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Computer Simulation of the L_{III} -Edge Densitometer

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CONTENTS

ABSTRACT	1
I. INTRODUCTION	2
II. MODELING THE L_{III} -EDGE DENSITOMETER	3
A. Step 1 - The Incoming Beam	5
B. Step 2 - Transmitting the Beam Through a Solution	6
C. Step 3 - Analyzing the Transmitted Spectrum . .	7
III. RESULTS	7
A. Detection System Resolution	9
B. Small-Angle Scattering	9
C. Variations in Solution Matrix	12
D. Elemental Contaminants	14
E. Shape of the Incoming Beam	15
F. Mixed Solutions--The Effect of the Uranium/ Plutonium Ratio	17
G. Assay Analysis Technique	18
IV. CONCLUSIONS AND RECOMMENDATIONS	22
REFERENCES	25

COMPUTER SIMULATION OF THE L_{III} -EDGE DENSITOMETER

by

Diana Langner

ABSTRACT

Since the L_{III} -edge densitometer was first fielded in 1977, it has displayed a 1 to 1.5% nonlinear deviation from its theoretical linear calibration response. In an effort to explain this nonlinear deviation, this study used a simple, closed-form computer simulation to examine the effects of several variables on the densitometer's measurement of uranium and plutonium in solution. The results of this simulation suggest that the variables that contribute to this nonlinearity include the effects of small-angle scattering and the detection system resolution function. The simulation also examined the effects of matrix contaminants, the shape of the incoming beam, the uranium-to-plutonium ratio for mixed solutions, and the data-reduction technique. All of these variables were found to have some effect on the assay results, although these were generally small. The calculations demonstrate that using a new edge-extrapolation data-reduction technique reduces the instrument's sensitivity to many of these variables.

I. INTRODUCTION

Since 1977 the Nuclear Safeguards Assay Group of Los Alamos National Laboratory has been designing and fielding L_{III} -edge densitometers. The operation and performance of these instruments are well described in the literature.^{1,2} These densitometers measure uranium, plutonium, or a uranium-plutonium mixture in solutions whose concentrations typically range from 1 to 100 g/l. Ideally, a system will have a linear response to the special nuclear material (SNM) in solution over most of this range of concentration. In both field and in-house tests of uranium solutions, the system response has been observed to be linear except for a small but statistically significant non-linearity at high concentrations. As seen in Fig. 1, this nonlinearity is at most 1 to 1.5%.

Fig. 1. Ratio of typical uranium assay results to known uranium solution concentrations. These measurements were performed on an L_{III} -edge densitometer that was fielded at the New Brunswick Laboratory. For comparison, the ratio of destructive analysis of these solutions to their known concentrations is also plotted.

This small, nonlinear characteristic of the L_{III} -edge densitometer's response to uranium solutions at high concentrations has been persistent enough to invite wide speculation as to its cause. Because of the expense of generating standard solutions, of chemically analyzing them after measuring them on the densitometer, and of decoupling the many physical parameters that were identified as possible causes of the nonlinearity, it has proven difficult to identify the exact source of the nonlinearity experimentally. For this reason, this calculational study was undertaken.

The purpose of this study was to analyze the L_{III} -edge densitometer's response using simple, closed-form computer simulations that could be run on a PDP-11/60 computer. This approach made it possible to quickly calculate a theoretical assay value for a large number of solutions while varying parameters one at a time. It also made it possible to identify those variables that will be important in later, more complex Monte Carlo-type calculations. The large data base, which has been generated over the years in the many field tests of the densitometer, served to tie the simulations to reality. In particular, the L_{III} -edge densitometer that was fielded at the New Brunswick Laboratory (NBL)^{3,4} was used as a standard for these calculations.

II. MODELING THE L_{III} -EDGE DENSITOMETER

The beauty of the L_{III} -edge densitometry technique is its theoretical simplicity. The measurement is essentially a transmission measurement. A well-collimated continuous x-ray beam, I_0 , passes through a solution of thickness x . For simplicity we will assume the solution contains uranium, U, and some solvent matrix, M. The exiting beam, I , is measured using a Si(Li) detector.

