a(\gamma_1)]} \quad (1)$$

Here t_a is the wall thickness, R is the measured count rates, I_w is the weight fractions of the isotopes giving rise to the gamma rays of interest (e.g., ²³⁵U and ²³⁸U), ε is the relative detector efficiencies for the two gamma rays, S is the specific activities, μ_s is the mass attenuation coefficients in the source material, ρ_a is the density of the passive absorber, and μ_a is the mass attenuation coefficients in the absorber.

As applied to cylinder-wall thickness measurements, the relative detection efficiencies (ε) are dependent only upon the energy of the gamma rays. The specific activities (S), the UF₆ mass attenuation coefficients (μ_s), the cylinder density (ρ_a), and the cylinder mass attenuation coefficients (μ_a) are all physical constants. Assuming that the material properties of the UF₆ storage cylinders are similar from cylinder to cylinder, the only variables in a thickness determination are the count rates of the two gamma rays, $R(\gamma_1)$ and $R(\gamma_2)$, and the weight fractions of the isotopes giving rise to the gamma rays, $I_w(\gamma_1)$ and $I_w(\gamma_2)$. If the measured gamma rays arise from decay of the same isotope, for example, the 258- and 1001-keV gamma rays from ^{234m}Pa (²³⁸U) decay, then the weight fractions cancel and the only variable is the count-rate ratio.

To measure t_a with good sensitivity, the absorber attenuation coefficients, $\mu_a(\gamma_1)$ and $\mu_a(\gamma_2)$, should differ by as much as possible. Because the attenuation coefficients are strongly dependent on energy only below 300 keV, at least one of the measured gamma rays should have an energy less than

300 keV. Candidate gamma rays are the 186-keV gamma ray from ^{235}U decay and the aforementioned 1001-keV and 258-keV gamma rays. Because of the weak dependence of the attenuation coefficients on energy above 300 keV, it is not possible to determine absorber thicknesses with sufficient accuracy from the ratio of the relatively intense 766- and 1001-keV $^{234\text{m}}\text{Pa}$ (^{238}U) gamma rays.

Detailed knowledge of detector efficiencies and the material properties of the cylinders may be difficult to obtain accurately. Alternatively, a calibration curve may be constructed from the ratio $R(\gamma_1) I_w(\gamma_2)/R(\gamma_1) I_w(\gamma_2)$ and the cylinder-wall thicknesses determined from the ultrasound measurements. As shown by Equation 1, a plot of thickness versus the natural log of this ratio results in a straight line. Using this line, the cylinder-wall thickness can be determined directly from the measured [count rate–weight fraction] ratio. The feasibility of this gauging method is discussed in the following section.

4.2 EXPERIMENTAL MEASUREMENTS

A 45 percent HPGe detector was used to obtain gamma-ray spectra from five UF_6 storage cylinders in the E Yard and one cylinder in the K Yard. Four of the cylinders had nominal wall thicknesses of 5/16 inch (312.5 mils), one had a nominal thickness of 5/8 inch (625 mils); and the cylinder in the K Yard had areas with wall thicknesses as thin as 1/4 inch (250 mils). The detector was shielded by a 2 5/8-inch-thick steel collimator in order to attenuate the background radiation in the cylinder yards. Calculations indicated that this collimator reduced the background from 186-keV gamma rays by about a factor of 2000 and the background from 1001-keV gamma rays by about a factor of 20. The area viewed by the detector inside the collimator was a circle having a diameter of two inches. The detector was positioned 4 3/4 inches deep in the collimator and typically placed approximately five inches from the cylinder wall. Because the analysis technique relies on the ratios of count rates, differences in the solid angle of the detector do not affect the measurements.

In conjunction with each gamma-ray measurement, a corresponding ultrasound thickness gauging of the cylinder surface was obtained. As the ultrasound thickness gauge was sensitive to a smaller area than that of the HPGe detector, five ultrasound measurements were taken: at the top, bottom, left side, right side, and center of the two-inch area viewed by the gamma-ray detector. In all but two cases (two thinned areas on the cylinder in K Yard), the five ultrasound measurements agreed to within seven mils.

Each gamma-ray spectrum was accumulated for a live time of ten minutes. In this time, typical peak-area uncertainties arising from counting statistics were approximately 5 percent for the 186-keV gamma ray, 15 percent for the 258-keV gamma ray, and less than 1 percent for the 1001-keV gamma ray.

Several background spectra were also collected by aligning the collimator and detector vertically so that gamma rays emitted from the cylinders could not strike the detector directly.

4.3 RESULTS

It was immediately determined that the 258-keV gamma ray was not intense enough to allow a sufficiently precise determination of the count-rate ratio $R(258 \text{ keV})/R(1001 \text{ keV})$ within a ten-minute

collection time. The 15 percent statistical uncertainty in the 258-keV count rate lead to an unacceptably large uncertainty in thickness of ± 100 mils for a 5/16-inch (312.5-mil) cylinder wall. This complicates the measurements; to determine cylinder-wall thicknesses from the $R(186\text{-keV})/R(1001\text{-keV})$ count-rate ratio, the weight fraction of ^{235}U must be known for each cylinder.

The data obtained from the gamma-ray measurements are listed in Table 2. For each measurement, the cylinder identification number, run identification number, ^{235}U enrichment fraction [$I_w(^{235}\text{U})$], count rates of the 186-, 258-, and 1001-keV gamma rays [$R()$], ratio $R(1001) \cdot I_w(^{235}\text{U}) / R(186) \cdot I_w(^{238}\text{U})$, natural logarithm of this ratio, and average cylinder-wall thickness from the ultrasound measurements are given in Table 2.

A plot of the natural logarithm of the ratios versus the average thicknesses obtained from the ultrasound measurements is given in Figure 23. With the assumption (better than one percent) that, for natural or depleted uranium, the weight fraction of ^{238}U equals one, the data points in such a plot should exhibit a straight line as shown by Equation 1. The observed deviations from straight-line behavior are discussed in the following paragraphs.

Cylinder 7340 in the K Yard was thermally imaged during March 1996 and was shown to have a non-horizontal UF_6 fill line. Results obtained on the north side of this cylinder with the detector situated well below the fill line (leftmost three open triangles), follow the expected trend between cylinder-wall thickness and count-rate ratio. On the south side of the cylinder, however, the detector was situated very close to the fill line. The outlying open triangles at the far right of the graph are measurements taken on the south side of the cylinder. Because the material viewed by the detector did not fulfill the requirements of an "infinite" source, the points taken on the south side of Cylinder 7340 were eliminated from the data set.

The top two data points (solid squares), which also show rather poor agreement, are measurements taken on Cylinder 597 in the E Yard. This cylinder had a nominal 5/8-inch storage cylinder-wall thickness. Because of the increased gamma-ray attenuation in the thicker cylinder wall, the statistical uncertainties in the 186-keV count rates were increased to approximately 15 percent in a ten-minute count time. The increased statistical uncertainty is indicated by the larger error bars associated with these data points. The east side of Cylinder 597 was sited close to a double-stacked row of UF_6 storage cylinders while the west side of the cylinder was open. The count-rate ratio $R(1001)/R(186)$ was found to be approximately 35 percent larger on the east side of the cylinder than on the west side.

To investigate the discrepancy between the measurements on the east and west sides of Cylinder 597, two background measurements were taken. In these measurements, the detector and collimator were positioned as close to the measurement location as possible but were pointed vertically so that gamma rays emitted from the UF_6 storage cylinders interacted with the detector by passing through the collimator. The background count rates are listed in Table 2 as Cylinder 597 bck (background), runs "w" (west) and "e" (east). The 1001-keV background on the east side of the cylinder was a factor of 1.75 larger than the background on the west side. No statistically significant background for the 186-keV gamma ray was observed on either side of Cylinder 597. As described above, the steel collimator surrounding the detector was much more effective at attenuating the background from low-energy gamma rays than at attenuating the background from high-energy gamma rays.

