14.7-Mev Neutron-Induced Fission of U^{238}†

K. M. Broom
Department of Chemistry, University of Arkansas, Fayetteville, Arkansas
(Received October 2, 1961; revised manuscript received January 17, 1962)

Fission yields of twelve mass chains from 14-Mev neutron-induced fission of U^{238} were measured with the following results: $A=89$, 2.0%; 90, 3.4%; 91, 2.16%; 111, 0.6%; 113, 0.6%; 131, 2.7%; 132, 4.5%; 133, 2.6%; 134, 4.7%; 135, 5.0%; 139, 4.4%; 140, 4.3%. Fine structure in the mass yield curve in the region $A=131$–135 was observed with a distinct peak at $A=132$. This data can be explained at least qualitatively by the method proposed by Pappas and agrees with the fine structure observed by Ashizawa and Kuroda for U^{238} spontaneous fission.

I. INTRODUCTION

THE mass distribution of the fission products from spontaneous fission of U^{238} has been investigated in this laboratory during the past several years,[1–8] and it was felt highly desirable to carry out a detailed comparative study on the mass-yield curves for the spontaneous and fast neutron-induced fission of U^{238}.

The only mass yields reported for 14-Mev neutron fission of U^{238} to date are the papers by Ames *et al.*[9] and Cuninghame.[10] However, in neither of these investigations were yields for more than two consecutive mass numbers measured so that it was impossible for them to observe fine structure. Wahl,[11] on the other hand, has reported the yields for 14-Mev neutron fission of U^{235} for five consecutive mass chains ($A=131$–135). He observed practically no fine structure at this energy in sharp contrast to the marked peak at $A=134$ observed for thermal neutron induced fission of U^{235}.[12,13]

We have irradiated $U_3{}^{238}O_8$ with 14.7±0.4-Mev neutrons and measured the fission yields for $A=89$–91, 111, 113, 131–135, 139–140.

II. EXPERIMENTAL

Target Material

The U^{238} used in this work was supplied by Dr. H. M. Roth of the Research and Development Division, U. S. Atomic Energy Commission, Oak Ridge, Tennessee. The isotopic composition, determined by Oak Ridge, was found to be 99.989% U^{238} and 0.011% U^{235}. This material was received chemically impure and was exhaustively purified by standard techniques and converted to the oxide. Gravimetric analysis showed that the oxide contained 84.75% uranium.

Bombardment Techniques

The $U_3{}^{238}O_8$ obtained as described above was irradiated with 14.7±0.4-Mev neutrons produced by the $T(d,n)He^4$ reaction in the University of Arkansas 400-kv Cockcroft-Walton positive ion accelerator.[14] Total neutron fluxes in the range 10^{10}–10^{11} neutrons per sec were obtained.

The sample to be irradiated was wrapped in cadmium foil 0.06-cm thick (13.2 half-thicknesses) to eliminate any thermal neutron component. After irradiation, the length of which varied from about twenty minutes up to several hours depending upon the half-lives of the particular nuclides for which fission yields were to be determined, the sample ($U_3{}^{238}O_8$) was dissolved in dilute HNO_3 containing appropriate amounts of inactive carriers of the various elements which were isolated at varying times after bombardment. In every case Ba^{140} was isolated as a reference yield after allowing sufficient time for short-lived barium isotopes 139 and 141 to decay. The chemical separation procedures employed were standard methods.[15–17]

Counting Technique and Counting Efficiency

All the beta counting was done in a 2π end-window methane-flow proportional counter. The window was

† This work was supported by the U. S. Atomic Energy Commission.

