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Delayed neutron yields: Time dependent measurements and a predictive model

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The delayed neutrons from neutron-induced fission in ^{232}U, ^{237}Np, ^{238}Pu, ^{241}Am, $^{242}Am^{m}$, ^{245}Cm, and ^{249}Cf were studied for the first time; those from ^{232}Th, ^{233}U, ^{235}U, ^{238}U, ^{239}Pu, ^{241}Pu, and ^{242}Pu were measured again. The data were used to develop an expression for the prediction of the absolute delayed neutron yield, and the prediction of delayed neutron emission with time. This approach accurately predicts observed delayed neutron yields and decay characteristics. A fission product yield model was used in conjunction with delayed neutron emission probability to analytically predict delayed neutron characteristics. The results of this analysis are in excellent agreement with experimental values.

[RADIOACTIVITY Delayed neutron yield from thorium and transuranium elements, measured and calculated values.]

I. INTRODUCTION

Predictive models for fission yields have been based primarily on yield data taken on isotopes near stability. These models and the data have been reviewed in detail elsewhere.[1-6] However, less attention has been given to the time-dependent-delayed neutron yields from fission and how they affect the overall fission yields. This area, as reviewed recently in the IAEA Vienna meeting,[7] is of importance in the design and estimation of performance characteristics of reactors with reprocessed fuels. In order to construct a reliable model for yield predictions in a wide variety of fissioning systems a data base as large as possible is necessary.

We have modified and extended a computer-controlled system for the automated measurement of time dependent neutron emissions. Using ^{3}He ionization chambers and BF_3 counters we have measured the time dependent gross beta-delayed neutron yield for a variety of systems ranging from ^{232}Th to ^{252}Cf. Using these data we have then constructed a fission-yield model which reliably predicts the yields of neutron-rich nuclei. These data and the modeling have allowed us to investigate the odd-even effect over a broad range of fissioning nuclides. Section II presents the details of our experimental measurements; Sec. III, the results; and in Sec. IV we discuss our modeling of the fission yields.

II. EXPERIMENTAL PROCEDURES

In order to measure delayed neutron yields of the transuranium nuclides which were available only in small quantities we utilized the high neutron flux available at the Livermore Pool-Type Reactor (LPTR). It should be noted that previous studies, such as those of Keepin,[8] required gram quantities. In contrast, samples used in this work ranged from micrograms for nuclides with large thermal fission cross section to several milligrams for materials with low thermal fission cross sections.

A. Irradiation system

The irradiation, transportation, and measurement of samples were performed under computer control. A block diagram of our transportation system is shown in Fig. 1. This system, under control of a PDP-8 computer, was used to send rabbits containing samples to the core of the LPTR. The rabbit's flight in the pneumatic tube system was monitored by several photosensors that reported the rabbit's passage to the monitors under PDP-8 control. Arrival of the rabbit at the core irradiation position was detected by a sound sensor. After predetermined time the rabbit was sent to a counting station located 46 m from the core and 2.40 m below ground level. This location provided sufficient shielding to

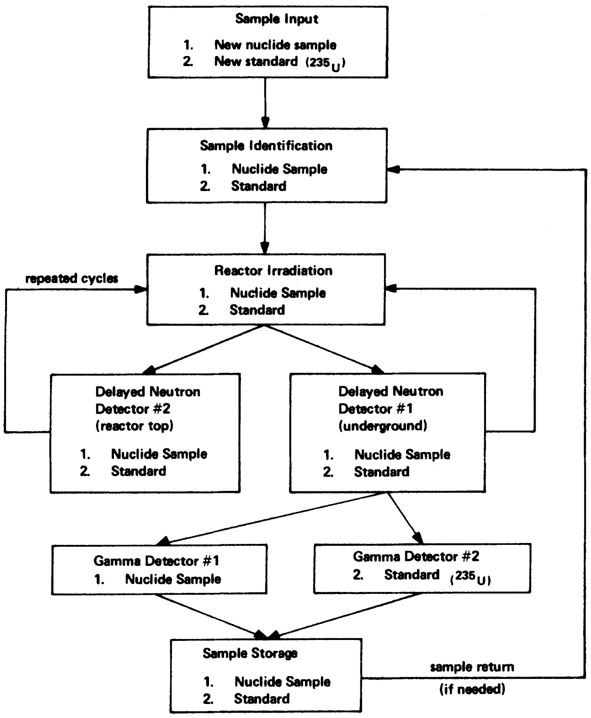

FIG. 1. Block diagram of automated sample irradiation system.

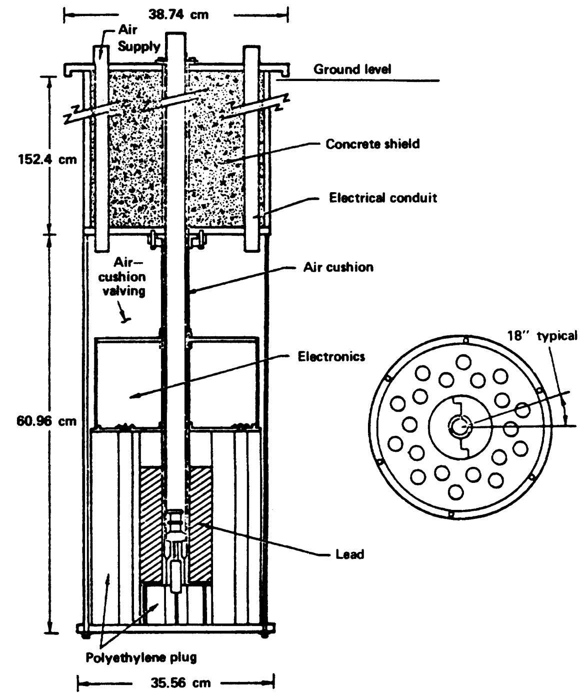

FIG. 2. Diagram of the delayed-neutron detector (taken from Lawrence Livermore Lab Report No. UCID 16911-76-3).

eliminate any background neutron activity. The counting station, consisting of 20 ^{3}He ionization chambers placed concentrically around the sample and embedded in polyethylene, is shown in Fig. 2. The 20 detector tubes were connected so that the dead time of one counter did not affect any of the others. This gave an efficiency of about 30% with a dead-time constant of about 3.1 μ sec that was capable of tolerating count rates up to 100 000 counts per second.

