

LA-UR-

09-04489

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Title: Los Alamos Fission Basis

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Intended for: Joint LANL/LLNL Fission Product Evaluation Review Panel



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ABSTRACT

The purpose of this document is to provide the committee with an overview of the experimental methods used to establish the Los Alamos National Laboratory (LANL) fission basis. A final comprehensive report will be submitted for committee review following the meeting.

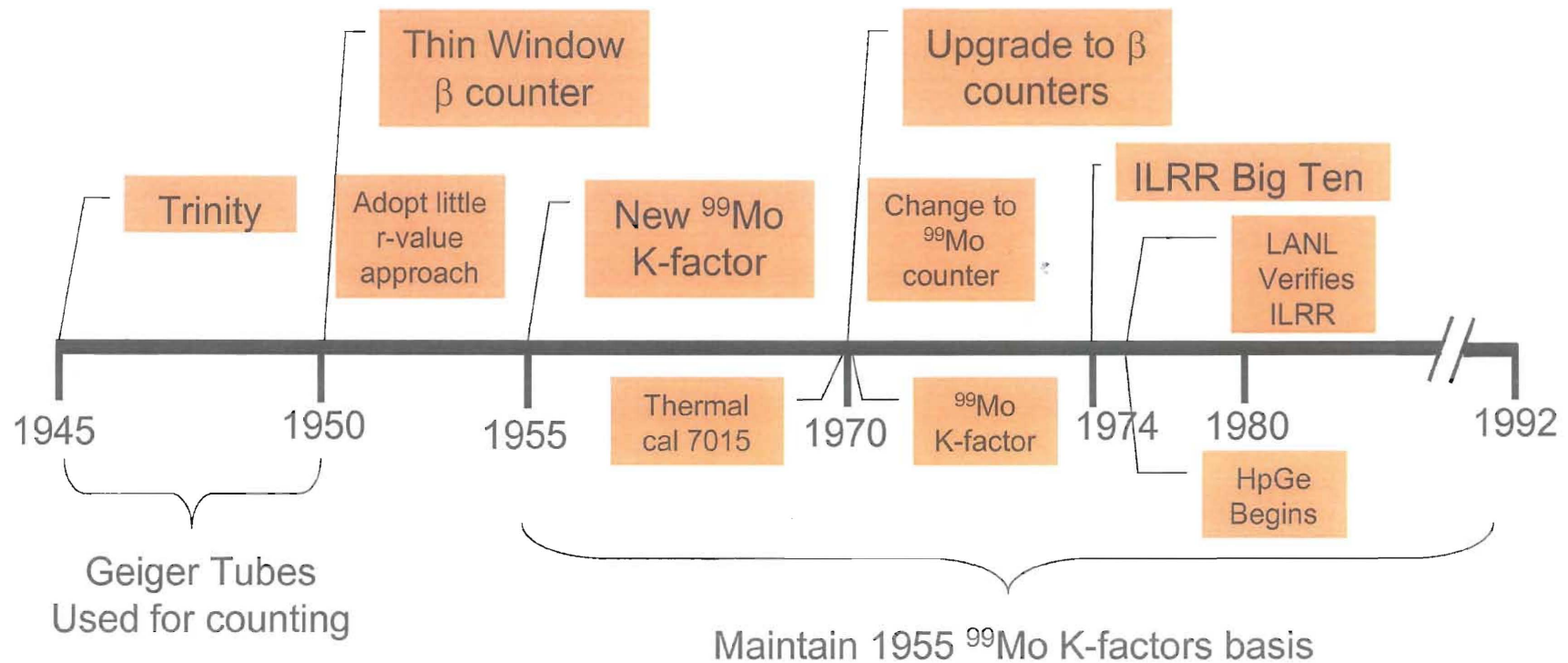
Previous reports have detailed the evolution of LANL fission quotation, focusing primarily on the changes in the fission basis as a function of improvements in technology and our understanding of the physics of fission itself [Mac07, Mac07B]. These reports span the period from the Trinity experiment [And45] to the cessation of nuclear testing in 1992, demonstrating LANL's painstaking effort to maintain the continuity and quality of the LANL fission basis. The present report will highlight the measurements required to establish, maintain, and update the LANL fission basis. Particular emphasis will be placed on the modern standardized measurements performed with the National Bureau of Standards (NBS, now NIST) -sponsored Inter-Laboratory Reaction Rate (ILRR) program and the subsequent LANL verification experiments. Elaboration of the methods will allow estimates of the uncertainties in the LANL experiments, providing an overall statement of the precision of LANL fission assessments.

Los Alamos Fission Basis

Carol Burns
Michael Mac Innes
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August Keksis

July 16-17 2009
Los Alamos NM

History of Fission Basis



GRAPHITE

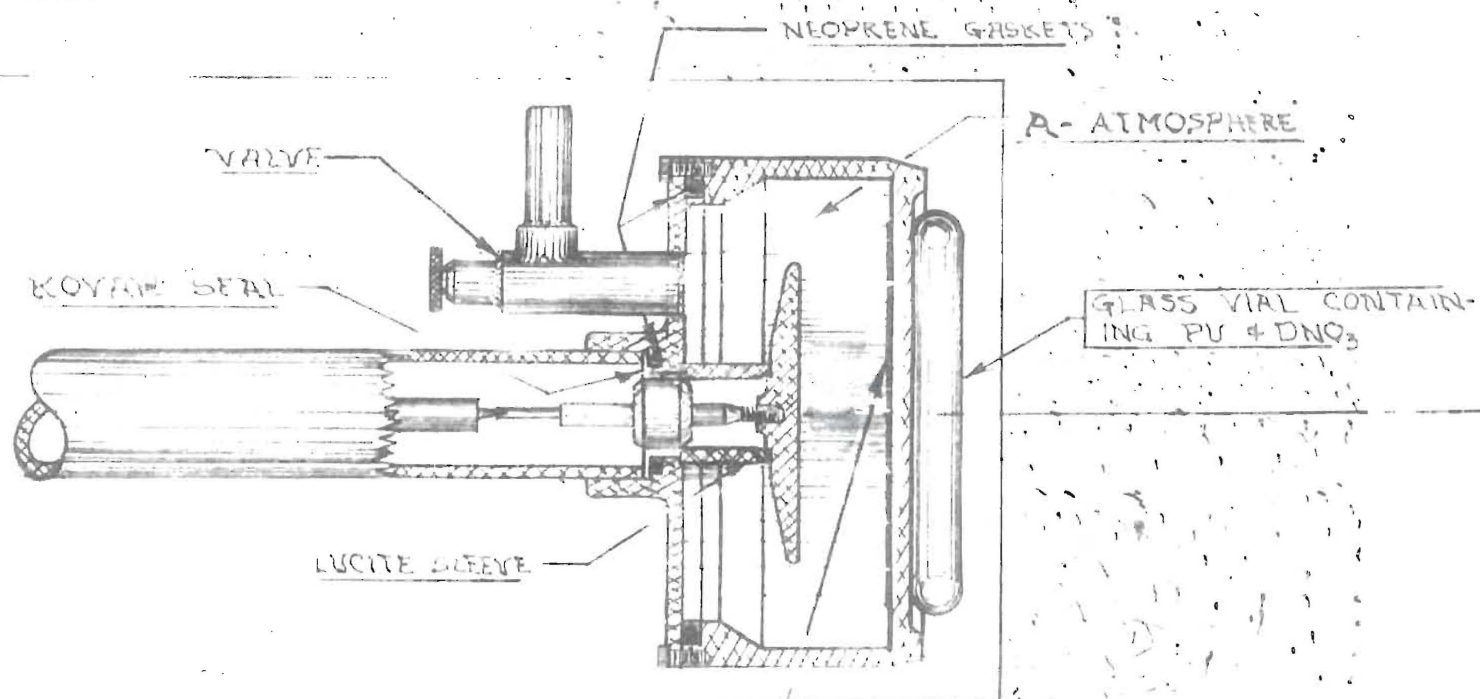


Fig. 2

"FISSION CALIBRATION"
MTL - AL. EXCEPT WHERE NOTED SCALE - 1" = 1"

DR.	DATE	CHK.	DATE
9/10/45	9/10/45		

LOS ALAMOS SCIENTIFIC LABORATORY
of
THE UNIVERSITY OF CALIFORNIA

May 2, 1951

LA-1238

THIN WINDOW PROPORTIONAL BETA CHAMBER

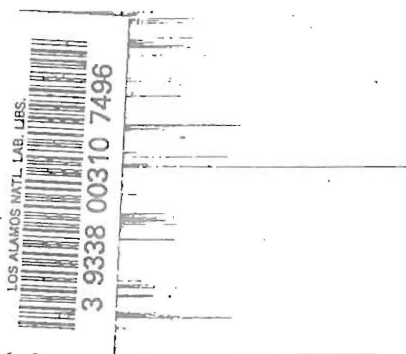
Work done by:

John H. Larkins

Report written by:

