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**MOUND LABORATORY QUARTERLY PROGRESS REPORT:
JULY-SEPTEMBER, 1966**

AEC Research and Development REPORT

MONSANTO RESEARCH CORPORATION

A S U B S I D I A R Y O F M O N S A N T O C O M P A N Y

M O U N D L A B O R A T O R Y

MIAMISBURG, OHIO

OPERATED FOR

UNITED STATES ATOMIC ENERGY COMMISSION

U.S. GOVERNMENT CONTRACT NO. AT-33-1-GEN-53

Printed in the United States of America
Available from
Clearinghouse for Federal Scientific and Technical Information
National Bureau of Standards, U. S. Department of Commerce
Springfield, Virginia 22151
Price: Printed Copy \$3.00; Microfiche \$0.65

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MLM-1388

TID-4500

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Progress Reports

NOV 3.00, MN 65

**MOUND LABORATORY QUARTERLY PROGRESS REPORT:
JULY-SEPTEMBER, 1966**

Date: September 30, 1966

Issued: April 28, 1967

The Mound Laboratory Progress Report, issued quarterly, is intended to be a means of reporting items of current technical interest in research and development programs. To issue this report as soon as possible after the end of the quarter, editorial work is limited; and since this is an informal progress report, the results and data presented are preliminary and subject to change.

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SUMMARY

RADIOELEMENTS

Half-Life Determination by Decaying Pairs Redetermination of the half-life of polonium-210 yielded inconclusive results, and the experiment will be repeated after some modifications are made to the counting equipment. After 281 days and 44 observations, the half-life was found to be 138.519 ± 0.039 days by the decaying pair method, 138.305 ± 0.166 days by both the variational and the general method of computation. It is believed that changes in the ionization gas used and deterioration of the electronic components caused the half-life to deviate from the accepted value. Satisfactory results were obtained in applying the decaying pair method of computation to the decay of 36-min lead-211. (page 7)

Determination of Coincidence Corrections A live-timer system was designed and constructed which permits the direct determination of the resolving time of a proportional counter. (page 14)

Thick Target Yields for the $^{19}\text{F}(\alpha, n) ^{22}\text{Na}$ Reaction Studies were made of several old polonium-fluorine neutron sources to determine the quantities of sodium-22 resulting from the α, n reactions on fluorine. Previous counting data showed that the α, n yield of 5.3 Mev alpha particles on a thick target of fluorine may be much lower than the reported value of 12 neutrons per million alphas. To explain the quantities of sodium-22 that were detected, it is necessary to postulate a yield of approximately three neutrons per million alphas on fluorine. (page 21)

Uranium-234 Mound Laboratory is separating and purifying uranium-234 from aged plutonium-238. By the anion exchange-hexone extraction-anion exchange method, 0.79 g of uranium-234 contaminated with less than four milligrams of plutonium was produced and is presently awaiting the final anion exchange purification step. Part of the raffinates from previous anion exchange purification runs, estimated to contain 0.9 g of uranium-234, were analyzed and processed; 0.1 g of uranium-234 was recovered. The waste solutions contained a negligible amount of uranium-234. Preliminary design and engineering work for a facility using the TOPO (tri-n-octylphosphine oxide) extraction process was begun. (page 23)

Thorium-229 Screening tests of solvent extraction systems continued. The best extractant tested was 0.1M di sec-butyl phenyl phosphonate (DSBPP) dissolved in diethyl benzene (DEB) which quantitatively extracted only the uranium from uranium-thorium mixtures in 1.0, 2.0, and 4.0M nitric acid. The uranium was easily stripped with 0.03M nitric acid. (page 24)

The results were confirmed by DSBPP-DEB extraction of the uranium from 0.5 g of aged uranium-233 dissolved in 2.0M nitric acid. The uranium-thorium ratio was reduced from 2.9×10^4 to 6.5 in six extraction stages. This could be further reduced to less than 0.1 in three more stages. Less than 0.1% of the thorium was extracted. Solvent extraction methods are being investigated for the purification of the thorium product, which still contains metallic ions other than uranium.

ANALYTICAL AND INSTRUMENTATION

Neutron Activation Analysis A neutron activation method for determining total boron and isotopic ratio of elemental boron is being developed. The boron-11 is determined by irradiating a sample and standard in a fast neutron flux of $\sim 1 \times 10^9$ n/cm²/sec. The nuclear reaction which occurs is $^{11}\text{B}(n,p)^{11}\text{Be}$. Beryllium-11 has a half-life of 13.6 sec and emits a 2.12 Mev gamma. (page 30)

The boron-10 is determined by neutron absorption using a plutonium-beryllium neutron source. A solution of the sample is put into a doughnut-shaped container and placed in a paraffin-filled drum with a helium-3 filled neutron detector through the center of the doughnut-shaped container. The source is placed so the neutrons pass through the sample to the detector. Boron-10 absorbs neutrons, thus the greater the boron-10 concentration, the lower the count rate.

Nuclear Magnetic Resonance The electron inductive effect of substituents in polystyrene is being studied by nuclear magnetic resonance spectroscopy to discover a correlation between the ease of polymerization of substituted polystyrenes and the substituent. The NMR spectra of polystyrene, p-methylstyrene, p-methoxystyrene, m-chlorostyrene, m-nitrostyrene, and p-nitrostyrene revealed that para-substitution has a greater inductive effect than meta-substitution and that the effect of substituents in the polystyrene ring is to decrease the electron density about the vinyl group making the polystyrene molecule more vulnerable to nucleophilic attack by basic reagents. (page 31)

Stilbene Crystal, Fast Neutron Spectrometer The stilbene crystal, fast neutron spectrometer is being used for routine measurement of the neutron energy spectra of a variety of sources. Refinements to the method are being incorporated by allowing for variation in light output of the crystal as a function of direction of incidence of the neutron beam on the crystal. Although this is a small correction, it should result in significant improvement in measured spectra. (page 31)

RADIOELEMENTS

HALF-LIFE DETERMINATION BY DECAYING PAIRS

The half-life of polonium-210 (138.3763 ± 0.0017 days)¹ is being redetermined by alpha-counting, primarily to test the decaying pair method of computation² and to compare it with the variational and general methods.³ The progress of the experiment during the first 230 days (38 observations) was summarized in the last quarterly report.⁴ After 281 days and 44 observations, the half-life of polonium-210 was found to be 138.519 ± 0.039 by the decaying pair method, 138.305 ± 0.166 days by the variational method, and 138.305 ± 0.166 days by the general method. Details of the analyses are given in Tables 1 through 6.

The experiment was terminated with inconclusive results; although the decaying pair method yielded a rather small probable error in the weighted average, the absolute error (based on the calorimetric value¹) was nearly four times as large. The general method, on the other hand, yielded a small absolute error but a large probable error (Table 1). The data were broken down into segments of 20 observations each in an effort to determine if a constant source of bias existed (Table 3). These results were also inconclusive, as were analyses with and without weighting, with and without Chauvenet's criterion, and with and without a normalizing standard.

