

The X-ray, K-edge Heavy Metal Detector

Final Report for Project CH-15C251

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ABSTRACT

Better quantification of heavy metal contamination in weapons lab facilities can yield savings in cleanup costs and benefits of reduced risk during dismantlement operations. The X-ray, K-edge heavy metal detector was developed under the Department of Energy Characterization Monitoring and Sensor Technology Crosscutting Program to address these needs. The K-edge inspection technology is based on the measurement of the energy spectrum of a broadband X-ray beam transmitted through a sample. The X-ray source is an industrial X-ray tube, and the detector is a high purity Ge crystal. Both source and detector are collimated to define a narrow (<1mm dia.) beam used to probe the sample. Unlike other assay techniques, the K-edge method is not sensitive to sample geometry.

The K-edge inspection system was successfully field demonstrated at several sites. During a Large Scale Demonstration at the Savannah River Site a series of exhaust ducts containing uranium deposits were inspected, and the results were compared with conventional passive gamma NDA measurements. Good agreement between the two techniques was observed, with the K-edge method providing greater precision and much better spatial mapping of the deposits. Other demonstrations of the technology have included detection of mercury, thorium and uranium in drain pipes, and measurement of uranium deposits ranging from 10 mg/cm² to 6000 mg/cm² in 1/8 inch wall monel pipes. In another application, measurement of uranium concentration in spent nuclear fuel plate assemblies (up to 3000 mg/cm² for 18 fuel plates) was demonstrated with better than 5% precision within five minutes measurement time.

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1.0 INTRODUCTION

The Department of Energy has thousands of buildings across the weapons complex which are being Deactivated and Decommissioned (D&D)[1]. Heavy metal contaminants, including mercury, lead, and radioactive metals such as thorium, uranium, and plutonium, are present at many of these facilities. Significant deposits often remain in processing equipment after it has been shut down, and surrounding building materials have also often become contaminated. Efficient, safe, cost-effective methods of characterizing these materials are needed to assist in the timely cleanup of these sites. Due to the hazardous nature of the contaminants, a nondestructive non-invasive technique is preferred for characterization and for monitoring decontamination processes.

Current nondestructive inspections often do not provide the desired accuracy and sensitivity. Passive gamma and neutron assay measurements are generally used for radioactive materials, but uncertainties can be as large as 100% for complex material geometry. X-ray K-edge densitometry is a nondestructive inspection technique that can provide accurate, sensitive quantification of heavy metal contamination that is generally not dependent on assumptions made about container geometry or material.

The method of X-ray absorption edge spectrometry was first developed by Glocker and Frohnmayer[2] in 1925. Early applications of this technique used modified X-ray diffraction spectrometers based on analyzing crystals. As energy-sensitive germanium detectors and multi-channel analyzers came into widespread use, dedicated X-ray densitometry units were developed. These were used extensively for monitoring nuclear fuel processing operations, and were optimized to measure several tens of grams per liter of uranium and/or plutonium in solution[3,4].

In the course of this project we have built on this early research work to develop a robust K-edge technology for application in the field. In this report we summarize the theory behind the K-edge technique and describe the instrumentation that we have developed. We follow that with brief descriptions of several applications of the technology and projections for future improvements. A brief history of the project is provided in appendices to this report. A record of the project funding is given in Appendix A, the project milestones are described in Appendix B, and a listing of project-related reports and publications can be found in Appendix C.

2.0 THEORY

The absorption of X-rays in materials is governed by the binding energies of atomic electrons. Each element has a unique distribution of electrons, with the K-shell having the highest binding energy. If an X-ray has just enough energy to liberate a K-shell electron, there will be an increased likelihood that the X-ray will be absorbed. The rate of absorption can be described by an attenuation coefficient that depends on the X-ray energy (see Fig. 1). If an X-ray source having a broad spectrum (such as an industrial X-ray tube) is directed through a sample to an energy-sensitive detector on the opposite side, one will observe an abrupt drop in the transmitted intensity at energies corresponding to the K-shell binding energies of elements in the sample. Figure 1 shows a spectrum for a calibration sample consisting of foils of thorium and uranium. The thorium K-edge appears at 109.7 keV, while the uranium K-edge is observed at 115.6 keV.

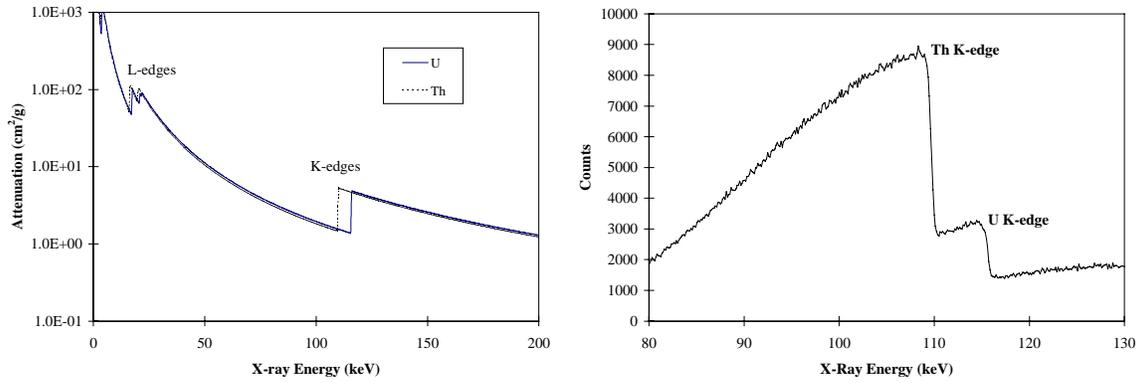


Figure 1. X-ray attenuation coefficients for thorium and uranium (left), and example spectrum from K-edge measurement of thorium and uranium calibration foils located behind 1/8 inch of steel (right). (Note that the energy scale is different for the two plots.)

The energy at which the abrupt change in transmitted intensity occurs identifies the type of contaminant. The amount of contaminant present can be calculated based on the magnitude of the intensity change. The thickness, x , will be given by

$$x = \frac{\ln \left[\frac{I(E_+)}{I(E_-)} \right]}{\mu(E_-) - \mu(E_+)}, \quad (1)$$

where $I(E_-)$, and $I(E_+)$, are the transmitted X-ray intensities at energies very close to the edge from the left, and the right, respectively (see Fig. 2), and $\mu(E_-)$ and $\mu(E_+)$ are the attenuation coefficients of the contaminant material at these energies. The precision of the K-edge technique is largely insensitive to the thickness or geometry of the container. The measurement uncertainty will be determined primarily by the statistics of the intensity measurements. Typically, a precision of 10% or better can be achieved within a few minutes measurement time.

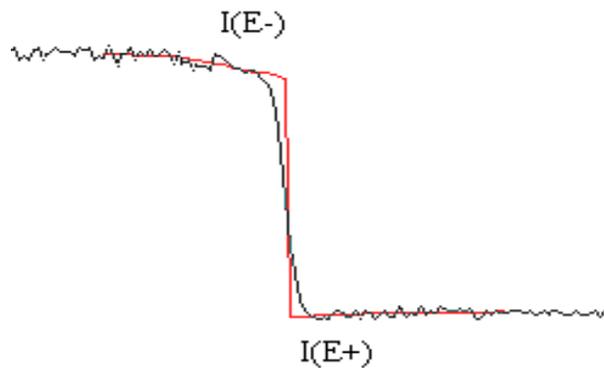


Figure 2. Blowup around the region of a K-edge drop in an X-ray spectrum, showing a fitted curve (smooth line) superimposed on the data points.

Due to the energy resolution of X-ray detectors (typically ~ 0.5 - 1.0 keV FWHM for high purity germanium), the spectrum will not exhibit a perfectly sharp transition at the K-edge. Thus, to determine $I(E_-)$ and $I(E_+)$, it will be necessary to fit a curve to the spectrum to extrapolate the measurements to the K-edge. A second order polynomial generally fits quite well

to a restricted interval on either side of the K-edge. In selecting the intervals to be used in the fits, it is important to avoid regions too close to the K-edge, where the detector resolution affects the slope of the curve, as well as regions that correspond to the K-edge energies of other elements that might be present in the sample.

It was found that for small amounts of a contaminant the above method did occasionally yield spurious results. A more robust fitting procedure requires calculating the transmission,

$$T(E) = I(E)/I_0(E), \quad (2)$$

where $I_0(E)$ is the incident intensity measured with no sample present. As pointed out by Ottmar and Eberle[4], a plot of $\log(\log(1/T))$ vs. $\log(E)$ can be treated as a linear function over the energy range around a K-edge. In this method, one must first make a high statistics measurement of the incident spectrum using the same X-ray tube voltage setting as used during inspection of the sample. In practice, $I_0(E)$ does not have to be measured without material in the beam. It is in fact better to measure a spectrum corresponding to a nominal matrix material. For example, if one were looking for heavy metal contamination in pipes, the I_0 spectrum would best be represented by a measurement through a clean section of similar pipe. Figure 3 shows an example of an incident spectrum, the spectrum through a sample containing uranium, and the resulting transmission function used in the fitting procedure. Note the linear behavior of the data on either side of the K-edge in Fig. 3c.

To attain good accuracy, one must assure that background radiation does not contribute significantly to the spectrum. This can usually be achieved by setting the X-ray tube voltage as low as possible while still maintaining an adequate transmission rate at the K-edge energy. Also, scattered X-rays are minimized by collimating the X-ray source and detector to define a narrow beam (~1 mm diameter) that penetrates the sample. Thus, in order to measure the total contamination in a large object, one must make measurements at a number of points and interpolate between them.

