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Application of Curium Measurements for Safeguarding at Reprocessing Plants

Study 1: High-Level Liquid Waste

and

Study 2: Spent Fuel Assemblies and Leached Hulls

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ABSTRACT

In large-scale reprocessing plants for spent fuel assemblies, the quantity of plutonium in the waste streams each year is large enough to be important for nuclear safeguards. The wastes are drums of leached hulls and cylinders of vitrified high-level liquid waste. The plutonium amounts in these wastes cannot be measured directly by a nondestructive assay (NDA) technique because the gamma rays emitted by plutonium are obscured by gamma rays from fission products, and the neutrons from spontaneous fissions are obscured by those from curium. The most practical NDA signal from the waste is the neutron emission from curium. A diversion of waste for its plutonium would also take a detectable amount of curium, so if the amount of curium in a waste stream is reduced, it can be inferred that there is also a reduced amount of plutonium.

This report studies the feasibility of tracking the curium through a reprocessing plant with neutron measurements at key locations: spent fuel assemblies prior to shearing, the accountability tank after dissolution, drums of leached hulls after dissolution, and canisters of vitrified high-level waste after separation. Existing pertinent measurement techniques are reviewed, improvements are suggested, and new measurements are proposed. We integrate these curium measurements into a safeguards system.

I. INTRODUCTION

A. Overview

International Atomic Energy Agency (IAEA) safeguards have been applied for many years to the locations in reprocessing plants with high concentrations of plutonium (such as the spent fuel assemblies, the accountability tank, and the separated plutonium). The throughputs of new and proposed reprocessing plants have grown to the point where the annual mass of plutonium in the waste streams has significance, although recovery would required a determined effort. For example, even a highly efficient plant that processes 800 t of U per yr and recovers 54.5 t of plutonium per year may still release 2 to 4 kg of plutonium per year in 400 drums of leached hulls and 5 to 6 kg of plutonium per year in 800 canisters of vitrified high-level liquid waste. The studies reported here deal with safeguarding these waste streams through nondestructive assay (NDA) techniques to ensure that the plutonium flowing through them is in the expected amounts and that the streams are not used for diversion paths.

The extremely high emission rates of gamma rays from fission products and neutrons from curium make it impossible to perform NDA measurements for plutonium directly. The heterogeneity and the nature of the waste materials make it impossible to apply destructive analysis methods. So it is proposed here that the neutrons emitted by curium be measured at key points for one or both of these purposes:

(1) check for a balance of curium at pertinent process points and in waste streams, implying a proper amount of plutonium in the waste stream; and
(2) estimate the plutonium amounts in the waste containers by combining curium measurements with curium-to-plutonium ratios from destructive analyses of samples, where practical.

Curium enters the plant through the spent fuel assemblies and leaves the plant only through the waste streams. The key measurement points include the spent fuel assemblies before shearing, the leached hulls drums, and the canisters of vitrified high-level liquid waste. By also measuring the curium masses in the accountability tank, plant operators and inspectors close the curium balance for the plant's head end.

B. Process Models

1. Head-End

The basics of the head-end of a reprocessing plant are shown very simplistically in Fig. 1. A spent fuel assembly is sheared into many short sections, which are guided into a dissolver tank. Almost all the spent fuel is dissolved and temporarily stored in the accumulator tanks (not shown) and eventually passes through the accountability tank. The empty fuel pin sections (leached hulls) and assembly end plates are transferred to a drum for disposal as waste, but they still hold small quantities of fissile materials on their surfaces. Many assemblies are processed together as a batch, so the dissolver solutions and the drums contain portions of many different assemblies. (In the existing La Hague plants and the proposed Rokkasho-Mura plant the dissolver tank has a slowly revolving wheel, half submerged in the acid, with internal segments holding batches of hulls. Baskets are replaced by the segments and the leached hulls simply fall into a drum as they approach the high point in the wheel. There is no conceptual difference with the scheme shown in Fig. 1 that affects this report.)

A model of the material flow in the head-end region is shown in Fig. 2. A reactor's core discharge is treated as a completed batch completely isolated from other batches; tanks and process lines are cleaned between batches to prevent any mixing of materials between batches. The dissolver solutions are stored in buffer tanks (not shown) and eventually go into the accountability tank in sub-batches. Following accountability measurements, the contents of the accountability tank are transferred out of the head-end and into the plutonium and uranium separations facilities (which is outside the scope of this study). The leached hulls are rinsed (not

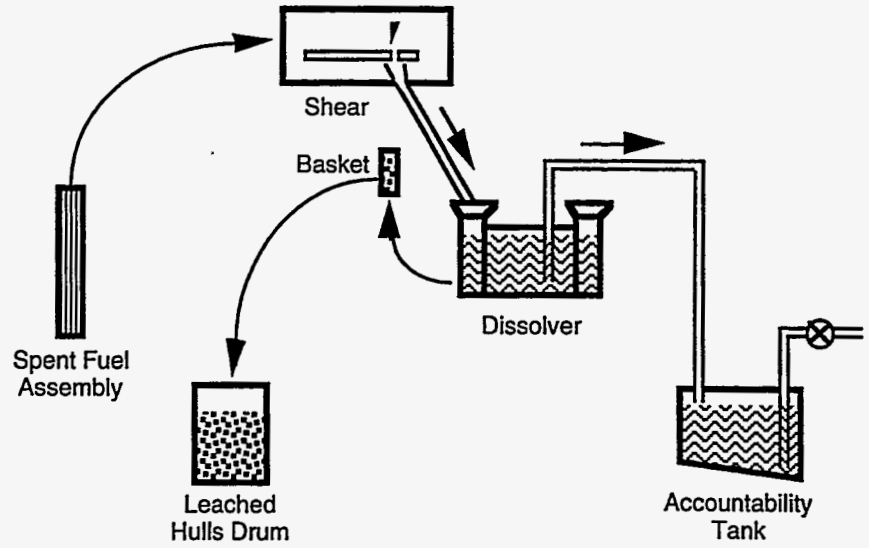

Fig. 1. The elements of a reprocessing plant's head-end that are involved in this study are shown in a schematic fashion. Spent fuel assemblies are cut into short segments in the shear and fall into a basket in the dissolver tank. The dissolver solution containing the fissile materials, the curium, and other actinides is transferred to an accountability tank. The leached hulls with residues of these same materials are accumulated in a drum as waste.

shown) before being placed in a waste drum and the effluent is returned to the dissolver tank to recover the plutonium in solution. The drums of leached hulls leave the head-end as waste.

A safeguards scheme for a plant's head-end based on curium measurements has three key measurement points: the curium entering through spent-fuel assemblies, the curium leaving through the accountability tank, and the curium leaving through the leached hulls. Measurement experiences and development expectations are discussed for these three items and then brought together to form a system of head-end measurements based on ^{244}Cm.

In principle the ^{244}Cm masses at the three locations could be entered into a ledger and accountancy techniques applied to check that no ^{244}Cm and associated plutonium are missing. However, the small ^{244}Cm values in the leached-hulls drums of Fig. 2 show that such a scheme is impractical. The difference between the curium masses measured on the assemblies and the tank cannot be sufficiently precise for a meaningful comparison with the curium measured in the drums. This simple but insufficient accountancy scheme will not be considered further in this report.

2. High-Level Liquid Waste

Another greatly simplified diagram is presented in Fig. 3 for the vitrification of the high-level liquid waste. The slurry from the separations facilities is chemically conditioned for vitrification and then calcined before joining a glass additive in a melter. The molten glass is poured into a canister. This process is likely to be done in batches, with two batches filling a canister.

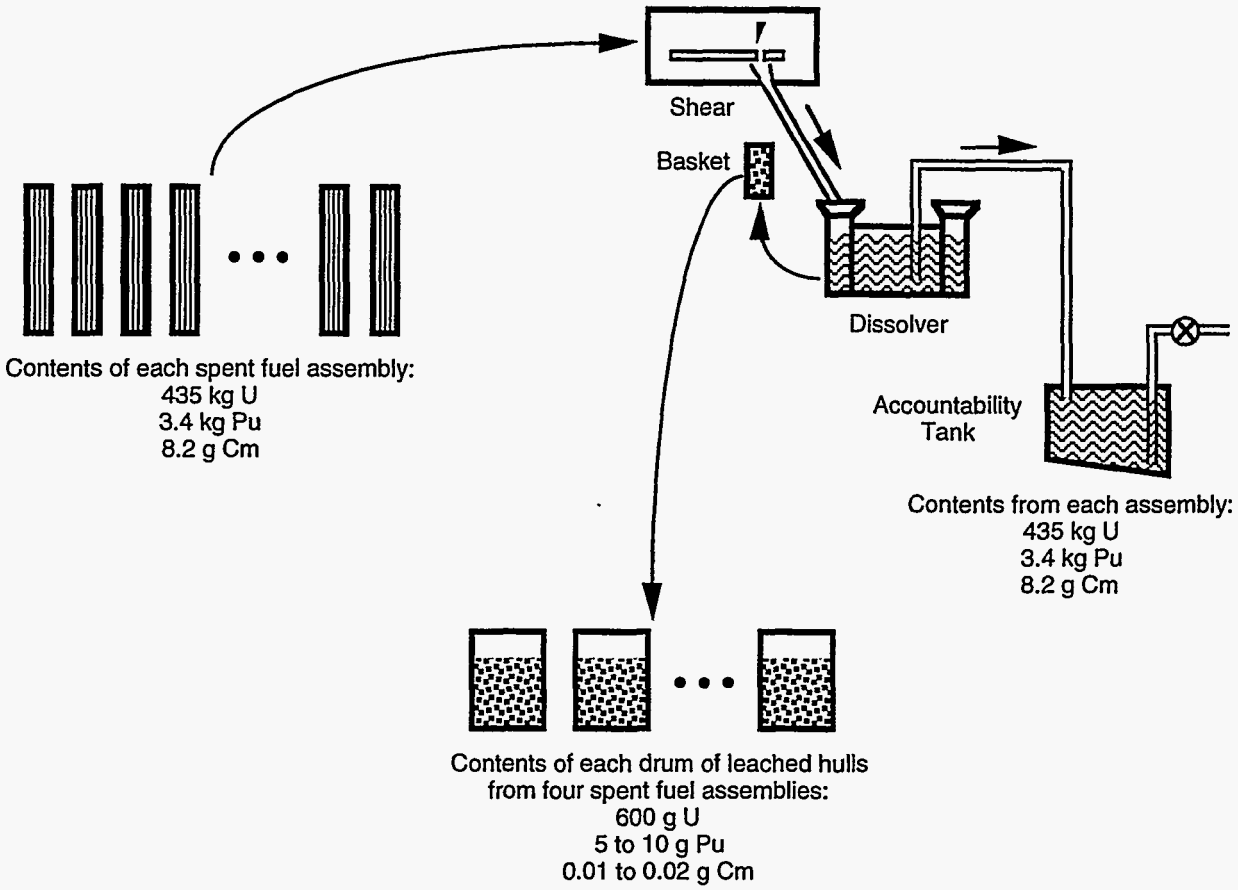

Fig. 2. A simplified model of the material flow through a plant's head-end is shown. Spent fuel assemblies are the only input containing fissile materials to the head-end; many assemblies are treated as a batch. The dissolver solution rich in uranium, plutonium, and curium passes through the accountability tank on its way to the separations area. The batch's leached hulls with relatively low amounts of these elements leave the head-end as waste in large drums.

The masses of uranium, plutonium, and curium are reference values for pressurized water reactor (PWR) assemblies with 33 GWd/tU and 3 years of cooling. The ^{235}U fraction of all the uranium at this moment is 1%. The plutonium is taken to be 24% ^{240}Pu and the curium is all ^{244}Cm. Any significant holdup in the shear or piping is swept into the accountability tank as part of the batch.

After filling, the canisters are cooled, welded shut, and then placed in an interim storage vault. They are highly radioactive from the fission products and curium and contain waste quantities of plutonium and uranium.

Curium measurements here could establish a balance between the curium entering the vitrification facility and the curium leaving in the canisters. The curium amounts can be converted into plutonium masses from a measurement of the plutonium-to-curium ratio before vitrification.

C. Organization of This Report

After this introduction each of the three measurement points of the head-end (Study 2) are discussed individually and then brought together into an integrated analysis. Two measurement points for the high-level waste (Study 1) are then discussed as a safeguards NDA system for the vitrification section of a reprocessing plant.

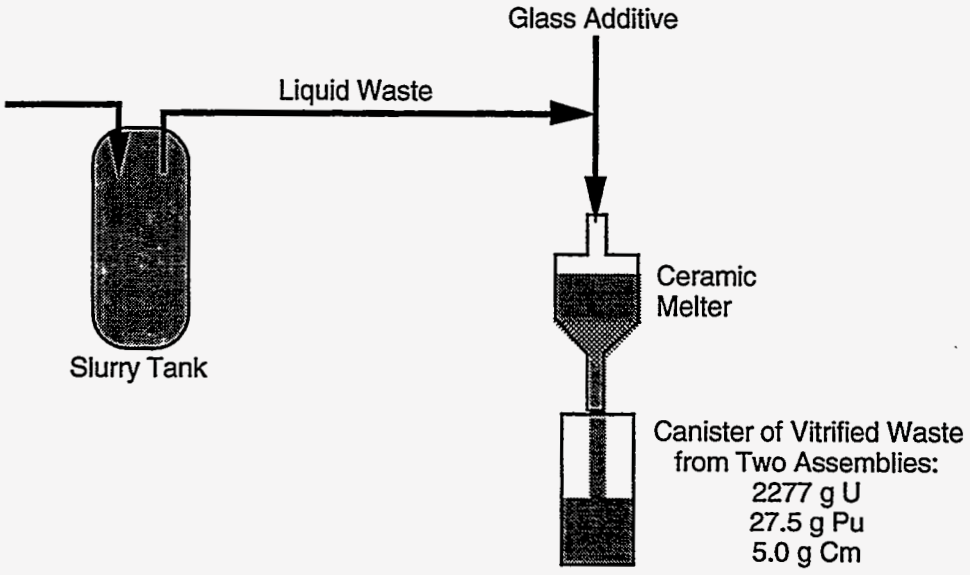

Fig. 3. The elements of a vitrification facility pertinent to this study are indicated. Many other tanks and a calciner are not shown. The slurry is transferred from the separations facility and after conditioning (not shown) is mixed with glass, melted, and poured into canisters, losing some volatile components in the process. Each canister holds the waste from two PWR assemblies.

II. HEAD-END OF A PLANT

A. Spent Fuel Assemblies

The inputs to a reprocessing plant are light-water-reactor spent fuel assemblies with cooling times greater than three years. The dominant source term of neutrons is spontaneous fission from ^{244}Cm. However, additional neutrons will be produced by the multiplication rocess from induced fissions inside the fuel assemblies. This multiplication is significantly increased if the PWR assemblies are underwater.

We performed Monte Carlo calculations to evaluate an NDA system for underwater neutron multiplicity measurements of a PWR fuel assembly. For multiplicity measurements the detection efficiency must be high enough to count a reasonable number of triple events (three neutrons counted during the electronics gate width). The ^{244}Cm spontaneous fission neutrons will be the dominant neutron driving term and the ^{235}U and plutonium fissile content will determine the amount of neutron multiplication. Neutron coincidence counting can be used to determine the ^{244}Cm mass and the multiplication, and for a particular PWR fuel configuration the fissile content in the fuel can be measured. This is a form of self-interrogation in which the interrogation source is ^{244}Cm.

The curium-to-plutonium ratio grows as the irradiation in a reactor increases, so the center of a fuel assembly has a higher ratio than the ends. To properly use an approach based on ^{244}Cm, the entire active length of an assembly must be scanned through an NDA system to obtain the total ^{244}Cm for the entire fuel assembly. This is especially crucial for boiling water reactor (BWR) assemblies where the burnup may vary widely along the assembly.

The spent fuel assemblies have one to three cycles of exposure history, where a single cycle normally generates* 15 GWd/tU for a period of about 300 days. After more than 3 years of cooling, the neutron emission rate from ^{242}Cm is negligible compared with that of ^{244}Cm, as are the emission rates from all other spontaneous fission neutron sources.

Figure 4 shows the relative sources of neutrons from a spent PWR fuel assembly with 30 GWd/tU as a function of cooling time. The dominant source of neutrons is ^{244}Cm and it is decaying with an 18.1-yr half life. Therefore the spontaneous fission of ^{244}Cm is the primary source of neutrons that we use in the present curium-based safeguards approach, although we shall see that neutrons from (α,n) reactions in the vitrified waste cannot be ignored. Table I gives the percentage of neutrons from the different isotopes in spent PWR fuel as a function of exposure.