[1] P. K. Kuroda and R. R. Edwards, J. Chem. Phys. **22**, 1940 (1954).
[2] P. K. Kuroda and R. R. Edwards, J. Inorg. & Nuclear Chem. **3**, 345 (1957).
[3] P. K. Kuroda, R. R. Edwards, and F. T. Ashizawa, J. Chem. Phys. **25**, 603 (1956).
[4] P. L. Parker and P. K. Kuroda, J. Chem. Phys. **25**, 1084 (1956).
[5] F. T. Ashizawa and P. K. Kuroda, J. Inorg. & Nuclear Chem. **5**, 12 (1957).
[6] P. L. Parker and P. K. Kuroda, J. Inorg. & Nuclear Chem. **5**, 153 (1958).
[7] R. H. Heydegger and P. K. Kuroda, J. Inorg. & Nuclear Chem. **12**, 12 (1959).
[8] M. P. Menon and P. K. Kuroda, Nuclear Science Eng. **10**, 70 (1961).
[9] D. P. Ames, J. W. Barnes, J. P. Balagna, A. A. Comstock, G. A. Cowan, P. B. Elkin, G. P. Ford, J. S. Gilmore, D. C. Hoffman, G. W. Knobeloch, E. J. Lang, M. A. Melnick, C. D. Minkkinen, B. D. Pollack, J. E. Sattizahn, C. W. Stanley, and B. Waren, Los Alamos Scientific Laboratory Report LA-1997, 1958 (unpublished).
[10] J. G. Cuninghame, J. Inorg. & Nuclear Chem. **5**, 1 (1957).
[11] A. C. Wahl, Phys. Rev. **99**, 730 (1955).
[12] L. E. Glendenin, Phys. Rev. **75**, 337 (1949).
[13] L. E. Glendenin, E. P. Steinberg, M. G. Inghram, and D. C. Hess, Phys. Rev. **84**, 860 (1951).
[14] W. L. Bronner, K. W. Ehlers, W. W. Eukel, H. S. Gordon, R. C. Marker, F. Voelker, and R. W. Fink, Nucleonics **17**, 94 (1959).
[15] C. D. Coryell and N. Sugarman, *Radiochemical Studies: The Fission Products* (McGraw-Hill Book Company, Inc., New York, 1951), Book 3, Parts V–VIII.
[16] D. Sunderman, thesis, University of Michigan, 1956 (unpublished).
[17] P. K. Kuroda, Argonne National Laboratory Report ANL-5829, 1958 (unpublished), p. 170.

1.4 mg per cm² singly aluminized Mylar film. Over a period of two years, during which this work was done, the background activity in the counter remained constant at 20±2 cpm. The background was taken before and after samples were counted and subtracted from the observed activity. The count-rate vs voltage plateau for the counter used was 600 v in length with less than one percent slope and did not shift appreciably during the entire two year period.

All samples were mounted by using the filter stick technique to provide an evenly distributed precipitate over a well-defined area (3.14 cm²). The samples were counted on a saturation-backscattering thickness of stainless steel ($\frac{1}{2}$-in. thick).

In order to convert observed counting rates to absolute activities, self-absorption curves were obtained for 4-π counted samples of four of the longer-lived nuclides using the method suggested by Steinberg.[18]

Such a technique is exact only where the radioactive species to be counted has a relatively long half-life so that a self-absorption curve may be measured. For short-lived nuclides an alternative method for calculating the detection efficiencies may be employed as described at the Conference on Absolute Beta Counting.[19]

This technique was used for those nuclides for which self-absorption curves did not apply. Since f_{ssA} as given by Nervik and Stevenson[20] varies only ~5% for betas from 0.6 to 2.8-Mev maximum energy at the sample densities employed here, the efficiencies determined in this manner varied little (less than 5%), for the nuclides in question, from the self-absorption curves for the long-lived isotope determined experimentally.

Treatment of Data

For each sample the gross beta decay was observed and resolved into the various components using reported half-lives. From a knowledge of the absolute activity at the end of the irradiation, the total number of atoms of a particular species produced during the irradiation was determined by applying a correction for the decay of the species during the irradiation.

$$A_{\text{total}}=\frac{A\lambda_A}{1-e^{-\lambda_A t}}t \qquad (1)$$

where A_{total} = total number of atoms of species A produced in the bombardment, $A\lambda_A$ = activity of A at end of bombardment, and t = length of bombardment.

The absolute yield of Ba^{140} was determined using an aluminum monitor, and all other yields were measured relative to Ba^{140} by means of the equation

$$\text{yield of } A=\frac{\text{yield of } Ba^{140}}{Ba^{140}\text{ total}}(A_{\text{total}}). \qquad (2)$$

Sources of Error

(1) An error in fitting the decay curve to the measured activity introduces an uncertainty in the total number of atoms of the radioactive species produced by the irradiation. Such errors amounted to ~3% for the long-lived nuclides (Ba^{140}, I^{131}, Ag^{111}, Sr^{89}, and Sr^{90}) and ~5% for the short-lived species (Ba^{139}, I^{132}–I^{135}, Ag^{113}, and Sr^{91}).

(2) Corrections were made for fluctuations in the neutron flux during the irradiations where possible, and in other cases agreement between triplicate samples was required.

(3) Errors introduced by thermal neutrons, 3-Mev neutrons, or chemical yield determinations were insignificant.