If there is any background radiation present at the K-edge energy, this must be accounted for before applying Eq. 1. Figure 4 shows the spectrum observed for a thorium foil of thickness 1500 mg/cm² behind ¼ inch stainless steel. The X-ray flux observed at low energies is dominated by background processes, primarily due to Compton scattering in the detector itself. This background can extend into the K-edge region, producing an erroneous result if the standard analysis procedure is applied (background present at the K-edge energy will generally bias the measurement to lower values). To account for this background, an iterative variation of Eq. 1 is applied. First, a measure of the initial spectrum $I_0(E)$ with no sample present is obtained. The spectrum observed for the sample (Fig. 4) can be expressed as

$$I(E) = I_0(E)\exp\{-\mu_c(E)t_c\}\exp\{-\mu_m(E)t_m\} + I_B(E), \quad (3)$$

where μ_c and t_c are the attenuation coefficient and thickness of the contaminant, and μ_m and t_m are the attenuation coefficient and thickness of the matrix material. $I_B(E)$ is the background energy spectrum. Attenuation coefficients for different materials are tabulated as a function of X-ray energy. One must make an assumption about the composition of the matrix material to determine values for μ_m to be used in Eq. 3. As long as the matrix material is relatively light compared to the contaminant (aluminum or iron matrix, for example), μ_m will change relatively slowly for

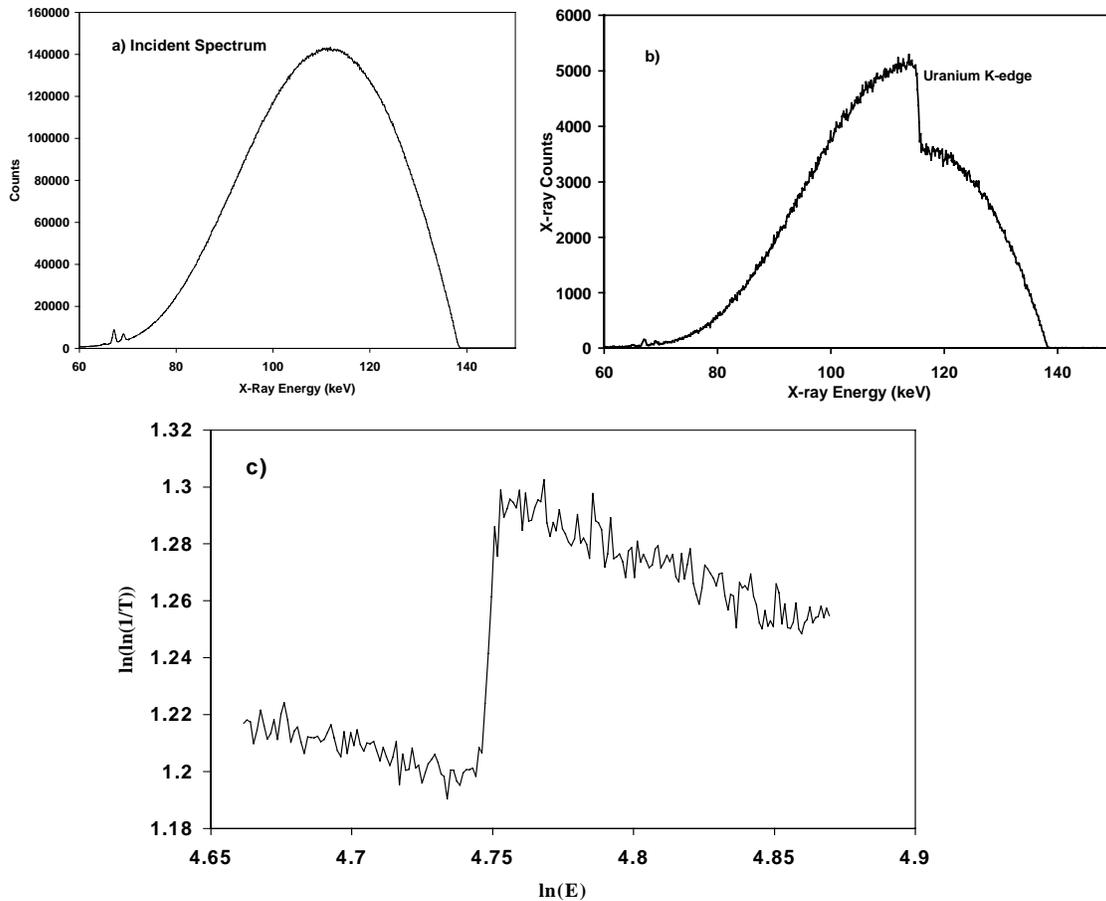


Figure 3. Example of transmission measurement: a) incident spectrum measured through 0.125 inch steel, b) spectrum for uranium inside a steel pipe, and c) the transmission function calculated from these two spectra.

energies greater than the contaminant K-edge energy, and bias introduced by the wrong choice of matrix material will be small. With these assumptions the iterative calculation proceeds as follows: An initial estimate of the contaminant thickness is obtained from Eq. 1 assuming no background. Using this value for t_c and setting $I_B=0$, a fit of Eq. 3 is made to the data near the high-energy endpoint of the spectrum (little background is expected in this region) to determine the matrix thickness, t_m . Eq. 3 can then be used to predict the signal at the K-edge energy, with observed excess counts being attributed to background. This background value is then subtracted from the signal at the K-edge energy and the procedure is iterated until it converges. In practice it has been found that this procedure generally converges after a few iterations, and repeat measurements have shown that the method is very robust and precise (see next section). Most processing equipment encountered in D&D operations was constructed from alloys of steel, aluminum, copper, or nickel, all of which satisfy the assumptions stated above with regard to matrix materials.

Finally, it should be noted that the X-ray K-edge technique cannot distinguish between different isotopes of an element. K-edge measurements yield total elemental content in a sample. External information must be applied to convert these results to specific isotope concentrations.

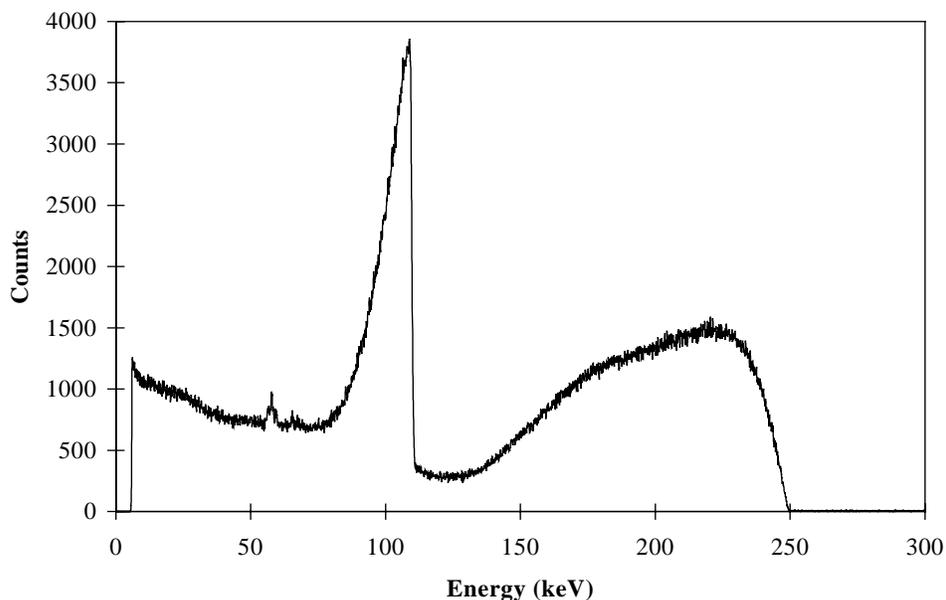


Figure 4. Transmission spectrum for 1500 mg/cm² thorium behind ¼ inch steel.

3.0 K-EDGE PROTOTYPE SYSTEM

A schematic diagram of the prototype X-ray K-edge system is shown in Fig. 5. It consists of three major components: an inspection head, a high voltage and cooling unit for the X-ray tube, and a control rack. The inspection head can be configured for different applications. Two different X-ray tubes are available: a relatively compact 160 kVp, 3.0 kW tube, and a more powerful 320 kVp, 4.5 kW unit. Figure 6 shows a photograph of the system configured with the 160 kVp tube mounted on an adjustable C-frame at the end of a mechanical boom. The inspection head will be positioned around the object to be inspected (the opening of the C-frame can be adjusted to accommodate different size objects). On the left end of the frame (as seen in Fig. 6) is the X-ray tube, and on the opposite side are two X-ray detectors. One detector is a high-purity germanium (HPGe) detector used to measure the X-ray energy spectrum for quantitative analysis as described in the previous section. The other detector is a real-time X-ray imaging system that provides a quick qualitative indication of heavy metal location, and can also be used to guide the interpolation between the spectroscopic measurements. Either detector can be centered on the X-ray beam by remote control. The total weight of the inspection head is 60 kg, and it can be wrapped in plastic to protect against contamination. Cables 20 m long connect the inspection head to the control rack and high voltage supply. The operator can sit at the control rack, a safe distance from the X-ray source, while making the measurements.