John H. Larkins

INSTRUMENTATION



Counters where fabricated to be identical

Typical Counter Ratios Calibrated by Mo-99 to Counter 6

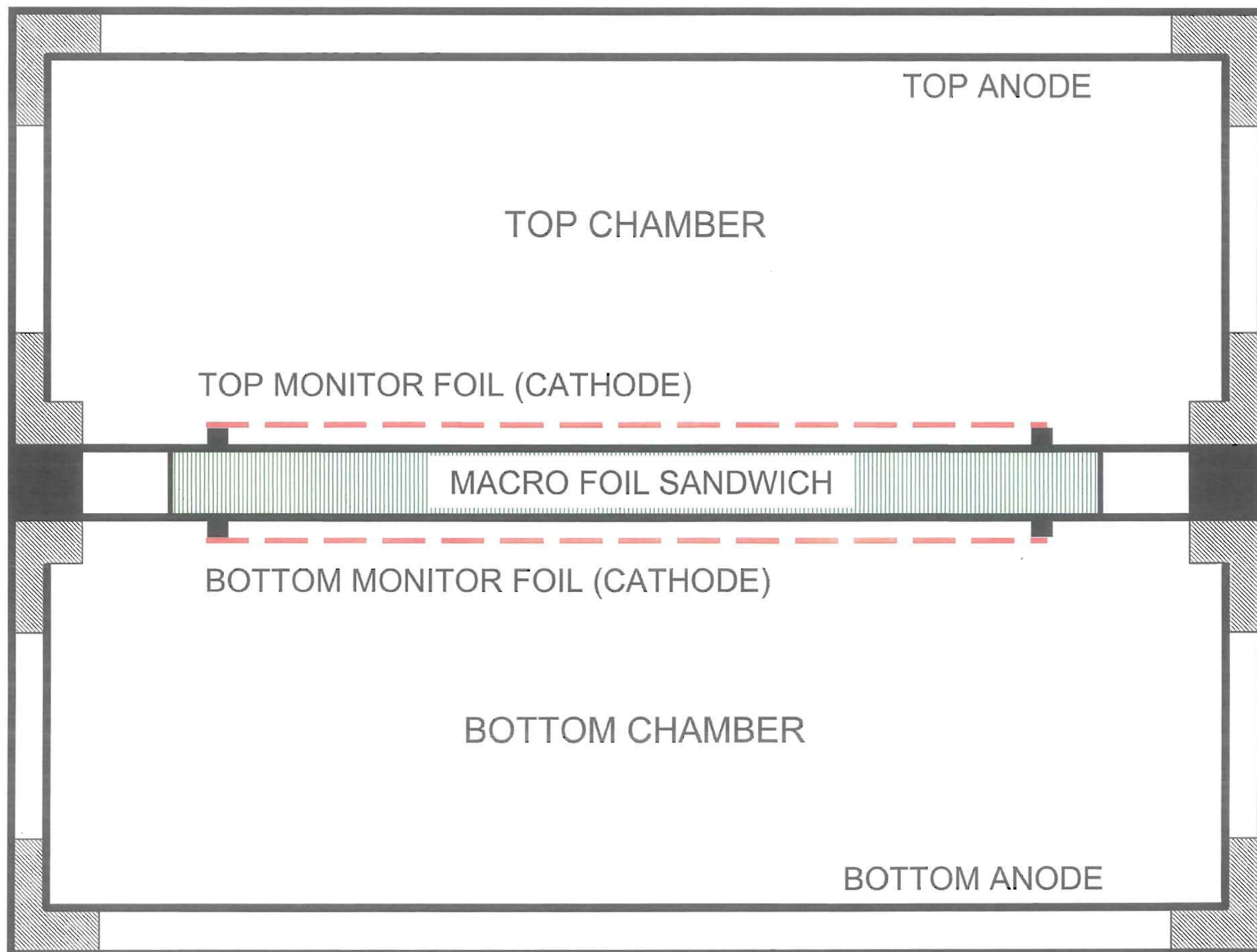
Counter Number	Count Rate Ratio to Counter 6
----------------	-------------------------------

2	1.0191
3	1.0183
4	1.0100
5	0.9993
6	1.0000
8	1.0108
9	1.0055
10	0.9922
12	1.0041
13	1.0022
14	0.9838
15	1.0018

Browne evaluated the standard deviation of radiochemical analyses over the period 1955 through 1958 for fission product measurements using the thin window limited-proportional beta counters. He determined that the average standard deviation for ^{99}Mo samples was 0.962 percent based on 348 cases studied. The average standard deviation calculated by Browne included the uncertainties associated with the counters and the chemical processing of the samples.

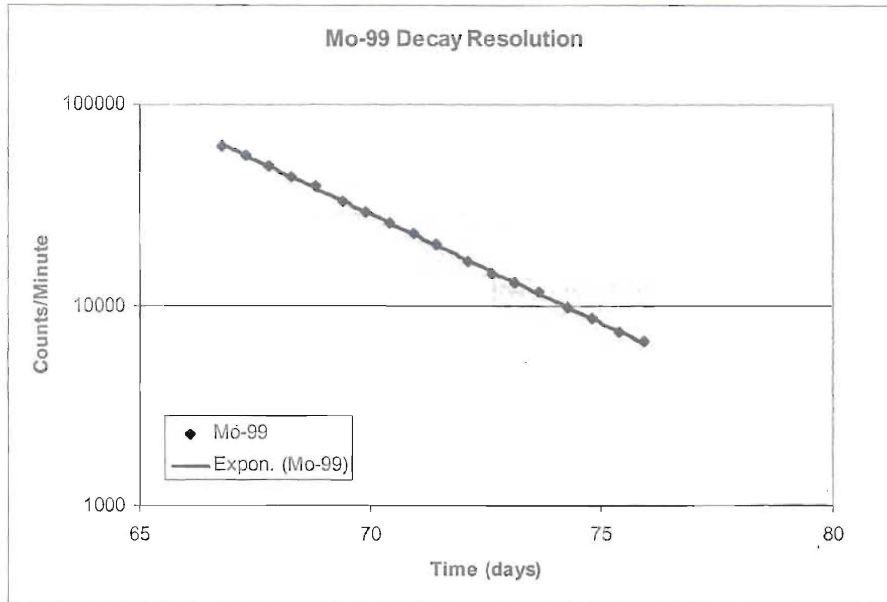
Larkins counter design “new” counter #6 circa 1975





UNCLASSIFIED

Fission Yield Basis Experimentally Determined by Mo-99 K-factor Method at LANL



1955 experiments with Balagna fission chamber used to define basis for entire experimental program (1951-1992)

K-factor results
(10^5 fission/Mo-99 cpm on old counter #6)

	U-235	Pu-239
K-1	2.358	2.451
K-2	2.340	2.437
K-3	2.419	2.426
K-4	2.363	2.494
K-5	2.365	
K-6	2.369	
Ave	2.369±1.1%	2.452±1.2%

Beta counting technique used for first 30 years of experiments at LANL

- Thus the historic ratio of K-factors, i.e. $Q_{49,th} = 0.966$, indicated the FPY of Pu-239 was smaller than that of U-235 for Mo-99.

Reproducibility of count rate ratios demonstrated for Nd/Mo

Little r for Nd

Date	cpm Nd/cpm Mo			
8/21/1962	0.03101			
1/24/1963	0.03083			
8/16/1963	0.03018			
9/4/1963	0.03054			
9/19/1963	0.03064			
1/13/1964	0.03034			
		0.0306	1.00%	0.33%
9/18/1964	0.03073			
7/19/1965	0.03064			
6/11/1966	0.03145			
average	0.03071	1.21%	0.40%	

Change in precipitate thickness correction

5/8/1967	0.02942			
1/4/1968	0.02932			
6/7/1968	0.02992			
1/3/1969	0.02982			
11/12/1968	0.0307			
4/24/1970	0.02999			
6/1/1970	0.03107			
10/1/1970	0.03038			
average	0.03008	2.01%	0.67%	

Los Alamos measured R-values for Zr-95 fast

U-235 fast Zr-95 R-values

Date

4/1/1971	0.9871
11/16/1971	0.9542
10/23/1973	0.9527
1/17/1974	0.9607
10/17/1974	0.9704
9/17/1974	0.9865
6/7/1976	0.9922

		std point	std mean
average	0.9720	1.71%	0.65%

Pu-239 fast R-values

Date

4/1/1971	0.747
11/16/1971	0.7181
10/23/1973	0.7251
10/17/1974	0.6983
6/7/1976	0.7286

		std point	std mean
average	0.7187	2.45%	0.93%

Los Alamos measured R-values for Nd-147 fast

U-235 fast Nd-147 R-values

Pu-239 fast Nd R-values

Date

1/25/1962	0.999	0.925
4/23/1962	0.986	0.894
8/5/1963	0.974	
	0.981	
	0.963	
	0.966	
	0.977	
	0.994	
4/19/1971	0.9740	0.902
	0.967	0.916
	0.962	0.902
	0.977	0.923
	0.954	0.915
	0.965	0.900
	0.972	0.904
	0.97	0.895
	0.979	
11/16/1971	0.977	0.974
10/23/1973	0.981	0.934
1/17/1974	0.965	
1/22/1974	0.9400	0.913
7/9/1974	0.968	0.889
	average	0.9133
	std point	2.39%
	std mean	0.51%

Counter cross calibration for New Counter 6

Mo-99 cross counter calibration

cpm ratio to 15

Date 8/20/1969 2/24/1970

Counter		average of 3	std
2		1.031	0.19%
old 6	1		
new 6		1.054	0.30%
9	1.033	1.034	0.20%
10	0.993	0.989	0.23%
13	1.025	1.028	0.06%
15	1	1	
16	1.003		
17		1.023	0.00%
19	1.035	1.030	0.30%
21		1.023	0.26%
24	0.985	0.985	0.10%
26	0.979	0.979	0.06%



