

USING X-RAY, K-EDGE DENSITOMETRY IN SPENT FUEL CHARACTERIZATION

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ABSTRACT

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. To obtain accurate estimates of these parameters will require the combination of information from different inspection techniques. A method which can provide an accurate measure of the total uranium in the spent fuel is

X-ray K-edge densitometry. To assess the potential for applying this method in spent fuel characterization, we have measured the amount of uranium in stacks of reactor fuel plates containing nuclear materials of different enrichments and alloys. We have obtained good agreement with expected uranium concentrations ranging from 60 mg/cm² to 3000 mg/cm², and have demonstrated that these measurements can be made in a high radiation field (>200 mR/hr).

1. INTRODUCTION

At Idaho National Engineering and Environmental Laboratory (INEEL) there are many diverse types of spent nuclear fuel stored, including production fuels, experimental fuels, research reactor fuels, and commercial fuels¹. Records for some of the older inventory are not complete, leading to compliance problems with NRC, DOT, and IAEA requirements for qualifying the spent fuel for repository acceptance. In particular, the fissile material content, source term, and burnup of the spent fuel must be known. There is a similar need for spent fuel characterization at the Westinghouse Savannah River Company, where spent aluminum-clad fuel from foreign reactors is received for storage². The enrichment and burnup of this fuel must be confirmed. If credit for U-235 burnup can be established, the packing density of spent fuel can be increased, yielding substantial savings in storage space and cost.

Presently, nondestructive assay (NDA) methods based on gamma and neutron emissions are used to characterize spent fuel. However, these measurements require corrections based on the geometry and material properties, and incomplete knowledge of these quantities can lead to large uncertainties in the results. This is further complicated by the fact that some of these materials have corroded over time and some storage canisters contain water.

K-edge densitometry is a technique that could be used to improve the characterization of spent nuclear fuel. It was

demonstrated by Canada et al.³ in 1977 that this technique could be used to determine the number of fuel plates in a Materials Test Reactor fuel bundle. K-edge densitometry was originally developed for monitoring fuel processing operations, and optimized to measure tens of grams per liter of uranium and/or plutonium in solution^{4,5}. We have previously demonstrated the validity of K-edge analysis under a variety of circumstances⁶⁻⁸. In this report we describe the application of the K-edge technique to measurement of the total uranium content in reactor fuel plates. We give a brief description of the theory behind K-edge densitometry, and describe a series of measurements made on fuel plates from the Iowa State University UTR-10 reactor. We conclude with an analysis of the potential for applying the technique to characterization of spent nuclear fuel.

2. THEORETICAL BACKGROUND

The absorption of photons in materials is governed by the binding energies of atomic electrons. Each element has a unique distribution of atomic electrons, with the K-shell having the highest binding energy. When a photon has enough energy to liberate one of the electrons there will be an increase in the rate of absorption. In K-edge densitometry analysis, a X-ray beam is made to pass through a sample to a collimated energy-sensitive X-ray detector. At an energy corresponding to an absorption edge for an element, there will be an abrupt change in the transmission

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rate, resulting in a step in the transmitted X-ray spectrum, as indicated in Fig. 1. This change in transmitted intensity at the K-edge energy (115.6 keV for uranium) identifies the element. The amount of that element present can be calculated based on the size of the step according to the equation,

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

where $I(E_+)$ and $I(E_-)$ are the measured intensities on either side of the K-edge, as indicated in Fig. 2, and $\mu(E_+)$ and $\mu(E_-)$ are the corresponding absorption coefficients. (Note that K-edge analysis cannot distinguish between different isotopes of an element.)

To first order the accuracy is determined by the statistics of the intensity measurements. It is generally possible to achieve a precision of 5-10% within a few minutes measurement time. The intensities, I and I_+ are determined by extrapolating measurements to the K-edge energy. Proper selection of data for extrapolation is very important. Absorption edges from other elements which might be present must be avoided. For uranium, the adjacent elements in the periodic table, Pa and Np, have K-edges at 112.6 keV and 118.7 keV, respectively. The next-nearest neighbors, Th and Pu, have K-edges at 109.7 keV and 121.8 keV, respectively. For spent enriched uranium fuel, these elements will generally be present in such small concentrations that they will not be observable in K-edge spectra. Good detector energy resolution is essential to separate any effects from adjacent edges.

