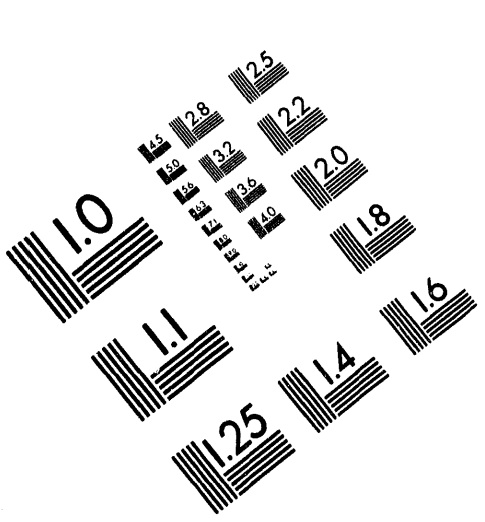

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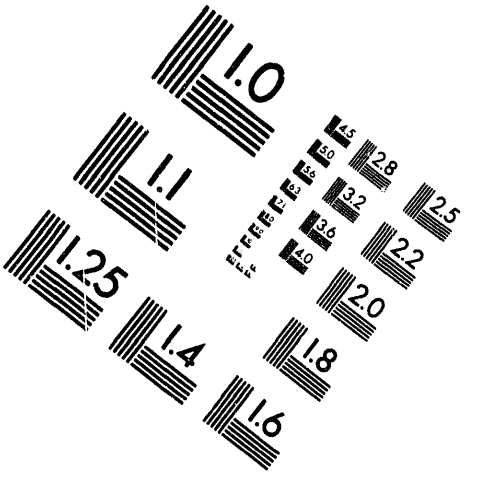

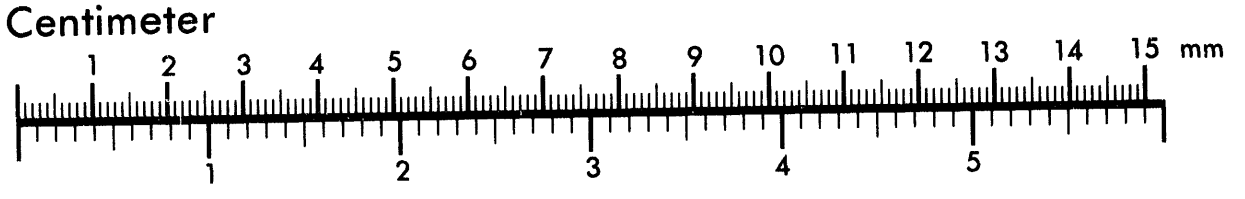

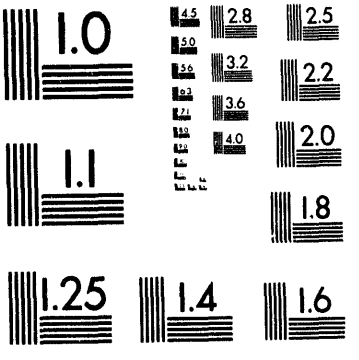

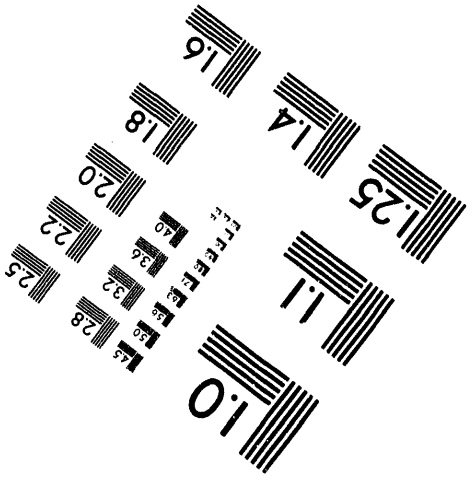

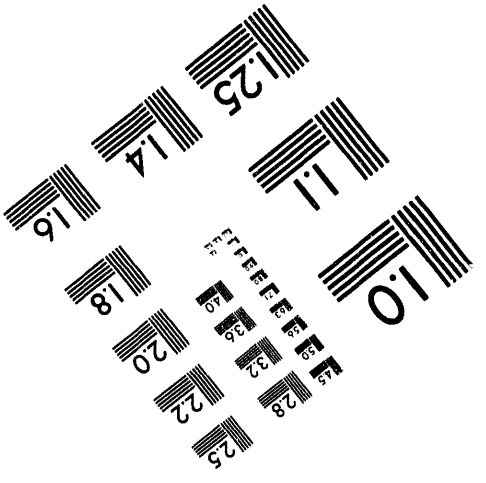

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The Use of Curium Neutrons to Verify Plutonium in Spent Fuel and Reprocessing Wastes

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THE USE OF CURIUM NEUTRONS TO VERIFY PLUTONIUM IN SPENT FUEL AND REPROCESSING WASTES

by

N. Miura and H. O. Menlove

ABSTRACT

For safeguards verification of spent fuel, leached hulls, and reprocessing wastes, it is necessary to determine the plutonium content in these items. We have evaluated the use of passive neutron multiplicity counting to determine the plutonium content directly and also to measure the $^{240}Pu/^{244}Cm$ ratio for the indirect verification of the plutonium. Neutron multiplicity counting of the singles, doubles, and triples neutrons has been evaluated for measuring ^{240}Pu, ^{244}Cm, and ^{252}Cf. We have proposed a method to establish the plutonium to curium ratio using the hybrid k-edge densitometer x-ray fluorescence instrument plus a neutron coincidence counter for the reprocessing dissolver solution. This report presents the concepts, experimental results, and error estimates for typical spent fuel applications.

I. INTRODUCTION

The verification of plutonium in spent-fuel reprocessing has focused on the main process streams because they contain a relatively large amount of plutonium and require more timely and sensitive verification procedures. However, the minor streams such as leached hulls and vitrified high-level wastes are now becoming important because they are increasing as the scale of reprocessing is becoming large. Also, it is desirable to remove the waste products from safeguards oversight after the plutonium has been measured.

Nevertheless, the measurement of plutonium in reprocessing wastes is difficult because the wastes are accompanied by a lot of fission products with extremely strong gamma radiation.

The measurement of plutonium in spent fuel is also important not only for safeguards but also for safely controlling their storage, transportation, and reprocessing. Burnup monitoring techniques have been developed to measure burnup and estimate amounts of plutonium. Most of these techniques are indirect methods and require other information about the fuel, such as initial enrichment, to calculate the amount of plutonium.

We have studied the assay methods that measure plutonium in leached hulls, high-level liquid wastes, vitrified wastes, and spent fuel by using passive neutrons emitted from the plutonium. Because neutrons penetrate well, they can be measured through thick shielding. In most cases after a sufficient cooling period for the decay of ^{242}Cm, ^{244}Cm is the dominant neutron source. Therefore, the curium information must be separated from the information

about plutonium. We studied a new method using neutron multiplicity to separate plutonium and curium.[1] We also evaluated another method using the plutonium to curium (Pu/Cm) ratio. The needs for further study are proposed.

II. NEUTRON SOURCES IN SPENT FUEL AND REPROCESSING WASTES

Neutron sources in several kinds of spent fuel were calculated by using the nuclear composition calculation code ORIGEN2.[2] Table I lists the calculated ratios of neutron yield from each neutron source after a 5-year cooling period. Curium-242 has decayed to a negligible level at this time. The spontaneous fissions of ^{244}Cm are the dominant neutron sources except for the low-burnup radial blanket of liquid-metal fast breeder reactors (LMFBRs).