TABLE V. Run 7015 r Values

Nuclide	Analyst	r Value Based on Old Conditions	Agreement with "old" r	r Value Based on New Conditions of ¹⁰⁸ Mo on #6 (Current) and ¹⁴⁷ Nd on #6	Agreement with "new" r
⁸⁸ Sr	BW/VA	4.781×10^{-2}	- 2.2%	5.040×10^{-2}	- 2.2%
⁸⁸ Sr	EAB/FO	$4.89 \times 10^{-2*}$	0%	$5.15 \times 10^{-2*}$	0%
⁹¹ Y	EAB/FO	$5.21 \times 10^{-2*}$	- 0.4%	$5.49 \times 10^{-2*}$	- 0.4%
⁹⁵ Zr	EAB/FO	$9.29 \times 10^{-3*}$	- 0.3%	$9.79 \times 10^{-3*}$	- 0.3%
¹¹¹ Ag	EAB/FO	$8.92 \times 10^{-4*}$	- 6.7%	$9.14 \times 10^{-4*}$	- 6.7%
¹¹⁵ Cd	BW/VA	3.688×10^{-3}	+ 0.7%	3.887×10^{-3}	+ 0.7%
¹¹⁵ Cd	EAB/FO	$3.64 \times 10^{-3*}$	- 0.4%	$3.85 \times 10^{-3*}$	- 0.5%
^{115m} Cd	BW/VA	9.136×10^{-8}	+ 2.3%	9.632×10^{-8}	+ 2.3%
^{115m} Cd	EAB/FO	$9.27 \times 10^{-8*}$	+ 3.8%	$9.77 \times 10^{-8*}$	+ 3.8%
¹²⁵ Sn	EAB/FO	$6.32 \times 10^{-4*}$	---	$6.74 \times 10^{-4*}$	---
¹³² Te	BW/VA	0.655	0%	0.670	+ 0.1%
¹³⁶ Cs	BW/FN	7.019×10^{-5}	- 0.4%	7.400×10^{-5}	- 0.4%
¹³⁷ Cs	BW/FN	1.711×10^{-4}	- 3.3%	1.804×10^{-4}	- 3.5%
¹⁴⁰ Ba	BW/VA	0.442	- 0.2%	0.448	- 0.2%
¹⁴⁰ Ba	EAB/FO	0.429*	- 3.2%	0.435*	- 3.2%
¹⁴⁴ Ce	BW/VA	3.919×10^{-3}	- 2.8%	3.481×10^{-3}	- 2.8%
¹⁴⁷ Nd	BW/JH	2.999×10^{-2}	- 2.0%	3.018×10^{-2}	- 2.0%
¹⁵⁰ Eu	BW/JH	3.243×10^{-4}	- 0.3%	2.942×10^{-4}	- 0.3%
¹⁶¹ Tb	BW/JH-FN	2.946×10^{-6}	- 13.1%	3.105×10^{-6}	- 13.1%

* Calculated on the basis of a sample diameter of 1.6 cm for nuclide of analysis but 1.9 cm for ⁸⁸Mo.

Counter-dependent K-factors and R-values

TABLE IV. K and R Values

J-11 Component	Old Value in Use Since 1964 or Earlier; Based Upon Listed # Counters	New Value Based Upon Listed # Counters
$K_{\text{Mn-}^{55}\text{Fe}}$	2.369×10^5 , ^{55}Fe on #6 (old)	2.497×10^5 , ^{55}Fe on #6 (current) 2.440×10^5 , ^{55}Fe on #19
$K_{\text{Mn-}^{55}\text{Co}}$	7.764×10^5 , ^{55}Co on #5	8.116×10^5 , ^{55}Co on #5 (current) 7.937×10^5 , ^{55}Co on #13
$R_{^{88}\text{Sr}}$	4.889×10^{-2} , ^{88}Sr on #6 (old), ^{88}Sr on #13	5.153×10^{-2} , ^{88}Sr on #6, ^{88}Sr on #13 5.036×10^{-2} , ^{88}Sr on #19, ^{88}Sr on #13
$R_{^{90}\text{Sr}}$	3.158×10^{-4} , ^{90}Sr on #6 (old), ^{90}Sr on #13	3.329×10^{-4} , ^{90}Sr on #6, ^{90}Sr on #13 3.255×10^{-4} , ^{90}Sr on #19, ^{90}Sr on #13
$R_{^{91}\text{Y}}$	5.23×10^{-2} , ^{91}Y on #6 (old), ^{91}Y on #21	5.51×10^{-2} , ^{91}Y on #6, ^{91}Y on #21 5.39×10^{-2} , ^{91}Y on #19, ^{91}Y on #21
$R_{^{95}\text{Zr}}$	9.32×10^{-3} , ^{95}Zr on #6 (old), ^{95}Zr on #31	9.52×10^{-3} , ^{95}Zr on #6, ^{95}Zr on #31 9.60×10^{-3} , ^{95}Zr on #19, ^{95}Zr on #31
$R_{^{111}\text{Ag}}$	9.515×10^{-4} , ^{111}Ag on #6 (old), ^{111}Ag on #9 pig	9.756×10^{-4} , ^{111}Ag on #6, ^{111}Ag on #21 9.534×10^{-4} , ^{111}Ag on #19, ^{111}Ag on #9
$R_{^{115}\text{Cd}}$	3.661×10^{-3} , ^{115}Cd on #6 (old), ^{115}Cd on #13	3.859×10^{-3} , ^{115}Cd on #6, ^{115}Cd on #13 3.771×10^{-3} , ^{115}Cd on #19, ^{115}Cd on #13
$R_{^{115m}\text{Cd}}$	8.935×10^{-6} , ^{115m}Cd on #6 (old), ^{115m}Cd on #13	9.415×10^{-6} , ^{115m}Cd on #6, ^{115m}Cd on #13 9.201×10^{-6} , ^{115m}Cd on #19, ^{115m}Cd on #13
$R_{^{127}\text{Sb}}$	1.42×10^{-3} , ^{127}Sb on #6 (old), ^{127}Sb on #31	1.43×10^{-3} , ^{127}Sb on #6, ^{127}Sb on #31 1.40×10^{-3} , ^{127}Sb on #19, ^{127}Sb on #31
$R_{^{132}\text{Te}}$	0.635 , ^{132}Te on #6 (old), ^{132}Te on #31	0.669 , ^{132}Te on #6, ^{132}Te on #31 0.654 , ^{132}Te on #19, ^{132}Te on #31
$R_{^{137}\text{Cs}}$	7.048×10^{-5} , ^{137}Cs on #6 (old), ^{137}Cs on #3	7.423×10^{-5} , ^{137}Cs on #6, ^{137}Cs on #3 7.259×10^{-5} , ^{137}Cs on #19, ^{137}Cs on #3
$R_{^{137}\text{Cs}}$	1.763×10^{-4} , ^{137}Cs on #6 (old), ^{137}Cs on #3	1.863×10^{-4} , ^{137}Cs on #6, ^{137}Cs on #3 1.821×10^{-4} , ^{137}Cs on #19, ^{137}Cs on #3
$R_{^{140}\text{Ba}}$	0.443 , ^{140}Ba on #6 (old), ^{140}Ba on #13	0.443 , ^{140}Ba on #6, ^{140}Ba on #13 0.430 , ^{140}Ba on #19, ^{140}Ba on #13
$R_{^{144}\text{Ce}}$	4.010×10^{-3} , ^{144}Ce on #6 (old), ^{144}Ce on #9 pig	3.578×10^{-3} , ^{144}Ce on #6, ^{144}Ce on #9 3.453×10^{-3} , ^{144}Ce on #19, ^{144}Ce on #9
$R_{^{147}\text{Nd}}$	3.059×10^{-2} , ^{147}Nd on #6 (old), ^{147}Nd on #5	3.076×10^{-2} , ^{147}Nd on #6, ^{147}Nd on #5 3.080×10^{-2} , ^{147}Nd on #19, ^{147}Nd on #5
$R_{^{156}\text{Eu}}$	3.252×10^{-4} , ^{156}Eu on #6 (old), ^{156}Eu on #18	2.949×10^{-4} , ^{156}Eu on #6, ^{156}Eu on #19 2.882×10^{-4} , ^{156}Eu on #19, ^{156}Eu on #18
$R_{^{161}\text{Tb}}$	3.332×10^{-6} , ^{161}Tb on #6 (old), ^{161}Tb on #16	3.512×10^{-6} , ^{161}Tb on #6, ^{161}Tb on #24 3.432×10^{-6} , ^{161}Tb on #19, ^{161}Tb on #24

1970 K-factor results with uncertainties

A) Fissions

For the 180 minute run, monitor foil A (63.77 nanograms) was in side 1 of the fission counting chamber.

