Field Test and Calibration of Neutron Coincidence Counters for High-Mass Plutonium Samples

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FOR HIGH-MASS PLUTONIUM SAMPLES

by

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ABSTRACT

Five different neutron coincidence systems were evaluated and calibrated for high-mass PuO_2 samples. The samples were from 2 to 7.2 kg of PuO_2 in mass, with a large range of burnup. This report compares the equipment and the results, with an evaluation of deadtime and multiplication corrections.

I. INTRODUCTION

During the past several years, new equipment has been developed to measure high-mass plutonium samples using neutron time-correlation (coincidence) counting. It was necessary to test this equipment and calibrate it for high-mass (2 to 7.2 kg) plutonium samples. To this end, an exercise was organized at the Sellafield Nuclear Facility in the United Kingdom during the week of January 14, 1985. The following organizations participated in the exercise:

BNFL	British Nuclear Fields, Risley, Warrington, Cheshire, United Kingdom, and Sellafield;
NMACT	Nuclear Materials Accounting Control Team, Harwell, United Kingdom;
EURATOM	Directorate of EURATOM Safeguards, Kirchberg, Luxembourg;
IAEA	International Atomic Energy Agency, Vienna, Austria; and
Los Alamos	Los Alamos National Laboratory, Los Alamos, New Mexico, United States.

The exercise at Sellafield had two primary purposes:

- to evaluate the performance of the gamma-ray and the neutron coincidence detectors for plutonium assay at very high counting rates and
- to calibrate several nondestructive assay systems for PuO_2 sample masses from 2 to 7.2 kg.

These two goals are covered in separate sections of the report. The first section covers the topics of neutron detector comparison, deadtime, bias, geometric effects, and data consistency.

The second section covers the calibration, multiplication corrections, moisture effects, and outliers. The more detailed multiplication and calibration results are presented for only one of the neutron detector systems. These results are correlated with prior results obtained at Los Alamos National Laboratory and Belgonucleaire in Belgium using the same detector but covering a lower range of mass.

The high precision and consistency of the neutron detector results made it possible to isolate all outliers as "sample problems" and not "instrument

problems." That is, all instruments identified the same three to four samples as outliers, and the direction and magnitude of the discrepancies were used to help evaluate the source of the problem.

This report gives the neutron results but not the gamma-ray spectroscopy results for plutonium isotopic ratios. The preliminary gamma-ray results have been evaluated separately by EURATOM and BNFL. The plutonium isotopic ratios supplied by the facility operator were used for all of the data reduction in this report.

II. EQUIPMENT DESCRIPTION

Five neutron counters and two gamma-ray systems were used in the exercise. These seven counting systems are listed in Table I and described in this section.

All of the samples were measured in each of the neutron systems, which provided a good check of the consistency and precision of the different systems. The sample masses and neutron counting rates were higher than had been measured previously. These high rates gave a good check on the validity of deadtime and multiplication corrections. A comparison of the detector results is given in Sec. V.

TABLE I
EQUIPMENT USED FOR THE PuO_2 MEASUREMENTS AT SELLAFIELD

<u>Item</u>	<u>Signal</u>	<u>Organization</u>	<u>Detectors</u>	<u>Coincidence Electronics</u>
Hex counter	Neutrons	BNFL	12-- ^3He tubes	Computer chip
PPNCC	Neutrons	NMACT	18-- ^3He tubes	Harwell shift register
HLNC-II (JCC-31)	Neutrons	IAEA (1)	18-- ^3He tubes	Shift register (JSR-11)
HLNC-II (JCC-31)	Neutrons	IAEA (2)	18-- ^3He tubes	Shift register (JSR-11)
HLNC-II (Los Alamos)	Neutrons	EURATOM/Los Alamos	18-- ^3He tubes	Shift register (HEC-100)
Ge (EURATOM)	Gamma rays	EURATOM	Ge	--
Ge (BNFL)	Gamma rays	BNFL	Ge	--

A. HLNCC-II

The HLNCC-II* detector is a portable neutron counter developed¹ for high-mass plutonium samples. A schematic diagram of the detector is shown in Fig. 1, and details on the system are given in Ref. 1. Three of these systems were used to obtain independent calibration data and to study differences between the systems and electronics. The two IAEA systems used the new commercial shift register (JSR-11) produced by JOMAR Systems** based on the Los Alamos design. These units used the AMPTEK[†] amplifiers (six per system) and a derandomizing buffer in the shift-register board. The older EURATOM/Los Alamos system used an IRT^{††} (HEC-100) shift register that had been modified to operate the AMPTEK detector head.

Each of the three systems had a Hewlett-Packard HP-85 minicomputer for data collection and analyses using the software program CAL4 (Ref. 2).

B. PPNCC

The portable passive neutron coincidence counter (PPNCC) system has been used by NMACT at Harwell for several years for the coincidence counting of bulk plutonium samples. This system is hexagonal with three ³He tubes in each side, giving a total of 18 tubes. The sample cavity is larger than that of the HLNC-II, resulting in more neutron leakage and a smaller detection efficiency. The system uses conventional preamplifiers and amplifiers (six each) developed at Harwell, and the electronic deadtime is similar to the older HLNC-I.³

Fig. 1. Diagram of HLNC-II.

*Further usage of this acronym in this report will be the IAEA four-letter code HLNC-II.

**JOMAR Systems, 1143 18th Street, Los Alamos, NM 87544.

[†]AMPTEK, Inc., 6 DeAngelo Drive, Bedford, MA 01730.

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The deadtime used during the Sellafield exercise was found to be too small, so it was subsequently modified⁴ by Harwell staff. Only the modified data results are used in this report.

C. BNFL Coincidence System

The BNFL counter has a hexagonal detector head similar in size to that of the HLNC-I, but it had the following changes:

- The detector walls were thicker than those of the HLNC-I and had only two ³He tubes in each side.
- Each of the 12 detector tubes was connected to its individual preamplifier and amplifier to reduce deadtime.
- The BNFL coincidence electronics replaced the shift register with a computer chip and a fast clock. These replacements allowed more flexibility in selecting the predelay and gate width.

III. SAMPLE DESCRIPTION

The facility operator provided a variety of high-mass plutonium samples for the measurements. These samples covered a wide range of age since separation and ²⁴¹Am content.

Table II describes the samples. The ²⁴⁰Pu-effective is defined as

$$^{240}\text{Pu-effective} = 2.49 M_{238} + M_{240} + 1.57 M_{242} ,$$

where M_{238} , M_{240} , and M_{242} are the masses of ²³⁸Pu, ²⁴⁰Pu, and ²⁴²Pu. The parameter α is defined as the ratio of (α ,n) neutrons to spontaneous-fission neutrons as calculated from the plutonium isotopics and the (α ,n) yields from pure PuO₂ (see Sec. VII.A).

TABLE II

PuO₂ SAMPLE SPECIFICATIONS
(Updated to January 1, 1985)

Sample No.	Pu (g)	²³⁸ Pu (%)	²³⁹ Pu (%)	²⁴⁰ Pu (%)	²⁴¹ Pu (%)	²⁴² Pu (%)	²⁴¹ Am (ppm)	²⁴⁰ Pu-eff. (g)	α	²³⁹⁺²⁴¹ Pu (g)
1	7 156.5	0.188	72.09	22.87	3.91	0.943	2 978	1 776	0.365	5 439
2	6 807.4	0.188	71.22	23.59	4.00	1.004	4 162	1 745	0.368	5 120
3	6 704.5	0.200	70.35	23.83	4.59	1.041	670	1 740	0.334	5 024
4	6 531.7	0.118	73.02	22.45	3.53	0.874	5 057	1 575	0.359	5 000
5	6 254.6	0.114	75.80	20.97	2.49	0.638	12 210	1 391.7	0.467	4 897
6	5 951.9	0.135	74.78	21.52	2.84	0.737	9 973	1 369	0.440	4 620
7	5 753.9	0.124	75.54	21.05	2.61	0.678	11 680	1 289.9	0.463	4 497
8	5 647.4	0.133	75.03	21.47	2.64	0.729	12 496	1 295.8	0.469	4 386
9	5 432.9	0.105	75.01	21.46	2.69	0.738	13 748	1 243	0.434	4 221
10	5 332.6	0.113	76.47	20.62	2.17	0.628	12 817	1 167	0.481	4 194
11	4 968.4	0.469	76.65	17.66	3.71	1.512	28 143	1 053	0.883	3 993
12	4 768.0	0.113	75.88	20.96	2.39	0.658	12 813	1 062	0.474	3 732
13	4 597.6	0.114	75.57	21.11	2.56	0.649	13 173	1 030	0.476	3 592
14	4 488.4	0.122	76.72	20.34	2.21	0.608	12 845	969.5	0.492	3 543
15	3 811.6	0.113	75.47	21.31	2.40	0.710	14 069	865.4	0.480	2 968
16	3 821.8	0.178	73.12	22.35	3.51	0.842	2 904	921.5	0.368	2 929
17	2 671.8	0.915	67.55	23.32	4.59	3.63	35 452	836.0	0.855	1 927
18	2 541.3	0.946	66.31	22.90	6.28	3.57	47 630	784.2	0.979	1 845
19	2 476.0	0.915	67.55	23.32	4.59	3.63	35 452	774.8	0.955	1 786

The samples were all doubly encapsulated in metal cans, with a polyethylene bag around the inside can. The substantial wall thickness of the cans resulted in a correction of

$$\frac{T(\text{Sellafield can})}{T(\text{negligible wall thickness})} = 1.013$$

and

$$\frac{R(\text{Sellafield can})}{R(\text{negligible wall thickness})} = 1.027$$

These wall-thickness corrections were not made to the comparison data, but the corrections were made before doing the calibration fit for the EURATOM/Los Alamos detector. This method makes it possible to compare the results with the prior data where the wall thicknesses were negligible or corrected for.