A second neutron counter was constructed closer to the reactor to allow the start of counting at times as early as possible. The detector system is shown in Fig. 3. This system provided a flight time of 300 msec for the rabbit. It consisted of a single BF_3 tube embedded in polyethylene and shielded from the sample by 13 mm of lead.

B. Target material composition

Since fission cross sections at thermal energy are normally much larger than those at high energy, small impurities of fissile nuclides could bias the results of fast fission studies. As an example, the presence of 0.1% ^{235}U in a ^{238}U sample would bias the results and even lesser amounts would lower the observed yield. This is because the total yield of ^{235}U delayed neutrons is about one-third the yield of ^{238}U. Samples with very large thermal cross sections, such as ^{249}Cf, ^{245}Cm, and ^{242}Am^m required little concern about impurity contamination. In other cases, such as ^{232}Th, ^{237}Np, ^{241}Am, ^{239}Pu, and ^{233}U, pure samples were obtained by chemical means. Mass separation was required in the other cases. For example, ^{235}U (93% enriched) was readily available but a special ultrapure ^{238}U (99.999% ^{238}U cadmium covered) was found suitable for study. Ultra pure ^{242}Pu (0.032% fissile impurities) was studied with and without cadmium covering and no difference was observed, so it was considered acceptable. Mass separated standards of ^{241}Pu, ^{232}U, and ^{238}Pu were also obtained.

Some samples had to be rejected as summarized in Tables I and II. A ^{240}Pu sample with 1% ^{239}Pu impurity was rejected and a ^{244}Pu sample contained enough ^{241}Pu to prevent obtaining any useful results. A very pure sample of ^{243}Am still contained enough ^{241}Am to bias the results so it was rejected. Finally, a ^{251}Cf sample contained enough ^{252}Cf to cause an unacceptable spontaneous fission neutron background.

It is important to note that the data provide a check on the sample purity. As discussed later in this paper, it is possible to predict with good accuracy the total delayed neutron yield a nuclide will have. This yield varies dramatically from nuclide to nuclide and so a sample which gives a

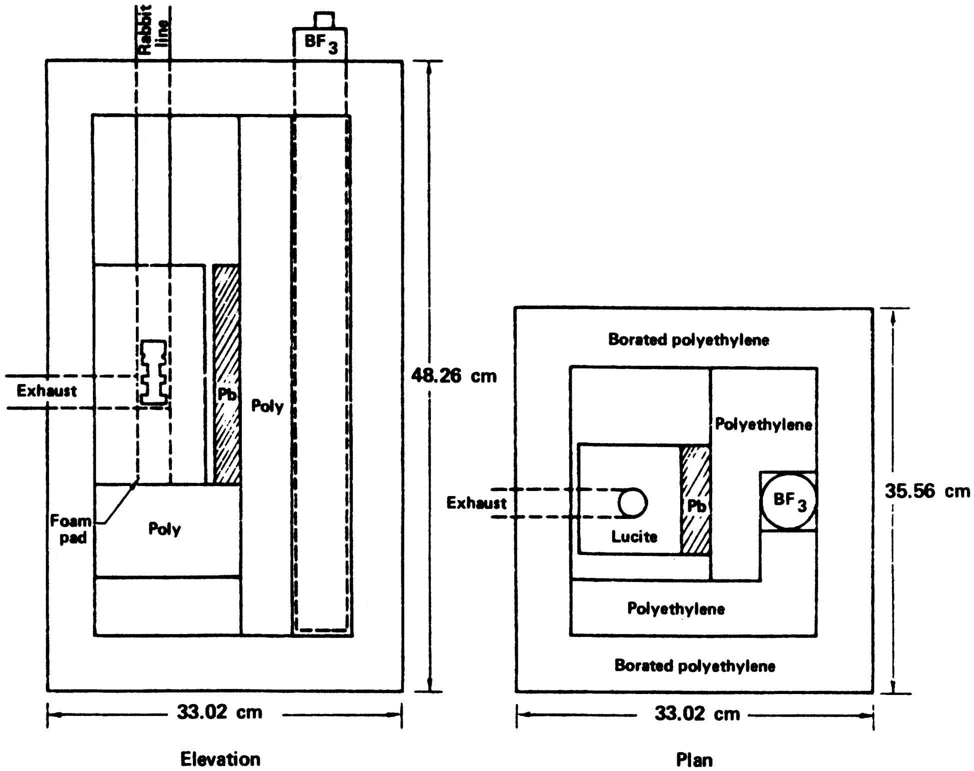

FIG. 3. Diagram of the neutron detector located on the reactor top.

TABLE I. Simple samples.

Nuclides of interest	Assay
^{232}Th	Chemically purified thorium (> 99.5% ^{232}Th)
^{232}U	Isotopically purified ^{232}U (99.99% ^{232}U)
^{233}U	Sample contained 4 ppm ^{232}U, 95.1% ^{233}U, 0.5% ^{239}U, 0.8% ^{235}U, 0.1% ^{236}U, and 3.5% ^{238}U.
^{235}U	^{235}U enriched to 93.7%.
^{237}Np	Sample contained 0.7% Th, 0.1% U, 0.01% Pu, and the rest Np.
^{238}Pu	Isotopically purified ^{238}Pu containing < 0.1% ^{238}U, 0.1% ^{239}Pu, and the rest ^{238}Pu.
^{239}Pu	Chemically purified sample containing 0.01% ^{238}Pu, 93.6% ^{239}Pu, 5.7% ^{240}Pu, and 0.65% ^{241}Pu.
^{241}Pu	Isotopically separated plutonium containing < 0.1% ^{240}Pu, 0.1% ^{242}Pu, and the rest ^{241}Pu.
^{241}Am	Chemically purified ^{241}Am from the decay of ^{241}Pu containing 1.8% ^{237}Np, < 0.1% all other fissile impurities, and the rest ^{241}Am.
^{242}Am^{m}	Isotopically purified ^{242}Am^{m} containing 0.79% ^{241}Am, 99.21% ^{242}Am^{m}, and < 0.007% ^{243}Am.
^{245}Cm	Isotopically purified ^{245}Cm containing 0.281% ^{244}Cm, 0.215% ^{246}Cm, 0.013% ^{247}Cm, 0.231% ^{248}Cm, and the rest ^{245}Cm.
^{249}Cf	Chemically purified ^{249}Cf from the decay of ^{249}Bk containing < 0.1% fissile impurities.

TABLE II. Difficult samples.

