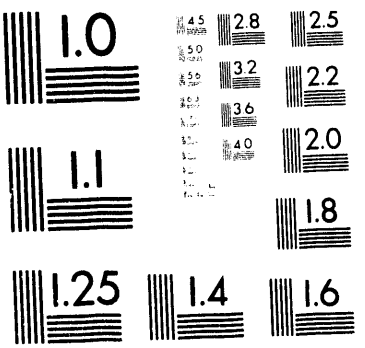
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EVALUATION OF IN-PLANT NEUTRON COINCIDENCE COUNTERS FOR THE MEASUREMENT OF MOLTEN SALT EXTRACTION RESIDUES*

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ABSTRACT

Americium is extracted from plutonium by a molten salt extraction (MSE) process. The residual americium-laden salts are a significant waste stream in this pyrochemical purification process. Rapid assay of MSE residues is desirable to minimize the exposure of personnel to these often high-level emissions. However, the quantitative assay of plutonium in MSE residues is difficult. Variable, unknown (α,n) rates and variable emitted-neutron energy spectra preclude the use of standard neutron coincidence counting techniques with old-generation neutron coincidence counters. Gamma-ray assay methods have not been successful with some residues because of random lumps of plutonium metal.

In this paper, we present measurements of MSE residues with two state-of-the-art neutron coincidence counters at the Los Alamos Plutonium Processing Facility: an in-line counter built for the assay of bulk waste material and the pyrochemical multiplicity counter that underwent test and evaluation at that facility. Both of these counters were designed to minimize the effects on measurements of variations in the sample geometry and variable energy spectra of emitted neutrons. These results are compared to measurements made with an HLNCII and with a 20-yr-old in-line well counter. The latter two counters are not optimized in this sense. We conclude that the newer counters provide significantly improved assay results. The pyrochemical multiplicity counter operated in the conventional coincidence mode provided the best assays overall.

INTRODUCTION

Molten salt extraction (MSE) residues are an important waste stream in recovering plutonium from scrap. These residues may contain up to several hundred grams of plutonium in a heterogeneous mixture of chloride salts. The plutonium exists in both chloride and metallic forms. The americium content of the salt is typically quite high: from a few to on the order of tens of weight percent relative to the plutonium. Thus, the radiation dose associated with these residues is also quite high.

Accountability numbers for these residues are difficult to obtain. Numbers obtained "by difference" (residue = feed value - product value) have large uncertainties because the residue value is small relative to the large values for the feed and the product. Destructive analysis to account for these residues is expensive and can result in significant radiation exposures to personnel. Gamma-ray assay techniques for accountability are often unsuccessful because of the lumpy nature of these salts,[1] and calorimetric techniques are hindered by the large americium content and heterogeneity of these residues.

Rapid assay of these salts is clearly desirable to minimize personnel radiation exposure. Assays using state-of-the-art neutron counting techniques are rapid, can be performed in the process line, and are not susceptible to problems resulting from sample inhomogeneity. In this work, we compare neutron assay results obtained for well-characterized MSE residues measured in four different neutron counters. The data used for these comparisons come from several MSE measurement studies and counter test and evaluations that have been performed at the Los Alamos Plutonium Processing Facility.[1-4]

DESCRIPTION OF THE NEUTRON COUNTERS

The neutron counters in which the measurements were made were as follows: a 20-yr-old well counter referred to as "N22"; an HLNCII; an in-line thermal-neutron coincidence counter built for Waste Isolation Pilot Plant (WIPP) certification measurements, referred to as the "WIPP" counter[6]; and the pyrochemical multiplicity counter.[7]

The N22 counter is a large, in-line thermal neutron counter built for the Los Alamos Plutonium Processing

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Facility to provide neutron measurements when the facility first started up. It has ample moderator but relatively poor uniformity in its response over its sample cavity.

The HLNCII is a much newer, portable, high-level neutron coincidence counter.[5] In this counter, aluminum is used in the end plugs to achieve greater uniformity in response as a function of sample position. However, the HLNCII is more sensitive to variations in the energies of neutrons emitted by a sample because it has a minimum of moderating material to reduce its weight.