Data acquisition is handled by a PC running a Windows[®]-based interface program to display the real-time X-ray images and the X-ray transmission spectra. A view of the user interface is shown in Fig. 7. In the normal mode of operation, the user would first view the real-time image (see examples in Sec. 4.5) for potential deposits. At a location of interest the operator would then switch to a spectroscopic measurement. Different contaminant elements of interest could be selected and the concentration of each one monitored as data accumulates. Data can be collected for a preset time, or until a desired level of precision is reached. Images and spectra can be stored for reference and further analysis.

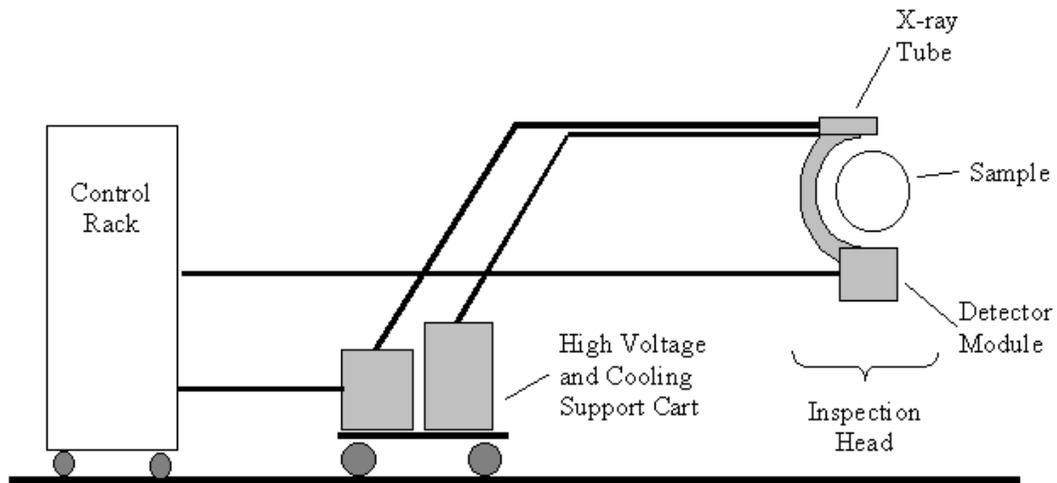


Figure 5. Schematic Diagram of the X-Ray, K-Edge Heavy Metal Detection System



Figure 6. The prototype X-ray K-edge system. The control rack is to the left, and the high-voltage supply/cooling unit is to the lower right. The C-frame inspection head is at the end of an adjustable boom.

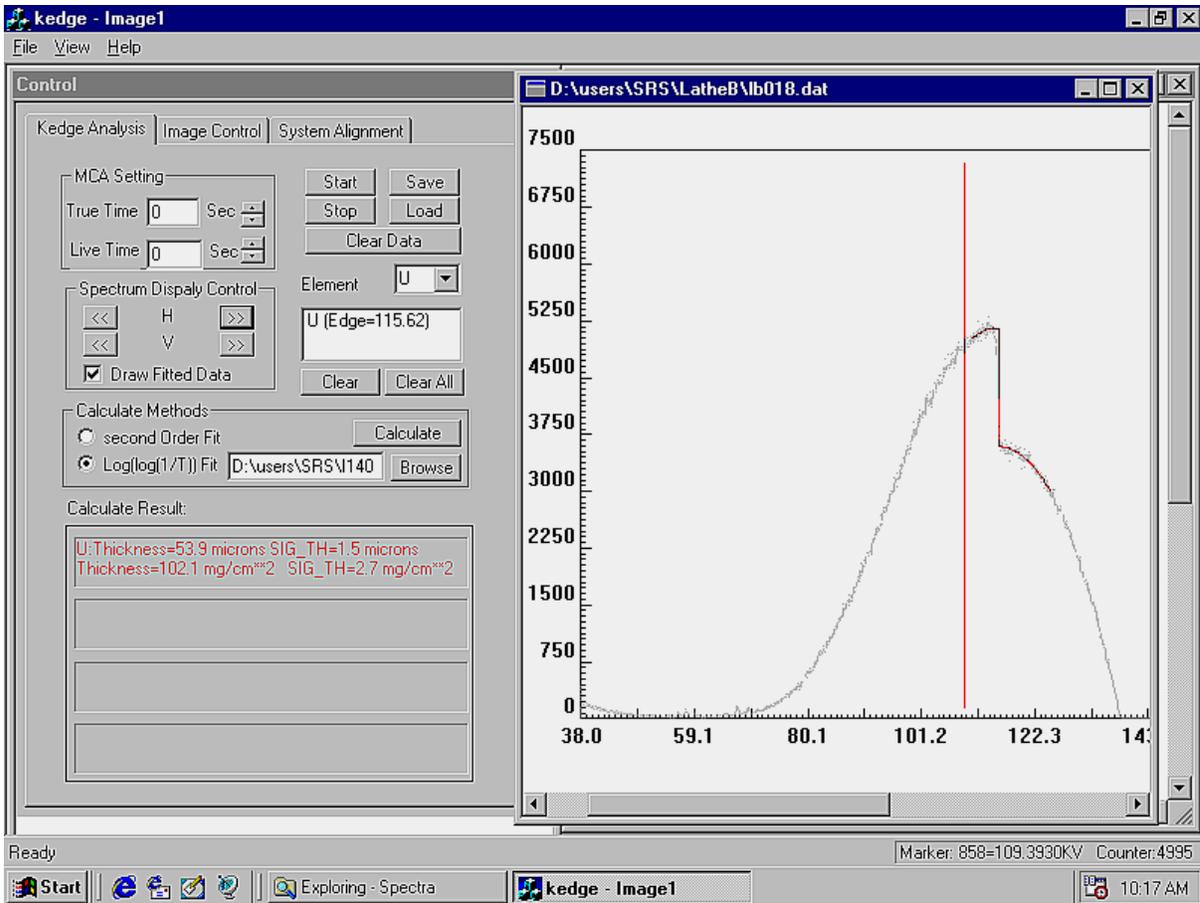


Figure 7. User interface for K-edge data acquisition and analysis program.

Operation of the K-edge detector has been validated using a variety of calibration samples. For small amounts of a heavy metal, Inductively Coupled Plasma (ICP) standard solutions in polyethylene bottles of dimensions known within 3% were used. For larger amounts of materials, foils of different elements having thickness measured with 5% accuracy were used. An example of the agreement between known and K-edge measured thickness is shown in Fig. 8 for uranium samples behind a steel plate.

Each data point represents the average of 5-10 measurements of duration a few minutes each. The error bars indicate the r.m.s. deviation of these measurements. For the thin samples, greater precision could be achieved with longer data acquisition. Note that the axes are logarithmic, and good agreement is observed over three decades. In most cases the precision of the K-edge measurement was as good, or better, than that of the method used to establish the reference sample thickness. Similar validation measurements have been obtained for gold, lead, and thorium samples for different container materials and thicknesses.

If one is looking for a specific contaminant, it is desirable to measure the transmission spectrum only around the K-edge of that element. There is a limit to the rate at which an X-ray detector can analyze incoming signals. X-rays at energies far from the K-edge contribute no information but limit the throughput. A crystal monochromator can be used to select a narrow band of energies around the K-edge. This method was tested in the early stages of the project[5], but, as the monochromator adds bulk and weight to the system, and is somewhat difficult to

align, it was not used very extensively. In many cases, a narrowing of the transmission energy band can be achieved by appropriate selection of the X-ray tube voltage and filtering materials.

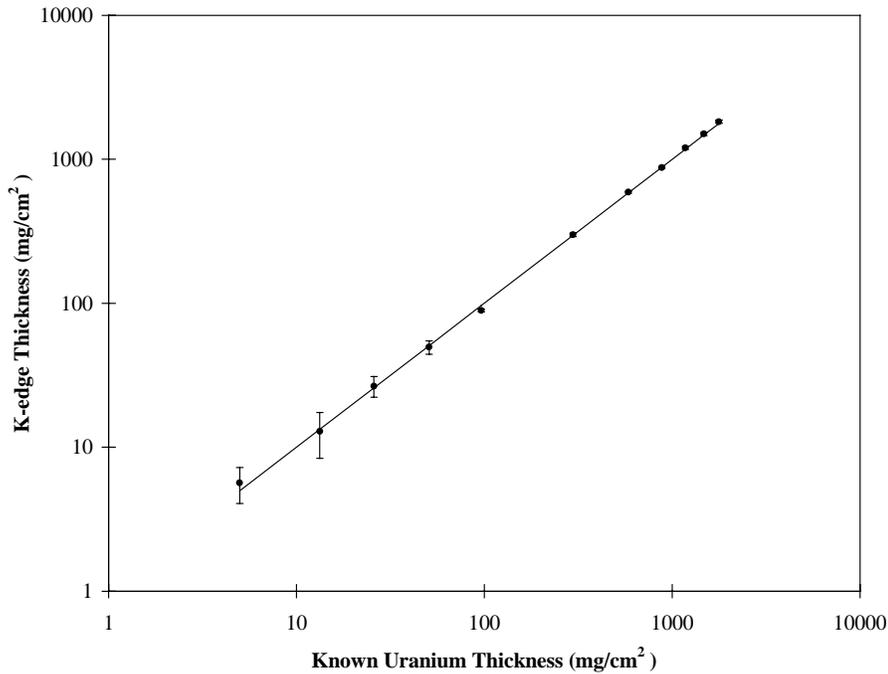


Figure 8. Comparison between K-edge measured and known values for different thicknesses of uranium calibration samples located behind ¼ inch steel. The solid line indicates a one-to-one relationship.