The exiting beam spectrum is compared to a reference beam spectrum, I_{ref} , which has been generated previously by measuring the same well-collimated x-ray beam passed through a reference solution. This reference has the same geometry as the uranium solution but contains only a matrix solution M' , which is similar to but is not necessarily identical to that of the uranium solution. Simple equations describe this situation. For a discrete energy, i ,

$$I = I_0 e^{-\mu_{iU}\rho_U x} e^{-\mu_{iM}\rho_M x}, \quad (1)$$

where μ_{iU} and μ_{iM} are the mass attenuation coefficients for uranium and the matrix at energy i , and ρ_U and ρ_M are the concentrations of uranium and the matrix. Similarly, for the reference solution,

$$I_{\text{ref}} = I_0 e^{-\mu_{iM}\rho_M x}. \quad (2)$$

The transmission spectrum, T , is computed by the L_{III} -edge densitometer's computer. At energy i ,

$$T_i = \frac{I}{I_{\text{ref}}} = e^{-\mu_{iU}\rho_U x} e^{-\mu_{iM}\rho_M x} e^{+\mu_{iM}\rho_M x}. \quad (3)$$

Consider now another energy, j , which with i brackets the large discontinuity in μ_U at the L_{III} edge. If we take the ratio of the transmissions measured at these two energies, we can define

$$R = \frac{T_i}{T_j} = e^{-\Delta\mu_U\rho_U x} e^{-\Delta\mu_M\rho_M x} e^{+\Delta\mu_M\rho_M x}, \quad (4)$$

where $\Delta\mu$ is the difference in the respective mass attenuation coefficients over this energy range. Because the reference is tailored so that its makeup is similar to the matrix solvent in the uranium solution, and because any residual matrix $\Delta\mu$ will be small compared to $\Delta\mu_U$ for energies close to the L_{III} edge, we have the very good approximation that

$$\ln R \approx -\Delta\mu_U \rho_U x \quad . \quad (5)$$

In fact, this treatment is quite general and can easily be extended to other SNM solutions including plutonium and mixed SNM solutions.

If all the above conditions are met, we should expect from Eq. (5) that an experimental plot of $\ln R$ versus uranium concentration will be linear. From Fig. 1 we see that except for the small downward tail at high concentrations the approximations made to reach Eq. (5) are indeed very good ones.

To simulate the L_{III} -edge measurements, calculations were done in three steps. First, an incoming beam function was computed and transmitted through a reference solution using equations similar to Eq. (2). Second, the incoming beam function was transmitted through a solution containing uranium, plutonium, or both. Finally, the calculated SNM solution spectrum and the calculated reference spectrum were input to an analysis program, which produced an assay result using exactly the same techniques as used by the L_{III} -edge densitometer's analysis routines. In each of the first two steps calculational assumptions were kept simple so that all computations could be made on a PDP-11/60 computer in reasonable processing times. These assumptions are described below.

A. Step 1 - The Incoming Beam

Two restrictions were applied in choosing the function to be used in calculating the incoming beam. As with all functions used in this study, it had to be simple. Second, it had to be a good approximation of the actual x-ray beam used by the NBL L_{III} -edge densitometer. Because this beam has never been measured directly because of its high intensity, the criterion used to decide on a beam function was that the function produced a good approximation of an experimental spectrum obtained by transmitting the L_{III} -edge densitometer's x-ray beam through a water sample. A plot of the experimental water spectrum and the chosen beam function calculationally transmitted through water is shown in Fig. 2. This function consists of two separate Gaussians, which describe the upward and downward tails, and a quadratic, which describes the peak region. The peak counts for the two Gaussians were chosen to match the experimental spectrum. The quadratic parameters were chosen to smoothly link the two Gaussian tails.