IV. EXPERIMENTAL PROCEDURE

For the measurements, all of the equipment was set up in a shielded room near the storage area as shown in Fig. 2. The room had thick concrete walls with inside dimensions of ~4 by 8 m, and the average distance between detectors was ~2 m. Each sample was counted in each detector for approximately 3 x 200 s, or a total of 10 min per system. The sample was then moved clockwise to the next detector as shown in Fig. 2. The gamma-ray counts typically took 20 min. Only one sample was allowed in the room at a time for criticality and safety reasons. Thus, each sample required about 1.5 h to complete the cycle.

Normalization measurements using two different ^{252}Cf sources were made in the detectors that were not occupied by the PuO_2 sample. The ^{252}Cf sources were D-372 from Los Alamos and Cf-2 from Harwell. They had the following yields (based on Los Alamos ^{252}Cf source CR-5):

$$\text{D-372} = 9.00 \times 10^3 \text{ n/s on 85-02-01 , (year-month-day)}$$

and

$$\text{Cf-2} = 7.39 \times 10^4 \text{ n/s on 85-02-01 .}$$

Room-background neutron rates were measured both with and without the PuO_2 sample in the room. Essentially all of the background was from the PuO_2 assay sample, so the detector containing the sample had negligible background (~50 counts/s) from other sources during the PuO_2 assay. However, for some of

Fig. 2. Position of nondestructive assay equipment in the measurement room at Sellafield.

the ^{252}Cf measurements, a significant background was induced from the large PuO_2 sample in one of the neighboring detectors. The normal data reduction included an appropriate room-background subtraction.

Separate measurements were performed to determine (1) the effect of the vault-wall reflection on the neutron counting efficiency and (2) the variability in can positioning in the detector sample cavity.

Gamma-ray dose measurements were made to determine the exposure to the operators and the ^3He detectors. In general, the gamma dose in the tubes should not get above ~ 1 R/h to avoid gamma-ray pileup into the neutron spectrum. This limit differs for the three types of systems. That is, the shorter amplifier time constants ($\sim 0.15 \mu\text{s}$) in the AMPTEK-based systems (HLNC-II) give better neutron/gamma-ray discrimination. Also, the large number (12) of amplifiers on the BNFL system reduces the gamma-ray pileup problem. When gamma-ray pileup is a problem (above the neutron discrimination threshold), the observed totals rate increases and the coincidence rate decreases.

V. SYSTEM COMPARISON RESULTS

A. Operating Parameters

Each of the neutron systems had different operating parameters, although the three HLNC-II systems were almost identical. Table III lists the five neutron systems and their totals efficiencies, counting and predelay gates, die-away times, and deadtime coefficients.

The efficiencies were determined by a calibrated ^{252}Cf source (CR-5)¹ measured at Los Alamos in IAEA-1, and the totals rate ratio of the other counters was compared with the ratio for this unit for source D-372 measured at Sellafield.

TABLE III
OPERATING PARAMETERS FOR THE NEUTRON DETECTORS

Counter	Predelay (μ s)	Gate (μ s)	Totals, ϵ (%)	Coincidence Deadtime, δ (μ s)	Totals Deadtime
IAEA-1 (blue)	4.5	64	18.3	$0.68 + 0.22 \cdot 10^{-6} T$	$\delta/4$
IAEA-2 (silver)	4.5	64	17.9	$0.62 + 0.20 \cdot 10^{-6} T$	$\delta/4$
EURATOM/Los Alamos (silver)	4.5	64	17.5	$0.68 + 0.22 \cdot 10^{-6} T$	$\delta/4$
NMACT (PPNCC)	4.5	32	18.5	$2.8 + 0.0 T$	$\delta/4$
BNFL (hex)	4.5	95.94	11.9	$1.38 + 0.58 \cdot 10^{-6} T$	$>\delta/4.5$

The deadtime coefficients were obtained for each detector system either before or after the Sellafield exercise. This was done using ^{252}Cf and/or AmLi sources. Because the Sellafield measurements pushed the counting rates to higher than normal levels ($\sim 500\,000\text{ s}^{-1}$), checking for consistency in the deadtime coefficients between the different systems was possible. The coincidence deadtime is about four times larger than the totals deadtime, so an error in the deadtime shows up in the comparison of R/T ratios and in the high-rate and low-rate sample ratios.

B. Experimental Data

The basic counting rates before and after multiplication corrections are listed in Tables IV-VIII. All of the rates have been corrected for room background (small) and deadtime using

$$R_c = R(\text{meas})e^{\delta T(\text{meas})}$$

and

$$T_c = T(\text{meas})e^{\delta T(\text{meas})},$$

TABLE IV
NEUTRON DATA SUMMARY FOR HLNC-II COUNTER IAEA-1

Sample	Deadtime Correction (a)	T_c (s ⁻¹) (b)	R_c (s ⁻¹) (b)	Multiple Correction R_{mc}	$\frac{R_{mc}}{q}$	M_L (c)
1	1.504	561 138	102 162	32 529	18.31	1.302
2	1.493	551 788	99 393	32 017	18.35	1.297
3	1.482	541 445	99 177	32 180	18.49	1.299
4	1.444	506 150	90 174	29 700	18.86	1.291
5	1.405	469 342	76 806	25 879	18.59	1.273
6	1.377	449 219	70 415	25 678	18.76	1.251
7	1.374	439 651	65 634	25 037	18.29	1.236
8	1.361	427 097	65 313	24 031	18.54	1.246
9	1.337	403 124	59 789	23 562	18.96	1.229
10	1.318	383 376	55 266	21 579	18.49	1.227
11	1.378	443 365	57 298	19 650	18.66	1.234
12	1.306	370 803	48 973	21 753	20.48	1.192
13	1.268	331 377	44 861	19 259	18.70	1.201
14	1.259	321 649	44 568	18 326	18.90	1.212
15	1.214	272 214	34 488	16 082	18.58	1.178
16	1.216	273 454	40 485	16 914	18.35	1.217
17	1.285	348 737	34 789	16 882	20.19	1.147
18	1.275	339 041	34 842	15 102	19.26	1.168
19	1.267	329 937	33 428	15 907	20.53	1.152
Average					18.91	
Scatter (1σ)					±0.71	
1σ(%)					±3.7	
CF (D-372)	1.001	1 624	317.9			

(a) The coincidence deadtime coefficient was $\delta = (0.68 + 0.22 T \cdot 10^{-6}) \mu s$.

(b) The data have been corrected for wall reflection by 1.015 for totals and 1.03 for reals.

(c) M_L = leakage multiplication.