TABLE I. Neutron Sources in Spent Fuels[a]

Neutron Sources		Light Water Reactor (LWR)			LMFBR[e]	
		UOX[b] 20 GWd/tu	UOX[c] 45 GWd/tu	MOX[d] 50 GWd/tu	CORE +AX.BLKT 80 GWd/tu[f]	RAD. BLKT 5 GWd/tu
Alpha,n	**Total**	6.6%	1.9%	1.3%	5.2%	57.2%
	^{238}Pu	2.3	0.8	0.4	2.2	4.7
	^{239}Pu	0.7	0.0	0.0	0.5	43.6
	^{240}Pu	0.8	0.1	0.0	0.7	7.8
	^{241}Am	2.1	0.2	0.1	1.1	1.1
	^{242}Cm	0.0	0.0	0.0	0.0	0.0
	^{244}Cm	0.7	0.8	0.8	0.7	0.0
Spontaneous Fission	**Total**	93.3%	98.1%	98.7%	94.8%	42.7%
	^{238}U	0.0	0.0	0.0	0.0	0.4
	^{238}Pu	0.4	0.0	0.1	0.4	0.8
	^{240}Pu	4.1	0.5	0.2	3.7	41.3
	^{242}Pu	0.9	0.2	0.1	1.1	0.0
	^{242}Cm	0.2	0.0	0.0	0.2	0.0
	^{244}Cm	87.6	96.8	97.6	89.4	0.2
	^{246}Cm	0.1	0.6	0.6	0.0	0.0
Total		100	100	100	100	100

[a]After a 5-year cooling time. Calculated by ORIGEN2.
[b]Initial uranium enrichment = 3.0 wt%.
[c]Initial uranium enrichment = 4.1 wt%.
[d]Initial fissile enrichment = 6 wt%. Recycled plutonium from an LWR with a burnup of 30 GWd/tu is used.
[e]Plutonium enrichment of core is 16 wt%. AX.BLKT=axial blanket, RAD.BLKT=radial blanket. Blankets are depleted uranium.
[f]This value is the burnup of CORE. The burnup of AX.BLKT is 5 GWd/t. A core fuel assembly consists of CORE (60%) and AX.BLKT (40%).

The neutron sources in spent fuel will be distributed into different streams during reprocessing. Most of ^{244}Cm will go into high-level liquid wastes and will be vitrified. A small amount of ^{244}Cm will go to solid wastes such as leached hulls. Most of the plutonium will become product material and only a little will go to solid wastes such as leached hulls or liquid wastes. Therefore the ratio of neutron sources is different at most of the process points. However, it is still likely that the ratio of Pu/Cm in spent fuel is the same in leached hulls because plutonium and curium behave similarly during dissolution at reprocessing.

III. CURIUM-PLUTONIUM NEUTRON SEPARATION

Two methods are proposed to assay plutonium by using neutrons from a mixture of curium and plutonium. One uses the neutron multiplicity distribution, the other uses the Pu/Cm ratio determined by destructive or nondestructive analysis.

A. Multiplicity Method

Moments Concept

A spontaneous fission nuclide has its own neutron multiplicity distribution. Figure 1 illustrates this distribution for ^{240}Pu, ^{244}Cm, and ^{252}Cf. These distributions have their own moments. The singles, doubles, and triples count rates, or S, D, and T, measured by a multiplicity counter, can be expressed with the moments.[3] If the neutron multiplication of the source can be regarded as unity, and if the (α,n) neutron source is negligible, then

$$S = mF\varepsilon\nu_{S1} = a \bullet m \ ,$$

$$D = \frac{mF\varepsilon^2 f_d \nu_{S2}}{2} = b \bullet m \ , \qquad (1)$$

$$T = \frac{mF\varepsilon^3 f_t \nu_{S3}}{3} = c \bullet m \ ,$$

where

m	=	mass (mg),
F	=	spontaneous fission yield counts/(s • mg),
ε	=	detector efficiency,
f_d	=	doubles gate fraction,
f_t	=	triples gate fraction,
ν_{s1}, ν_{s2}, ν_{s3}	=	first, second, and third reduced moments of the spontaneous fission neutron distribution,

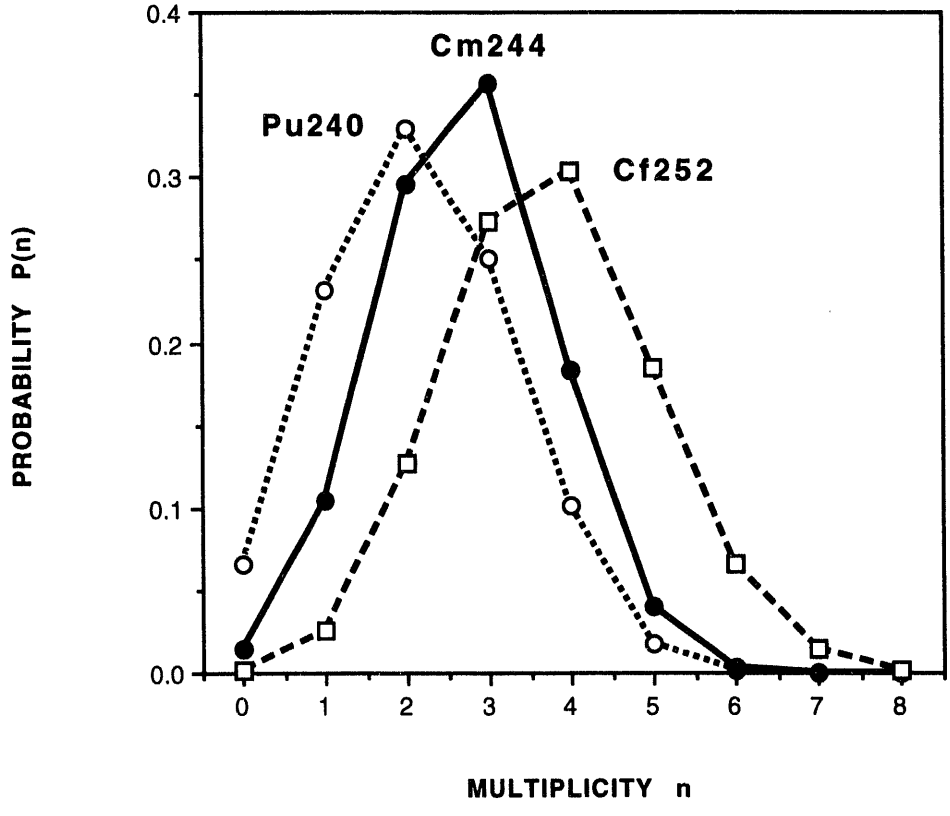

Fig. 1. *The multiplicity distributions of spontaneous fission neutrons from ^{240}Pu, ^{244}Cm, and ^{252}Cf. The distribution of ^{244}Cm was calculated as a Gaussian.*[5]

a, b, c = calibration coefficients counts/(s • mg),
S, D, T = singles, doubles, and triples count rates (counts/s).

For a mixture of two spontaneous fission nuclides, ^{240}Pu and ^{244}Cm, these equations reserve linearity for cases with low multiplication. The count rates of the mixture can be expressed as

$$S_{Pu+Cm} = a_{Pu} \bullet m_{Pu} + a_{Cm} \bullet m_{Cm} \ ,$$
$$D_{Pu+Cm} = b_{Pu} \bullet m_{Pu} + b_{Cm} \bullet m_{Cm} \ , \text{ and} \qquad (2)$$
$$T_{Pu+Cm} = c_{Pu} \bullet m_{Pu} + c_{Cm} \bullet m_{Cm} \ ,$$

where

m_{Pu}, m_{Cm} = mass of ^{240}Pu and ^{244}Cm (in mg),

$a_{Pu}, a_{Cm}, b_{Pu}, b_{Cm}, c_{Pu}, c_{Cm}$ = calibration coefficients of ^{240}Pu and ^{244}Cm [in counts/(s • mg)],

$S_{Pu+Cm}, D_{Pu+Cm}, T_{Pu+Cm}$ = singles, doubles, and triples count rates of mixture of ^{240}Pu and ^{244}Cm (in counts/s).

The calibration coefficients can be obtained by calculations with known parameters or by separate measurements of known samples. By using two of the three measured count rates, S_{Pu+Cm}, D_{Pu+Cm}, and T_{Pu+Cm}, the two unknowns, m_{Pu} and m_{Cm}, can be calculated. As an example, when S_{Pu+Cm} and D_{Pu+Cm} are used to obtain m_{Pu} and its error, they can be expressed as

$$m_{Pu} = \frac{b_{Cm}S - a_{Cm}D}{a_{Pu}b_{Cm} - a_{Cm}b_{Pu}} \quad \text{and} \tag{3}$$

$$\frac{\sigma_{m_{Pu}}}{m_{Pu}} = \frac{\sqrt{b_{Cm}^2\left(\frac{\sigma_S}{S}\right)^2 + a_{Cm}^2\left(\frac{\sigma_D}{D}\right)^2}}{\left|a_{Pu}b_{Cm} - a_{Cm}b_{Pu}\right|} \; . \tag{4}$$

This error does not include the errors of the calibration coefficients.