	<u>Σ Fissions above Bkg.</u>	<u>Efficiency</u>	<u>Fiss/mg ²³⁵U</u>
Scaler #1	8.3126×10^8		
Scaler #2	8.3250×10^8		
Avg.	8.3192×10^8	97.93%	1.332×10^{11}
	Spread = 0.2%		

Side 2 Foil B (64.14 nanograms)

Scaler #1	8.4044×10^8		
Scaler #2	8.4160×10^8		
Avg.	8.4105×10^8	97.45%	1.346×10^{11}
	Spread = 0.2%		

Avg. 1.339×10^{11}

Sp = 1.0%

σ = 0.5%

B) ²³⁵U in Sandwich

Sample	vs Monitor A mg/ml A	vs Monitor B mg/ml A
1908-1	4.706×10^{-2}	4.709×10^{-2}
-2	4.650×10^{-2}	4.657×10^{-2}
-3	4.690×10^{-2}	4.696×10^{-2}
-4	4.683×10^{-2}	4.693×10^{-2}
Avg.	4.682×10^{-2} Sp = 1.2%	4.689×10^{-2} Spread = 1.1%
	Average = 4.686×10^{-2}	Spread = 1.3% σ = 0.2%

C) ⁹⁹Mo in Sandwich

cpm/ml A Corrected to EOB and Decay during Bombdt

Sample #	P. Norman	P. Oliver
1908-1	2.666×10^4	2.746×10^4
-2	2.653×10^4	2.703×10^4
-3	2.641×10^4	
-4	2.649×10^4	

2.652×10^4 2.724×10^4

Spread 0.9% Spread 1.6%

Average = 2.676×10^4 Sp = 4.0% σ = 0.6%

D) ¹⁴⁷Nd in Solution

Sample #	cpm/ml A Corrected to EOB and Decay during Bombdt
1908-1	8.219×10^2
-2	8.358×10^2
-3	8.422×10^2
-4	8.275×10^2
	8.314×10^2 Spread = 2.5% σ = 0.5%

$$E) K_{Mo} = 2.345 \times 10^5 \quad \sigma = 0.8\%$$

$$K_{Mo} = \frac{\text{Fiss/mg } ^{235}\text{U} \times ^{235}\text{U/ml A}}{^{99}\text{Mo cpm/ml A}}$$

$$= \frac{1.339 \times 10^{11} \times 4.686 \times 10^{-2}}{2.676 \times 10^4} = 2.345 \times 10^5$$

K-factor Summary in 1970 Knobeloch Memo

<u>Date</u>	<u>K_{Mo}</u>	<u>Number of determinations</u>	<u>σ</u>	<u>Remarks</u>
Jan 1951	2.501×10^5	6	1.9%	Glass vial and discriminator plateau method
Jan 1953	2.50×10^5	1	-	" " " "
Jan 1955	2.369×10^5	6	1.0%	1 mil Ni sandwich $\frac{1}{2}$ mil Pt -15 mil Al monitors and Pulse height analysis method.
Mar 1956	2.350×10^5	4	1.0%	" " "
Mar 1958	2.431×10^5	3	1.1%	" " "
Oct 1960	2.398×10^5	2	2.4%	" " "
Feb 1961	2.398×10^5	3	0.6%	" " "
Sept 1965	2.349×10^5	2	0.9%	1 mil Ni sandwich $\frac{1}{2}$ mil Pt 2 mil Al monitors Pulse height anal. method
June 1970	2.349×10^5	1	0.8%	1/4 mil mylar sandwich $\frac{1}{2}$ mil Pt 2 mil Al monitors Pulse Height anal. method
Wt'd Avg.	2.378×10^5	21	1.1%	Sandwich and pulse height analysis method

Mo K-factor Summary 1970 counting conditions

Counter # 19	U-235	U-238	Pu-239
Historic K-factor	2.440E+05	2.372E+05	2.526E+05
Counter # 6	U-235	U-238	Pu-239
Historic K-factor	2.497E+05	2.427E+05	2.585E+05

ILRR Big Ten Irradiation

- Collaborative effort led by NBS involving several laboratory engaged in reactor fuel burn-up measurements
- Establish highly accurate fission reaction rates for interlaboratory calibration
- Produced consensus fission product yields as measured by high-resolution gamma spectroscopy at participating laboratories

Test: BIG-10 Fission Foil Set (Test 74-14, 8th Quarterly; Test C, April 1974 Test Plan Letter)

HEDL Capsule No.: LASL 74-14

Container Ident.: 74-14

Set Ident.: See attached Figure 2

By: WN McElroy, GE Shook

Date

Completed: 5/23/74

Material	Isotopic Enrichment (Elemental Composition)	Source (ORNL)	Sample Number	Material			Encapsulated Foil			HEDL Weight (mg)
				Weight (mg)	Dia. (Inch)	Thick. (mils)	Dia. (Inch)	Thick. (mils)	ORNL Weight (mg)	
235-U	99.89% (99.44%)	U-235-264-CM	U5-7	277.972	0.5	5.1	0.75	21.5	530.28	530.29
237-Np	99.99% (97.50%)*	Np-237-NP21M	N7-12	467.431	0.5	9.1-9.2	0.75	22.7	725.99	726.05
238-U	99.998% (99.48%)	U-238-ES-ZM	U8-10	304.007	0.5	5.2	0.75	21.5	557.7	557.55
239-Pu	99.11% (98.25%)	Pu-239-453BM	P9-10	267.977	0.5	5.7	0.75	21.0	522.12	522.13

Final Measured Capsule Dimensions:**

Total Weight = 2.43839 gm.

Diameter = 0.75-0.76"

Thickness = 0.10-0.11"

* ORNL result based on two foil samples, HEDL obtained a result of 99.70% on a single foil sample; further samples will be analyzed to resolve the difference between the ORNL and HEDL results.

** The nominal weights of the ~0.75" diameter Al foil holder bottom and top are ~0.0445 gm and ~0.0152 gm, respectively. Al foil spacers were used to separate each of the four fission foils. These spacers were made the same as the foil holder top. The thickness of the Al foil was ~0.8 mils for all of these items.

NBS Reactions Rates for Big Ten Experiments

TABLE NBS-20
FISSION RATE PER ATOM IN BIG-10*

		Principal Isotope of Fission Foil			
		U-235	U-238	Pu-239	Np-237
Perturbed Flux/ Flux in Cavity-Free BIG-10		.994 ±.25%	1.000 ±.25%	.995 ±.25%	1.001 ±.25%
Combined Errors in Fiss. Rate/Atom †		1.5%	1.9%	1.7%	3.0%
Run-to-run Normalization Error		0.3%	0.3%	0.3%	0.3%
Test 74-15 C-ANC	10 ⁻¹⁵ fission per second per atom of principal isotope	111.18	4.1720	133.33	35.491
Test 74-16 C-ANL		111.11	4.1694	133.24	35.469
Test 74-13 C-HEDL		111.33	4.1776	133.51	35.540
Test 74-17 C-MGL		110.88	4.1608	132.97	35.396
Test 74-14 C-LASL		110.35	4.1410	132.34	35.228

* The fission rates given include calculated flux perturbations so that these values represent the average flux seen by the fission activation foil.

† The uncertainty in the fission rate due to uncertainty in the chemical purity of the activation foils has been included in the combined errors given here.

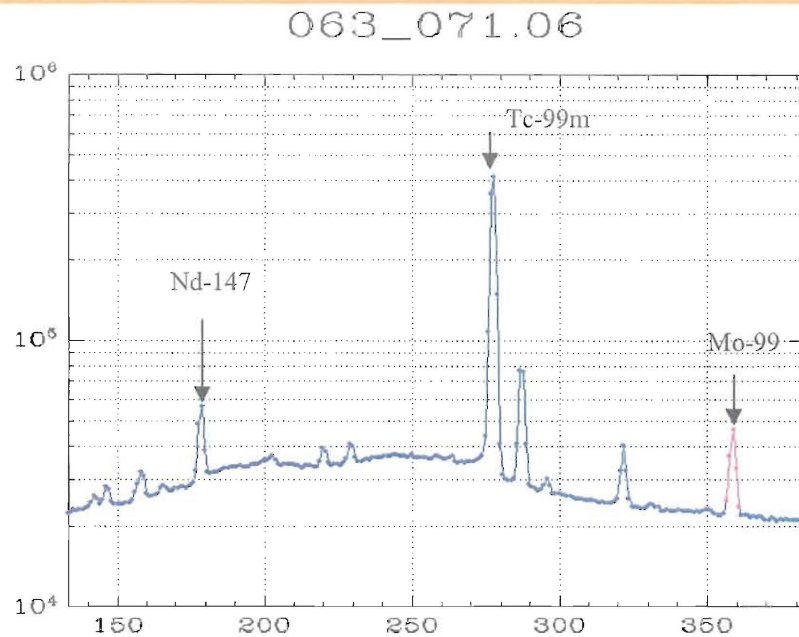
Comparison of NBS and LASL Reactions Rates for Big Ten Experiments

Test 74-14 C-LASL Fission Rate per Atom

	Principle Isotope			
	U-235	U-238	Pu-239	Np-237
NBS	1.104E-13	4.141E-15	1.323E-13	3.523E-14
LASL	1.127E-13	4.212E-15	1.420E-13	3.526E-14
LASL/NBS	1.021	1.017	1.073	1.001

LANL historic K-factor is 7% higher than NBS fission chamber results

Fission basis using gamma-ray measurements tied to Mo-99 K-factor method through thermal calibrations



Early LANL gamma-ray measurements were converted using K-factor method

Analysis of measurement is a count rate. Still need a K-factor to determine fissions.