Figure 5 shows the increase in neutron emission rate as a function of exposure, as calculated using the ORIGEN2 code. The neutron emission rate for a PWR MOX fuel

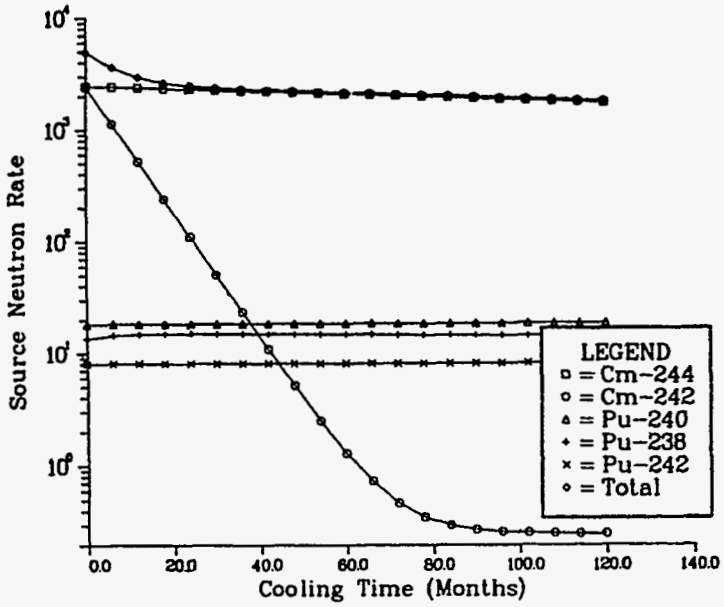

Fig. 4. The relative neutron emission rates from the major isotopes in a PWR spent fuel assembly are plotted here as a function of cooling time.[2] The assembly's exposure was 30 GWd/tU. The two curium isotopes easily dominate the plutonium isotopes initially, and after two years only the ^{244}Cm is important. This pattern holds for all exposures above 15 GWd/tU; at lower exposures ^{240}Pu cannot be ignored.

* A distinction is made between exposure as an atom percent of fissile material fissioned and exposure as thermal production per unit mass of fissionable fuel.[1] One atom percent burnup is approximately 9.6 GWd/tU of exposure, but this conversion varies with the ratio of uranium and plutonium fissions. Exposure in gigawatt-days per metric ton of initial uranium (GWd/tU) is used in this report.

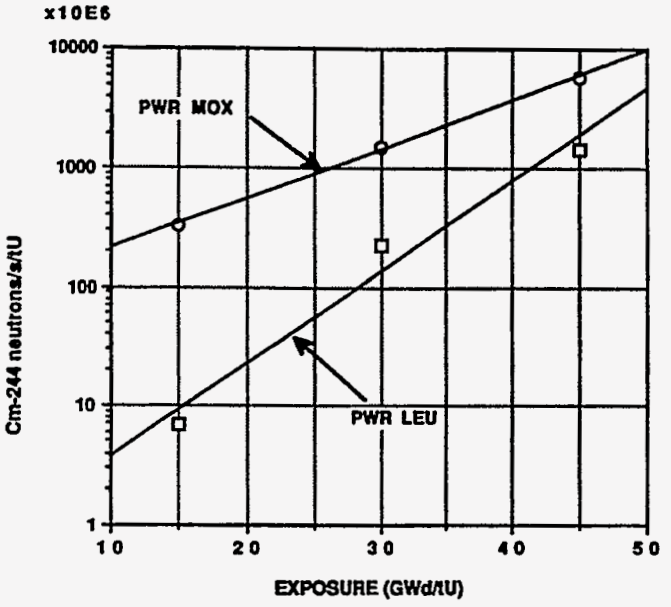

Fig. 5. The neutron emission rates from ^{244}Cm in PWR LEU and mixed oxide (MOX) fuels are shown as functions of exposure. The MOX fuel has higher rates because the plutonium in the fresh fuel enhances the production of ^{244}Cm through neutron captures in the fuel.

assembly is approximately an order of magnitude higher than the rate for a PWR low-enriched uranium (LEU) fuel assembly for similar irradiation conditions. However, the fission product inventories for the MOX and LEU fuels of the same exposure are approximately the same.

1. Specifications of Assembly Types

Some relevant nuclear properties of fuel assemblies from different reactor types are quite different and are in summarized Table II.

2. Measurement Experience

a. LWR Assemblies

Various instruments have been used to measure the neutron and gamma-ray emissions from LWR spent fuel assemblies while underwater.[1,4-11] They have served both safeguards[4,9-11] and criticality control purposes.[7,12-15] Only total neutrons and gross gamma rays have been counted in these applications. As illustrated in Fig. 3, the neutron emission rates from ^{242}Cm and ^{244}Cm easily dominate all other sources in LWR assemblies with more than 15 GWd/tU exposure and less than 3 years of cooling; when the cooling time is longer than 3 years only the emission from ^{244}Cm is significant.

Data analyses to determine exposures from neutron emissions usually apply correction factors for these parameters:

- initial ^{235}U enrichment,
- irradiation history,
- cooling time, and
- fraction of the total neutron emission rate originating from ^{244}Cm.

TABLE I

NEUTRON SOURCES IN LWR SPENT FUELS

After 5 years of cooling.
Taken from Ref. 3.

Neutron Source	Isotope	UOX[a] 20 GWd/tU	UOX[b] 45 GWd/tU	MOX[c] 50 GWd/tU
(α,n)	Total	6.6%	1.9%	1.3%
	^{238}Pu	2.3	0.8	0.4
	^{239}Pu	0.7	0.0	0.0
	^{240}Pu	0.8	0.1	0.0
	^{241}Am	2.1	0.2	0.1
	^{242}Cm	0.0	0.0	0.0
	^{244}Cm	0.7	0.8	0.8
Spontaneous Fission	Total	93.3%	98.1%	98.7%
	^{238}U	0.0	0.0	0.0
	^{238}Pu	0.4	0.0	0.1
	^{240}Pu	4.1	0.5	0.2
	^{242}Pu	0.9	0.2	0.1
	^{242}Cm	0.2	0.0	0.0
	^{244}Cm	87.6	96.8	97.6
	^{246}Cm	0.1	0.6	0.6
Total		100	100	100

[a] Initial uranium enrichment = 3.0 wt%.

[b] Initial uranium enrichment = 4.1 wt%.

[c] Initial fissile enrichment = 6 wt%. Recycled plutonium from an LWR with an exposure of 30 GWd/tU is used.

TABLE II

SELECTED SPECIFICATIONS OF SPENT FUEL ASSEMBLIES

LWR = light water reactor
UOX = uranium oxide fuel
MOX = mixed uranium oxide and plutonium oxide fuel

Reactor Type	Initial U enr. (%)	Initial Pu/(Pu+U)	Exposure Range (GWd/tU)	^{244}Cm wt. % of Initial Heavy Metal
LWR UOX	1.5-4.5	0.000	12-50	0.0001-0.026[a]
LWR MOX	0.2	0.052	12-50	≈0.0001-0.03

[a] From Ref. 2.

The neutron rate from (α,n) reactions is usually ignored as a minor contributor for LWR fuel with normal exposures; it is important only for extremely low exposures where virtually no curium is produced. The corrected total neutron count rate n is related to the exposure E by a simple but accurate expression:

$$n = \alpha E^{\beta} . \tag{1}$$

The power β is typically near 4 for exposures greater than 15 GWd/tU.

Neutron count rates have also been correlated with the k_{eff} for an assembly for criticality considerations[5,7] with this expression:

$$k_{eff} = 1 - \kappa E / n , \tag{2}$$

where κ is a constant. Neutron count rates have also been correlated with the mass of plutonium m_{Pu} for safeguards purposes:[6]

$$m_{Pu} = \kappa n^{p} [1 + \varepsilon_1(e)] [1 + \varepsilon_2(T_c)] , \tag{3}$$

where $\varepsilon_1(e)$ is a correction factor for variations in the initial enrichment e, and $\varepsilon_2(T_c)$ is a correction factor for variations in cooling time T_c. The power p is typically about 0.125.

Cooling times T can be estimated from gross gamma-ray measurements using ionization chambers that produce a response g, as follows:[16]

$$g/E = a T_c^{-b} , \tag{4}$$

where b is about 0.9.

An even better measure of the ^{244}Cm neutron rate would be obtained with an assembly in air rather than water. Neutron multiplication within the assembly would be reduced because the fission rate would decrease and effects from any neutron "poison" (such as boron) in the water would be eliminated. The detector head might need a little more moderator around the detector tubes (compared to the underwater equipment) to optimize the detection efficiency, but the change would be slight.

Precisions of neutron counts from spent fuel assemblies under water are excellent, even when the exposure is lower than normal and the cooling time is long. If during a total neutron count T the estimated background count is B, the relative precision of the totals count is

$$\sigma_T/T = \sqrt{(1/T + 2B/T^2)}. \quad (5)$$

The background B is generally much smaller than T and the last term is negligible. This might not be true if measurements were made near one or more other assemblies, depending on the intervening shielding. Table III gives some representative measured count rates and calculated relative precisions after 100-s counts. These data were taken with the Los Alamos Fork Detector[4] with a low detection efficiency.

Comparison of French LWR neutron measurements with their burnup code calculations shows that the plutonium masses are correlated well with the ^{244}Cm neutron count rates; the overall uncertainty of the plutonium mass in an assembly is about 3%.[6] This same inaccuracy for ^{244}Cm in a normal PWR assembly amounts to about 0.25 g of ^{244}Cm (out of 8.2 g, see Appendix A).

The fissile content of spent LWR fuel has also been measured directly by irradiating the underwater assembly with a ^{252}Cf source and observing the increase in neutron emission.[6,7,17,18] The uncertainty in the total fissile mass is about 4%.

TABLE III

PRECISIONS OF TOTAL NEUTRON COUNT RATES FROM LWR FUELS WITH THE FORK DETECTOR

Assembly Type	Exposure (GWd/tU)	Cooling Time (y)	Count Rate (counts/s)	Relative σ after 100 s	Reference
PWR	16	9.0	95	1.0%	16
	30	6.0	424	0.5%	16
BWR	12	4.8	50	1.4%	17
	28	0.2	240	0.7%	17

b. Research Reactor Rods

Fuel rods from research reactors have been measured while underwater.[19] The exposures were low (under 1.5 GWd/tU) so the amounts of ^{244}Cm produced were very small and the ^{242}Cm had decayed away, so the neutron emission rate was dominated by plutonium.

Cooling times were 1 to 7 years, but the long half-lives of the important plutonium isotopes reduce the importance of cooling time in this case.

The instrument was calibrated for plutonium at Los Alamos with fresh fuel rods of MOX and UOX (to quantify the ^{238}U contribution). No new calibration was done at the measurement site. The original plan included correction factors based on computed isotopics for each individual rod, but only a single set of isotopics for a core average was actually available so it was used for all rods regardless of exposure. This instrument measured both total and coincidence neutron rates, but the smaller coincidence count rates offered no advantage over the total count rates because the (α,n) neutron rate was negligible for the metal fuel.

The precision of a total neutron count rate was quite good with this detector. The efficiency for counting neutrons from a ^{252}Cf source was 14%. Assays were done on baskets generally containing five rods (sometimes less). A scanning technique was necessary because the rods were much longer than the detector head; ten measurements were taken at equally spaced locations along a rod. Count rate profiles were strongly peaked in the center and count rates averaged over the lengths were about 70 counts/s. The count time at each scan location was 100 s, so the total count time was 700 s for each basket. The relative precision of such a count rate was 0.45%.

The accuracy of this instrument under these conditions was subsequently known because the research reactor rods were dissolved in batches for reprocessing. The cumulative nondestructive assay result over 223 rods differed from the destructive assay value by 5%. On a couple of occasions an individual nondestructive assay differed from the declared value by more than 10%, but in each case the declared exposure was much lower than usual and the generic plutonium isotopics assumed in the data analysis were known to be inaccurate.

Gross gamma-ray data were also taken to check the integrity of the rods along their lengths. These data were not used in any quantitative manner because cooling time was not an issue for these rods.

c. Variations Among Assemblies

Some factors tend to reduce variations among the assemblies being reprocessed as a batch: (1) similar fabrication specifications (e.g., geometry, initial enrichment), (2) irradiation in the same reactor, (3) irradiations during the same core cycles, and (4) irradiation in three cycles of that core at three different locations within the core to deliberately produce uniform exposures. Nevertheless, there are small variations in exposures among assemblies even in the best case. In a US facility where 25 assemblies met the above conditions, their exposures had an average of 29.139 GWd/tU with a standard deviation of 2.54 GWd/tU (the minimum and maximum exposures were 24.065 GWd/tU and 31.615 GWd/tU).[10] Another set of 36 BWR assemblies meeting the above conditions had an average exposure of 26.675 GWd/tU and standard deviation of 0.689 GWd/tU; the exposures ranged from 25.344 to 28.048 GWd/tU.[11]

Variations among batches of assemblies from different reactors, or the same reactor at different times, will be larger. But this is unimportant to curium balancing if processing is done in batches of assemblies from segregated reactors.

Experience with neutron measurements from the Cadarache Center for Nuclear Studies on assemblies at La Hague shows that there is less than a 1% variation in count rates from assemblies with identical declared exposures.

3. Possible Advances in Spent-Fuel Measurements

a. Coincidence Counting

For the normal LWR spent fuel assembly (>15 GWd/tU exposure, >3 years cooling) coincidence counting gives an advantage over total neutron counting in that the neutron multiplication within an assembly can be determined and the strength of the neutron source can

be calculated from spontaneous fissions. Furthermore, coincidence counting is hardly affected by changes in background rates, unlike total neutron counting.

Normally, three parameters of an assembly affect the coincidence count rate but only two rates are measured (total and coincidence rates). The three parameters are (a) the mass of spontaneously fissioning isotopes, (b) the ratio of the (α,n)-reaction and spontaneous fission production rates (called α, for short), and (c) the neutron multiplication factor M [or, equivalently, k_{eff} because $M = 1/(1 - k_{eff})$]. However, for medium-to-high exposures in LWR fuel the (α,n) term is negligible compared to the spontaneous fission neutrons from ^{244}Cm and induced fissions in the fissile fuel. With only two unknowns remaining, the total and coincidence count rates can be used to determine the spontaneous fission rate and the multiplication.

However, for low exposures (<15 GWd/tU), the neutron rates from ^{238}U and plutonium through spontaneous fissions and (α,n) reactions become important because curium production is greatly reduced. At extremely low exposures (<1 GWd/tU) the curium contribution is secondary. In low-exposure cases, coincidence counting could separate the spontaneous fission neutrons from the (α,n)-reaction neutrons, thereby improving the accuracy of the measurement. The detector head of a coincidence counter should be closely coupled to an assembly to prevent as much water as possible from coming between them; this would help preserve the time correlations among the fission neutrons that form the basis of coincidence counting.

The precision of a coincidence count rate is calculated from this expression:[1]

$$\sigma_R/R = \sqrt{(R + 2A + 2B)} / (R \sqrt{t}) , \tag{6}$$

where R is the real coincidence rate, A is the accidental coincidence rate (both corrected for detector deadtime), B is the coincidence background rate, and t is the count time. The background is normally from cosmic-ray events in the instrument and is negligible compared to the other rates. The real coincidence rate from spontaneous fissions is estimated by another expression:

$$R = S_0 \varepsilon^2 e^{-P/\tau} (1 - e^{-G/\tau}) \langle \nu(\nu - 1)/2 \rangle , \tag{7}$$

where S_0 is the neutron emission rate (n/s), ε is the detector efficiency, P is the detector's predelay time, τ is the die-away time, G is the coincidence gate length, and $\langle \nu(\nu - 1) \rangle$ is the average second moment of the fission neutron distribution. The accidental coincidence rate has a simpler expression:

$$A = G R_T^2 , \tag{8}$$

where R_T is the total neutron count rate corrected for electronics deadtime losses.

These expressions are evaluated in Table IV for ^{244}Cm using plausible values of the parameters. Details on the neutron emission rate are given in Appendix A, but the rate has less impact on the precision than the detection efficiency. Table IV shows that a detection efficiency of only about 1 or 2% is needed for 1% precision in the count rate from curium in a spent PWR fuel assembly in water. (The coincidence background rate of 0.1 counts/s is higher than would be encountered, but even this conservatively high value has negligible effects on the precision.)

TABLE IV

PRECISIONS OF REAL COINCIDENCE COUNT RATES FROM ^{244}Cm IN A PWR ASSEMBLY

2.6% initial enrichment. 33 GWd/tU. 1 yr of cooling
30-cm length of fuel. B = 0.1 count/s
$P = 2$ μs. $\tau = 40$ μs. $G = 64$ μs. $t = 400$ s

ε (%)	σ_R/R (%)
1	1.25
2	0.63
5	0.25
10	0.13
15	0.08
20	0.06

Would coincidence counting improve the precision and accuracy of the measured ^{244}Cm mass in a normal LWR assembly beyond that of total neutron counting? Comparing Tables III and IV shows that excellent precision (better than 1%) is possible with either method; reasonable detector designs and count times can be used in either case. Background rates are actually higher with coincidence counting, if accidental coincidences are considered to be a form of background, but do not interfere with good precision. Accuracy depends primarily on the quality of the calibration process. If only total counting is used, the contribution to the neutron rate from neutron multiplication is unknown so the absolute ^{244}Cm content cannot be determined. Therefore, coincidence counting should be used on fuel assemblies to separately determine the ^{244}Cm masses and the multiplications.

