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THE EFFECTS OF FISSION PRODUCTS ON DEMONSTRATED X-RAY AND GAMMA-RAY NDA TECHNIQUES APPLIED TO NUCLEAR MATERIALS ACCOUNTING IN REPROCESSING

by

P. A. Russo and S. T. Hsue

ABSTRACT

The application of demonstrated high-resolution techniques of energy-dispersive x-ray and gamma-ray spectroscopy to the assay of samples derived from plutonium reprocessing is reported. K-edge absorption densitometry with a continuum x-ray transmission source is shown to be effective for materials accounting of uranium and plutonium in fast-breeder reactor dissolver solutions. The L-edge technique with a continuum transmission source can be applied similarly beyond the first extraction. Fluorescence of K x rays with a continuum source for excitation shows some promise for uranium and plutonium materials accounting in light-water reactor dissolver samples. The success of this technique in these applications relies on the ability to improve the statistical precision for the assay of plutonium using a new technology of high-resolution gamma-ray spectroscopy at count rates an order of magnitude greater than those previously possible. Passive gamma-ray assay techniques for determination of plutonium isotopic fractions are shown to be ineffective for all but product materials in reprocessing.

I. INTRODUCTION

The presence of varying levels of fission products in spent fuel material reprocessed for recovery of plutonium has motivated studies to ascertain the capabilities of demonstrated nondestructive assay (NDA) techniques for plutonium assay in the presence of interfering gamma-ray backgrounds. The evaluated x-ray and gamma-ray NDA techniques include passive gamma-ray spectroscopy, L-edge absorption densitometry, K-edge absorption densitometry, and K x-ray fluorescence (K-XRF). All are high-resolution, energy-dispersive methods.

The following sections review the gamma-ray techniques used in these studies and summarize the results of measurements performed on samples representative of process solutions and oxide products of plutonium reprocessing. The interfering backgrounds caused by the fission products have been simulated, in some cases, with ^{137}Cs sources. The 662-keV gamma rays produced by this radioisotope are about midway in the energy spectrum of fission products (500 to 800 keV). Because the Si(Li) and germanium detectors used with the NDA instruments have low photopeak efficiencies at these energies, it is mainly the Compton continuum in the low-energy region that determines the effect of the fission product background on the assay capabilities.

The absolute fission product levels are highest in the fast-breeder reactor (FBR) dissolver solutions, but the uranium-to-plutonium ratio (∿3:1) is compatible with techniques designed to assay plutonium. The light-water reactor (LWR) dissolver solutions are lower in plutonium concentrations so that the lower absolute fission product levels are higher, relative to plutonium, than in the FBR solutions. The large uranium-to-plutonium ratios (100:1) greatly reduce the sensitivity of the plutonium assay. Separation factors for fission products are approximately 100:1 or better at the first extraction. For the plutonium products, fission product levels are typically less than 10 µCi/g plutonium in reprocessing plant streams; exceptions are the streams of conceptual processes that have been proposed as alternatives to traditional separations technology. These processes have been discussed in terms of (1) reduced separation factors for fission product extraction and (2) the addition of radioactive spikes (such as ^{60}Co, ^{95}Zr, ^{106}Ru, ^{110}Ag, ^{144}Ce, and ^{46}Sc) beyond the extraction stages to restore high levels of high-energy gamma-ray activity as a deterrent to diversion.

Table I gives the flow-sheet values for the special nuclear material (SNM) and fission product contents of reprocessing solutions at various stages in the process. Both LWR (Tokai, Barnwell) and FBR reprocessing are represented.