Table 2. Gamma-Ray Measurements

Cylinder	Run ID	I_w (^{235}U)	R (186)	R (258)	R (1001)	Ratio	ln(Ratio)	Thick	Comment	Sig
119130	e	0.001967	6962	1544.8	62406.7	0.017632	-4.03804	343	South end of cylinder	0.0442
119130	e2	0.001967	6693	1446	63975.7	0.018802	-3.973805	325	Nearly same as 119130e	0.0467
5277	e	0.001962	6604	1183	63924	0.018991	-3.963772	341	South end of cylinder	0.0471
5277		0.001962	6708	1593	65459	0.019146	-3.955668	343	West side of cylinder	0.0483
5277	s	0.001962	6700	1600	65122	0.01907	-3.959636	346	East side of cylinder	0.0486
5277	s2	0.001962	6146	1355	65380	0.020871	-3.869376	346	East side of cylinder (repeat)	0.0526
152675	b	0.00711	23052	1247	64410	0.019866	-3.918737	328	West side of cylinder	
152675		0.00711	23712	1182	61910	0.018564	-3.986553	334	East side of cylinder	0.0153
114995	a	0.002487	7673	1283	65381	0.021192	-3.854154	335	Cylinder in rows (right) side	0.0387
114995	b	0.002487	7192	1174	64519	0.022311	-3.802688	350	Cylinder in rows (left) side	0.048
597		0.001271	2058	480	39340	0.024296	-3.717444	625	West side of cylinder	0.134
597	e	0.001271	1675	921	43118	0.032718	-3.419824	620	East side of cylinder	0.176
7340		0.001956	7992	1485	68117	0.016671	-4.094068	250	K Yard, thin spot	0.0404
7340	b	0.001956	6929	1422	65608	0.018521	-3.988872	290	K Yard, thicker spot	0.0461
7340	f	0.001956	4191	685	33527	0.015648	-4.157442	290	K Yard, north side thicker (repeat)	0.0552
7340	c	0.001956	7032	887	40909	0.011379	-4.475975	319	K Yard, south side of cylinder	
7340	d	0.001956	6531	930	40592	0.012157	-4.409843	319	K Yard, south side (repeat)	0.0409
7340	e	0.001956	7130	929	44543	0.01222	-4.40471	318	K Yard, south side (left)	0.0376
597	w		~0	~0	8947				West side background	
597	e		~0	~0	15681				East side background	
114995	bck		~0	~0	23228				Background (in stacks)	
597	w_corr	0.001271	2058	480	30393	0.01877	-3.975474	625	West background-corrected	
597	e_corr	0.001271	1740	921	27437	0.020042	-3.909944	620	East background-corrected	
114995	a_corr	0.002487	7673	1283	42153	0.013663	-4.29308	335	Background-corrected	
114995	b_corr	0.002487	7192	1174	41291	0.014278	-4.249003	350	Background-corrected

FIGURE 23. PLOT OF THE NATURAL LOGARITHM OF RATIOS VERSUS AVERAGE THICKNESSES FROM ULTRASOUND MEASUREMENTS

After correcting the measured count rates for background, the $R(1001)/R(186)$ count-rate ratio was reduced to only 11 percent larger on the east side of the cylinder, which is well within the uncertainty based on counting statistics. The corrected 1001-keV count rates and the corrected count-rate ratios are given in Table 2. Note that the "background" used for the subtraction was not quantitatively correct because the background and measuring geometries were slightly different (*i.e.*, the detector was pointed at the cylinder while measuring the count-rate ratio and pointed vertically while measuring the background). However, it is clear that the east side/west side count-rate discrepancy near Cylinder 597 can be qualitatively understood as arising from different background conditions.

An absolute determination of the cylinder-wall thickness was attempted using Equation 1. With γ_1 equaling 1001 keV and γ_2 equaling 186 keV, the following values were taken for the physical constants: $S(\gamma_1) = 74.2 \text{ s}^{-1}$, $S(\gamma_2) = 4.68 \times 10^4 \text{ s}^{-1}$, $\mu_s(\gamma_1) = 0.0704 \text{ cm}^2/\text{g}$, $\mu_s(\gamma_2) = 1.027 \text{ cm}^2/\text{g}$, $\rho_a = 7.86 \text{ g/cm}^3$, $\mu_a(\gamma_1) = 0.0594 \text{ cm}^2/\text{g}$, and $\mu_a(\gamma_2) = 0.1447 \text{ cm}^2/\text{g}$. Using a detector-efficiency ratio $\epsilon(\gamma_2)/\epsilon(\gamma_1)$ of 3, an absolute cylinder-wall thickness of approximately 610 mils for the 5/16-inch

(312.5-mil) Cylinder 114995 was obtained. However, as previously discussed, background from the 1001-keV gamma ray skews the observed count-rate ratio and must be taken into account.

Correcting for the 1001-keV background measured near Cylinder 114995 (see Table 2) gave an absolute thickness of approximately 350 mils for this cylinder which was in much better agreement with the nominal (312.5 mils) thickness and with the thickness determined from the ultrasound measurements (334 mils). The background-corrected count rates and count-rate ratios for runs "a" and "b" on Cylinder 114995 are given in Table 2.

A total of ten measurements (diamond-shaped points shown in Figure 23) were taken on four cylinders: 119130, 5277, 152675, and 114995. Each had a nominal wall thickness of 312.5 mils. The ultrasound measurements gave an average thickness of 339 mils $\pm$ 8, corresponding to a relative uncertainty of 2.4 percent. The gamma-ray measurements gave a relative uncertainty of about 12 percent, a much less precise result and a significantly larger uncertainty than the 5 percent due to counting statistics alone. Much of the additional uncertainty was likely caused by differences in background conditions. Measurements taken close to neighboring cylinders exhibited an increase in the 1001-keV yield and a correspondingly larger extracted value for the container-wall thickness.

4.4 CONCLUSIONS

Two preliminary studies were performed to determine the feasibility of gauging the wall thicknesses of UF₆ storage cylinders with gamma-ray detection techniques. The results with a 45 percent HPGe detector viewing a two-inch circle on the cylinder walls can be summarized as follows:

- A. Measurements with a single gamma-ray detector required a ten-minute count time to obtain five percent counting statistics on the thin-walled (5/16-inch) cylinders. Several (five-ten) ultrasound measurements could be taken during this counting time.
- B. Within the ten-minute counting time, the 258-keV ^{234m}Pa (²³⁸U) gamma ray was not intense enough to be useful. This complicates the measurement in that it is necessary to know the ²³⁵U enrichment fraction along with the count-rate ratio.
- C. The measurements were very sensitive to background 1001-keV gamma rays from nearby cylinders. To obtain accurate and precise results, corrections need to be applied for each source-detector-background geometry.
- D. The measurements were sensitive to the presence of the UF₆ fill line. To ensure consistency, measurements should be taken in an infinite source geometry away from the fill lines.

4.5 FOLLOW-ON WORK – Sodium-Iodide Array for Gamma-Ray Attenuation

To decrease the scanning time of the gamma-ray attenuation measurement technique, the single high-resolution HPGe detector could be replaced by an array of low-cost scintillation (sodium iodide or cesium iodide) detectors. More than 20 such detectors could be instrumented for the cost of a single HPGe detector, vastly increasing the area that can be scanned at one time. In addition, some scintillation detectors can be packaged more compactly than HPGe detectors, allowing better access to the hard-to-reach areas around the cylinders.