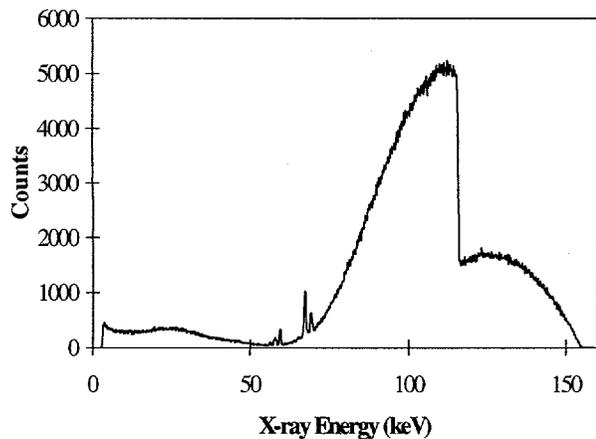


Figure 1. X-ray transmission spectrum observed for six plates of HEU fuel. The uranium K-edge is at 115.6 keV.

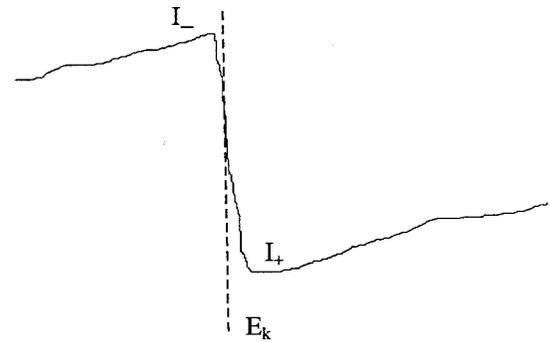


Figure 2. X-ray transmission spectrum in the vicinity of the K-edge of an element.

One must also assure that background gamma rays and X-ray fluorescence lines do not contribute to the signal regions selected for extrapolation. A number of the transuranic elements fluoresce at energies near the uranium K-edge, and the uranium K_β lines appear in the range 111-114 keV. These are reduced to insignificant levels through collimation of the detector. The solid angle accepted by the detector is typically $<10^{-4} \times 4\pi$. As can be seen in Fig. 1 there is no evidence for any fluorescence lines near the uranium K-edge. The only noticeable fluorescence lines are in the 60-70 keV range, from the X-ray tube tungsten target.

Finally, any background present at the K-edge energy must be accounted for before applying Eq. 1. For relatively thick amounts of material, background due to escape of compton-backscatter X-rays from the detector becomes significant. An example of this effect is shown in Fig. 3. We have developed techniques to account for this background by analyzing the entire transmission spectrum⁶. Further details on K-edge densitometry analysis can be found in Refs. 4-6.

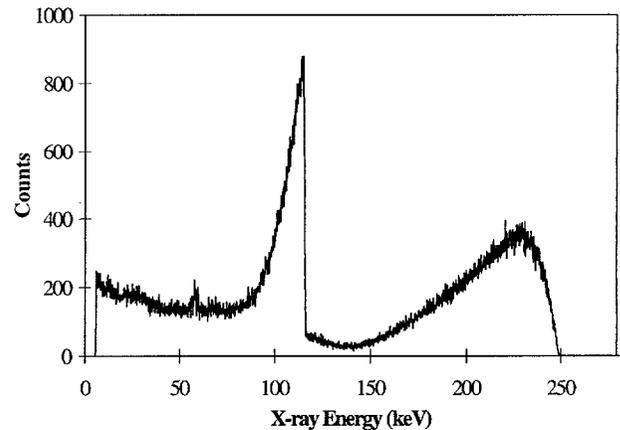


Figure 3. X-ray transmission spectrum for 14 plates of LEU fuel, showing significant background at the K-edge energy.