TABLE I

SOLUTION CONCENTRATIONS AT VARIOUS REPROCESSING PLANTS

Processing Stage	Process Solution Characteristics	Fuel Reprocessing Plant		
		Tokai[a]	FBR[b]	Barnwell[c]
Dissolver accountability tank	g U/ℓ	180	123	310
	g Pu/ℓ	2	35	3
	Ci/ℓ	340	2800	1000
	Ci/g Pu	170	80	330
After 1st FP extraction	g U/ℓ	55	69	100
	g Pu/ℓ	0.6	19	1
	Ci/ℓ	0.3	4.5	1-10
	Ci/g Pu	0.5	0.24	10
After FP & uranium extraction	g U/ℓ	0.8(3BP)	43(1AF)	10(1BP)
	g Pu/ℓ	1.9	12	5
	mCi/ℓ	3.4	288	46
	mCi/g Pu	1.8	24	9
	g U/ℓ		39(2AP)	
	g Pu/ℓ		19	
	mCi/ℓ		10	
	mCi/g Pu		0.53	
After plutonium purification	g U/ℓ	0	44	0
	g Pu/ℓ	15	22	60
	mCi/ℓ	0.24	0.22	Trace
	mCi/g Pu	0.016	0.01	
Product	g U/ℓ	0	269	0
	g Pu/ℓ	250	89	250
	mCi/ℓ	Trace	0.89	Trace
	mCi/g Pu		0.01	

[a] L. L. Lowry and R. H. Augustson, Comps., Tokai Advanced Safeguards Technology Exercise Task T-F: "Study of Selected Capabilities Needed to Apply DYMAC Principles to Safeguarding the Tokai Reprocessing Plant," Los Alamos Scientific Laboratory report LA-8070-MS (ISPO-74) (October 1979).

[b] "SK-B-200 Series Process Flow Diagrams, Revision 1," Bechtel National, Inc., San Francisco, California (June 29, 1979).

"CFRP-HEF Conceptual Design, Process Flow Diagrams, Revision A," Bechtel National Inc., San Francisco, California (April 21, 1980).

[c] E. A. Hakkila, D. D. Cobb, H. A. Dayem, R. J. Dietz, E. A. Kern, E. P. Schelonka, J. P. Shipley, D. B. Smith, R. H. Augustson, and J. W. Barnes, "Coordinated Safeguards for Materials Management in a Fuel Reprocessing Plant," Los Alamos Scientific Laboratory report LA-6881 (September 1977).

II. PASSIVE GAMMA-RAY SPECTROSCOPY

Passive gamma-ray assay techniques have been demonstrated for determination of the plutonium isotopic composition of solution and solid samples in reprocessing. The techniques have been applied to product solutions and oxide products where fission products are absent at detectable levels.

The possibility of extending these methods to the assay of samples that originate further upstream has been evaluated recently for a general assay technique that applies to plutonium samples of arbitrary size, geometry, age, and (chemical or isotopic) composition.[1,2] This technique obtains the complete plutonium isotopics distribution based on measured ratios of areas of closely spaced peaks in the passive gamma-ray spectrum. Table II gives the particular gamma-ray peak ratios used to determine the isotopic ratios from which the isotopic fractions are derived. For materials accounting applications, the timely NDA results for plutonium isotopics have been coupled with calorimetry measurements of the same sample to give total plutonium determinations. The introduction of fission products (by simulation with ^{137}Cs) has the greatest effect on the results for the even isotopes (^{238}Pu and ^{240}Pu) because the passive gamma-ray peaks (at 153 and 160 keV) used for determination

TABLE II

GAMMA-RAY PEAK RATIOS USED TO DETERMINE ISOTOPIC RATIOS

Isotope 1/Isotope 2	Peak 1/Peak 2 (keV)
^{238}Pu/^{241}Pu	152.7/148.6
^{239}Pu/^{241}Pu	345.0/332.4
	203.5/208.0
^{240}Pu/^{241}Pu	160.3/164.6
^{241}Am/^{239}Pu	125.3/129.3
	169.6/171.3

of these isotopic fractions are weak relative to those for ^{239}Pu and ^{241}Pu. Furthermore, the fractions of the even isotopes (^{240}Pu, in particular) are required to convert the calorimetry results to plutonium mass. Figure 1 shows the relative precision (arbitrary units) in the isotopic fractions for 15% ^{240}Pu material (solid lines) plotted vs equivalent fission product content (in μCi/g plutonium). The loss of precision for ^{240}Pu with increasing background activity is dramatic--more than twice that for low burnup samples (6% ^{240}Pu, dashed line in Fig. 1). Therefore, the passive assay techniques show an extremely low practical tolerance [$\sim$20 μCi/g plutonium for higher burn-up samples; $\sim$10 μCi/g plutonium for samples with <15% ^{240}Pu (Ref. 2)] to fission product backgrounds. These limits of practical tolerance refer particularly to the 125- to 375-keV assay region used for plutonium isotopics. Use of the higher energy (600-keV) region would, in principle, be less tolerant of fission products because of direct interferences. The more intense plutonium