4.6 REFERENCES

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Chapter 5. Compton Scatterometry

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5.1 INTRODUCTION

The Compton scatterometry technique¹ employs a highly collimated and intense source of gamma rays or X rays for high-resolution, three-dimensional imaging. Compton-scattered photons are detected through a focusing collimator that selects a volume of the material to be imaged (Figure 24). The detector and collimator are moved laterally to image parallel to the surface of the material and moved transversely to image the depth. State-of-the-art, high-resolution systems² allow a 70-cubic-centimeter volume to be scanned in about one minute, with a spatial resolution approaching 25 micrometers.

This technique could be applied to thickness measurements of UF₆ storage cylinder walls. The difference in electron density between the cylinder material and solid UF₆ gives rise to an approximate factor of two difference in the Compton scattering probability. (The difference in electron density between the cylinder wall and gaseous UF₆ produces a much greater difference in scattering probability.) A highly collimated system scanning transversely could locate the point where the scattering yields changes.

5.2 TEST

Typical source activities used in Compton scatterometry are in the range of 40–600 curies. To avoid handling such a highly-active source, a simple test of the technique was performed using an uncollimated, 0.5-millicurie, cobalt-60 (⁶⁰Co) source. Two 10-minute, HPGe gamma-ray spectra were measured in an identical geometry on Cylinder 597 with and without the source present (Figure 25). The spectrum taken with the source in place (upper curve) gave a background that was more than two times greater. This increase in background was due primarily to multiple scattering from the UF₆, the cylinder, the detector mount, etc. The broad peak at 265 keV in the upper spectrum arises from gamma rays scattered by the cylinder or the UF₆ directly into the detector. Quantifying this peak as a function of scanning position would allow a precise determination of the location where the interface between the UF₆ and the cylinder wall occurs.

FIGURE 24. COMPTON SCATTEROMETRY DETECTOR AND COLLIMATOR

Reference Spectrum

Datasource: 597SORCX.CNF
 Live Time: 600 sec
 Real Time: 930 sec
 Acq. Start: 5-23-96 4:31:05 PM
 Start: 1 : 0.95 (keV)
 Stop: 2048 : 1338.93 (keV)

Compare Spectrum

Datasource: 597NOX.CNF
 Live Time: 600 sec
 Real Time: 657 sec
 Acq. Start: 5-23-96 4:51:06 PM
 Start: 1
 Stop: 2048

FIGURE 25. TEN-MINUTE HPGE GAMMA-RAY SPECTRA WITH (UPPER CURVE LABELED "DATA") AND WITHOUT (LOWER CURVE LABELED "COMPARE") THE ^{60}Co SOURCE PRESENT

5.3 CONCLUSIONS

Compton scatterometry holds promise as a simple, quick method to determine cylinder-wall thicknesses to an accuracy of approximately ten mils.

5.4 FOLLOW-ON WORK

A typical UF_6 storage cylinder has a surface area of approximately 1.3×10^5 square centimeters. At the scanning rate quoted previously for the high-resolution Compton-scatterometry system, a complete scan of a cylinder having a wall thickness over 2.5 millimeters (100 mils) would require approximately eight hours. In practice, the scanning time may be increased due, for instance, to space limitations near the bottom of the cylinders. However, if a precision of 25 micrometers (1 mil) is not required, a complete scan may require less time. A LANL group¹ having expertise in Compton scatterometry studies is interested in applying this technique to measure wall thicknesses of UF_6 storage cylinders.

5.5 REFERENCES

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Chapter 6. Active Infrared Inspection

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6.1 INTRODUCTION

This chapter describes the results of a field demonstration conducted during March 25–29, 1996, at the Oak Ridge K-25 gaseous diffusion plant. The objectives of this field demonstration are listed below:

- Demonstrate the operational capability of a commercial, active IR system.
- Inspect UF₆ storage cylinders under actual field conditions.
- Evaluate the measurement performance of the commercial system for quantitative inspection of cylinder-wall thickness.
- Determine what changes, if any, are needed to make the system suitable for inspecting UF₆ storage cylinders.

6.2 TECHNICAL APPROACH

A pulsed thermal-wave imaging (PTWI) system was used for the field demonstration. This system was set up to heat a cylinder surface area of approximately one square foot using a brief (less than five-millisecond) pulse from a set of high-energy optical flash lamps. The flash heating at the surface caused a thermal pulse to propagate into the material. Discontinuities in the subsurface thermal properties of the material (*i.e.*, a steel-oxide boundary or the steel–air or steel–UF₆ interface at the interior wall of the cylinder) caused some of the pulse energy to be reflected back toward the sample surface, affecting the local cooling rate at the surface. By monitoring these anomalies in the cooling rate, subsurface defects can be identified. Additionally, the time, relative to the flash, when these anomalies occurred was correlated to the depth of the subsurface scatterer.

The PTWI method has been used successfully to detect hidden corrosion or material loss on the interior surface of steel pipes and storage tanks. However, the problem under study is somewhat different. Corrosion that initiates from the inside of the cylinder and then propagates out does not appear to be a significant problem at the K-25 Site. In fact, it is the opposite problem that is prevalent, that is, corrosion on the outside of the tank propagates through the cylinder wall to the interior. The situation is complicated because the tanks have been exposed to weather for long periods of time and thus may appear uniformly corroded. Therefore, the challenge was to discriminate surface corrosion from much deeper corrosion where the accumulated byproduct may mask severe thinning in the cylinder wall.

The cylinder data were viewed as a time sequence of images or as a set of amplitude-versus-time plots where discrete 5- × 5-pixel regions of high and low amplitude could be compared directly. In particular, the early and asymptotic cooling behavior in response to flash heating was examined. Whenever possible, regions that displayed anomalous behavior in the time sequence were chosen for plotting.

6.3 EQUIPMENT

A modified version of the commercial EchoTherm® system was used for the field demonstration. The system included two 6.4-kilojoule, 8-inch linear flash lamps that were used to instantaneously heat the

cylinder surface over a 10- × 12-inch area. The flashtubes were housed in parabolic reflectors designed to provide a uniform beam profile at the target surface. Light from the flash was contained by a reflective shroud that also redirected stray light onto the target surface. Each flashtube was powered by a Balcar Starflash 3, 6.4-kilojoule power supply. The duration of the flash pulse was approximately five milliseconds. The EchoTherm sequential-charging controller was used to allow both power supplies to charge from a single 110-volt alternating-current source without drawing excessive current.

A forward-looking IR (FLIR) Prism DS camera was used to acquire the images. Twelve-bit, digital data (60-hertz-frame rate) from the camera was captured by the computer using the EchoTherm data-acquisition card, which was housed in a portable, 133-megahertz Pentium, personal computer with a 64-megabyte random access memory (RAM). The computer was used to synchronize flash heating and data acquisition and to analyze the data after acquisition. All operations were performed within the EchoTherm program.

6.4 PROCEDURE

The cylinders were not painted or otherwise prepared for imaging. The tank surfaces were typically covered with an enamel paint or, more commonly, covered with a layer of corrosion. During the imaging process, caution was taken, in compliance with health physics restrictions, to avoid contact between either the operators or the equipment and the cylinders. At each cylinder, the computer, power supplies, and support electronics were positioned approximately five feet from the cylinder, and the imaging head was mounted on a tripod and moved to within a one-inch proximity of the cylinder surface.