The errors reported for all the yields measured in this work are most probable errors propagated in the usual way, assuming the Ba^{140} yield to be 4.3% as measured in this work.

Absolute Fission Yield of Ba^{140}

Ba^{140} was chosen as the reference nuclide for the work. In order to convert the relative yields of the other nuclides to absolute yields, the Ba^{140} yield was measured using the $Al^{27}(n,\alpha)Na^{24}$ cross section 115 mb as reported by Poularikas and Fink.[21]

A sample of $U_3^{238}O_8$ was "sandwiched" between double layers of aluminum foil and irradiated with 14.7-Mev neutrons for two hours. Ba^{140} was isolated after allowing sufficient time for the shorter lived barium activities to decay.

By counting both the Ba^{140} and the aluminum monitor, the activity of Ba^{140} and Na^{24} at the end of the irradiation was obtained. Since these nuclides have different half-lives, it was necessary to correct these data to saturation bombardment time.

Using the thin target approximation,[22] the cross section for the reaction may be obtained from

$$\sigma_{\text{sample}}=\frac{N_{\text{sample}}Q_{\text{monitor}}}{N_{\text{monitor}}Q_{\text{sample}}}\sigma_{\text{monitor}} \qquad (3)$$

since both monitor and sample were of equal area and were irradiated under identical conditions. In this equation σ is the cross section for the process in question, expressed in cm²; N is the absolute disintegration rate for saturation bombardment; and Q is the number of target nuclei per cm². Since all of the terms on the right-

[18] E. P. Steinberg, Argonne National Laboratory Report ANL-5622, 1956 (unpublished).
[19] *Conference on Absolute Beta Counting*, No. 8, Nuclear Sci. Series (National Research Council, 1950).
[20] W. E. Nervik and P. C. Stevenson, Nucleonics **10**, 18 (1952).
[21] A. Poularikas and R. W. Fink, Phys. Rev. **115**, 989 (1959).
[22] G. Friedlander and J. Kennedy, *Nuclear and Radiochemistry* (John Wiley & Sons, Inc., New York, 1955), pp. 60–62.

hand side of the equation are known, σ_{sample} may be calculated. This quantity was used to calculate the absolute yield of Ba^{140} by assuming the value for the 14-Mev neutron induced fission cross section of U^{238} to be 1.13±0.05 b.[23]

The results of three such determinations gave a value of 0.0491±0.0045 b for partial fission cross section of U^{238} leading to mass number 140 or (4.3±0.4)% for the fission yield of Ba^{140}.

III. RESULTS

Yield of Ba^{139}

By separating barium samples quickly after irradiation, it was possible to observe the 85-min Ba^{139}. The Ba^{140}-La^{140} component was subtracted from the gross decay, and the resulting curve was used to calculate the total number of atoms of Ba^{140} and Ba^{139} produced during the irradiation as described previously.

The results of duplicate determination gave a value for the Ba^{139} fission yield of (4.4±0.5)%.

Strontium Yields

The gross beta decay curve for the strontium isotopes was resolved into a 50.4-day Sr^{89} component and a 9.7-hr Sr^{91} component. The results of three such determinations gave (2.0±0.2)% for Sr^{89} and (2.6±0.3)% for Sr^{91}.

After allowing the Sr^{89} to decay for 1.5 yr, the samples were redissolved and Sr^{90}-Y^{90} separation was performed by carrying the Y^{90} on $Fe(OH)_3$.

By using a Tracerlab Ce-14 low-level counting system (background activity: counter $A=0.59\pm0.03$ cpm, counter $B=1.10\pm0.04$ cpm), it was possible to determine the Sr^{90} yield. Results of three samples gave (3.4±0.3)% for the Sr^{90} fission yield.

Iodine Yields

Samples of iodine were separated at various times after irradiation, and from the gross beta decay curves it was possible to calculate the fission yields of I^{131}, I^{132}, I^{133}, I^{134}, and I^{135}. Sealed sources were used to prevent any escape of the Xe daughters. The yields calculated from three sets of samples are listed below.

I^{131} (2.7±0.2)%
I^{132} (4.5±0.4)%
I^{133} (2.6±0.3)%
I^{134} (4.7±0.5)%
I^{135} (5.0±0.5)%

Silver Yields

Using standard chemical separation procedures referred to previously, silver samples were isolated and the yields of Ag^{111} and Ag^{113} were calculated. Results of two determinations gave fission yields of (0.6±0.1)% for Ag^{111} and (0.6±0.1)% for Ag^{113}.