Nuclide of interest	Impurity	Solution
^{238}U	^{235}U	Use of high purity ^{238}U covered with cadmium (99.999% ^{238}U).
^{240}Pu	^{239}Pu	Sample not used.
^{242}Pu	^{241}Pu	Use of high purity ^{242}Pu, checked with cadmium covered sample (99.90% ^{242}Pu).
^{244}Pu	^{241}Pu	Data dominated by ^{241}Pu fission. Data not used.
^{243}Am	^{241}Am	Data dominated by ^{241}Am fission. Data not used.
^{244}Cm	^{245}Cm	Sample not used.
^{251}Cf	^{252}Cf	Data dominated by spontaneous fission neutron background. Data not used.

different absolute yield required further investigation. Such an example was the ^{242}Pu sample. The absolute yield observed is considerably lower than expected (probably due to the even-odd effect). This could have been due to impurities (^{239}Pu and ^{241}Pu) with lower absolute yields and large thermal cross sections. However, covering the sample with cadmium would have decreased this effect and the change would have been noticed. Covering a sample of pure ^{242}Pu with cadmium would have no effect since thermal neutrons do not cause fission in ^{242}Pu. In a nuclide with a large thermal fission cross section (such as ^{239}Pu or ^{241}Pu) the ratio of fission in a bare sample to a cadmium covered sample is about 10. Since no change was observed in the delayed neutron yield of the ^{242}Pu sample, the low absolute yield was assumed to be real.

C. Transit time correction

Due to the relatively long transit time, a fraction of the shortest delayed neutrons was missed. We analyzed Keepin's delayed neutron yields and found that the fraction of the delayed neutrons observed at $t = 0.4$ sec to that at $t = 0.0$ sec was a function of the quantity A_c/Z_c, where A_c and Z_c were the composite mass and charge of the fissioning material, respectively. This assumption has some basis in that the larger the ratio A_c/Z_c the more neutron rich the fission products that are produced. Specifically, the fraction of delayed neutrons missing at $t = 0.4$ sec was derived from the equation

$$F = 1 - \frac{y(t=0)}{y(t=0.4)} = 1.9207\,\frac{A_c}{Z_c} - 4.788 \pm (2\%).$$

This correction was then applied to the observed delayed neutron yield at $t = 0.4$ sec to give an absolute yield at $t = 0$ sec. The correction was of the order of 10% and so the error associated with this correction was small (0.2%). Typically, this did not change the result at all. In all cases except ^{232}Th and ^{238}U the effect was minimal. For nuclides where a short-lived group was observed (that is, 0.7 sec^{-1}) no correction was applied. For nuclides where a short-lived group was not observed it was assumed that a short-lived group with low yield was missed due to the long transit time. In these cases the correction was applied. The difference between the observed yield (extrapolated to $t = 0$) and the calculated absolute yield using this correction was assigned to a short-lived group. In all cases this group was very small. We adopted Keepin's suggested value of 0.514 ± 0.013 sec for the average half-life of his group V neutrons ($= 1.35$ sec^{-1}) to this short lived group.

^{232}Th and ^{238}U were treated somewhat differently. Because these two nuclides were extremely neutron rich, the correction needed was significant. However, both of these nuclides were studied by Keepin so that accurate information was available about the decay from $t = 0$ to $t = 0.4$ sec. In these two cases the shortest-lived group yield and decay constant was modified to accurately reflect the decay observed from $t = 0$ to $t = 0.4$ sec.

D. Energy sensitivity of detector

Energy sensitivity of the delayed neutron counters was of serious concern since a change in counter efficiency with neutron energy would give a distorted count rate. This is because delayed neutrons are similar to gamma rays in that those with the highest energies also tend to have the shortest half-lives. Thus, if a counter had a higher counting efficiency for low energy neutrons one would observe an increase of the 55 and 22 sec groups relative to the shortest-lived (most energetic) groups. Plots of data taken with the two delayed neutron detectors used in this work were compared with plots of Keepin's data (taken with an energy insensitive detector). No variation was observed for any nuclide. Since there is a very large change in relative group yields from ^{232}Th and ^{238}U to ^{233}U and ^{239}Pu this was considered a sufficient check on the energy sensitivity of the counter.

E. Yield normalization

The γ-ray spectra used in the absolute yield normalization was taken with one of the four mated coaxial Ge (Li) detectors associated with the pneumatic system. Each counter was set in an identical movable holder to provide variable but reproducible counter geometry. Measurements were begun about 30 min after irradiation and extended over eight hours. Another measurement was taken 24 h later to emphasize the longer-lived fission products. A similar measurement was taken on a ^{235}U standard so that the ratio of fissions in the sample to those of the ^{235}U standard was found. This ratio incorporated the published fission yields of the fission product in question for both ^{235}U fission and fission in the sample. In general, we were able to identify and use over a dozen different fission products to establish this ratio. The one exception was ^{237}Np. In this case, the decay of ^{238}Np formed from neutron capture precluded gamma counting below 1 MeV. Fortunately, several fission products have gamma energies above this energy and these were used to find the fission ratio. The 60 keV gamma peak associated with ^{241}Am caused counter deadtime problems. This was reduced by shielding the gamma counters with 1.5 mm of cadmium.

III. EXPERIMENTAL RESULTS

We measured the following time-dependent beta-delayed neutron yields: ^{232}Th, ^{232}U, ^{233}U, ^{235}U, ^{238}U, ^{237}Np, ^{238}Pu, ^{239}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{242}Am^m, ^{245}Cm, and ^{249}Cf. For these, a least squares fit of the time-dependent neutron data was made and the results are given in Table III. In addition, several nuclides were measured but had to be discarded because of severe contaminant problems. The discarded nuclei were ^{240}Pu, ^{244}Pu, ^{243}Am, ^{244}Cm, and ^{251}Cf. Details of contaminants and specific reasons for discarding each case is given in detail elsewhere.[9]

Our data compare well with Keepin[8] and Cox[10,11]; however, for the long-lived groups our uncertainties are much smaller. This is due to better statistics available from the high fluxes used. The count rate several seconds after irradiation in this work was several hundred times that obtained by Keepin from his multiple irradiations. For the shorter-lived groups this work had larger uncertainties due to the nonpulsing nature of the experiments and the significant transit times.

In all cases, the longest-lived group (group I or ^{87}Br) agrees well with the values quoted elsewhere. For group II the agreement is also excellent except that in the cases of ^{233}U and ^{242}Pu our analysis has separated the ^{137}I and the ^{88}Br contributions into two groups. Beyond group II agreement is not expected unless the same precursors are incorporated into the same groups. The total yield, however, is not affected by the groupings.