Two IR cameras were initially used to image the surface, an Inframetrics Thermacam and a FLIR Prism DS. Each camera was equipped with a 32-millimeter, wide-angle lens and a standard 16-millimeter lens. At the standoff distance provided by the imaging head, an area of approximately 8 × 6 inches could be viewed with uniform illumination. At the low-ambient temperature in the yards, the FLIR camera offered more contrast and higher signal-to-noise content in the images, so the FLIR was used to acquire the images included in this report.

The image data was acquired continuously beginning 64 milliseconds before flash heating and lasting 6.6 seconds after flash heating. Ten discrete gates of image sums were then formed from the accumulated images with each gated image comprising the sum of four consecutive image frames of 16.6-millisecond duration. The purpose of the summing and gating was to increase signal-to-noise content of the images and to reduce the storage requirements for archiving the images (the size of the raw data set was 54 megabytes while the size of the gated set was approximately 2 megabytes). The exact time when the gates were acquired was -0.067 milliseconds (*i.e.*, immediately before the flash) and then 0.083, 0.234, 0.384, 0.534, 0.684, 0.934, 1.268, 3.270, and 6.607 milliseconds. Continuous amplitude-versus-time data was also acquired for discrete points in the data set.

6.5 OPERATIONAL CONSIDERATIONS

At the site, several issues emerged that impacted the outcome of the demonstration. In general, the problems that emerged included (a) positioning of the imaging head on high- and low-cylinder sections, (b) electrical power constraints, (c) lack of registration markings on the cylinders, and (d) limited

mobility of the operator station on the gravel and dirt surfaces. These problems can be corrected with only minor modifications to the system for future applications.

6.6 RESULTS

For the feasibility study, it was not possible to make independent measurements of cylinder-wall thickness. Instead, data were acquired and, when possible, compared to quantitative ultrasonic measurements that were made at the site. Data were acquired on seven cylinders. Figures 26 and 27 are examples of data acquired on uncorroded and corroded cylinders, respectively. Figure 26 illustrates the rapid cooling and asymptotic convergence characteristic of uncorroded regions. Regions A and B in Figure 27 correspond to regions where severe corrosion had occurred. In these regions, cooling was retarded and the traces did not converge in the 6.6-second-sampling interval. Several cylinders exhibited more complex behavior than shown in either of these figures. For example, areas of moderate surface corrosion may display some retarded cooling and offset, but asymptotically, they converge and display similar behavior to uncorroded regions. Numerous examples of such intermediate behavior are presented in this report.

In most cases, sufficient thermal contrast could be extracted without painting or otherwise preparing the cylinders. In particular, corroded regions provided sufficient signals for further analysis. On painted cylinders, relatively quiescent regions yielded lower contrast and were difficult to discriminate from each other. However, such regions were unlikely candidates for significant cylinder-wall thinning. Regions that displayed the most rapid return to equilibrium after flash heating had generally completed

FIGURE 26. *TEMPERATURE-TIME PLOTS OF AN UNCORRODED CYLINDER. Each trace represents the average of a 25-pixel region over a 6.6-second interval. The average cylinder-wall thickness in this area was measured ultrasonically to be 0.317 inches compared to the nominal cylinder-wall thickness of 0.345 inches. The curves are nearly indistinguishable except for their initial amplitudes.*

FIGURE 27. TEMPERATURE-TIME PLOTS OF A CYLINDER WITH CORROSION. Each trace represents the average of a 25-pixel region over a 6.6-second interval. Regions A and B (red and blue, respectively), which are over apparent regions of corrosion, exhibit retarded cooling behavior. Region B exhibits the slowest cooling at earlier times (less than one second) but appears to converge to equilibrium more quickly than Region A.

the cooling cycle in less than one second. Both apparent corroded and uncorroded regions could be seen to exhibit this rapid cooling behavior. However, regions of severe corrosion cooled much more slowly.

6.7 RECOMMENDATIONS FOR FUTURE WORK

In order to perform more effective quantitative measurements of cylinder-wall thicknesses, more detailed study of the temperature-time response of the cylinder walls will be required. A library of calibration curves, based on sample cutouts of known thicknesses and with known amounts of corrosion, should be developed for reference. Both painted and unpainted cutouts should be included. Development of a predictive model based on this library will then be used to allow actual thickness measurements to be made in the field.

Better resolution of deeper features and improved measurement capability could be obtained by increasing the amount of energy used to heat the cylinder walls. For the demonstration, two 6.4-kilojoule flash lamps were used. For future applications, four lamps should be used.

An alternate configuration of the system, based on a moving continuous heat source, should be investigated. Such a system could be mounted on a robotic crawler and would image axial stripes. This configuration would allow access to the entire cylinder surface, including the underside, and could significantly decrease the time required to inspect a single cylinder. Furthermore, the use of a robotic system would reduce personnel exposure to radiation.

Chapter 7. Passive Thermal Infrared Imaging

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7.1 INTRODUCTION

7.1.1 Background

Thermal infrared (IR) imagers detect energy that is emitted from a surface. This energy is a function of the surface temperature, the efficiency or emissivity of the surface, and the effects of the atmosphere between the surface and the camera.

A UF₆ storage cylinder consists of three main components. The cylinder itself is a heavy steel tank with walls on the order of 0.5 inches thick. A filled cylinder normally contains 10 or 14 tons (depending on the type of cylinder) or just more than 60 percent by volume of solidified residue from the uranium enrichment process. Just more than a third of the cylinder volume is a low-pressure, gas-filled space. A simplified model of the energy transfers might consist of these three cylinder components and the surrounding environment.

The steel wall of the cylinder conducts heat well and can thus disperse temperature gradients rather quickly. The wall is in close contact with the environment, the solid material, and the gas space and will respond to their influences. The solid contents of the cylinder store energy in a large mass, are not directly exposed to the environment, and change temperatures more slowly than the steel cylinder. The solid contents conduct heat to (night) and from (day) the cylinder. Additionally, the material inside the cylinder produces some heat. Thus at night the metal is warmed where it contacts the solid contents. The amount of warming is affected by the thermal conductivity of the metal. This varies with thickness and, possibly, with changes to the integrity of the steel due to corrosion, cracks, and other damage.

During the day, sunlight presents a strong control on the surface temperatures of the cylinder and can obscure weaker signals from internal heat-transfer processes. At night, thermal IR transfers between the cylinder and its environment are significant. With good weather, the air is cool and the ground is relatively warm. In the instance of an isolated empty cylinder near the ground, the bottom of the cylinder would be warm and the top would be cool. In general, small temperature changes resulting from differences in target properties are easiest to observe at night when extraneous energy fluxes, especially those between the subject and its environment, are minimized.

For cylinder inspection purposes, it is not so much the absolute temperature that is of interest but rather the variation of temperatures caused by changes in the cylinder surface, wall, or contents. The cylinder itself is examined to detect changes in the amount of heat conducted through the cylinder wall. Heat conduction is only one of the heat-transfer mechanisms, but it is the only one likely to vary at small spatial scales at night. If one is looking for localized temperature differences, such as those due to handling damage, temperature changes can be expected to be dominated by changes in heat conductivity. If one is looking for gradual changes, such as the result of wall thinning due to weathering, temperature differences must be separated from variations induced by environmental factors. Both of these types of changes are important.

7.1.2 Objectives

The broad objective of this study was to provide an initial demonstration and evaluation of the ability of passive thermal IR imaging to support the management of the UF₆ storage cylinders. The following issues were addressed:

- Cylinder-wall thicknesses (the effect of thinning or weathering).
- Detection of handling damage or localized weathering damage.
- Determination of the fill level and distribution of cylinder contents.

This study employed one particular sensor operated under a limited set of environmental conditions. Many of the tests were performed on targets of opportunity. It was not anticipated that thermal IR would be successful in all aspects of the evaluation. Nevertheless, the tests were of value by enabling future focus to be placed on the more successful and beneficial uses of the technology.