TABLE V
NEUTRON DATA SUMMARY FOR HLNC-II COUNTER IAEA-2

Sample	Deadtime Correction (a)	T_C (s ⁻¹)	R_C (s ⁻¹)	Multiple Correction R_{mc}	$\frac{R_{mc}}{g}$	M_L (b)
1	1.435	549 062	97 864	32 055	18.05	1.292
2	1.426	539 365	94 787	31 558	18.08	1.287
3	1.417	529 737	94 564	31 762	18.25	1.288
4	1.385	496 389	86 096	29 389	18.66	1.280
5	1.350	459 164	73 627	25 485	18.31	1.265
6	1.335	441 746	67 306	25 484	18.62	1.240
7	1.325	430 581	62 403	24 750	19.19	1.225
8	1.314	419 202	63 049	23 711	18.30	1.240
9	1.293	395 194	57 576	23 224	18.69	1.220
10	1.276	374 938	53 280	21 385	18.33	1.220
11	1.328	434 871	54 786	19 414	18.44	1.225
12	1.266	363 353	46 958	21 455	20.20	1.184
13	1.233	324 105	42 630	19 004	18.45	1.190
14	1.226	315 708	43 033	18 078	18.65	1.206
15	1.187	267 028	33 400	15 837	18.30	1.173
16	1.189	268 749	38 818	16 752	18.18	1.208
17	1.248	341 519	33 530	16 601	19.86	1.142
18	1.240	332 811	33 565	14 896	18.79	1.163
19	1.234	324 701	32 032	15 764	20.34	1.144
Average					18.73	
Scatter (1σ)					±0.69	
1σ(%)					±3.7	
CF (D-372)	1.001	1 600	308.0			

(a) The coincidence deadtime coefficient was $\delta = (0.62 + 0.2 T \cdot 10^{-6}) \mu s$.

(b) M_L = leakage multiplication.

TABLE VI

NEUTRON DATA SUMMARY FOR HLNC-II COUNTER EURATOM/LOS ALAMOS

Sample	Deadtime Correction (a)	T_c (s ⁻¹)	R_c (s ⁻¹)	Multiple Correction R_{mc}	$\frac{R_{mc}}{q}$	M_l (b)
1	1.470	538 267	93 726	31 670	17.83	1.282
2	1.462	530 964	92 168	31 190	17.87	1.282
3	1.450	519 551	90 763	31 373	18.03	1.278
4	1.415	486 152	82 667	28 971	18.39	1.271
5	1.379	451 564	72 054	25 101	18.03	1.263
6	1.359	430 887	64 535	24 989	18.25	1.234
7	1.352	424 253	61 172	24 424	18.93	1.223
8	1.336	407 951	59 916	23 247	17.94	1.230
9	1.314	385 079	54 754	22 807	18.35	1.213
10	1.298	368 076	51 459	20 914	17.92	1.215
11	1.353	425 209	52 723	19 067	18.11	1.220
12	1.285	354 233	44 850	21 048	19.82	1.177
13	1.250	316 010	40 808	18 630	18.09	1.184
14	1.244	309 202	41 610	17 775	18.33	1.201
15	1.202	262 131	32 142	15 637	18.07	1.166
16	1.203	263 410	37 669	16 473	17.88	1.204
17	1.265	332 751	32 257	16 229	19.41	1.139
18	1.259	326 596	32 524	14 667	18.70	1.159
19	1.251	317 652	31 214	15 438	19.93	1.143
Average					18.41	
Scatter (1 σ)					± 0.65	
1 σ (%)					± 3.5	
Cf (D-372)	1.001	1 581	297.7			

(a) The coincidence deadtime coefficient was $\delta = (0.68 + 0.22 T \cdot 10^{-6}) \mu s$.

(b) M_l = leakage multiplication.

TABLE VII
NEUTRON DATA SUMMARY FOR NMACT COUNTER PPNC

Sample	Deadtime Correction (a)	T_c (s ⁻¹)	R_c (s ⁻¹)	Multiplication Correction (b)	R_{mc} (b)	M_L (b)	R_{mc} (c)	M_L (c)
				R_{mc}	$\frac{R_{mc}}{q}$			
1	1.997	298 543	13 045	8 504	4.79	1.097	4 949	1.246
2	1.996	298 470	12 925	8 509	4.88	1.094	4 949	1.243
3	1.975	292 889	13 089	8 538	4.91	1.098	4 952	1.250
4	1.910	275 708	12 594	7 798	4.95	1.110	4 527	1.263
5	1.840	256 907	10 745	6 792	4.88	1.100	3 964	1.246
6	1.807	247 856	10 117	6 768	4.94	1.088	3 941	1.232
7	1.784	241 661	8 884	6 655	5.16	1.061	3 894	1.196
8	1.759	234 926	9 454	6 274	4.84	1.089	3.664	1.231
9	1.715	222 656	8 710	6 151	4.95	1.075	3 605	1.215
10	1.676	211 773	8 134	5 669	4.86	1.077	3 291	1.217
11	1.799	245 794	6 851	5 319	5.05	1.046	3 184	1.156
12	1.655	205 755	7 117	5 703	5.37	1.046	3 349	1.176
13	1.679	212 436	11 043	5 246	5.09	1.173	3 035	1.338
14	1.561	178 851	6 982	4 727	4.88	1.083	2 765	1.223
15	1.473	153 219	5 082	4 270	4.93	1.036	2 511	1.162
16	1.470	152 278	6 491	4 362	4.73	1.089	2 537	1.237
17 ^(d)	1.623	(196 696)	(3 678)	(4 714)	(5.64)	(0.96)	(2 860)	(1.046)
18	1.705	219 839	6 896	4 376	5.58	1.083	2 605	1.202
19	1.679	212 661	6 578	4 584	5.92	1.067	2 726	1.186
Average					5.04			
Scatter (1σ)					±0.30			
1σ(%)					±5.6			
Cf (D-372)		940	76.0					
Cf-2		8 100	624					

(a) The coincidence deadtime coefficient was $\delta = 2.8 \mu\text{s}$.

(b) M_g = leakage multiplication; multiplication calculated by NMACT with $\rho_0 \approx 0.0427$.

(c) Multiplication calculated using IAEA CAL4 program with $\rho_0 \approx 0.0282$.

(d) Faulty measurement with the NMACT counter omitted from average.

TABLE VIII
NEUTRON DATA SUMMARY FOR BNFL COUNTER

Sample	Deadtime Correction (a)	T_c (s ⁻¹)	R_c (s ⁻¹)	Multiple Correction (b) R_{mc}	$\frac{R_{mc}}{q}$ (b)	R_{mc} (b)	M_q (b)
1	1.660	363 926	43 132	12 054	6.79	13 638	1.304
2	1.648	359 078	42 185	11 963	6.86	13 460	1.301
3	1.632	351 702	41 899	12 006	6.90	13 516	1.301
4	1.580	328 476	36 740	11 195	7.11	12 611	1.278
5	1.532	306 597	32 300	9 708	6.97	10 968	1.271
6	1.513	297 455	30 263	9 846	7.19	11 003	1.252
7	1.494	288 294	27 638	9 573	7.42	10 660	1.233
8	1.477	280 259	27 858	9 106	7.03	10 200	1.248
9	1.449	266 270	25 777	9 034	7.27	10 053	2.232
10	1.415	249 274	22 653	8 319	7.13	9 241	1.215
11	1.490	286 638	23 006	7 519	7.14	8 324	1.220
12	1.403	243 163	20 403	8 461	7.97	9 291	1.185
13	1.353	217 238	18 734	7 453	7.24	8 217	1.195
14	1.332	205 898	17 754	6 969	7.19	7 689	1.197
15	1.278	176 420	14 145	6 369	7.36	6 796	1.170
16	1.276	175 036	16 154	6 532	7.09	7 096	1.202
17	1.372	227 469	14 606	5 316	6.36	7 142	1.145
18	1.354	217 450	13 653	5 353	6.83	6 367	1.151
19	1.343	211 968	13 127	5 559	7.17	6 718	1.135
Average					7.10		
Scatter (1 σ)					± 0.32		
1 σ (%)					± 4.5		
cf (D-372)			128.9				

(a) The coincidence deadtime coefficient was $\delta = (1.38 + 0.58 T \cdot 10^{-6}) \mu s$.

(b) The multiplication-corrected rates and leakage multiplication (M_q) are based on the IAEA CAL4 program (new method) and $\rho_0 = 0.0667$.

where the coincidence coefficient δ and the totals deadtime coefficient δ_T are given in Table III. A slightly different equation was used for the BNFL counter totals correction.

The counter IAEA-1 was positioned close to the concrete wall, and an empirical correction (see Sec. V.H) was applied to the data listed in Table IV. The measured results were divided by 1.015 for the totals rates and 1.03 for the reals.

The data from sample 17 in the PPNCC counter were faulty by a factor of ~ 2 in the coincidence rate and a factor of 1.14 in the totals rate. The original data were examined and all six runs of 100 s each were consistently low, so the faulty data were not caused by a transmission/recording error or a momentary electronic failure. The sample may have been mispositioned, or the lid may have been left off the counter, or gamma-ray pileup problems occurred. This result was omitted from subsequent averages and comparisons.

Details concerning the multiplication results are given in Sec. VII. The reals rate after multiplication corrections (R_{mc}) should be a linear function of the spontaneous-fission rate or of $g^{240}\text{Pu}$ -effective. Thus, the R_{mc}/g ratio should be a constant. If there is moisture in the sample or an (α, n) contaminant such as fluorine, then this ratio, as well as the R_c/g ratio, will be high. Four of the systems indicated that samples 12, 17, and 19 gave high results.