3. REACTOR FUEL PLATE MEASUREMENTS

To demonstrate the viability of using the K-edge technique in characterizing spent nuclear fuel, we performed a series of measurements on fuel plates from the UTR-10 University Teaching and Research reactor at the Nuclear Engineering Laboratory on the campus of Iowa State University. This reactor can be configured to operate with either high-enrichment uranium (HEU) or low-enrichment uranium (LEU) fuel. Both types of fuel plates have the same transverse dimensions (3.0 in. x 26.0 in.), but are of different thickness. Each LEU fuel plate is composed of U_3Si_2 in an aluminum matrix 0.020 inch thick clad on both sides with 0.015 inch aluminum. The enrichment is 19.75%, and from the manufacturer's specifications the nominal concentration of uranium is calculated to be 176.5 mg/cm^2 . The HEU plates contain a uranium-aluminum alloy 0.040 inch thick clad with 0.015 inch thick aluminum. The enrichment is 93%, implying a nominal uranium concentration of 58.7 mg/cm^2 . Most of the plates that we used had not been irradiated, which simplified measurement procedures. To demonstrate that background radiation from spent fuel would not bias the K-edge measurements, we also made some measurements using irradiated fuel plates.

The arrangement used in these experiments is indicated in Fig. 4. The X-ray tube has a maximum voltage of 320 kV and a maximum tube load of 4.2 kW. The beam is collimated to a diameter less than one cm at the sample, and the detector is further collimated to less than one mm diameter active area. The detection system consists of a high

purity germanium (HPGe) detector and a real-time X-ray imaging system. The HPGe detector and imaging system set next to each other on a platform that can be moved to center one system or the other on the X-ray beam. Because heavy metals are strong X-ray absorbers, they will generally produce good contrast in X-ray images. The real-time X-ray image provides a quick qualitative indication of heavy metal location. The HPGe detector can then be positioned to obtain a precise measure of the heavy metal. Operation of the K-edge system is controlled through a remotely located PC. Results on heavy metal content are provided in real time to the operator. Further details on the K-edge system can be found in Ref. 6.

Stacks of differing numbers of fuel plates were placed on the sample positioner for inspection. Additional matrix materials were placed around the plates to represent different storage conditions. The stack could be moved across the X-ray beam by remote control to obtain an image or a K-edge measurement at desired locations.

For all HEU plates, and for up to 7 LEU plates, the uranium concentration was determined from Eq. 1. The intensities were determined by performing least-squares fits to the data within the energy windows 111-114 keV and 117-121 keV. For cases of 8 or more LEU plates it was necessary to operate the X-ray tube at higher voltage resulting in significant background, which was corrected for as described in Ref. 6. Additional validation of these procedures was performed using depleted uranium foils of known thickness.

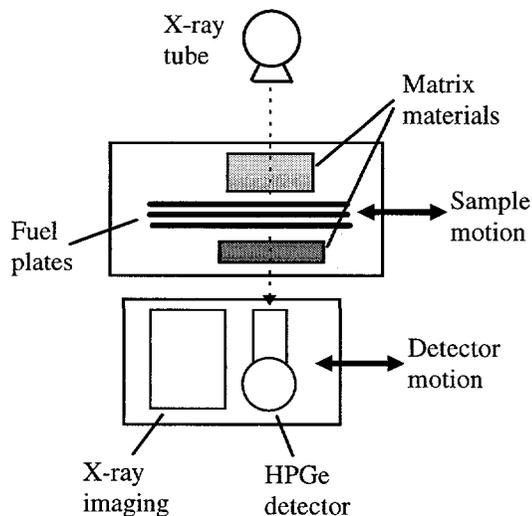


Figure 4. Setup for K-edge measurements on UTR-10 fuel plates.

Figure 5 shows a real-time X-ray image at one end of a LEU fuel plate. It can be seen that the ends of the fuel plates are somewhat irregular. This could introduce some error into the calculation of the nominal uranium concentration, which was based on the assumption of a rectangular fuel element. It is also possible that the uranium is not distributed uniformly throughout the plate. To test this we have scanned along several of the plates measuring the concentration at different points. Measurements taken near the end of a LEU plate are plotted in Fig. 6. Excluding the first four points at the edge of the plate, the average concentration is 174.8 mg/cm^2 , which compares very well with the predicted value (176.5 mg/cm^2). A coarse scan along a HEU plate yielded an average uranium concentration of 57.2 mg/cm^2 , also in good agreement with the predicted value (58.7 mg/cm^2). Based on a series of such scans, we conclude that the uranium is uniformly distributed in the fuel plates to within 10% of the nominal concentration.