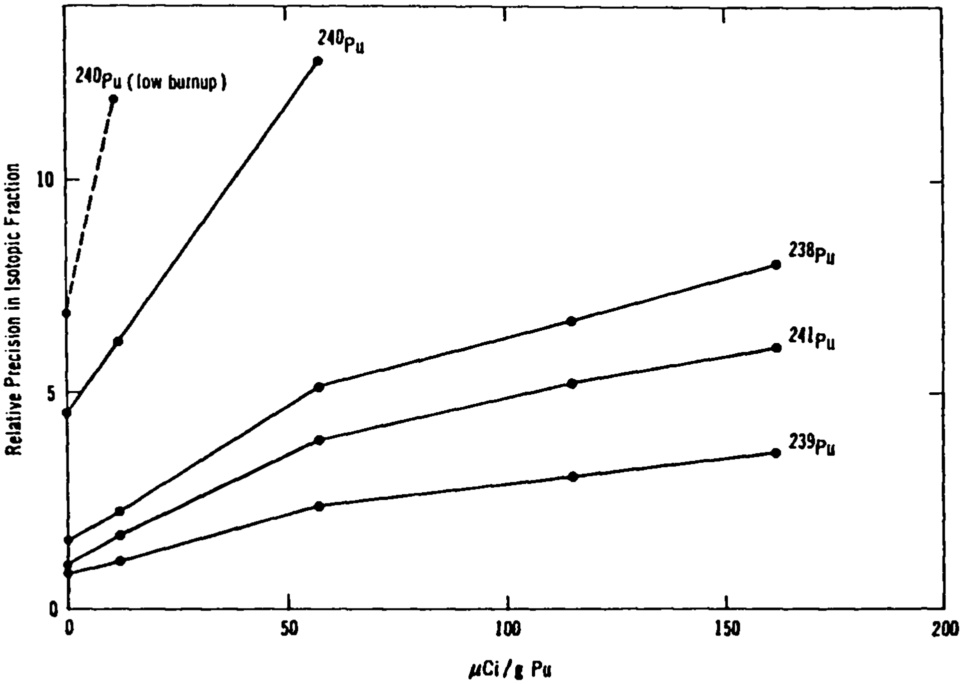

Fig. 1.
Relative precision (arbitrary units) in the isotopic fraction determined by passive gamma-ray assay plotted vs level of fission product activity (relative to plutonium). The solid lines correspond to data obtained using oxide samples of intermediate-burnup (15% ^{240}Pu) material. The dashed line corresponds to the results obtained with low-burnup (6% ^{240}Pu) samples.

gamma-ray peaks in the 40- to 51-keV region may also be less tolerant because of the higher Compton backgrounds at lower energies and because of the presence of high levels of americium at those stages in the process where fission products are present.

Other estimates[3] of tolerable levels of fission products for passive assay techniques have exceeded 200 µCi/g plutonium. Only the relatively intense gamma-ray peaks of the odd-mass plutonium isotopes have been used in these studies to determine the levels at which fission products can be tolerated. The passive assay results have not been applied, in these cases, to determination of the mass of plutonium using coupled results of neutron coincidence counting or calorimetry.

III. L-EDGE ABSORPTION DENSITOMETRY

Techniques of x-ray absorption-edge densitometry are equivalent to active gamma-ray assay techniques in that passive gamma-ray emissions from the sample are not used as the assay signature. Therefore, background gamma rates from fission products can be reduced by restrictive sample collimation combined with increased external source strength to achieve the optimum detector count rates. This approach was applied using the L-edge densitometer.[4,5]

The L-edge instrument uses a 50-kV x-ray generator, operated at 22 kV, as the external transmission source. The solution samples are positioned, typically, between the source and the Si(Li) detector. Figure 2 shows a cross section of the transmission path for this instrument. The distances from the x-ray tube to the sample and the sample to the detector are 12 and 9 cm, respectively. The original 3-mm-thick brass collimators on the detector and x-ray beam had 5-mm-diam holes. These hole diameters were reduced to 2.5 mm for the measurements described here. Two tungsten collimators 25 mm in length were inserted into the beam path inside the alignment frame. The downstream and upstream collimator hole diameters were 3 and 1.5 mm, respectively. A brass collimator with a 1-mm-diam hole was positioned immediately downstream from the sample to prevent the x-ray beam from striking the tungsten.