Fig. 2. A calculated water spectrum compared to a spectrum obtained on the NBL densitometer.

B. Step 2 - Transmitting the Beam Through a Solution

To compute a calculational spectrum for a solution containing matrix and SNM, the atomic constituents of the solution, the molecular fractions for the matrix solution, and the densities of the solution components were input. A transmitted spectrum was then computed on a 100-point energy grid from 15- to 20-keV energy using total mass absorption coefficients and equations similar to Eq. (1). The intensity "lost" because of incoherent scattering was next calculated by comparing the incoming beam intensity at a given energy with a transmitted intensity computed using only incoherent mass absorption coefficients.

For the sake of simplicity, the scattered intensity was assumed to be distributed uniformly in energy from the energy of interest to the Compton edge

for that energy. Scattering was assumed to be equally likely in all directions, but only scattering that originated on the axis of the incoming beam was counted. Multiple scatterings were allowed, and the size of the detector aperture as well as its distance from the solution were taken into account. The solution thickness, the path length from the solution, ℓ , and the detector aperture diameter, d , were chosen to match the values for the L_{III} -edge densitometer, which was fielded at NBL. To observe the effect of scattering on these computations, the ratio, ℓ/d , was varied from 10 to 30. The value of this ratio for the NBL machine is about 19.

Once the absorbed and scattered intensity have been computed, the effect of the detector-counting electronics system is simulated by convolving the computed transmitted intensity with a normalized Gaussian. A Gaussian was chosen for its simplicity. It was assumed the system response did not change over the 15- to 20-keV range of the calculations. The full width at half maximum (FWHM) for this Gaussian was chosen to match that observed from the stabilization sources of the NBL L_{III} -edge densitometer. This was about 0.3 keV. The effect of this resolution function was also computed for FWHM = 0.1 and 0.5 keV.

C. Step 3 - Analyzing the Transmitted Spectrum

The analysis technique used to compute an assay result from the calculational spectra was identical to that used by previous L_{III} -edge densitometers. Briefly, a transmission spectrum is computed using the calculated spectrum for the SNM-bearing solution and the calculated reference spectrum. The natural logarithm is taken and a two-point running difference computed. This produces a spectrum as shown in Fig. 3. The area under the peak less the background is proportional to the elemental concentration. When a calibration constant is applied to this area, an assay value results.

III. RESULTS

When this study was conceived, three phenomena were chosen for study using the above-described calculational technique. These phenomena were thought to be the most likely sources of the observed nonlinearity in the L_{III} measurements. They were

Fig. 3. A typical difference spectrum for a uranium-bearing solution. A running difference is computed from the natural logarithm of the smoothed transmission spectrum. The assay value is deduced from the area under the difference peak.

- (1) the detection system resolution,
- (2) small-angle scattering, and
- (3) variations in the makeup of the matrix solvent solution compared to the reference solution.

As the study progressed, however, and confidence in the computer simulations increased, the effects of several other parameters were studied in the hope of further understanding the L_{III} -edge densitometry technique and of improving the analysis technique. These were

- (4) elemental contaminants,
- (5) the shape of the incoming beam as a function of energy,
- (6) the uranium/plutonium ratio for mixed SNM solution assays, and
- (7) the method used to reduce the experimental data to an assay value.

To present the results of the computations for these parameters, several conventions were adopted. When one parameter was varied, all others were kept at nominal values as defined by the L_{III} -edge densitometer, which was fielded at NBL. All assay results were divided by the known SNM concentration so that the variation from linearity caused by the parameter of interest could be observed. In addition, these normalized concentration results were sometimes further normalized to that result obtained at the midpoint concentration of the nominal operating range of the NBL densitometer. For uranium, this midpoint is 50 g/l. Henceforth, the results normalized in this way will be called the normalized assay response.

Except for mixed solutions, the results given below are for uranium solutions, which were varied in concentration from 5 to 90 g/l. Plutonium or other SNM would yield identical results, providing system parameters were optimized for their measurement.