The concept of effective ^{240}Pu mass is defined the same as for the conventional neutron assay of plutonium. Because D and T are not affected by (α,n) neutrons, the combination of D and T can be used even if the sample has unknown (α,n) neutrons.

Discrimination Ratio

The discrimination ratio is an index of the performance of the separation method that is defined in Ref. 4. A larger discrimination ratio gives a better separation performance. The discrimination ratio of m_{Pu} and m_{Cm} with the measurements of S and D can be expressed as

$$DR = \frac{a_{Pu}/a_{Cm}}{b_{Pu}/b_{Cm}} \; . \tag{5}$$

B. Plutonium to Curium Ratio Method

Concept

The multiplicity method of separating plutonium from curium results in large errors for the minor constituent if one of the two mixed nuclides dominates the neutron yields. Actually, ^{244}Cm is the dominant neutron source in most spent fuels except for blanket assemblies of fast breeder reactor (FBR) or low-burnup research reactor fuels.

For these cases of typical burnup, we prefer to get the Pu/Cm ratio from a supplemental measurement. The method uses the ratio of Pu/Cm together with the measured *D* or *S*. The ratio can be determined by destructive analysis (DA) or nondestructive analysis (NDA) of small samples taken from the actual wastes or the feed materials. The neutron count rates are dominated by ^{244}Cm. The plutonium amount in the actual wastes can be obtained by multiplying the *D* or *S* of actual wastes by the Pu/Cm ratio.

We prefer to use *D* to avoid the effect of (α,n) neutrons. However, *S* may be more practical than *D* when the neutron detection efficiency is low and when it is confirmed that the (α,n) neutron ratio of the sample used to determine the Pu/Cm ratio is the same as that of actual wastes.

Measurement of the Plutonium to Curium Ratio in Dissolver Solutions

The Pu/Cm ratio of spent fuel can be determined by DA or NDA of dissolver solutions and should represent the ratio in leached hulls. DA will consist of chemical separation and mass analysis or radiation analysis. It will be accurate but would cost more than NDA. DA should be used to verify the results of NDA that would be done more frequently.

The hybrid k-edge densitometer/x-ray fluorescence (KED/XRF) instrument is available to measure the plutonium concentration of dissolver solutions.[5] A Kernforschungszentrum Karlsruhe-designed hybrid KED/XRF instrument[6] has been working at La Hague, France, for several years. LANL designed and fabricated a hybrid system[7] that was installed at the Tokai facility operated by the Japanese Atomic Energy Research Institute in Japan. Power Reactor and Nuclear Fuel Development Corporation, Japan, is also developing a hybrid system.[8]

Measuring neutrons from the sample of dissolver solution is not difficult. The inventory sample counter (INVS),[9] developed at LANL, can measure neutrons from the dissolver solution. The INVS can be located underneath a sample-handling glove box. The sample is set into a sample well in the bottom of the glove box. The neutron detector is located around the outside of the sample well. To achieve a high neutron detection efficiency, eighteen ^{3}He tubes surround the sample well in

two concentric rings. The detection efficiency of the INVS is about 43%. The measurement is done with the shift register coincidence counter.

The INVS must be modified to measure dissolver solutions. Lead shielding is necessary between the sample well and the ^{3}He tubes to reduce the gamma-ray dose to less than ~10 R/h. The maximum gamma-ray dose that a ^{3}He tube with a CO_2 mixture can tolerate is approximately 10 R/h. Designs are being evaluated to increase the limit to ~50 R/h.

With these two NDA systems, the ratio of the plutonium mass to the ^{244}Cm can be obtained.

Plutonium to Curium Ratio in Wastes

Before applying the Pu/Cm ratio method to the measurement of wastes, it is necessary to establish the Pu/Cm in the waste stream of interest.

The leached hulls may have the same ratio as the dissolver solution, because the chemical behavior of curium and plutonium during irradiation and dissolution are similar. However, the ratio of (α,n) neutrons to spontaneous fission neutrons may be different for dissolver solutions and leached hulls. For normal oxide fuel, the (α,n) contribution is only a small fraction (1 to 3%) of the spontaneous fission neutron yield (see Table I) so possible changes in the (α,n) yield should be unimportant. Therefore, some destructive analysis will be needed to confirm the constancy of the ratio. Any neutron multiplication in the leached hulls or the ~3-mℓ grab sample from the dissolver solution is negligible.

It is clear that the Pu/Cm ratio of the liquid effluent from extraction cycles and vitrified wastes is not the same as that of the dissolver solution. In these cases the ratio must be measured for each waste stream. Because the ratio for these wastes is much smaller than that of the dissolver solution, the hybrid KED/XRF might not be applicable. DA or some other NDA will be required. The accuracy required for the plutonium in the vitrified waste is low so the error for the Pu/Cm ratio can be large and still satisfy the safeguards requirements.

Anticipated Errors

We assumed that 45 GWd/t UOX spent fuel from an LWR approximated the measurement error of the Pu/Cm ratio method for measuring leached hulls.

The plutonium concentration of a sample of dissolver solution from the spent fuel will be about 2 g/ℓ. It can be measured with about 1% error with the hybrid KED/XRF.[5] The sample volume is assumed to be 3 mℓ. The volume can be determined with less than 1% error. The sample contains neutron sources that emit about 350 neutrons per second. If the same sample can be measured by the INVS, the S will be about 120 counts per second and D will be about 20 counts per

second. The counting statistical error of the D will be about 1% after 1000 s of measurement.[5] Finally, the Pu/Cm ratio can be measured with, at most, 2% error. This value is small enough that we can apply the ratio method for verification of plutonium in the reprocessing wastes.

However, the overall measurement error depends on the error in measuring neutrons from the actual leached hulls as well as from the grab sample. The measurement error for the actual leached hulls should depend on the conditions of each particular case. The usefulness of this method must be evaluated for the particular applications by studying the actual neutron measurement system.

IV. EXPERIMENTAL

The feasibility of the method for separating curium and plutonium by neutron multiplicity counting was studied experimentally. Two combinations of sources were measured. One was ^{240}Pu and ^{252}Cf; the other was ^{244}Cm and ^{252}Cf.

The combination of ^{244}Cm and ^{240}Pu was not measured because the source of ^{244}Cm was strong and a comparative plutonium source would have been more than 1 000 g of plutonium. This amount of plutonium will give a high multiplication and many (α,n) neutrons. With this condition, the equations described in the previous section are not valid. Therefore, we studied this combination and used the results of the other two combinations to extrapolate to the ^{240}Pu and ^{244}Cm mixture.

A. Equipment

The detector used in this experiment was the dual-mode multiplicity counter, which was built in 1985 and still is used for research and development. This counter consists of 130 helium-3 tubes arranged in five concentric rings in an aluminum/polyethylene body. The sample cavity is 16.5 cm in diameter and 25.4 cm high. The overall diameter is about 80 cm and the counter is 85 cm high with its wheels.

The detection efficiency was about 53% and the die-away time was about 57 μs.[10] The values of these parameters were measured with a ^{252}Cf source for this experiment.

The data were collected by the MULTI software[11] and a Los Alamos MSR-4 multiplicity shift register coincidence counter.[12] The count losses from dead time were corrected with the analytic procedure developed by Dytlewski.[13] At higher count rates (>250 kHz), the procedure does not provide enough correction, so additional correction factors are applied to the doubles and triples rates.[12] The ^{244}Cm source used in this experiment gave a high count rate (ca. 200 kHz), so we reviewed the dead-time correction parameters and used a new set of correction parameters for the measurements of the combination of ^{244}Cm and ^{252}Cf. These parameters were based on the measurement of known rates from combinations of ^{252}Cf and americium-lithium.

The detector parameters used in this experiment are listed in Table II.