4.0 APPLICATIONS

As the K-edge development project unfolded, a number of opportunities arose to demonstrate the technology. Each one placed different requirements on the equipment or data analysis. Due to the flexibility of the system, it was possible to quickly adapt the K-edge technology to a variety of situations. The major demonstrations carried out during this project are summarized below.

4.1 Uranium Holdup at the Oak Ridge Gaseous Diffusion Plant

The first field demonstration of the prototype K-edge system was carried out at the Oak Ridge K-25 Site in February 1997[6]. Five samples of one inch inside diameter and 1/8th inch wall monel pipes that had been cut out of a UF₆ feed facility, were inspected. Contamination levels in the pipes ranged from barely detectable to nearly full with deposits that appeared to be oxide and fluoride compounds of uranium. A picture of one of the pipes is shown in Fig. 9. During inspection the pipes were contained in sealed polyester bags to avoid contamination of the equipment.

The pipes inspected ranged in length from 2-5 feet. Real-time images and K-edge measurements were acquired at intervals of 4-8 inches for two perpendicular orientations of each pipe. Most real-time images indicated a fairly uniform coating around the circumference of the pipes. Occasionally a small region was noted where the uranium deposit had flaked off. In most

cases accurate measurements were obtained within a few minutes. An example spectrum is shown to the right in Fig. 9.

In Fig. 10 we plot the K-edge measurements as a function of position along one of the pipes. In general we see that the deposit thickness is changing gradually along the length of the pipe and the data can be fit to a straight line. These fits were used in conjunction with the known diameter of the pipes to calculate the total amount of uranium in each pipe. These results are presented in the second column of Table 1. These same samples were also inspected by personnel at Oak Ridge using a baseline passive gamma NDA technology estimated to have a $\pm 30\%$ accuracy[7]. These results are shown in the third column of Table 1, and are in good agreement with the K-edge measurements.

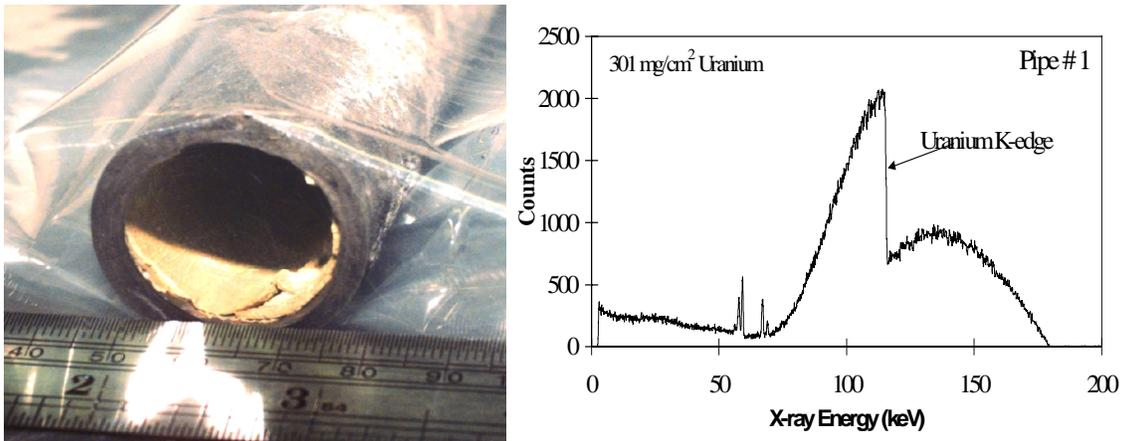


Figure 9. Photograph of one of the pipes from the K-25 Site UF_6 feed facility, and an example K-edge spectrum acquired at one point along the pipe.

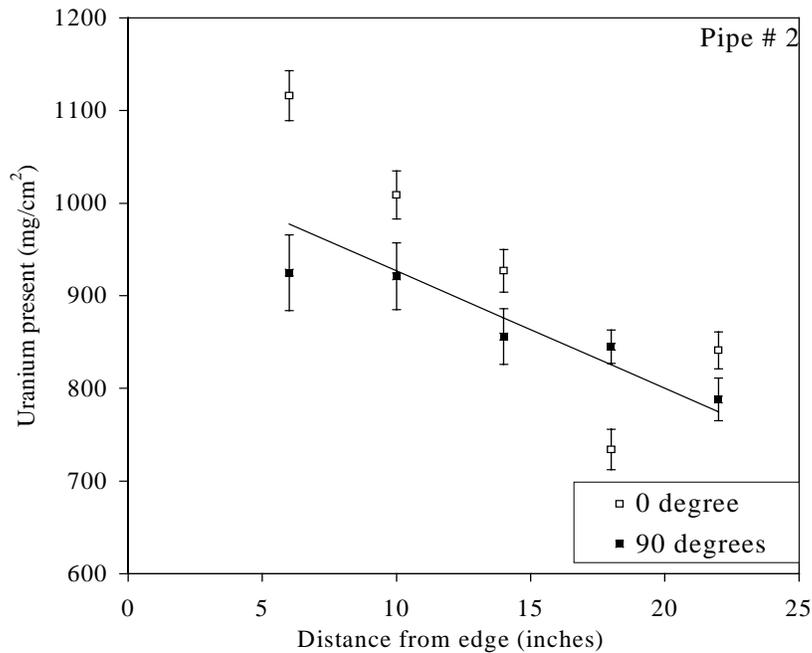


Figure 10. Measured Uranium concentration as a function of position along the pipe.

Table 1. Amount of uranium contamination in each of the samples as measured using the K-edge technique and a passive gamma NDA method.

	K-Edge inspection (grams)	NDA technique (grams)
Pipe # 1	72 ± 7	78 ± 23
Pipe # 2	259 ± 15	342 ± 103
Pipe # 3	806 ± 104	1000 ± 300
Pipe # 5	5.8 ± 3	2.54 ± 0.76
Pipe # 6	6.2 ± 3	5.9 ± 1.8

4.2 Uranium Concentration in Reactor Fuel Plates

There are instances where records for spent nuclear fuel are incomplete, as well as cases where fuel assemblies have deteriorated during storage. To bring these materials into compliance for long term storage will require determination of parameters such as enrichment, total fissionable material, and burnup. Accurate estimates of these parameters can be obtained only through the combination of information from different inspection techniques.

To demonstrate the viability of using the K-edge technique in characterizing spent nuclear fuel, the prototype K-edge system was set up in the Nuclear Engineering Laboratory on the campus of Iowa State University, where a series of measurements were made on fuel plates from the UTR-10 University Teaching and Research reactor[8]. This reactor can be configured to operate with either high-enrichment uranium (HEU) or low-enrichment uranium (LEU) fuel. Most of the plates used in these studies had not been irradiated, which simplified measurement procedures. To demonstrate that background radiation from spent fuel would not bias the K-edge measurements, several tests were made using irradiated HEU fuel plates.

In performing the K-edge measurements we placed varying numbers of fuel plates in the X-ray beam and then placed differing amounts of material around the plates (1 inch aluminum, ½ inch aluminum plus 4 inches water, ¼ inch steel, or ½ inch steel). For each arrangement we made five repeat measurements, collecting data for 2-3 minutes each time. Examples of these measurements are plotted in Fig. 11 as a function of the number of plates being inspected. The K-edge measurements show good agreement with predictions for uranium concentrations ranging from 60 mg/cm² to 3000 mg/cm² under a variety of matrix conditions.

K-edge measurements were also made on a single HEU plate and on a stack of six HEU plates that had been irradiated. As seen in Fig. 11, the results are in good agreement with the measurements on the unirradiated plates. The radiation field in the vicinity of the detector (~10 cm from the plates) was 20 mR/hr for the single plate and 200 mR/hr for the six-plate stack (1R/hr on contact). It was possible to easily shield the detector so that this background radiation did not affect the K-edge measurements, and there is room for improvement to enable operation in even higher radiation fields.