C. Totals Rate Ratios

One parameter of interest in the consistency checks is the ratio of totals counting rates in the different systems for the same PuO_2 sample and the much weaker ^{252}Cf source. These consistency checks are primarily on the totals deadtime correction, sample positioning, and electronic stability. However, the PuO_2 cans have different fill heights, so if the detector efficiency varies as a function of sample fill height, this variance can also affect the ratios.

Table IX gives the ratios of counter IAEA-2 to all of the other counters. The IAEA-2 counter was chosen as the reference counter because it has the smallest deadtime coefficient, and it was carefully measured before the exercise.

The large totals rates ($150\,000$ to $550\,000\text{ s}^{-1}$) and long counting times ($\sim 600\text{ s}$) resulted in very small statistical errors in the totals rates. The

TABLE IX
CONSISTENCY RATIOS FOR DEADTIME-CORRECTED TOTALS AND REALS RATES
FOR THE FIVE NEUTRON SYSTEMS

Sample No.	$\frac{\text{IAEA-2 } T_c}{T_c}$					$\frac{\text{IAEA-2 } R_c}{R_c}$				
	IAEA-2					IAEA-2				
	T_c (s ⁻¹)	IAEA-1	EURATOM/ Los Alamos	PPMCC	BNFL	R_c (s ⁻¹)	IAEA-1	EURATOM/ Los Alamos	PPMCC	BNFL
1	549 062	0.9785	1.020	1.839	1.509	97 864	0.9579	1.044	7.502	2.269
2	539 365	0.9775	1.016	1.807	1.502	94 787	0.9537	1.028	7.334	2.247
3	529 737	0.9784	1.020	1.809	1.506	94 564	0.9535	1.042	7.225	2.257
4	496 389	0.9807	1.021	1.800	1.511	86 096	0.9548	1.042	6.836	2.343
5	459 164	0.9783	1.017	1.787	1.498	73 627	0.9586	1.022	6.852	2.279
6	441 746	0.9834	1.025	1.782	1.485	67 306	0.9558	1.043	6.653	2.224
7	430 581	0.9794	1.015	1.782	1.494	62 403	0.9508	1.020	7.024	2.258
8	419 202	0.9815	1.028	1.784	1.496	63 049	0.9653	1.052	6.669	2.263
9	395 200	0.9803	1.026	1.775	1.484	57 575	0.9630	1.051	6.610	2.234
10	374 950	0.9780	1.019	1.771	1.504	53 280	0.9641	1.035	6.550	2.352
11	434 871	0.9808	1.023	1.769	1.517	54 786	0.9362	1.039	7.997	2.381
12	363 353	0.9799	1.026	1.766	1.494	46 958	0.9585	1.047	6.598	2.302
13	324 105	0.9781	1.026	1.526	1.492	42 630	0.9503	1.045	3.860	2.276
14	315 708	0.9815	1.021	1.765	1.533	43 033	0.9656	1.034	6.163	2.423
15	267 028	0.9809	1.019	1.743	1.514	33 400	0.9685	1.039	6.572	2.361
16	268 749	0.9828	1.020	1.765	1.535	36 818	0.9588	1.031	5.980	2.403
17	341 519	0.9793	1.026	(1.736) ^a	1.501	33 539	0.9638	1.040	(9.116) ^a	2.926
18	332 811	0.9816	1.019	1.514	1.531	33 565	0.9633	1.032	4.867	2.458
19	324 701	0.9841	1.022	1.527	1.532	32 032	0.9582	1.026	4.870	2.440
<hr/>										
Average		0.9803	1.021	1.739	1.507		0.9591	1.037	6.453	2.319
Scatter (1σ)		±0.002	±0.004	±0.099	±0.016		±0.005	±0.009	±1.02	±0.074
1σ(%)		±0.2	±0.4	±5.7	±1.1		±0.5	±0.9	±15.8	±3.2
CF (D-372) 1 600		0.985	1.012	1.703	--	308.0	0.969	1.035	4.053	2.389

^aFaulty measurement with PPMCC counter omitted from average and σ.

statistical errors ranged from 0.01 to 0.005%, and they were completely negligible when compared with other sources of error, such as

- electronic stability,
- sample positioning, and
- variable reflection from bodies in the measurement room.

The observed scatter in the totals ratios was only 0.2% and 0.4% for IAEA-1 and EURATOM/Los Alamos systems, respectively, and 5.7% and 1.1% for the NMACT and BNFL systems, respectively.

D. Reals Rate Ratios

The scatter in the reals ratio is expected to be a factor of 2 larger than the totals error for efficiency instabilities such as the above three items. Deadtime errors also are larger for the reals than the totals by a factor of ~4. In addition, the deadtime errors will show a systematic trend in going from the low-totals-rate samples (for example, sample 16 and the ²⁵²Cf source) to the high-rate samples (for example, sample 1).

The observed scatter in the reals ratio is 0.5% and 0.9% for the IAEA-1 and EURATOM/Los Alamos systems, respectively, and 15.8% and 3.2% for the NMACT and BNFL systems, respectively. This scatter in the reals is a factor of 2 larger than the totals for the IAEA-1 and EURATOM/Los Alamos systems and a factor of 3 larger for the NMACT and the BNFL systems, as expected.

The statistical counting errors in the reals rates are given in Table X. These standard deviations were calculated from the expression

$$\sigma\% = \sqrt{(R + A) + A} \cdot \frac{100\%}{R} ,$$

where A is the accidental count rate. This expression gives a σ that is too large when $A > T$, as is the case with the present data. These standard deviations will be used later to help quantify the sources of errors on the basis that the total error is the combination of the statistical error and the other sources of error.

TABLE X

STATISTICAL COUNTING ERRORS FOR THE RANGE OF SAMPLES

Counter	1σ for 600-s Count ^a	
	Sample 1 (%)	Sample 19 (%)
IAEA-1,2	0.22	0.43
EURATOM/Los Alamos	0.23	0.44
NMACT	0.60	0.91
BNFL	0.43	0.58

^aThe standard deviations were calculated from the equation

$$\sigma = \sqrt{(R + A) + A \cdot \frac{100\%}{R}} .$$

The sample with lowest deadtime correction is ²⁵²Cf (D-372); the correction is negligible. If there are no deadtime or geometry problems, the totals and reals ratios (Table IX) for ²⁵²Cf should be the same as for the high-rate samples. That is the case for all of the systems except the PPNCC counter, where the ²⁵²Cf ratio is 4.053 and the average sample ratio is 6.453. Also, both the totals and reals ratios for the PPNCC counter are systematically lower for the lower-mass than for the higher-mass samples. These ratios indicate the deadtime correction in the PPNCC counter overcorrects at the lower rates and undercorrects at the higher rates.

These ratio differences cannot be accounted for by the difference in the functional form of δ(T). That is,

$$\sigma(T) = a + bT$$

vs

$$\delta = 2.8 \text{ (for the NMACT system) ,}$$

where a and b are the deadtime coefficients. To illustrate these differences, Table XI covers the counting rate of interest, and the last column gives the comparison in the two methods of calculating deadtime.

The observed real ratio problem of 4.0 (low rate) vs 7.5 (high rate) must be caused by a more serious problem, such as an electronic bias in the coincidence rate. For a normal range of samples, the calibration curve contains the same bias, and it cancels out in the final assay result. Thus, it could easily go unnoticed at lower counting rates.

E. Outliers

The ratios of T and R given in Table IX are consistent for all of the samples (including the outliers) with only minor exceptions for the PPNCC system. Thus, the outliers are caused by sample problems rather than by instrument problems. Table XII gives some examples of instrument and sample problems that could lead to outliers.

This experiment was designed to identify and eliminate possible instrumentation problems. This goal was accomplished because we showed that instrument problems will change the T and R ratios for the different equipment. On the

TABLE XI
CONSTANT DEADTIME VS RATE-DEPENDENT DEADTIME

Totals Counting Rate (s ⁻¹)	$\delta(T)$ ^a	$e^{\delta(T) \cdot 10^{-6} T}$	$e^{2.8 \cdot 10^{-6} T}$	$\frac{e^{2.8 T}}{e^{\delta T}}$
1 000	2.302	1.002	1.003	1.001
50 000	2.381	1.126	1.150	1.022
100 000	2.462	1.279	1.323	1.035
200 000	2.624	1.690	1.751	1.036
300 000	2.786	2.307	2.316	1.004
400 000	2.948	3.252	3.065	0.943

^aDeadtime coefficient from HLNC-I ($2.3 + 1.62 \cdot 10^{-6} T$) that should be similar to PPNCC counter.