A. Fission yields for ^{87}Br and ^{137}I

The groups I and II, the longest in time, are due solely to ^{87}Br for group I and predominantly ^{137}I for group II. The half-life obtained for ^{87}Br from our neutron measurements is 55.23 ± 0.13 sec, which agrees reasonably well with the mean measured value[12] of 55.6 ± 0.2 sec.

The best P_n value for ^{87}Br is $2.38 \pm 0.08\%$.[7] P_n is defined as the fraction of decays which result in neutron emission. By dividing the observed group I yield by 2.38% the cumulative fission yield of ^{87}Br is obtained. The results of this calculation are listed in Table IV. Also included are experimentally measured cumulative yield values listed in Rider and Meek.[6] In some cases the cumulative fission yield of ^{87}Br is not known or uncertain so in these cases the cumulative fission yield of the beta decay daughter ^{87}Kr is included with a less than sign (<) because the yield of ^{87}Kr is definitely greater than that of ^{87}Br.

The corrected group yields were obtained by correcting for the difference between λ_1 and the decay constant for ^{87}Br (0.01247 sec^{-1}). The time at which group I and group II yields are the same were taken as the reference time. At this time the neutrons being observed are those of ^{87}Br. By extrapolating back using the decay constant for ^{87}Br, instead of that for group I. The equations used were

$$Y_{\mathrm{Br}} \exp(-\lambda_{\mathrm{Br}} t_0) = A_1 \exp(-\lambda_1 t_0) \ ,$$

so that

$$Y_{\mathrm{Br}} = A_1 \exp[-(\lambda_1 - \lambda_{\mathrm{Br}}) t_0] \ .$$

The agreement in cumulative yields is excellent except for ^{238}U and ^{241}Pu. The latter case may be

TABLE III. Measured delayed neutron groups.

Nuclide	λ_1	A_1	λ_2	A_2	λ_3	A_3	λ_4	A_4	λ_5	A_5	λ_6	A_6
^{232}Th	0.012 51	0.180 9	0.032 41	0.704	0.132 7	1.330	0.437	2.02	1.79	0.79		
	±0.000 02	±0.006 9	±0.000 12	±0.027	±0.002 5	±0.059	±0.020	±0.12	±0.64[a]	±0.29[a]		
^{232}U	0.012 76	0.052 4	0.035 02	0.131	0.143 9	0.134	0.396	0.113	1.35[b]	0.007		
	±0.000 04	±0.0040	±0.000 29	±0.010	±0.005 9	±0.014	±0.045	±0.012	±0.40	±0.039[b]		
^{233}U	0.012 39	0.055 1	0.025 9	0.070	0.039 8	0.160	0.161	0.175	0.287	0.188	1.32	0.084
	±0.000 04	±0.003 7	±0.001 9	±0.027	±0.002 4	±0.024	±0.010	±0.024	±0.028	±0.030	±0.40	±0.013
^{235}U	0.012 55	0.056 6	0.030 9	0.356	0.114 0	0.346	0.328	0.672	2.06	0.303		
	±0.000 03	±0.001 1	±0.000 1	±0.007	±0.000 1	±0.011	±0.007	±0.018	±0.31	±0.045		
^{238}U	0.012 54	0.048 7	0.030 32	0.557	0.086 91	0.358	0.245 3	1.656	0.705	1.212	2.5	0.82
	±0.000 03	±0.004 0	±0.000 10	±0.042	±0.003 10	±0.035	±0.003 5	±0.140	±0.051	±0.124	±1.1[a]	±0.50[a]
^{237}Np	0.012 58	0.036 8	0.030 60	0.244	0.065 3	0.070	0.139	0.153	0.328	0.424	1.62	0.132
	±0.000 04	±0.003 4	±0.000 34	±0.024	±0.016 0	±0.033	±0.019	±0.065	±0.030	±0.053	±0.69	±0.031
^{238}Pu	0.012 62	0.019 7	0.030 26	0.142	0.085 1	0.052 8	0.197	0.081 5	0.356	0.151	1.35	0.015
	±0.000 13	±0.003 1	±0.000 35	±0.022	±0.012 0	±0.031 0	±0.023	±0.013 0	±0.051	±0.024	±0.40	±0.087[b]
^{239}Pu	0.012 46	0.018 95	0.029 41	0.182 5	0.071 4	0.078 0	0.212	0.158	0.324	0.147	1.28	0.119
	±0.000 01	±0.000 90	±0.000 80	±0.008 9	±0.003 6	±0.008 7	±0.018	±0.031	±0.048	±0.031	±0.25	±0.015
^{241}Pu	0.012 96	0.019 5	0.029 6	0.324	0.066 3	0.086 0	0.196	0.473	0.694	0.598	1.35[b]	0.058
	±0.000 10	±0.001 2	±0.000 2	±0.017	±0.007 9	±0.018 0	±0.009	±0.036	±0.047	±0.035	±0.40	±0.089[b]
^{242}Pu	0.013 40	0.022 2	0.029 5	0.316	0.040 9	0.061 6	0.127 0	0.322	0.397	0.721	2.22	0.523
	±0.000 27	±0.002 7	±0.001 5	±0.104	±0.014 0	±0.009 7	±0.005 6	±0.030	±0.033	±0.071	±0.87	±0.169
^{241}Am	0.012 71	0.018 5	0.029 85	0.146	0.151 9	0.154	0.446	0.154	2.63	0.036		
	±0.000 03	±0.002 2	±0.000 04	±0.018	±0.003 0	±0.019	±0.022	±0.020	±0.11	±0.048		
^{242}Am^{m}	0.012 73	0.017 6	0.030 02	0.195	0.093 0	0.082 2	0.246 2	0.244	0.656	0.119	1.35[b]	0.030
	±0.000 05	±0.001 2	±0.000 11	±0.013	±0.005 4	±0.009 2	±0.006 7	±0.026	±0.083	±0.013	±0.40	±0.045[b]
^{245}Cm	0.013 35	0.013 97	0.030 31	0.179 3	0.104	0.054	0.211	0.174	0.537	0.136	1.35[b]	0.035
	±0.000 09	±0.000 90	±0.001 14	±0.012 0	±0.014	±0.017	±0.011	±0.031	±0.073	±0.016	±0.40	±0.056
^{249}Cf	0.012 851	0.007 65	0.030 37	0.094 0	0.167 8	0.102 0	0.541	0.062 8				
	±0.000 021	±0.000 56	±0.000 01	±0.006 9	±0.003 7	±0.008 6	±0.063	±0.006 9				

[a] This group was modified to fit Keepin's data from $t = 0$ to 0.4 sec.