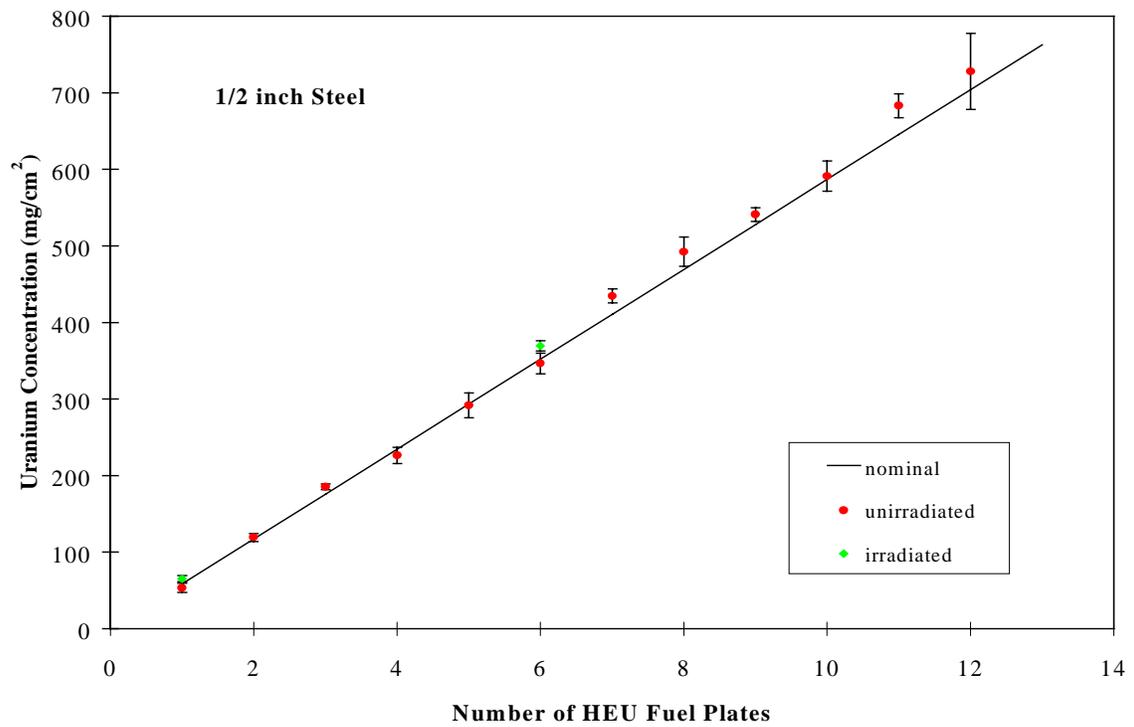
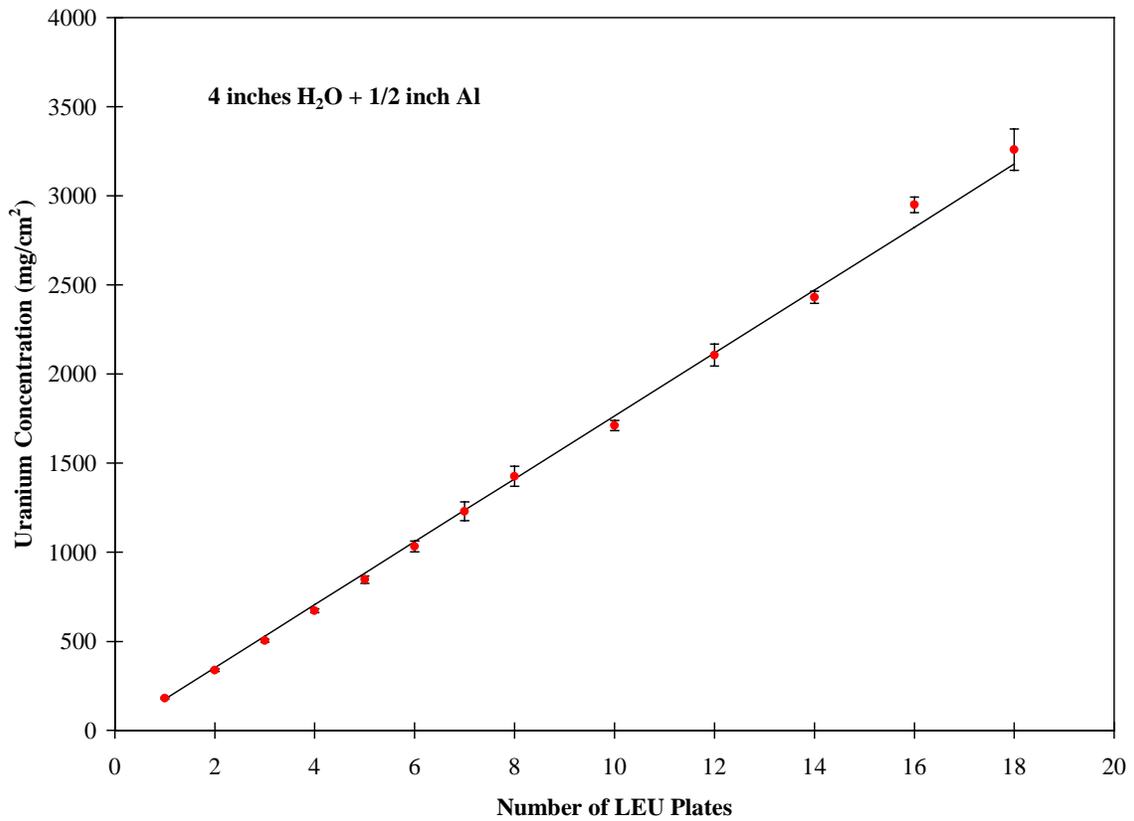


Figure 11. K-edge measured uranium concentration as a function of the number of fuel plates in a stack in the presence of additional aluminum and water (top) and additional steel (bottom).

4.3 RCRA Heavy Metal Concentration in Waste Drums

There are currently in storage substantial volumes of mixed waste resulting from over 40 years of nuclear weapons production at Department of Energy sites. For example, it is estimated that there are more than 135,000 55-gallon drums of transuranic (TRU) waste at INEEL[9]. Much of this material is in the form of sludge containing RCRA-identified heavy metals such as cadmium, mercury and lead. Plans are being developed for disposal of this waste in a permanent repository such as the Waste Isolation Pilot Plant (WIPP). Prior to disposal, it must be verified that these wastes meet the Waste Acceptance Criteria for the storage site.

Three identical sets of 14 simulated waste drums were prepared for Lockheed Martin Idaho Technologies Company by Rust Geotech. The simulated waste consisted of a mixture of water, sand, sodium nitrate, and Portland cement spiked with various amounts of sulfides of the RCRA metals cadmium, mercury and lead. These drums were used to evaluate different prompt gamma neutron activation analysis systems in a series of blind tests[9]. One set of these surrogate waste drums was shipped to Ames Laboratory for the purpose of determining the feasibility of using an X-ray K-edge technique to measure RCRA heavy metal contamination.

X-ray K-edge measurements were made on 13 of the 14 simulated waste drums (drum #14 contained only Cd which could not be observed with the K-edge technique under these circumstances)[10]. The setup for these measurements is shown in Fig. 12. In this case, the 320 kVp X-ray tube was used, and the waste drums were positioned relative to a stationary inspection head. An example spectrum is shown in Fig. 13. All data were acquired without specific knowledge of the drum contents. The known concentration from one drum (#12) was used as a reference calibration to normalize the rest of the measurements. The statistical precision of the K-edge signal was monitored to determine the data acquisition time. The goal was to obtain a precision of at least 10%, which for most drums could be reached in 5-10 minutes. Where no significant signal was observed, a three sigma upper limit was estimated. The results are summarized in Table 2 for the 13 drums.

Multiple measurements were made on most drums, with the results being consistent. The scan time for each analysis is also shown in Table 2. As can be seen, it is possible to achieve improved precision with longer scans. The errors reflect statistical uncertainty only. Additional error will be introduced by the uncertainty in the measurement of the distance of the X-ray beam from the edge of the drum, and in the estimation of the density of the sludge.

The X-ray K-edge technique performed very well on this set of simulated waste drums. It was possible to obtain measurement accuracy better than 10% for most samples within 20 minutes. Although cadmium could not be detected, its presence did not affect the measurement of lead contamination in the drums. Furthermore, there were no instances where false positive signals for lead or mercury were observed.

A limitation of this technique must be pointed out. The simulated waste drums were fabricated to have very homogeneous contents, and this homogeneity was crucial for the K-edge analysis. The X-ray beam could not penetrate a thick region of the drum, so only a small volume near the wall of the drum was sampled. If contaminants in the DOE legacy waste drums are not uniformly distributed, X-ray K-edge analysis will not give an accurate measure of the average concentration in the drum. However, if the sludge can be assumed to be homogeneous within a given layer in a drum, the K-edge technique could be used to scan along the edge of the drum to map a vertical profile of the contamination.



Figure 12. Setup for measuring RCRA metal contamination in 8-gallon waste drums. The X-ray tube is in the foreground and the collimated HPGe detector is behind the drum.

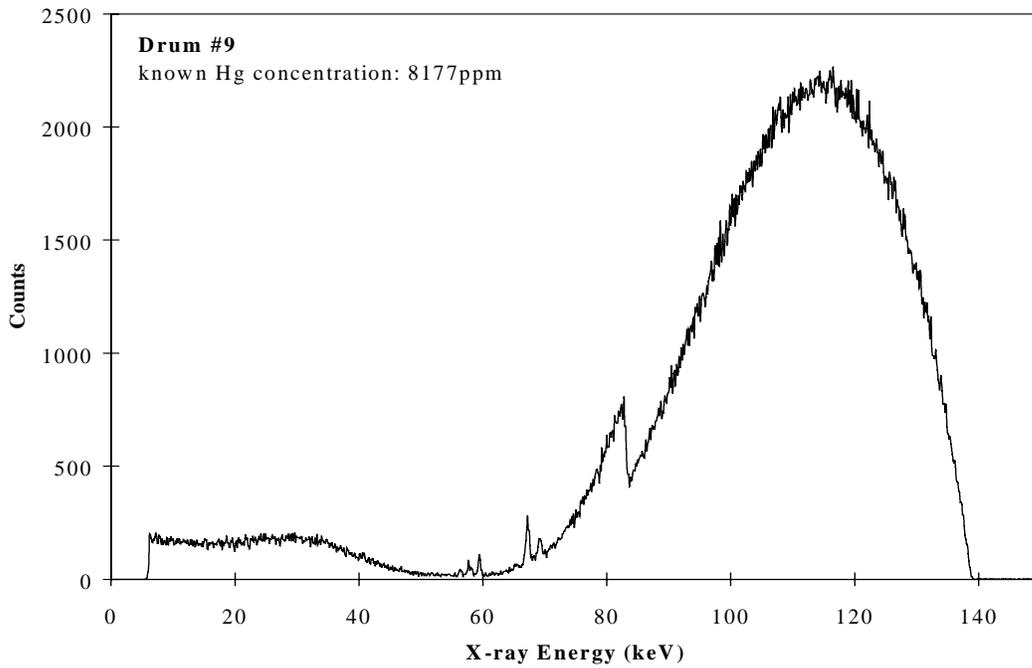


Figure 13. X-ray transmission spectrum observed for a waste drum containing mercury.