During the 281 days of the experiment, three different tanks of P-10 gas* were used, and while there was apparently no significant difference in the composition of the gas (as determined by mass spectrometry), a progressive increase was noted in the counting rate of the plutonium-239 source used as a standard. This increase was slight (of the order of 0.5% overall), and may represent the decay of a small amount of americium-241. However, it may also be indicative of a progressive change in instrument response due to the change of ionization gas or to aging of

*Commercial mixture of 90% Ar + 10% CH₄

¹Mound Laboratory Progress Report for July, 1964, MLM-1209 (July 20, 1964) p. 12.

²Mound Laboratory Progress Report for April, 1965, MLM-1254 (April 30, 1965) p. 7.

³H. W. Kirby and J. Z. Braun, Determination of Coincidence Corrections in a Proportional Counter, III - Least Squares Analysis by the Variational and General Methods, MLM-1263 (January, 1966).

⁴Mound Laboratory Quarterly Progress Report: April-June, 1966, MLM-1342 (June 30, 1966) p. 9.

Table 1

HALF-LIFE OF POLONIUM-210 AS COMPUTED BY
THE DECAYING PAIR AND GENERAL METHODS

Number of Observations	Time Span (days)	Decaying Pair ^a		General ^b	
		$T_{1/2} \pm \text{P.E.}$ (days)	Absolute Error (days)	$T_{1/2} \pm \text{P.E.}$ (days)	Absolute Error (days)
39	233	138.517 $\pm$ 0.037	+ 0.141	138.256 $\pm$ 0.170	- 0.120
40	246	138.504 $\pm$ 0.036	+ 0.128	138.239 $\pm$ 0.160	- 0.137
41	254	138.519 $\pm$ 0.034	+ 0.143	138.369 $\pm$ 0.161	- 0.007
42	260	138.516 $\pm$ 0.033	+ 0.140	138.315 $\pm$ 0.149	- 0.061
43	268	138.534 $\pm$ 0.034	+ 0.158	138.295 $\pm$ 0.165	- 0.081
44	281	138.519 $\pm$ 0.039	+ 0.143	138.305 $\pm$ 0.166	- 0.071

^a Weighted average of six pairs

^b Weighted average of four samples

Table 2

HALF-LIFE OF POLONIUM-210 BY THE DECAYING PAIR METHOD
(44 Observations - 281 Days)

Pair	Ratio	Half-Life (days)	Probable Error (days)	Absolute Error (days)
1-2	0.618	138.436	$\pm$ 0.103	+ 0.060
1-3	0.387	138.549	$\pm$ 0.068	+ 0.172
1-4	0.261	138.499	$\pm$ 0.055	+ 0.123
2-3	0.627	138.738	$\pm$ 0.116	+ 0.361
2-4	0.422	138.568	$\pm$ 0.063	+ 0.192
3-4	0.673	138.256	$\pm$ 0.113	- 0.120
Weighted Average		138.519	$\pm$ 0.039 (External) + 0.143 $\pm$ 0.031 (Internal)	

Table 3

HALF-LIFE OF POLONIUM-210 BY THE DECAYING PAIR METHOD
(Analyzed in Segments of 20 Observations)

<u>Observation Numbers</u>	<u>Days</u>	<u>Time Span</u>	<u>Weighted Average Half-Life (days)</u>
1 - 20	0 - 94	94	138.283 $\pm$ 0.207
5 - 24	15 - 122	107	138.212 $\pm$ 0.097
10 - 29	34 - 162	128	138.619 $\pm$ 0.092
15 - 34	63 - 202	139	138.678 $\pm$ 0.132
20 - 39	94 - 233	139	138.496 $\pm$ 0.086
21 - 40	104 - 246	142	138.382 $\pm$ 0.085
22 - 41	111 - 254	143	138.508 $\pm$ 0.128
23 - 42	115 - 260	145	138.551 $\pm$ 0.135
24 - 43	122 - 268	146	138.573 $\pm$ 0.128
25 - 44	129 - 281	152	138.416 $\pm$ 0.155

Table 4

HALF-LIFE OF POLONIUM-210 AND COUNTER RESOLUTION TIME BY
THE GENERAL METHOD
(44 Observations - 281 Days)

<u>Sample</u>	<u>Half-Life (days)</u>	<u>Probable Error (days)</u>	<u>Absolute Error (days)</u>	<u>Resolution Time (min $\times 10^7$)</u>
1	138.21	$\pm$ 0.25	- 0.16	1.17 $\pm$ 0.20
2	139.00	$\pm$ 0.22	+ 0.63	0.69 $\pm$ 0.11
3	137.84	$\pm$ 0.20	- 0.54	1.12 $\pm$ 0.06
4	138.25	$\pm$ 0.20	- 0.13	0.98 $\pm$ 0.04
Weighted Average	138.30	$\pm$ 0.17	- 0.08	1.00 $\pm$ 0.05

the electronic components. It is noteworthy that, if the half-life had been computed in the usual way (assuming no error in the resolution time), a quite satisfactory solution would have been obtained (See Tables 5 and 6).

After some modifications are made to the counting equipment, the experiment will be repeated and an effort will be made to control (or, at least, to identify) the various suspected sources of experimental bias.

Although the method of decaying pairs, as originally derived, is based on the counting of two separate sources of the same radioelement, it can be applied to a single short-lived source if successive observations are made at equally spaced intervals. This requirement can be met by operating a multi-channel analyzer as a multi-scaler. Counts are accumulated for a fixed period in a single channel; then, upon receipt of a channel advance signal from a crystal-controlled Time Base Generator, the counts are accumulated in the next channel, and so on. If the data stored in the odd- and even-numbered channels are separated into two groups, they may be treated as though they had originated from two different sources of the same radioelement, counted alternately by the same detector. As in the case of two physically different sources, the requirement that they be counted "simultaneously" is met by adjusting the starting time (T_0) of one of the series of observations.

In actual practice, the computation of a half-life from a single source by the decaying pair method has considerably more versatility than when physically different sources are used, since the relative magnitudes of the sources to be paired can be selected almost arbitrarily by the simple expedient of selecting various values for the difference in T_0 (ΔT). The number of such pairings is limited only by the number of observations.

Thus, for example, in one experiment with 36.1-min lead-211, the multi-scaler accumulated alpha counts for 50.00 sec in each of 332 channels, resulting in 166 pairs of observations. To analyze the data, the minimum value of ΔT was set at 550 sec (eliminating the first five even-numbered observations) and the maximum ΔT was set at 10,000 sec, or approximately 4.6 half-lives, corresponding to a pair ratio of 0.04. The half-life was computed for every possible value of ΔT between those limits, yielding a total of 95 solutions. The probable errors of the individually computed half-lives were obtained by the usual methods,⁵ and a weighted average was found, with the weights set equal to the squared reciprocals of the probable errors. However, since the data were obtained from a single source, rather than from 95 independent sources, the probable error of the mean was computed from the expression:

$$P.E. = 0.6745 \sqrt{\sum \frac{(\bar{X} - \bar{X})^2}{n}}$$

⁵R. T. Birge, Phys. Rev., 40, 207 (1932).