To simulate fission products, the sample cell was filled with 0.5N nitric acid solutions with various concentrations of ^{137}Cs. The SNM samples were inserted, as metal foils, into the slot labeled "calibration foil" in Fig. 2. Because the K absorption edge of zirconium is at nearly the same energy as the

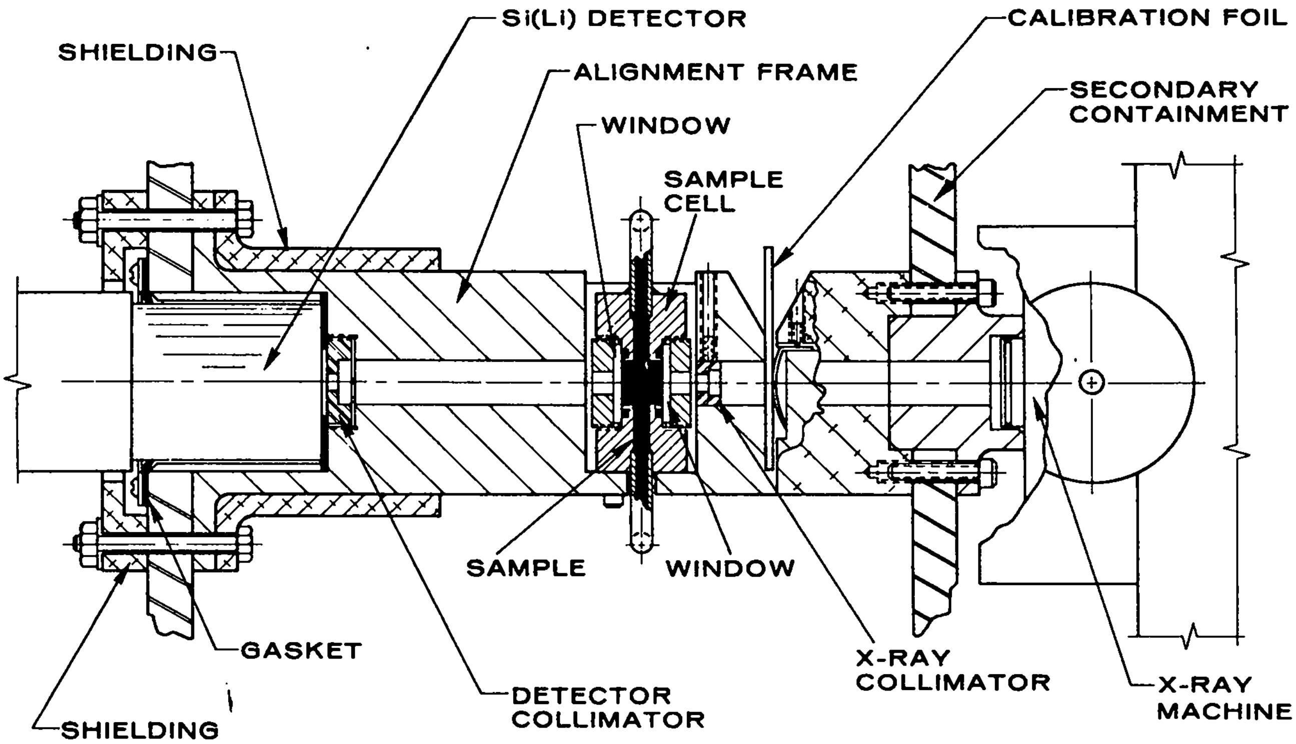

Fig. 2.
Cross section of L-edge densitometer transmission path before collimation for measurement of uranium and plutonium concentrations with gamma-spiked solutions.

plutonium L_{III} edge, zirconium was used to simulate plutonium for these measurements. Foils of uranium and zirconium (mixed) and of zirconium alone were measured with and without the simulated fission product interferences. The software for data acquisition and analysis was modified to accumulate and store a background spectrum, which was later subtracted from the transmission spectrum. The background caused by the 662-keV gamma rays of ^{137}Cs is flat in the low-energy region of the L_{III} absorption edges. However, the analysis uses the ratio of transmissions above and below the absorption edge of the element assayed. Therefore, failure to correct for significant backgrounds causes a bias in the measurement results.