Table 2. Comparison of known and K-edge measured concentrations of mercury and lead in the different simulated waste drums. Upper limits are at 99.7% CL.

Drum #	SCAN TIME (min)	Known Cd (mg/kg)	Known Hg (mg/kg)	Measured Hg (mg/kg)	Known Pb (mg/kg)	Measured Pb (mg/kg)
1	20	0	0	<380	0	<276
1	30	0	0	<342	0	<295
2	10	0	18,110	18,128 +/- 440	0	<1232
2	20	0	18,110	17,974 +/- 308	0	<880
3	10	0	5,835	5,258 +/- 222	0	<563
3	20	0	5,835	5,728 +/- 171	0	<420
4	20	0	583	518 +/- 106	0	<305
5	10	4,970	0	<1024	18,080	19,160 +/- 292
5	30	4,970	0	<568	18,080	18,824 +/- 172
6	10	808	0	<568	8,844	7,957 +/- 166
6	20	808	0	<502	8,844	7,890 +/- 116
7	10	291	0	<509	0	<367
7	30	291	0	<294	0	<298
8	5	0	19,820	18,563 +/- 744	0	<1981
9	5	0	8,177	8,228 +/- 351	0	<891
10	5	0	1,942	1,915 +/- 250	0	<590
11	10	0	487	803 +/- 235	0	<853
11	60	0	487	399 +/- 72	0	<175
12	5	8,181	0	<1403	19,950	CALIB.
13	5	2,123	0	<909	9,927	10,342 +/- 285

4.4 Heavy Metal Contamination in Drain Pipes

From 1949 to 1953 the U.S. Government conducted research activities on thorium-232 in a building located on the Iowa State University campus, which is now called Harley Wilhelm Hall, and is under the control of Ames Laboratory, U.S. DOE. As a result of these activities various areas of this building were contaminated. Since cessation of thorium research activities in Wilhelm Hall, several surveys of the building have been performed and mitigation activities have been taken where feasible[11]. Although remaining contamination does not represent a hazard for normal day-to-day activities in the building, it presents risks that must be controlled whenever any modifications are made to the building, or when maintenance is done on some of the building infrastructure. One area of contamination is in many of the sink traps and drain lines in the building. Measurements made external to the pipes using portable radiation survey meters indicate significant contamination in several locations. However, quantitative estimates of contamination are difficult to obtain due to the unknown amount of scale inside the pipes, and the fact that the pipes are wrapped in asbestos insulation.

For K-edge inspections in Wilhelm Hall[12], the source and detector were mounted on a mechanical lift so they could be positioned around a drain line as indicated in Fig. 14. There was not enough space to mount the real-time X-ray imager in this instance. Instead, radiographs of the drain traps were taken. These were of mixed quality due to the constraints in locating the film and X-ray tube, but generally gave a feel for the location of scale and other deposits in the traps. Most traps showed quite a variation in X-ray absorption, with some having wires or large pieces of absorbing material embedded in the scale. For each of the drain lines a series of K-edge measurements were made to map out any contamination. An example spectrum from one of the drain traps is shown in Fig. 15. Note the ability to identify multiple contaminants in a single measurement.

The X-ray K-edge detector proved very capable of quantifying heavy metal deposits in these drain lines. The results are in qualitative agreement with radiation survey measurements. It will not be possible to obtain quantitative verification without removing the pipes. The K-edge measurements provided very good spatial resolution to define the limits of contamination. Deposits of mercury were also identified, a task that is not possible with most other non-invasive inspection techniques. This information should prove valuable in deciding how to remove these pipes in the future. Decisions on where to cut the pipes can be made so as to minimize the potential for contaminating the surroundings.

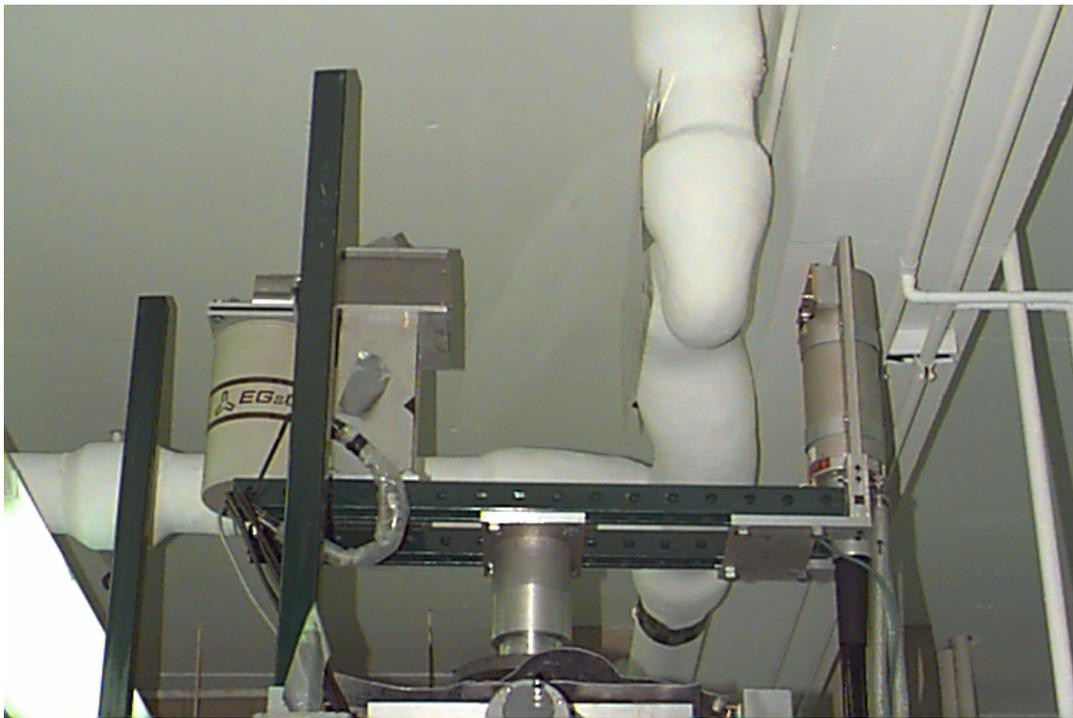


Figure 14. K-edge setup for inspecting a drain trap. The X-ray source is to the right, and the detector is to the left.

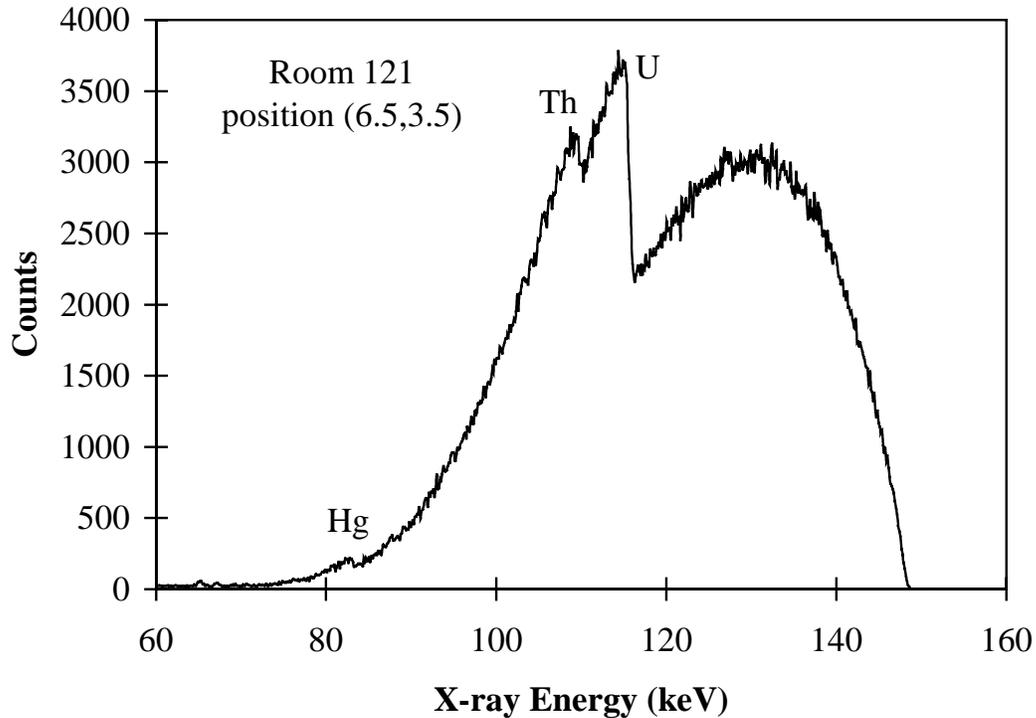


Figure 15. Spectrum from a region of the drain trap in room 121 showing signals for uranium, thorium and mercury.

4.5 Uranium Holdup in Machining Ducts at the Savannah River Site

From 1956 to 1992, the 321-M Facility at the Savannah River Site (SRS) fabricated fuel assemblies and extruded target assemblies for the SRS production reactors. The manufacturing process, combined with high ventilation flow rates, left dust, cuttings, and other forms of highly enriched uranium (HEU) in the building ventilation ducts and in the process equipment enclosures.