A. Detection System Resolution

Varying the width of the detection system resolution function has a definite effect on the assay. Figure 4 shows the normalized assay response for three Gaussian system resolution functions whose FWHMs were 0.1 keV, 0.3 keV, and 0.5 keV. The NBL densitometer's response has a FWHM of ~0.3 keV. If we compare Fig. 4 with a replot of the data taken at NBL shown in Fig. 5, we see that the simulation predicts the shape of the assay response very well. As the system resolution degrades (FWHM = 0.5 keV), the assay response also degrades. However, if the system resolution improves (FWHM = 0.1 keV), the assay response flattens and becomes closer to the ideal of a linear response.

B. Small-Angle Scattering

Small-angle scattering also contributes to the nonlinearity of the assay response function. Figure 6 shows the simulation results for four scattering configurations. The amount of scattering entering the detector was varied by changing the ratio of the path length, ℓ , from the solution to the detector aperture to the diameter of the aperture, d . For no scattering, but for all other parameters set at values determined by the NBL densitometer, the assay response is quite flat. However, it has a small negative slope caused by the effect of the system's resolution function. As scattering increases for $\ell/d = 30, 19, \text{ and } 10$, the assay response becomes increasingly nonlinear.

Fig. 4. The calculated normalized assay response to variations in the system resolution function.

Fig. 5. The NBL calibration data from Fig. 1 replotted. These data are normalized to the average response at 50 g/l of uranium.

Fig. 6. The calculated normalized assay response to variations in the amount of small-angle scattering that can get into the densitometer's detector.

The nominal value of l/d for the NBL densitometer was 19. At this nominal value, the simulations compare very well qualitatively with the assay response data obtained at NBL (Fig. 5). However, the simulations underestimate the magnitude of the nonlinearity.

The calculational model allows for only on-axis scattering. It also assumes that scattering is equally likely in all directions. In the NBL densitometer, however, the incoming beam has a finite width determined by a 5-mm-diam brass collimator, and thus off-axis scattering is likely. Furthermore, in reality, scattering is more probable for forward angles. Thus, the model underestimates the amount of scattering entering the detector. To increase the

amount of scattered intensity reaching the detector using this model, one must decrease the ratio ℓ/d . This suggests that the "effective" ℓ/d for the NBL densitometer relative to the modeling assumptions is smaller than 19. From Fig. 6, decreasing ℓ/d results in larger tails at low and high-uranium concentrations. Thus, correcting the scattering underestimations of the model would likely produce an assay response curve more in quantitative agreement with the data taken at NBL.

C. Variations in Solution Matrix

Perhaps the most fundamental assumption made in using the L_{III} -edge densitometry technique is that the reference standard is so similar in makeup to the SNM solution matrix that matrix effects can be neglected. In practice, however, a user may wish to measure SNM-bearing solutions that are different in matrix qualities from that of the reference standard currently available. Thus, it seemed valuable to examine computationally the effect of variations in solution matrix on the assay result.

Two scenarios were examined:

- (1) Calibrating the instrument using a reference matrix such as water and measuring SNM solutions of a different matrix type such as nitric acid (HNO_3); and
- (2) Calibrating the instrument for solutions having a certain acid matrix but measuring solutions having a different acid molarity.

Figures 7 and 8 show the results. In Fig. 7, the instrument is computationally calibrated using an H_2O reference and uranium-water solutions. This calibration produces the curve with the least nonlinearity. When uranium-nitric acid solutions are measured for three molarities of acid, but using a water reference, much greater nonlinearity is observed, particularly at low uranium concentrations. This nonlinearity increases as the molarity of the acid increases.

In Fig. 8, the instrument is calibrated using 3M HNO_3 -uranium solutions and a 3M HNO_3 reference. The resulting curve is virtually identical to the H_2O curve in Fig. 7. If the molarity of the acid is increased but the

Fig. 7. The calculated effect of variations in solvent matrix on the measurement of uranium solutions for a system calibrated for uranium-water solutions.