Table 5

COUNTER RESOLUTION TIME BY THE KNOWN HALF-LIFE METHOD^a
(44 Observations - 281 Days)

<u>Sample Number</u>	<u>Resolution Time (min x 10⁷)</u>	<u>True Initial Counting Rate (cpm)</u>
1	1.047 ± 0.034	151,248 ± 56
2	1.015 ± 0.019	244,797 ± 82
3	0.960 ± 0.011	390,320 ± 121
4	0.968 ± 0.007	579,628 ± 175

Weighted

Average 0.9727 ± 0.0075 (External)
 ± 0.0057 (Internal)

^a Based on $T_{1/2} = 138.3763$ days

Table 6

HALF-LIFE OF POLONIUM-210 ASSUMING RESOLUTION TIME (τ)
IS EXACTLY 1.000×10^{-7} MINUTE
(44 Observations - 281 Days)

<u>Sample Number</u>	<u>Half-Life (days)</u>	<u>True Initial Counting Rate (cpm)</u>
1	138.428 ± 0.041	151,148 ± 26
2	138.424 ± 0.038	244,697 ± 39
3	138.237 ± 0.034	390,910 ± 57
4	138.227 ± 0.034	580,635 ± 85

Weighted

Average 138.315 ± 0.037 (External)
 ± 0.018 (Internal)

Figure 1. Half-Life of Lead-211 by the Decaying Pair Method as a Function of the Selected ΔT .

Table 7

HALF-LIFE OF LEAD-211 AS A FUNCTION OF COUNTING RATE LIMIT

Maximum Counting Rate (cps)	By Decaying Pairs		By General Method	
	Half-Life at $\Delta T = 4350$ (sec)	Weighted Average Half-Life (sec)	Half-Life (sec)	Resolution Time (μ sec)
13,515	2168.77 $\pm$ 0.85	2168.55 $\pm$ 0.61	2167.62 $\pm$ 0.49	20.09 $\pm$ 0.04
12,202	2169.58 $\pm$ 1.01	2169.48 $\pm$ 0.72	2168.58 $\pm$ 0.53	19.94 $\pm$ 0.05
10,782	2170.62 $\pm$ 1.12	2169.82 $\pm$ 0.68	2169.01 $\pm$ 0.58	19.87 $\pm$ 0.07
9,471	2170.62 $\pm$ 1.26	2170.17 $\pm$ 0.66	2169.43 $\pm$ 0.64	19.78 $\pm$ 0.09
8,344	2170.10 $\pm$ 1.40	2170.41 $\pm$ 0.73	2169.60 $\pm$ 0.72	19.75 $\pm$ 0.11
7,311	2170.64 $\pm$ 1.60	2170.56 $\pm$ 0.95	2170.07 $\pm$ 0.80	19.63 $\pm$ 0.14
6,335	2171.72 $\pm$ 1.81	2170.66 $\pm$ 1.81	2169.82 $\pm$ 0.89	19.69 $\pm$ 0.18
5,536	2172.21 $\pm$ 2.07	2170.87 $\pm$ 1.21	2170.35 $\pm$ 0.98	19.53 $\pm$ 0.23
4,813	2169.15 $\pm$ 2.06	2169.41 $\pm$ 1.24	2169.52 $\pm$ 1.09	19.83 $\pm$ 0.29
4,139	2169.88 $\pm$ 2.63	2171.03 $\pm$ 1.68	2170.25 $\pm$ 1.22	19.53 $\pm$ 0.37
3,566	2171.90 $\pm$ 3.07	2171.22 $\pm$ 2.17	2170.51 $\pm$ 1.38	19.40 $\pm$ 0.48
3,041	2170.55 $\pm$ 3.63	2170.97 $\pm$ 2.62		
2,672	2172.31 $\pm$ 3.33	2169.22 $\pm$ 3.00		
2,282	2169.92 $\pm$ 3.74	2169.96 $\pm$ 6.68		
1,950	2172.58 $\pm$ 4.54	2170.33 $\pm$ 7.64		
1,664	2178.72 $\pm$ 7.56	2172.84 $\pm$ 5.32		

which allows for only one degree of freedom.⁶ The weighted average was found to be 2168.55 $\pm$ 0.61 sec or 36.14 $\pm$ 0.01 min. The results of these computations are shown in Figure 1, where it can be seen that all of the individual values fall within less than two (and usually less than one) probable error of the weighted average. The computed half-lives tend to be low for ΔT less than one half-life and high for ΔT more than four half-lives. What is not immediately apparent from Figure 1 is that the probable errors pass through a minimum value, the smallest probable error occurring at ΔT equal to 3750 sec. The significance of this trend is not clear and will be the subject of further investigation.

The decaying pair method of computation is relatively insensitive to the absolute magnitudes of the counting rates used. Table 7 gives the half-life and probable error computed for each of several maximum counting rate levels at an arbitrarily selected ΔT of 4350 sec and the weighted average of all values computed with the same counting limit. Neither list shows a significant trend in the half-life, although the probable error increases as the number of data points is reduced. For the purpose of comparison, the results of treating the same data by the general method³ are also shown in Table 7.

⁶Mound Laboratory Progress Report for April, 1965, MLM-1254 (April 30, 1965) p. 7.

Although the inherent dead-time of the multichannel analyzer, operating in the time mode, is only $11 \pm 1 \mu\text{sec}$, the resolution time computed by the general method reflects the fact that the pulse counted by the multiscaler was generated by the variable monostable of the live time system described in the following section (Cf. Determination of Coincidence Corrections). Since the monostable was preset for a dead time of $20 \mu\text{sec}$, the values computed by the general method are in good agreement with the value determined directly by the live time system. (H. E. Kirby)

DETERMINATION OF COINCIDENCE CORRECTIONS

The dead-time of a multichannel pulse height analyzer depends upon the number of the channel in which the incoming information is stored. Typically, it is of the order of $10 \mu\text{sec}$ plus $0.5 \mu\text{sec}$ per channel. Since correction of results for counts lost during the dead-time of the instrument would involve rather complicated mathematical treatment, this correction is usually made automatically by the incorporation of "live time" circuitry.

A pulse train, generated by an accurately calibrated quartz crystal, is gated by the analyzer so that the pulses are counted in a scaler only during the period when the analyzer is "live"; that is, ready to accept pulses from a radiation detector. During the time that the analyzer is engaged in receiving, examining, and storing information, the timing pulses are diverted, usually to a dead-time meter, and are not counted by the timing scaler. Thus, the total count accumulated in the timing scaler represents only that portion of the real time during which the analyzer was available for receiving incoming pulses. The live time may be as little as 10% of the real time, although the live timers incorporated in most analyzers are inaccurate when the dead time exceeds 50-75%.