For a large-scale demonstration of the X-ray K-edge technology[13], a subset of equipment at the SRS 321-M Fuel Fabrication Facility was selected. The items targeted for K-edge inspection were exhaust ducts from two machining lathes (designated Lathe A and Lathe B) in the 321-M building. Figure 16 shows the placement of the inspection head around one of the ducts. The ducts are made of painted rolled steel approximately 1/16 inch thick. The larger portion of the ductwork is 20-inch diameter, and smaller 14-inch diameter ducts tee off of the main ducts. Approximately 84 feet of ventilation duct on the roof of the building were surveyed.

The typical measurement cycle for this demonstration entailed two to eight images and spectroscopic shots per linear foot of ventilation duct. Example images and spectra are shown in Fig. 17. In the images, darker regions correspond to areas where the duct wall is thicker, or where there is holdup material. The first image shows a large variation in density, and filaments of the type expected from lathe machining can be distinguished. The corresponding spectrum shows a strong signal for uranium. The second image shows a small, isolated dense object, and the corresponding spectrum indicates a very high concentration of uranium.



Figure 16. Method of manually positioning the K-edge inspection head around an exhaust duct. The X-ray tube is at the top of the C-frame, and the HPGGe detector and imaging detector are at the bottom.

The X-ray images provided rapid qualitative information that could be used to indicate areas where holdup material was likely located, and to distinguish between dust, lathe turnings, and chips. In general, it was noted that the holdup in the Lathe A duct was primarily dust, whereas that in the Lathe B duct was in the form of lathe turnings and small chips.

It was not feasible to make spectroscopic measurements at every point where there was an indication from the X-ray image for the presence of uranium. To obtain the total amount of uranium holdup in a region, a calibration was performed to relate the image density (in greyscale units ranging from 0 to 255) to the uranium density (in mg/cm^2) obtained from Eq. 1. Then, by also applying the distance scale for the image, it was possible to sum up, pixel by pixel, the total amount of uranium in that region. Each image covered approximately a 12 cm by 12 cm region of the duct. The ducts were divided into sequentially labeled sections corresponding to physical joints. The total uranium holdup in a section was obtained by adding the contributions from all images obtained for that section. To convert these results to an amount of U-235 holdup, the K-edge numbers must be multiplied by the enrichment fraction for the alloy mixture. For the lathe exhaust ducts, an enrichment of 70% U-235 was deduced from historical information on the materials processed by the lathes[14].

The K-edge results are displayed in the second column of Table 3 for each section of the ductwork. The uncertainties in these measurements are dominated on the low side by uncertainty in the conversion from image density to uranium density, and on the high side by the limit in sensitivity to small amounts of uranium spread over a large area. For those cases where no significant indication of uranium was found in a section of duct, an upper limit on the holdup at 95% confidence level is indicated. Those cases with relatively large uncertainties or upper limits are regions where few measurements were made. With more measurements these results would improve.

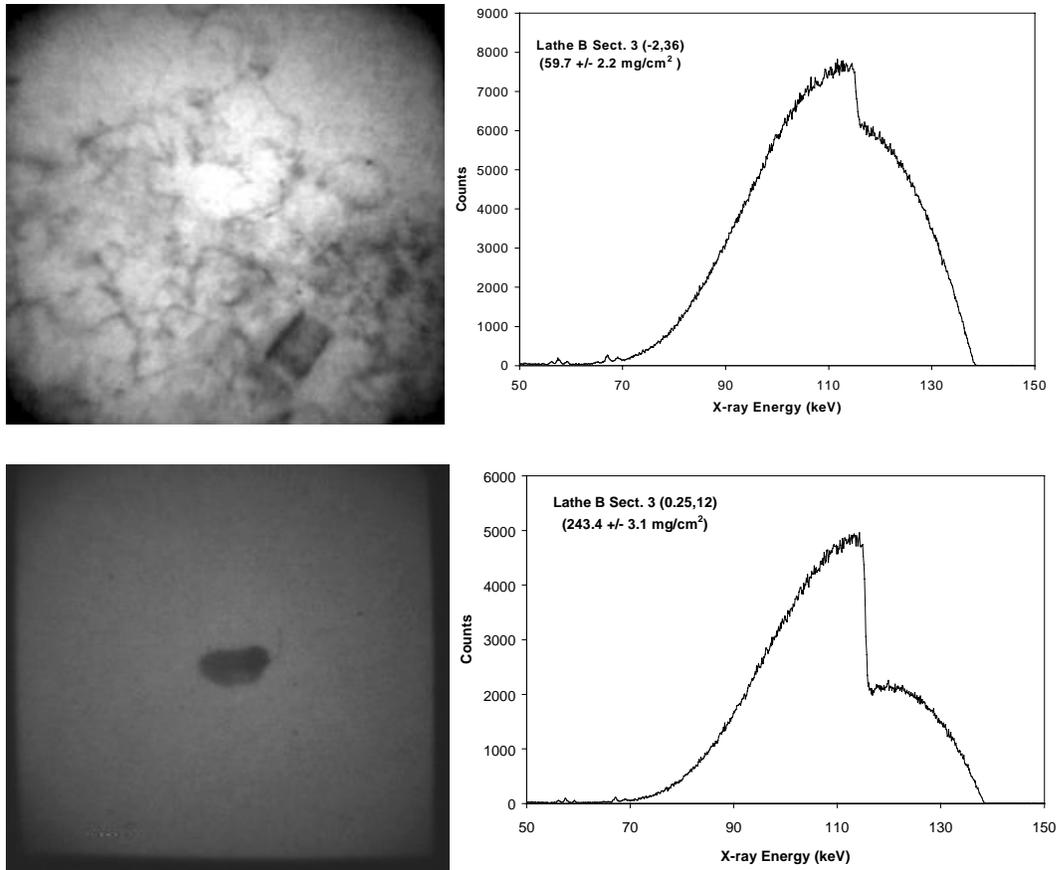


Figure 17. Example X-ray images from different positions along the ducts. Darker regions correspond to greater material thickness or density. To the right of each image is shown the X-ray spectrum observed at the center of the corresponding image.

In 1995 as part of the deinventory process for the 321-M Fuel Fabrication Facility, a survey was made of U-235 held up in ventilation ducts and process equipment in the building[14]. In this survey a 2x2 inch NaI detector was used with a lead collimator producing a 45 degree field of view about the axis of the detector. Along the regions of the ducts covered by the K-edge measurements a total of 18 readings were made using the NaI detector held at a typical distance of three feet from the duct. The results of this survey are shown in the third column of Table 1. (Sections 4 and 5 were surveyed together with the NaI detector for each duct.)

These results are in agreement with the K-edge measurements. However, there is a general trend in the NaI results indicating a relatively large holdup in the lower-numbered sections. As this survey was done four years before the K-edge measurements were made, there is a possibility that the material has moved downstream under the continuous operation of the exhaust system. To check this hypothesis, measurements were made using a radiation survey meter (Bicron μ rem) held against the bottom of the ducts. Average readings for each section of the ductwork are displayed in the fourth column of Table 3, and show the same trend as the K-edge measurements. On both ventilation systems, the highest radiation readings were found in the region where the large duct tapers down to the smaller duct.

Table 3. Comparison of K-edge and passive gamma NDA measurements of U-235 contamination in each section of the SRS 321-M exhaust ducts.

Duct Section	K-edge measurement (g)	NDA measurement (g)	Survey meter ($\mu\text{R/hr}$)
A-2	2.9 (+3.5/-1.4)	8.1 (+8.1/-4.0)	39
A-3	11.9 (+6.3/-5.6)	12.8 (+12.8/-6.4)	156
A-4	5.7 (+2.8/-2.1)	5.7 (+5.7/-2.9)	217
A-5	<7.0		53
B-2	<16	11.2 (+11.2/-5.6)	27
B-3	10.0 (+5.0/-3.0)	7.4 (+7.4/-3.7)	140
B-4	2.2 (+1.8/-0.7)	4.1 (+4.1/-2.0)	110
B-5	0.3 (+1.7/-0.1)		50

5.0 FUTURE PROSPECTS

Over the course of this project great improvements in the robustness of the equipment and in the speed and reliability of the analysis software have been made. As the technology for X-ray sources and detectors advances, one can anticipate further improvements in the K-edge technology.

The most serious drawback of the current K-edge prototype system is its size and weight. This has implications for accessibility as well as higher costs for transport and setup of the equipment. There are technological advances on the horizon that could dramatically improve this situation.

The energy-sensitive high purity germanium X-ray detector currently in use requires liquid nitrogen for cooling and occupies approximately one cubic foot volume. A detector based on a cadmium-zinc-telluride (CZT) crystal can operate at -30°C instead of at liquid nitrogen temperature (-196°C). This temperature can be maintained by a relatively small thermoelectric cooler, resulting in a detector package that occupies only a few cubic inches. However, in our tests of an early model of this type of detector, we found that it could not operate reliably at the high flux rates required for the K-edge analysis[15]. As CZT crystal fabrication improves one can expect this problem to disappear.

The real-time X-ray imager is also rather bulky (~ 1 cubic foot) and somewhat fragile. In the past few years significant advances have been made in the development of flat-panel amorphous silicon imagers that are much more compact and more rugged. These detectors also yield more uniform images having greater contrast.