7.2 MEASUREMENTS AND ANALYSIS

Thermal IR images were obtained on individual cylinders selected by the K-25 cylinder yard staff. Additional imagery was obtained from an elevated position to demonstrate phenomena of interest.

7.2.1 Instrumentation

An AMBER Instruments Radiance PM thermal IR camera was leased from a sales representative for this study. This camera uses a 256- × 256-pixel InSb (indium antimonide) focal plane array and was chosen because it provides the highest thermal sensitivity on the commercial market. The sensitivity, as stated in the manufacturer's specifications, is 0.025 degrees Celsius. The array detects energy in the three- to five-micrometer spectral band, and the system is internally calibrated so that conversion from radiance to effective radiant temperature can be performed. A lens with a 50-millimeter focal length was supplied providing a field of view of 11 degrees square. The 12-bit precision data were recorded digitally (in 16 bits) on a PCMCIA Type II memory card and downloaded to a computer for analysis and backup. The memory card uses a proprietary format and a software package developed by AMBER Instruments called AmberTherm, which is used to read the card and download the data to the computer.

7.2.2 Locations and Targets

Imagery was obtained at three cylinder storage yards at the K-25 Site: E, K, and J Yards. In the E Yard, seven cylinders were selected by the K-25 staff and moved to an area with easy access. One of these cylinders was empty and the other six were chosen to represent a variety of conditions. Three of them were the same type of cylinder (48OM) having different assay contents while the others were different cylinder types. Other images of the E Yard were obtained both at ground level and from an elevated position. In the K Yard, the main topic of study was Cylinder 7340. This cylinder had experienced considerable weathering corrosion, and the bottom was considerably thinned. Another cylinder, Cylinder 7953, had breached and was patched. Images of the K Yard from an elevated position were also obtained. At the J Yard, five cylinders with known problems were examined: two were severely damaged, one had a deep erosion pit, one had a high-pressure gas space, and one contained a "heel"

(the remnants of material within an empty cylinder). Other imagery was obtained that is not discussed. These additional images did not contribute significantly to the study objectives.

7.2.3 Weather Conditions

Images were obtained during the nights of March 25–29, 1996. Measurements were made under favorable conditions on March 25 and 26. However, the night of March 26 was considerably colder than March 25, and many surface temperatures fell to below freezing by the end of the night. March 27 and 28 were generally damp with scattered showers. With one exception, data from these nights were not reported. Data were taken under cloudy but otherwise good conditions on March 29.

7.2.4 Analysis

AmberTherm is a proprietary software package designed for analyzing data from AMBER products. A demonstration copy of the software was provided. It allowed data to be read from the memory card and the measurements converted into apparent radiative temperatures. It did not allow for the export of analysis products. Therefore, data could be analyzed in temperature units, but no output through a computer was possible. The image data, in the form of raw digital counts, were transferred to the Remote Sensing Laboratory (RSL) image-analysis systems and analyzed using the ERDAS Imagine, Version 8.2, software. However, analysis in temperature units was not possible.

Most of the analyses were qualitative. In some cases, apparent temperatures of the cylinder surfaces were reported. In these instances, temperatures were determined using the AmberTherm "spot" function or cursor using an assumed emissivity of 0.95. More detail will be given in the following section.

7.3 RESULTS

7.3.1 Thinning

Cylinder 7340 in the K Yard had undergone severe thinning due to ground-contact weathering. The bottom had thinned to less than half of the original thickness as indicated by ultrasound measurements. This cylinder was rolled on its side in order to expose its thinned bottom. Unfortunately, space was so constrained that a significant area of the cylinder wall could not be imaged from the side. Weak temperature gradients were observed, but these may have well been caused by other factors. It is difficult to predict in any detail just how temperatures would vary when a filled cylinder is rotated on its side. The thermal effects of thinning are likely to be weak and difficult to separate from other variations. The collected data were inconclusive.

7.3.2 Localized Damage

7.3.2.1 E Yard

Local variations of temperature were observed, but with a single exception, these could be easily associated with variations of the surface: surface stains, paint, corrosion flakes, etc. In the field, thermal IR did not seem to provide any advantage over a careful visual inspection for localized damage.

One dramatic exception was found on Cylinder 119486, which is shown in Figure 28. The top portion of this figure shows the thermal IR image, and the bottom shows a scanned color photograph of the same area on the cylinder. A scrape is visible at the level of the lifting tabs that presumably occurred during handling. This scrape was virtually invisible to the eye until the oxidation was removed for ultrasound measurements. Even then, it was barely visible and would have likely remained unnoticed by inspectors unaware of its presence. Ultrasound measurements did not indicate localized thinning, so the damage was either superficial or too recent to initiate corrosion. The phenomenon causing the apparent temperature change has not been identified but seems to be associated with a decrease in the emissivity of the surface. While it is likely that the damage is of little consequence in this case, impact on the cylinder wall during handling is known to accelerate localized corrosion and deterioration of the cylinder integrity.

7.3.2.2 K Yard

An image of Cylinder 116797, taken through a chain link fence, is shown in Figure 29 (see page 57). The cylinder was breached and patched. Although partially obscured in the image by the fence and the patch straps, the cylinder beneath the patch was warmer than nearby parts of the cylinder. The reason for the temperature difference has not been determined. It might be due to materials or surface preparation related to the patch as opposed to any leakage of the contents from the cylinder.

7.3.2.3 J Yard

Three damaged cylinders in the J Yard were examined, and the images are presented in Figure 30 (see page 58). The damage due to a severe dent is indistinguishable from other thermal variations in the image at the top right (Cylinder 151642). A dent on Cylinder 151863 is weakly visible in the close-up image. Finally a deep corrosion pit on the end of Cylinder 153676 is not apparent. The observational conditions were not optimal: the cylinders were covered with tarpaulins because of rainy weather until late afternoon before the measurements were made. Under these conditions, passive thermal IR was not an effective method for detecting cylinder damage.

7.3.3 Fill

7.3.3.1 E Yard

The seven test cylinders in the E Yard were examined closely. Additionally, distant imaging of the E Yard stacks was done from both the ground and an elevated position. The test cylinders and the imagery from the stacks are discussed separately.

A. Test Cylinders

It was immediately apparent that thermal IR imagery could provide important information on the level and distribution of the fill material in the cylinders. Figure 31 (see page 59) shows the ends of the seven test cylinders. Cylinder 143997 was empty and served as a control. Cylinder 114921 was rolled several months before this study, and its contents were seen to have regained a horizontal fill line. Cylinder 152675 was rolled counterclockwise slightly, and the general fill pattern was seen to be rotated by the same amount. However, the solid-gas boundary was somewhat

CYLINDER 119486

FIGURE 28. *THERMAL IR IMAGE AND PHOTOGRAPH OF CYLINDER SCRAPE*

more diffuse than in the other cylinders and appeared to be less uniform across the cylinder end. To better illustrate the thermal transition between the filled and empty parts of a cylinder, the original gray-scale image can be displayed as a color image. Both color and gray-scale images

CYLINDER 116797 (480M)

FIGURE 29. CLOSE-UP VIEW OF A CYLINDER BREACH PATCH

are shown in Figure 32 (see page 60). The fill boundary follows a curved line at the end of the cylinder.