Fig. 8. The calculated effect of variations in the molarity of a nitric-acid solvent matrix on the measurement of uranium solutions for a system calibrated for uranium - 3M HNO₃ solutions.

3M HNO_3 reference is used, the assay result decreases. If the molarity decreases, the assay result increases. Again this effect is most dramatic at low concentrations.

These results suggest that the accuracy of the L_{III} -edge densitometry technique is sensitive to variations in the matrix of the SNM-bearing solution relative to the reference solution used. Whenever possible, the reference should match the matrix of the SNM-bearing solutions. If it is known that the molarity of the acid in the SNM solutions will vary in a process, but the exact molarity of the acid in the solution to be measured is not known precisely, the midpoint molarity of the variation should be used for the reference standard. Also, in this situation, the user must remember that measurements of solutions containing low concentrations of SNM can result in large biases, in addition to the usual large uncertainty associated with poor precision at low concentrations.

In addition to the scenarios mentioned above, calculations were also performed to observe the effect of the matrix solution on the assay result when the reference is matched to that of the SNM-bearing solutions. This was done for uranium- H_2O solutions, uranium-100% HNO_3 solutions, and uranium-hydroxylamine solutions. The three curves produced were almost identical and displayed no more than a 0.15% variation at 5 g U/l, 0.05% variation at 50 g U/l, and were calculationaly identical at 90 g U/l. This confirms that if the matrix of the SNM solution is known and is matched to the reference standard used for the measurement, the matrix can be varied a great deal without substantially affecting the accuracy of the measurement.

D. Elemental Contaminants

Ideally, L_{III} -edge densitometry is insensitive to elemental contaminants whose electronic energy levels are distinct from the SNM element being measured. As shown above, however, variations in the solution matrix can affect a densitometry measurement to varying degrees. Because of these results, calculations were performed to study the effect on the measurement of small amounts of elemental contaminants whose atomic numbers (Z) were higher than those usually encountered in the solvent matrix. Three contaminants were chosen: aluminum (Z = 13), iron (Z = 26), and gadolinium (Z = 64). Figure 9 shows the results for a 50 g/l uranium-water solution with varying amounts of these three elements. Each of the three contaminants causes the assay to decrease with

Fig. 9. The calculated effect of elemental contaminants on the L_{III} -edge densitometer's measurement of a 50-g/l uranium solution.

increasing contaminant concentration. As the Z of the contaminant increases, this effect becomes more dramatic. This effect is approximately linear for a given element.

E. Shape of the Incoming Beam

The x-ray beam spectrum used by the NBL L_{III} -edge densitometer was tailored so that the L_{III} edges for uranium and plutonium fall in the flattest region. One question that arose during this study was how the assay results obtained using the current beam compare to those for an idealized, flat beam. Another question was how a narrower, less well tailored beam would effect the assays.

Fig. 10. The calculated effect of variations in the shape of the incoming x-ray beam on the normalized assay response.

Figure 10 shows the calculated normalized assay response for the nominal beam shape for the NBL instrument, for an ideal flat beam, and for a beam that is 20% narrower than the nominal beam but has the same peak position. The flat beam and the narrow beam responses are normalized to the nominal beam response at 50 g/l. These calculations show that the nominal beam is very well tailored and that the shape of the beam contributes only minimally to the non-linearity of the assay response. These calculations also show that narrowing the beam by 20%, which causes the background regions used for calculation of the assay to be more sloped, results in only a 20-30% increase in the nonlinear variation of the assay response.

F. Mixed Solutions—The Effect of the Uranium/Plutonium Ratio

When measuring mixed uranium-plutonium solutions, the L_{III} -edge densitometer has performed well. Figures 11 and 12 show assay results obtained at NBL for uranium-plutonium solutions having uranium/plutonium ratios from 1 to 5. These results, as well as those obtained before the instrument was fielded at NBL, show a very small tendency for the assay result to decrease as the uranium/plutonium ratio increases. The statistics on these measurements are poorer than for single-substance measurements because of the difficulty in resolving the region between the uranium and plutonium L_{III} edges. For plutonium in mixed solutions, the statistics are reduced even more because of the reduction in counting precision caused by the uranium matrix.