While live timers are standard equipment in multichannel analyzers, they are not commercially available for routine use with single-channel analyzers or with G-M and proportional counters. Dead-time corrections for these instruments can usually be made rather easily (if the resolving time is known) by application of the Ruark and Brammer equation:⁷

$$N = \frac{n}{1 - n \tau}$$

where:

$\underline{N}$ = corrected count

$\underline{n}$ = observed count

τ = resolving time found independently by any of a number of methods.^{8, 9}

⁷A. E. Ruark and F. E. Brammer, Phys. Rev. 52, 322 (1937)

⁸H. W. Kirby, Determination of Coincidence Corrections in a Proportional Counter, I - Double Source Methods, MLM-1197 (November, 1964).

⁹H. W. Kirby and J. Z. Braun, Determination of Coincidence Corrections in a Proportional Counter, II - The Known Half-Life Method, MLM-1202 (November, 1964)

In a continuing study of the problem of precisely determining τ , a live timer system was designed and constructed which makes possible a direct determination of the value of N , from which the value of τ can be computed. Thus far, the system has been used only to count pulses originating in a Nuclear Measurements Corp. PC-3A proportional counter, but it is believed capable of operating from the output of G-M tubes and scintillation counters as well.

The live timer system is shown in block diagram form in Figure 2. The system consists of an accurately calibrated and thermostated 100 mcs quartz crystal oscillator, four scalers, and two gates. The oscillator pulse train is fed simultaneously to a real time scaler and to a time gate, which permits the timing pulses to be counted in a live time scaler only while the count gate is "live".

The radiation source is counted by a conventional detector-preamplifier which feeds pulses to an amplifier-discriminator, and these pulses are counted directly in a pulse scaler and simultaneously fed to the count and time gates. Triggering of the gates by a discriminator pulse causes a count to be recorded in the gate pulse scaler and switches the oscillator pulse train to a dead time scaler (not shown in Figure 2). After a pre-set time, which may be varied from one microsecond to two milliseconds, the gates are again open, and the live time scaler resumes counting the oscillator pulses until another discriminator pulse triggers the count gate.

Figure 2. Block Diagram of Live Timer System

Table 8

COUNTING OF LEAD-211 IN REAL AND LIVE TIME

Time from T_0 (min)	ETC/ETS ^a (cps)	LTC/LTS ^b (cps)	LTC/ETS ^c (cps)	τ_1 ^d (μ sec)	τ_2 ^e (μ sec)
1	19257.20	21666.22	15136.02	5.77	19.91
10	17016.10	18870.50	13715.08	5.78	19.92
20	14269.62	15571.25	11880.86	5.86	19.95
30	11961.34	12854.42	10232.12	5.81	19.94
40	9986.38	10611.22	8758.50	5.90	19.93
50	8345.48	8773.64	7468.12	5.85	19.93
60	6930.40	7227.73	6317.04	5.94	19.95
70	5752.00	5951.83	5320.94	5.84	19.92
80	4783.70	4926.58	4486.14	6.06	19.93
90	3954.96	4050.91	3747.90	5.99	19.96
100	3283.08	3352.11	3141.60	6.27	19.99
110	2703.70	2748.69	2605.76	6.05	19.96
120	2242.58	2275.85	2177.08	6.52	19.93
130	1861.60	1883.13	1814.96	6.14	19.95
140	1536.98	1552.24	1505.36	6.40	20.06
150	1272.14	1281.15	1249.12	5.53	20.01
160	1053.34	1060.06	1038.22	6.02	19.84
170	873.38	878.25	863.14	6.34	19.93
180	719.56	723.29	713.02	7.17	19.92

^a ETC/ETS - Elapsed time counts/elapsed time sec

^b LTC/LTS - Live time (Gate pulse) counts/live time sec

^c LTC/ETS - Gate pulses/elapsed time sec

^d τ_1 - (ETS/ETC) - (LTS/LTC); Conventional counter resolution time

^e τ_2 - (ETS/LTC) - (LTS/LTC); gate resolution time

The four scalers yield the following information:

- Scaler 1 - conventional (elapsed time) counts
- Scaler 2 - Live (gate pulse) counts
- Scaler 3 - Live time in 0.01 sec
- Scaler 4 - Elapsed time in 0.01 sec

The conventional counting rate (n) is determined from scalers 1 and 4; the live time counting rate (N) is determined from scalers 2 and 3. The dead time (τ_d) of the gate can be observed on an oscilloscope, but it can be computed more precisely from the counts recorded in scalers 2, 3, and 4.

The system was automated by using preset printing scalers, a printing programmer, and a paper tape printer. In addition, to control the time lost in printing out information, a fifth scaler, also driven by the crystal oscillator, can be preset for a selected off-time. Thus, when a half-life experiment is in progress, the time interval between counts is the sum of the two times preset in the off-time scaler and the elapsed (real) time scaler.

To test the system, a source of 36.1-min lead-211 was counted in real and live time. Counts were obtained at 60-sec intervals, with the off-timer preset at 10.00 sec and the elapsed timer preset at 50.00 sec. The gate was adjusted for a dead-time of 20 μ sec, as determined by an oscilloscope trace and by counting a standard plutonium-239 source. A total of 250 observations was recorded, of which a sampling is given in Table 8.

The most striking feature of the results was the relative reproducibility of the gate dead-time (τ_d) over the entire counting range, as compared with the trend towards higher resolution times (τ_d) in the conventional proportional counter as the counting rate declined. The explanation for this difference is probably to be found in the shape of the pulse being counted in the two systems. In Figure 3, it is seen that the output pulse from the one-shot multivibrator has a long recovery time, during which occasional pulses from the detector-preamplifier may be sufficiently high to trigger a new pulse, thereby decreasing the apparent resolution time. The probability of such pulses occurring would be greater at higher counting rates. The preamplifier pulse (Figure 4) exhibits an opposite effect; that is, the dead-time tends to be high at high counting rates and to decrease as the counting rate becomes lower. In this case, occasional pulses from the detector, which are too weak to trigger the preamplifier, may be large enough to extend its recovery time.

The gate pulse has a sharply defined square wave form whose rise and decay times are of the order of 0.1 μ sec, essentially eliminating the possibility that the gate can be triggered by a new pulse or its dead time extended before the circuit has completely recovered from the preceding pulse.

Figure 3. One-Shot Multivibrator Output Wave Form

Figure 4. Preamplifier Pulse Shape

In the light of these findings, it should be possible to develop a circuit which, at relatively little additional cost, could be interposed between the amplification and scaling systems of a proportional counter (or other radiation pulse counters) to extend its useful counting range. Such a circuit is under development and will probably be available for testing in the near future.