[b] The decay constant for this group was arbitrarily set at 1.35 sec^{-1} and the group yield was calculated using the corrected absolute yield.

TABLE IV. Calculated cumulative fission yield for ^{87}Br.

Fissioning nuclide	^{87}Br delayed neutron yield per fission	Derived ^{87}Br fission yield %	Rider and Meek[a] recommended yield %
^{232}Th	0.180(7)[a]	7.61(39)	<7.15(20)
^{232}U	0.052(4)	2.81(18)	
^{233}U	0.055(4)	2.31(19)	2.20(13)
^{235}U	0.056(1)	2.35(09)	2.27(14)
^{238}U	0.048(4)	2.02(18)	1.36(44)
			<1.44(4)
^{237}Np	0.036(3)	1.51(14)	<1.73(7)
^{238}Pu	0.019(3)	0.84(13)	
^{239}Pu	0.190(9)	0.80(5)	0.73(4)
^{240}Pu	0.022(3)	0.92(13)	<1.01(16)
^{241}Pu	0.018(1)	0.76(5)	0.61(5)
			<0.80(6)
^{242}Pu	0.019(3)	0.80(13)	<0.86(14)
^{241}Am	0.018(2)	0.76(9)	
^{242}Am^m	0.017(1)	0.71(5)	
^{245}Cm	0.0122(9)	0.51(4)	
^{249}Cf	0.0072(6)	0.30(3)	

[a] Number in parentheses is error $\times 10^3$, e.g., $0.180(7) \equiv 0.180 \pm 0.007$.

due to a problem in the group I yield data reported here. In the case of ^{238}U there is very poor agreement. The problem is not in the group I yield because Keepin's group I yield gives even worse agreement. We suggest that the reported cumulative fission yield in Rider and Meek is in error. In view of the difficulty in making cumulative yield measurements this could be possible. A more severe problem is that the cumulative yield of ^{87}Kr, which must be larger than that of ^{87}Br, is also reported as a lower figure.

B. Fission yield for ^{137}I

A similar, though more complicated, analysis can be made for the yield of ^{137}I from group II data. In general, in group II the major contributor is ^{137}I with smaller contributions from ^{88}Br and ^{136}Te. The contribution of ^{88}Br decreases with increasing fissioning nuclide mass so that in most instances well over 80% of the contributions come from ^{137}I. The contribution of ^{88}Br and ^{136}Te was estimated using the fission yield model described later and was subtracted from the observed group II yield ($T_{1/2} = 23$ sec). The fission yield of ^{137}I is then calculated by dividing by the P_n value of ^{137}I of 6.6 ± 0.6.[7] The results of these calculations are shown in Table V.

Rider and Meek[6] list a few experimentally measured cumulative fission yields which agree well with the values obtained by this analysis. They also list recommended fission yields using calculations where measurements are not available. These values also agree with the values obtained by analyzing delayed neutrons. For several nuclides, however, no report of fission yields has been made and this analysis provides new information.

C. Other values

Among the values reported by Keepin[8] for absolute delayed-neutron yields was the absolute yield of ^{238}U fast fission. His other data agreed with prior and subsequent work but the absolute yield of ^{238}U differed significantly. Work by Evans[13] indicated a higher yield even after corrections for a miscalibration of a ^{99}Mo source was applied. Keepin relied on ^{99}Mo counting to determine the number of fissions. It appears that the same value of ^{99}Mo yield[14] was used for ^{235}U and ^{238}U. The value for ^{238}U is 1.08 ± 0.03 times larger than that for ^{235}U. If his value is corrected by this amount, it agrees with all other published values for ^{238}U fission. Table VI shows the published values and a mean value weighted by the quoted uncertainties of the individual reported values. The values are taken from Tuttle.[15] The yield from fast neutron fission of ^{238}U is 4.44 ± 0.23 neutrons per 100 fissions.

IV. DISCUSSION

A. Parametrization of total delayed neutron yield

For fissioning systems, the yield of beta-delayed neutrons follow the quantity $(A_c - 3Z_c)$, where

TABLE V. Calculated cumulative yield for ^{137}I.

Fissioning nuclide	Group 2 yield %	^{88}Br and ^{136}Te contribution (±20%)	Calculated[a] ^{137}I yield %	Observed % ^{137}I yield	Rider and Meek (Ref. 6) recommended value %
^{232}Th	0.704(27)	0.433	4.1(1.4)	5.15(82)	5.39(59)
^{232}U	0.131(10)	0.0995	0.48(34)		
^{233}U	0.230(36)	0.140	1.4(7)	1.67(10)	1.65(7)
^{235}U	0.358(7)	0.158	3.0(6)	3.46(21)	3.22(19)
^{238}U	0.557(42)	0.162	6.0(1.0)	5.31(85)	
^{237}Np	0.244(24)	0.087	2.4(5)		2.90(67)
^{238}Pu	0.142(22)	0.040	1.5(4)		
^{239}Pu	0.183(9)	0.048	2.0(3)	2.57(21)	2.43(14)
^{240}Pu	0.238(16)	0.059	2.7(4)		2.58(59)
^{241}Pu	0.324(17)	0.065	3.9(5)	3.86(23)	4.13(25)
^{242}Pu	0.316(104)	0.086	3.5(1.6)		3.70(85)
^{241}Am	0.146(18)	0.024	1.8(3)		
$^{242}Am^m$	0.195(13)	0.039	2.4(3)		
^{245}Cm	0.179(12)	0.032	2.2(3)		
^{249}Cf	0.094(7)	0.012	1.25(17)		
^{252}Cf	0.0347(9)	0.020	3.0(3)		2.29(73)

[a] Number in parentheses is error $\times 10^3$, e.g., $0.704(27) = 0.704 \pm 0.027$.