- After adopting common gamma analysis practice, thermal FP yields were defined to maintain consistency with Mo-99 K-factor method

	Historic FPY _{25,th}	Modern FPY _{25,th}
Mo-99	0.0595	0.0606
Nd-147	0.0219	0.0223

LANL Experimental Determination of Thermal Fission Chain yields

Calibration	Mo-99	Nd-147	Zr-95	Ba-140	Ce-141	Ce-143
2468	0.0597					
2693	0.0619					
7512	0.0597					
7625	0.0597					
8004	0.0615	0.0221	0.0642	0.0627	0.0582	0.0602
8105	0.0646	0.0228	0.0655	0.0646	0.0612	0.0638
8202	0.0641	0.0227	0.0656	0.0641	0.0608	0.0641
8391	0.0631	0.0223	0.0642	0.0628	0.0594	0.0612
8501	0.0623	0.0224	0.0632	0.0627	0.0588	0.0609
8606	0.0608	0.0223	0.0643	0.0633	0.0596	0.0612
8606	0.0600	0.0227	0.0652	0.0638		
8801	0.0587	0.0219	0.0628	0.0618	0.0582	0.0594
9001	0.0602	0.0217	0.0635	0.0622	0.0583	0.0598
9207	0.0599	0.0225	0.0664	0.0601	0.0601	0.0621
Average	0.0606	0.0223	0.0645	0.0631	0.0594	0.0614
std point	2.15%	1.61%	1.80%	1.45%	1.90%	2.69%
std mean	0.60%	0.49%	0.54%	0.46%	0.60%	0.85%

Comparison of Los Alamos and Livermore Nd-147 atoms using gamma spectroscopy

¹⁴⁷Nd atoms/ml

cal	LLNL	LANL	LANL/LLNL
8004	2.341E+12	2.311E+12	9.872E-01
8105	1.167E+12	1.145E+12	9.811E-01
8202	1.101E+12	1.074E+12	9.755E-01
8391		1.044E+12	
8501	9.823E+11	9.735E+11	9.910E-01
8606	5.199E+11	5.141E+11	9.888E-01
8801	6.576E+11	6.371E+11	9.688E-01
9001		6.398E+11	
9207	4.301E+11	4.160E+11	9.672E-01
		average	9.800E-01
		stdev	0.99%

History of Mo K-factors at LANL

Mo-99 Thermal k-factor experiments
(old counter 6)

Year	Exp #	K(Mo)	Mg of U	target cm ²	mg/cm ²	blackness or attenuation correction	corrected K(Mo)
1951		2.501E+05					
1952	k-1	2.354E+05	11.26	5.067	2.2222	0.9962	2.345E+05
	k-2	2.371E+05	11.45	5.067	2.2597	0.9923	2.353E+05
	k-3	2.383E+05	10.61	5.067	2.0939	0.9888	2.356E+05
1953		2.500E+05					
1951, 52, 53 used glass vial slug and were external to the thermal column. 1955 and later used the foil packet sandwich.							
1955	k-1	2.358E+05	6.328	5.067	1.2489	0.9925	2.340E+05
	k-2	2.340E+05	4.638	5.067	0.9153	0.9942	2.326E+05
	k-3	2.419E+05	5.735	5.067	1.1318	0.9931	2.402E+05
	k-4	2.363E+05	4.707	5.067	0.9290	0.9941	2.349E+05
	k-5	2.365E+05	4.877	5.067	0.9625	0.9940	2.351E+05
	k-6	2.369E+05	4.931	5.067	0.9732	0.9939	2.355E+05
1956	k-7	2.339E+05	8.888	5.067	1.7541	0.9899	2.315E+05
	k-8	2.367E+05	10.31	5.067	2.0347	0.9885	2.340E+05
	k-9	2.358E+05	8.992	5.067	1.7746	0.9896	2.334E+05
	k-10	2.336E+05	10.16	5.067	2.0051	0.9887	2.310E+05
1958	k-1	2.371E+05	8.575	5.067	1.6923	0.9902	2.348E+05
	k-2	2.337E+05	8.962	5.067	1.7687	0.9898	2.313E+05
	k-3	2.376E+05	8.826	5.067	1.7419	0.9900	2.352E+05
1960	k-1	2.342E+05	9.23	5.067	1.8216	0.9896	2.318E+05
	k-2	2.454E+05	9.19	5.067	1.8137	0.9896	2.429E+05
1961	k-1	2.369E+05	4.174	5.067	0.8238	0.9947	2.357E+05
	k-2	2.403E+05	4.026	5.067	0.7946	0.9949	2.391E+05
	k-3	2.421E+05	3.948	5.067	0.7792	0.9950	2.409E+05
1965	k-1	2.361E+05	7.288	5.067	1.4383	0.9915	2.341E+05
	k-2	2.336E+05	7.37	5.067	1.4545	0.9914	2.316E+05
beginning with 1970, all results converted to old counter 6.							
1970	k-1	2.345E+05	4.68	5.067	0.9236	0.9942	2.331E+05
1975-78	2662	2.462E+05	2.825	0.7854	3.5969	0.9815	2.416E+05
	2667	2.334E+05	4.754	3.941	1.2063	0.9927	2.317E+05
	2693	2.326E+05	4.201	3.941	1.0660	0.9934	2.311E+05
	2725	2.309E+05	0.5147	1.27	0.4053	0.9972	2.303E+05
	2755	2.310E+05	0.5628	1.27	0.4431	0.9969	2.303E+05
average							2.333E+05
x 1.054							2.459E+05

The value used by CIB and GWK was 2.369E+05 for old counter 6.

In January 1970 a new body was put in old counter six lowering the count rate by a factor of 1.054. The counter became known as new counter 6 with a k of 2.497E+05.

Holy Counter 6 and Summary of Mo K-factors

Year	Experiment	Counter	$K_{99}^{235,th(1)}$ ($\times 10^5$ f/cpm)	$K_{99}^{235,th(2)}$ ($\times 10^5$ f/cpm)
1951 ^c		6		2.501
1952 ^c	k-1	6	2.354	
	k-2	6	2.371	
	k-3	6	2.383	
1953 ^c		6		2.500
1955	k-1	8	2.358	2.408
	k-2	8	2.340	2.385
	k-3	8	2.419	
	k-4	8	2.363	
	k-5	8	2.365	
	k-6	8	2.369	
1956	k-7	6	2.339	
	k-8	6	2.367	
	k-9	6	2.358	
	k-10	6	2.336	
1958	k-1	6	2.371	
	k-2	6	2.337	
	k-3	6	2.376	
1960	k-1	6	2.342	
	k-2	6	2.454	
1961 ^e	k-1	6	2.369	
	k-2	6	2.403	
	k-3	6	2.421	
1965	1241	6	2.361	
	1242	6	2.336	
1970	1908	new 6	2.472	
Mar-76	2662	new 6	2.595	
Apr-76	2667	new 6	2.460	
Sep-76	2693	new 6	2.452	
Oct-76	2725	new 6	2.434	
May-77	2755	new 6	2.435	



The “Modernization” Experiments of 1974-1977

A Look at the Methods, Materials, and Results

From LANL Radiochemistry’s Efforts

Post-ILRR

Hugh D. Selby

Scientist, C-NR

Definition of Terms

The K-factor

$$K_i = F / A_i$$

The Relationship to FPY

$$A_i = N_i \varepsilon_i \lambda_i = F Y_i \varepsilon_i \lambda_i$$

$$K_i = F / F Y_i \varepsilon_i \lambda_i = 1 / Y_i \varepsilon_i \lambda_i$$

Q-value Construction from K-factors

$$Q_i = K_i^* / K_i^p = Y_i^p / Y_i^*$$

FPY interconversion with Q-values

$$Y_i^p = Q_i Y_i^*$$

The R-value

$$R_j = \frac{A_j^{f,e} / A_i^{f,e}}{A_j^* / A_i^*} = \frac{r_j^{f,e}}{r_j^*}$$

Interconversion and Scaling of K_i and Q_i

$$K_j^{f,e} = \frac{K_{99}^{f,e}}{r_j^* R_j^{f,e}}$$

$$Q_j^{f,e} = R_j^{f,e} \frac{K_{99}^*}{K_{99}^{f,e}} = R_j^{f,e} Q_{99}^{f,e}$$

Definition of Terms 2

$$Y_i = \frac{R_i}{F}$$

$$\frac{A_i}{(\epsilon\lambda)_i} = Y_i F$$

$$\frac{A_i^*}{(\epsilon\lambda)_i} = Y_i^* F^*$$

$$\frac{A_i/F}{A_i^*/F^*} = \frac{Y_i}{Y_i^*} = Q_i$$

$$\frac{A_i/A_R}{A_i^*/A_R^*} = \frac{A_i/A_R}{\epsilon} = R = \frac{Q_i}{R} = \frac{Y_i/Y_i^*}{Y_R/Y_R^*}$$