The WIPP counter is part of an integrated gamma-ray neutron-coincidence counting assay system that was built for the Los Alamos Plutonium Processing Facility to measure and certify waste destined for storage at the WIPP. The sample well of the WIPP counter is larger than that of the HLNCII, and there is more moderator. Also the moderator is nonuniform in the axial dimension to further flatten the geometric response of the WIPP counter compared to the HLNCII.

Finally, the pyrochemical multiplicity counter (PMC) is a very high-efficiency counter designed to do in-plant thermal neutron multiplicity counting. In 1992 it underwent a short evaluation at the Los Alamos Plutonium Processing Facility.[4] This counter was designed using Monte Carlo calculations to provide uniformity in both spatial and energy response.

MSE RESIDUE MEASUREMENTS

On several occasions, MSE residues at the Los Alamos Plutonium Facility have been selected for careful characterization and study. Three sets of neutron measurements have been obtained. We will refer to these as Sets 1, 2, and 3. These data sets are given in Tables I, II, and III. The process used to characterize the residues is described in detail in Ref. 2. Briefly, the residues are pulverized and any metal chunks removed. The metal chunks are carefully oxidized to retain their plutonium mass, and then the oxide is returned to the pulverized residue. Finally, the residue is blended and sampled for destructive analysis.

Set 1 consists of measurements made on 14 residues before characterization. These measurements were performed in the HLNCII and N22 counters. After characterization, eight of these residues were remeasured in both of these counters and then, later, measured in the WIPP counter after it was delivered to the Los Alamos Plutonium Processing Facility. We will call these measurements Set 2.

Finally and most recently, eight more residues were selected and characterized. We will call these measurements Set 3. Five of these residues were low-mass residues representative of the MSE process before its recent refinement. The remaining three residues are from the "new generation" process whose by-product residues generally contain less plutonium and have less total mass. All of the residues measured in Sets 1 and 2 represent the "old generation" process. Because of the time scales over which these characterizations were done, these last eight residues were only available for measurement in the WIPP counter and the PMC. Also, the earlier residues were not available to the PMC.

MSE RESIDUE ASSAY COMPARISON

The neutron assay of MSE residues poses a difficult problem because of the variable matrix of this material. The matrix of these residues consists of elements that serve as good (α,n) targets for the high alpha-particle flux emitted by the large amounts of americium typically present. This results in a large, variable, random neutron rate relative to the rate for the plutonium in the material. The energy spectrum of these (α,n) neutrons will also vary from sample to sample as the matrix and density vary. Furthermore, for residues that contain larger amounts of plutonium or contain plutonium in dense layers or chunks, the random (α,n) neutrons induce fissions, and the coincidence rate increases in a variable and unpredictable manner.

Few standard neutron assay techniques succeed for materials such as these. The simplest method to assay materials whose matrix and self-multiplication are unknown employs a calibration using well-characterized standards and a quadratic relationship between the background-corrected coincidence rate and the effective mass of ^{240}Pu in a sample. This technique is appropriate if the self-multiplication in the sample is small and the (α,n) neutron rate is not varying too greatly. This technique is very sensitive to sample geometry if the detector is not properly optimized.

Other neutron assay techniques that have the potential for success for these residues also use the measured total neutron rate[8] and perhaps sample geometry information.[9] These techniques are sensitive to changes in the energy spectrum of the neutrons emitted by the sample as well as the sample geometry.