The values of the yields measured in this work as well as the yields determined by Cuninghame[10] and Ames *et al.*[9] are listed in Table I and are plotted in Fig. 1. The spontaneous fission yields[1–8,24,25] are plotted for comparison in Fig. 2.

TABLE I. 14-Mev fission yields.

Nuclide	Percent this work	Percent Cuninghame[10]	Percent Ames *et al.*[9]
Sr^{89}	2.0±0.2	2.30±0.12	3.0
Sr^{90}	3.4±0.3		
Sr^{91}	2.6±0.3		
Y^{91}		2.78±0.14	
Zr^{97}			4.8
Mo^{99}		5.58±0.28	5.7
Rh^{105}			3.4
Ag^{111}	0.6±0.1	0.81±0.04	0.87
Ag^{113}	0.6±0.1		
Cd^{115}		0.64±0.04	0.71
Sn^{121}			0.73
Sn^{125}			0.83
Sb^{127}			1.43
I^{131}	2.7±0.2		
I^{132}	4.5±0.4		
Te^{132}			4.7
I^{133}	2.6±0.3		
I^{134}	4.7±0.5		
I^{135}	5.0±0.5		
Ba^{139}	4.4±0.5		
Ba^{140}	4.3±0.4	4.41±0.22	4.6
Ce^{143}		3.91±0.27	
Pr^{143}		3.16±0.16	
Ce^{144}		2.68±0.16	3.4
Nd^{147}		1.99±0.10	
Sm^{153}		0.39±0.02	
Eu^{156}		0.13±0.01	0.22

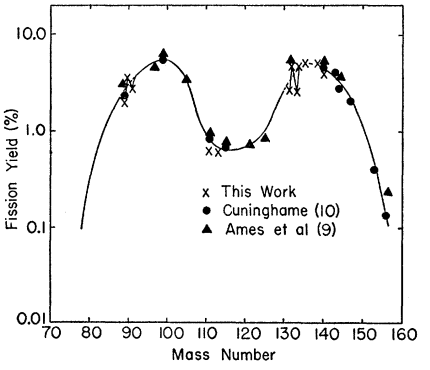

FIG. 1. Mass yield curve for 14-Mev neutron-induced fission of U^{238}.

[23] N. N. Flerov, A. A. Berezin, and I. E. Chelnokov, Atomnaya Energ. **5**, 657 (1958).

[24] J. MacNamara and H. G. Thode, Phys. Rev. **80**, 471 (1950).
[25] G. W. Wetherill, Phys. Rev. **92**, 907 (1953).

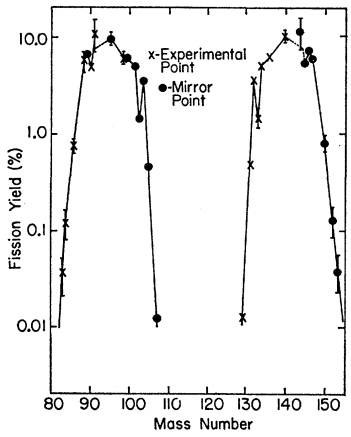

FIG. 2. Mass yield curve for spontaneous fission of U^{238}.

IV. DISCUSSION

The mass-yield curve for 14-Mev fission of U^{238} exhibits the usual effects of increased energy of incident particle when compared to the spontaneous fission curve, Fig. 2. The peak-to-valley ratio decreases to ~10 to 1, the peaks are lowered from ~10% to ~5%, and the wings are splayed out. Such effects are quite common and are qualitatively explained by considering symmetric fission and asymmetric fission to be two separate, competing modes. As the energy is increased, the broad, symmetrical peak gradually rises, filling up the valley in the mass-yield curve.

One of the most interesting features of the spontaneous fission curve is the presence of a pronounced peak at mass number 132. It has been suggested by Kuroda and Ashizawa[5] that this is related to the stability of the doubly magic nuclide $_{50}Sn_{82}^{132}$. In the case of 14-Mev U^{238} fission, Cuninghame[10] has suggested that fine structure at $A=134$ should be observed similar to that observed in U^{235} thermal fission.[13] A slight amount of fine structure may be present in the U^{238} mass yield curve at $A=134$, but, if present, it is much smaller than the peak observed at $A=132$.