TABLE XII
POTENTIAL OUTLIER CAUSES FOR NEUTRON COUNTERS

<u>Number</u>	<u>Instrument Problems</u>	<u>Error Direction</u>	<u>Sample Problems</u>	<u>Error Direction</u>
1	Electronic noise pickup	High	H ₂ O in PuO ₂	High
2	Gain change	Either	Contaminants (fluorine) in PuO ₂	High
3	Variable efficiency profile	Either	Container change	Either
4	Loss of tubes/amplifier	Low	Matrix change	High
5	High-voltage breakdown	High		
6	Geometry/reflection change	Either		
7	Incorrect deadtime	Either		
8	Gamma-ray sensitivity	Low		

other hand, the sample-related problems listed in Table XII will be common to all of the neutron systems.

Before we get to the specific calibration curves, we should mention that we can spot outliers by a high R_{mc}/g ratio. This ratio is proportional to the specific spontaneous-fission yield, and it should be a constant for each detector.

In Tables IV-VIII, we list the R_{mc}/g values with their averages and scatter about the mean. Three high outliers were identified (samples 12, 17, and 19) as being more than 2σ above the mean. There were no low outliers. Two of these outliers (17 and 19) have high burnups and high ²⁴¹Am contents, indicating a longer time since chemical separation. This long separation time would give more opportunity for moisture absorption.

Samples 17-19 had a very high gamma-ray dose on contact, which may be related to their assay variability.

F. Gamma-Ray Sensitivity

If the gamma-ray dose from the sample gets above ~ 1 R/h, the pileup of gamma-ray pulses can get above the neutron discrimination threshold. When this happens, the T value increases rapidly and the R value decreases slowly.

Figure 3 shows the totals rate vs high voltage for the ^3He tubes in a neutron system similar to the AMPTEK system being evaluated. The operating high voltage should be set below the point where the gamma pileup is significant.

The gamma-ray dose was measured for two Sellafield sample cans, using one high-dose can and one low-dose can. The measurements were made at positions A, B, and C, shown in Fig. 4, where A is in contact with the can and B and C were measured with the can inside the HLNC-II.

Table XIII, which lists the measured doses as a function of position, shows that dose is getting close to the 1 R/h limit at the location of the ^3He tubes. In retrospect, a high-voltage plateau curve should have been measured for the highest dose sample.

Most of the gamma dose is coming from the ^{241}Am (60-keV) gamma ray that is effectively shielded by the cadmium liner on the interior of the neutron detector cavities. Because the high dose is 2000 mrem/h on the inside of the detector wall and 100 mrem/h on the outside of the wall, we can estimate the dose as ~500 mrem/h at the tube location.

Fig. 3. Typical totals rate vs high voltage on the ^3He tubes for an HLNC-II system with different levels of gamma-ray exposure to the detectors.

Fig. 4. Geometry for gamma-ray and neutron dose measurements where points A, B, and C represent the measurement positions.

TABLE XIII
GAMMA-RAY DOSE MEASUREMENTS
FROM HIGH- AND LOW-DOSE CANS

<u>Can Number</u>	<u>Position</u>	<u>Gamma (mrem/h)</u>	<u>Neutron (mrem/h)</u>
18	A	2000	60
	B	100	8
	C	8	0.9
7	A	250	60
	B	40	13
	C	4	1.2

The different neutron detectors should have a different sensitivity to gamma-ray pileup. The BNFL system should be the least sensitive because it has a separate preamplifier and amplifier on each ^3He tube, thus reducing the pileup load by a factor of 3. The totals ratio for the high- and low-dose samples is the same for the different detectors, so it would appear that there is no pileup problem.

G. Sample-Positioning Errors

To examine the variability in the result from the radial position of the sample can, using IAEA-1 we counted one of the samples in the normal center position and then repeated the count with the can touching the wall of the sample cavity. The results were as follows:

$$\frac{T(\text{on wall})}{T(\text{center})} = \frac{343\ 585}{342\ 930} = 1.002$$

and

$$\frac{R(\text{on wall})}{R(\text{center})} = \frac{35\ 806}{35\ 464} = 1.010 \quad ,$$

and after multiplication correction,

$$\frac{R(\text{on wall})}{R(\text{center})} = \frac{15\ 248}{15\ 230} = 1.001 \quad .$$

Thus, the error from radial variability in positioning the cans was negligible for the HLNC-II counters. Because the multiplication correction uses the R/T ratio, the results after multiplication correction show even less wall perturbation than the results before multiplication correction.

H. Wall-Reflection Effects

Many of the neutrons that are emitted from the sample pass through the detector body and escape to the room. If the detector is positioned close to a high-density wall, such as concrete, neutrons will be reflected back into the detector, giving a small increase in the counting rate.

This efficiency change was measured with a ^{252}Cf source in an HLNC-II positioned at various distances from the concrete wall in the vault. The results of the measurements are shown in Fig. 5, where the distance is from the outside edge of the detector to the surface of the wall. The coincidence rate has been normalized to unity at large distances from the wall. For the work at Sellafield, a wall-reflection correction (0.97) was applied only to the IAEA-1 counter that was operated at ~15 cm from the wall. The other counters were far enough away from the wall to make the correction negligible.

VI. CALIBRATION RESULTS

For brevity, this section dealing with calibration results examines only one of the five systems. The results for any of the other systems can be

Fig. 5. Coincidence rate increase (HLNC-II) from concrete wall reflection vs distance between side of detector and wall.

obtained from the ratios given in Table IX. The EURATOM/Los Alamos system is used as the reference system, so the present results can be combined with measurements at Los Alamos¹ and Belgonucleaire⁸ to extend the mass that the calibration covers from a few grams to 7.2 kg of PuO₂.

We calibrated the detector for the data both before and after multiplication corrections, and we used the alpha-corrected totals rate to help identify outliers.

A. Sample-Container Correction

The thick-walled sample containers give a small percentage of increase in the neutron rate, so we corrected for this effect before comparison with results from thin-walled cans.

To do this, the ²⁵²Cf source (D-372) was measured inside a double-walled container. The results were

$$\frac{R(\text{Cf inside can})}{R(\text{Cf no can})} = 1.027 \quad ,$$

and the totals correction was 1.013. Thus, the T and R rates given in Table VI should be divided by these factors for the calibration.

B. Californium-252 Source Normalization

The source (D-372) gave a rate in counter IAEA-2 of

$$R(D-372) = 297.7 \pm 2.0 \text{ (on 85-01-30).}$$

This is our reference rate; we normalized the Sellafield data to the data taken at Belgonucleaire and Los Alamos using the EURATOM/Los Alamos counter.

Belgonucleaire (EURATOM/Los Alamos Counter). The neutron source reference rate at Belgonucleaire was

$$R(D-372) = 329.6 \pm 1 \text{ (on 84-09-24).}$$

The time interval between the Belgium work and the measurements in the United Kingdom was

$$\text{Time lapse} = 0.345 \text{ y,}$$

$$e^{-\lambda t} = 0.913, \text{ so}$$

$$R(\text{corr}) = 301.1 \text{ (on 85-01-30).}$$

The source normalization constant is

$$\frac{R(\text{Sellafield})}{R(\text{Belgonucleaire})} = \frac{297.7}{301.1} = 0.9887 \quad .$$

To directly compare the prior data with the present data, we need to divide the Sellafield reals rates by a factor of

$$\underbrace{(1.027)}_{\text{Sample Container}} \cdot \underbrace{(0.9887)}_{\text{Cf Norm.}} = 1.015$$

and the totals rates by 1.008.

C. Deadtime Correction

The data taken at Belgonucleaire with the EURATOM/Los Alamos counter were evaluated using the erroneous deadtime coefficient of

$$\delta = (0.62 + 0.2 \cdot 10^{-6} T) \mu s .$$

The value used at Sellafield (based on more recent measurements) was

$$\delta = (0.68 + 0.22 \cdot 10^{-6} T) \mu s .$$

Thus, a small correction was applied to the prior Belgonucleaire data.

D. Updated Results Covering Lower Mass Range

The data taken at Belgonucleaire using the EURATOM/Los Alamos counter have been updated with the new value of $\delta = 0.68 + 0.22 \cdot 10^{-6} T$, and the results are listed in Table XIV. The "green" samples with high moisture content were omitted from the table.

Similar data taken at Los Alamos for masses of 10 to 150 g ^{240}Pu effective are given in Table XV. In this case, the counting rates were so low that the change in δ made a negligible difference in the results.