TABLE I. Measurement Data for Original MSE Samples

						Measurements in HLNCII			Measurements in N22		
Sample ID	Pu (g)	$^{239}Pu_{eff}$ (g)	$^{240}Pu_{eff}$ (g)	Can Radius (cm)	Fill Height (cm)	Reals (counts/s)	Sigma R (counts/s)	Totals (counts/s)	Reals (counts/s)	Sigma R (counts/s)	Totals (counts/s)
XBLP120	112.0	110.5	6.8	6.1	16.8	185.0	2.0	33 671	566.7	2.8	53 971
XBLP267	126.0	124.9	7.5	5.3	22.7	230.0	2.0	43 936	780.4	3.6	71 521
	126.0	124.9	7.5	5.3	22.7	228.0	2.0	44 009	772.4	3.5	71 454
	126.0	124.9	7.5	5.3	22.7	233.0	2.0	44 057			
	126.0	124.9	7.5	6.1	14.2	244.0	2.0	44 330	768.6	2.2	72 507
XBLPs300	199.0	193.6	12.3	5.3	19.6	278.0	1.0	15 940	808.0	0.7	26 398
	199.0	193.6	12.3	6.1	14.0	291.0	1.0	16 239	804.0	1.4	26 661
XBLP270	99.0	98.1	5.8	5.3	20.3	167.0	2.0	27 876	549.9	2.7	46 073
	99.0	98.1	5.8	6.1	13.6	179.0	1.0	28 683	546.3	2.4	46 745
XBLP121	155.4	153.9	9.0	5.3	28.4	262.0	2.0	47 123	855.4	4.0	76 127
	155.4	153.9	9.0	5.3	28.4	258.0	2.0	46 756			
	155.4	153.9	9.0	6.1	16.8	291.0	1.0	47 644	841.4	3.9	77 535
	155.4	153.9	9.0	6.1	16.8	296.0	2.0	47 943			
XBLPs301	247.0	240.3	15.2	5.3	22.1	356.0	1.0	17 001	1056.6	1.5	28 497
	247.0	240.3	15.2	5.3	22.1	357.0	1.0	16 917			
	247.0	240.3	15.2	5.3	22.1	367.0	1.0	17 122			
	247.0	240.3	15.2	6.1	14.1	381.0	1.0	17 355			
	247.0	240.3	15.2	6.1	14.1	386.0	1.0	17 652	1052.5	1.5	28 872
XBLP278	90.4	89.8	4.8	5.3	20.8	129.0	2.0	30 038	443.1	2.5	49 086
	90.4	89.8	4.8	6.1	14.2	138.0	2.0	30 541	432.5	1.3	49 567
	90.4	89.8	4.8	6.1	14.2	141.0	2.0	30 711			
	90.4	89.8	4.8	6.1	14.2	140.0	1.0	30 655			
RFMSE1	243.8	244.3	14.4	5.2	11.2	606.0	2.0	85 944	1792.2	7.5	142 763
	243.8	244.3	14.4	5.2	11.2	611.0	4.0	86 001	1776.5	7.1	141 627
RFMSE2	372.7	372.1	22.5	5.2	11.2	1079.0	5.0	100 443	3071.4	6.4	167 963
	372.7	372.1	22.5	5.2	11.2	1127.0	5.0	101 106	3099.8	6.2	167 222
RFMSE3	55.6	54.2	3.3	5.2	8.0	74.0	0.4	6 983	205.6	0.5	11 480
	55.6	54.2	3.3	5.2	8.0	75.2	0.2	6 990	205.4	0.5	11 475
	55.6	54.2	3.3	5.2	8.0	75.7	0.3	6 921	205.8	0.6	11 467
RFMSE4	408.7	407.8	24.3	5.2	11.2	1258.0	6.0	116 262	3593.9	6.0	194 123
	408.7	407.8	24.3	5.2	11.2	1287.0	7.0	117 192	3548.8	7.0	192 982
RFMSE5	141.2	137.4	8.4	5.2	8.1	205.0	1.0	11 203	541.0	1.0	18 491
	141.2	263.0	8.4	5.2	8.1	206.0	1.0	11 336	542.4	1.0	18 454
ARF595	263.6	263.0	15.9	5.2	11.2	643.0	4.0	78 661	1880.8	3.2	132 222
	263.6	263.0	15.9	5.2	11.2	657.0	4.0	79 333	1871.8	6.4	131 712
ARF642	219.5	218.8	12.0	5.2	11.2	464.0	3.0	67 779	1404.3	5.8	112 735
	219.5	218.8	12.0	5.2	11.2	473.0	3.0	68 088			

To assess each counter's performance in the measurement of MSE residues, we analyzed the neutron data in two or three ways depending on the information that was available about the residues. Assays were performed based on coincidence rate alone, coincidence rate and total neutron rate, and coincidence and total rates plus sample geometry information. This latter technique could not be applied to the data obtained from the PMC because sample geometric information was not available for the residues that were measured in it.

Assays were performed by deducing a calibration for the measurements from the reference values obtained by characterizing the residues. Then using these calibrations we deduced an assay value for each residue. We computed the standard deviation from the mean of the difference between assay results and reference values for each set in each counter for each assay method. Comparison of the standard deviations obtained for different counters for a set of measurements made on the same residues then provides a measure of how well the counters are performing relative to each other.