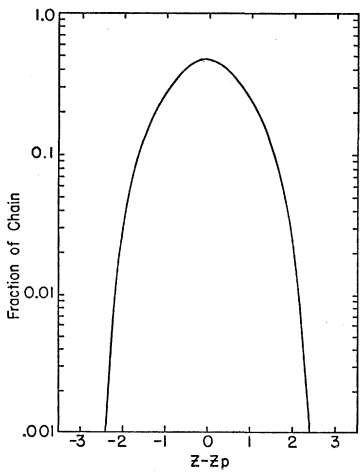

FIG. 3. Variation of yield with nuclear charge for 14.7-Mev neutron fission of U^{235}.

TABLE II. Independent yields calculated by Pappas' method.[a]

A	Z_p	Z	$Z-Z_p$	Fraction of chain	Yield assumed from Fig. 1 (%)	Independent yield (%)
131	50.7	49	−1.7	0.06	3.2	0.19
	50.7	50	−0.7	0.34	3.2	1.09
	50.7	51	0.3	0.47	3.2	1.44
	50.7	52	1.3	0.17	3.2	0.54
132	51.1	49	−2.1	0.01	3.8	0.06
	51.1	50	−1.1	0.22	3.8	0.84
	51.1	51	−0.1	0.48	3.8	1.82
	51.1	52	0.9	0.26	3.8	0.99
	51.1	53	1.9	0.03	3.8	0.10
133	51.5	50	−1.5	0.11	4.2	0.46
	51.5	51	−0.5	0.40	4.2	1.68
	51.5	52	0.5	0.40	4.2	1.68
	51.5	53	1.5	0.10	4.2	0.42
134	52.0	50	−2	0.02	4.7	0.10
	52.0	51	−1	0.40	4.7	1.08
	52.0	52	0	0.48	4.7	2.26
	52.0	53	1	0.40	4.7	1.08
	52.0	54	2	0.02	4.7	0.10
135	52.5	51	−1.5	0.11	5.0	0.55
	52.5	52	−0.5	0.40	5.0	2.00
	52.5	53	0.5	0.40	5.0	2.00
	52.5	54	1.5	0.10	5.0	0.50

[a] See reference 28.

An attempt was made to calculate the fission yields based on two different methods: (1) *Postulate of Equal Charge Displacement*—Glendenin, Coryell, and Edwards[26] proposed that the most probable charge for a given mass chain is equally distant from the most stable charge for both fission fragments. The most probable charge Z_p is given by

$$Z_p=Z_A-\tfrac{1}{2}(Z_A-Z_{239-A}-Z_{239}), \qquad (4)$$

where Z_A is the most stable charge for a given mass number A, Z_{239-A} is the most stable charge for the complementary fission fragment, and Z_{239} is the atomic number of the compound nucleus (92 in the case of uranium). Z_A values were calculated from the Bohr-Wheeler theory[27] by Glendenin *et al.*[26]

Also, these investigators proposed that the charge

TABLE III. Fission yield (%).

A	Calculated[a]	Observed
131	3.33	2.74
132	4.21	4.46
133	3.78	2.63
134	4.62	4.70
135	5.0	5.01

[a] Calculated by Pappas' method.[28]

[26] L. E. Glendenin, C. D. Coryell, and R. R. Edwards, *Radiochemical Studies: The Fission Products* (McGraw-Hill Book Company, Inc., New York, 1951), paper 52, Book 1.
[27] N. Bohr and J. A. Wheeler, Phys. Rev. **56**, 426 (1939).

distribution is very nearly symmetrical and, using experimentally determined independent yields from thermal fission of U^{235}, they obtained a charge distribution curve similar to the one in Fig. 3. By means of such a curve, it is possible to calculate the charge distribution along any particular mass chain A. The curve shown was obtained by Ames *et al.*[9] for 14-Mev fission of U^{235} using a value of three for the number of prompt neutrons emitted by the heavy fragment. This curve was used to estimate the charge distribution for 14-Mev U^{238} fission because there is insufficient independent yield data for U^{238}. Since Cuninghame[10] and Ames *et al.*[9] have shown that the heavy fragment from 14-Mev U^{238} fission emits ~ 3 prompt neutrons, the curve for U^{235} is considered a good approximation. By using the smooth curve drawn through the yields in Fig. 1, the independent yields were determined for mass numbers 131 to 135. In each case Z_p falls short of the 82-neutron closed shell and the independent yields provide too few nuclides beyond the closed shell to account for the peak at $A=132$.