The multiplication-corrected reals per gram value is slightly less for these samples because the small mass gives less neutron scattering than the

TABLE XIV

MEASUREMENT RESULTS FOR BELGONUCLEAIRE PuO₂
IN HLNC-II COUNTER EURATOM/LOS ALAMOS

Sample	²⁴⁰ Pu-eff. (q)	Totals (s ⁻¹)	Reals ^a (s ⁻¹)	s _R ^b (%)	R _c (s ⁻¹)	$\frac{R_c}{q}$ ²⁴⁰ Pu
S00720	364.9	108 717	11 249	0.56	6 798	18.63
S00849	413.2	121 160	13 398	0.53	7 423	17.96
S00850	421.4	124 036	13 734	0.43	7 596	18.03
XN 1066	558.9	213 516	18 288	0.55	10 524	18.83
XN 1064	974.8	377 874	35 204	0.53	18 237	18.71
XN 1059	1 025.6	401 348	38 174	0.52	19 267	18.79
XN 1062	1 046.8	409 971	39 881	0.51	19 567	18.69
XN 1055	809.1	318 017	29 132	0.57	15 468	19.11
XN 1025	837.4	315 461	32 468	0.46	15 858	18.93
XN 1123	936.9	341 329	32 718	0.51	17 220	18.38
XN 1076	957.5	371 049	34 897	0.45	17 855	18.65
XN 1097	1 026.8	396 554	37 984	0.51	18 913	18.42
					Av =	18.59
						±0.34
Cf (D-372) ^c		1 744	329.6	0.04		

^aThe totals and reals rates have been corrected for steel double containers, room background (40 counts/s), and deadtime, $\delta = (0.68 + 0.22 \cdot 10^{-6} T) \mu s$.

^bThe s_R(%) is the observed standard deviation of repeat runs about the mean value of R.

^cMeasured on 9/24/84; the ratio of D-372/CR-5 = 0.1578 ± 0.001 .

TABLE XV

MEASUREMENT RESULTS FOR LOS ALAMOS PuO₂ IN HLNC-II

Sample No.	q ²⁴⁰ Pu-eff	Run Time (s)	Totals Rate (s ⁻¹)	Reals Rate (s ⁻¹)	σ_R (%)	Corrected Rate R_{mc} (s ⁻¹) ^a	$\frac{R_{mc}}{q}$
1	10.11	900	2 520	208.3	0.53	182.9	18.09
2	29.33	800	7 469	663.3	0.48	529.8	18.06
3	54.42	800	14 063	1 322	0.44	988.9	18.17
4	65.19	800	16 868	1 636	0.43	1 173	18.00
5	92.31	600	24 236	2 441	0.47	1 654	17.92
6	105.33	600	27 841	2 832	0.46	1 907	18.11
7	144.53	600	38 467	4 137	0.44	2 604	18.02
8	149.29	600	39 833	4 320	0.44	2 689	18.01
Av =							18.05
							±0.07
CF (CR-6) ^b		900	31 050	5 890	0.20		

^aMultiplication correction made with Method 2.^bMeasured on 6/6/84.

larger samples do. The scattered neutrons have lower energy and thus a higher detection efficiency (ϵ) in the EURATOM/Los Alamos counter.

E. EURATOM/Los Alamos Calibration Results

Table XVI gives the Sellafield data for the EURATOM/Los Alamos counter corrected for deadtime, ²⁵²Cf normalization, and sample containers (wall thickness). This amounts to dividing the Table VI base data by a factor of 1.015 for R and 1.008 for T (see Sec. VI.B). The R_{mc} values decrease by a factor of only ~1.006.

The calibration curves for R and R_{mc} are shown in Fig. 6, where in addition to the Sellafield data, the lower-mass data from Belgonucleaire and Los Alamos are shown for comparison. The three outliers (samples 12, 17, and 19) are shown on the graph, but they were not included in the calibration curve

TABLE XVI

SELLAFIELD CALIBRATION SUMMARY FOR COUNTER EURATOM/LOS ALAMOS

Sample	T_c (s ⁻¹) ^a	R_c (s ⁻¹) ^a	R_{mc} (s ⁻¹)	$\frac{R_{mc}}{q}$	$\frac{T_c}{(1 + \alpha)}$	$\frac{T_c}{(1 + \alpha)q}$	$\frac{T_c}{(1 + \alpha)q M_L}$
1	533 995	92 341	31 481	17.72	391 205	220.3	171.8
2	526 750	90 806	31 003	17.76	385 051	220.7	172.2
3	515 427	89 422	31 186	17.92	386 377	222.1	173.8
4	482 293	81 445	28 798	18.28	354 888	225.3	177.3
5	447 981	70 989	24 951	17.92	307 468	220.9	174.9
6	427 467	63 581	24 840	18.14	296 852	216.8	175.7
7	420 886	60 268	24 278	18.82	287 687	223.0	182.3
8	404 714	59 031	23 108	17.83	275 503	217.1	176.5
9	382 023	53 945	22 671	18.24	266 404	214.3	176.7
10	365 155	50 699	20 789	17.81	246 559	211.3	173.9
11	421 834	51 944	18 953	18.00	224 022	212.7	174.3
12	351 422	44 187	20 922	19.70	238 414	224.5	190.7
13	313 502	40 205	18 519	17.98	212 400	206.2	174.2
14	306 748	40 995	17 669	18.22	205 595	212.1	176.6
15	260 051	31 667	15 544	17.96	175 710	203.0	174.1
16	261 319	37 112	16 375	17.77	191 023	207.3	172.2
17	330 110	31 780	16 132	19.29	177 960	212.9	186.9
18	324 004	32 043	14 580	18.59	163 721	208.8	180.2
19	315 131	30 753	15 346	19.81	169 882	219.3	191.9
Av (omit 3 outliers) =							175.4
1σ							±1.6%
CF (D-372) ^b 1 556 301.1							

^aThe coincidence deadtime coefficient was $\delta = (0.68 + 0.22 \cdot 10^{-6} T) \mu s$, the sample-container correction was 0.974, and the efficiency correction was 1.011.

^bValues reflect efficiency correction on 85-01-30.

Fig. 6. The coincidence rate vs ^{240}Pu -effective mass before (R) and after (R_{mc}) multiplication corrections for the extended data set.

fit. Several of the high points on the R curve are not the identified "outliers," but they are legitimately high because of very high α values. The extra (α, n) neutrons induce additional fissions (multiplication) in the plutonium. The standard multiplication correction adequately corrects for this effect, and thus those samples are not necessarily outliers on the curve for R_{mc} vs mass.

The calibration parameters are listed in Table XVII for the EURATOM/Los Alamos counter. The Sellafield data set (high mass) and the extended data set (36 samples) were both fit with a quadratic function of the form

$$R = aM^2 + bM + c ,$$

TABLE XVII

CALIBRATION PARAMETERS FOR EURATOM/LOS ALAMOS COUNTER^(a)

Sample Group	a	b	c	(b)
	$\pm\sigma_a$	$\pm\sigma_b$	$\pm\sigma_c$	$ \bar{\Delta} $ (%)
(1) 36 samples (no mc)	0.014963	25.283	-528.7	3.57
	± 0.000803	± 0.7033	± 15.6	--
(1) 36 samples (mc)	0	18.168	0	1.70
	0	± 0.0658	0	--
(2) 16 Sellafield (no mc)	0.008287	42.527	-8 611	2.96
	± 0.00827	± 21.73	$\pm 13\ 630$	--
(2) 16 Sellafield (mc)	0	18.020	0	1.28
	0	± 0.0748	0	--

(a) The calibration function is $R = aM^2 + bM + c$.

(b) The average absolute mass residual, $|\bar{\Delta}|$, was obtained from the least-squares fit of the data to the calibration curve.

where M is the mass of ^{240}Pu -effective and a , b , and c are the fitting constants. For the multiplication-corrected response we used a straight line through the origin,

$$R_{mc} = bM.$$

The fits were reasonably good for all of the data sets; with an average absolute mass residual of 3.57% before multiplication correction and 1.70% after the correction. This is very good considering that the following variables were included:

- plutonium masses from 60 to 7100 g,
- burnup values from 82% to 58% ^{239}Pu ,

- americium-241 contents from 670 to 33 814 ppm,
- four different container types and geometries, and
- three different facilities and dates for the data collection.

A ^{252}Cf source was used to normalize the detector for electronic changes, but these corrections were always $<1.2\%$.

When the more limited Sellafield data set (16 samples) was fit, the average absolute mass residual from R_{mc} was reduced to 1.28% . The three outliers were 6% to 8% above the calibration curve. Figure 7 shows the R_{mc} values for the Sellafield data set with an expanded scale to show the three outliers.

The following section examines the totals rates to see if they are anomalous for the outliers.