Furthermore, if the assembly of interest is not sufficiently isolated from neighboring assemblies, a coincidence count would be unaffected by neutrons from the other assemblies (even if it changes with time), unlike a simple total neutron count.

b. Multiplicity Counting

Additional information can be determined about the multiplication in an assembly by measuring the triple coincidence rate in addition to the total and double coincidence rates. This is the motivation for multiplicity counting. The counting terminology now shifts from total and coincidence counts to "singles," "doubles," and "triples."

The mathematical basis for triples counting is well developed but it is quite complex.[20-23] Only through experience can some of its nuances be uncovered. For example, in principle the individual masses of two spontaneously fissioning isotopes (such as ^{240}Pu and ^{244}Cm) can be determined even though they are mixed together. However, when the neutron emission rate of one of two isotopes is much larger than the other the precision of the result for the minor constituent will be uselessly poor; Table V shows such a case for assay errors of plutonium in spent fuel where curium dominates the neutron emissions.

TABLE V

ASSAY ERRORS FOR PLUTONIUM IN SPENT FUEL ASSEMBLIES BY THE MULTIPLICITY METHOD

Taken from Ref. 3

		LWR			LMFBR	
		UOX 20 GWd/t %	UOX 45 GWd/t %	MOX 50 GWd/t %	CORE+ AX. BLKT 80 GWd/t %	RADIAL BLKT 5 GWd/t %
Spontaneous Fission Neutron Emission Rate	U	0.0	0.0	0.0	0.0	0.9
	Pu	5.8	0.7	0.5	5.5	98.6
	Cm	94.2	99.3	99.5	94.5	0.5
Assay Error 1 Sigma	Pu	60	508	711	63	2.2

The uncertainty in the triples rate is another important limitation for multiplicity counting. The expression for this uncertainty is quite complex and cannot be written as a closed-form equation; it is evaluated with numerical techniques by a computer code.[23] Experience has shown that as the singles rate increases the uncertainty in the triples rate first improves but then degrades because the accidental triples rate builds. There is a range of count rates within minimum and maximum limits that gives useful results.

Spent fuel assemblies are prolific emitters of neutrons, which places multiplicity counting in a dilemma. Multiplicity counting is best done with a detector of high efficiency to give good precision for the real triples rate. But high emission rates and high detection efficiency also create high singles rates and therefore high accidental triple coincidence rates that degrade the precisions of the triples rates. The detector should have a compact volume so that the geometric coupling to the fuel assembly is limited to a small section of the fuel assembly, thus reducing the magnitude of the source emission-rate term; the detector's efficiency could be kept high in this case.

Even if the triples count rate has excellent precision, what does multiplicity offer to spent fuel assembly measurements beyond normal neutron coincidence counting? If two neutron emitters in the fuel were of nearly equal intensities, multiplicity could deduce their individual intensities.[3] But for normal LWR fuels, the ^{244}Cm neutron emission rate completely dominates that of plutonium. For assemblies of any type with very low exposure the plutonium neutron intensities could rival or even exceed those of curium; multiplicity counting for such cases could be of definite value in obtaining the ^{244}Cm and $^{240}Pu_{eff}$ masses.

4. Measurement Protocols

Measurements can be done underwater, in air, on a whole assembly at once, on separate sections, or while the assembly is in motion. The measurements could also be done in the shearing cell.

a. Measurements Under Water

Most of the spent-fuel measurements have been of passive neutrons and gamma rays while the assembly is under water, which provides shielding from other assemblies. The neutron count rates are generally very high and the conditions are quite favorable for getting good results.

When under water, neutrons from a source can be moderated by the water and induce fissions in the remaining fissile materials (uranium and plutonium). A source of ^{252}Cf has been placed next to the assembly to do this.[6] Water will also increase the induced fission rate in the uranium and plutonium caused by neutrons from curium, and this forms part of the background rate of an active-neutron technique.

Boron in the water has dramatic effects on neutron count rates,[24] but the concentration can be readily measured[25] and corrections applied, if necessary.

Water also provides effective shielding among neighboring assemblies; about 1 m of water is enough to isolate one assembly from others. Boron in the water enhances this shielding, although the shielding is very effective even without boron.

b. Measurements in Air

If the fuel assembly were measured in air, there would be less neutron multiplication than under water. Shielding equivalent to a hot cell would be needed in place of the water. The existing detectors are optimized for underwater measurements, so for use in air some additional moderator could be added around the detector tubes to enhance the detection efficiency.

Neutrons from nearby assemblies would seriously interfere with a total neutron measurement, but they would affect a coincidence measurement only by raising the accidental coincidence count rate. Massive shielding would certainly be needed to isolate nearby assemblies for total neutron counting, but little or no shielding would be needed for coincidence counting.

c. Scanning an Assembly

The existing detectors receive radiations from only a small portion of an assembly at any one time. Count rates decrease rapidly for sources away from the detector and are small after about 20 cm.[7,26] A whole assembly is examined by either taking measurements at several locations along an assembly or passing the assembly continuously through a stationary detector.

d. Whole Assembly

The measurement process can be simplified and the time reduced by measuring the whole assembly at once, rather than scanning. When scanning by taking measurements at several locations, the count time at each location is 30 to 60 s. The total count time is about 5 to 10 min, depending on the length of the assembly. Another 5 to 10 min is needed to move the assembly to the various positions, depending on the design of the assembly handler. The time used in counting while slowly moving an assembly through a detector is again about 10 min.

The time would be reduced to about 1 min by using multiple sets of detectors along the assembly's length and taking data from them simultaneously. The initial cost for fabrication is greater, but such a detector would be simpler to use and not be so susceptible to positioning errors.

e. Measurement Location

The most timely information could be obtained by measuring an assembly immediately before shearing. But if the shearing proceeds and then the measurement is found to reveal an anomaly, there is no chance for a remeasurement and information about that assembly is lost or at least uncertain. So a time lag between the measurement and the shearing is advantageous.

Measurements at the storage location can be done more leisurely before assemblies are removed for shearing. Equipment problems are more easily handled in this area than inside the shearing cell. The disadvantage is that there is less certainty that the assembly when sheared has not been tampered with. However, physical protection devices can monitor assemblies' movements and give assurance that no assembly could have been changed between the measurement and shearing times. Radiation monitors with interpretive logic at key pathways are one example of monitors that could be applied without requiring an inspector's intensive effort.

The practical considerations of measuring assemblies before they enter the shearing cell seem to easily outweigh the extra assurance of measuring them as they are sheared. A compromise is to measure an assembly while still under water in the storage pond but as near the shear as possible.

B. Accountability Tank

An accountability tank is routinely sampled to measure the plutonium content for safeguards purposes; new uses for the results will be proposed later in this report. Various measurement techniques will be considered first.

1. Destructive Analysis

Isotope dilution mass spectrometry (IDMS) gives the most accurate results for the plutonium concentrations in a sample from an accountability tank, but the disadvantages of IDMS are the time and cost involved. An international evaluation program of the IDMS technique gave one-sigma uncertainties of 0.6% for uranium and 1% for plutonium, using conventional spiking techniques.[26] Technical improvements may lower these uncertainties to 0.5% each.

Titrimetry is commonly used for liquids in the separations operations, so these are outside the scope of this study.

2. Nondestructive Analysis

The nondestructive techniques described here are much quicker and much less expensive than a destructive technique. Their precisions and accuracies will be estimated and applied to the curium measurements. The hybrid X-ray instrument can give the plutonium concentration in a sample and an Inventory Sample Neutron Counter (INVS) can give the curium mass in the sample. From this information the curium and plutonium masses in the accountability tank can be deduced and the curium-to-plutonium ratio might be applied to the leached hulls.

a. Hybrid X-Ray Instrument and Plutonium Mass

The hybrid K-edge densitometer (KED) and K X-ray fluorescence (K-XRF) instrument was developed by Kernforschungszentrum Karlsruhe to assay for plutonium when mixed in dissolver solutions with uranium.[26-28] The K-XRF technique was the basis for the assay, with the KED serving as an external reference standard to improve the accuracy and reliability of the K-XRF result. The average of 15 ratios of uranium to plutonium from the hybrid X-ray instrument differed from the average destructive assay result by only 0.02%; the average difference for the uranium concentration was 0.31% and the average plutonium concentration differed by 0.32% for the test conditions. Standard deviations of the 15 concentrations of uranium and plutonium were 0.25% and 0.7%, respectively.

b. Hybrid X-Ray Instrument and Curium-to-Plutonium Ratios

More recently the hybrid instrument has been extended to give the ratio of two elements in a solution.[29] Ratios between 0.001 and 1000 can be determined. One application has been for

the ratio of thorium to plutonium in solutions from anion exchange columns. The detection limit for thorium was found to be 25 mg/ℓ (1000-s assay) and the minimum useful Th:Pu ratio was 0.001 (5% precision in 1000 s for thorium concentrations greater than 200 mg/ℓ).

Expected ratios of curium to plutonium have been estimated through calculations. A standard computer code to calculate atom densities in fuel assemblies is ORIGEN and the version for desktop personal computers, ORIGEN2 (version 2.1), was used here.[30] The results are in Table VI, where all isotopes of curium and plutonium have been included. The accountability tank has atom densities smaller than these because of dilution, but the ratio of curium to plutonium should still be representative. If the enrichment or irradiation schedule are changed within normal bounds, the effects are apparent but secondary compared with the effects of exposure. The ratio of the values in Fig. 2 is 2.4×10^{-3}, which is consistent with the values in Table VI.

The growth of curium with exposure is approximately the power law of Eq. (1), while the growth of plutonium is more nearly linear. Therefore the curium-to-plutonium ratio also follows a power law; for the data in Table VI the power is 4.40.

TABLE VI

CALCULATED CURIUM-TO-PLUTONIUM RATIOS IN AN ACCOUNTABILITY TANK

UOX PWR 17 x 17 fuel, 3.3% enriched.
Three successive cycles and a cooling time of 491 days.
Atom densities are before dissolution.

Exposure	Atom Density (atoms/tU)		
(GWd/tU)	Cm	Pu	Cm/Pu
15	1.57×10^{21}	1.36×10^{25}	1.16×10^{-4}
30	4.27×10^{22}	2.00×10^{25}	2.13×10^{-3}
45	2.99×10^{23}	2.39×10^{25}	1.27×10^{-2}

The most useful result of Table VI for the purpose of this study is to define the range of the normal PWR curium-to-plutonium ratio. For exposures between 30 and 45 GWd/tU and enrichments from 2.5% to 4%, the ratio ranges from 1×10^{-3} to 2.5×10^{-2}.

Curium-to-plutonium ratios for this range are within the grasp of the hybrid instrument, but lower values could be difficult for the instrument. The precision of the ratio degrades as the ratio shrinks; at the small ratio of 2.4×10^{-3} the precision of the ratio measurement is only about 5% which is much inferior to the other measurements throughout the head-end. The next two sections give a way around this difficulty.

c. Inventory Sample Counter

The curium concentration in a dissolver solution may be measured through its neutron emission with an INVS.[1,31] Plutonium solutions have been used in this instrument and a nearly linear calibration curve was found up to 100 mg of $^{240}Pu_{eff}$ (the largest amount used). The ^{244}Cm has no multiplication itself (because fissions are not significantly induced in it) and the plutonium in the dissolver solution is very dilute so its multiplication in the small sample (a few cm^3) is negligible.

A sample from an accountability tank would have curium as a small fraction of plutonium; the curium-to-plutonium mass ratio would be from 0.001 to 0.025. Table VII (Part B) compares the spontaneous fission rates of these two elements as a function of this fraction.

The precision of an INVS instrument as a coincidence counter for ^{244}Cm alone is shown in Table VIII, as estimated with Eq. (6). The ^{244}Cm concentration and sample volume have little or no individual consequences in this case, so just the ^{244}Cm mass in the sample is indicated. There is clearly no problem in achieving better than 1% precision. The data in Fig. 2 show that a 1-cm^3 sample of the dissolver solution would contain 1.45×10^{-5} g of ^{244}Cm; the calculated precision of the coincidence count rate is then about 1% with a 400-s count.

TABLE VII

SPONTANEOUS FISSION RATES
FROM CURIUM AND PLUTONIUM IN
DISSOLVER SOLUTIONS

PART A. Nuclear Data.

Isotope	Relative Isotopic Mass	Specific Fission Rate [fissions/(s•g)]	Relative Isotopic Fission Rates
^{238}Pu	0.02	1.17×10^{3}	2.21×10^{1}
^{239}Pu	0.58	1.01×10^{-2}	5.86×10^{-3}
^{240}Pu	0.20	4.72×10^{2}	9.44×10^{1}
^{241}Pu	0.15	2.22×10^{-2}	3.26×10^{-3}
^{242}Pu	0.06	8.00×10^{2}	4.80×10^{1}
^{all}Pu	1.00		1.65×10^{2}
^{242}Cm	0.001	8.27×10^{6}	8.67×10^{3}
^{243}Cm	0.023	3.43×10^{1}	7.82×10^{-1}
^{244}Cm	0.939	3.97×10^{6}	3.73×10^{6}
^{245}Cm	0.037	1.35×10^{1}	4.97×10^{-1}
^{246}Cm	0.000	2.97×10^{6}	0.00×10^{0}
^{all}Cm	1.000		3.74×10^{6}

PART B. Dissolver Solution Curium-to-Plutonium Fission Rates.

3 years of cooling.

Mass Ratio (Cm:Pu)	Cm-to-Pu Ratio of Fission Rates	Cm Fraction of the Total Rate
0.001	22.7	0.958
0.005	113.6	0.991
0.010	227.2	0.996
0.025	567.9	0.998

To decide whether the INVS counter can measure these low masses of ^{244}Cm, the detection limit can be calculated. The detection limit is determined by the rate of cosmic-ray events within the detector body and the accidental coincidence rate from the room's background neutrons. The computational details are in Appendix B. Table IX shows detection limits for a range of plausible background rates.

TABLE VIII

CALCULATED COINCIDENCE COUNT RATE PRECISIONS FROM AN INVS COUNTER FOR ^{244}Cm

3.97×10^6 fissions/(s•g), 1.08×10^7 neutrons/(s•g), $\varepsilon = 43.5\%$, $P = 4.5$ μs, $\tau = 64$ μs, $G = 128$ μs, $t = 400$ s.

^{244}Cm Mass (g)	σ_R/R (%)
0.00001	1.17
0.0001	0.37
0.00015	0.31
0.001	0.13
0.01	0.065
0.1	0.055
0.500	0.054

TABLE IX

DETECTION LIMIT OF THE INVS COUNTER FOR ^{244}Cm AS A FUNCTION OF THE COINCIDENCE BACKGROUND RATE

$\sigma_R/R = 0.25$.

3.97×10^6 fissions/(s•g), 1.08×10^7 neutrons/(s•g), $\varepsilon = 43.5\%$, $P = 4.5$ μs, $\tau = 64$ μs, $G = 128$ μs, Count time = 400 s.

Coincidence Background Rate (counts/s)	Detection Limit (μg ^{244}Cm)
0.000	0.0081
0.001	0.0085
0.010	0.0111
0.100	0.0226
1.000	0.0614

The plausible background coincidence rates are less than one, but in any case the detection limit is a small fraction of a microgram and thus is not a limiting factor in curium measurements. All the ^{244}Cm masses in Table VIII are well above the microgram range.

The count rate from the ^{3}He neutron-detector tubes in an INVS counter can be affected by intense gamma rays, but lead or tungsten shielding may be added to mitigate the problem. Dissolver solutions from fuel with 30-35 GWd/tU exposure and 3 years of cooling have been measured to have 100 to 190 Ci/ℓ from beta and gamma decays.[28] A sample of a few milliliters would thus have only about 0.15 Ci and only a small thickness of gamma-ray shielding would be necessary.

3. Precision of the Curium-to-Plutonium Ratio

The curium-to-plutonium ratio would be immediately useful in the measurements on drums of leached hulls if the ratio is the same for the hulls and the accountability tank solution. The precision of the ratio can be readily calculated from measurements with the X-ray and INVS instruments. If the precision of the plutonium concentration is 0.7% and that of the curium concentration is 1.0%, the precision of the ratio is 1.2%.