The combination of these two types of detectors would produce a much lighter system that could be mounted on a small robotic arm. This would greatly reduce the manual setup time and enable much greater inspection throughput.

6.0 CONCLUSIONS

In the course of this project we have demonstrated that X-ray K-edge densitometry can be successfully applied to a wide range of heavy metal inspection applications. The coupling of an X-ray image with the quantitative K-edge analysis yields much more information about unknown samples than can be obtained from baseline nondestructive assay techniques.

Because the technique uses a high-power X-ray tube, special attention must be paid to radiation safety. The situation is really no different than what is encountered in the construction industry (for example, radiographic inspection of pipeline welds). In all of the field demonstrations we were easily able to assure personnel safety through use of mobile shields, interlock circuitry, and radiation monitors. The only drawback is that other work cannot be carried out in the area while inspections are being made.

Field applications of the K-edge technology have been carried out for inspection for the heavy metals mercury, lead, thorium, and uranium. The K-edge results compared favorably with baseline measurements from other techniques when they were available. Furthermore, the technique will work equally well with other heavy metals, such as plutonium, and it was demonstrated that multiple elements could be measured simultaneously.

For detection of uranium, good accuracy was demonstrated over a dynamic range from 10 mg/cm² to 8000 mg/cm² in a matrix of 1cm thick steel. It was further shown that the results are not sensitive to the matrix material or geometry. The improved information provided by the K-edge inspection technique can be used to reduce risk and increase the efficiency of cleanup operations.

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9. R. J. Gehrke and W. A. Propp, "Performance Test Results of Noninvasive Characterization of RCRA Surrogate Waste By Prompt Gamma Neutron Activation Analysis", Idaho National Engineering and Environmental Laboratory Report INEEL/EXT-97-01164 (October 1997).
10. T. Jensen, "Feasibility Study of X-ray, K-edge Analysis of RCRA Heavy Metal Contamination of Sludge Packaged in Drums", Ames Laboratory Internal Report IS-5139 (October 1999).
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12. T. Jensen and C. Whitmore, "X-Ray, K-Edge Analysis of Drain Lines in Wilhelm Hall, Ames Laboratory", Ames Laboratory Internal Report IS-5135 (January 1999).
13. "Portable X-ray, K-edge Heavy Metal Detector", *Innovative Technology Summary Report*, Deactivation and Decommissioning Focus Area & Characterization, Monitoring, and Sensor Technology Program, DOE/EM-0519 (October 1999). (This report is available on the OST web site at <http://OST.em.doe.gov> under "Publications.")
14. U-235 Holdup in Building 321-M Contamination Areas and Associated Exhaust Systems; WSRC Technical Report WSRC-TR-95-0492 (Rev. 0); December 19, 1995.
15. T. Jensen, T. Aljundi, and J. N. Gray, "X-ray Detector Selection for K-edge Heavy Metal Detection", Project Internal Memo (July 1995).

APPENDIX A. Project Funding History

Fiscal Year	OST Funding (\$K)	Other Funding (\$K)
1995	390	
1996	325	
1997	250	
1998	200	10 (Ames Lab demonstration support)
1999	200	34 (subcontract) + 70 (SRS onsite support)
2000	carryover only	

APPENDIX B. Project Milestones

FY95

1. Survey Oak Ridge needs:
Met with representatives from the K-25 Site and Y-12 Plant. Concluded that the K-edge technique could be useful in quantifying uranium deposition in the many miles of steel pipe at the gaseous diffusion plant.
2. Monochromator specification:
Experiments carried out using a highly oriented pyrolytic graphite crystal to select a narrow band of X-ray energies from a broad bremsstrahlung spectrum. Designed a monochromator that could be incorporated into the prototype K-edge detector.
3. Technique demonstration with gold:
Accurately known ICP solutions of gold were used to experimentally verify the sensitivity and accuracy of the K-edge technique. A gold sample 7 microns thick (13 mg/cm^2) behind 1.5 inches of aluminum could be measured with 15% accuracy in 5 minutes. A 2.5 micron thick (4.7 mg/cm^2) sample required 15 minutes measurement time for comparable accuracy.
4. Optimal detector selection:
NaI(Tl), room temperature CdZnTe, Peltier cooled CdTe, gas proportional, and Ge detectors were evaluated for application in the K-edge technique. The Ge detector proved to be the best choice for the K-edge technique.
5. Determine sensitivity for uranium:
Accurately known ICP solutions of uranium were used to experimentally verify the sensitivity and accuracy of the K-edge technique. A uranium sample 13 microns thick (24.6 mg/cm^2) behind 1.0 inch of steel could be measured with 17% accuracy in one minute. A 7 micron thick (13.3 mg/cm^2) sample behind one inch of steel could be detected in 5 minutes. Consistent results were obtained when a steel pipe was substituted for flat plates, indicating insensitivity to geometry.
6. Specification of portable device:
Based on the initial experiments, specifications were made for the design of the prototype X-ray K-edge detector.

FY96

1. Lab demonstration of the prototype K-edge detector:
The assembled prototype detector was tested in the lab using calibration samples of gold and uranium and different matrix materials.
2. Field test of the prototype:
A field test at the Oak Ridge K-25 Site was scheduled, but had to be postponed due to a failure of the X-ray high voltage transformer.

FY97

1. Field demonstration of the K-edge detector:
Two field demonstrations were carried out- measurement of uranium holdup in pipes at Oak Ridge K-25 Site, and determination of uranium concentration in reactor fuel plates at the ISU Nuclear Engineering Lab.

FY98

1. Operate K-edge detector in D&D Focus Area Large Scale Demonstration:
Due to changes at the D&D Focus Area and at field sites, it was not possible to schedule a Large Scale Demonstration during FY98. However, a field demonstration was carried out at Ames Laboratory using a more compact X-ray source, where measurements were made of heavy-metal contamination in drain lines.

FY99

1. Assemble modified compact source-detector package:
A compact source-detector package was designed and assembled, making the K-edge system more mobile.
2. Operate K-edge detector in D&D Large Scale Demonstration Project:
The K-edge system was operated for five days at the Savannah River Site 321-M Facility where 84 feet of ventilation ducting were inspected for uranium holdup.

FY00

1. Complete K-edge data acquisition and analysis software package:
Based on experience gained during the Large Scale Demonstration, the user interface was modified and updated to be compatible with the Windows-98 operating system.

APPENDIX C. Project Publications and Reports

The following is a chronological listing of the publications and reports generated throughout this project.

1. T. Jensen, T. Aljundi, and J. N. Gray, "X-ray Detector Selection for K-edge Heavy Metal Detection", Project Internal Memo (July 1995).
2. T. Aljundi, J.N. Gray, T. Jensen, C. Whitmore, and D. Robinson, "Development of a Portable X-ray, K-edge Heavy Metal Detector", Project Annual Report (September 1995).
3. Taher Aljundi, Terrence Jensen, Doug Robinson, and Joseph Gray, "Heavy metal contamination detection using X-rays", *Review of Progress in Quantitative Nondestructive Evaluation*, Vol. 15A, pp. 465, Edited by D.O. Thompson and D.E. Chimenti. Plenum Press, New York, N.Y. (1996).
4. Taher Aljundi, Terrence Jensen, Doug Robinson, and Joseph Gray, "Development of an X-ray K-edge Heavy Metal Detector", *Proceedings of the International Topical Meeting on Nuclear and Hazardous Waste Management, Spectrum '96*, pp. 418, The American Nuclear Society, La Grange Park, IL (1996).
5. T. Jensen, T. Aljundi, C. Whitmore, H. Zhong, and J.N. Gray, "Field Demonstration of a Portable, X-Ray, K-Edge Heavy-Metal Detector", Ames Laboratory Internal Report IS-5131 (March 31, 1997).
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7. T. Jensen, T. Aljundi, and J.N. Gray, "Application of X-ray K-Edge Densitometry in D&D Operations", *Proceedings of the International Conference on Decommissioning and Decontamination and on Nuclear and Hazardous Waste Management, Spectrum '98*, pp. 902, The American Nuclear Society, La Grange Park, IL (1998).
8. T. Jensen, "Feasibility Study for X-ray, K-edge Analysis of Concrete Core Samples from Wilhelm Hall, Ames Laboratory", Project Internal Memo, (December 1998).
9. "X-Ray, K-Edge Analysis of Drain Lines in Wilhelm Hall, Ames Laboratory", T. Jensen and C. Whitmore, Ames Laboratory Internal Report IS-5135 (January 5, 1999)
10. T. Jensen, "Feasibility Study of X-ray, K-edge Analysis of RCRA Heavy Metal Contamination of Sludge Packaged in Drums", Ames Laboratory Internal Report IS-5139 (October 1999).
11. Terrence Jensen, Craig Whitmore, and Jeffrey W. Lee, "X-ray K-edge Measurement of HEU Holdup in the Savannah River Site 321-M Facility", *Proceedings of the American Nuclear Society 2nd Topical Meeting on Decommissioning, Decontamination and Reutilization of Commercial and Government Facilities*, September 1999.
12. "Portable X-ray, K-edge Heavy Metal Detector", *Innovative Technology Summary Report*, Deactivation and Decommissioning Focus Area & Characterization, Monitoring, and Sensor Technology Program, DOE/EM-0519 (October 1999). (This report is available on the OST web site at <http://OST.em.doe.gov> under "Publications.")
13. Huaifu Lu, "Implementation of integrated computer control for X-ray material characterization systems", Masters Thesis, Iowa State University, June 2000, (unpublished).