Fig. 11. The NBL calibration data for uranium in mixed uranium-plutonium solutions having a uranium/plutonium ratio that varied from 1 to 5.

Fig. 12. The NBL calibration for plutonium in mixed uranium-plutonium solution having a uranium/plutonium ratio that varied from 1 to 5.

Figures 13 and 14 show calculated assay responses for mixed solutions having uranium/plutonium ratios from 2 to 5. These results confirm the observed tendency of the experimental results to decrease as the uranium/plutonium ratio increases. These calculations suggest that in future L_{III} -edge systems, the assay value should be assigned using a two-pass procedure. During the first pass, a uranium-to-plutonium ratio would be deduced based on a single calibration constant for uranium and plutonium, as in the current method. The calculated uranium-to-plutonium ratio would then be used to correct the initial calculations for the uranium/plutonium effect observed in Figs. 13 and 14.

G. Assay Analysis Technique

The difference technique used to calculate the assay result for an L_{III} -edge measurement has an advantage in that it is easy to detect the

Fig. 13. The calculated effect of the variation of uranium/plutonium ratio on the uranium measurement for mixed solutions.

Fig. 14. The calculated effect of the variation of the uranium/plutonium ratio on the plutonium measurement for mixed solutions.

region in which the edge and resolution effects are influencing the data. The difference spectrum, shown earlier in Fig. 3, is in fact a mapping of the resolution-smearing function of the detection system convolved with the edge discontinuity. A criticism of this technique is that in using the standard simple net-peak area algorithms in the difference spectrum to arrive at an assay result, relatively little of the collected information is used. Three sums are generated to reduce the data to an assay, but since these are sums of differences, effectively only six data points are used. (Actually, a few more points than this are used because the data is smoothed using a five-point window before differences are taken. However, the number of data points used in the calculation is still much less than the number collected).

Another technique that has been proposed to reduce the L_{III} -edge data involves fitting the data after the log of the transmission has been computed. Regions are selected on both sides of the L_{III} edge. They are chosen far enough away from the edge to avoid resolution effects, but close enough to still be operating on the relatively flat portion of the incoming beam spectrum. These regions are then fit with a linear least-squares routine, and the fits are extrapolated to the edge to yield the transmission values to be used to deduce an assay result. This linear fit edge-extrapolation method has the advantage that it uses more of the collected data and minimizes the statistical random uncertainty.

The edge-extrapolation analysis method has been tested using data obtained with a compact L_{III} -edge densitometer.⁵ These results display a nonlinear characteristic similar to that observed in the NBL data, which was analyzed using the difference method. However, the edge-extrapolation data show a smaller statistical scatter than the difference data. For solutions containing small uranium concentrations, the edge-extrapolation results also display a smaller deviation from the ideal linear calibration than the difference results.

The spectra that were calculated to study the effects of small-angle scattering, system resolution, and solution-matrix makeup were reanalyzed using the edge-extrapolation analysis method. Figure 15 shows the effect of small-angle scattering on calculational data analyzed using the edge-extrapolation technique. Figure 16 compares the results reduced using the difference technique to those obtained with the edge-extrapolation method. It is interesting that while there is still an increase in the nonlinearity as the degree

Fig. 15. The calculated normalized assay response to variations in small-angle scattering for the edge-extrapolation technique.

Fig. 16. A comparison of the calculated normalized assay response to variations in small-angle scattering for the difference analysis technique and the edge-extrapolation technique.

of scattering increases (Q/d decreases), the increase in the nonlinearity is smaller for the extrapolation technique than for the difference technique. It is also interesting that the nonlinear behavior of the assay response for the extrapolation technique is opposite that for the difference technique. For example, with the extrapolation method at low concentrations, as the scattering increases the assay decreases, whereas for the difference technique, the assay increases as scattering increases. For high concentrations the opposite is true.