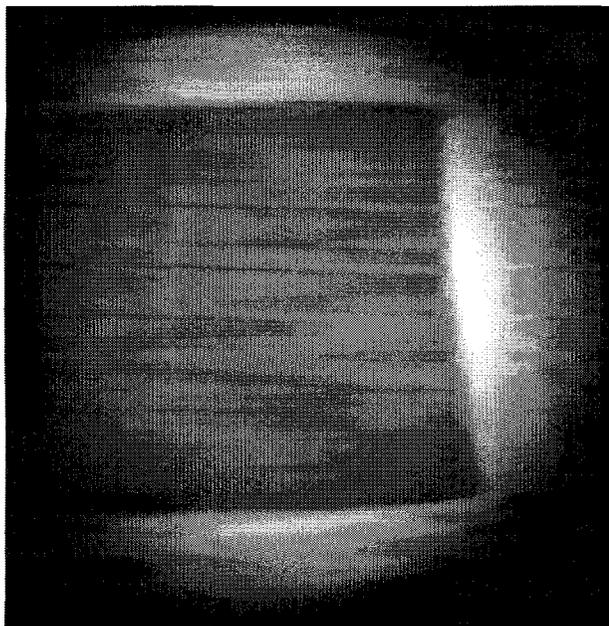


Figure 5. Real-time X-ray image near one end of a LEU fuel plate.

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, 1/2 inch aluminum plus 4 inches water, 1/4 inch steel, or 1/2 inch steel). For each arrangement we made five repeat measurements, collecting data for 2-3 minutes each time. The average of these five measurements is plotted in Fig. 7 as a function of the number of LEU plates being inspected. The error bars on these points correspond to the rms deviations for the five measurements. A similar set of measurements acquired for HEU plates is presented in Fig. 8.

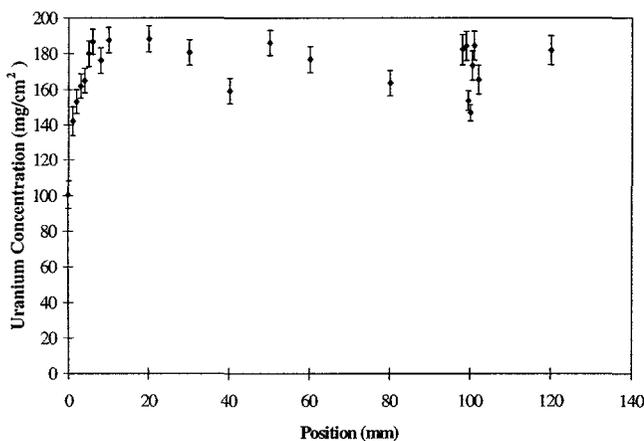


Figure 6. K-edge measurements as a function of position along a LEU fuel plate.

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. Least squares fits to the data yield average uranium concentrations of 182.4 mg/cm², and 61.4 mg/cm², for a single LEU, and HEU plate, respectively. These are within 5% of the calculated nominal concentrations.

We also had access to some HEU plates which had been irradiated. K-edge measurements were made on a single plate and on a stack of six plates as indicated in Fig. 8. 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 (1 R/hr on contact). It was possible to shield the detector so that this background radiation did not affect the K-edge measurements. The nominal shielding on the HPGe detector is 1/2 inch tungsten on the front and 1/4 inch tungsten on the sides. Under these conditions the worst case background observed at the K-edge energy was a few percent. By adding an additional 1/2 inch lead to the sides of the detector and two inches lead with a one cm diameter collimation at the front, this background was made negligible.

4. CONCLUSIONS

We have demonstrated that K-edge densitometry is a fast, accurate characterization technique for determining the total amount of uranium present in nuclear fuel plate assemblies. Accurate measurements of uranium concentration ranging from 60 mg/cm² to 3000 mg/cm² were obtained within a few minutes inspection time. The results were shown to be insensitive to a variety of matrix conditions, and operation of the system in a high radiation field was demonstrated.

The application of K-edge densitometry to characterization of spent nuclear fuel at INEEL or Savannah River would require integration of the K-edge results with NDA measurements. The real-time X-ray image could provide a good indication of the geometry and condition of the fuel assembly. K-edge measurements made at selected points would yield the total uranium content, which, when combined with the geometric information and NDA measurements, could be used to improve the accuracy of the measurements of enrichment and burnup.