ASSAY BASED ON COINCIDENCE RATE ALONE

Table IV gives the results for assays based on neutron coincidence rate alone. Figures 1 and 2 illustrate the

TABLE II. Homogenized MSE Samples—Measurement Data Set 2

Sample ID	Can Radius (cm)	Fill Height (cm)	Measurements in HLNCII			Measurements in N22			Measurements in WIPP		
			Reals (counts/s)	Sigma R (counts/s)	Totals (counts/s)	Reals (counts/s)	Sigma R (counts/s)	Totals (counts/s)	Reals (counts/s)	Sigma R (counts/s)	Totals (counts/s)
XBLPs301	6.1	14.2	383.5	1.0	22 707	1 012.2	1.8	36 700	433.0	2.8	24 285
RFMSE1	6.1	7.9	667.0	2.0	95 181	1 891.3	7.9	154 953	720.4	4.8	101 039
RFMSE2	6.1	7.6	1 149.0	4.0	116 052	3 268.1	9.5	189 763	1 291.5	7.2	122 998
RFMSE3	6.1	8.6	74.9	0.3	7 989	205.7	0.7	12 820	87.6	0.7	8 407
RFMSE4	5.2	7.9	1 408.5	7.0	141 393	3 886.7	11.3	230 655	1 480.4	6.7	149 418
RFMSE5	6.1	9.4	196.0	1.0	13 695	541.3	1.0	22 485	225.8	2.0	14 464
ARF595	5.2	9.5	662.0	2.0	86 343	1 916.1	6.7	141 453	717.6	1.7	91 643
ARF642	5.2	9.2	535.0	3.0	80 466	1 546.7	6.5	131 701	570.7	16.4	84 789

TABLE III. MSE Samples—Measurement Data Set 3

Sample ID	Pu (g)	^{239}Pu-eff (g)	^{240}Pu-eff (g)	Measurements in WIPP			Measurements in PMC		
				Reals (counts/s)	Sigma R (counts/s)	Totals (counts/s)	Reals (counts/s)	Sigma R (counts/s)	Totals (counts/s)
"Old Generation" MSE Residues									
XBLP 272	109.9	107.1	5.84	184.0	2.2	32 477	1 014	14	83 772
XBLP 280	158.6	154.3	9.13	306.4	2.2	32 412	1 663	26	86 252
XBLP 265	168.7	164.4	9.11	332.8	3.6	52 649	1 851	62	136 789
XBLP 260	108.4	105.7	5.65	191.8	2.1	43 338	1 049	34	112 275
XBLP 268	115.3	112.2	6.34	196.8	2.2	32 237	1 070	14	82 786
"New Generation" MSE Residues									
XBLS 97	42.1	41.07	2.24	68.8	0.6	8 226	366	6	24 975
XBLS 96	86.5	84.37	4.59	131.2	0.9	13 010	743	10	49 135
XBLS 94	130.6	127.4	6.78	259.0	2.1	25 860	1 397	10	64 808

calibration technique. In these figures, the background-corrected neutron coincidence rate is plotted vs the ^{240}Pu-effective mass for the eight characterized residues measured in the old N22 and the new WIPP counter.

The differences in the spatial characteristics of each counter are evident in these results. In the figures, it is evident that although the scatter in the data is primarily caused by variations in the induced-fission component of the coincidence rate, the counter also affects the scatter. From the table, the old N22 performs worse for the residues before characterization (Set 1) because of the non-uniformity of its spatial response. When the residues are homogeneous (Set 2), the N22 performs slightly better. The HLNCII, on the other hand, does almost equally well whether the samples are chunky or not. The WIPP counter measured the homogeneous residues better than either the N22 or the HLNCII. This is due to the WIPP counter's excellent, uniform response.

Finally, the PMC performed slightly better than the WIPP counter on the final eight residues (Set 3). This is probably due to the larger sample cavity of the PMC relative to the WIPP counter, which in turn results in a more uniform radial response over the sample.

