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National Laboratory

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# KED/KXRF Hybrid Densitometer

May 1996

**Safeguards Science  
and Technology  
NIS-5**

MS E540  
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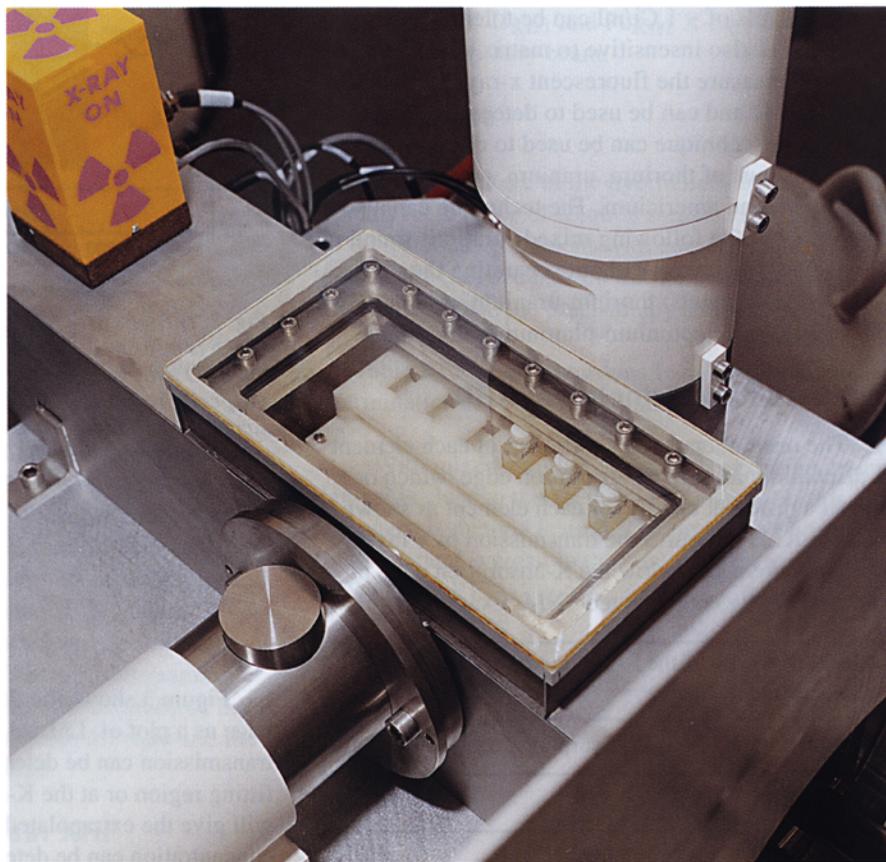
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**LALP-96-49**

Los Alamos National Laboratory, an affirmative action/equal opportunity employer, is operated by the University of California under contract W-7405-ENG-36 for the US Department of Energy.

This work was supported by the US Department of Energy, Office of Safeguards and Security.



*Fig. 1. The vertical cylinder on the right houses the x-ray tube. The x-ray beam shines through the collimator and the solution sample. The glass window in the middle shows the sample tray (with six sample positions), controlled by the computer. The sample tray moves in a tunnel attached to the glove box. The K-edge densitometer detector on the left measures the transmission beam. The XRF detector behind the x-ray tube measures the fluorescent x-ray.*

**T**he hybrid K-edge/K x-ray fluorescence (XRF) densitometer is a unique nondestructive assay (NDA) instrument for determining the concentrations of the isotopes of special nuclear material (SNM) in solutions. The technique is ideally suited to assay the dissolver solutions as well as the uranium and plutonium product solutions from reprocessing plants. The densitometer is an important instrument for safeguarding reprocessing plants; it is also useful in analytical laboratories because it can analyze mixed solutions of SNM without chemical separation. Figure 1 shows the hardware of a hybrid system developed at Los Alamos.

The hybrid densitometer employs a combination of two complementary assay techniques: absorption K-edge densitometry (KED) and x-ray fluorescence (XRF). The KED technique measures the transmission of a tightly collimated photon beam through the sample; KED is therefore insensitive to radiation emitted by the sample material. Fission product levels of  $\sim 1$  Ci/ml can be tolerated. The technique is also insensitive to matrix variation. XRF can measure the fluorescent x-rays from the same sample and can be used to determine the SNM ratios. The technique can be used to determine the concentrations of thorium, uranium, neptunium, plutonium, and americium. The technique can also be applied to the following mixed solutions found in the nuclear fuel cycle without separating the fissionable elements: thorium-uranium, uranium-plutonium, and neptunium-plutonium-amerium.