B. Sources

All the sources used in these experiments were sealed in small stainless steel capsules.

TABLE II. Detector Parameters		
Detector Parameters	Pu-Cf Combination	Cm-Cf Combination
High Voltage (V)	1680	1680
Gate Length (μm s)	32	32
Predelay (μm s)	3	3
Die-Away Time (μm s)	57	57
Dead Time (ns)	77.5	86.5
Dead-Time Coefficient *c*	9.72×10^{-8}	5.59×10^{-8}
Dead-Time Coefficient *d*	1.12×10^{-7}	6.57×10^{-8}
Efficiency	0.5349	0.5341
Doubles Gate Fraction	0.3979	0.3982
Triples Gate Fraction	0.1685	0.1693

Plutonium

A small ^{240}Pu-oxide source (FZC-158) was used for the initial experiments. The specifications are shown in Table III. The effective ^{240}Pu mass at the date of measurement was calculated with the MULTI software.

Curium

Two ^{244}Cm-oxide sources were available; however, only the smaller source was used to make combinations with californium sources in this experiment because the larger source was too large. The specifications of the sources are shown in Table IV. The reference mass of ^{244}Cm was determined with the measured *S*, the detector efficiency, and the known specific neutron yield.[5]

The neutrons from ^{240}Pu are negligible because the specific neutron emission rate is very small compared with ^{244}Cm [SFn/s-g means spontaneous fissions/(s • g) and (α,n)n/s-g means (α,n) neutrons/(s • g)].

^{240}Pu $\quad 1.02 \times 10^3$ SFn/s-g + 1.41×10^2 (α,n)n/s-g and
^{244}Cm $\quad 1.08 \times 10^7$ SFn/s-g + 7.73×10^4 (α,n)n/s-g .

TABLE III. Specifications of Plutonium Source (Dec. 1978)							
PuO_2 Mass (g)	Isotypes (wt %)						
	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{243}Pu	^{241}Am
0.7879	0.014	0.955	93.76	0.699	4.56	0.009	not known
Reference mass of effective ^{240}Pu at the date of this experiment was 0.704 g.							

TABLE IV. Specifications of Curium Sources					
ID	Name	Composition (wt %)		Metal Total Mass (mg)	Metal ^{244}Cm Mass (mg)
		^{244}Cm	^{240}Pu		
DET 743902	^{244}Cm *S*	37.3	42.6	90	33.6
DET 743906	^{244}Cm *L*	37.3	42.6	122	45.5
The date of specification was not available. Reference mass of ^{244}Cm *S* used in this study was 32.9 mg.					

Californium

The Los Alamos reference californium sources CR1 through CR11 were used. The reference mass of ^{252}Cf was calculated from the absolute mass of CR5. For the other sources, the ratio of the measured doubles to the measured doubles of CR5 was used to get the reference mass. The absolute mass of CR5 was 0.0151 μg (as of 1/1/87), and the decay constant was 0.2623 y^{-1}.[14] The reference mass values used in this study at the date of the experiment are listed in Table V.

C. Measurement Procedure

Calibration

Each source was measured separately to get its calibration coefficients counts/(s • mg).

The sources were set at the center of the sample cavity of the multiplicity counter. The ^{252}Cf sources were attached to an aluminum rod so they could be set precisely at the center of the cavity. The other sources were set on an empty can adjusted so that they were in the center of the cavity.

The measurement time was one hour divided into 120 intervals of 30 s to allow a statistical quality control test on the data and to eliminate infrequent cosmic-ray events with high multiplicity.

The measured multiplicity distributions were converted by correcting for dead time and subtracting background to obtain *S*, *D*, and *T*, automatically with MULTI software. These measured count rates and the reference mass values were used to obtain the calibration coefficients.

Combination Sources

Two sources were set in the sample cavity of the multiplicity counter and measured simultaneously. The ^{252}Cf source was attached to the rod and the other was set on an empty can.

TABLE V. Reference Mass of Californium Sources				
Sources	Pu-Cf Combination Measurement		Cm-Cf Combination Measurement	
	Date	Reference Cf Mass (mg)	Date	Reference Cf Mass (mg)
CR1	9/21/93	6.40E-08	—	—
CR2	9/21/93	1.75E-07	—	—
CR3	9/20/93	4.20E-07		—
CR4	9/21/93	9.94E-07	—	—
CR5	—	—	12/8/93	2.46E-06
	—	—	12/7/93	2.46E-06
CR6	9/21/93	6.69E-06	11/9/93	6.46E-06
	—	—	12/6/93	6.33E-06
CR8	9/21/93	3.82E-05	—	—
CR9	—	—	11/9/93	8.27E-05
	—	—	12/5/93	8.12E-05
CR10	—	—	11/12/93	2.10E-04
	—	—	12/4/93	2.08E-04
CR11	—	—	11/12/93	5.54E-05
	—	—	12/3/93	5.47E-04

The measurement time was normally 1 h. Twenty-four-hour measurements were done for the second run of the ^{244}Cm and ^{252}Cf combination. This second run was done to get better counting statistics and to study why some of the assay results from the first run were far from the reference mass.

The measured count rates (S, D, and T) and the calibration coefficients were used to calculate the mass value of each source. The counting statistical errors and the calibration coefficients were used to calculate the errors of obtained mass values.

V. RESULTS

The results of the experiments with the two combinations and the calculations for the $^{244}Cm/^{240}Pu$ combination are described below.

A. ^{240}Pu and ^{252}Cf Combination

Calibration Coefficients

The measured and calculated calibration coefficients (a, b, and c) are listed in Table VI.

The calculation was done by Eq. (1) with the known moments[5] and with an assumption of nonmultiplication and negligible (α,n) neutrons. Actually, the ^{240}Pu source had M = 0.007 and an (α,n) ratio of 0.15. This is the reason some measured data do not agree with the calculated values. The measured a of ^{240}Pu is larger than the calculated value because the (α,n) source is not zero. The measured c of ^{240}Pu is larger than the calculated value because M is not 1.

Discrimination Ratios

The discrimination ratios of the combination of ^{240}Pu and ^{252}Cf are listed in Table VII.

The measured discrimination ratio of *S-D* is larger than the calculated one because the calculated discrimination ratio does not include (α,n) neutrons, as mentioned above.

Assay

The measured count rates of the plutonium and californium sources are listed in Table VIII. The assay results are shown in Table IX. Generally, the assay results agree with the reference values very well. In the case of FZC-158 + CR8, the assay result of effective ^{240}Pu mass with D and T measurements was more than three times larger than the reference value. However, the error of the assay was also large enough to cover the reference value. In this particular case, the plutonium source emitted less than 1% of the neutrons and the californium source emitted over 99%. Thus the error for the dominant californium source is small.

Figure 2 shows the assay errors as a function of *S*-Ratio. The *S*-Ratio is the ratio of the *S* (singles neutron rate) from plutonium to the total *S*. The discrimination ratios

TABLE VI. Calibration Coefficients of ^{240}Pu and ^{252}Cf

	^{240}Pu		^{252}Cf	
[counts/(s • mg)]	Measured	Calculated	Measured	Calculated
a	6.17E-01	5.46E-01	1.25E+09	1.25E+09
b	1.03E-01	1.03E-01	4.26E+08	4.24E+08
c	2.31E-02	2.17E-02	1.72E+08	1.70E+08

TABLE VII. Discrimination Ratios of the ^{240}Pu and ^{252}Cf Combination

Assay by	Measured	Calculated
S-D	2.0	1.8
S-T	3.7	3.4
D-T	1.8	1.9

TABLE VIII. Measured Count Rates of ^{240}Pu and ^{252}Cf Combination by Multiplicity					
Sources	Date	Measurement Time	Singles (counts/s)	Doubles (counts/s)	Triples (counts/s)
FZC-158+CR1	9/21/93	1 h	514 ± 1	99 ± 0	27 ± 0
FZC-158+CR2	9/21/93	1 h	655 ± 1	147 ± 0	46 ± 0
FZC-158+CR3	9/20/93	1 h	962 ± 1	252 ± 0	89 ± 1
FZC-158+CR4	9/21/93	1 h	1684 ± 1	496 ± 1	187 ± 1
FZC-158+CR6	9/21/93	1 h	8836 ± 3	2919 ± 2	1156 ± 4
FZC-158+CR8	9/21/93	1 h	48316 ± 5	16342 ± 10	6547 ± 29