Fig. 7. The multiplication-corrected real rate vs ^{240}Pu -effective for the Sellafield samples, with an expanded scale to show the outliers.

F. Totals Rate Analysis

The totals rate can indicate a variable (α, n) problem in a sample because the observed T will increase when (α, n) emitters (for example, fluorine or H_2O) are present. The observed totals rate can be written as

$$T = T(\text{spontaneous fission}) + T(\alpha, n) + T(\text{induced fission}) ,$$

or

$$T = T(SF) + T(\alpha, n) + T(IF) . \quad (1)$$

The spontaneous-fission rate is directly proportional to the g ^{240}Pu -effective. However, we have measured T, so we must correct it for (α, n) neutrons and induced-fission neutrons as follows:

The calculated ratio α is defined as

$$\alpha \equiv \frac{T(\alpha, n)}{T(SF)} ,$$

and the leakage multiplication, M_l , is

$$M_l \equiv \frac{T}{T(SF) + T(\alpha, n)} .$$

Equation (1) can be solved to give

$$T(SF) = \frac{T}{(1 + \alpha)M_L}$$

and also

$$\frac{T(SF)}{g \text{ } ^{240}\text{Pu-effective}} = \text{const} = \frac{T}{(1 + \alpha)g M_L} \quad (2)$$

This ratio should be a constant for all samples with correct values of α , M_L , and $g \text{ } ^{240}\text{Pu-effective}$. If there are (α, n) contaminants in the sample, the calculated values of α and M_L will be too small and the totals rate ratio from Eq. (2) will be too large.

This corrected totals rate ratio is directly related to the corrected reals ratio per unit mass by the multiplication constant ρ_0 (see Sec. VII.A).

Table XVI gives this totals per gram ratio for the 19 Sellafield samples, and we see that the value is high for all of the identified outliers. This ratio, corrected for multiplication and (α, n) neutrons, should be a constant in the EURATOM/Los Alamos counter for all normal samples. The same constant (175.4) should be valid for the other low-mass data sets given in Tables XIV and XV, which correspond to the same detector.

The average value of $T/(1 + \alpha)g M_L$ for the Sellafield data set was 175.4 ± 2.8 . This value can be compared with the average value of 174.4 ± 1.4 for the Los Alamos samples (10 to 150 g $^{240}\text{Pu-effective}$) and 179.2 ± 5.3 for the Belgonucleaire samples (365 to 1027 g $^{240}\text{Pu-effective}$). The stated uncertainties are the scatter of the ratios in the individual data sets. The typical sample at Belgonucleaire had a moisture content $>5000 \text{ ppm H}_2\text{O}$.

Figure 8 plots the ratio $T/(1 + \alpha) g M_{\ell}$ vs ^{240}Pu -effective for the EURATOM/Los Alamos detector. The outliers (samples 12, 17, and 19) are identified by circles on the plot. The deviations from the fitted horizontal straight line were 8.7%, 6.6%, and 9.4% for samples 12, 17, and 19, respectively. The average deviation of the other samples was 1.6%.

The present values of M_{ℓ} were determined from the measured R/T ratios and the calculated value of α . For the outliers, α is in error, so the subsequent calculation of M_{ℓ} is also incorrect. On the other hand, if we calculated M_{ℓ} using Monte Carlo computer codes and the operator's values of sample mass and size, then the constant value of Eq. (2) could be used to estimate α .

The utility of the constant ratio plotted in Fig. 8 or the equivalent Fig. 7 is to help identify outliers from their anomalous totals rates. High rates can be explained by high (α, n) contamination or moisture in the samples.

Fig. 8. The spontaneous-fission totals rate, $T/(1 + \alpha) g M_{\ell}$, vs the ^{240}Pu -effective mass for the EURATOM/Los Alamos detector.

VII. MULTIPLICATION ANALYSIS

The results after multiplication correction have a scatter that is a factor of ~ 2 less than before the correction. This difference occurs because the correction takes care of variations in sample density, shape, and α (induced fissions). These effects are more important as the sample mass increases.

A. Multiplication Equations

The multiplication corrections were made using the new equations given by Böhnel⁷ and Ensslin.⁸ They are

$$R_{mc} = \frac{R}{CF} ,$$

$$CF \equiv M \cdot r ,$$

$$r \equiv \frac{\frac{R}{T}}{\rho_0} (1 + \alpha) ,$$

$$\rho_0 = \left(\frac{R}{T} \right)_0 (1 + \alpha_0) = 0.103 \quad (\text{for the HLNC-II}) ,$$

$$M = \frac{-B + \sqrt{B^2 - 4AC}}{2A} ,$$

$$A = 2.074(1 + \alpha) ,$$

$$B = -(2.074\alpha + 1.074) ,$$

$$C = -r ,$$

$$\alpha = \frac{134 f_{238} + 0.381 f_{239} + 1.41 f_{240} + 0.013 f_{241} + 0.02 f_{242} + 26.9 f_{241Am}}{10.22(243 f_{238} + f_{240} + 1.69 f_{242})} ,$$

where the f_m values are the weight fractions of the plutonium isotopes with mass number m .

The value of M_l is calculated in the above equations as M if the correct values of ρ_0 and α are used. M_l is a parameter related to the mass and size of the sample, so it should have nothing to do with the particular detector (neglecting differences in neutron reflection from the detector walls). The detector-wall reflection of neutrons gives a small increase to M_l .

B. Multiplication Constant

The parameter ρ_0 is defined by

$$\rho_0 = \left(\frac{R}{T} \right)_0 (1 + \alpha_0)$$

for a nonmultiplying sample. The value of ρ_0 was measured to be ~ 0.103 for the HLNC-II. This parameter is proportional to the singles efficiency and the fraction of time-correlated neutrons that fall within the coincidence gate.

The value of ρ_0 for one detector can be related to a different detector by the R/T ratios in both detectors for the same sample. That is,

$$\rho_0(\text{BNFL}) = \rho_0(\text{HLNC-II}) \frac{\frac{R}{T}(\text{BNFL})}{\frac{R}{T}(\text{HLNC-II})} \quad (3)$$

Different values of ρ_0 can be used for any detector, but it is essential that the same value of ρ_0 be used in both the calibration fit and in the actual assay. If incorrect values of ρ_0 are used, they cancel out in the calibration curve; however, M_l will be incorrect.

For this analysis, we wanted to be able to directly compare the M_l values determined from the R/T measurements for each detector, so we used Eq. (3) to calculate consistent ρ_0 values for the PPNC and BNFL detectors.

There was so little change in the ρ_0 values for the three HLNC-II detectors that the original value ($\rho_0 = 0.103$) was maintained for each of these systems.

A summary of the multiplication parameters is given in Table XVIII. The listings of the R_{mc}/g ratios are given in Tables IV-VIII. If there are no unknown (α, n) or moisture problems, this ratio should be a constant for each detector. Also, Table XVIII shows the scatter of this ratio about the mean for the five systems. The three outliers (samples 12, 17, and 19) were identified.

C. Multiplication Consistency Checks

To investigate the consistency in R_{mc} and M_q , we processed the base data from each of the five systems using the IAEA CAL4 computer code to give consistent R_{mc} and M_q values. For each detector, the ratio of R_{mc} and ($M_q - 1$) was compared with the reference system (IAEA-2).

The ($M_q - 1$) values were compared because they give a more sensitive indication of the multiplication than M_q would. For example, $M_q = 1.250$ means that there is a 25% increase in the net neutron leakage caused by multiplication.

TABLE XVIII

MULTIPLICATION PARAMETERS SUMMARY

Detector	Original ρ_0	CAL4 ρ_0	$\frac{R_{mc}}{g}$ Av	$\frac{R_{mc}}{g}$ Omit Outliers ^a	Scatter $1\sigma(\%)$ ^b
IAEA-1	0.103	0.103	18.91	18.59	1.2
IAEA-2	0.103	0.103	18.73	18.43	1.6
EURATOM/Los Alamos	0.103	0.103	18.41	18.13	1.6
NMACT	~0.0437	0.0282	5.04	4.92	2.3
BNFL	~0.060	0.0667	7.10	7.11	2.5

^aAverage based on original ρ_0 values for all systems.

^bOmitted outlier sample numbers 12, 17, and 19 in the average and scatter.

If there are no problems of the type listed in Table XII, this R_{mc} ratio to the reference detector will be constant for all of the samples. The variation in the ratio outside of counting statistics is an indication of deadtime or geometry problems.