K-edge densitometry is not applicable to characterization of all types of spent fuel. It will work best on fuels in the form of plates of the type used in these studies. For fuel in the form of rods there is the difficulty of knowing where on the cross section of the rod the measurement was made. A scan in fine steps across the rod may be necessary to determine

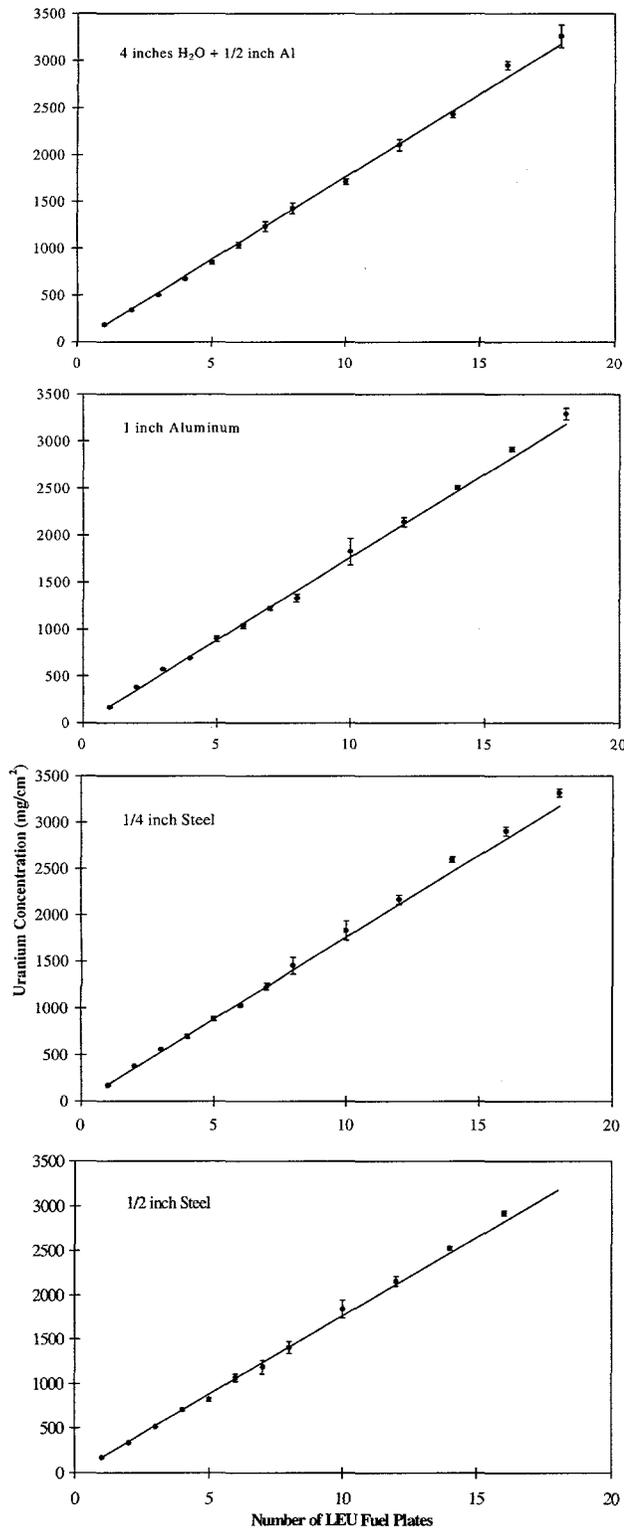


Figure 7. K-edge measurement of uranium concentration as a function of the number of LEU fuel plates in a stack for different matrix materials. The solid line indicates expected concentration based on the manufacturer's specifications.