The data of Table 8 were plotted to show the effect of counting a decaying source in real and live time (Figure 5). After the first five minutes (to allow for the growth of bismuth-211), the live time counting rate falls on a straight semi-logarithmic line, while the uncorrected counting rates in elapsed time approach the same line asymptotically. The three decay curves become congruent only after approximately three hours, or about five half-lives of lead-211.

Another test of a live timer system is to count paired sources separately and together and to compare the sum of the individual counting rates with the counting rate of the combined sources. The results of such a test with plutonium-239 sources are given in Table 9. All sources were counted for a total of either 1×10^6 or 5×10^5 present gate pulses; since the timing scalars record the time in units of 0.01 sec, count rates from combined sources tend to be slightly higher than the sums of those from the individual sources. Nevertheless, the agreement between the summed and combined counting rates is excellent, indicating that the live timer is performing the function for which it was designed. (H. W. Kirby)

Figure 5. Decay of Lead-211 in Real and Live Time

Table 9
LIVE TIME COUNTING RATES OF PAIRED SOURCES

Source Number		Counting Rates (cps)				Ratio of Counting Rates (Sum to Combined)
		A	B	Sum A + B	Combined A & B	
1	2	4919.81	4901.48	9821.29	9832.84	0.9988
3	4	4951.97	4891.89	9843.86	9889.24	0.9988
1	5	4919.81	10768.90	15688.71	15693.66	0.9997
1	6	4919.81	14196.48	19116.29	19120.46	0.9998
5	6	10768.90	14196.48	24965.38	24987.51	0.9991
6	7	14196.48	14501.16	28697.64	28735.63	0.9987
1	9	4911.83	13616.54	18528.28	18563.19	0.9981
11 ^a	9	8725.99	13616.54	22342.53	22326.39	1.0007
5	9	10716.95	13616.54	24333.49	24360.51	0.9989
6	9	14150.26	13616.54	27766.80	27777.75	0.9996
10	9	15959.3	13616.54	29575.67	29620.82	0.9985

^a Over-sized plate; off-centered.

THICK TARGET YIELD

Thick Target Yield for the $^{19}\text{F}(\alpha, n)^{22}\text{Na}$ Reaction

A search for polonium-208 and polonium-209 in neutron and alpha sources prepared at Mound Laboratory many years previously, yielded three neutron sources that exhibited the characteristic gamma-ray spectrum of sodium-22. The records maintained for all neutron sources showed that the three sources were fabricated with polonium-210 and various fluorine-containing compounds. Preliminary calculations indicated that the neutron yield for the $^{19}\text{F}(\alpha, n)^{22}\text{Na}$ reaction was considerably below the value given by Roberts¹⁰, 12×10^{-6} neutrons per alpha particle (n/α).

The sodium-22 activity of a neutron source as a function of time can be expressed as:

$$\frac{dN_{22}}{dt} = Y I_0 e^{-\lambda_1 t} - \lambda_{22} N_{22} \quad (1)$$

where Y = $^{19}\text{F}(\alpha, n)^{22}\text{Na}$ thick target yield.
 I_0 = Initial alpha flux per unit time.
 N_{22} = Number of atoms of ^{22}Na at any time, t .
 λ_1 = ^{210}Po decay constant.
 λ_{22} = ^{22}Na decay constant.

This equation is similar to the Bateman equations, and the solution is:

$$N_{22} = \frac{Y I_0}{\lambda_{22} - \lambda_1} (e^{-\lambda_1 t} - e^{-\lambda_{22} t}) \quad (2)$$

Equation (2) gives the number of atoms of sodium-22 in the source as a function of time. All the variables are known except the yield, Y , and the number of atoms of sodium-22, N_{22} . The number of atoms of sodium-22 can be determined by comparing the gamma-ray counting rates of the sources with a standard source of sodium-22.

The gamma-ray spectra of the three neutron sources were somewhat different from that of the sodium-22 standard, because the neutron sources were enclosed in rather heavy metal containers. The counting errors involved, however, are probably less than 20%. The ages of the sources, t , and the initial polonium-210 alpha activity values, I_0 , were obtained from the records. The yield values obtained from Equation (2) were corrected for the composition of the fluorine-containing thick-target, and the stopping powers of the various constituents. The target material consisted of a 24:1 mole ratio mixture of $\text{Na}_2\text{BF}_4/\text{NaBeF}_4$. Roughly 70% of the alpha particles emitted were stopped in fluorine nuclei. The data on these three sources are given in Table 10; the average yield for these sources is $2.48 \times 10^{-6} n/\alpha$.

¹⁰James H. Roberts, U.S.A.E.C. Unclassified Report MDDC-731 (1947)

Table 10

DATA ON POLONIUM-210 - FLUORINE -19 SOURCES USED TO COMPUTE THE
THICK TARGET YIELD FOR THE $^{19}\text{F}(\alpha, n)$ REACTION

<u>Source</u>	<u>Initial ^{210}Po Content (curies)</u>	<u>t (days)</u>	<u>^{22}Na activity (microcuries)</u>	<u>Yield, Y (n/$10^6\alpha$)</u>
JR-9	35.54	4124	0.570	2.81
PN594E	28.75	1463	3.14	2.68
PN594A	31.16	1495	2.93	1.95

The disparity between the present results and the value reported by Roberts prompted a search for additional information on the fluorine-19 reaction. In 1959, Geiger¹¹ determined the neutron flux of a source (prepared at Mound Laboratory) containing 1.6 curies of polonium-210 mixed with one gram of calcium fluoride powder, using a technique similar to the one presented here; Geiger's results can be converted to a yield of 1.81×10^{-6} n/ α . In 1955, Breen and Hertz¹² measured the neutron flux of a source which was similar to Geiger's source. Their value is equivalent to 2.83×10^{-6} n/ α . These data of Geiger and Breen and Hertz were not corrected for thick target composition. This correction may increase the yield by about a factor of two.

There is still some uncertainty concerning the distribution of the polonium-210 within the fluorine-containing targets. In most cases, the sources were heated above the volatility point of polonium metal, so that the polonium vapor might diffuse through the target material. Rapid cooling might cause the metal to deposit preferentially on the inside walls of the source container, so that a significant fraction of the emitted alpha particles would not enter the target material at all. However, if this factor is ignored, it seems clear from the data presented here that the thick-target yield of the fluorine-19(α, n)sodium-22 reaction is much smaller than the often-quoted value of Roberts, and is probably close to 3×10^{-6} n/ α . (R. C. Lange)

¹¹K. W. Geiger, Can. J. Phys. 37, 550, (1959)

¹²R. J. Breen and M. R. Hertz, Phys. Rev. 98, 599, (1955)

URANIUM-234

Mound Laboratory is separating and purifying uranium-234 from aged plutonium-238. A separation and purification process consisting of an initial separation by anion exchange in nitric acid media, hexone extraction to further purify the uranium, and a final anion exchange in both nitric and hydrochloric acid media to remove the traces of impurities, was developed previously.¹³ A second process using TOPO (tri-n-octylphosphine oxide) extraction to recover and concentrate both uranium and plutonium from large volume, low level ion exchange raffinate solutions was also developed previously.¹⁴

Approximately 3.5 liters of solution containing one gram of plutonium and 0.25 g of uranium were extracted into hexone from an aluminum nitrate salted medium. The loaded organic phase was stripped with 1.5 liters of 0.35N nitric acid, and the strip solution evaporated to 100 ml before analysis. Analyses indicated that the strip solution contained 17 mg of plutonium and 140 mg of uranium. Additional plutonium decontamination was obtained by adjusting this solution to one normal nitric acid and contacting with a 0.5M thenoyltrifluoroacetone (TTA) xylene solution. Alpha counting indicated less than two milligrams of plutonium remained.