$$Q_i = \sum_j \epsilon_j Q_{ij}$$

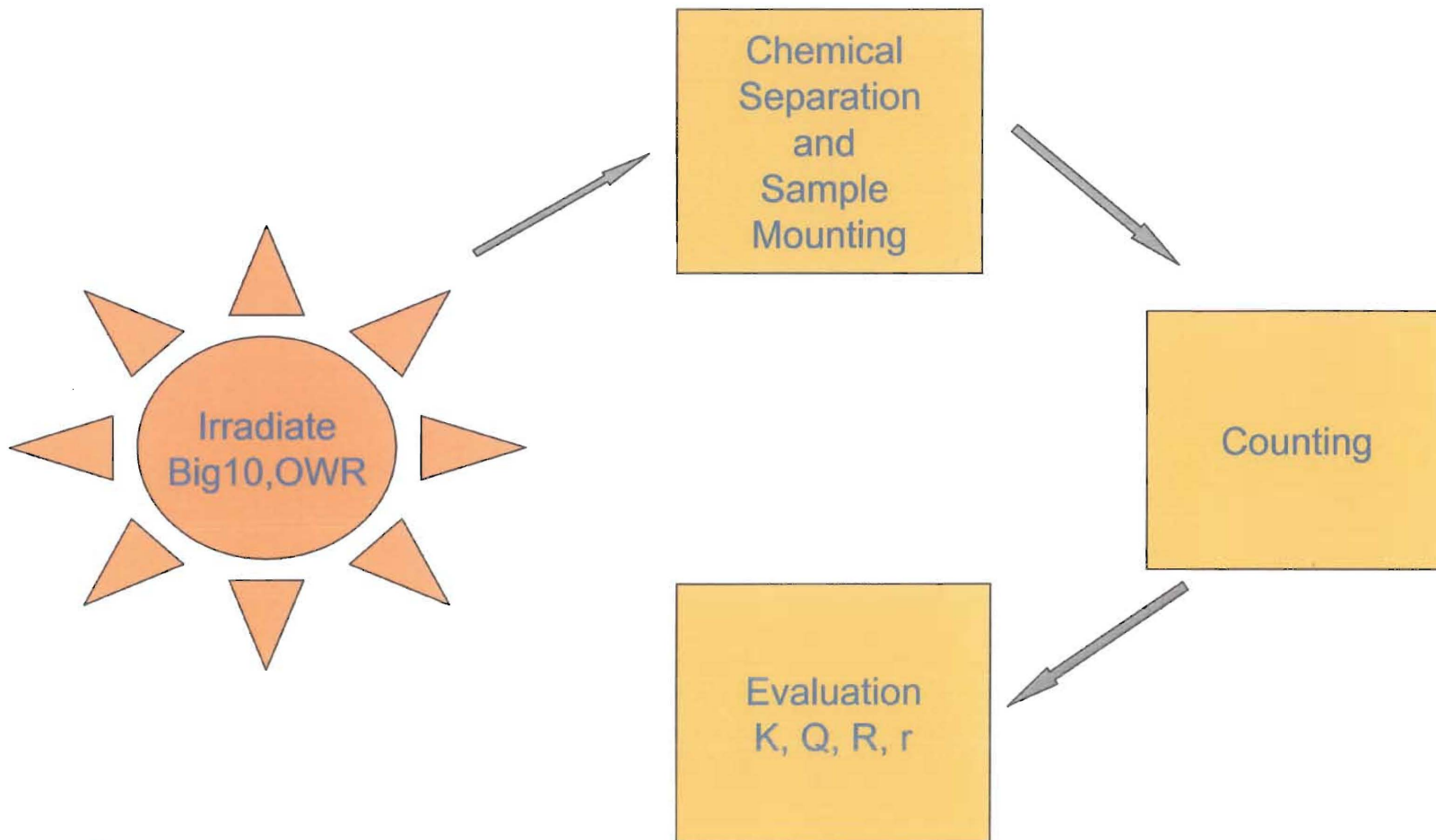
for the case of j
mixed fission types
each contributing fraction ϵ ($\sum_j \epsilon = 1$)

NECESSARY:

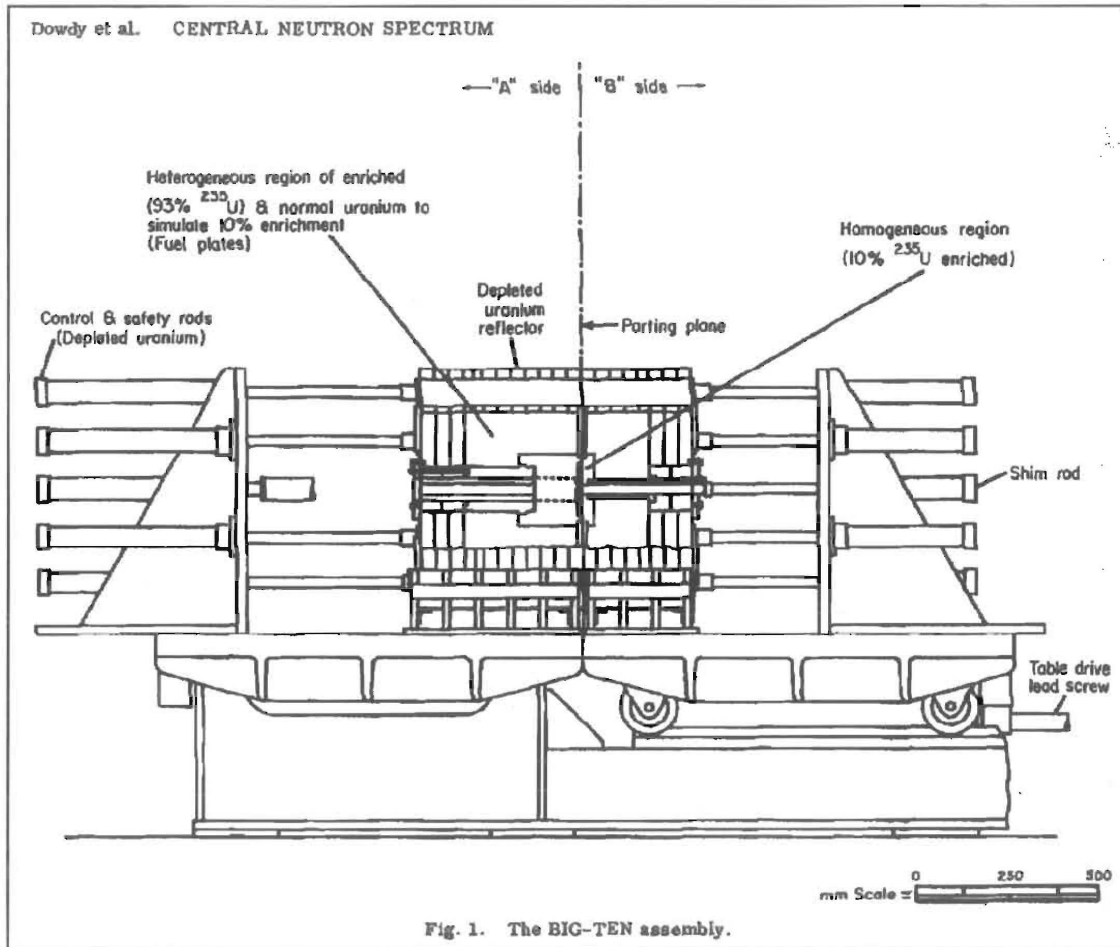
Y = (Radioactive) fission product yield
 R = " " " " atoms
 λ = " " " " decay constant
 A = " " " " instrument count rate
 ϵ = " " " " and decay scheme efficiency

F = number of fissions
 sub * = reference spectrum (^{235}U + "thermal" neutrons)
 sub R = " radionuclide (^{89}Po for LASL)
 sub i = generalized (radioactive) fission product
 Q, K, ϵ, R as defined above