### Measurement Principles: KED

The mass attenuation coefficient of each element is discontinuous at its K-absorption edge, which occurs at a different energy for each element as shown in Table I. KED measures the transmission of x-rays at energies above and below the K-absorption edge in the most highly concentrated SNM. Examples of the

Element	K-Absorption Edge (keV)
Thorium	109.651
Uranium	115.606
Neptunium	118.676
Plutonium	121.818
Americium	125.027

spectra recorded by the K-edge detector in this system are shown in Fig. 2. The figure displays, in logarithmic scale, the spectra from a blank (3-M HNO<sub>3</sub>) solution (reference spectrum) and from a uranium solution with a 197 g/L concentration. The photon transmission decreases at the K-absorption edge of uranium (115.6 keV); the amount of the decrease depends on the uranium concentration. The transmission spectrum is the uranium solution spectrum divided by the reference spectrum, adjusted for the counting time difference.

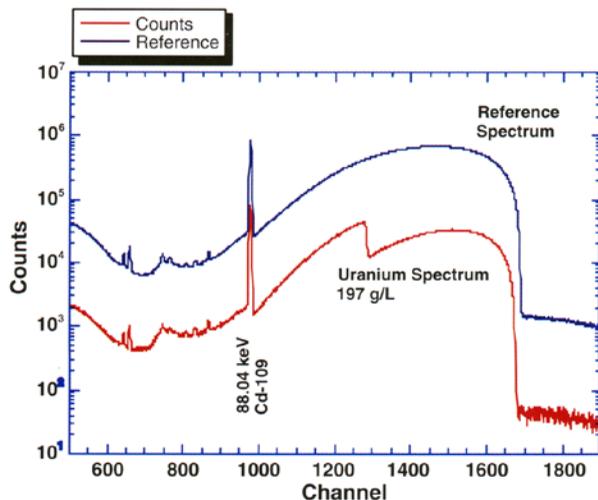


Fig. 2. The top curve (blue) shows the reference spectrum, which is measured with a blank (3-M HNO<sub>3</sub>) solution by the KED detector; it shows the continuous spectrum and the peaks from <sup>109</sup>Cd. The bottom curve (red) shows the sample spectrum from a uranium solution with a concentration of 197 g/L. The ratio of the sample spectrum to the reference spectrum gives the transmission of the solution.

Figure 3 shows the fitting region near the uranium edge as a plot of  $\text{LnLn}(1/T)$  versus  $\text{Ln}(E)$ . The transmission can be determined at the edge of the fitting region or at the K-absorption edge; the latter will give the extrapolated assay results. The concentration can be determined by the equation

$$\rho(\text{SNM}) = \frac{1}{\Delta\mu \cdot d} \ln\left(\frac{T_l}{T_u}\right)$$

where

$\rho$  (SNM) = concentration of the SNM in g/cm<sup>3</sup>,

$\Delta\mu$  = difference of the mass absorption coefficients at the transmission energies,

$d$  = solution sample thickness,

$T_l$  = transmission below the absorption edge,

$T_u$  = transmission above the edge.

The same method can be used to determine thorium, neptunium, plutonium, and americium concentrations using transmissions measured at the appropriate energies.

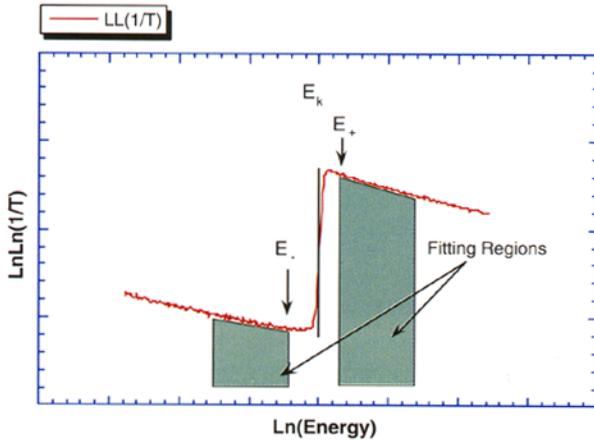


Fig. 3. This figure shows the fitting region for a single-element solution. The fitting regions are 6 keV wide. The  $K$ -absorption-edge energy of the SNM is  $E_k$ . The  $E_+$  begins 1.65 keV above the  $E_k$ ;  $E_-$  begins 2.35 keV below  $E_k$ .