The transition (during the night) from the warm part of the cylinder containing the solid UF_6 to the cooler part of the cylinder adjacent to the gas-filled space is referred to as the fill line or fill boundary. The observed fill boundaries were not horizontal. Material sublimates near the walls of a cylinder and tends to recondense near the center. Hence a meniscus-like feature forms. The actual level of fill was not defined in the images with precision; a transitional area a few inches wide was observed. Even if the boundary between the solid and gaseous constituents was distinct, it would be blurred thermally by heat conduction along the cylinder wall resulting from the temperature gradient. It was not possible to inspect the inside of the cylinder, and it is not exactly known how this transitional area correlates to the actual fill boundary. Therefore, the accuracy with which passive thermal IR determines the fill-line position quantitatively is still unknown.

The characteristics of the fill line were examined quantitatively using vertical temperature profiles. To illustrate the general trends, Figure 33 (see page 61) shows vertical profiles of Digital Numbers (DN) from three of the cylinders shown in Figure 31. (The positions are in reference to arbitrary points on the ends of the cylinders. Comparisons of position between cylinders are only grossly accurate.) The empty cylinder (Cylinder 143997) had a linear decrease of temperature of about 6 degrees Celsius (8.5–2.5 degrees Celsius) from bottom to top. The filled portions of the other

CYLINDER 151863 DENT

CYLINDER 151642 DENT

CYLINDER 153676 DEEP PIT

FIGURE 30. DAMAGED CYLINDERS IN J YARD

cylinders showed weak decreasing temperatures with increasing height. The upper portion of the cylinders showed a strong, nearly linear, decrease of temperature with height.

If it is hypothesized that the weak trend is where the cylinder contacts the solid material and the stronger linear trend is where the cylinder is adjacent to the gas space, then the point where the two trends meet should be the interface. Figures 34, 35, and 36 (see pages 61, 62, and 62) illustrate an effort to exploit this hypothesis for the three type-48OM cylinders in the test set. For each cylinder, linear regression equations were fit to the two segments of the vertical temperature profile: one for the filled portion and one for the "empty" portion of the cylinder. The segments that were used are indicated by the vertical lines in the figures. The resulting regression lines are also

143997
(EMPTY)

5277

119130

119486

128209

152675

114921

E YARD TEST CYLINDERS

FIGURE 31. THERMAL IR IMAGES OF THE SEVEN E YARD TEST CYLINDERS

THERMAL IR IMAGE

PSEUDO COLOR VERSION

CYLINDER 152675

FIGURE 32. THERMAL IR IMAGE AND PSEUDO-COLOR
VERSION OF SLIGHTLY ROTATED CYLINDER
(CYLINDER 152675) FILL LINE

FIGURE 33. VERTICAL TEMPERATURE PROFILES OF CYLINDER-ENDS. Three E Yard 480M test cylinders.

FIGURE 34. TEMPERATURE PROFILES AND LINE-FITTING RESULTS OF CYLINDER 119130

FIGURE 35. *TEMPERTURE PROFILES AND LINE-FITTING RESULTS OF CYLINDER 114921*

FIGURE 36. *TEMPERTURE PROFILES AND LINE-FITTING RESULTS OF CYLINDER 119486*

shown. The percent of fill by volume is given by the intersection of the two lines and is 42, 46, and 42 percent for Cylinders 119130, 114921, and 119486, respectively. The actual fill amounts are 60, 63, and 59 percent, respectively. The intersection positions corresponding to these fill amounts are indicated by the vertically dashed lines; they are located in areas bereft of features in the temperature profiles.

The differences between calculated and measured fill levels in the three cylinders were nearly constant (17 percent). This apparent high degree of precision (but low accuracy) may be exploitable. The realistic distribution of solid UF_6 within the cylinders was described earlier. It remains to be determined whether the material deposited in the middle of the cylinders can account for an additional 17 percent of the volume. If it can, then the technique used to derive the fill line, or more precisely, the contact of the fill boundary with the cylinder end, may be an appropriate way to quantitatively measure fill volumes.

The regression-based procedure indicated a fill level less than halfway up the cylinder end; however, the fill levels on the raw thermal IR images appeared to be more than halfway up. To explore this apparent difference between the displayed image and the surface temperature analysis, the black and white image of Cylinder 119130 was artificially colored by assigning ten colors to the measured range of temperatures. The original and the colored images are shown in Figure 37. The colored image shows that a strong temperature gradient exists at the middle and upper portions of the cylinder: the visual fill line is actually in the midst of a region of temperature change, not at the transition. This effect is shown again in Figure 38 where an image of stacked cylinders in the E Yard is used. In this image, the upper row of cylinders shows much stronger temperature changes than the bottom row, but the effect is similar.

The variation of cylinder-end temperatures with the assay of the fill materials was examined for the E Yard test cylinders. No correlation was observed with temperature for ^{235}U assay values ranging from 0.1967 to 0.7110 percent. Several images were obtained from a position about 35 feet above the cylinders as shown in Figure 39 (see page 65). Once again, there was no correlation with the ^{235}U assay. The top surfaces of all filled cylinders were within about one degree Celsius of each other. The temperatures of the cylinder tops seemed to be controlled by heat loss to the environment.

THERMAL IR IMAGE

PSEUDO COLOR VERSION

FIGURE 37. THERMAL IR IMAGE AND PSEUDO-COLOR VERSION OF THE TEMPERATURE PATTERN ON THE END OF CYLINDER 119130

THERMAL IR IMAGE

PSEUDO COLOR VERSION

FIGURE 38. THERMAL IR IMAGE AND PSEUDO-COLOR VERSION OF TWO ROWS OF CYLINDERS IN E YARD

The visual fill line along the edge of the cylinder was as distinct as on the ends. This is seen in the oblique views of some of the cylinders in Figure 31. However, because of the curvature of the cylinder, it is exposed to significantly different thermal environments from bottom to top, making comparisons much harder. Also, the narrow field of view of the camera combined with access limitations made it difficult to image much of the lengths of the cylinders from the side.

B. E Yard Stacks

Images of the E Yard stacks were obtained to get a more realistic look at the cylinders within their usual environment. Two examples are shown in Figure 40 (see page 66). The image on the top was obtained from ground level and shows a clear difference between the upper and lower rows. (This is the same image as the left panel of Figure 38.) The cylinders in the top row had end-temperature differences between their tops and bottoms of about 5–6 degrees Celsius. The bottom-row cylinders were thermally more uniform; there was a difference of about 2.5 degrees Celsius between top and bottom. The filled portions of the top-row cylinders were slightly warmer than those in the bottom row. These differences may be due to the shading and thermal insulation given to the bottom-row cylinders by those in the top row. The other image in Figure 40 was taken from an elevated position. Several anomalous cylinders were identified in the stacks: one cylinder in the bottom row was seen to be cool while another had a lower than normal fill level. Other cylinders were filled to levels slightly less than normal, especially the isolated set of cylinders in the foreground.

7.3.3.2 K Yard

A. Cylinder 7340

Images of the fill line along the side of Cylinder 7340 were obtained from an elevated position. This cylinder was rotated so that the fill lines could be viewed from above. The fill lines are clearly evident in the images in Figure 41 (see page 67). Only a very weak indication of the fill line was obtained from imaging the end of the cylinder. This was one of only a few cases where an image at the end of a skirted cylinder did not readily identify the fill line.

CYLINDER 5277 (48T)

CYLINDER 152675 (48H)
CYLINDER 119486 (48OM)

FIGURE 39. E YARD TEST CYLINDER VIEWED FROM
THE LIFT

B. K Yard Stacks

Images were made of the K Yard from an elevated position at twilight. Some "cold" spots were visually associated with paint on cylinders. A few cases of lower-than-normal fill levels were identified. Other cylinders were seen to be abnormally warm. Representative examples are shown in Figure 42 (see page 68).