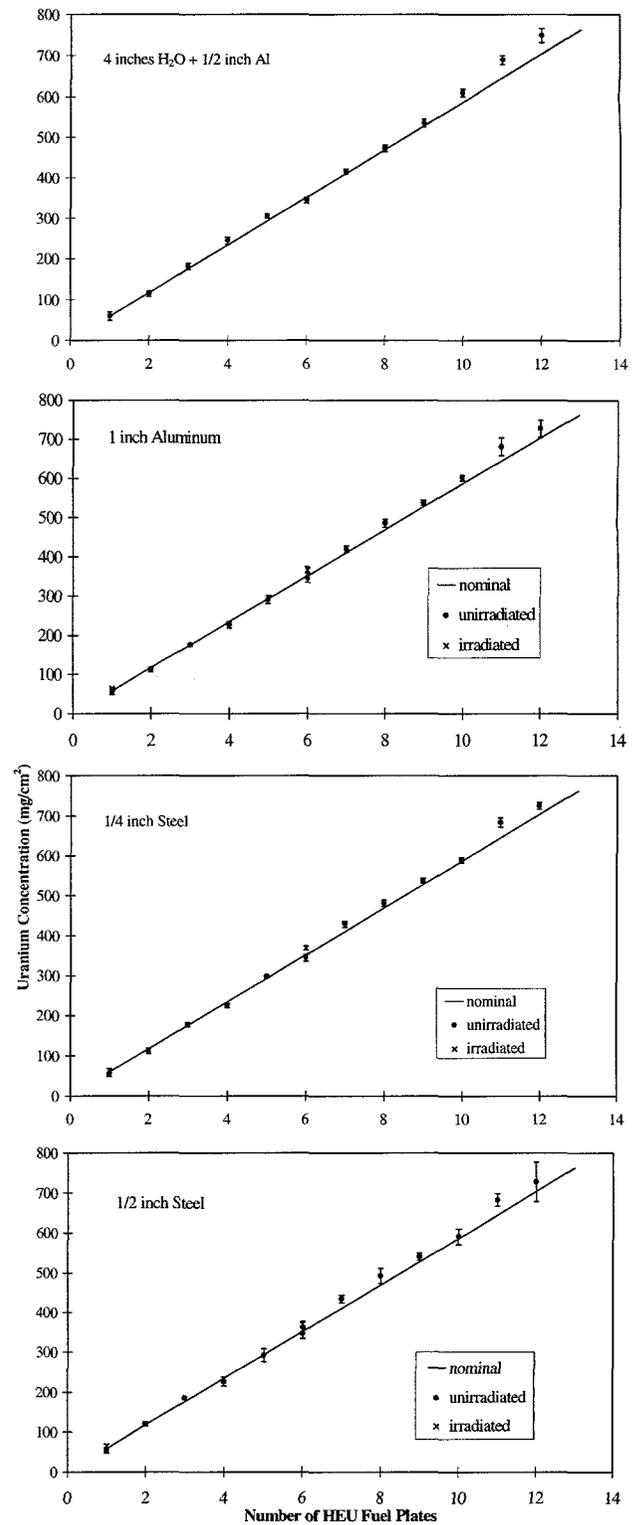


Figure 8. K-edge measurement of uranium concentration as a function of the number of HEU fuel plates in a stack. The solid line indicates expected concentration based on the manufacturer's specifications.

the uranium content. There is also a limit to the amount of material that can be penetrated. As seen in Fig. 7 we were able to measure up to 18 plates of LEU fuel plus surrounding matrix materials. In a previous demonstration⁶ we were able to measure up to 6000 mg/cm² of uranium holdup in a 1/8 inch wall monel pipe. Beyond these limits the signal to background decreases rapidly, and substantially longer acquisition times are required to obtain accurate measurements.

The fuel plates that we measured were relatively cool by spent fuel standards. Radiation levels up to several thousand R/hr are encountered in some fuel assemblies. It was fairly easy to shield the HPGe detector for 1 R/hr radiation levels, and there is room for improvement. Since the K-edge analysis interrogates the sample with a narrow pencil beam, the solid angle seen by the HPGe detector can be reduced to a very small value without affecting the K-edge signal. Some additional work is needed to optimize this collimation and verify operation at very high radiation levels. Of more serious concern is the operation of the real-time X-ray imaging system in such a high radiation field. The phosphor conversion screen may be overwhelmed by the background radiation.

In spite of these limitations there are a number of situations (especially characterization of some of the experimental reactor fuels) that could benefit from K-edge analysis. Given the improvements in technology since Canada et al.³ demonstrated that K-edge densitometry could be used to determine the number of fuel plates in a MTR fuel bundle, we feel that it is timely to reconsider K-edge densitometry as a useful aid in characterization of spent nuclear fuel.

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