The KED method can also be used to determine both the uranium-plutonium or thorium-uranium concentrations simultaneously if the ratio does not exceed 8 to 1. The new analysis method that we have developed improves the measurement precision by a factor of two over previous methods. For ratios exceeding 8 to 1, XRF is a much more precise method to determine the ratios of SNM concentrations. Once the ratio is determined, the concentration of the minor isotope can be calculated from the ratio and the concentration of the major isotope.

## Measurement Principles: XRF

The XRF measurement determines the ratios of SNM concentrations. Figure 4 shows a typical spectrum from a solution containing both uranium and plutonium. The broad "bump" of counts in the middle portion of the spectrum is caused by inelastic scattering of the primary x-ray beam, the largest contribution of which comes from scattering off the low- $Z$  elements of the sample.

The U/Pu weight ratio can be determined from the measured net peak areas of the fluoresced  $U_{K\alpha 1}$  and  $Pu_{K\alpha 1}$  X-rays:

$$\frac{U}{Pu} = \frac{At(U)}{At(Pu)} \cdot \frac{Area(U_{\alpha 1})}{Area(Pu_{\alpha 1})} \cdot \frac{R_{eff}(Pu_{\alpha 1})}{R_{eff}(U_{\alpha 1})} \cdot \frac{1}{R_{U/Pu}}$$

where

$At$  = atomic weight of uranium and plutonium,

$Area$  = net peak area of the  $K\alpha 1$  X-rays,

$R$  = relative detection efficiency curve,

$R_{U/Pu}^{eff}$  = calibration factor describing the ratio of excitation probabilities for emission of  $U_{K\alpha 1}$  and  $Pu_{K\alpha 1}$  X-rays in the primary beam.

## Performance

The performance of the hybrid densitometer depends on the type of solution sample. Table II lists the typical assay precisions with an assay time of 1000 s. The bias depends on the calibration and is often smaller than the precision by a factor of 2-5.

Table III lists the precisions for a single-element determination for 2-cm and 4-cm cells.

With mixed solutions, the precision of the major element is close to that of the single element if this new analysis method is used. The precision of the minor element is equal to the combined error of the XRF and the KED.

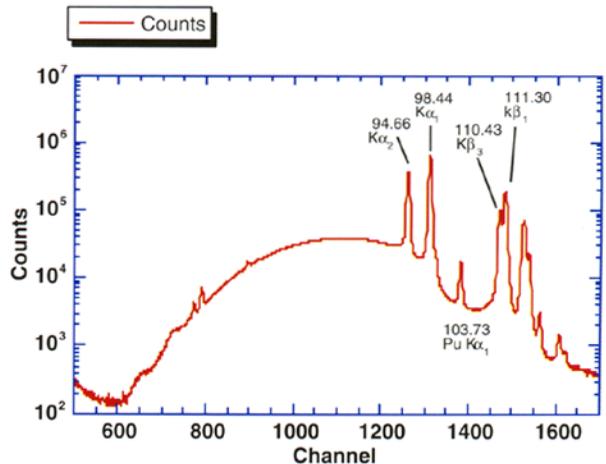


Fig. 4. This figure shows the fluorescent spectrum from a sample containing a mixture of uranium and plutonium.

## Calibration

Calibration of the hybrid densitometer should be planned carefully. First, the hybrid system should be calibrated with well-characterized solution standards. Because the precision of the hybrid system is  $\sim 0.2\%$ , the standards should be known to 0.05%. Second, the concentration of a standard solution is known to

increase with time. Standards should be used soon after preparation or stored in such a way that the concentration does not change. Plutonium solutions should be freshly prepared for each calibration. The saving grace is that because both KED and XRF are ratio methods, the calibration is quite stable. Several hybrid systems in plant applications have not changed in calibration in 5 years.

Table II. Typical Assay Precisions for the Hybrid Densitometer for 1000-s Assays.

Solution	Concentration	Meas. method	Precision (%)
LWR dissolver	200 gU/L	KED	0.2
	2 gPu/L	XRF + KED	0.75
LWR product	200 gPu/L	KED	0.2
LWR product	200 gU/L	KED	0.2

Table III. Precision of the Hybrid Densitometer with 2- and 4-cm Cells for Single Elements.

Concentration (g/L)	Precision (%) 2-cm cell	Precision (%) 4-cm cell
10	2.64	1.35
20	1.08	0.71
50	0.472	0.338
100	0.285	0.234
150	0.231	0.224
200	0.212	0.247
250	0.210	0.297
300	0.216	0.376

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