Table XIX lists the ratios of R_{mc} and $(M_q - 1)$ for all of the detectors. The scatter (1σ) of R_{mc} about the mean is only $\pm 0.26\%$ for IAEA-1 and $\pm 0.36\%$ for EURATOM/Los Alamos. This scatter is very good and consistent with the counting statistics on R ($1\sigma \approx 0.4\%$). It is interesting that the scatter in R_{mc} is less than half that observed in the R values (Table IX) of $\pm 0.5\%$ and $\pm 0.9\%$ for the same two systems.

The BNFL detector ratios are consistent with those of the HLNC-II with about twice the scatter ($\pm 0.68\%$) in R_{mc} .

The PPNCC detector shows a systematic problem in the deadtime correction that results in a scatter of $\pm 3.83\%$.

The average M_q ratios for all of the systems are remarkably close to unity. The differences in the average values for the three HLNC-II systems just reflect that they have slightly different ($\sim 2\%$) efficiencies (see Table III), but they all used the same ρ_0 value (0.103).

When there is a deadtime problem, the R/T ratio will be in error because the deadtime correction is approximately four times larger in R than in T . This incorrect R/T ratio will give nonphysical values for M_q calculated by the equations in Sec. VII.A. Thus, the $(M_q - 1)$ ratios for the PPNCC detector have a large scatter of $\pm 18.7\%$.

D. Multiplication Investigation

Because M_q is an intrinsic property of the sample, it is interesting to examine it for all three sample sets (Los Alamos, Belgonucleaire, and Sellafield). Figure 9 plots the M_q values as a function of plutonium fissile mass from 50 to 5439 g. The measured R/T ratios and calculated α values were used to determine the values of M_q using the multiplication equations given in Sec. VII.A and the EURATOM/Los Alamos counter.

Figure 10 shows the inside diameter and maximum fill heights of the three different PuO_2 containers. The low-mass Los Alamos samples fill only a few centimeters in the bottom of the can, so the sample has the shape of a disk or pancake, with very low neutron multiplication. However, as the sample mass increases to the largest Los Alamos sample (876 g of plutonium), the height is

TABLE XIX

COMPARISON OF MULTIPLICATION-CORRECTED RESULTS FOR THE FIVE NEUTRON SYSTEMS

Sample No.	IAEA-2 R_{mc} ^(a)					IAEA-2 $(M_2 - 1)$ ^(a)				
	R_{mc}					$M_2 - 1$				
	IAEA-2 T_c (s ⁻¹)	IAEA-1	EURATOM/ Los Alamos	PPNCC	BNFL	IAEA-2 M_p (s ⁻¹)	IAEA-1	EURATOM/ Los Alamos	PPNCC	BNFL
1	32 055	0.985	1.012	6.477	2.350	1.292	0.967	1.036	1.187	0.961
2	31 558	0.985	1.012	6.377	2.345	1.287	0.966	1.018	1.181	0.953
3	31 762	0.987	1.012	6.414	2.350	1.288	0.963	1.036	1.152	0.957
4	29 389	0.989	1.014	6.492	2.330	1.280	0.962	1.033	1.065	1.007
5	25 485	0.984	1.015	6.429	2.231	1.265	0.971	1.008	1.077	0.978
6	25 484	0.992	1.020	6.466	2.316	1.240	0.956	1.026	1.034	0.952
7	24 750	0.988	1.013	6.356	2.322	1.225	0.953	1.009	1.148	0.966
8	23 711	0.986	1.020	6.471	2.325	1.240	0.976	1.043	1.039	0.964
9	23 224	0.985	1.018	6.442	2.310	1.220	0.961	1.033	1.023	0.948
10	21 385	0.991	1.023	6.498	2.314	1.220	0.969	1.023	1.014	1.023
11	19 414	0.988	1.018	6.097	2.332	1.225	0.962	1.023	1.442	1.023
12	21 455	0.986	1.019	6.406	2.309	1.184	0.958	1.040	1.045	0.995
13	19 004	0.987	1.020	6.262	2.313	1.190	0.945	1.033	0.562	0.974
14	18 078	0.986	1.017	6.538	2.351	1.206	0.972	1.025	0.924	1.046
15	15 837	0.984	1.013	6.307	2.330	1.173	0.972	1.042	1.068	1.018
16	16 752	0.990	1.017	6.603	2.361	1.208	0.959	1.020	0.878	1.030
17	16 601	0.983	1.023	(5.805) ^(b)	2.324	1.142	0.966	1.022	(3.087) ^(b)	0.979
18	14 896	0.986	1.016	5.718	2.340	1.163	0.970	1.025	0.807	1.079
19	15 764	0.991	1.021	5.783	2.347	1.144	0.947	1.007	0.774	1.067
Average		0.987	1.017	6.341	2.331		0.963	1.026	1.023	0.996
Scatter (1σ)		±0.003	0.004	0.243	0.016		±0.008	±0.011	±0.191	0.040
1σ(%)		±0.26	0.36	3.83	0.68		±0.87	±1.08	18.7	4.06

(a) All of the multiplication-corrected rates and M_2 are based on ρ_0 values given in Table XVIII.

(b) Faulty measurement with PPNCC counter omitted from average.

Fig. 10. The inside dimensions of the three different sample cans used in the extended sample set.

VIII. SUMMARY

Samples of PuO_2 powder with mass values as high as 7.2 kg have been successfully measured using the five neutron counters. The deadtime corrections were adequate for the HLNC-II and BNFL systems, but some additional adjustment is needed for the PPNCC system. In general, a single constant cannot be used for δ unless the true counting rate is used in the exponential function.

The ratios of T , R , R_{mc} , and M_l were consistent to better than 1% for the three HLNC-II systems. Table XX summarizes the expected standard deviations in the ratios compared with the observed standard deviations. Because of the good statistical precision in the data, the differences between the observed scatter (from all sources) and the calculated statistical standard deviation (counting statistics) give us an estimate of the nonstatistical errors, such as electronic instability, sample positioning, and background and neutron

TABLE XX
SUMMARY OF STATISTICAL SCATTER IN THE RATIO
WITH THE IAEA-2 COUNTER

Parameter	IAEA-2 Ratio			
	IAEA-1 (%)	EURATOM (%)	NMACT (%)	BNFL (%)
1σ in T ratio calc. ^a	0.008	0.008	0.01	0.01
Scatter in T observed	0.2	0.4	5.7	1.1
1σ in R ratio calc. ^a	0.4	0.4	0.7	0.5
Scatter in R observed	0.5	0.9	15.8	3.2
Scatter in R _{mc} ratio observed	0.26	0.36	3.83	0.68
Scatter in (M _λ - 1) ratio observed	0.87	1.08	18.7	4.06

^aThe ratio error corresponds to the average value calculated from $\sigma(T) = \sqrt{T/T}$ or $\sigma(R) = \sqrt{(R+A)/R}$ for both IAEA-2 and the counter in question.

reflection changes. For example, with the EURATOM/Los Alamos counter the observed scatter in the coincidence is

$$\sigma^2(\text{observed}) \approx \sigma^2(\text{statistics}) + \sigma^2(\text{geometry}) + \sigma^2(\text{electronics}) + \dots ,$$

$$(0.9)^2 \approx (0.4)^2 + \sigma^2(\text{geometry}) + \sigma^2(\text{electronics}) + \dots .$$

Thus, the sum of nonstatistical components is

$$\sigma^2(\text{nonstatistical}) \approx (0.9)^2 - (0.4)^2$$

$$\sigma(\text{nonstatistical}) \approx 0.8\%$$

This value is smaller (0.4%) for the IAEA-1 counter.

The calibration curve (EURATOM/Los Alamos) showed an average absolute mass residual of 2.96% for the data before multiplication corrections and 1.28% after multiplication corrections. If the three outliers are included in the data, the average residuals get a little bigger, but not much because of the large number of samples.

A single constant slope for R_{mc} was found adequate to fit the entire range of masses from 60 to 7200 g of PuO_2 for the HLNC-II systems. This slope gives a baseline (slope ≈ 18.17 for EURATOM/Los Alamos), and all outliers (R_{mc}) fell above this line. Outliers from high moisture or impurity content will fall above the R_{mc} baseline and the ratio R_{mc}/g can be used to identify outliers.

Concerning equipment reliability, five NDA systems were shipped to Sellafield for this exercise and two were home based (BNFL). All of the equipment arrived at Sellafield in good working order, and no repair, maintenance, or replacement was required during the 5-day exercise.

Although the equipment was shut off each night because of plant safety regulations and restarted the next day, the good precisions quoted in Table XX were nonetheless achieved.

Measurements repeated a few days apart on the same samples (9 and 10) in the IAEA-2 counter gave good agreement, with an average difference of only 0.2% for the two sets of measurements. The exercise showed that achieving excellent precision is possible using the neutron counting systems under field operating conditions.

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