Two additional batches of uranium-234 from previous hexone extractions containing a total of 0.65 g of uranium-234 were also extracted with TTA, and the plutonium contamination reduced to approximately one milligram in each batch. These three batches, containing 790 mg of uranium, are presently being prepared for the final anion exchange purification step.

Raffinates from previous final anion exchange purification runs are being analyzed and the uranium-234 recovered. These solutions, consisting of nine normal hydrochloric acid or 1.6M aluminum nitrate or mixtures of the two, were analyzed by evaporating a small aliquot to dryness to remove the chloride ion, then dissolving in one normal nitric acid and diluting 10-100 fold. The uranium-234 content was determined by gross alpha counting and alpha pulse height analysis. To date, over 500 ml of raffinate solution have been processed and discarded, and 0.1 g of uranium-234 recovered. Reprocessing of additional raffinate solutions is expected to yield an additional 0.8 g of uranium-234.

Preliminary design and engineering work for a facility using the TOPO extraction process was begun. This facility will be capable of processing large volumes of low-level, ion exchange raffinates from plutonium-238 recovery operations, and will recover and concentrate the uranium-234 for further processing in laboratory scale equipment. (P. E. Figgins)

¹³P. E. Figgins and R. J. Belardinelli, J. Inorg. Nucl. Chem. 28, 2193 (1966)

¹⁴Mound Laboratory Quarterly Progress Report: Jan-March, 1966, MLM-1325 (March 31, 1966)

THORIUM-229

Solvent Extraction Screening tests of uranium-thorium separation using thorium-230 and uranium-233 tracers were continued; the method was described previously.¹⁵ The present results using a nitric acid feed solution containing 25 g/liter uranium and 12.5 g/liter thorium are given in Table 11.

Table 11

SOLVENT EXTRACTION SCREENING TESTS

<u>HNO₃</u> <u>(N)</u>	<u>Extractant^a</u>	<u>Remarks</u>
2	DSBPP ^b	Complete extraction of uranium. No detectable thorium extracted.
4	DSBPP	Complete extraction of uranium. No detectable thorium extracted.
1	Alamine-336 ^c	No uranium or thorium extracted. (Increased acid to be investigated.)
2	TIOA ^d	Some differential extraction between uranium and thorium. Better results at 1 <u>N</u> HNO ₃ . (See Reference 16)

^a Extractant was 10 Volume Percent in DEB (Diethyl Benzene)

^b Di-Secondary Butyl Phenyl Phosphonate

^c A Mixed Tertiary Amine

^d Tri Iso-Octyl Amine

DSBPP-DEB looked promising and another run was made to test the DSBPP-DEB extraction method on a sample of uranium-233 nitrate plus daughters. Approximately 0.5 g of aged uranium-233 nitrate, containing an unknown amount of uranium-232, was dissolved in 10 ml of 2M HNO₃. This aqueous phase was extracted successively with six 10 ml portions of DSBPP-DEB organic by the following procedure:

¹⁵ Mound Laboratory Quarterly Progress Report: April-June, 1966, MLM-1342
(June 30, 1966) p. 32.

The 10 ml aqueous phase plus 10 ml of 10% DSBPP-DEB were placed in a 60 ml separatory funnel and shaken for two minutes. After separation, the two phases were withdrawn into separate polyethylene bottles. Triplicate one lambda alpha slides were mounted from each phase; the aqueous phase was returned to the funnel, fresh organic added, and the procedure repeated. After six extractions, it was evident from gross alpha counts that the activity of the aqueous phase had become constant and a negligible amount of radioactive material was being extracted into the organic phase. The experiment was discontinued at this point.

Two alpha pulse height curves were run on one of the planchets from each sampling; these covered alpha energy ranges of 4-9 MeV and 4.3-5.8 MeV. The latter range allowed close examination of the uranium and thorium peaks. At the same time, data for each channel were printed out and an oscilloscope photograph was obtained.

The gross alpha counts are given in Table 12. Material balances were calculated for each extraction and are shown in Table 13.

Table 12

ALPHA COUNTING RESULTS

Remarks	Aqueous Phase		Organic Phase	
	Sample Number	Alpha cpm ^a	Sample Number	Alpha cpm ^a
U-233 Mother		5.34 x 10 ⁵		
After 1st. Ext.	QA-1	2.90 x 10 ⁵	QO-1	2.84 x 10 ⁵
After 2nd. Ext.	QA-2	1.37 x 10 ⁵	QO-2	1.61 x 10 ⁵
After 3rd. Ext.	QA-3	8.40 x 10 ⁴	QO-3	5.29 x 10 ⁴
After 4th. Ext.	QA-4	7.30 x 10 ⁴	QO-4	1.02 x 10 ⁴
After 5th. Ext.	QA-5	7.21 x 10 ⁴	QO-5	2.35 x 10 ³
After 6th. Ext.	QA-6	7.40 x 10 ⁴	QO-6	4.3 x 10 ²

^aCounts shown are the average from three one-lambda slides

For the material balance the sum of QA-1 and QO-1 was used rather than the mother solution count because the extremely high count of the mother solution resulted in a high coincidence and, therefore, a low result.

Several quantitative deductions can be made. First, the thorium fraction constitutes

$$\frac{73}{574} \times 100 = 12.7\% \text{ of the total alpha count.}$$

Table 13

MATERIAL BALANCE

Sample Number (QA-n + QO-n)	Sum of Alpha cpm	QA-(n-1) Sample No.	Alpha cpm	Percent Difference
QA-1 + QO-1	5.74×10^5	Mother soln.	5.34×10^5	7.2
QA-2 + QO-2	2.98×10^5	QA-1	2.90×10^5	2.8
QA-3 + QO-3	1.37×10^5	QA-2	1.37×10^5	0
QA-4 + QO-4	8.32×10^4	QA-3	8.40×10^4	0.95
QA-5 + QO-5	7.44×10^4	QA-4	7.30×10^4	1.9
QA-6 + QO-6	7.40×10^4	QA-5	7.21×10^4	2.6
QA-6 + Sum QO	5.84×10^5	QA-1 + QO-1	5.74×10^5	1.73

If the thorium content was not altered by previous processing an estimate of the original uranium-232 content of the uranium-233 could be obtained from this percentage. The following table, Table 14, was developed. It can be seen that the 12.7% thorium alpha calculation coincides with the 13% calculated value for 15 ppm uranium-232.