TABLE IV. Percent Standard Deviation from the Mean in Assay Results Relative to Reference Values Based on Neutron Coincidence Rate

Counter	Set 1	Set 2	Set 3
N22	16.3	14.9	N/A
HLNCII	13.9	13.8	N/A
WIPP	N/A	12.9	6.6
PMC	N/A	N/A	5.8

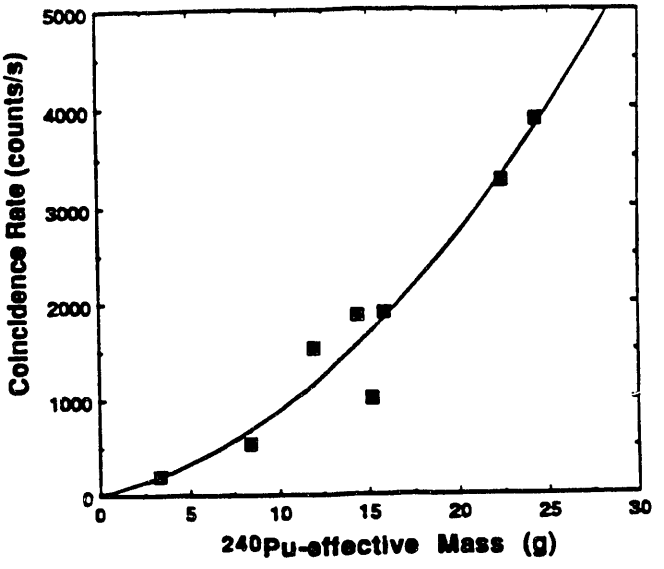

Fig. 1. The measured neutron coincidence rate vs ^{240}Pu-effective mass for eight homogeneous, characterized MSE residues measured in the N22 counter.

ASSAY BASED ON COINCIDENCE RATE AND TOTAL NEUTRON RATE

Table V gives the assay results obtained with a "Known M" assay technique. This method uses both the total neutron rate and the coincidence rate to deduce an assay. Calibration parameters for this technique were deduced by calculating the self-multiplication for each residue from the measured neutron rates and known ^{240}Pu-effective mass, relating this multiplication to the known ^{239}Pu-effective mass of the residue, and finally applying the multiplication correction equations given by Ensslin.[10] The Known M assay method is described in more detail in Ref. 8.

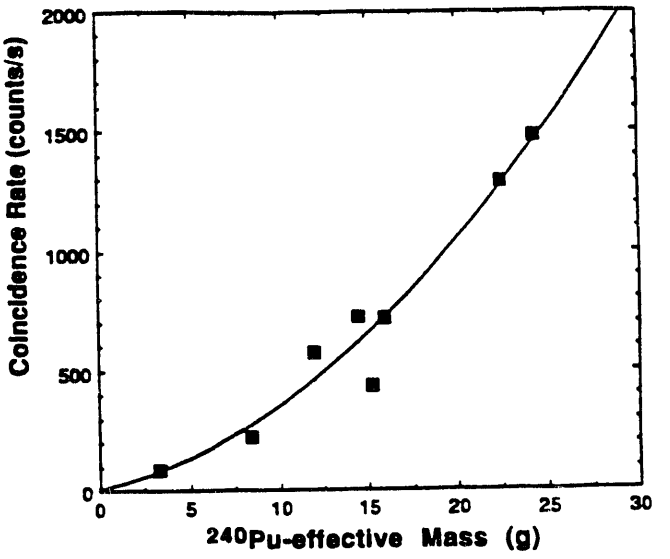

Fig. 2. The measured neutron coincidence rate vs ^{240}Pu-effective mass for eight homogeneous, characterized MSE residues measured in the WIPP counter.

TABLE V. Percent Standard Deviation from the Mean in Assay Results Relative to Reference Values Based on Coincidence and Total Neutron Rates

Counter	Set 1	Set 2	Set 3
N22	7.7	6.5	N/A
HLNCII	9.9	9.7	N/A
WIPP	N/A	6.3	12.2
PMC	N/A	N/A	10.0

From these results we conclude that the MSE residues will assay best in counters that are optimized relative to their response to neutrons of different energies. The N22, which is amply moderated, performs better for this kind of assay method than does the undermoderated HLNCII. The nonuniformity of the N22's spatial response is still evident, however, if one compares the results for the residues after homogenizing to those obtained before. Again the WIPP counter performs the best for the homogenized residues, and the PMC provides the best assays for the final eight residues in measurement Set 3.