The precision of the curium concentration from the INVS instrument can be improved with count times longer than the 400 s assumed in Table VIII and the preceding paragraph. For example, the precision would be 0.6% after 1000 s and 0.5% after 1500 s. The corresponding precisions of the curium-to-plutonium ratios are 0.92% and 0.86%. A precision for the curium concentration of less than 0.5% is probably not worth the time required if the precision of the hybrid X-ray instrument is still 0.7%.

4. Fines (Undissolved Solids)

A small fraction of a spent fuel assembly is left as very small undissolved particles called "fines." The final disposition of the fines and the amounts of plutonium and curium they may carry are important to curium balancing.

An extensive study has been done[32] to characterize fines from PWR spent fuel assemblies with exposures from 7 to 39 GWd/tU, with the following pertinent results.

(a) Amount of Fines. The amount of fines grows with exposure in a nonlinear fashion. In the 30 to 40 GWd/tU range, the weight percentage of fines (per initial uranium) grows from about 0.2 to 0.4% fairly linearly. In a plant with an annual throughput of 800 tU for assemblies with 30 to 40 GWd/tU exposures, the mass of the fines produced each year is about 2.5 t.

(b) Particle Size. The fines are quite small (less than 0.10 μm) immediately after dissolution and then grow by coagulation to larger sizes (5 to 50 μm) after about a day.

(c) Composition. Five elements make up more than 70% of the material in the fines: molybdenum, technetium, ruthenium, rhodium, and palladium. Their relative amounts are not the same as calculated by ORIGEN2 for spent fuel because the elements have different solubilities. These elements, and most of the other 30%, do not directly affect curium balancing, but the fines also were found to have plutonium; curium was not sought in the study. The plutonium fractions for assemblies with more than 17 GWd/tU exposure were from 0.03 to 0.08% of the fines by weight, with no clear correlation with exposure. The fraction of plutonium in the fines compared to the total plutonium in the spent fuel was estimated to be 0.005 to 0.02%. Cesium-137 was found on the fines in about the same fraction and it is considered to be totally dissolved, so apparently the plutonium is in fact dissolved and is merely on the surface of the fines as a contaminant.

Some small fraction of the fines is no doubt carried on the leached hulls as the hulls leave the dissolver tank. The mass of plutonium in a year of fines leaving the dissolver tank is 0.27 to 1 kg in an 800-tU plant. But the hulls are washed and the effluent returned to the dissolver tank, so the fraction of the fines bearing plutonium or curium that reaches the leached hull drums is assumed to be negligibly small and the fines should not affect the plutonium and curium amounts in the hulls.

The fines are carried through the separation facility and stream and leave the plant in the vitrified high-level waste.

C. Leached Hulls

1. TASTEX Experience

The Tokai Advanced Safeguards Technology Exercise (TASTEX) of 1978-1981 resulted in a 1982 report[33] that included a section on leached hulls. Progress has been made since then, but the report is still useful and informative.

Experience with nondestructive measurements of the fissile contents of leached hulls through 1981 was summarized in the report. Passive gamma-ray measurements had been done at La Hague and Tokai, and an active neutron technique was in use at La Hague. The TASTEX report also noted that for "longer" cooling times a passive neutron count could be used and would be preferable; this could only mean a measurement of the curium neutron emissions with an inference of the plutonium content.

The active neutron instruments measure the fissile material directly, but they cannot distinguish between uranium and plutonium. Passive gamma-ray instruments reveal the fission product load, and the amount of fissile material must be inferred.

The leached hulls in the TASTEX report were measured in baskets (La Hague: 10 cm in diameter, 95 cm long. Tokai: 20 cm in diameter, 180 cm long). Results with active neutron and passive gamma-ray techniques were equivalent. Random errors were about 20% and systematic errors were unknown.

2. Active Neutron Assay of Leached Hulls

Active neutron measurements of leached hull containers are difficult because of the high gamma-ray and neutron backgrounds. The gamma-ray dose rates are several thousands of R/hr and the neutron background rate is typically about 2×10^5 n/s (primarily from ^{244}Cm). If ^3He tubes are used to count the induced fission neutrons from the active interrogation, a large amount of lead shielding is required to shield the ^3He tubes from the gamma rays.

For the LWR fuel considered in this study, the induced fission reactions take place in both the ^{235}U and the plutonium. Thus, the analysis must have additional information to separate these two components and determine the plutonium in the leached hulls.

Neutron sources that have been used for leached hull active assays include ^{252}Cf spontaneous fissions (shuffler) and neutron generators (14-MeV neutrons from D-T reactions). The D-T generators can be used in the pulsed mode for the differential die-away technique (DDA)[34] to count prompt-fission neutrons rather than delayed neutrons (as with the shuffler). This increases, by about 2 orders of magnitude, the signal-to-background ratio. However, the DDA method has about two orders of magnitude less source yield. The lead shielding that is needed for gamma-ray shielding and the low source yield ($\approx 2 \times 10^8$ n/s) makes the DDA neutron interrogation less efficient for spent fuel than for low-level wastes.

If ^{252}Cf shufflers are used to assay leached hulls, large sources ($\approx 1 \times 10^{10}$ n/s) must be used to compete with the ^{244}Cm background neutrons. The neutrons in both the shufflers and DDA are moderated in energy so the fission rate in the ^{238}U is negligible compared with the fissile fission rate.

The most important problem in the active assay of leached hulls is that the induced signal comes from both ^{235}U and plutonium, whereas the safeguards focus is on the plutonium. The plutonium-to-uranium ratio (≈ 0.01) can be measured with the hybrid X-ray densitometer for a liquid sample from the accountability tank. However, the ^{235}U enrichment would need to be determined using a mass spectrometer. This ratio of ^{235}U to plutonium could be applied to the hulls to calculate their plutonium content. If this approach is used, it is much simpler to use a passive neutron measurement of the hulls plus the curium-to-plutonium ratio from the accountability tank to determine the plutonium in the hulls.

3. Active Assay Examples

Since the TASTEX work, new active neutron instruments for leached hulls have been placed in operation at La Hague and Dounreay.

a. La Hague

Leached hulls from LWR assemblies are now measured at La Hague in large 800-ℓ drums, not small baskets, which increases the attenuation of the fission-product gamma rays and makes uniform irradiation with neutrons more difficult. The experience with these drums in a Cadarache passive-active shuffler has given us the following information.

- There are about 5 to 10 g of plutonium in a drum and about 600 g of uranium; the plutonium-to-uranium ratio in the waste is thus about 0.010 to 0.015.
- The neutron emission rate from a drum is about 1.7×10^5 n/s (2×10^3 to 3×10^3 counts/s with 1.5% detection efficiency). The 5 to 10 g of plutonium produce only about 2.5×10^3 n/s (with 24% ^{240}Pu), so if the remaining emissions are from ^{244}Cm, the mass of ^{244}Cm is 0.0155 g. The curium-to-plutonium ratio is then about 0.002.
- A ^{252}Cf shuffler (active neutron interrogation) irradiating a drum with 1.2×10^{10} n/s gives about 40 delayed-neutron counts/s (the detection efficiency is 1.5%). The detection limit for an 800-ℓ drum is about 1 g of fissile material in a 3-hour assay. The active neutron interrogation signal comes from both the plutonium and the ^{235}U that is 1 to 2% of the residual uranium. Knowledge about the input fuel, the fuel's irradiation history, and passive neutron measurements are used to deduce the plutonium content.
- A DDA instrument is also used to irradiate drums and stimulate fissions in the plutonium. The output of the D-T neutron generator is much lower than that of ^{252}Cf, but its performance will equal that of the shuffler with a shorter assay time because it counts prompt-fission neutrons instead of the less numerous delayed neutrons. Self-shielding could adversely impact accuracy because the drum does not rotate and thermal neutrons will travel through many thin layers of liquid on the hulls as they reach the farthest points of a drum.

b. Dounreay

The Dounreay plant[35,36] uses a shuffler to measure hulls from fast breeder reactor fuel assemblies in a basket not very different from the basket at Tokai (20 cm diameter, 100 cm long). The shuffler has been calibrated up to 63 g of plutonium; a typical plutonium mass in a basket is about 30 g. For the Dounreay fast breeder fuel the ^{235}U content is small relative to the plutonium, so the active neutron assay can be used to directly measure the plutonium. A very large ^{252}Cf neutron source is used to generate a signal large enough to override the curium neutron background rate. The passive mode is used to screen a basket for plutonium and curium; only if a passive count is significantly above background is an assay made in the active mode. Cooling times are generally short enough for ^{242}Cm to significantly contribute to the neutron emission rate.

TABLE X

MATERIALS IN THE REFERENCE DRUM OF LEACHED HULLS

800-ℓ drum with hulls and end pieces from 4 PWR assemblies
with more than 3 years of cooling,
600 g of uranium with 0.83% ^{235}U,
8 g Pu with isotopics from Ref. 1 for 25% ^{240}Pu, and
0.15 g of ^{244}Cm with no significant ^{242}Cm.

Isotope	Mass (g)	Spontaneous Fission Passive Neutron Emission Rate[a] (n/s)
^{235}U	5.00	0.00150
^{238}U	595.	8.09
^{238}Pu	0.126	326.
^{239}Pu	4.59	0.100
^{240}Pu	2.00	2040.
^{241}Pu	0.845	0.042
^{242}Pu	0.4436	763.
^{241}Am	0.0927	0.109
^{242}Cm	0.000	0.000
^{244}Cm	0.015	162000.

[a] The (α,n) neutron rates are negligible compared with the spontaneous fission rate of ^{244}Cm.

c. Reference Leached-Hulls Drum

The large drum of leached hulls at La Hague is taken to be more representative of the situation being studied here than is the Dounreay basket. The assumed isotopic mixture in the reference drum is given in Table X.

4. Passive Neutron Measurements of Leached Hulls

The passive neutron measurement of the ^{244}Cm in the leached hulls is easier and more accurate than the active neutron interrogation methods for the fissile materials. The application problem is in establishing the accuracy of the curium-to-plutonium ratio in the hulls. Table X shows that the ^{244}Cm mass is very small, but the neutron emission rate is much larger than the combined rate from all other isotopes. An efficiency of only 5% would give a coincidence count rate of 230 counts/s and an imprecision less than 1% would be reached in a few minutes. The imprecision of the totals count rate would be much less than 1% in only 1 s.

The accuracy of a curium mass deduced from a passive count is limited by the accuracy of the calibration and nonuniformity of the detection probability with the distribution of curium

in a drum. This second problem can be reduced by a careful design of the assay chamber, if the reprocessing facility's design allows a reasonable space for the detector. Calibration standards could be very accurate, in principle, so the accuracy of the curium assay should be good (e.g., 2%).

Curium itself can be detected with excellent precision and accuracy but calculating a corresponding plutonium mass will introduce more uncertainties. There are bias-inducing issues beyond the usual calibration and ratio determinations. Before we can establish the accuracy in using the curium-to-plutonium ratio from the accountability tank for the leached hulls, we must determine if there has been any change in the curium-to-plutonium ratio caused by the shearing, the dissolution process, and the rinsing. Future collaborations with the reprocessing facilities are needed to quantify these potential sources of inaccuracy.

D. Integrated Curium Measurements At A Plant's Head-End

Individual curium measurements at the three locations (spent fuel assemblies, accountability tank, and leached hulls) for the plant's head-end of Fig. 2 have been discussed up to this point. The usefulness of the measurements individually and when used in concert at the head-end MBA will now be examined. The result is three major improvements:

- ♦ more assurance of the integrity of the spent fuel assemblies;
- ♦ more assurance that no fuel was diverted from the dissolver tank; and
- ♦ an accurate assay of the plutonium in the leached hulls drums.

The process by which these improvements are achieved has the following steps, which are also summarized in Fig. 6.

(a) Spent Fuel Assemblies. A high-quality curium measurement on a spent fuel assembly will verify the integrity of the assembly, assuring that no material has been diverted. The measurement should be able to quantitatively detect the absence of irradiated fuel.

The quality of a measurement would be improved by scanning the full length of the assembly and making a multiplication correction based on neutron coincidence counting. This eliminates the contribution to the neutron count rate from the uranium and plutonium, leaving only the count rate from curium. The curium present is virtually all ^{244}Cm because the cooling time is 3 years or more and the ^{242}Cm has decayed away. Only the ^{244}Cm is a significant neutron emitter from spontaneous fissions in this case. Coincidence counting eliminates the neutrons from (α,n) reactions and neutrons from induced fissions in the uranium and plutonium.

The relative imprecision of a measurement for one assembly would be much less than 1%, but the relative systematic error in correlating count rate with ^{244}Cm mass is about 4% for one assembly.

(b) Accountability Tank: Plutonium. The existing analysis of a sample from the accountability tank gives an accurate concentration and mass of plutonium in the tank. The relative inaccuracy of the present measurements of the plutonium mass is a little under 1% (perhaps 0.5 to 0.8%); 0.7% will be used here.

(c) Accountability Tank: Curium. A new neutron coincidence measurement on the accountability tank sample with an INVS counter can give the curium concentration and mass in the accountability tank. This should have a relative inaccuracy of about 1% or a little less.

(d) Assemblies and Tank. The curium in the spent fuel assemblies and the accountability tank should be statistically equal. This checks that all the fuel has reached the tank (except for very small waste quantities in the leached hulls). The expected ratio of curium amounts from the two measurements is extremely close to unity. The relative uncertainty in the ratio is about 1.6%, using the results from (a) and (c).

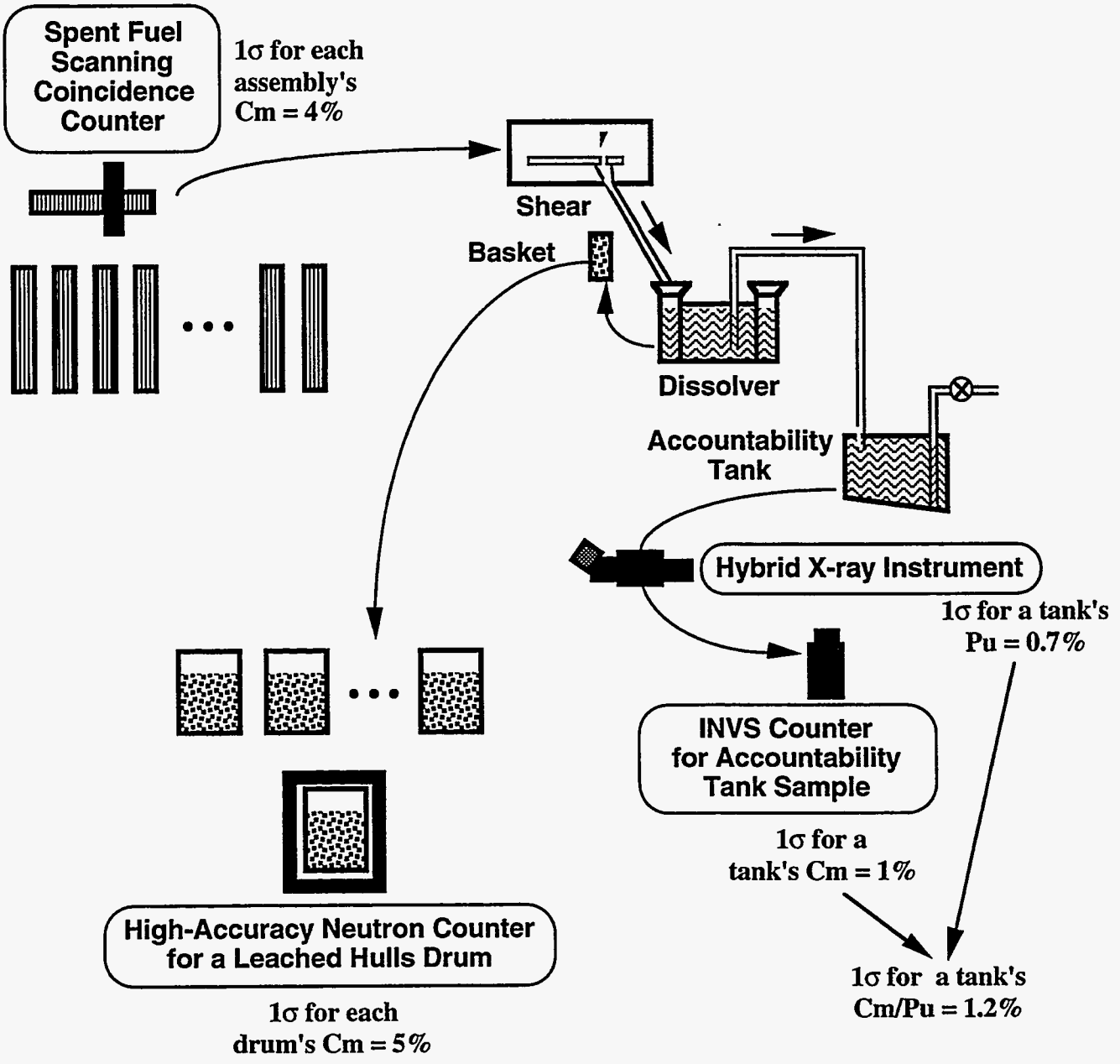

Fig. 6. The three measurement points are indicated here: spent fuel assemblies, samples from the accountability tank, and the drums of leached hulls. The full length of a fuel assembly is measured by either scanning the counter along the assembly or moving the assembly through the counter. The sample from the accountability tank normally used to determine the plutonium concentration is also placed in the INVS counter to measure its ^{244}Cm concentration. A passive neutron coincidence counter was designed for high-accuracy assays for ^{244}Cm on each leached hulls drum. Relative one-sigma inaccuracy estimates for ^{244}Cm masses are indicated at each measurement point. Relative inaccuracy values for plutonium-related measurements on samples from the accountability tank are also shown.