Figures 17 and 18 show the results of calculations in which the system resolution was varied and the edge-extrapolation analysis technique was employed. Again, the nonlinearity becomes more severe as the system resolution degrades, but the effect on the assay for the extrapolation method is smaller than for the difference technique.

Figure 19 shows the effect on the assay of variations in the molarity of solvent matrix when the extrapolation technique is used. The instrument is calibrated using a 50 g/l uranium - 3M HNO_3 solution. A 3M HNO_3 solution is used as a reference but the unknown solutions have solvent matrices which vary from 1M HNO_3 to 6M HNO_3 . In Fig. 19, the effects of these variations are approximately 10 times smaller than in Fig. 8, where the calculations were done using the difference technique. As with the scattering computations presented above, the behavior of the assays obtained using the edge extrapolation analysis method is opposite from that observed for the difference technique.

IV. CONCLUSIONS AND RECOMMENDATIONS

On the basis of the calculations performed during this study, the nonlinear characteristic for the L_{III} -edge densitometer's response appears to be the result of a combination of the effects of small-angle scattering and the detection system's response function. The shape and extent of this nonlinearity is influenced by many factors, including the densitometer's geometry, its instrumentation, and the analysis technique used to reduce the data. The calculations predict the shape of the nonlinearity very well. However, the magnitude of the nonlinearity at high concentrations is not predicted. Because of the simplifying assumptions used to model scattering in the densitometer

Fig. 17. The calculated normalized assay response to variations in the system resolution function for the edge-extrapolation technique.

Fig. 18. A comparison of the calculated normalized assay response to variations in the system resolution function for the difference analysis technique and the edge-extrapolation technique.

Fig. 19. The calculated effect of variations in the molarity of a nitric acid solvent matrix on the measurement of uranium solutions for a system calibrated for uranium - 3M HNO₃ solutions using the edge-extrapolation technique.

system, the model underestimates the amount of scattered intensity that reaches the densitometer's detector. This, in turn, results in an underestimate of the magnitude of the nonlinearity. A more complex scattering model is necessary to investigate this effect more quantitatively.

This study has demonstrated that the L_{III} -edge technique is quite insensitive to elemental contaminants in solution with the SNM to be measured, but it is sensitive to the molarity of the matrix solvent used in the solution. This sensitivity is most dramatic for low SNM concentrations. These results emphasize the need for using caution when interpreting the assay results for low-concentration solutions. Not only do the assays of low-concentration

solutions display a greater random error as a result of the poorer precision of the instrument in this range, but variations in the matrix solvent of the solutions relative to the reference solution may result in large measurement biases.

For mixed solutions, the calculations demonstrate that the L_{III} -edge densitometer's response is a function of the uranium-to-plutonium ratio of the solutions measured. To date, relatively few mixed solutions have been measured by the densitometer because of the difficulty in obtaining such solutions. The measurements that have been performed have hinted that such an effect exists but are not sufficient to verify its existence. It is hoped that future mixed-solution measurements will strive to demonstrate this dependence on the uranium/plutonium ratio so that the magnitude of its effect on the assay can be determined and the accuracy of the technique improved.

The calculations performed in this study have also shown that the assay results obtained with the L_{III} -edge densitometer may be improved by changing the technique used to reduce the data. The edge-extrapolation technique appears to be less sensitive to the effects of small-angle scattering and to resolution effects than is the differencing technique. The edge-extrapolation technique is also less sensitive to changes in the molarity of the solvent matrix. It is hoped that future experimental measurements will further evaluate the edge-extrapolation technique. In particular, future experimental measurements will compare the results obtained using both analysis techniques. Because the assay results calculated using these techniques behave in an opposite manner with respect to the effects of changing matrix solvent molarity and small-angle scattering, such a comparison may prove useful in identifying changes in these parameters.

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