A_c and Z_c are the composite mass and charge of the fissioning material.[16] *A priori* dependence on the mass to charge ratio of the parent nuclide (A_c/Z_c) which is normally about 2.57 might be expected. However, another effect is also observed. Since the heavy fission peak is approximately constant, an increase in A_c causes the light fission yield peak to shift. Delayed-neutron precursors are concentrated in two groups near the light and heavy fission yield peaks ($A = 90$ and $A = 140$). Increasing the mass of the fissioning material, A_c, causes the light fission yield peak to shift away from the light delayed neutron precursors. The result is a decrease in the delayed neutron yield. To compensate for this loss, the mass to charge ratio must be increased by more than 2.57. It is not surprising then that leaving the quantity ($A_c - 3Z_c$) constant leaves the delayed-neutron yield constant. Increasing the quantity increases the delayed-neutron yield exponentially. A least-squares fit of the observed delayed-neutron data in the form[15]

$$Y_{\mathrm{DN}}(\text{per 100 fissions}) = \exp(a + bZ_c + cA_c)$$

was made. It was found that the neutron-induced fission data fits very well, whereas some of the photofission and spontaneous-fission data fit but with greater dispersion. This could be because the quoted uncertainties on these data are too

TABLE VI. Reported ^{238}U delayed-neutron yields (Ref. 14).

Investigator	Neutron energy	Yield (neutrons/100 fissions) (Δy)
Keepin (1957)	Fission	4.12(25)[a]
Tomlinson (1972)	Fission	4.40(12)
Manero and Konshin (1972)	Fission	4.60(25)
Brunson (1955)	2.7 MeV	4.76(74)
Maksyutenko (1959)	2.4 MeV	4.37(35)
Maksyutenko (1959)	3.3 MeV	4.15(38)
Masters *et al.* (1969)	3.1 MeV	4.84(36)
Cox and Whiting (1970)	0.9–2.4 MeV	4.46(29)
Clifford (1972)	1.8 MeV	4.72(25)
Cox (1974)	2.0 MeV	4.39(26)
Cox (1974)	3.0 MeV	4.35(26)
Keepin (adjusted)	Fission	4.45(30)
This work (1980)	Fission	4.65(35)

[a] Number in parentheses is error ×100, e.g., $4.12 \pm 0.25 = 412(25)$.

small or that these are somewhat different processes.

The least-squares fit of the available data (excluding ^{237}Np photofission, ^{234}U photofission, and ^{252}Cf spontaneous fission) gives the following expression with an accuracy of $\pm 9\%$:

$$Y_{\mathrm{DN}}(\text{per 100 fissions}) = \exp(16.698 - 1.144 Z_c + 0.377 A_c) \; .$$

Figure 4 is a plot of the measured delayed-neutron yields versus Y_{DN} that have been reported in this work or elsewhere. Correlations such as this are quite useful in estimating delayed neutron yields for unmeasured nuclides. For example, the contribution of ^{238}Pu fission or ^{236}U fission in reactors with these minor contributors can be estimated using such a correlation. If actinide burning reactors are ever designed, unmeasured delayed-neutron yields will have to be estimated in this way.

The time dependent nature of the beta delayed neutrons can be estimated. In Fig. 5 we have shown the relative delayed-neutron yield (normalized to unity) with time for all avaiable data.

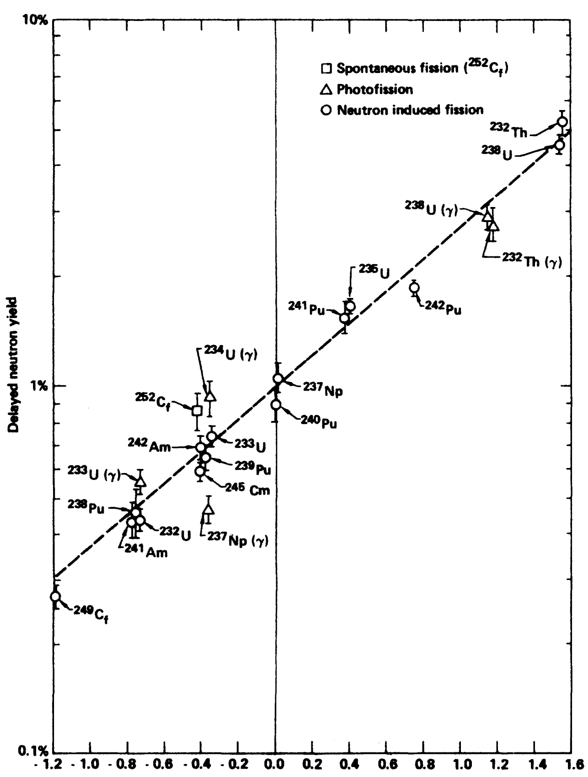

FIG. 4. Plot of the total delayed neutron yield for various nuclides versus the quantity $(16.698 - 1.144 Z_c + 0.3769 A_c)$, where Z_c and A_c are the composite charge and mass of the fissioning nuclide.

From this plot we can calculate the uranium equivalent mass for all the nuclides studied where the uranium equivalent mass of a nuclide of mass A and charge Z is $92(A/Z)$. If this quantity is calculated for each nuclide, an orderly progression exists from ^{232}U to ^{238}U including the non-uranium nuclides. Thus, it is possible to not only estimate the total delayed-neutron yield for a given nuclide, but the time dependent nature of the delayed neutrons as well. Reactors utilizing recycled fuel or burning actinides are likely to have inventories of fissioning nuclides which have not been studied. The delayed neutrons from these nuclides can have a perturbing effect on the stability of such reactors.[17]

B. Generalized fission yield model

Generally independent yields can be fit to a Gaussian distribution[6]; that is, the relative independent yield is divided by the chain yield and plotted as a function of fission product for a given mass. The resulting Gaussian has been found to have a width parameter of $\sigma_z = 0.56$ while its centroid is *quoted* by the parameter Z_p.[2] Recent studies indicate a value of 0.53 may be more exact. For a fission product of mass A and charge

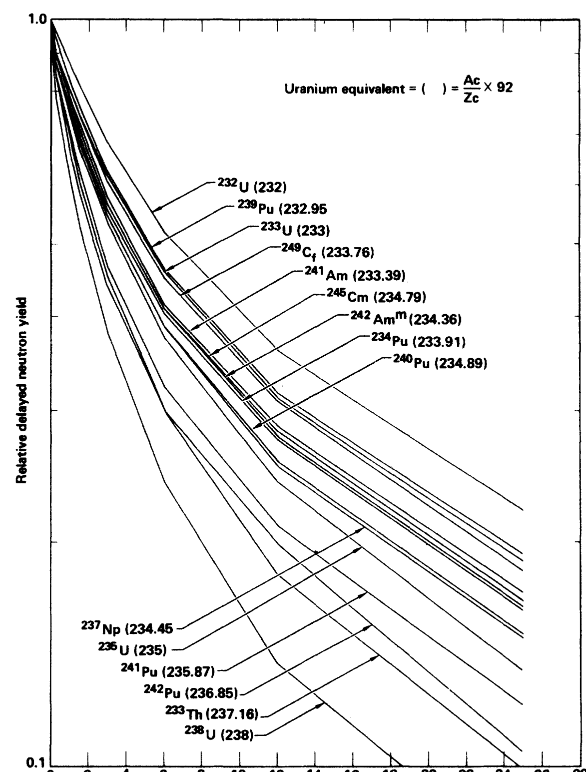

FIG. 5. Plots of the relative delayed neutron yield for various nuclides with time.