(e) Curium-to-Plutonium Ratio. The curium-to-plutonium ratio in the accountability tank can be calculated from the results of (b) and (c). A likely ratio is about 0.00242 with a relative uncertainty of about 1.2%.

(f) Leached Hulls: Curium. The curium amount in the drums of leached hulls can be measured with passive neutron counting with a relative imprecision smaller than 1%. An inaccuracy of about 5% could be expected from the calibration process. The total relative uncertainty is then about 5% for one drum. After measuring four such drums containing hulls from 16 assemblies that might form a batch, the combined relative uncertainty is about 2.5%.

(g) Leached Hulls: Plutonium. The plutonium mass in the leached hulls drums can be found from the results of (e) and (f). This will verify that only normal waste amounts of plutonium leave the MBA through this waste stream. The relative uncertainty of this mass (using the preceding uncertainties) is about 2.8%. However, for this to be correct it must first be proven that the curium-to-plutonium ratio in the accountability tank is also accurate for leached hulls; investigations at reprocessing facilities are anticipated in the future to resolve this matter.

E. Summary of Head-End Curium Balancing

Plutonium cannot be tracked by direct observation through the head-end of a reprocessing plant, so we consider the alternative of improving and extending measurements of the neutron-emitting curium at key locations. A model of the essentials of the head-end of a reprocessing plant is shown in Fig. 6; a batch of assemblies and their products are indicated along with the three key measurement locations (spent fuel assemblies, accountability tank, and leached hulls drums). Plutonium is measured directly in samples from the accountability tank and the plutonium mass in the tank is inferred from this measurement. But this direct technique cannot be applied to the spent fuel assemblies and the drums of leached hulls.

Curium can be assayed readily at the three locations because of its intense emission of neutrons through spontaneous fissions. The precisions of these measurements are always better than 1% because of the high neutron emission rates from even small amounts of ^{244}Cm. Measurement inaccuracies are primarily caused by systematic errors from calibrations.

We suggest the following improvements in existing measurements and a new measurement of curium to enhance the safeguarding of the fissile materials in the head-end of a reprocessing plant.

- Spent Fuel Assemblies. The accuracy of neutron measurements on spent fuel assemblies can be improved to determine the ^{244}Cm masses prior to shearing by using coincidence neutron counting while scanning the entire fuel length of each assembly. Coincidence counting allows a correction for the multiplication of neutrons by the residual fissile materials and greatly reduces the problem of interference in the measurement by neighboring assemblies.

The relative inaccuracy in the ^{244}Cm mass for one assembly would be about 4%. The inaccuracy for a batch of assemblies would be smaller because the average would be more accurate than any single measurement.

- Accountability Tank. The ^{244}Cm mass in the accountability tank can be measured by counting neutron coincidences from the sample routinely taken for the plutonium mass determination. The relative inaccuracy of this mass would be about 1%. The precision of the ratio of curium and plutonium concentrations would also be about 1%, depending on the count time used for the coincidence neutron counter.
- Assemblies and Tank. By comparing the results of the ^{244}Cm measurements on assemblies and the accountability tank, there would be increased safeguards confidence that all the fuel has been dissolved and resides in the accountability tank, except for the small waste in the leached hulls. The ratio of these two ^{244}Cm masses calculated from the neutron measurements would have a relative inaccuracy of about 1.6%. There would be a small additional error related to the volume or mass determination of the sample.

♦ Leached Hulls. The ^{244}Cm mass in a drum of leached hulls can be measured with improved accuracy using passive neutron counting and a specially designed detector (more constant response to neutrons originating throughout the assay chamber's volume and coincidence counting for reduced interference). The relative inaccuracy in this mass would be about 5% for one drum and 2.5% for four drums that might form a batch. The curium-to-plutonium ratio (calculated from the plutonium and curium measurements on the accountability tank sample with a relative imprecision of about 1.2%) can be applied to the drum of leached hulls to calculate the drum's plutonium content. The inaccuracy in the plutonium mass in the four drums is 2.8% plus an increase from any change (yet to be established) in the curium-to-plutonium ratio between the tank and the drums because of the shearing, dissolution, or rinsing processes. The plutonium leaving the plant in this waste stream is thus very closely monitored.

III. HIGH-LEVEL LIQUID WASTE MEASUREMENTS

Three measurement techniques are considered for safeguarding the plutonium in high-level liquid wastes: the direct assay for plutonium, the indirect safeguarding technique of curium balancing, and confirmation measurements before and after vitrification. But first the nature of the waste is discussed.

A. Characteristics of High-Level Liquid Waste

The waste is assumed to have the actinide amounts shown in Table XI before and after vitrification. After preparation stages (including calcining) and the loss of volatiles in the melter, the volume of waste is reduced by about a factor of 7.5; the concentration of the waste is therefore increased. The relative amounts of oxides in the liquid waste are given in the upper half of Table XII. The lower half of that table shows the assumed glass feed composition. The waste oxide and glass feed are mixed in the ratio of 1:3 to form the vitrified waste.

The contents of a canister of vitrified waste are modeled as occupying a right-circular cylinder with a radius of 20 cm and a height of 87.5 cm, giving a volume of 1.10×10^5 cm^3 (or 110 liters). A canister is taken to contain 300 kg of vitrified waste, so the density of the waste is 2.73 g/cm^3.

TABLE XI

ACTINIDE CONTENTS OF THE WASTE

	U[a]	Pu[b]	Am[c]	Cm[d]
Entering Vitrification Plant (g/m^3)	2820	33.3	360	6.1
Entering Melter (g/m^3)	4570	55	584	9.9
After Vitrification (g/m^3)	20700	250	2660	45

(a) ≈1% ^{235}U. (b) ≈24% ^{240}Pu.

(c) Eight years decay of ^{241}Pu. (d) All ^{244}Cm; no ^{242}Cm.

TABLE XII

WASTE AND GLASS COMPOSITIONS

	Compound	wt. %
Waste Oxides	Na_2O	10.0
	P_2O_5	0.3
	Fe_2O_3	2.0
	NiO	0.2
	Cr_2O_3	0.1
	Fiss. Prod.	9.9
	Actinides	2.5
	Subtotal	25.0
Glass Feed	SiO_2	46.7
	B_2O_3	14.3
	Al_2O_3	5.0
	Li_2O	3.0
	CaO	3.0
	ZnO	3.0
	Subtotal	75.0
Vitrified Waste	Total	100.0

B. Problems With Direct Nondestructive Assay Of Plutonium

1. Passive Gamma-Ray Technique

Gamma rays from plutonium are much less intense than those from the fission products, so a direct assay based on the passive emission of characteristic gamma rays is not possible. Furthermore, the waste container is much too large and the vitrified waste too dense to allow gamma rays to escape from all but the surface of the waste. An assay based on gamma-ray measurements would have to assume homogeneity of the waste. The example below illustrates these problems.

By the time of vitrification (≥5 years of cooling) the gamma rays from short-lived fission products have become negligible and the remaining gamma rays are predominately from ^{137}Cs. Can the 662-keV gamma ray from ^{137}Cs be used to track the plutonium waste? Attenuation in the large volumes of liquid and glass is severe, making it impossible to be assured that a container has more than a thin outer layer of material. If the 662-keV gamma rays are created uniformly throughout a cylinder (20 cm radius, 87.5 cm high) of the waste in Table XI, about 26% of them will escape without a significant energy loss, according to a Monte Carlo calculation. This still produces a large signal, but of the gamma rays that start within 5 cm of the cylinder's center, only about 6% escape with nearly their full energy. So attenuation by the glass makes it difficult to fully track even the ^{137}Cs. Furthermore, the ^{137}Cs cannot be used to track the plutonium because of the volatility of the ^{137}Cs, which changes the ratio of ^{244}Cm to plutonium at different vitrification process steps.

2. Active Gamma-Ray Techniques

The absorption-edge technique for measuring a concentration requires transmission of gamma rays through the material; this is impossible with a vessel as large as the waste canister. If the technique were applied to a small sample of the material, it would have to be assumed that the sample is representative. In any case, the plutonium concentration is too low in this waste for this technique.

X-ray fluorescence[1,29] can give the ratio of elements when the ratio is within 3 orders of magnitude of unity. In the present case the plutonium-to-curium ratio is expected to be about 5.5 (Table XI), which is well within the range for X-ray fluorescence. The uranium-to-plutonium ratio is about 82 and the uranium-to-curium ratio is about 450, so the concentrations of all three elements can be measured in principle. The relative precision for curium will be about 3% while the relative precisions for uranium and plutonium will be much better. Furthermore, this technique can be used through the vessel walls and in the presence of intense gamma-ray backgrounds if a sufficiently intense X-ray generator is used. There must be space against the vessel's surface for a detector with a bulky collimator.

3. Passive Neutron Techniques

Total passive neutron counts will be dominated by neutrons from spontaneous fissions of ^{244}Cm (Table XIII) and (α,n) reactions with the matrix [notably oxygen before vitrification and boron after vitrification (Table XIV)]. The contribution by (α,n) reactions to the total neutron emission is approximately 3% before vitrification and about 12% afterwards. Coincidence neutron counts will be dominated by neutrons from the spontaneous fissions of ^{244}Cm.

About 80% of the (α,n) yield is from reactions with the boron isotopes; 12% is from ^{23}Na, and 3% from oxygen. Essentially all (99.5%) of the (α,n) yield is caused by alpha particles from ^{241}Am and ^{244}Cm, with the smaller number of alpha particles from plutonium isotopes being negligible.

a. Coincidence Counting

The feasibility of coincidence counting for plutonium in high-level liquid waste can be decided by comparing coincidence rates for neutrons from the relative amounts of plutonium and curium given in Table XI. There is no hope of a direct assay for plutonium if the coincidence rate from the plutonium is much smaller than the rate from curium.

TABLE XIII

SPONTANEOUS FISSION NEUTRON PRODUCTION

	Yield [n/(g•s)]	Mass (g)	Production [n/(s•canister)]
Pu[a]	3.41×10^2	27.8	9.48×10^3
^{244}Cm	1.08×10^7	5.0	5.40×10^7

(a) 1.2% ^{238}Pu, 24% ^{240}Pu, 3.8% ^{242}Pu

TABLE XIV

NEUTRON PRODUCTION BY (α,n) REACTIONS IN 300 kg OF VITRIFIED WASTE

Vitrified Waste Composition from Tables XI and XII.
Yields Calculated by W. B. Wilson, LANL.

Target Element	(α,n) Yield (n/s•canister)	Percent of Total Yield
^{7}Li	7.70×10^{4}	1.1
^{10}B	3.25×10^{5}	4.7
^{11}B	5.26×10^{6}	75.2
^{17}O	1.79×10^{4}	0.2
^{18}O	1.95×10^{5}	2.8
^{23}Na	8.37×10^{5}	12.0
^{27}Al	1.02×10^{5}	1.5
^{29}Si	1.21×10^{5}	1.7
^{30}Si	5.82×10^{4}	0.8
Total	7.00×10^{6}	100

Details of calculating the plutonium-to-curium ratio of count rates are in Appendix C. The resulting ratio of coincidence rates is only 0.000162 for a plausible mixture of plutonium isotopes and ^{244}Cm. It is clearly impossible to use coincidence counting to assay for plutonium directly when mixed in this manner with curium.

b. Multiplicity Counting

An analysis of applying the neutron multiplicity technique to waste is reported in detail in Ref. 3 (pages 16-21). In principle, the technique can determine the individual amounts of plutonium and curium when mixed together because the neutrons from their spontaneous fissions have different multiplicity distributions. However, with waste the accuracy for the plutonium amount is poor because the neutron count rate is dominated by neutrons from curium; relative assay errors for the plutonium mass would be too large to be useful.

4. Active Neutron Techniques

Fissions can be induced in the waste plutonium (principally ^{239}Pu) by neutrons from an external interrogating neutron source, while the probability of inducing fissions in ^{244}Cm is very small. The increase in the neutron count rate above background is approximately proportional to the mass of ^{239}Pu present with very significant interference from other isotopes. These interferences are demonstrated with the $^{239}Pu_{eff}$ principle.[37] A can of plutonium oxide would produce an active signal from fast-neutron interrogation equal to this much ^{239}Pu:

$$^{239}Pu_{eff} = 0.786\ ^{238}Pu + {}^{239}Pu + 0.515\ ^{240}Pu + 1.414\ ^{241}Pu + 0.422\ ^{242}Pu + 0.545\ ^{241}Am + 0.671\ ^{235}U + 0.082\ ^{238}U\ , \quad (9)$$

where the isotope symbols stand for the masses of those isotopes. The individual terms in this sum are evaluated in Table XV for the plutonium in Table XIII with 2.3 kg of uranium (1% enriched). The large masses of ^{238}U and ^{241}Am cause them to generate 90% of the signal. Only 4.5% of the signal is from ^{239}Pu and all the plutonium isotopes contribute only 6.4%.

Several instruments are based on an active neutron principle, but use different neutron sources:

- active-well counters (AmLi neutron source);[38,39]
- shufflers (^{252}Cf source);[38-40] and,
- differential die-away time instruments (D-T generator).[34]

TABLE XV

ISOTOPIC CONTRIBUTIONS TO ACTIVE NEUTRON SIGNALS

Isotope	Mass (grams)	Relative Count Rate	Fractional Contribution
^{235}U	23.	15.4	0.0397
^{238}U	2277.	186.7	0.4818
^{238}Pu	0.33	0.3	0.0007
^{239}Pu	17.61	17.5	0.0452
^{240}Pu	6.67	3.4	0.0088
^{241}Pu	2.14	3.0	0.0077
^{242}Pu	1.06	0.4	0.0010
Total Pu	27.81	24.6	0.0635
^{241}Am	295.	160.8	0.4150
Total		387.5	1.0000

Even if an AWCC could be designed for a large waste canister, the number of typical AmLi sources needed to overcome the background rate would be in the thousands and would increase the background rate even further. An AWCC simply is not practical.

The ^{252}Cf source in a shuffler would also be impossible to implement because of the huge neutron background rate. To achieve just a relative precision of 10% in the delayed-neutron count could require 100 mg (or more) of ^{252}Cf.

The DDA instrument has a more favorable signal-to-noise ratio for a given background rate, but the typical D-T generator tube is limited to about 10^8 neutrons/s (compared to $2x10^{10}$ from a 7.5-mg sample of ^{252}Cf). Multiple tubes, or a larger accelerator, could be used, but the number (and cost) again would be prohibitive. Furthermore, the DDA technique uses thermal-neutron interrogation and for this energy the boron in the glass mixture is highly absorbent.

In summary, there are two complications in applying any of these techniques to the present case. The large background rate requires an unrealistically strong external neutron

source, and the signal would be dominated by isotopes other than ^{239}Pu. Even if a useful signal-to-noise ratio could be achieved, the signal would somehow still have to be interpreted to separate the small ^{239}Pu contribution from those of other isotopes and this would introduce sources of inaccuracy. Active neutron techniques simply are not useful for this type of waste.

C. Curium Balancing for High-Level Waste

1. Basis of the Technique

In the absence of a feasible direct assay technique, an indirect technique must be sought. The fission-product gamma rays and neutrons from ^{244}Cm that prohibit a direct technique are obvious candidates for indirect methods. With known correlations between the ^{244}Cm and the plutonium at measurement points, tracking the ^{244}Cm becomes equivalent to tracking the plutonium. The ratio of the two elements in the vitrification facility could be known from either an XRF or destructive analysis measurement on a sample taken at a point in the waste stream near the liquid or vitrified waste containers.

The liquid waste also contains the fines. The mass of plutonium carried annually by the fines is 0.27 to 1 kg in an 800-tU plant, which is 1 to 5% of the plutonium in the vitrified waste (Fig. 3). This could have a small impact on curium balancing if the ratio of plutonium to curium in the fines is greatly different from the ratio in the solution. Assume that the ratio in only the solution is measured just prior to vitrification and that there is no curium on the fines, for a worst case; the measured ratio is in error by the same fraction of plutonium that is on the fines and this is only 1 to 5%, at worst. The fines do not present a serious problem to curium balancing of the high-level liquid waste.