TABLE IX. Assay Results of ^{240}Pu and ^{252}Cf Combination by Multiplicity Method

Sources	Measurement Time	*S*-Ratio Pu/(Pu+Cf) %	*D*-Ratio Pu/(Pu+Cf) %	Assay/Reference ± 1σ					
				Assay by *S-D*		Assay by *S-T*		Assay by *D-T*	
				^{252}Cf	eff. ^{240}Pu	^{252}Cf	eff. ^{240}Pu	^{252}Cf	eff. ^{240}Pu
FZC-158+CR1	1 h	84.5	72.7	0.96 ± 0.02	1.01 ± 0.00	0.99 ± 0.03	1.00 ± 0.01	1.00 ± 0.05	0.99 ± 0.02
FZC-158+CR2	1 h	66.4	49.4	0.99 ± 0.01	1.01 ± 0.00	1.00 ± 0.02	1.00 ± 0.01	1.00 ± 0.03	1.00 ± 0.03
FZC-158+CR3	1 h	45.1	28.9	1.00 ± 0.01	1.01 ± 0.01	1.00 ± 0.01	1.00 ± 0.01	1.00 ± 0.01	0.99 ± 0.04
FZC-158+CR4	1 h	25.8	14.6	1.00 ± 0.00	1.02 ± 0.01	0.99 ± 0.01	1.03 ± 0.02	0.99 ± 0.01	1.03 ± 0.06
FZC-158+CR6	1 h	4.9	2.5	1.00 ± 0.00	1.12 ± 0.03	0.99 ± 0.00	1.29 ± 0.09	0.98 ± 0.01	1.68 ± 0.31
FZC-158+CR8	1 h	0.9	0.4	1.00 ± 0.00	1.06 ± 0.14	0.99 ± 0.01	1.76 ± 0.66	0.99 ± 0.01	3.41 ± 2.21

rates should be *S-T* > *S-D* > *D-T*. Actually, the order is *S-D* > *S-T* > *D-T* because the statistical error for *T* is much larger than for *S* and *D*.

Ratio Analysis

Figure 3 shows the measured ratios of *T/D* plotted as a function of the *D*-Ratio. The *D*-Ratio is defined as the ratio of the *D* from ^{240}Pu to the total *D*. The solid line connects the *T/D* of a pure ^{240}Pu source and that of a pure ^{252}Cf source. The *T/D* of a mixture of ^{240}Pu and ^{252}Cf should be on the line. Therefore, the *D*-Ratio of the mixture can be obtained from the measured *T/D*. The ratio of mass can be calculated from the obtained *D*-Ratio. This relation is useful when the absolute calibration is not available. The ratios of *D/S* and *T/S* with *S*-Ratio are related similarly.

B. ^{244}Cm and ^{252}Cf Combination

Moments of ^{244}Cm

The moments of the multiplicity distribution of ^{244}Cm were obtained from the measured *S*, *D*, and *T* of a ^{244}Cm source. The moments were calculated with Eq. (1) and the detector parameters listed in Table II. Because the absolute quantity of our ^{244}Cm source was not accurately known, the second and the third

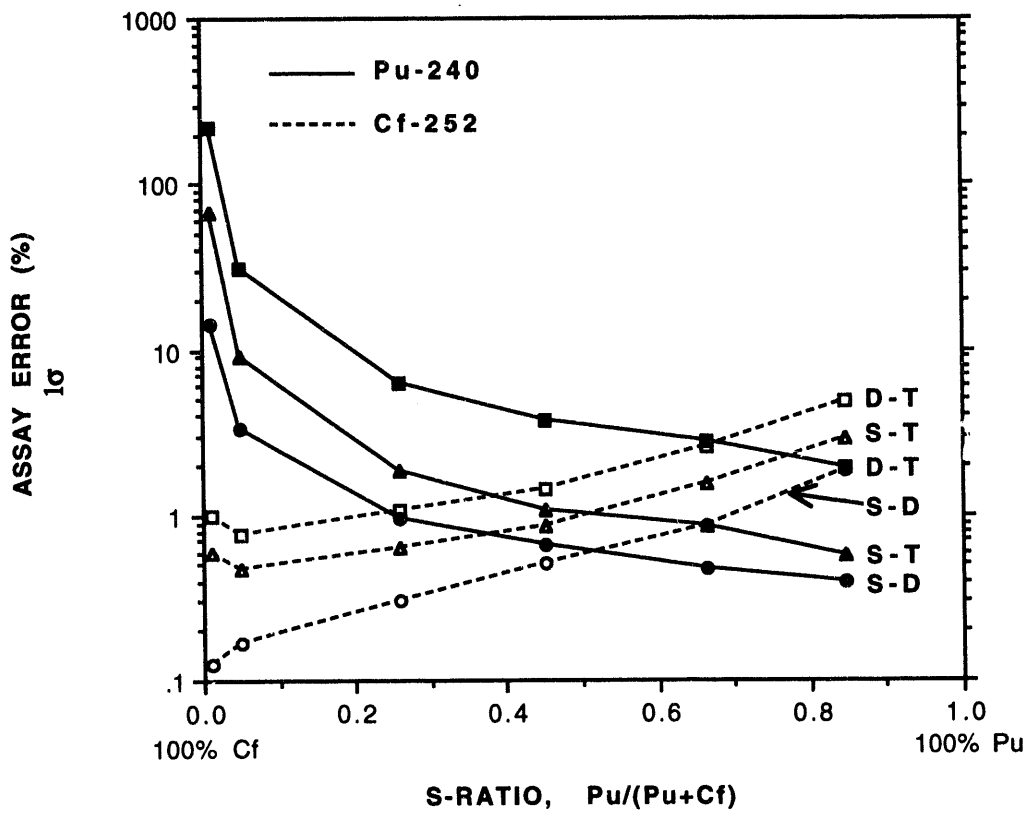

Fig. 2 *The relative assay error for a 1-h measurement of the ^{240}Pu and ^{252}Cf combination as a function of the* S-*Ratio. The* S-*Ratio is defined as the ratio of the* S *(singles neutron rate) from ^{240}Pu to the total* S.

moments were calculated based on the known mean multiplicity from reference data.[5] The measured moments are listed in Table X.

Calibration Coefficients

The measured and calculated calibration coefficients a, b, and c are listed in Table XI.

The calculated calibration coefficients of ^{252}Cf are different from those in Table VI because the detector parameters used for the calculations are not the same. The detector parameters are listed in Table II.

In this combination, the measured and calculated values agree well because (α,n) neutrons are completely negligible for this combination.