Z the relative independent yield (RIY) is given by the equation

$$RIY = C(1+a)\int_{Z-0.05}^{Z+0.5} \exp\left[-\frac{(Z-Z_p)^2}{2\sigma_z^2}\right]dz\ ,$$

where C is a normalizing constant (so that the total of all relative independent yields in a chain is unity) and a parametrizes the even-odd effect. If the variable X is defined by the equation

$$X = \frac{Z-Z_p}{\sigma_z}\ ,$$

then $F(X)$ may be defined as the integral quantity above so that

$$RIY = (1+a)F(X)\ .$$

$F(X)$ values are tabulated by Bevington.[18] Independent yields may be calculated by multiplying the relative value by the chain yield and the cumulative yield may be obtained by summing from $z=0$ to $z=Z$, where Z is the charge of the nuclide of interest.

In the case of the even-odd effect, if the charge is even, a is positive and if it is odd, a is negative. For thermal fission of ^{233}U and ^{235}U, for instance, the even-odd effect is about 22%.

An accurate formula for Z_p would allow the calculation of fission yields for any fissioning nuclide and any fission product of interest. Sufficient experimental data[16] are available to calculate Z_p values for thermal fission of ^{233}U and ^{235}U. A plot of the resulting values as a function of mass is shown in Fig. 6. In calculating these values, the even-charged nuclides result in Z_p values that are on the average 0.11 charge units less than the average. Odd-mass nuclides give a Z_p of 0.11 charge units larger than the average value Z_p. We have parametrized the quantity Z_p for U^{235} as follows:

$$Z_p = 0.4153A - 1.19,\ A < 116$$

and

$$Z_p = 0.4153A - 3.43,\ A > 116\ .$$

For thermal fission of ^{233}U, the equation has the form

$$Z_p = 0.4153A - 0.856,\ \ A < 116$$

and

$$Z_p = 0.4153A - 2.94,\ \ A > 116.$$

A least squares fit of the value listed in Rider and Meek[6] gives essentially the same result. We assumed that deviations of Z_p for other nuclides would only depend upon the composite mass to charge ratio; hence, for any composite system of mass A_c and charge Z_c we used the following:

$$\overline{Z}_p = 0.4153A - 1.19 + 0.167\left(236 - 92\,\frac{A_c}{Z_c}\right),\ \text{for } A < 116$$

and

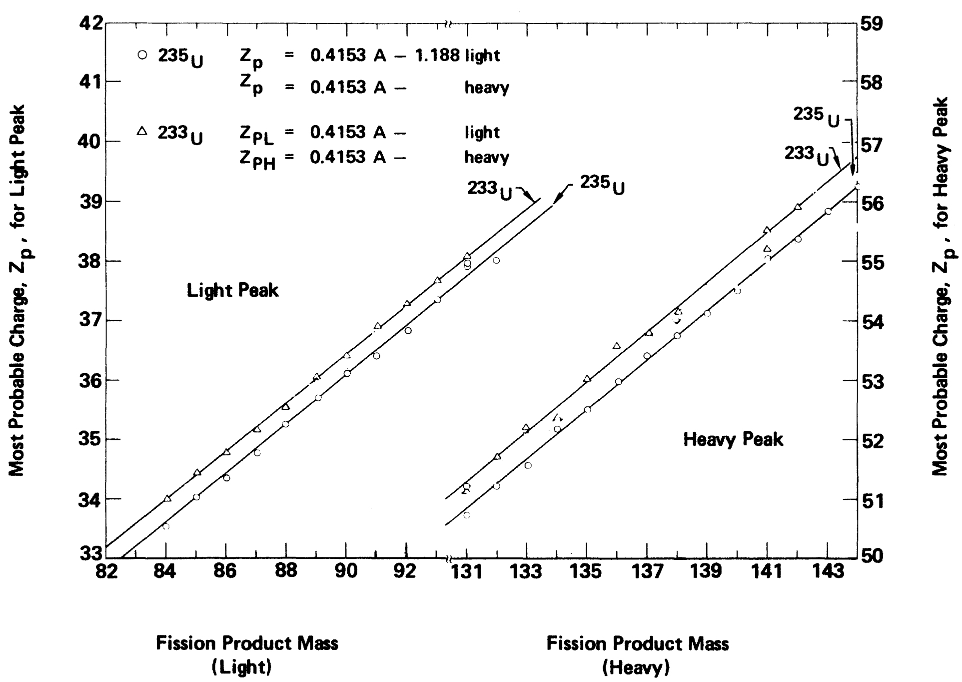

FIG. 6. Plot of Z_p values versus mass for ^{233}U and ^{235}U for light and heavy fission products.

$$\bar{Z}_p = 0.4153A - 3.43 + 0.243\left(236 - 92\frac{A_c}{Z_c}\right), \quad \text{for } A > 116.$$

The values chosen fit the observed values for ^{233}U and ^{235}U. Thus, for a fissioning material of mass A_c and charge Z_c the fission yield of a fission product of mass A and charge Z can be calculated. No attempt has been made to insert an even-odd effect which is the major difference between the approach used here and that used by Rider and Meek. Their formulation for Z_p was derived by using a correlation reported by Nethaway.[5]

We used the cumulative fission yield model described above and the most current p_n and half-life values for all the known delayed-neutron precursors to calculate not only the total delayed-neutron yield from fission but the time dependence of this yield. The p_n and half-life values we used here are given in Table VII and were taken from Rudstam[7] and from Rider and Meek.[6] The results of these calculations are summarized in Table VII. Not only is the calculated absolute yield compared to measured values, but the relative yield (normalized to unity) at several times is also compared to observed values after a continuous irradiation.