Table 14

EFFECT OF VARYING AMOUNTS OF ^{232}U ON THE ALPHA ACTIVITY OF
8 YEAR OLD ^{232}U

Line	Alpha Contributor	Disintegrations/min/gram U-233		
		10 ppm ^{232}U	15 ppm ^{232}U	20 ppm ^{232}U
1	^{233}U	2.11×10^{10}	2.11×10^{10}	2.11×10^{10}
2	^{232}U	0.04×10^{10}	0.06×10^{10}	0.08×10^{10}
3	Total U (Ln 1+2)	2.15×10^{10}	2.17×10^{10}	2.19×10^{10}
4	^{229}Th + Daughters	0.01×10^{10}	0.01×10^{10}	0.01×10^{10}
5	^{228}Th + Daughters	0.21×10^{10}	0.32×10^{10}	0.42×10^{10}
6	Total Th + Daughters (Line 4 + 5)	0.22×10^{10}	0.33×10^{10}	0.43×10^{10}
7	Total alphas/gram (Line 3 + 6)	2.36×10^{10}	2.50×10^{10}	2.62×10^{10}
8	Percent alphas from Th + Daughters (Line 6)/(Line 7) x 100	9.3	13.0	16.5

The sample used for this experiment had been recently used for uranium-thorium ion exchange separation experiments and the 15 ppm uranium-232 content calculated above is not necessarily a true value. The thorium content at the time of the extractions may not represent true transient equilibrium with the uranium. The calculation does, however, demonstrate a method for determining the uranium-232 content of a sample which has not been recently processed.

From the information in the preceding table, the weight of uranium in the original solution can be determined. The gross alpha count of the original solution was 5.74×10^5 counts per minute per one lambda. The geometry of a "2π" hemispherical gas flow alpha chamber is estimated at 51%. The total disintegrations per minute for the 10 ml solution can then be calculated as:

$$\frac{5.74 \times 10^5 \times 10^4}{0.51} = 1.125 \times 10^{10} \text{ dpm.}$$

The uranium contained in the solution is:

$$\frac{1.125 \times 10^{10}}{2.50 \times 10^{10}} = 0.45 \text{ g}$$

or $4.5 \times 10^4 \text{ } \mu\text{g/ml.}$

As the decay constant for uranium-233 is $4.28 \times 10^{-6} \text{ year}^{-1}$, thorium-229 is formed at the rate of 4.21 ppm/year by weight. For a few years the thorium-229 accumulation curve approximates a straight line and the eight-year-old sample could therefore be expected to contain

$$4.21 \times 8 \text{ yr} = 33.7 \text{ ppm thorium-229}$$

For the 0.45 g sample, the total thorium-229 content would be:

$$0.45 \times 33.7 = 15.2 \text{ } \mu\text{g}$$

or 1.52 $\mu\text{g/ml}$ of the aqueous solution. In terms of cpm/ λ , one μg thorium-229/ml is:

$$\frac{7.3 \times 10^4}{1.52} = 4.8 \times 10^4 \text{ cpm}/\lambda$$

For the pure uranium in the organic phases the cpm/ λ / μg of uranium/ml is:

$$\frac{4.99 \times 10^5}{4.5 \times 10^4} = 11.1 \text{ cpm}/\lambda$$

Data from the uranium removal are given on a weight basis in Table 15 and are plotted in Figure 6. In addition, the ratio of uranium to thorium by weight in the aqueous phase is given in Table 15. With the system used, nine extraction steps would be needed to obtain the thorium at more than 90% purity with respect to uranium. If the ratio of organic to aqueous volume was increased, the number of steps could be reduced.

Beyond six stages, alpha pulse height analysis does not yield useful data but gross alpha counts on the organic phases should be useful in estimating the number of additional extractions necessary to reach any desired purity of the thorium product. Since thorium appears to be insoluble in the organic phase, a number of additional extractions could be made without loss of thorium. The absence of thorium in the organic is so definite that, even if there were 2000 ppm thorium-

Figure 6. Uranium Left in Aqueous 232 in the original uranium, there would not be a loss of thorium to the organic.

Table 15

URANIUM LEFT IN 10 MILLILITERS AQUEOUS PHASE

Number of Extractions	Cumulative Organic Volume (ml)	Uranium in Aqueous (μg/ml)	Uranium/Thorium-229 Weight Ratio Aqueous
0	0	4.5 x 10 ⁴	2.93 x 10 ⁴
1	10	1.95 x 10 ⁴	1.25 x 10 ⁴
2	20	0.4 x 10 ³	4.12 x 10 ³
3	30	1.0 x 10 ³	6.45 x 10 ²
4	40	2.0 x 10 ²	129
5	50	50	32
6	60	10	6.5
(Values below were obtained by extrapolation)			
7	70	2	1.3
8	80	0.5	0.3
9	90	0.1	0.06

It is concluded that DSBPP-DEB solvent extraction is a feasible method for removing the thorium-229 daughter from quantities of aged uranium-233. Since the separation removes both thorium isotopes and all their daughters the uranium radiation level is temporarily reduced to a negligible value for further processing. (M. R. Hertz)

ANALYTICAL AND INSTRUMENTATION

NEUTRON ACTIVATION ANALYSIS

A neutron activation method for determining total boron and the isotopic ratio of elemental boron is being developed.

The boron-11 is determined by using the reaction: $^{11}\text{B} (n,p)^{11}\text{Be}$. Samples and standards are simultaneously irradiated in a fast neutron flux of $\sim 1 \times 10^9$ n/cm²/sec in the pneumatic transfer system. Beryllium-11 has a 13.6 sec half-life and emits a gamma energy at 2.12 MeV. A sensitivity of 400 c/mg of boron-11 was obtained.

The boron-10 is determined by neutron absorption using a plutonium-beryllium neutron source. One hundred milliliters of sample solution is placed in a doughnut-shaped container located in a paraffin-filled drum with a helium-3 filled detector through the center of the doughnut-shaped container. The source is situated so the neutrons pass through the sample to the detector. Since boron-10 absorbs neutrons, an increase in boron-10 concentration causes a decrease in count rate. Count rate decreases of ~ 3600 cpm per milligram of boron-10 per 100 ml of solution were obtained.

Initial work was done with HNO_3 - H_2SO_4 sample solutions with 10-min counts. A reagent blank was used for obtaining a zero boron count rate. Good precision was obtained with this technique; however, impurities present in the sample absorbed enough neutrons to make the total boron results high compared to those obtained by the titration method. A Dowex-50W ion exchange column used to remove impurities required that sample solutions be neutralized; this increased the solution to a volume greater than the sample cell.