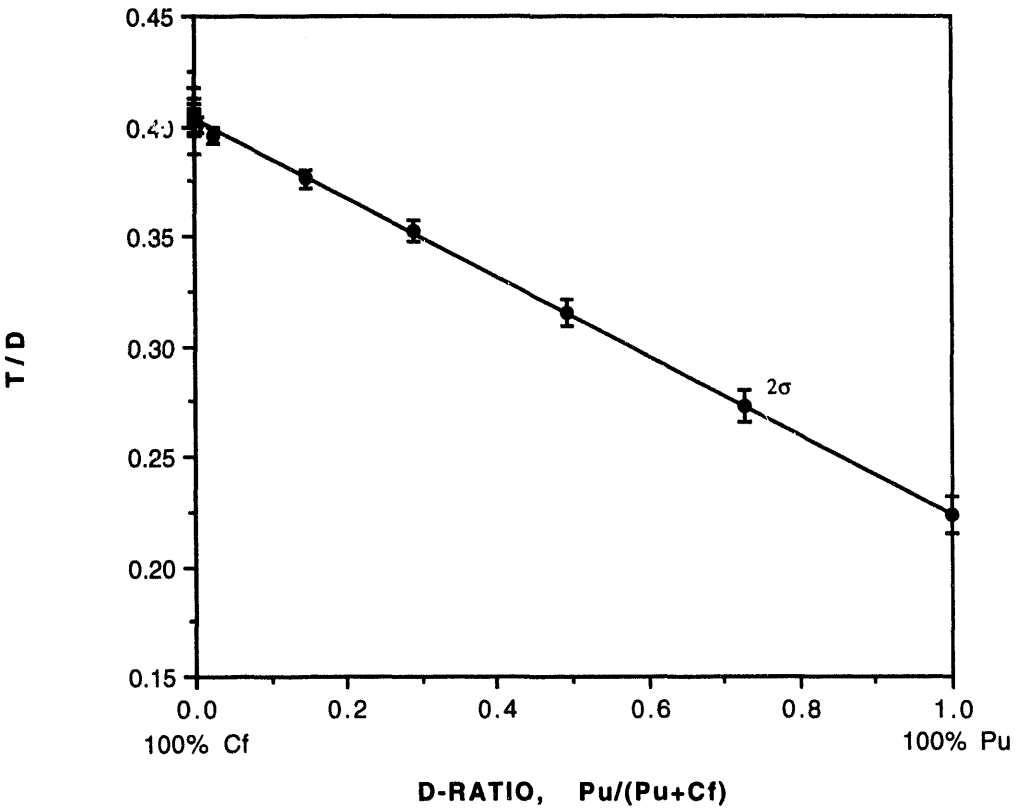

Fig. 3 *The measured* T/D *ratio of the* ^{240}Pu *and* ^{252}Cf *combination as a function of the* D-*Ratio. The* D-*Ratio is defined as the ratio of the* D *(doubles neutron rate) from* ^{240}Pu *to the total* D.

TABLE X. Measured Moments of ^{244}Cm	
Order	Moments
First	2.72 (known)
Second	5.99
Third	10.6

Discrimination Ratios

The discrimination ratios of the combination of ^{244}Cm and ^{252}Cf are listed in Table XII.

In comparison with the ^{240}Pu and ^{252}Cf combination, the discrimination ratios of the ^{244}Cm and ^{252}Cf combination are smaller. This means the ^{244}Cm and ^{252}Cf

TABLE XI. Calibration Coefficients of ^{244}Cm and ^{252}Cf				
	^{244}Cm		^{252}Cf	
$\frac{\text{counts}}{(s \cdot mg)}$	Measured counts $\frac{}{(s \cdot mg)}$ $x10^3$	Calculated counts $\frac{}{(s \cdot mg)}$ $x10^3$	Measured counts $\frac{}{(s \cdot mg)}$ $x10^9$	Calculated counts $\frac{}{(s \cdot mg)}$ $x10^9$
a	5.79	5.70	1.25	1.25
b	1.35	1.33	0.425	0.423
c	0.358	0.357	0.172	0.170

combination is more difficult to separate than the ^{240}Pu and ^{252}Cf combination because the multiplicity distributions of ^{244}Cm and ^{252}Cf are closer than those of ^{240}Pu and ^{252}Cf (see Fig. 1).

Assay

The measured count rates of the curium and californium sources are listed in Table XIII. The assay results are shown in Table XIV. Generally, the assay results agree well with the reference values. These results suggest this method is practical to use if the neutron emission rate from ^{244}Cm is similar to that from

TABLE XII. Discrimination Ratios of ^{244}Cm and ^{252}Cf Combination		
Assay by	Measured	Calculated
S-D	1.5	1.4
S-T	2.2	2.2
D-T	1.5	1.5

TABLE XIII. Measured Count Rates of ^{244}Cm and ^{252}Cf Combination by Multiplicity Counter					
Sources	Date	Measurement Time	Singles (counts/s)	Doubles (counts/s)	Triples (counts/s)
^{244}Cm + CR5	12/8/93	1 h	193337 ± 11	45559 ± 36	12387 ± 154
^{244}Cm + CR6	11/9/93	1 h	199100 ± 10	47533 ± 38	13148 ± 160
^{244}Cm + CR9	11/9/93	1 h	295136 ± 14	80119 ± 59	26215 ± 295
^{244}Cm + CR10	11/12/93	1 h	454762 ± 17	133862 ± 97	49110 ± 581
^{244}Cm + CR11	11/12/93	1 h	892049 ± 25	279646 ± 216	112801 ± 1557
^{244}Cm + CR5	12/7/93	24 h	193323 ± 2	45561 ± 7	12374 ± 30
^{244}Cm + CR6	12/6/93	24 h	198250 ± 3	47223 ± 8	13133 ± 32
^{244}Cm + CR9	12/5/93	24 h	292458 ± 4	79032 ± 13	25802 ± 52
^{244}Cm + CR10	12/4/93	24 h	449711 ± 8	132001 ± 17	47002 ± 117
^{244}Cm + CR11	12/3/93	24 h	880425 ± 16	275576 ± 42	107695 ± 340

TABLE XIV. Assay Results of ^{244}Cm and ^{252}Cf Combination by Multiplicity Method.									
Sources	Measurement Time	*S*-Ratio Cm/(Cm+Cf) %	*D*-Ratio Cm/(Cm+Cf) %	Assay/Reference ± 1σ					
				Assay by *S-D*		Assay by *S-T*		Assay by *D-T*	
				^{252}Cf	^{244}Cm	^{252}Cf	^{244}Cm	^{252}Cf	^{244}Cm
^{244}Cm + CR5	1 h	98.4	97.7	1.68 ± 0.11	0.99 ± 0.00	1.82 ± 0.66	0.99 ± 0.01	1.90 ± 1.05	0.99 ± 0.03
^{244}Cm + CR6	1 h	95.9	94.2	1.37 ± 0.04	0.99 ± 0.00	1.35 ± 0.26	0.99 ± 0.01	1.34 ± 0.42	0.99 ± 0.03
^{244}Cm + CR9	1 h	64.8	56.0	1.03 ± 0.01	0.99 ± 0.00	1.01 ± 0.04	1.00 ± 0.02	1.00 ± 0.06	1.02 ± 0.05
^{244}Cm + CR10	1 h	42.0	33.2	1.00 ± 0.00	1.01 ± 0.00	1.05 ± 0.03	0.94 ± 0.04	1.08 ± 0.05	0.84 ± 0.09
^{244}Cm + CR11	1 h	21.4	15.9	0.97 ± 0.00	1.15 ± 0.01	1.10 ± 0.03	0.70 ± 0.11	1.17 ± 0.05	0.10 ± 0.25
^{244}Cm + CR5	24 h	98.4	97.7	1.70 ± 0.02	0.99 ± 0.00	1.77 ± 0.13	0.99 ± 0.00	1.81 ± 0.20	0.99 ± 0.00
^{244}Cm + CR6	24 h	96.0	94.3	1.27 ± 0.01	0.99 ± 0.00	1.44 ± 0.05	0.98 ± 0.00	1.55 ± 0.08	0.97 ± 0.01
^{244}Cm + CR9	24 h	65.1	56.4	1.01 ± 0.00	1.00 ± 0.00	1.00 ± 0.01	1.00 ± 0.00	0.99 ± 0.01	1.01 ± 0.01
^{244}Cm + CR10	24 h	42.3	33.5	0.98 ± 0.00	1.02 ± 0.00	0.97 ± 0.01	1.04 ± 0.01	0.97 ± 0.01	1.06 ± 0.02
^{244}Cm + CR11	24 h	21.6	16.1	0.97 ± 0.00	1.16 ± 0.00	1.02 ± 0.01	0.94 ± 0.02	1.06 ± 0.01	0.66 ± 0.06

^{252}Ct. However, if one of them becomes dominant, the assay of the other becomes difficult.

In the case of ^{244}Cm + CR11, the assay result of ^{244}Cm with the *D* and *T* measurements was only 66% of the reference value. The error of this assay was about 6% (1σ) and did not cover the difference between the assay and the reference value. We think the discrepancy is caused by insufficient dead-time correction for this high-counting-rate combination. The total neutron count rate of this combination is about 900 kHz. The dead-time correction is difficult at this high count rate, and the assay is very sensitive to the *T* for this condition. Only a 1% change in *T* causes a 20% change in the assayed mass.