Neutrons from spontaneous fissions of ^{244}Cm are abundant (Table XIII) and sufficiently penetrating to escape from even the center of bulk vessels. Our Monte Carlo calculation showed that 93% of the ^{244}Cm fission neutrons starting uniformly throughout a container of vitrified waste will escape the vessel. (Capture in boron accounted for most of the other 7%.) Tracking essentially all the ^{244}Cm with these neutrons will also track the plutonium, assuming it can be assured that no further plutonium separation is possible.

Counting neutrons from spontaneous fissions of ^{244}Cm is complicated by interfering neutrons from (α,n) reactions, the high-intensity gamma rays, and the very high total neutron emission rate. The next section shows that the (α,n) background is not a problem. Some lead shielding would be needed to reduce the gamma-ray dose rate in the neutron detector, but this has been done before and the impact on the neutron counting would be small. Given the neutron production rates of Tables XIII and XIV, an inefficient detector is needed to avoid saturating the electronics. If 6.1×10^7 neutrons/s are released from a container (Tables XIII and XIV) and it is necessary to keep count rates below 2×10^6 counts/s, the detection efficiency must be no more than 3.3%. Such a low efficiency would normally make coincidence counting difficult and eliminate multiplicity counting, but with such a highly correlated count rate even multiplicity counting is still a possibility. The proper proportion of singles, doubles, and triples from a multiplicity count would give assurance that the neutrons originated from ^{244}Cm as expected (more on this in a later section).

2. Performance Estimate

The performances of coincidence and multiplicity instruments on ^{244}Cm were calculated under the assumption that neutrons released anywhere within a container have a high probability of escaping the container. A Monte Carlo calculation gave a probability of 93%, so the assumption is appropriate for this feasibility study.

a. Coincidence Counting

The first concern is the sensitivity of coincidence counting to neutrons from ^{244}Cm in the presence of a very high (α,n) background rate. In other words, how much ^{244}Cm must be present in a canister before it is detectable? For coincidence counting to be useful, the answer must be a small fraction of a gram.

The details of calculating a minimum detectable ^{244}Cm mass using coincidence counting are given in Appendix D. For the reasonable conditions in Table D-I the minimum detectable mass is in the microgram range and there is clearly no problem with sensitivity.

In borated glass the spontaneous fission yield is almost ten times larger than the (α,n) yield (Tables XIII and XIV) and the detection efficiency must be small enough to keep the total count rate at 2×10^6 counts/s or less. If the emission rate is $(5.4 \times 10^7 + 7.0 \times 10^6)$ neutrons/s, the maximum efficiency that can be used is 3.3%.

The Inventory Sample Counter (INVS) is a candidate for measuring the ^{244}Cm content in a small sample. The precision of this instrument for the waste is shown in Table XVI for a range of liquid volumes. The formulation in Appendix D was again used in these calculations.

TABLE XVI

PRECISION OF ^{244}Cm COINCIDENCE COUNTS WITH THE INVS COUNTER

^{244}Cm density = 9.96×10^{-6} g/cm^3, $G = 64$ μs, $\tau = 50$ μs, $P = 2$ μs, $\varepsilon = 40\%$, $t = 1000$ s.

Sample Volume (cm^3)	Relative Coincidence Rate Precision (%)
1	0.88
2	0.63
3	0.52
4	0.45
5	0.41

b. Multiplicity Counting

The original role for multiplicity counting was to determine, through neutron counting, three quantities: the mass of the spontaneously fissioning material, the neutron multiplication within the material, and the ratio of neutron production rates from (α,n) and spontaneous fission sources.[41,42] In this case the multiplication is known to be essentially unity because only small amounts of plutonium and curium are widely dispersed in a large container and furthermore the fission cross section for curium is low. (A Monte Carlo calculation gave a multiplication of 1.0016.) There are thus only two unknowns and normal coincidence counting is adequate to determine the spontaneous fission rate.

However, if multiplicity counting is feasible in this case, it could be used to verify that the isotope undergoing spontaneous fissioning is ^{244}Cm as expected, not some other isotope such as ^{252}Cf placed in the waste after the ^{244}Cm was removed along with the plutonium.

The equations in Appendix E show that the ratios of the first, second, and third moments are easily related to the measured singles, doubles, and triples rates. If the ratios have the expected values, the fissioning isotope is verified to be ^{244}Cm.

The relative precision of the singles count S, with a negligible background, is

$$\sigma_S / S = 1 / \sqrt{S} . \tag{10}$$

The relative precision of the doubles count D is calculated with Eq. (D-1) of Appendix D. The relative precision of a triples count T cannot be estimated by a closed-form equation; a computer program based on the work in Ref. 23 was used to calculate this and the other two relative precisions (Appendix F).

Table XVII shows these precisions for 5 g of ^{244}Cm as a function of the (α,n) production rate. (The parameters in the header are defined in Appendix D.) For the borated vitrified waste, the expected value of α is between 0.1 and 0.2; for the non-borated liquid waste, the value will be much smaller. The efficiency of 3% is taken from the preceding section to produce a total count rate from a canister of 2 x 10^6 counts/s, the maximum the electronics can handle.

TABLE XVII

RELATIVE PRECISIONS
OF MULTIPLICITY MEASUREMENTS

5 g ^{244}Cm, F = 3.97 x 10^6 fissions/(s•g), t = 1000 s,
<ν> = 2.72, <ν(ν-1)> = 5.99, <ν(ν-1)(ν-2)> = 10.6,
G = 32 μs, P = 2 μs, τ = 30 μs, ε = 3%.

		Relative Precisions (%)		
$S_{\alpha n}$ (n/s)	α	σ_S / S	σ_D / D	σ_T / T
1 x 10^3	1.85 x 10^{-5}	0.0025	1.56	34.6
1 x 10^4	1.85 x 10^{-4}	0.0025	1.56	34.6
1 x 10^5	1.85 x 10^{-3}	0.0024	1.57	37.4
1 x 10^6	1.85 x 10^{-2}	0.0024	1.59	38.3
1 x 10^7	1.85 x 10^{-1}	0.0023	1.85	44.5

The relative precision for the triples count when $S_{\alpha n}$ is about 10^6 n/s is rather poor at about 38%. However, the triples count would only be used to flag a substitution of an isotope such as ^{252}Cf for ^{244}Cm. The imprecision in the triples count at high count rates has long been recognized as a limit to the usefulness of multiplicity counting[41] and the problem is present here. So the doubles rate is used for the ^{244}Cm measurement in the present case of vitrified waste.

It is shown in Appendix E that ratios of multiplicities can be used to distinguish ^{244}Cm from other plausible neutron sources that might be used to match ^{244}Cm's large emission rate. However, if the curium and plutonium are removed from a slurry and the same amount of curium is added to the vitrified waste, neither multiplication nor any other passive neutron technique will know the difference. The best defense against this scenario is comprehensive knowledge of the plant's capabilities to manipulate curium.

D. Confirmation With Waste Signatures

Some signals could be used to simply confirm that the radioactive waste has indeed gone from the slurry into the vitrified form. The signals cannot be from plutonium itself but are again from ^{244}Cm and fission products and only indicate plutonium indirectly:

- total neutron count rate,
- neutron coincidence count rate,
- neutron multiplicity count rate, and
- fission product gamma rays.

1. Total Neutron Count Rate

The total neutron count rate found from the slurry will be increased by about 30% after vitrification because of the new (α,n) reactions with boron. A summary of total neutron production rates is given in Table XVIII for spent fuel waste in the form of UO_2 (as in an assembly) and in a vitrified canister. The spontaneous fission rate is roughly ten times larger than the (α,n) rates. A variation in the boron concentration would change the total neutron count rate even though the curium and plutonium amounts were constant. So to use the total count rate properly, the boron concentration should be measured and used to correct the measured count rate. The total count rate is also easily confused by a change in the background rate.

The total neutron count rate is not necessarily a simple signature in this case.

The coincidence and multiplicity count rates will be unaffected except by (a) the different neutron transports in the slurry and glass, and (b) the reduced precision with the glass because of higher accidental coincidence rates.

2. Coincidence Count Rate

Coincidence counting is quite feasible as a confirmation signature. Table XVII shows that the coincidence (doubles) precision is quite good. Coincidence count rates in the slurry and in the glass will not be the same, so it is necessary to learn how to correlate the two.

Coincidence rates are not seriously affected by the boron concentration; variations in the concentration are readily taken into account by the usual coincidence counting analysis.

The total neutron emission rate is enhanced by the (α,n) process, but this only increases the accidental coincidence rate and degrades the precision insignificantly (Table XVII).

The real coincidence rate is not affected by any neutron captures in boron. If a neutron has an energy low enough to be captured, it would not contribute to the real coincidence count rate anyway.

Coincidence counting does not provide a confirmation signature unique to ^{244}Cm but it clearly distinguishes between a fission source (such as ^{244}Cm) and more common non-fission sources (such as Am-Li or Po-Be).

3. Multiplicity Count Rates

Multiplicity measurements before and after vitrification could confirm the transfer of the ^{244}Cm from the slurry into the glass because the ratios of the three count rates (S, D, and T) from ^{244}Cm are unique to that isotope. Only the ratio of D and T may be of practical use because the singles rate is affected by the (α,n) rate. The main problem is the poor precision of the triples count rate because the same detector parameters must be used as with the curium assay; Table XVII shows that it may be only 35%, but this is probably sufficient. The scenario with plutonium and curium removal from the slurry and curium addition to the glass applies here again.

TABLE XVIII

NEUTRON YIELDS FOR TWO SPENT FUEL WASTE FORMS

Five years of cooling of the waste.
The (α,n) yields were calculated by W. B. Wilson, LANL.
One vitrified canister has waste from two PWR assemblies.
The masses are those in one nominal vitrified waste canister.

Isotope	Mass (g)	Spontaneous Fission Yields (neutrons/s)	(α,n) Yields (neutrons/s) UO_2	(α,n) Yields (neutrons/s) Vitrified Waste
^{238}Pu	0.3	7.77×10^{2}	4.02×10^{3}	2.56×10^{4}
^{239}Pu	17.5	3.82×10^{-1}	6.67×10^{2}	4.07×10^{3}
^{240}Pu	6.7	6.83×10^{3}	9.45×10^{2}	5.45×10^{3}
^{242}Pu	1.1	1.89×10^{3}	2.20×10^{0}	1.40×10^{1}
^{241}Am	295	3.48×10^{2}	7.94×10^{5}	4.69×10^{6}
^{244}Cm	5	5.37×10^{7}	3.87×10^{5}	2.27×10^{6}
Total		5.37×10^{7}	1.19×10^{6}	7.00×10^{6}

4. Gamma-Ray Measurements

The gamma-ray production rate from the fission products will change at the different steps of the vitrification process, so it has limited usefulness in verifying the plutonium.

Gamma rays from ^{244}Cm would readily confirm that this isotope is present, but these gamma rays are weak in energy and intensity compared to the gamma rays from fission products. After ^{244}Cm alpha decays, the resulting ^{240}Pu decays without emitting a gamma ray 76.7% of the time. The other 23.3% of the decays produce 42.8-keV gamma rays. A very small fraction of the decays (< 0.024%) give gamma rays with energies between 142 keV and 938 keV, but their intensities are very weak.

Even if the ^{244}Cm gamma rays were detectable amid those from fission products, self-attenuation within the large volume of waste would not allow any conclusions to be made about the existence of ^{244}Cm throughout more than a thin surface layer of the waste. The mass attenuation coefficient of a 40-keV gamma ray in oxygen is 0.218 cm^2/g and 0.592 cm^2/g in silicon (the two most abundant elements in the vitrified waste); the mean-free-path length in SiO_2 is thus about 4 mm, indicating the strong attenuation within the vitrified waste.

The gamma rays from fission products can be tracked more readily from slurry to glass because these are dominated by the 662-keV gamma ray from ^{137}Cs. But even at this energy the mean-free-path length in SiO_2 is only 4.7 cm, which is much smaller than the dimensions of slurry and glass containers. The value of fission products as a confirmatory signature is also questionable on the basis of the volatility of ^{137}Cs at high temperatures, resulting in a change in the ^{137}Cs-to-Pu ratio during the vitrification process.

E. Conclusions on High-Level Liquid Waste

♦ Direct nondestructive assays of plutonium in high-level liquid wastes are not possible.

Safeguarding waste quantities of plutonium in high-level liquid wastes (slurries and glass) poses important problems for nondestructive assays. The neutrons and gamma rays from plutonium are totally submerged in backgrounds of ^{244}Cm neutrons, (α,n) neutrons, and fission product gamma rays. The usual assay methods based on signals from plutonium simply cannot be applied.

A possible exception to this conclusion could arise. If the pipe carrying the liquid into the ceramic melter were sufficiently small so that K-edge densitometry could be done, and there is space around the pipe for the instrument near the melter, the density of plutonium could be determined. By combining this density with the flow rate or the volume of a holding tank, the mass of plutonium could be calculated. However, the problems of installing, operating, and maintaining a densitometer in a hot cell are probably prohibitive unless the hot cell has special design features for this instrument. A detailed verification of the plant's plumbing would still be the best way to ensure that all the liquid waste enters the vitrified canister.

♦ Curium balancing with neutron coincidence or multiplicity counting is feasible.

The indirect method of balancing the ^{244}Cm at different process stages takes advantage of the same strong neutron count rate from this isotope that makes it impossible to measure neutrons from plutonium. Coincidence counting will be very effective in separating neutrons from ^{244}Cm out of the background even though a low-efficiency detector (about 3%) must be used to keep the count rate below the upper limit set by the electronics. Such a detector can be rather simple in design, taking advantage of the rotation of the canister. Figure 7 shows a slab detector with only a few detector tubes behind gamma-ray shielding.

Neutron multiplicity counting has the same features as coincidence counting but goes further. Under these conditions it cannot be used to separate plutonium and curium signals (because the plutonium signal is too weak), but it can identify ^{244}Cm as the source of the neutrons and thereby strengthen the curium balancing process. The difference in cost for hardware and analysis between coincidence and multiplicity counting is minuscule.

The ratio of plutonium and curium would need to be verified at the input solution tank or at the glass melter with a frequency appropriate to the process throughput. The ratio could be established using destructive analysis techniques or a combination of destructive analysis for the plutonium and nondestructive assay for the curium. As shown in Table XVII, a 2-cm^3 input-solution sample would provide a precision of 0.6% for the ^{244}Cm mass after a 1000-s count.

The "fines" carry some plutonium and the plutonium-to-curium ratio for fines is not established. However, the quantity of plutonium on the fines is too small to affect the results of curium balancing by more than 5% in even the worst case.

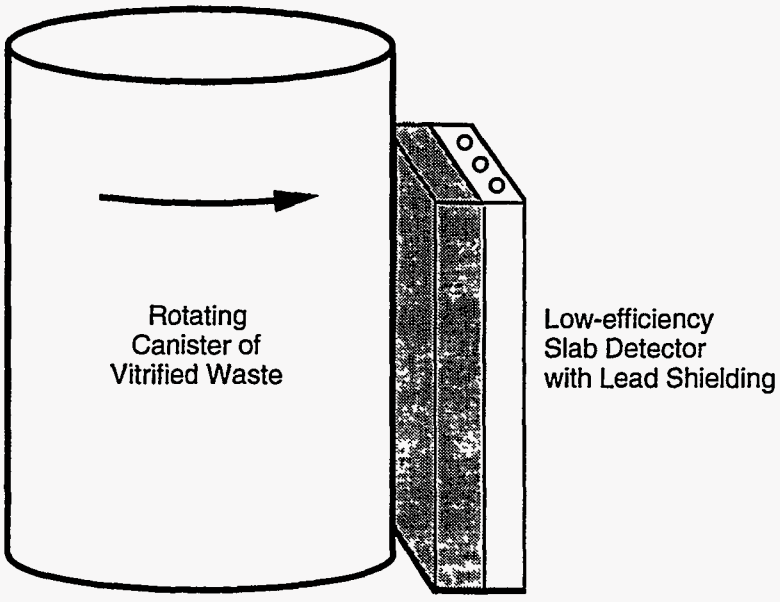

Fig. 7. A slab neutron detector with low efficiency is near a rotating canister of vitrified waste to measure the neutron emission rate. The detector has a front surface of metal (such as lead) to attenuate the gamma rays from the canister and reduce the signal production rate from gamma-ray interactions in the detector.

♦ Confirmatory signatures could be based on neutron coincidence or multiplicity counting.

If only a signature confirmation is considered adequate for safeguarding the waste, the best signal is still the count rate of neutrons from ^{244}Cm. The neutrons readily escape from the entire volume of the containers and emission rates are very high. The total count rate depends somewhat on the boron concentration in the glass, so coincidence or multiplicity counting should be used.