It is interesting to compare these Known M assays to those obtained using the coincidence rate alone. For the MSE residues measured in Sets 1 and 2, the assays that use the information from the total neutron rate are much

improved over those that used only the coincidence rate. However, for those measured in Set 3, the opposite is true. This is due to the nature of the samples measured in Set 3. Figure 3 gives the background-corrected neutron coincidence rates plotted vs the ^{240}Pu-effective mass for measurements made in the PMC. First, this residue set has less plutonium mass and thus less self-multiplication. The fit of these data is more linear than in Figs. 1 and 2. Thus, errors due to induced fission will be smaller. The residues measured in Set 3 also, in general, contain more americium relative to the plutonium than the earlier residues. Thus, for these residues, the ratio of (α,n) neutrons to spontaneous-fission neutrons, frequently called simply "α," is larger. The error in a Known M assay is strongly dependent on α. For a given self-multiplication and its accuracy, the assay accuracy will decrease as α increases. Thus, the residues measured in Set 3 by this method are worse than assays that use the coincidence rate alone.

ASSAY BASED ON COINCIDENCE RATE, TOTAL NEUTRON RATE, AND SAMPLE GEOMETRY

A variation in the Known M approach uses the sample dimensions as an additional piece of information from which to deduce an assay. This "Geometry-Based, Multiplication Correction" method is described in Ref. 9.

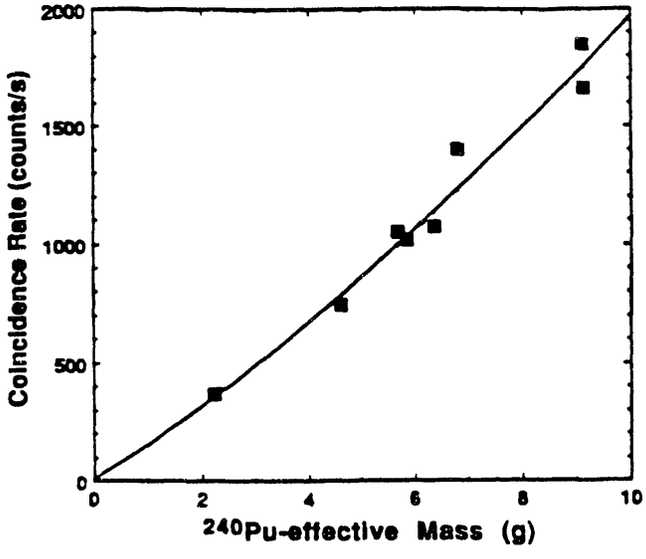

Fig. 3. The measured neutron coincidence rate vs ^{240}Pu-effective mass for eight low-mass, characterized MSE residues measured in the pyrochemical multiplicity counter.

Table IV gives these assay results for the measurement sets for which sample dimensions were available. The additional step needed to get this information about the residues increases personnel exposure to high-level emissions. However, this measurement does not need to be very precise for the technique to yield good results.

From Table VI, the same counter effects seen in the assays based on coincidence rate alone are evident; however, the results are greatly improved. That the assays obtained with the HLNCII are better than those from the N22 suggests that this assay method is not as sensitive to the energy response characteristics of the counter as the Known M method.

TABLE VI. Percent Standard Deviation from the Mean in Assay Results Relative to Reference Values Based on Coincidence Rate, Total Neutron Rate, and Sample Geometry

Counter	Set 1	Set 2	Set 3
N22	10.0	6.5	N/A
HLNCII	5.4	5.2	N/A
WIPP	N/A	2.3	N/A
PMC	N/A	N/A	N/A

CONCLUSIONS

The accuracy of neutron coincidence assay of MSE residues is strongly dependent on the characteristics of the counter used for the measurements. The best assays will be obtained with counters that have been optimized with respect to their spatial characteristics and their energy response.

The results presented here also strongly suggest that the requirements for counter optimization depend on the type of assay method that will be applied to the data and the desired accuracy in the assay. For waste materials that usually do not require very precise measurement, but rather need to be certified to contain less than some threshold amount of plutonium, less optimal counters may be adequate. For example, the HLNCII, which is an inexpensive counter, measured these residues quite well when sample dimension information was added.

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