Nevertheless, these high count rates are out of our range of interest for hulls and wastes. The neutron emission rate from 0.2 ton of spent fuel [~1 boiling-water reactor (BWR) fuel assembly] with 45 GWd/tU is about $1x10^8$ n/s. For the measurement of leached hulls, we expect at most, 10 kHz count rates. If the leached hulls represented 0.2 ton of spent fuel, the residual neutron source would be 0.1% of the input spent fuel, and the neutron detection efficiency would be 10%.

Figure 4 shows the assay errors as a function of *S*-Ratio. The *S*-Ratio is the ratio of *S* from curium to the total *S*. One hour of measurement time was required for the data on this figure. In comparison with Fig. 2, the assay errors of the ^{244}Cm and ^{252}Cf combination were larger than those of the ^{240}Pu and ^{252}Cf combination. This means that it is more difficult to separate the ^{244}Cm and ^{252}Cf combination than the ^{240}Pu and ^{252}Cf combination, as anticipated from the difference among multiplicity distributions of three nuclides.

Ratio Analysis

Figure 5 shows the measured ratios of *T/D* plotted as a function of the *D*-Ratio. The *D*-Ratio is defined as the ratio of the *D* from curium to the total *D*. The solid line connects the *T/D* of pure ^{244}Cm and that of pure ^{252}Cf. The measured *T/D* of

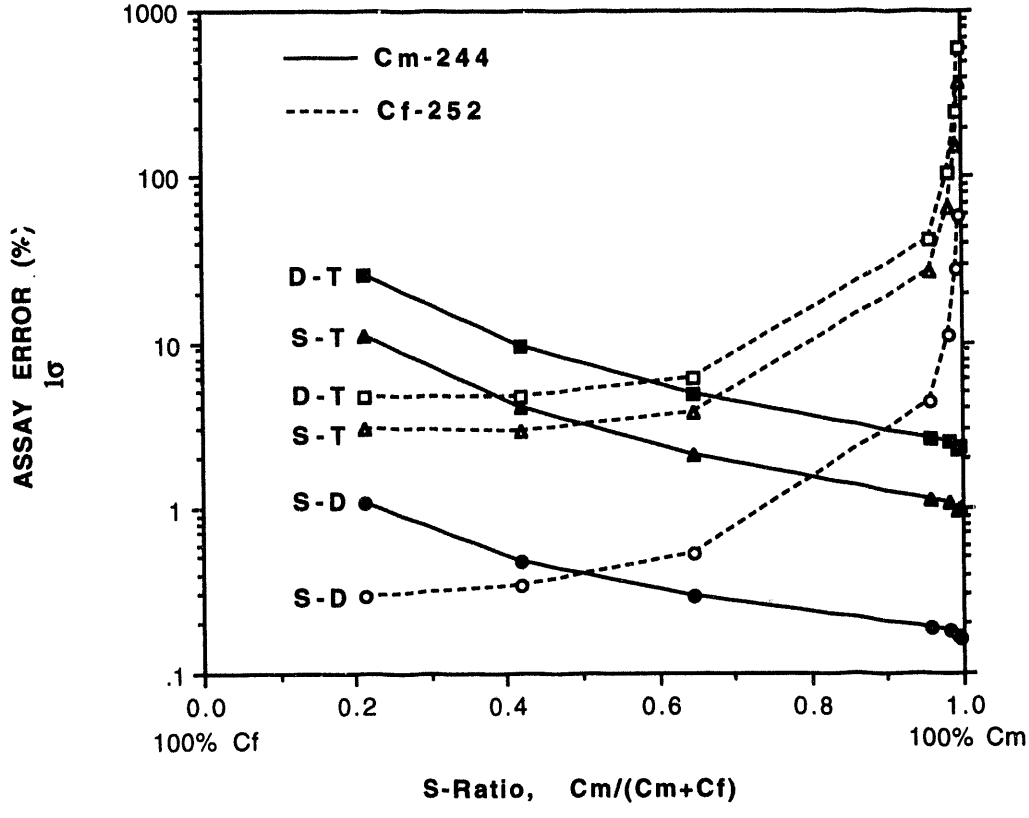

Fig. 4. *The relative assay error for a 1-hour measurement of the ^{244}Cm and ^{252}Cf combination as a function of the* S-*Ratio. The* S-*Ratio is defined as the ratio of the* S *(singles neutron rate) from ^{244}Cm to the total* S.

the mixture of ^{244}Cm and ^{252}Cf is on the expected solid line as well as the combination of ^{240}Pu and ^{252}Cf.

C. ^{240}Pu and ^{244}Cm Combination

To study the separability of this combination with the neutron multiplicity method, we calculated the assay errors with the measured calibration coefficients and the assumed measurement errors of S, D, and T.

Calculation Procedure and Conditions

We used Eq. (4) and similar equations to calculate the assay errors. The measured calibration coefficients are listed in Tables VI and XI. The measurement errors of S, D, and T were assumed to be 0.01%, 0.06%, and 0.4%, respectively. These

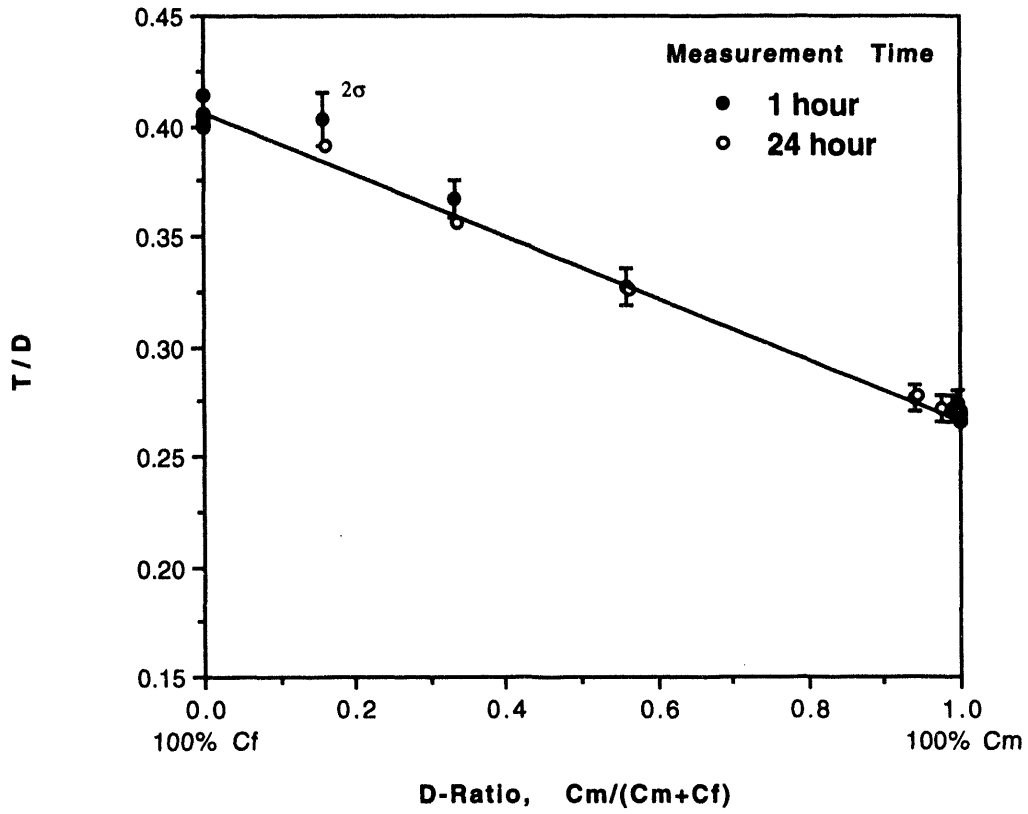

Fig. 5. *The measured* T/D *ratio of the* ^{244}Cm *and* ^{252}Cf *combination as a function of the* D-*Ratio. The* D-*Ratio is defined as the ratio of the* D *(doubles neutron rate) from* ^{244}Cm *to the total* D.

values are counting statistical errors of a ^{252}Cf source measured by the dual-mode multiplicity counter.

Discrimination Ratios

The discrimination ratios of all combinations are summarized in Table XV. These are obtained from the measured calibration coefficients. The ^{240}Pu and ^{244}Cm combination has the smallest discrimination ratios for the three combinations.