Table III gives the ^{137}Cs activity, relative to equivalent plutonium, for the five solution samples. The beam-off count rates in the Si(Li) detector are also listed. It was determined (by varying the collimator size) that 2 kHz of the background rate with the most radioactive ^{137}Cs solution was due to scattered radiation rather than to gamma rays transmitted through the

TABLE III

L-EDGE ASSAY RESULTS FOR URANIUM AND PLUTONIUM IN THE PRESENCE OF FISSION PRODUCTS

Relative Fission Product Activity (mCi/g Pu)	Beam-Off Rate (Hz)	Assay Result (g/ℓ ± 1σ)		
		Mixed		Separate
		Uranium	Plutonium	Plutonium
0	1	41.27 ± 0.11	19.97 ± 0.21	10.25 ± 0.05
2.5	17			
10	68			
60	410	41.29 ± 0.11	10.02 ± 0.21	10.26 ± 0.06
370	2600[a]	41.24 ± 0.15	10.05 ± 0.24	10.21 ± 0.06

[a] Caused primarily by scattered radiation. (Refer to text.)

collimator hole. The scattering originates in the aluminum alignment frame (Fig. 2), which is an adequate shield for 20-keV x rays but not for the 662-keV gamma rays of ^{137}Cs. Therefore, backgrounds can be reduced to well below 1 kHz by introduction of adequate shielding. (This is accomplished by replacing the aluminum alignment shield with tungsten.) The densitometry measurements were performed with beam-on count rates of 15 kHz.

The results[6,7] shown in Table III are the averages of ∿10 runs of 1000 s each performed on each sample. The foils were repositioned between measurements with and without ^{137}Cs in the sample cell. This can introduce small differences (0.1%) in the results. The errors given are the standard deviations obtained from the repeated measurements. The standard deviations in the mean results are approximately one-third of the stated errors. Therefore, the results for each sample agree, within statistical errors, over the indicated range of fission product background activities.

Background rates up to 10 kHz can be tolerated with only small losses in assay precision. Therefore, by replacing the aluminum alignment frame with a heavy metal, the maximum tolerable fission product level increases to several curies per gram of plutonium. This exceeds the maximum levels beyond the first extraction. Because the L-edge technique is suitable for simultaneous assay assay of both uranium and plutonium concentrations (in mixed solutions), the L-edge method, which has been demonstrated on-line with flowing solution streams,[8] shows great promise for far-upstream applications in reprocessing.

IV. K-EDGE ABSORPTION DENSITOMETRY

The use of K-edge absorption densitometry for assay of LWR and FBR dissolver solutions has recently been demonstrated.[9] The K-edge technique uses a 150-kV x-ray generator, operated at 145 kV, as the external transmission source. The equipment was designed to be optimized for the K-edge assay of dissolver samples. Therefore, the detector is a planar intrinsic germanium crystal, and the transmission path is long (114 cm from x-ray tube to detector) to produce a narrow well-collimated beam (3 mm in diameter). The detector is positioned 24 cm from the sample. Tungsten collimators 3 mm in diameter, along with tungsten shielding between the sample and detector, reduce the background count rates to negligible (1%) levels relative to 15 kHz, which is the detector count rate with the x-ray beam on.

The K-edge assay precision for uranium and plutonium in dissolver feed solutions is given in Table IV. These results correspond to count times of

TABLE IV

PRECISION OF K-EDGE ASSAY RESULTS FOR URANIUM AND PLUTONIUM IN DISSOLVER FEED SOLUTIONS

Feed Solution	Assay Precision (%,1σ)	
	Uranium	Plutonium
LWR (150-g U/ℓ, 3-g Pu/ℓ)	0.3	NA
FBR (150-g U/ℓ, 50-g Pu/ℓ)	0.3	0.9

1000 s and are essentially identical to those obtained for samples that do not have high levels of gamma-ray activity.

These K-edge results demonstrate that nuclear materials accounting of FBR dissolver feed solutions can be accomplished in a timely manner by a technique that, like the L-edge methods, can be used on-line for measurements of flowing streams. The disadvantages of the larger size, higher power, and greater cost of the x-ray generator required for these measurements are greatly outweighed by the savings in time and sample handling of the highly radioactive feed solutions. The high uranium-to-plutonium ratio in the LWR dissolver precludes use of this method for plutonium assay of the LWR feed material.