FIGURE 40. THERMAL IR IMAGE OF E YARD STACKS

BACK HALF

FRONT HALF

FIGURE 41. *FILL LINE OF CYLINDER 7340. Rotated view is from lift above the cylinder.*

7.3.3.3 J Yard

A. Cylinder 7767 (1500-Pound Heel)

A "heel" is fill material that remains stuck to the wall of a cylinder after it is emptied. Cylinder 7767, having a heel of approximately 1500 pounds, was imaged to determine if heels can be detected thermally. The image is presented in Figure 43 (see page 69). There was a detectable amount of material in the cylinder; however, the heel boundary was not very distinct. The lack of a clear signal could have been caused by a relatively thin and variable layer of fill material on the inside of the cylinder. Additionally, the outside surface of this cylinder appeared to be highly variable,

FIGURE 42. K YARD STACKS

which partially obscured the signature of the heel. There was an indication that a relatively large accumulation of material was located on the valve end of the cylinder on the side where the image was taken. However, the actual distribution of the heel material is unknown. These images illustrate the potential for thermally detecting heel distributions.

B. Cylinder 3097

Cylinder 3097 had an internal gas pressure more than an order of magnitude higher than normal. Images of Cylinder 3097 are shown in Figure 44 (see page 70). The cylinder had a very unusual temperature distribution. Viewed from the ends, no fill line was visible, and while there were strong local variations, no temperature trends were apparent. When viewed from the side, a warm band of about 9.5 degrees Celsius could be seen near the middle of the cylinder, and a nearly isothermal region about two degrees cooler was present below it. This cooler region is the result of old paint on the cylinder surface. Above the warm band, the temperature decreases linearly to about 6.3 degrees Celsius, only about one degree Celsius cooler than the bottom part of the cylinder. Cylinder 3097 had a 5/8-inch-thick wall, twice as thick as the other cylinders that were studied. Another cylinder that seemed to be warm on top appears in the background in Figure 43 behind Cylinder 7767. This cylinder, identified as Cylinder 360, also had a 5/8-inch-thick wall. Because Cylinder 3097 was the only cylinder with a 5/8-inch-thick wall selected for detailed study, there was no way to evaluate a hypothesis that the unusual temperature distribution was due to the thickness of the cylinder wall.

FIGURE 43. CYLINDER 7767 WITH 1500-POUND (10 PERCENT) HEEL

7.4 SUMMARY

More than 100 passive thermal IR images were obtained at the K-25 Site to allow an initial evaluation of the utility of the passive thermal IR imaging technology to support the K-25 cylinder yard program. These images were reviewed, and a subset was selected for more detailed study. Specific areas of interest included the (a) detection of cylinder-wall thinning due to weathering processes, (b) detection of localized damage to the cylinder, and (c) collection of information about the cylinder contents. As

FIGURE 44. CYLINDER 3097 WITH HIGH INTERNAL GAS PRESSURE

discussed in the following section, the most promising results were achieved in obtaining information about the cylinder contents.

7.5 CONCLUSIONS

7.5.1 Thinning

Adequate data to evaluate this application were not obtained. This was partly because access was limited, and the field of view of the camera was narrow. However, it is believed that detecting thinned areas will be a difficult task. Controlled experiments and a more thorough understanding of the background are needed.

7.5.2 Localized Damage

Success at detecting localized damage to cylinders was mixed. The local features seen by the IR camera, while plentiful, were dominated by surface characteristics. The detection of dents and pitting may be performed more easily by visual inspection. However, a scrape that was not observed by eye was detected on one cylinder by thermal IR imaging. This preliminary evaluation was not comprehensive, but it did indicate that thermal IR imaging may be a useful component of an inspection procedure.

7.5.3 Fill

Information about fill levels and material distributions within UF_6 storage cylinders can be obtained rapidly and simply using passive thermal IR imaging. With additional investigation, a more precise relationship between fill-line and temperature patterns may be determined. Occasionally there were problems determining the fill lines at the ends of skirted cylinders. Imaging of the ends was important because it is primarily the ends that are visible in the stacks.

There were notable temperature differences between neighboring cylinders in the stacks; as yet, these differences cannot be explained. In the E Yard test cylinders, no correlation was found between the reported assays and temperatures of the cylinders. Further investigation of this relationship is warranted.

In one test cylinder, the presence of a large heel was observed. Smaller heels in the B Yard were not detected. Additional work is required to determine the minimum amount of heel material that is detectable.

The UF_6 storage cylinder with the higher-gas pressure exhibited an unusual temperature distribution. No complete explanation for this anomalous distribution has been developed. It may be characteristic of thicker-walled cylinders (5/8 inch) or due to the paint on the surface.

The extent to which these conclusions can be extrapolated to other measurement conditions (*i.e.*, daytime) should be explored.

7.6 RECOMMENDATIONS

General recommendations for operational implementation and additional measurement and modeling studies are presented in the following sections. Development of detailed plans was beyond the scope of this study.

7.6.1 Operational Use

Semi-quantitative information on fill levels and material distributions can be obtained with good reliability. It would be straightforward to train teams of operators to use modern imaging and data-recording equipment for routine monitoring. It is believed that only a limited effort is needed to optimize system performance.

7.6.2 Measurement Studies

It is necessary to develop a better understanding of the relationship between the material distribution within the cylinders and the resulting surface temperature patterns. It is also necessary to expand the range of environmental conditions under which the techniques can be employed or, at least, to ascertain the limits of applicability. It would be desirable to determine if the parameters of the measurements can be manipulated in simple ways to achieve better results. A wider variety of cylinder types and surface conditions needs to be explored. Differences in the thermal patterns of cylinders in the stacks need to be better understood. Finally, two types of temporal changes are of interest: the cooling of freshly filled cylinders over time and the redistribution of fill material after a cylinder has been rolled.

To address these issues, three types of studies are recommended.

- A. A well-planned series of measurements on instrumented and monitored surrogate cylinders should be performed. One end of the cylinders would be open for access and a surrogate material, such as sand or concrete mix, would be inserted for fill. One or two cylinders with varying properties of the surrogate fill material, the cylinder, and the environment should be established at RSL for study. A similar surrogate cylinder and an actual UF₆ storage cylinder should be established as controls at K-25. This would allow the cylinder conditions to be modified in a known way while monitoring the response. It would also support the modeling studies recommended in Section 7.6.3.
- B. A series of carefully planned *in situ* measurements is recommended. These measurements would address variations in cylinder type, cylinder condition, and cylinder position in the stacks. The measurements are important for developing and refining operational procedures and for providing training to facilitate technology transfer. They also would provide a link from the measurements on the controlled surrogate cylinders to the real cylinders.
- C. It is recommended that measurements be taken over time of a freshly filled cylinder and one or more recently rolled cylinders. Measurements should be made periodically until change becomes negligible. For best efficiency, these cylinders should be located at the same site as the other test cylinders.

7.6.3 Modeling Studies

It is recommended that a comprehensive, three-dimensional, heat-transfer model of filled cylinders be developed. Theoretical simulations can help elucidate the roles of specific parameters in complex systems and allow extrapolations to conditions not yet encountered. They can be used to investigate variable scenarios and to optimize the implementation of technology.

Such a modeling effort will require quantitative information about the cylinders and their contents that is not readily available, such as the heat conductivity of the fill material and the surface emissivity of

the cylinders. However, initial investigations can be performed using approximations until more accurate information can be obtained. Parallel efforts can be made to develop the necessary knowledge base. Heat-transfer modeling capabilities existing at the national laboratories should be included in this effort.