The K-factor Measurement Process



Irradiation: Facilities



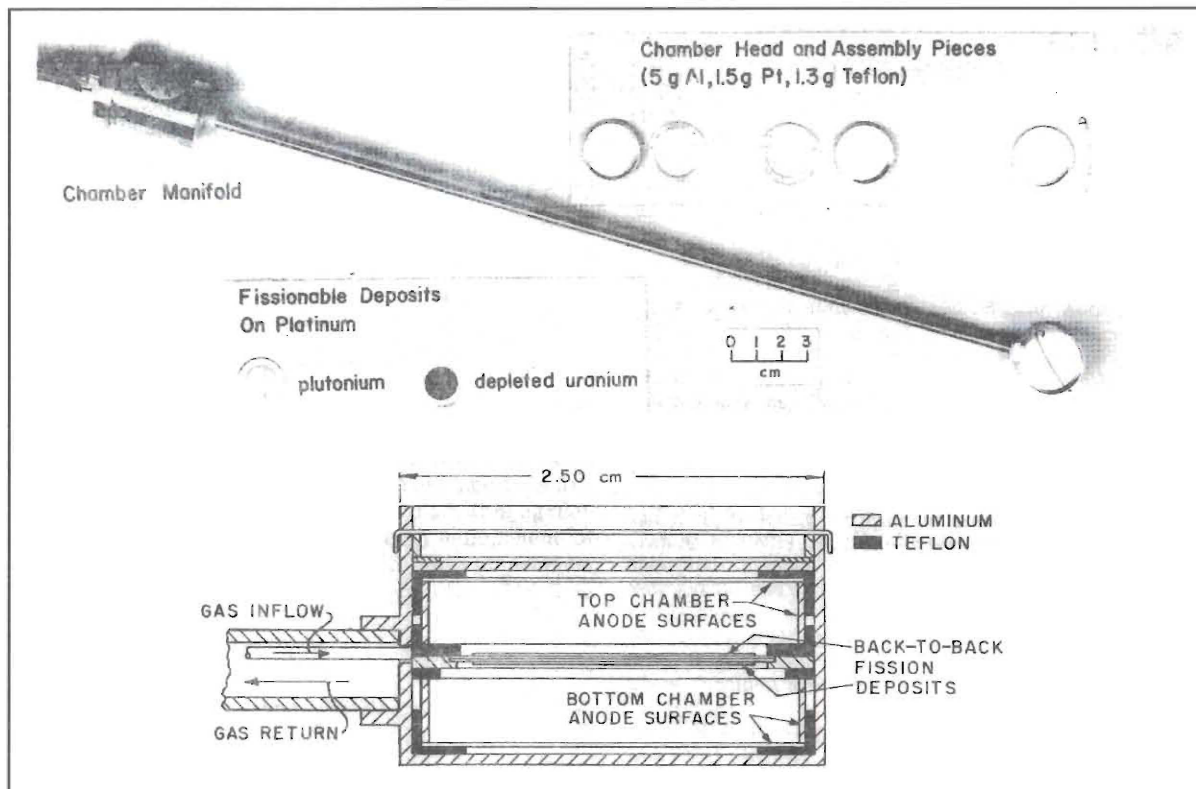
Big Ten Fast Critical Assembly



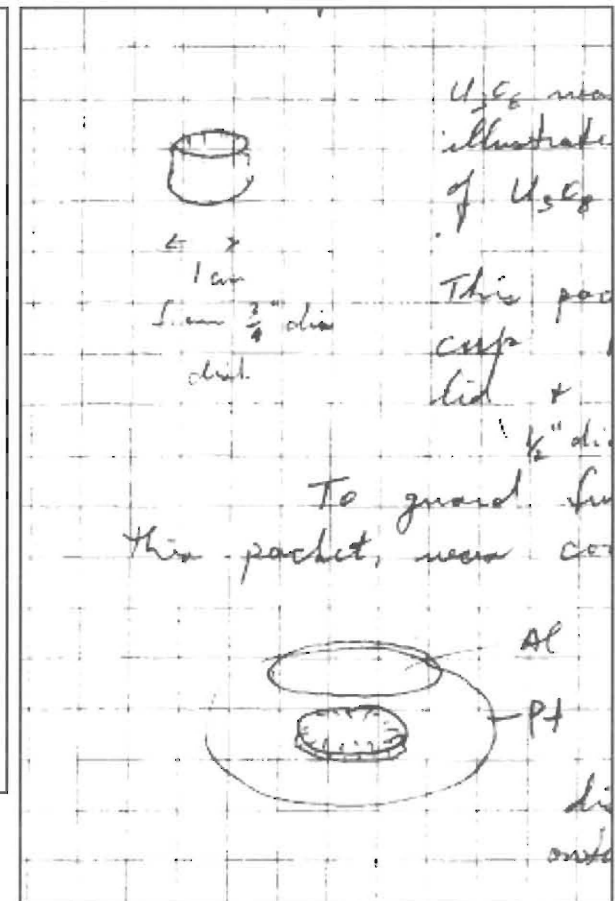
Thermal Column in OWR*

***NOT a picture of OWR!**

Irradiation Equipment and Materials

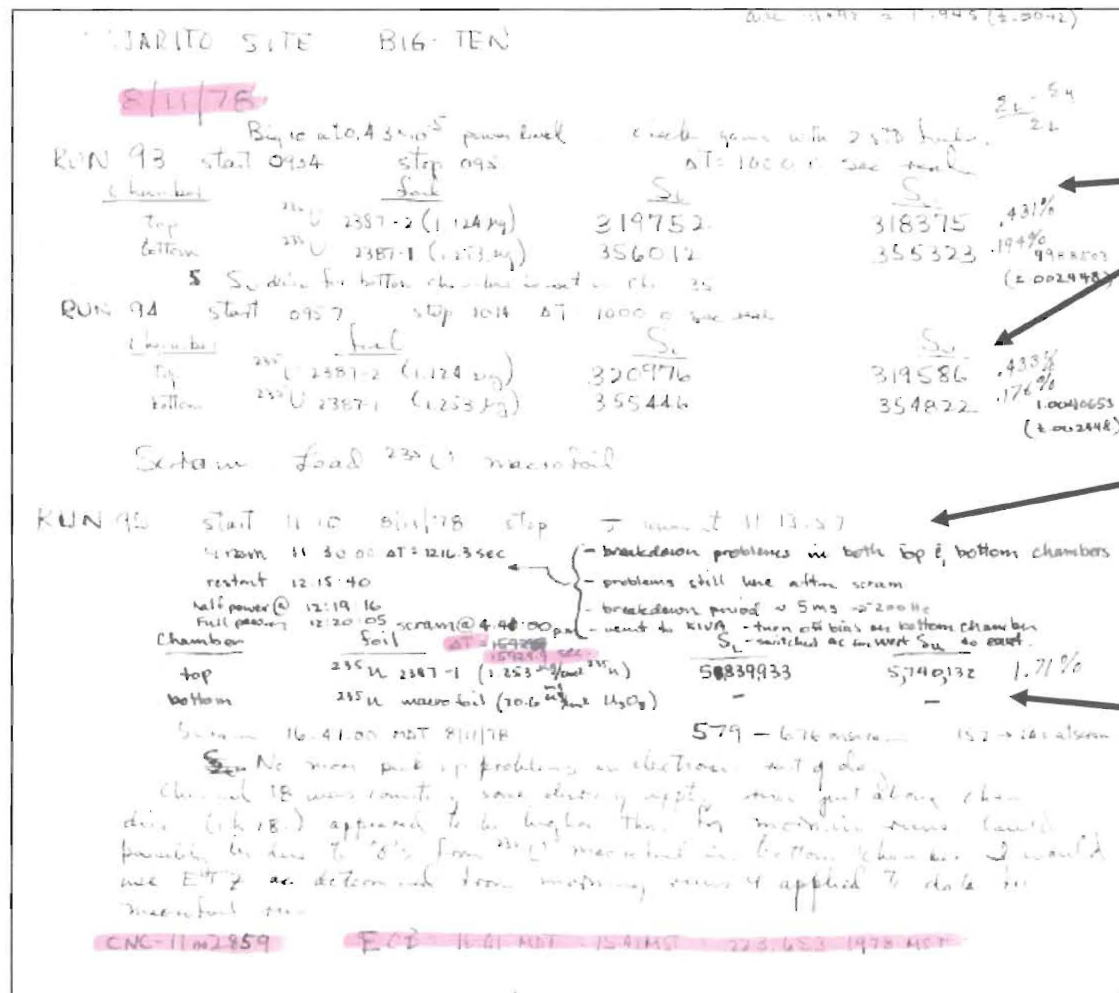


NBS (Grundl-type) Dual Fission Chamber



**Knobeloch Target Foil
Fabrication**

Irradiation "Checkout" and K-factor Measurement



Checkout Runs on Standard foils

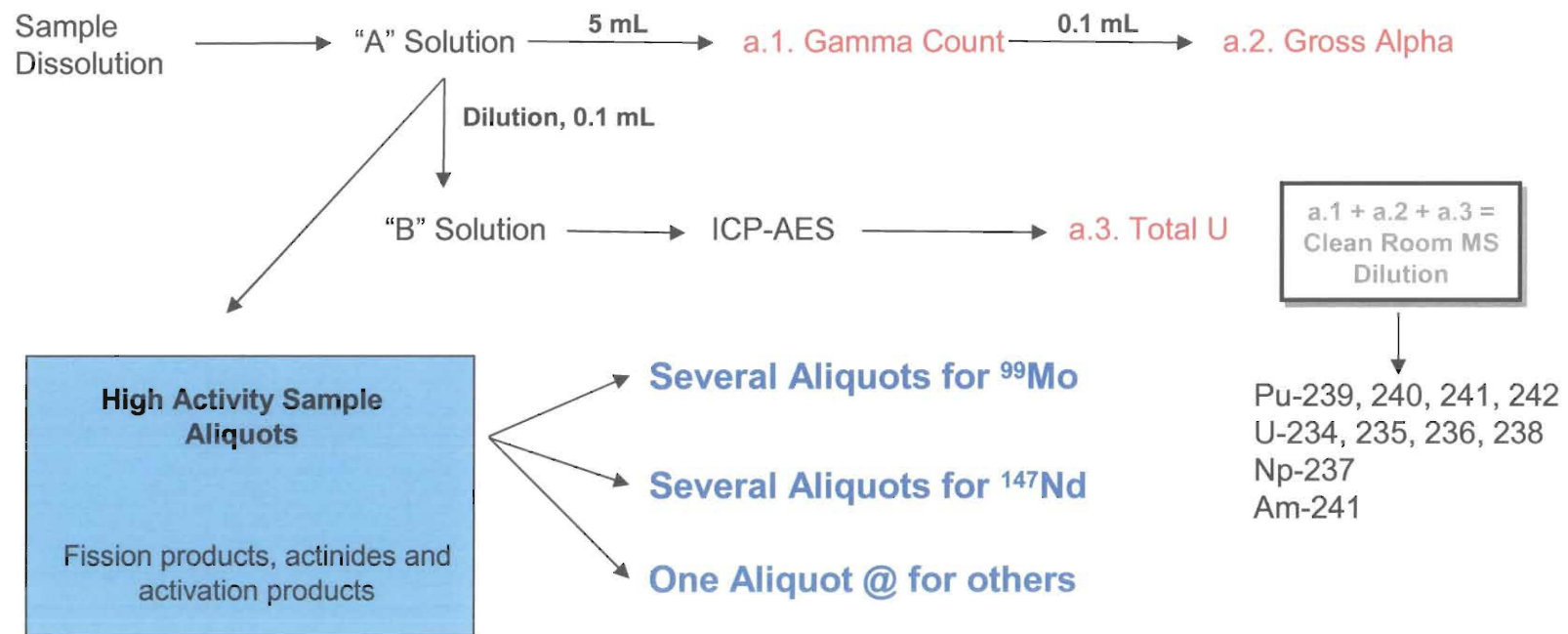
Bugs; Worked Until Stable Scaler Reading Observed