V. K X-RAY FLUORESCENCE

A recent demonstration of the use of K-XRF for measuring dissolver solutions that resemble LWR dissolver solutions has shown this technique to be useful, to a limited extent, in determination of the ratio of uranium to plutonium.[10] A 250-kV x-ray generator was operated at 180 kV for uranium and plutonium K-electron excitation. The x-ray tube was positioned 45 mm from the 0.6-cm-thick sample cell. The fluoresced x rays were detected at 90° to the incident beam. The planar intrinsic germanium detector was 15 cm from the sample. The sample collimation diameter was 6.35 mm.

One of the solution samples originated from advanced gas reactor spent fuel. The fission product levels in this sample were ∿100 Ci/ℓ with a uranium-to-plutonium ratio of ∿140:1; the plutonium concentration was ∿3 g/ℓ. Therefore, this sample was reasonably representative of LWR spent fuel in terms of SNM content, but the fission product levels were lower by factors of 3 to 10.

The $K\alpha_1$ peak of plutonium at 103.7 keV and the $K\alpha_2$ peak of uranium at 94.7 keV were used to obtain the assay result. Potential interferences from fission product gamma rays were safely within the resolution capabilities of the detector. Other passive interferences were negligible. The contribution to the uranium K_{α_2} peak caused by internal excitation resulting from the high radiation field was 7% of that excited by the x-ray generator. The corresponding contribution to the plutonium K_{α_1} peak could not be estimated because of the high backgrounds relative to the peak. The data were obtained at detector count rates of ∿20 kHz, corresponding to a data storage rate of 15 kHz.

Approximately 20% of this rate was due to passive background from the sample.

The linearity of the measurement result (that is, the relative intensity of the plutonium K_{α_1} peak to the uranium K_{α_2} peak) in the uranium-to-plutonium ratio was demonstrated within 10% with test solutions (from which fission products were absent) containing 250 g/ℓ uranium with uranium-to-plutonium ratios varying from 60:1 to 250:1. Although the assay peaks are closely spaced, large deviations from this linearity can be expected if the total SNM concentration varies substantially.

The precisions in the measured ratios of the uranium K_{α_2} and plutonium K_{α_1} peak intensities deduced from the statistics obtained in 2000-s count periods are shown in Table V for four samples, one of which is the dissolved spent fuel sample. These results represent the optimum precision in the measured ratios of uranium to plutonium that can be expected in 2000-s count periods if there are no contributions, other than statistical errors, to the random error. Furthermore, the 7% relative contribution from internal excitation of uranium K_{α_2} x rays must also be subtracted using data from a separate passive count if this relative contribution is found to vary substantially.

Reference 10 reveals the intention to upgrade the K-XRF counting system to allow an order-of-magnitude increase in data storage rates. This upgrade would improve the statistical precision by as much as a factor of 3. It would also enable reduction of the relative (internal and passive) background because

TABLE V

PRECISION IN RATIO OF URANIUM TO PLUTONIUM MEASURED BY K-XRF

Plutonium Concentration (g/ℓ)	Uranium-to-Plutonium Ratio	Fission Product Level (Ci/g Pu)	Precision (1σ) in Measured Intensity Ratio (%)
1.0	250	0	6
2.5	100	0	3
4.0	62.5	0	2
∿3	∿140	∿30	6

the x-ray tube could be moved closer to the sample. The success of this upgrade requires that no significant losses in detector resolution accompany the increased count-rate capabilities because this would result in an unresolved interference between a fission product gamma-ray line (at 105.3 keV, from ^{154}Eu) and the plutonium K_{α_1} assay peak.

Reference 10 also reports a proposal to combine the capabilities of K-edge absorption densitometry (for determination of uranium concentration) and K-XRF (for determination of uranium-to-plutonium ratio) into a single, two-detector instrument. This proposed instrument would use two detectors and a single x-ray generator and sample cell to perform the two assays in sequence. The two results combined would give both the uranium and plutonium concentrations.