7.6.4 System Specifications

It is recommended that efforts should be directed toward determining the necessary and optimal requirements for a thermal IR imaging system including camera specifications and data-recording and analysis requirements. Thermal IR-imaging technology has been available for many years, and the basic principles are well established. However, recent developments in detector and computer technology have greatly expanded the range of technical options. Complete systems are now available with varying wavelength sensitivities, detector types, data-recording schemes, and data-analysis systems. Although the range of options continues to expand, it is probably unnecessary that cutting-edge technology be implemented for this application.

7.6.5 Miscellaneous Topics

- A. Determine the quantitative relationship between ^{235}U assay, heat output, and the age of the fill material.
- B. Characterize the spectral emissivity of the cylinder surface in the relevant spectral regions. Changes in emissivity due to handling damage and weathering should be better understood.
- C. Undertake controlled laboratory and field measurements to determine the size of a heel that can be detected. This investigation could be extended to the detection of material on the interior walls of pipes.
- D. Conduct further studies to better understand the distribution and physical characteristics of the fill material in the cylinders, including probable changes occurring over time.

Appendix A

Typical Applications of Neutron Activation Analysis

1. Instrumental Neutron Activation Analysis (INAA)

This application involves only neutron activation of a given sample after some decay period by counting with a gamma-ray spectrometer. Because it does not involve post-irradiation radiochemical separations, a given induced activity must be measured in the presence of all other activities also induced in the sample matrix. Thus in general, any photopeak (total absorption peak) of interest must be measured on top of a cumulative Compton continuum produced by gamma rays of energies greater than the energy being measured.

2. Cold Neutron Activation

High-purity, low-background, gamma-ray-free, thermal neutron beams with a flux of approximately 10^7 n/cm²/sec are used for prompt-gamma NAA at Cornell University and the NIST laboratory in Gaithersburg, Maryland.

3. Determination of Trace-Element Impurities in High-Temperature Superconductor Starting Materials

The starting materials for high-temperature superconductor materials like the yttrium-barium-copper-oxygen systems are neutron activated, and trace elements are looked for at the level of less than 0.2 ng/g for iridium; 1.0 ng/g for cesium, etc.

4. Elemental Characterization of Size-Fractionated Ash from Municipal Solid Waste Incinerator

The presence of heavy metal in incinerator ash and its leachability into ground water is monitored by using the neutron activation method together with an anti-coincidence Compton-suppression detector to increase the sensitivity of elements that emit gamma rays between the energies of 200 and 800 keV.

5. Airborne Particles from Recent-Model Automobiles

Newer models of automobiles emit light rare earth elements (REEs) that are present in the "three way" catalyst used to remove nitrous oxide, carbon monoxide, and hydrocarbons from the exhaust of gasoline-powered vehicles. The samples are collected in micro-orifice impactors and activated with thermal neutrons for resultant gamma-ray analysis.

6. Determination of Trace Elements in Environmental Samples by NAA.

7. Investigation on Bromine-Binding Proteins in Bovine Kidneys by Biochemical Techniques in Conjunction with Neutron Activation.

8. Use of Neutron Activation Tracers for Studying the Chromatographic Separation of Strontium and Yttrium.

9. High-Level Neutron Coincidence Measurements in Conjunction with High-Resolution Gamma Spectroscopy

Nondestructive assay of plutonium in various forms, including nitrates, metal buttons, plutonium-oxide and mixed-oxide powder, fuel pellets, and pins can be performed for plutonium scrap and waste using thermal neutrons.

Appendix B

Thermal Neutron-Capture Gamma-Ray Data

Energy (keV)	Elements	Intensity (γ /capture)	Thermal Cross Section (barns)	Percent Nuclides (% abundance)
212.0	Mn	10.6	13.3	100.0
229.8	Co	21.0	37.2	100.0
277.1	Co	12.0	37.2	100.0
278.2	Cu	19.0	4.5	69.1
352.3	Fe	11.2	2.55	100.0
465.1	Ni	13.0	4.43	100.0
585.2	Mg	25.6	6.3e-2	100.0
749.2	Cr	11.0	3.1	100.0
770.3	K	55.2	1.96	93.3
778.3	Mo	62.0	14.5	15.7
835.1	Cr	26.9	3.1	100.0
847.7	Mo	10.6	14.5	15.7
850.0	Mo	13.3	14.5	15.7
1077.4	Zn	18.9	1.1	100.0
1171.3	Sn	7.9	0.63	100.0
1261.7	C	29.9	3.4e-3	96.9
1293.3	Sn	12.8	0.63	100.0
1633.1	F	100.0	9.5e-3	100.0
1778.8	Al	100.0	0.23	100.0
1808.9	Mg	30.0	6.3e-2	100.0
2073.7	K	11.5	1.96	93.3
2223.2	H	100.0	5.3e-4	100.0
2238.9	Cr	8.3	3.1	100.0
2828.1	Mg	42.5	6.3e-2	100.0
3683.9	C	32.5	3.4e-3	98.9
4945.3	C	68.4	3.4e-3	98.9
5975.1	Co	7.3	37.2	100.0
6705.8	Co	8.5	37.2	100.0
6837.0	Ni	10.8	4.43	100.0
6876.8	Co	8.5	37.2	100.0
7631.1	Fe	27.7	2.55	100.0
7637.0	Cu	15.0	4.5	69.1
7645.5	Fe	24.4	2.55	100.0

Energy (keV)	Elements	Intensity (γ/capture)	Thermal Cross Section (barns)	Percent Nuclides (% abundance)
7723.9	Al	25.9	0.23	100.0
7863.3	Zen	10.6	1.1	100.0
7916.0	Cu	27.0	4.5	69.1
8884.1	Kr	27.0	3.1	100.0
8998.8	In	37.7	4.43	100.0

Appendix C

Neutron Generation

Neutrons are generated by accelerating protons and deuterons. Rubberized beryllium targets are used to produce neutron outputs ranging from 10^8 to more than 10^{13} neutrons per second. Beryllium(p,n) and beryllium(d,n) reactions produce neutrons with low-average energies, which simplifies moderator design and reduces shielding requirements for thermal neutron applications. Pulsed-beam systems are ideal for applications requiring high-peak neutron-flux variable pulse widths, and repetition rates make them ideal for activation analysis using short-lived reaction products. The specifications for five representative neutron generators based on the RF (radio frequency) LINAC technology are given in Table C-1.

Table C-1. Typical Neutron Generator LINAC Specifications¹

Model	DL-1	DL-1.5	PL-4	PL-6	PL-11
Accelerated particle	d ⁺	d ⁺	p ⁺	p ⁺	p ⁺
Beam energy (MeV)	0.9	1.5	3.9	6.5	11
Neutron yield (n/sec/μA)	7.8×10^7	3.9×10^8	1.3×10^9	7.5×10^9	3.0×10^{10}
Beam current/pulse (mA)	10	15	40	25	40
Beam pulse width (μsec)	30-120	30-180	25-215	35-120	35-215
Pulse repetition rate (Hz)	1-120	1-120	1-120	1-120	1-120
Maximum target current (μA)	140	325	1000	350	1000
Maximum target yield (n/sec/4π)	1×10^{10}	1×10^{11}	1.3×10^{12}	3×10^{12}	3×10^{13}
Accelerator length (ft)	4	5.25	12.5	11.25	16
Maximum thermal neutron flux (n/cm ² /sec)	5.0×10^7	2.6×10^8	1.9×10^9	1.5×10^{10}	2.0×10^{11}
Accelerator weight (lb)	400	500	5000	3500	5500
Electrical requirement (kVA)	12	15	80	60	130

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Report Number (14) DOE/NV/11718--049

Publ. Date (11) 199702

Sponsor Code (18) DOE/DP, XF

UC Category (19) UC-702), DOE/ER

DOE