K-Factor Run

Checkout Results Example

	Source	Run	Sample		Ratio	Wts (ug)		Fissile	
			Top	Bottom		Top	Bottom	Top	Bottom
T/B	2354/235u BIG-10	1	2387-1	2387-2	0.9981	1.252	1.124	u	u
		10	2387-1	235-B-1-1	1.0156	1.253	1.225	"	"
		1	"	"	1.0085	"	"	"	"
	239Pu/235u BIG-10	5	2396-1	2387-1	1.1925	1.792	1.253	Pu	u
		6	"	"	1.1912	"	"	"	"
		14	2553	"	1.1985	1.750	"	"	"
		15	"	"	1.1919	"	"	"	"
	238u/235u BIG-10	7	2393-2	2387-1	0.04041	2.511	1.253	28	25
		8	"	"	0.03652	"	"	"	"
		18	2394-2	"	0.03604	98.80	"	"	"
		19	"	"	0.03640	"	"	"	"
		21	2387-1	2394-2	0.03607	1.253	98.80	25	28
		22	"	"	0.03608	"	"	"	"

Standard Aliquot Chemical Separation Scheme

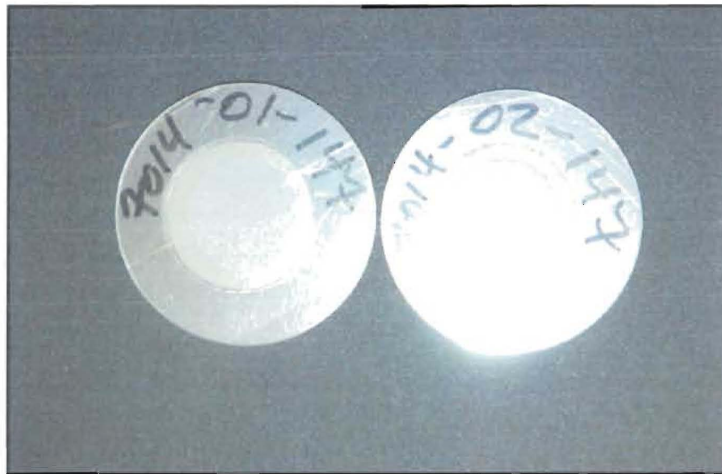


Unclassified

Example Chemical Mount Forms for Beta Counting



^{99}Mo Mounts on Al
Planchettes



^{147}Nd Mounts on Al
Planchettes

Unclassified

“Holy” Counter #6 and Counter #19



Unclassified

Unclassified

Evaluation of Post-ILRR Big Ten Verification Experiments: "Houston, we have a problem"

	Counter # 6	^{235}U	^{238}U	^{239}Pu
	Historic K-factors	2.497E+05	2.427E+05	2.585E+05
		U = fissions/mg; Pu = fissions/alpha		
		^{235}U	^{238}U	^{239}Pu
Experiment #		2547	2571	2554
Target foil areal density (mg/cm ²)		3.9080E+01	1267.1	33.4
Reference foil specific activity, directly measured		3.828E+09	1.319E+08	55.997
LANL historic thermal K-factor evaluation of specific activity		3.908E+09	1.382E+08	60.552
Historic K-factor/reference foil ratio		1.021	1.047	1.081
Original LANL/ILRR comparison (from table 1)		1.021	1.017	1.073

Thermal Series 2700 at OWR: A Hunch Confirmed

	Counter # 19	^{235}U	^{238}U	^{239}Pu	
	Historic K-factors	2.440E+05	2.372E+05	2.526E+05	
		U results are fissions/mg, Pu results are fissions/alpha			
		^{235}U	^{235}U	^{239}Pu	
Experiment #		2725	2755	2726	
Target foil areal density (mg/cm²)		0.4459	0.4004	0.2951	
Reference foil specific activity, directly measured		3.144E+11	3.354E+11	5061	
LANL historic thermal K-factor evaluation of specific activity		3.183E+11	3.403E+11	5414.668	
Historic K-factor/reference foil ratio		1.012	1.015	1.070	

1970 Mo Thermal K-factor Summary Corrected for Self-attenuation

Thermal	Counter	U-235
Modern K-factor	19	2.403E+05
Modern K-factor	6	2.459E+05

Based on the correction for self-attenuation the revised k-factors post 1970 are shown in the table

Barr/Mac Innes Corrected K-factor vs. 2700 Observed

		Counter # 19	²³⁵U
		Modern K-factor	2.403E+05
		U results are fissions/mg	
			²³⁵U
Experiment #		2725	2755
Target foil areal density (mg/cm²)		0.4459	0.4004
Reference foil specific activity, directly measured (contains ~ 0.3% self attenuation correction)		3.144E+11	3.354E+11
LANL <i>modern</i> thermal K- factor evaluation		3.144E+11	3.362E+11
Modern K-factor/reference foil ratio		1.000	1.002

More Comparisons for Selected FPs

2725	Calculated modern	Counter 19 cpm in	Observed	Observed/Expected ratio
	K-factor	target foil	K-factor	
⁹⁹ Mo	2.403E+05	663400	2.403E+05	1.000E+00
¹⁴⁰ Ba	5.509E+05	287560	5.545E+05	1.006E+00
¹⁴⁷ Nd	7.797E+06	20905	7.627E+06	9.782E-01

2755	Calculated modern	cpm in	Observed	Observed/Expected ratio
	K-factor	target foil	K-factor	
⁹⁵ Zr	2.351E+07	8058.6	2.350E+07	9.994E-01
⁹⁹ Mo	2.403E+05	789890	2.397E+05	9.976E-01
¹⁴⁰ Ba	5.509E+05	353960	5.349E+05	9.710E-01
¹⁴⁷ Nd	7.797E+06	24665	7.677E+06	9.846E-01

Fully Modernized Comparison Across Materials and FPs

	²³⁵ U		²³⁸ U		²³⁹ Pu	
	ILRR	LANL	ILRR	LANL	ILRR	LANL
	Q-value	Q-value	Q-value	Q-value	Q-value	Q-value
⁹⁵ Zr	0.9698	0.9916	0.8289	0.7705	0.7088	0.8289
⁹⁹ Mo	0.9996	0.9997	1.0245	1.0546	1.0148	1.0237
¹⁴⁰ Ba	0.9650	0.9442	0.9557	0.9284	0.8516	0.8117
¹⁴⁴ Ce	0.9402				0.6768	
¹⁴⁷ Nd	0.9669	0.9683	1.2351	1.1833	0.9016	0.9270

UNCLASSIFIED

Path Forward - LANL



Completing current work

- **Complete manuscript on LANL 1970s experiment(s)**
 - We are providing you with a current draft of this manuscript
- **Complete assessment of the historical memos**
- **Refine our recommendation for data values (K's, Q's)**
- **Thorough review of thermal irradiation data**

Resurrect older gamma data

- In addition to the LANL irradiations described in the manuscript, additional fission chamber experiments (2 fission, one thermal spectrum) were carried out, but data was not analyzed
- Gamma data associated with these experiments was electronically archived
- We have initiated an effort to “resurrect” this data electronically and to determine if we can associate it with calibration information (e.g. counter efficiencies)
- This would give all parties new data to evaluate in lieu of a new experiment

A new update to ENDF fission yields is due

- Next release is ENDF/B-VII.1
- Evaluation – see Chadwick methodology
- “Mini-CSEWG” meeting planned to help with peer review
- Involve people from the IAEA, JEFF, JENDL
- In addition to examining new evaluation, this the meeting will also examine other issues:
 - If cumulative fission product yields are changed for a few nuclei, how best to ensure consistency with requirements that they still must sum to 200%
 - delayed-neutron data must be consistent
 - etc.

Experimental program recommendations

- We have jointly discussed options for experimental validation approaches at past meetings
- Significant resources would likely be required to reconstitute key capabilities
- Any measurement should be part of a broader program to measure reaction rates in fast critical assemblies
 - Experimental campaign at DAF
 - Mentoring younger scientists in the program
 - Valuable to a broader range of program interests

LANL does not feel that one additional experiment will add to our approach



Another option: Re-establish a comparative calibration basis

Fission product yields can be inferred from data consistent with each laboratory's system of calibration

$$Y_i = Y_i^* \cdot R_i \cdot Q_{99}$$

Y_i^* : "Standard" Thermal yield of fission product i from ^{235}U :
LANL/LLNL ~ agree to within counting uncertainty/laboratory biases

R_i : Relative yield for i , compared to Mo-99 standard in fission spectrum (divided by same in thermal spectrum)
Both laboratories have measured R_i in the past, and these are in reasonable agreement

Use a value for Q_{99} backed by careful, appropriate experiments