Because the limit to the precision achievable for plutonium concentration is clearly in the K-XRF measurement, it is desirable, for simplification of the instrumentation, to obtain the absolute result for uranium concentration using the K-XRF technique. Furthermore, because the total SNM concentration may vary enough to destroy the linearity between the measured intensity ratio and the uranium-to-plutonium ratio, there is also a need to determine and correct for the effects of self-attenuation of the fluoresced x rays. Such a correction would enable determination of the absolute uranium concentration. The following paragraphs discuss a demonstrated method that might be used to accomplish the corrections for self-attenuation and thus give the K-XRF technique the added capability of providing uranium and plutonium concentration, regardless of dissolver sample composition and concentration.

The use of an external transmission foil that becomes fluoresced simultaneously with the sample has been described in previous reports[11,12] of L-XRF methods. The methods are illustrated in Fig. 3. A ^{109}Cd annular source excites the L electrons of uranium in uranium solution samples and of thorium in the thorium transmission foil. The observed intensities of the 13.6-keV uranium L_α x rays required corrections for self-attenuation of the exciting and fluoresced radiation because the concentration range for these 1.3-cm-thick samples varied between 1 and 20 g/ℓ. The self-attenuation correction factors were obtained from the measured transmissions (ratios of intensities with and without the solution sample) of the thorium L_α peak and of one of the thorium L_β peaks (these peaks bracket the uranium L_α peak). The uranium L_α intensities, normalized by uranium concentration, are plotted as solid data points in

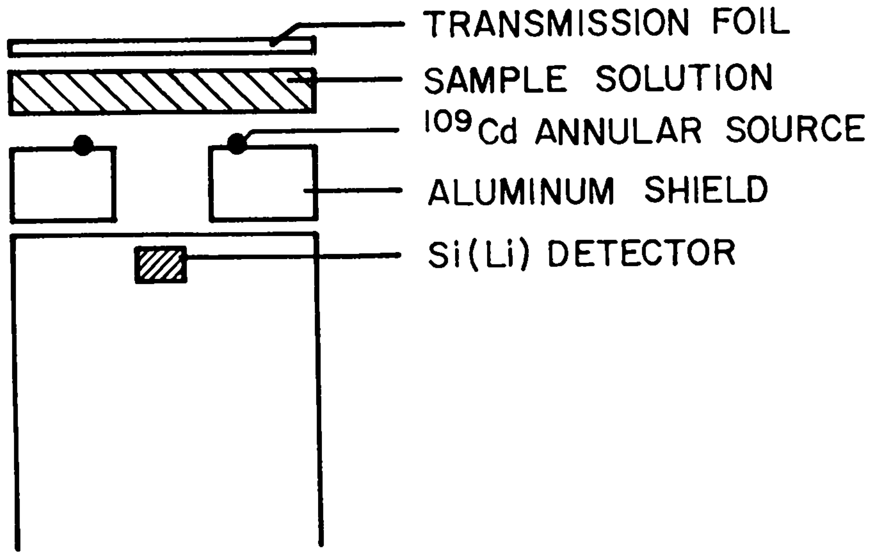

Fig. 3.
Experimental arrangement for L-XRF performed with a transmission foil for correction of exciting and fluoresced x-ray intensities for sample attenuation.

Fig. 4. The effects of sample self-attenuation result in substantial deviations from the horizontal line for uranium concentrations greater than 2 g/ℓ. The open data points correspond to the normalized uranium L_α intensities corrected for sample self-attenuation using the measured transmissions of the L_α and L_β peaks of thorium. These results demonstrate the ability to use empirical results to correct for the differential attenuation effects over a substantial range of SNM concentration.

Similar K-XRF measurements of uranium solutions with a thorium transmission foil[13] have demonstrated that precisions of better than 0.5% (1σ) for assay of uranium concentration in 50-g/ℓ solutions are achieved in 1000-s counts. These measurements used a ^{57}Co exciting source. The uranium K_{α_1} intensity was corrected for sample self-attenuation using the thorium 104.8- and 105.6-keV (K_{β_3} and K_{β_1}) peaks.