Gamma rays from fission products are readily measured, but only those originating very near a container's surface will be detectable; the interior of a canister is not probed. Fission products are more readily separated from the plutonium than is curium, further weakening their usefulness as a signature for the presence of plutonium.

The schematic in Fig. 8 indicates the flow of the liquid waste from a slurry tank into a canister becoming vitrified along the way. Possible measurement points are indicated for both destructive and nondestructive assays.

IV. DIVERSION SCENARIOS AND CURIUM BALANCING

What plausible schemes might be attempted to divert plutonium from a reprocessing plant's waste streams and how might curium balancing detect the attempts? Most of the potential diversion scenarios involve the removal of materials with plutonium and curium from items such as fuel pins and solutions, and replacing it by similar materials containing only the curium. The separated curium would come from the back end of the separation process. Some specific cases are given in Appendix G.

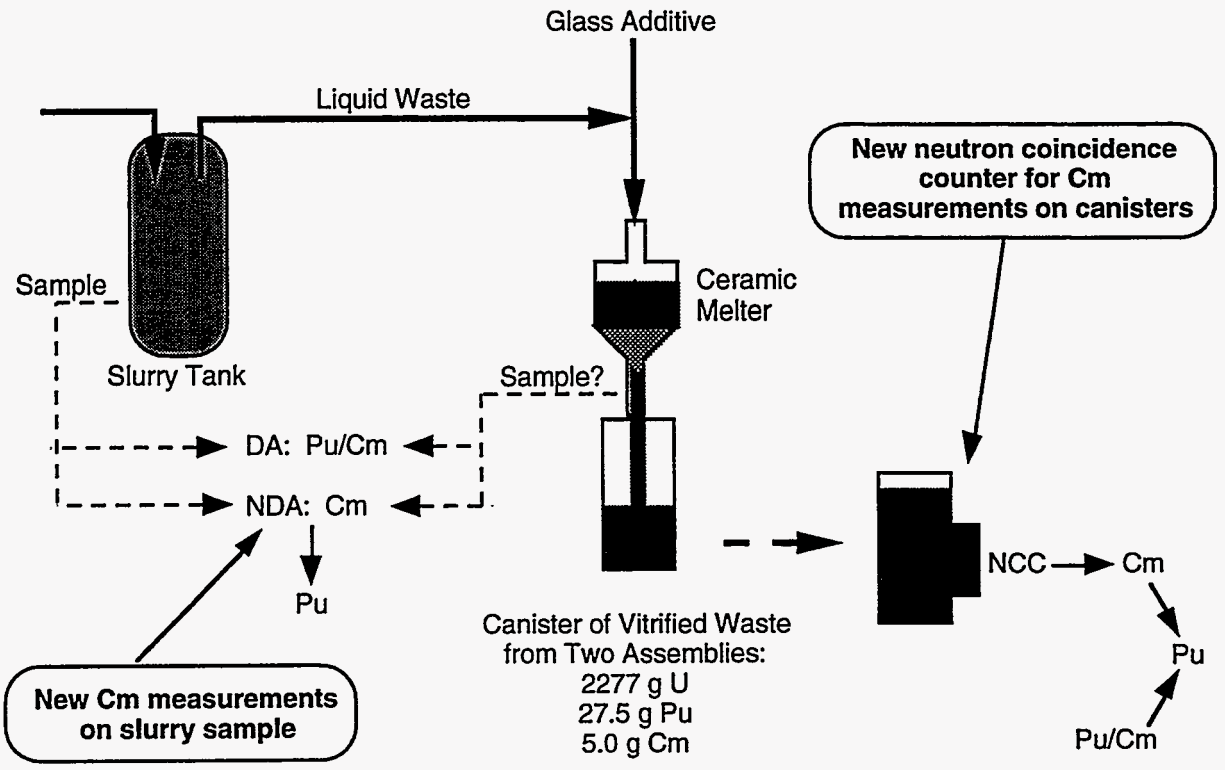

Fig. 8. This simplified flow diagram of the vitrification process shows sampling of the slurry tank to determine the ratio of plutonium to curium using a destructive assay process and the nondestructive assay to determine the curium content; from these results the plutonium content is calculated. A sample of the vitrified waste may be assayed similarly, although the sampling process is more complex. The canister of vitrified waste is measured with a neutron coincidence counter (NCC) for its ^{244}Cm content, from which the plutonium content is deduced with the help of the plutonium-to-curium ratio.

V. CONCLUSIONS ON CURIUM BALANCING

A. The Rationale for Curium Balancing

The waste streams of an efficient, large-scale reprocessing plant pass 7 to 10 kg of plutonium out of the plant each year while 54.5 t of plutonium are recovered in the separations facility. Some of the waste plutonium is fixed in the leached hulls and cannot be dissolved; a smaller amount may remain in a thin film on the surface of the pieces of hulls. Plutonium heels in solution after the separation and on the surface of the "fines" (undissolved solids) add another 5 to 6 kg of plutonium each year in 800 canisters of vitrified high-level liquid waste. Curium balancing is an approach to applying safeguards to the materials in these waste streams.

With these wastes it is not possible to perform a nondestructive assay based on signals from plutonium itself. The gamma rays from fission products and the neutrons from curium are too intense for gamma rays or neutrons from plutonium to be detected. Furthermore, gamma rays from the waste materials or from a transmission source are too highly attenuated by the contents of the large, dense containers to be reliable indicators of the complete contents. Fission products are not a reliable indicator of plutonium because they are too readily separable (unlike curium).

However, these waste containers are fairly transparent to neutrons (except for thermal neutrons in the case of the vitrified waste) and the ^{244}Cm is a prolific emitter of high-energy neutrons from spontaneous fissions. The long half life (18.1 yr) of ^{244}Cm has long made it a practical indicator for fuel assembly burnup and now gives the curium contents in the waste

containers. Curium can be measured at key points in a reprocessing plant to monitor its flow through the head-end and the waste streams, or plutonium can be correlated with curium through nondestructive and destructive measurements and the quantities of plutonium can be inferred at the key points.

B. Reprocessing Plant's Head-End

Plutonium and curium enter the head-end in spent fuel assemblies. Passive neutron counts should be taken of each assembly to establish the amount of curium (and plutonium, through correlations from calculations and operating experience). Such counts are already used to check the declared exposures for criticality control. We suggest that neutron coincidence counting be used while scanning the active length of each assembly. This makes it possible to correct for the neutron multiplication and essentially eliminates interference from any other neutron source that may be accidentally or deliberately nearby.

Almost all of the plutonium and curium pass through the accountability tank where excellent safeguards measurements for plutonium are already made. We propose that the ratio of plutonium to curium also be measured nondestructively using a Hybrid X-ray Densitometer (which is already routinely used for the plutonium concentration); the curium concentration can be best measured from the same sample with a passive neutron count in an INVS. Without this plutonium-to-curium ratio there can be no correlation between curium and plutonium for the leached hull measurement, but curium can still be balanced alone. The new INVS measurement would allow a safeguards comparison of the curium amounts in the spent fuel assemblies and the accountability tank; a diversion of material between these two key points would be detected only days or weeks after it has occurred.

It is necessary to improve the conditions under which measurements of leached hulls are made. If a plant cannot allow space for a high-quality detector where hulls are loaded into a drum, then there could be space elsewhere in the facility. The accuracy of any passive-active neutron instrument for a drum will be hindered unless the instrument has detectors that completely surround the assay chamber; rotating the drum during a measurement would improve the active measurement, but if detectors are not above and below the drum, the accuracy will be well below the state of the art. Drums should be measured before they are filled with water or concrete to allow the fission neutrons to escape readily from the entire volume of each drum. Compaction of the drums may actually be a safeguards advantage because the assay accuracy is usually improved by having a smaller object.

C. Vitrified High-Level Liquid Waste

Plutonium and curium enter the vitrification facility in a waste stream from the separations facility, along with the fission products. The plutonium-to-curium ratio is now much smaller than at the head-end.

The curium concentration in the incoming slurry could be measured by neutron coincidence counting on a small sample. If a destructive measurement would give the plutonium-to-curium ratio, the curium concentration could then be applied to give the plutonium concentration. The same neutron data could be given a multiplicity analysis to confirm that the neutron source is indeed curium and not a substitute.

Another neutron coincidence count on a vitrified waste canister would provide its curium mass. This concentration would be greater than with the slurry because of evaporative losses in a calciner and the glass melter. This neutron detector would require thick gamma-ray shielding between it and the canisters, but the shielding would not interfere with the neutron count.

Assurance that the curium and plutonium (by correlation) in the expected amounts are contained in the vitrified waste would be given by these measurements before and after vitrification.

APPENDIX A

THE ^{244}Cm NEUTRON EMISSION RATE FROM A PWR ASSEMBLY

To evaluate Eqs. (6) through (8) it is necessary to know the ^{244}Cm neutron emission rate from the portion of a PWR assembly that is within the detector head; this is the S_0 of Eq. (7). This rate has been estimated from information in Chapter 18 of Ref. 1 in the following manner.

(a) There are 9.8×10^4 kg of fuel in the 193 PWR assemblies in a reactor core.
(b) Each assembly therefore has 508 kg of UO_2 fuel.
(c) Each assembly therefore has 447.5 kg, or 0.4475 t, of uranium.
(d) After an exposure of 33 GWd/tU, there are 18.3 g of ^{244}Cm per ton of U.
(e) Each assembly therefore has 8.19 g ^{244}Cm.
(f) The active length of an assembly is 366 cm.
(g) Each assembly therefore has an average of 0.0224 g of ^{244}Cm per centimeter of active length.
(h) One gram of ^{244}Cm has 3.97×10^6 fissions/s.
(i) Each centimeter of the assembly therefore has 8.89×10^4 fissions/s.
(j) One gram of ^{244}Cm emits 1.08×10^7 n/s from spontaneous fissions [and 7.73×10^4 n/s from (α,n) reaction in oxides].
(k) A centimeter of the assembly has an average emission rate of 2.42×10^5 n/s.
(l) For a length of 30 cm within the detector head, the emission rate is 7.26×10^6 n/s.

For such an assembly the value of S_0 is 7.26×10^6 neutrons/s. This was used in calculating the values in Table IV.

APPENDIX B

INVS COUNTER DETECTION LIMIT FOR ^{244}Cm

The detection limit for ^{244}Cm, with coincidence counting, is a special case of Eq. (6). The relative uncertainty σ_R/R is set to an assigned value (generally 1/3 or 1/4, to make it more than 99% likely that a signal is not merely a statistical fluctuation in the background[43]), and R and A are expressed in terms of the mass of ^{244}Cm. The equation is then solved for the mass, which is the detection limit (or sensitivity).

The real coincidence rate is shown in Eq. (7) and is abbreviated here as

$$R = F\, m_{Cm244}\,, \tag{B-1}$$

where $F = (S_o/m_{Cm244})\, \varepsilon^2\, e^{-P/\tau}\, (1 - e^{-G/\tau})\, \langle \nu(\nu - 1)/2 \rangle$.

The accidental coincidence rate depends on the square of the mass:

$$A = G\, T^2 = G\, (r\, \varepsilon\, m_{Cm244})^2\,, \tag{B-2}$$

where r is the total neutron emission rate per gram of ^{244}Cm and ε is the detection efficiency. Spontaneous fissions contribute 1.08×10^7 neutrons/(s•g) to r, while (α,n) reactions in an oxide add a negligible 7.73×10^4 neutrons/(s•g).

Using these new expressions in Eq. (6) gives this quadratic equation for the smallest m_{Cm244} that will produce the given precision (σ_R/R).

$$[(\sigma_R/R)^2\, F^2\, t - 2\, G\, r^2\, \varepsilon^2]\, m_{Cm244}^2 - F\, m_{Cm244} - 2B = 0\,. \tag{B-3}$$

This m_{Cm244} is the detection limit of the INVS counter for ^{244}Cm.

APPENDIX C

RELATIVE COINCIDENCE RATES FROM PLUTONIUM AND CURIUM

The coincidence rate r for neutrons from an element is (Ref. 1, page 470)

$$r = \varepsilon^2 e^{-P/\tau} (1 - e^{-G/\tau}) \sum_i [m_i F_i \langle \nu(\nu-1) \rangle_i / 2] , \qquad \text{(C-1)}$$

where

m_i = mass of the ith isotope of the element (in grams),
F_i = specific fission rate of the ith isotope of the element [fissions/(s•g)],
ε = detection efficiency,
P = predelay time (in seconds),
τ = die-away time (in seconds),
G = coincidence gate width (in seconds),
$\langle \nu(\nu-1) \rangle_i$ = average coincidence multiplicity of neutrons from the ith isotope.

Only a single isotope of curium (^{244}Cm) is present, but for plutonium, the three isotopes with even atomic weights must all be considered. Table C-I gives information on the important isotopes.

TABLE C-I

PERTINENT FISSION PROPERTIES OF SIGNIFICANT ISOTOPES

Data from Ref. 1, pp. 339 and 342 and Ref. 3, page 15.

Isotope	F fissions/(s•g)	$\langle \nu(\nu-1) \rangle$
^{238}Pu	1172	3.957
^{240}Pu	472	3.825
^{242}Pu	800	3.794
^{244}Cm	3.97×10^6	5.990

The ratio of coincidence rates is

$$r_{Pu} / r_{Cm} = [\sum_i m_i F_i \langle \nu(\nu-1) \rangle_i]_{Pu} / [m F \langle \nu(\nu-1) \rangle]_{Cm} , \qquad \text{(C-2)}$$

which is independent of the parameters in Eq. (C-1) that describe the detector's operation (assuming the detection efficiencies for the two sets of neutrons are nearly equal). From Table XI, the ratio of plutonium and curium masses is 5.57 and the plutonium isotope mixture is taken to be (by weight percentages) 1.574% ^{238}Pu, 24.98% ^{240}Pu, and 5.545% ^{242}Pu.

With these data the ratio of the coincidence rates for the two elements is

$$r_{Pu} / r_{Cm} = 0.0001623 . \qquad \text{(C-3)}$$

APPENDIX D

MINIMUM DETECTABLE ^{244}Cm MASS IN VITRIFIED WASTE FROM COINCIDENCE COUNTING

Coincidence counting is feasible only if the relative precision of the real coincidence rate is sufficiently small (generally less than 33.33%); this requires having a certain minimum mass of ^{244}Cm present. The vitrified waste will have a higher minimum detectable mass than the liquid waste (Appendix B) because of the higher (α,n) production rate, so the vitrified case is considered here. Neutron producers other than ^{244}Cm are not important.

The analytical analysis begins with the statement that the relative precision (Ref. 1, page 477) is to be less than some maximum value:

$$\sigma_R/R = \sqrt{(R + 2A)}/R \leq (\sigma_R/R)_{max}, \tag{D-1}$$

where

R = real coincidence count,
σ_R = precision (or uncertainty) of R,
σ_R/R = relative precision of R, and
A = accidental coincidence count.

The real coincidence count depends on the ^{244}Cm mass and properties of the detector in this manner (Ref. 1, page 470):

$$R = F\, m\, \varepsilon^2\, e^{-P/\tau}\, (1 - e^{-G/\tau})\, \langle \nu(\nu-1) \rangle\, t\, /\, 2 = \beta\, t\, m\,, \tag{D-2}$$

where

F = specific fission rate [fissions/(s•g)],
m = mass of ^{244}Cm (grams),
ε = detector efficiency,
P = predelay time (in seconds),
τ = die-away time (in seconds),
G = coincidence gate width (in seconds),
$\langle \nu(\nu-1) \rangle$ = average coincidence multiplicity, and
t = count time (in seconds).

Under normal circumstances a high detector efficiency (20% or more) is desirable, but in this case a low value (such as 3%) may be needed to keep the total count rate within the limits of the electronics (which is about 2×10^6 counts/s).

The accidental coincidence count varies with the total count (Ref. 1, page 469):

$$A/t = G\, (T/t)^2, \tag{D-3}$$

where T is the total neutron count during time t. The total count originates from spontaneous fissions and (α,n) reactions:

$$T = T_{SF} + T_{\alpha n} = \varepsilon\, m\, r\, t + \varepsilon\, m\, s_{\alpha n}\, t\,, \tag{D-4}$$

where r is the neutron emission rate from spontaneous fissions in one gram of ^{244}Cm [neutrons/(s•g)] and $s_{\alpha n}$ is the specific (α,n) source production rate (neutrons/s/g ^{244}Cm). This latter rate is taken from Table XIV to be 7.00×10^6 neutrons/[s•(5 g ^{244}Cm)] or 1.40×10^6 neutrons/[s•(g ^{244}Cm)].