The sample dissolution technique was changed to a carbonate fusion. A good fusion was obtained using 0.1-g samples; however, the fusion mixture, dissolved in a minimum of water and passed through the column, still gave high results. An atomic absorption analysis of the solution showed that much of the sodium had passed through the column, causing a high result due to the large thermal neutron cross-section of sodium-23. Other techniques for sodium removal are being investigated. (B. D. Craft and E. B. Nunn)

NUCLEAR MAGNETIC RESONANCE

Electron Inductive Effects in Substituted Polystyrenes The electron inductive effect of substituents in polystyrene is being studied by nuclear magnetic resonance spectroscopy to discover a correlation between the ease of polymerization of substituted polystyrenes and the substituent.

The NMR spectra of polystyrene, p-methylstyrene, p-methoxystyrene, m-chlorostyrene, m-nitrostyrene, and p-nitrostyrene were obtained at 60 Mcps and the chemical shifts of the vinyl protons were extracted from the spectra.

The positions of the NMR absorption bands of nuclei of the same species (in this case, protons) depends upon the chemical environment in which the nuclei are found in the molecule. In the substituted polystyrenes, the determination of the relative positions (chemical shifts) of the absorption bands of the three vinyl protons as a function of ring substitution was of interest.

The substituent group alters the chemical shift of the vinyl protons by an inductive effect in which electrons are either withdrawn or donated to the polystyrene ring and passed along to the vinyl group. The results obtained here revealed that para-substitution has a greater inductive effect than meta-substitution, and that the effect of substituents in the polystyrene ring is to decrease the electron density about the vinyl group, making the polystyrene molecule more vulnerable to nucleophilic attack by basic reagents. (A. Attalla and R. Eckstein)

STILBENE CRYSTAL, FAST NEUTRON SPECTROMETER

Data Reduction A stilbene crystal, fast neutron spectrometer is being used for measuring neutron energy spectra of isotopic neutron sources. Measurements are made of protons which have been elastically scattered by neutrons. The protons being scattered are hydrogen nuclei of the crystal (stilbene being a hydrocarbon), and therefore, all recoil protons above a lower cutoff point are measured. If the light output of the crystal were a function only of proton energy, differentiation of the pulse height data would yield the desired neutron energy spectrum.

Since the light output is principally, but not exclusively, a function of proton energy, numerical differentiation of the pulse height data has been used at Mound Laboratory and elsewhere with a fair degree of success. Small corrections resulting from the presence of carbon, second scattering of neutrons by hydrogen, and the finite size of the crystal can be made for small size crystals.¹⁶ An example of some data obtained with our

¹⁶H. W. Broek and C. E. Anderson, Rev. Sci. Instr. 31, 1063 (1960)

stilbene spectrometer is given in Figure 7. For comparison, data for the same source, obtained with nuclear emulsions,¹⁷ are shown. The agreement between the two sets of data is reasonably good.

In numerical differentiation, the light output, L , is assumed to be a function of proton energy, P , only. It has been observed, though, that the light output for a given monoenergetic neutron beam incident on a stilbene crystal is a function of both the neutron energy and the direction of the neutron beam relative to the crystal. This is illustrated in Figure 8 where the light output for neutrons incident from the direction referred to as $\theta=90^\circ$ is 18% greater than it is for direction $\theta=0^\circ$. (The light output for the 90° case was found to be essentially constant as the crystal was rotated about its axis of symmetry.) Since measurements are being made on recoil protons, the crystal response is a function of both proton energy and the direction of travel of the proton within the crystal, or

$$L = L(P, \theta) \quad (3)$$

In elastic scattering by hydrogen, proton and neutron energies are related by

$$P = E \cos^2 \psi \quad (4)$$

where ψ is the laboratory angle between the initial direction of the recoil proton and the direction of the incident neutron. If we restrict irradiation to a direction parallel to the crystal axis of symmetry, i. e., $\theta = 0^\circ$, then $\theta = \psi$ and

$$L = L(P, E) \quad (5)$$

The most desirable method of reducing stilbene pulse height data would be the use of a set of experimentally determined response curves for several different neutron energies. Such curves would also account for secondary reactions within the crystal and for the crystal size. This is being done by some investigators,^{18, 19} but an accelerator is required to obtain such curves. In the absence of an accelerator, theoretical response curves will be used. To obtain these curves, the following initial expression for the light output is being used

$$L = a [1 + d \sin^2 \theta] P^b \quad (6)$$

¹⁷M. E. Anderson and W. H. Bond, Jr., Nucl. Phys. **43**, 330 (1963)

¹⁸V. V. Verbinski, J. C. Courtney, W. R. Burrus, and T. A. Love, Proceedings for a Special Session on Fast Neutron Spectroscopy at the 1964 Winter Meeting of the American Nuclear Society (ANS-SD-2), 189 (1964)

¹⁹B. Brunfelter, J. Kockum, and H. O. Zetterstrom, Nucl. Instru. and Meth. **40**, 84 (1966)

Figure 7. A Comparison of Neutron Energy Measurements for a ^{239}Pu -Be (α, n) Source Obtained by Using a Stilbene Spectrometer with Those Obtained with Nuclear Emulsions

Figure 8. Illustrating Crystal Irradiation for Measuring Dependence of Light Output on Manner of Irradiation

where d is the fractional increase of the light output at $\theta = 90^\circ$ over that at $\theta = 0^\circ$, and the angle factor assumes the variation of light output is a cosine function with a period of 180° . By combining Equations 4 and 6 we get

$$L = a [1 + d (1-P/E)] P^b \quad (7)$$

If $d = 0$, the expression is the same as used for reduction by numerical differentiation. Others have calibrated crystals by using only protons recoiled at 0° in which case $P = E$ (see Equation 4). In this case, which is equivalent to $d = 0$, Equation 7 reduces to

$$L = aP^b, \quad (P = E) \quad (8)$$

This last relationship has been found to give a satisfactory fit to experimental data.^{18, 20} Values for b have been reported from 1.3 to 1.5. By using the emulsion data shown in Figure 7 as a standard, and adjusting the values for a and b until the best fit for the locations of the several maxima was found, we obtained $b = 1.38$.

Figure 9 illustrates the differences between response functions obtained by using values of 0.00 and 0.18 for d in Equation 7 for neutron energy of 10 MeV and all other factors such as finite resolution, second scattering, etc., being ignored.

A computer program to reduce the pulse height data is now being written which will incorporate Equation 7. Matrix methods will be used for this program. Initially, the other secondary corrections, such as second scattering by hydrogen, will be accounted for in the manner of reference 15. (M. E. Anderson)

²⁰M. McN. Wasson, AERE - R 4269 (1963) unpublished

Figure 9. Response Functions for Monoenergetic Neutrons to Illustrate the Effect of the Factor d in Equation 7