The feasibility of measuring the uranium-to-plutonium ratios of LWR dissolver solutions by K-XRF with an x-ray generator as an exciting source has been demonstrated. Although these results are currently precise to only 5 to 10%, the investigation of techniques to improve the statistical precision by a factor of 3 is under way. If such improvements are accomplished, it would be

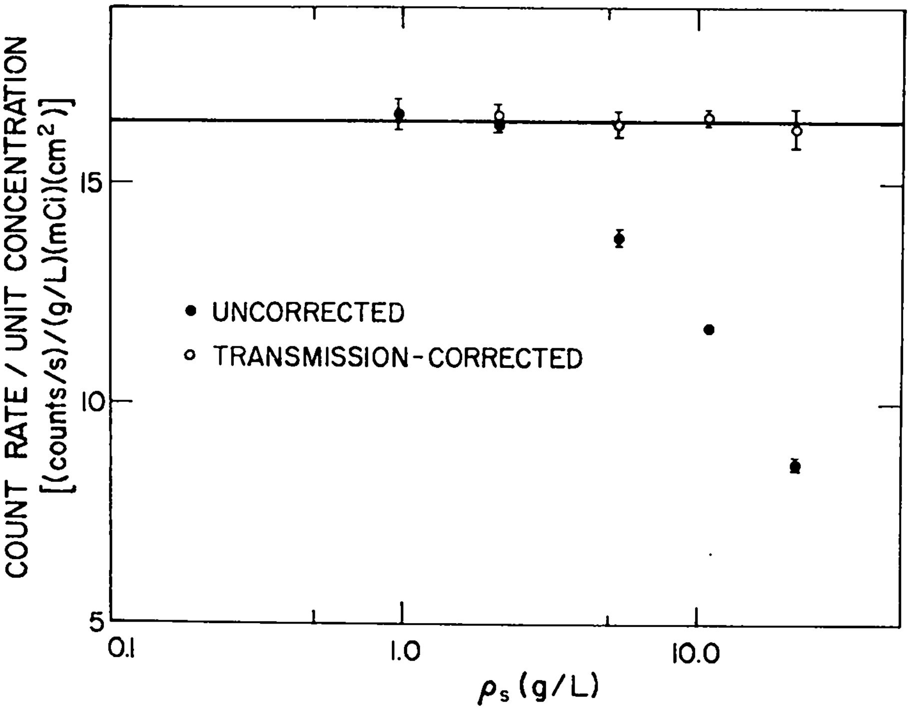

Fig. 4.
Plot of the intensity of the fluoresced uranium L_α peak (divided by uranium concentration) vs concentration of the uranium sample. The solid data points are the normalized intensities with no correction for sample self-attenuation. The open points are corrected for sample self-attenuation using the transmissions of the fluoresced thorium L_α and L_β lines.

desirable to modify these techniques to incorporate the capability to correct for sample attenuation of the exciting and fluoresced x-rays so that the single K-XRF technique would provide all the information (namely, uranium and plutonium concentrations) required for materials accounting in LWR dissolver feed.

VI. CONCLUSIONS

The following conclusions can be stated regarding the application of demonstrated NDA techniques (energy-dispersive, high-resolution gamma-ray spectrometry) to the assay of samples with high levels of fission products.

(1) Passive gamma-ray assay of plutonium is highly sensitive to the continuum backgrounds produced by high-energy fission product gamma rays. Complete determination of the plutonium isotopic fractions requires relative fission product levels that do not exceed 10 to 20 μCi/g plutonium (for low- and high-burnup materials, respectively) so that the isotopics information can be useful for materials accounting. The fission product levels of product streams and oxide products of conventional reprocessing meet these criteria.

(2) The use of L-edge densitometry for materials accounting of uranium and/or plutonium is practical for solutions where fission product levels do not exceed 5 Ci/g plutonium. Thus, the technique is suitable for solutions beyond the first dissolver. Note that precise L-edge assay results cannot be achieved for SNM concentrations below 1 g/ℓ and that assay of plutonium is also not practical if the uranium-to-plutonium ratio exceeds 10.[7]

(3) The capability of K-edge densitometry, performed with an x-ray generator as a transmission source, has been demonstrated for accounting of uranium and plutonium in FBR dissolver solutions. In LWR dissolver solutions, the technique can be applied to the assay of uranium but not plutonium (because of the unfavorable ratio of uranium to plutonium).

(4) Although K-XRF shows some potential for measurement of uranium-to-plutonium ratios in LWR dissolver feed solutions, the proposed improvements in fast-counting technology must be demonstrated before the measurement results can be considered sufficiently precise for purposes of materials accounting. A successful demonstration of this improved technology, and the added capability of performing absolute concentration measurements using a modified approach to the K-XRF assay technique, would give this technique the precision required for uranium and plutonium accounting in LWR dissolver solutions.

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