The minimum detectable mass of ^{244}Cm can be found from these expressions with the poorly known (α,n) production rate as a parameter. Eq. (B-1) is first written as a quadratic equation for R.

$$(\sigma_R/R)^2_{max} R^2 - R - 2A \geq 0 . \qquad \text{(D-5)}$$

Equations (D-2)-(D-4) are next used to expand R and A in terms of the ^{244}Cm mass m. The equality in Eq. (D-5) is taken because the minimum detectable ^{244}Cm mass is being sought.

$$m^2 [(\sigma_R/R)^2_{max} (\beta t)^2 - 2G(r + s_{\alpha n})^2 \varepsilon^2 t] - m\beta t = 0 . \qquad \text{(D-6)}$$

The minimum detectable ^{244}Cm mass is thus

$$m = \beta t / [(\sigma_R/R)^2_{max} (\beta t)^2 - 2G(r + s_{\alpha n})^2 \varepsilon^2 t] . \qquad \text{(D-7)}$$

The mass must be positive and this imposes a mathematical upper limit on the efficiency in Eq. (D-8). But in practice there is no limit.

Equation (D-7) has been evaluated for two sets of plausible parameters that bracket realistic detection limits and count times. The efficiency is only 3% so that up to 5 g of ^{244}Cm can be assayed in the counter. The results are shown in Table D-I and are small fractions of a milligram.

TABLE D-I

MINIMUM DETECTABLE ^{244}Cm MASS USING COINCIDENCE COUNTING

$F = 3.97 \times 10^6$ fissions/(s•g)
$r = 1.08 \times 10^7$ spontaneous fission neutrons/(s•g)
$\langle \nu(\nu-1) \rangle = 5.99$
$G = 32$ μs, $P = 2$ μ, $\tau = 30$ μs, $\varepsilon = 3\%$.

σ_R/R (%)	t (s)	m (μg)
33.3	1000	1.4
10.0	400	40

APPENDIX E

IDENTIFYING ^{244}Cm WITH MULTIPLICITY COUNTING

A multiplicity count results in three count rates: singles (S), doubles (D), and triples (T). The singles and doubles rates are the same as the total and coincidence rates used in coincidence counting. The triples rate is an additional measurement and makes it possible to determine three unknowns instead of only the two with coincidence counting.

The three expected count rates when multiplication is unity are these:[3,23]

$$S = m\,F\,(1 + \alpha)\,\varepsilon\,\nu_{s1}\,, \tag{E-1}$$

$$D = (m\,F\,\varepsilon^2 f_d\,\nu_{s2})\,/\,2\,, \text{ and} \tag{E-2}$$

$$T = (m\,F\,\varepsilon^3 f_t\,\nu_{s3})\,/\,3\,, \tag{E-3}$$

where

m = mass of fissioning isotope (in grams),
F = fission yield [in counts/(s•g)],
α = ratio of neutrons produced by (α,n) reactions and spontaneous fissions,
ε = detector efficiency,
f_d = doubles gate fraction = $e^{-P/\tau}\,(1 - e^{-G/\tau})$,
f_t = triples gate fraction $\approx f_d^2$,
ν_{s1} = first moment of the neutron distribution,
ν_{s2} = second moment of the neutron distribution, and
ν_{s3} = third moment of the neutron distribution.

These equations would normally use known values of the moments to find m, the multiplication M, and α. In waste, the multiplication is unity and is thus not even shown in Eqs. (E-1)-(E-3). This means that coincidence counting (involving only S and D) can be used to find m and α. Then ratios of S, D, and T rates can be used to check that the fissioning isotope is really ^{244}Cm and not another isotope such as ^{252}Cf that has been implanted after removing the ^{244}Cm along with the plutonium from the waste.

There are three ratios of counts from which three ratios of moments can be calculated:

$$\nu_{s2} / \nu_{s1} = 2\,(D/S)\,(1 + \alpha)\,/\,(\varepsilon f_d)\,, \tag{E-4}$$

$$\nu_{s3} / \nu_{s1} = 3\,(T/S)\,(1 + \alpha)\,/\,(\varepsilon^2 f_t)\,, \text{ and} \tag{E-5}$$

$$\nu_{s3} / \nu_{s2} = (3/2)\,(T/D)\,f_d\,/\,(\varepsilon f_t)\,. \tag{E-6}$$

These three ratios should match those known for ^{244}Cm (taken from Ref. 3, page 15):

$$\nu_{s2} / \nu_{s1} = 5.99 / 2.72 = 2.20\,, \tag{E-7}$$

$$\nu_{s3} / \nu_{s1} = 10.6 / 2.72 = 3.90\,, \text{ and} \tag{E-8}$$

$$\nu_{s3} / \nu_{s2} = 10.6 / 5.99 = 1.77\,. \tag{E-9}$$

However, ν_{s1} and ν_{s2} are used in the coincidence analysis to find m and α, so only the last two ratios involving ν_{s3} can be applied as independent indicators of ^{244}Cm.

The ratios in Eqs. (E-4) to (E-6) must be determined with sufficient precision and be significantly different from those of other isotopes to be a reliable indicator of ^{244}Cm. Precisions of the measured moments can be estimated from Eqs. (E-1) to (E-3) and the precisions of the counts in Table XVIII.

The square of the relative precision for the first ratio, $\nu_{s2} / \nu_{s1} \equiv \rho_1$, is

$$(\sigma_{\rho 1}/\rho_1)^2 = (\sigma_S/S)^2 + (\sigma_D/D)^2 + [\sigma_\alpha/(1+\alpha)]^2 . \qquad \text{(E-10)}$$

Similarly the squares of the relative precisions for the other two ratios are

$$(\sigma_{\rho 2}/\rho_2)^2 = (\sigma_S/S)^2 + (\sigma_T/T)^2 + [\sigma_\alpha/(1+\alpha)]^2, \text{ and} \qquad \text{(E-11)}$$

$$(\sigma_{\rho 3}/\rho_3)^2 = (\sigma_D/D)^2 + (\sigma_T/T)^2 . \qquad \text{(E-12)}$$

From Table XVII (at the higher $S_{\alpha n}$ values) it is seen that (σ_S/S) is negligibly small, (σ_D/D) is about 1.6%, and (σ_T/T) is about 35%. The α particles and the fission neutrons both originate from ^{244}Cm at known rates, so the value $[\sigma_\alpha/(1+\alpha)]$ is as well known as the density of boron in the glass [the most important target for the (α,n) reaction]; if the boron density is known to within 5%, the value of α is also known with an uncertainty of 5%. Values of α itself are shown in Table XVII; a likely value is close to 0.02. The relative precisions of the three ratios are thus about 5%, 35%, and 35%, in the sequence of Eqs. (E-10) through (E-12). Only the last two ratios are useful, as noted earlier, and they have the poor precisions related to the triples counts.

Ratios of multiplicities for ^{244}Cm, ^{240}Pu, and ^{252}Cf are compared in Table E-I.

TABLE E-I

RATIOS OF MULTIPLICITIES

Multiplicities from Refs. 1 and 3

	^{240}Pu	^{244}Cm	^{252}Cf
ν_{s2} / ν_{s1}	1.774	2.202	3.184
ν_{s3} / ν_{s1}	2.475	3.897	8.467
ν_{s3} / ν_{s2}	1.395	1.770	2.659

With a 35% uncertainty on the ^{244}Cm ratios, it is impossible to distinguish ^{244}Cm from ^{240}Pu, but the plutonium amount would never be increased to simulate the normal plutonium and curium mixture when the objective is to remove the plutonium.

Can the plutonium and curium be removed and another neutron emitter implanted to mimic the ^{244}Cm? Fortunately, the list of possible substitutes for the ^{244}Cm in waste is a short one because a substitute must match the large neutron production rate of ^{244}Cm. Neutrons from (α,n) reactions with a simple alpha emitter would be easily revealed by the lack of doubles (or

coincidence) and triples counts. Few spontaneously fissioning isotopes have neutron intensities that match thos from ^{244}Cm.

Emission rates from ^{252}Cf can readily match those of the ^{244}Cm; it takes only 0.5 mg of ^{252}Cf to achieve the same emission rate as 100 g of ^{244}Cm. But as seen above the ratios of moments could be used to detect a substitution of ^{252}Cf for ^{244}Cm. Furthermore, the acquisition of this much ^{252}Cf would attract attention because the manufacture is difficult (requiring a special high-flux reactor), the supply is limited, and the number of suppliers is very small. At current prices, the cost for 0.5 mg is about $50K, and this would be used for only one canister.

Curium-242 has a neutron production rate and multiplicities very close to those from ^{244}Cm, but its short half-life (163 days) means it has to be continually obtained from spent fuel assemblies with short cooling times, and it would be chemically inseparable from ^{244}Cm. So there is no reason to expect ^{242}Cm to be used as a substitute.

The isotope ^{249}Bk is another neutron emitter that is much more intense than average, but it would take 10 kg to match the neutron emission rate from 100 g of ^{244}Cm. Berkelium-249 can only reach the needed intensity in unreasonably large quantities. Other neutron sources are even less practical than ^{249}Bk.

Perhaps a scenario that would be the hardest to detect is to separate the plutonium and curium from the slurry and return curium from an earlier batch into the vitrification mixture. The multiplication ratios would be perfect because the neutrons are still coming from ^{244}Cm. Detection of this scenario would have to rely on finding the additional separation and handling facilities within the plant. The plutonium remaining in the waste is in a rather insoluble form, having passed through the normal separation process in the reprocessing plant, so any additional separation would require extensive equipment.

APPENDIX F

FIGURE OF MERIT CODE INPUT DATA

A "figure of merit" computer code was written by Norbert Ensslin of Los Alamos National Laboratory for multiplicity counting. The general topic is described in Ref. 23. Among the output of the code are the relative precisions (in percent) of the single, doubles, and triples counts. This code was used to generate the relative precisions given in Table XVII. The input data supplied to the code for these calculations is given in this appendix.

The code's name is FOM15. It has a creation date and time (in DOS USA format) of 7-12-93, 4:01p. Here is an annotated example of the input data given to FOM15:

2 [type of detector = thermal];
32 [G = gate width in microseconds];
100 [counting deadtime in nanoseconds];
03 [ε = detection efficiency as a fraction, not a percent];
1000 [count time in seconds];
0 [background trigger rate];
0.009203 [E1; see note 1];
0.003765 [E2; see note 1];
52941 [$^{240}Pu_{eff}$ mass (g) equivalent to 5 g of ^{244}Cm; see note 2];
1 [neutron multiplication; one is equivalent to no multiplication];
0.185 [α, the ratio of (α,n) to spontaneous fission yields; see note 3].

Note 1. $E1 = \varepsilon f_d/2$. f_d and other terms in this appendix are defined in Appendix E.
Other values used in calculating f_d were P = 2 ns and τ = 30 μs, giving $f_d = 0.6135$. In this case, E1 = 0.3068ε.
$E2 = \varepsilon f_t/3 \approx \varepsilon f_d^2/3$. In this case, E2 = 0.1255ε.
The efficiency chosen for Table XVIII is 3%, or $\varepsilon = 0.03$. For this efficiency, E1 = 0.009203 and E2 = 0.003765.

Note 2. One gram of ^{244}Cm generates 1.08×10^7 n/s from spontaneous fissions; five grams generates 5.40×10^7 n/s. One gram of $^{240}Pu_{eff}$ generates only 1020 n/s, so it takes $(5.40 \times 10^7)/(1020) = 52941$ g of $^{240}Pu_{eff}$ to equal the generation rate of 5 g of ^{244}Cm.

Note 3. It is assumed that the spontaneous fission yield is fixed at 5.40×10^7 n/s (from 5 g of ^{244}Cm). The value of α is then $S_{\alpha n} / 5.40 \times 10^7$. In the example in the data list above, the value of $S_{\alpha n}$ was taken to be 10^7 n/s, giving $\alpha = 0.185$.

APPENDIX G

DIVERSION SCENARIOS AND CURIUM BALANCING

Diversion scenarios can be imagined at many stages through a plant. The list below goes through the head-end and the waste treatment stages.

• A fraction of the pins in an assembly could be removed prior to shearing without affecting the neutron count rate by an amount outside the acceptable statistical fluctuation. Containment and surveillance would detect such gross operations on assemblies and the mechanical problem of removing only some of the pins is solved, the yield of plutonium from an assembly would be rather meager. Neutron count rates from assemblies with nearly identical histories differ from each other by only about 1%, so for a 15x15 PWR assembly with 204 pins even a 3% drop in the coincidence count rate, caused by removing about 10 pins (holding about 165 g of plutonium) would appear suspicious. A separate reprocessing line or facility would be needed for the diverted pins, further complicating the diverter's task.

• Curium-244 could be attached to assemblies in a random manner to expand the uncertainty of the measurements on intact assemblies. This might be used to conceal the removal of a larger number of pins from assemblies prior to shearing. But an increased uncertainty, especially among assemblies with very similar irradiation histories, would not appear normal, and adding ^{244}Cm to many assemblies would make the count rates from assemblies with missing pins even further from the expected values. If correct operation of the instrument has been verified, the only remaining cause would be a neutron-producing additive to the assemblies.

• The correct amounts of ^{244}Cm might be added to assemblies after removing some pins (despite the containment and surveillance and the mechanical difficulties). The neutron counter before shearing would not detect a problem, if this diversion were done properly. But the ratio of plutonium to curium in the dissolver-tank sample would be inconsistent with similar fuel assemblies. A quick NDA check of the sample using a hybrid X-ray densitometer and the INVS counter could screen such samples for more precise mass spectrometry examinations. How small a change in this ratio would be detectable nondestructively?

The accountability tank might be normally filled, sampled, and discharged once a day with about 13,000 liters of dissolver solution with 250 g-U/ℓ, or about 2.5 g-Pu/ℓ and 0.0024 g-Cm/ℓ; the amount of plutonium normally in a filled tank is thus about 33 kg (from 9 or 10 PWR assemblies). The uncertainty in the plutonium-to-curium ratio from a hybrid X-ray densitometer is perhaps 5%, which corresponds to 1.6 kg of plutonium in a single tank filling.

However, by using the X-ray instrument to give only the plutonium concentration and then using the INVS neutron measurement for the curium concentration, the uncertainty in the ratio is reduced to about 0.9% and the amount of plutonium that could be removed from a single filling of the accountability tank with little suspicion is reduced to 360 g. A curium measurement based on neutron counting makes the detection of a diversion much more likely and timely. A destructive analysis (e.g., mass spectrometry) for curium would be even more definitive, but would take longer. The combination of the X-ray and neutron measurements could be used to screen for accountability samples that are suspicious and deserve a mass spectrometry examination.

• Removing the small, dilute waste amounts of plutonium from leached hulls requires a technique that is more effective than the plant's dissolver process. But assuming the plutonium could be removed, the curium either stays with the hulls or could be replaced. In principle the passive neutron count could appear normal. An active neutron assay (with a shuffler or a DDA) would be suspiciously low unless a proper amount of ^{235}U is introduced to simulate the missing ^{239}Pu. This is an unattractive diversion path because it requires an obscure recovery process and yields, at most, only 5 to 10 g of plutonium from each drum.

• An inefficiency could be introduced into the head-end so that more plutonium is carried by the leached hulls than normal. Not rinsing the hulls would be one simple way to increase the plutonium carried into a drum. The time in the dissolver tank could also be shortened. Draining some of the dissolver solution directly into drums would be another way to increase the plutonium contents in drums in an attractive form. However, until the dissolver solution has gone through a separations facility, the curium travels with it. The passive count rate of neutrons from curium will increase linearly with the extra solution in the drum; the active assay for the plutonium will also give a larger result. The amount that could be added to a drum without arousing suspicion would be small (probably less than 10%); the better the quality of this instrument, the smaller the amount that could be successfully diverted.

• Dissolver solution could be diverted from the accountability tank and the curium could be replaced. The curium added could come from waste solutions generated by the plutonium separation process. The detection of this scheme (apart from noting the additional plumbing and tanks involved) would depend on the measurement of the plutonium-to-curium ratio in samples from the accountability tank, as discussed in the third scenario.

• A diversion in the vitrification facility would be mostly easily done with the slurry before the melter. The curium would have to be taken with the plutonium. A measure of the curium concentrations in the slurry (using an INVS neutron counter) compared to the curium concentrations in the canisters after vitrification (using a simple slab neutron detector) could detect a diversion. The concentrations would be converted into masses of curium batches before and after vitrification, allowing for the normal change in concentration caused by the vitrification process. The accuracy of the comparison could be a few percent, depending on the calibration standards for the canisters and the understanding of the changes introduced by the vitrification process. With only about 27.5 g of plutonium in a canister and 30 canisters for a batch of 60 assemblies, the most plutonium that could be diverted is 850 g during the week of reprocessing. A passive neutron count would show a reduced amount of curium within a canister about a day after it was filled, and the curium balance for the batch would reveal the full scope of a diversion within a day or two of the end of the batch operation (after the last canister has cooled and is welded shut).

The removal of significant quantities of plutonium from the waste streams requires some patience and determination on the part of a diverter. Monitoring the curium amounts in the waste streams would place additional severe restrictions on a potential diverter's scheme.

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