Assay Errors

Figure 6 shows the calculated assay errors as a function of *S*-Ratio. It is clearly shown that the assay error for the minor component increases sharply when one of the two nuclides dominates the total neutron emission rate. The errors of the assay

TABLE XV. Discrimination Ratios of Multiplicity Method			
Assay by	$^{240}Pu/^{252}Cf$	$^{244}Cm/^{252}Cf$	$^{240}Pu/^{244}Cm$
S-D	2	1.5	1.4
S-T	3.7	2.2	1.7
D-T	1.8	1.5	1.2

of ^{240}Pu by *D* and *T* increase to about 350% when only 1% of the total neutrons come from ^{240}Pu and the rest come from ^{244}Cm.

The calculated assay errors for leached hulls from different kinds of reactors are listed in Table XVI. The ratio of neutron sources in leached hulls is assumed to be the same as in the spent fuel. To eliminate the effect of (α,n) neutrons, we assumed that *D* and *T* were used. The values in Table I were used to obtain the ratios of spontaneous fission neutrons from plutonium and curium. The multiplicities of

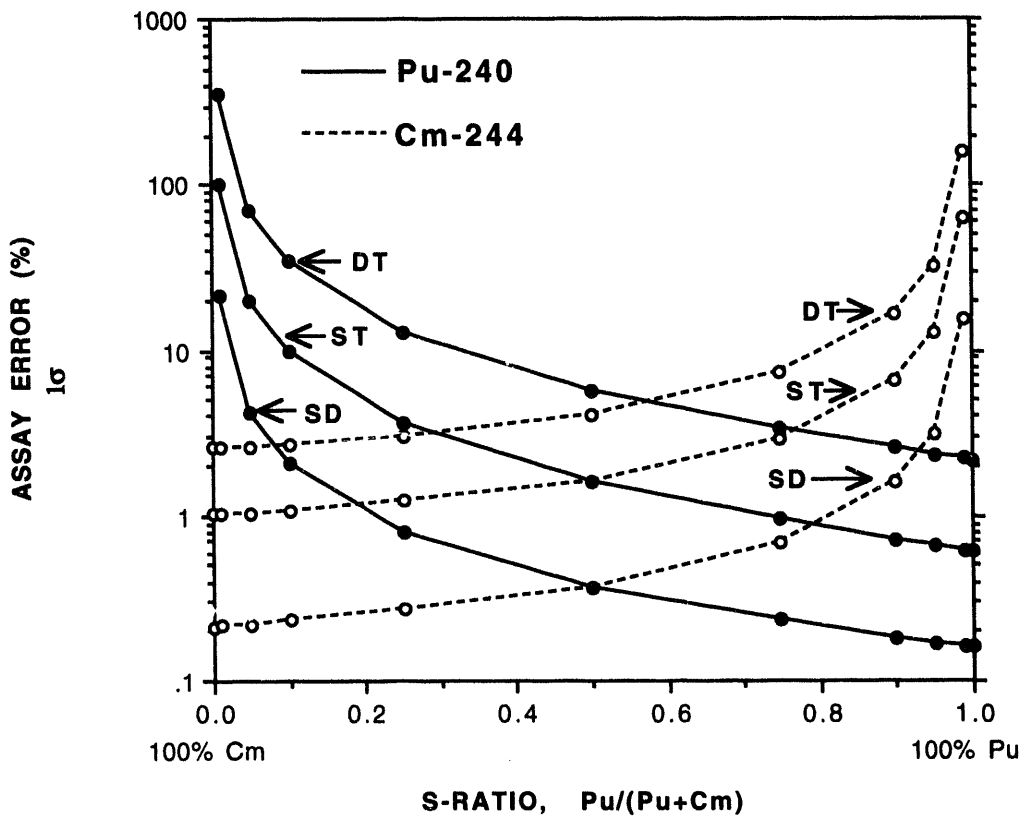

Fig. 6. *The calculated relative assay error of the ^{240}Pu and ^{244}Cm combination as a function of the* S-*Ratio. The* S-*Ratio is defined as the ratio of the* S *(singles neutron rate) from ^{240}Pu to the total* S.

TABLE XVI. Neutron Sources and Assay Errors of Plutonium in Leached Hulls by Multiplicity Method

Spent Fuels		LWR			LMFBR	
		UOX 20 GWd/t %	UOX 45 GWd/t %	MOX 30 GWd/t %	CORE+AX.BLKT 80 GWd/t %	RAD. BLKT 5 GWd/t %
Spontaneous Fission Neutrons	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	60.0	507.5	711.1	63.4	2.2

^{238}Pu and ^{242}Pu were assumed to be the same as that of ^{240}Pu. Similarly, the multiplicities of ^{242}Cm and ^{246}Cm were assumed to be the same as that of ^{244}Cm. For most practical applications, the ^{242}Cm will have decayed to negligible levels because of its short half-life (163 d). Because ^{240}Pu is dominant in the case of the radial blanket of a LMFBR, the assay error is small. However, this is a particular case that does not require the separation of curium and plutonium because the curium is negligible. Concerning the low burnup (20 GWd/t) UOX spent fuel of LWR and CORE+AX.BLKT spent fuel of LMFBR, the calculated assay error was smaller than 100%. As for the high-burnup LWR fuels, the calculated assay errors were more than 500%. For these cases, the Pu/Cm ratio should be determined by the supplemental measurement method and only the ^{244}Cm would be measured in the leached hulls.

VI. CONCLUSIONS

We examined the technique of separating plutonium and curium with neutron multiplicity counting through experiments and calculations. The feasibility of this technique was confirmed by the experiments. However, it is not practical to apply this method to measuring leached hulls from high-burnup LWR fuels because the assay error would be more than 500%. As for low-burnup LWR fuels or FBR fuels, the assay error would be smaller than 100%. The assay errors should be considered in evaluating the use of this method. It might be more economical to install a measurement system that performs similar to the dual-mode multiplicity counter.

Generally, a high neutron detection efficiency is required to measure a higher multiplicity, such as T. A careful study of geometry and many neutron detection tubes will be required to obtain a suitable efficiency. This may be the critical consideration in deciding if the multiplicity method will be used.

The supplemental measurement method of determining the Pu/Cm ratio may be much more practical for most applications. The assay error of this method will depend on the performance of the system in measuring singles and coincidence neutrons from actual waste materials. An evaluation of the measurement system for each case will be required.

The Pu/Cm ratio in the dissolver solution can be determined by DA. NDA would be preferable because of economical and operational reasons. A combination of the hybrid KED/XRF instrument and the INVS is proposed to measure the Pu/Cm ratio. The hybrid KED/XRF instruments have been developed and used for practical measurements of hot dissolver solutions. The INVS could measure neutrons with some modifications for additional shielding. The equipment must be developed and tested.

The constancy of the Pu/Cm ratio for actual wastes also needs to be checked. As for the leached hulls, it is reasonable that the Pu/Cm ratio would be the same as that of the dissolver solution. However, the ratio of (α,n) neutrons and the neutron multiplication might be different for dissolver solutions and leached hulls. Therefore, some destructive analysis will be needed to confirm that the neutron signal from the leached hulls accurately reflects the neutron signal from the dissolver solution. The Pu/Cm ratio should be much smaller than other wastes, such as vitrified wastes, for the dissolver solution. Proper systematic measurement of the waste stream will be necessary to obtain the Pu/Cm ratio for the wastes. It should be possible to apply neutron coincidence counting to the ^{244}Cm in the vitrified waste in bulk containers. Passive neutron counters[15] that have been designed to assay waste in 200-ℓ drums could be used for vitrified waste containers after modification with lead shielding to protect the detectors from high gamma-ray dose levels.

Because the spontaneous fission neutrons from ^{244}Cm will probably dominate all sources of neutrons in the vitrified wastes and most of the leached hulls, uncertainties in the (α,n) contribution to the neutron yield will be relatively unimportant. Thus, it is possible that singles neutron counting can be used to verify the plutonium content when the Pu/Cm ratio is obtained from the supplemental measurements. The coincidence count might be used mainly to verify that the chemical process is as expected.

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