(2) Pappas[28] has suggested a modification of the equal charge displacement hypothesis by proposing that equal charge displacement applies before neutron emission. Independent yields are calculated by his method in the following manner: First, Z_p is calculated by the modified form of Eq. (4).

$$Z_p = Z_{A+n} - \tfrac{1}{2}(Z_{238-A-n} + Z_{A+n} - 92). \qquad (5)$$

The most probable charges, Z_{A+n} and $Z_{238-A-n}$, are obtained from the maximum stability lines of Coryell.[29] Secondly, the fractional yield is read from Fig. 3. The The results of such calculations are shown in Table II and Fig. 4. Finally, a correction for the emission of prompt shell neutrons is made by transferring the independent yields of $_{50}Sn_{83}^{133}$ and $_{49}In_{83}^{132}$ to $_{50}Sn_{82}^{132}$ and $_{49}I_{82}^{131}$, respectively. It should be noted that no correction is made for neutron emission by $_{52}Te_{83}^{135}$ or $_{51}Sb_{83}^{134}$ because both of these nuclides (a) have measurable beta decay half-lives ($Sb^{134}T_{\frac{1}{2}}=45$ sec, $Te^{135}T_{\frac{1}{2}}<2$ min) and, (b) they lie closer to stability than do Sn^{133} or In^{132}.

Z \ N	79	80	81	82	83	84
54		Xe^{134} .104%	Xe^{135} .50%	Xe^{136}		
53	I^{132} .10%	I^{133} .42%	I^{134} 1.08%	I^{135} 2.00%	I^{136} (86 sec β^-)	
52	Te^{131} .54%	Te^{132} .99%	Te^{133} 1.68%	Te^{134} 2.26%	Te^{135} (<2 min β^-) 2.00%	
51		Sb^{131} 1.44%	Sb^{132} 1.82%	Sb^{133} 1.68%	Sb^{134} (45 sec β^-) 1.08%	Sb^{135} .50%
50			Sn^{131} 1.09%	Sn^{132} .84%	Sn^{133} .461%	Sn^{134} .104%
49				In^{131} .19%	In^{132} .068%	

FIG. 4. Independent yields calculated by Pappas' method.

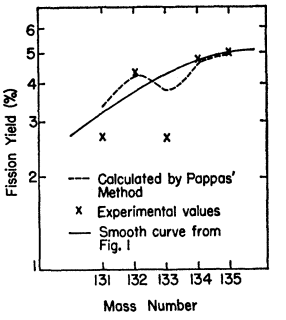

FIG. 5. Calculated and experimental values of fission yields for $A=131$–135.

The fission yields for $A=131$ to 135 may be obtained by summing the resulting independent yields. The values are listed in Table III and are plotted in Fig. 5.

The actual values of the calculated yields are subject to the large error in drawing a smooth curve through the experimental points; nevertheless, it seems significant that the calculated yields do show fine structure at mass 132 in precisely the same manner as observed experimentally.

In summary, the present work has revealed several significant facts:

(1) The expected changes in the fission yields for spontaneous vs 14-Mev fission were observed. For 14-Mev neutron fission the peak-to-valley ratio decreased, the yields of the peak nuclides decreased, the peaks broadened, and so forth.

(2) Fine structure was noted in 14-Mev fission at mass numbers 89–91 and 131–135, but the fine structure which Cuninghame[10] predicted was not observed. The yields for $A=131$–135 showed a high value for 132 similar to the effect reported by Kuroda and Ashizawa[5] and calculation of the yields for $A=131$–135 by the method proposed by Pappas[28] gave at least qualitative agreement with the experimental data.

ACKNOWLEDGMENTS

The author would like to thank the U. S. Atomic Energy Commission whose financial assistance made this work possible; Professor D. G. Gardner and Professor R. W. Fink for their many helpful suggestions; H. L. Hodges for his assistance in counting the Sr^{90} samples; J. E. Wray, the accelerator operator; and Dr. B. T. Kenna for his assistance in preparing the manuscript.

This paper represents a part of the dissertation submitted by K. M. Broom to the Graduate School of the University of Arkansas in partial fulfillment of the requirements for the degree of Master of Science. In conclusion, the author wishes to express his gratitude to Professor P. K. Kuroda for his kind guidance.

[28] A. C. Pappas, Massachusetts Institute of Technology Technical Report No. 63, 1953 (unpublished).
[29] C. D. Coryell, Ann. Rev. Nuclear Sci. 2, 325 (1953).