The even-odd effect is expected to be large in nonfissile systems such as ^{232}Th, ^{238}U, ^{240}Pu, and ^{242}Pu. Indeed, the measured yields for the plutonium isotopes do appear lower than calculated. However, for ^{232}Th and ^{238}U the measured yield does not indicate a significant even-odd effect. The odd Z nuclides ^{237}Np, ^{241}Am, and $^{242}Am^m$ should not have an even-odd effect and fission-yield measurements on ^{239}Pu and ^{241}Pu seem to show no effect either.[19] In general, the model does an excellent job predicting the total yield in all these cases. The only area where there is poor agreement is at very large masses (for ^{245}Cm, and ^{249}Cf.

C. Comparison of experimental and calculated total yields

The only comprehensive attempt at calculating delayed-neutron yields for a variety of nuclides has been the work of Rider and Meek.[6] The approach used here differs in that we use more current p_n values and our model for Z_p is different. Since Rider and Meek included a postulated even-odd effect in each calculation, we compare the experimentally determined values with those calculated by Rider and Meek in Table VIII. Also included is a calculation using Nethaway's correlation (used by Rider and Meek) but without the even-odd effect. For a Z_p correlation Nethaway uses

$$Z_p(Z_c, A_c, E^*) = Z_p(92, 236, 6.52) + a(Z_c - 92) + b(A_c - 236) + c(E^* - 6.52),$$

where for the light mass fission fragments

$a = 0.414 \pm 0.016$,

$b = -0.143 \pm 0.007$,

and

$c \simeq 0.0174$,

and for heavy fission fragments

$a = 0.547 \pm 0.010$,

$b = -0.188 \pm 0.004$,

and

$c \simeq 0.051 - 0.0023(A_H - 130)$,

where A_H is the mass of the heavy fission fragment, and Z_c, A_c, and E^* are the composite charge, mass, and excitation energy of the fissioning system.

In general, the model used in this work gave the best agreement with experimental data. Poor agreement was found only for ^{242}Pu, ^{245}Cm and ^{249}Cf. In these cases the calculated yields were too high, possibly because of a large even-odd effect in those nuclides. An indication of this is that the Nethaway correlation, which has no even-odd correction, gave worse agreement. Although the Rider and Meek results agree reasonably well, they gave poor agreement for ^{232}Np and ^{242}Pu. Again, in the case of ^{242}Pu a large even-odd effect was assumed and perhaps the real effect is smaller. It would appear the large effects assumed for nonfissile nuclides do not in reality exist. This only points out the fact that the even-odd effect is poorly understood.

D. Time dependent delayed neutron yields

Time dependent delayed neutron yields depend upon two separate quantities. First of all, experiments measure the time dependence of delayed neutrons normalized to unity. This is the relative delayed neutron yield with time. Secondly, the absolute delayed neutron yield must be measured.

The most accurate relative delayed neutron yields are those reported by Keepin,[8] Cox,[10,11] and in this work. The values reported in this work are more accurate several seconds after irradiation, but for times immediately after irradiation Keepin's and Cox's values are more accurate. Indeed the values for ^{232}Th and ^{238}U reported here were developed using Keepin's values for short times after irradiation. For this reason

TABLE VII. Delayed neutron parameters (P_n and half-life values) used in this work. (Refs. 6 and 7).

Precursor Charge	Precursor Mass	P_n (%)	Half-life (sec)	Precursor Charge	Precursor Mass	P_n (%)	Half-life (sec)
35	87	2.38	55.6	39	98	3.4	2.0
55	137	6.6	24.9	43	109	1.7	2.0
53	137	6.6	24.4	35	90	21.2	1.92
52	136	0.9	17.5	32	83	0.17	1.9
35	88	6.7	16.0	42	110	1.3	1.892
41	103	0.13	15.669	36	92	0.033	1.85
51	134	0.108	10.4	41	105	2.9	1.8
56	147	5.2	10.0	56	150	0.24	1.798
53	138	5.3	6.53	55	143	1.68	1.78
37	93	1.39	5.85	54	141	0.044	1.73
33	84	0.090	5.6	55	142	0.091	1.71
34	87	0.190	5.6	51	135	15.6	1.71
37	92	0.012	4.5	31	80	0.8	1.66
35	89	13.5	4.38	34	88	0.6	1.52
40	104	0.11	3.783	47	122	1.4	1.5
39	97	0.06	3.7	50	133	0.02	1.47
57	149	0.81	2.864	52	138	6.3	1.4
31	79	0.094	2.86	39	99	1.2	1.4
52	137	2.50	2.8	36	93	1.96	1.29
37	94	10.4	2.76	54	142	0.42	1.24
30	79	1.1	2.74	31	81	11.9	1.23
49	129	3.5	2.5	32	84	10	1.20
53	139	9.42	2.38	38	100	5	1.046
56	147	5.2	2.23	50	134	17	1.04
41	104	0.71	1.0	52	139	6.3	0.424
54	144	0.73	1.0	34	89	5	0.41
56	149	0.03	0.917	47	123	4.6	0.39
33	86	12.00	0.9	37	95	8.8	0.384
49	128	0.057	0.84	35	92	22	0.362
43	110	0.10	0.83	55	146	13.2	0.335
48	128	0.11	0.83	53	143	18	0.328
51	136	23.00	0.82	31	83	56	0.31
38	98	0.36	0.8	54	143	1.2	0.30
39	100	5.50	0.756	50	135	8.6	0.291
33	87	44.00	0.73	51	137	20	0.284
57	150	0.94	0.648	49	131	1.73	0.28
31	82	21.90	0.6	34	91	21	0.27
53	140	23.00	0.6	32	86	22	0.259
38	99	3.40	0.6	32	85	20	0.234
55	145	13.3	0.585	55	147	25.4	0.21
49	130	1.38	0.58	36	94	5.7	0.208
40	105	1.40	0.559	37	96	14.2	0.201
34	90	11.00	0.555	35	93	41	0.201
35	91	10.9	0.542	53	142	16	0.196
41	106	5.5	0.535	37	97	28	0.17
36	95	9.5	0.5	49	132	4.3	0.13
56	148	23.9	0.5	37	98	16	0.119
53	141	39	0.47	37	99	15	0.076
38	97	0.27	0.43				

and because normal uses of such data include times immediately after irradiation, Keepin's and Cox's relative values should be used where available.

Absolute delayed neutron yields have been measured by many experimenters for the import nuclides. Many of these are more accurate than Keepin's absolute yields. For this reason the ab-

TABLE VIII. Comparison of experimental and calculated total yields.

Nuclide	Observed yield	Calculated yield this work %	Rider and Meek yield %	Nethaway correlation %
^{232}Th	5.27(40)[a]	5.24	4.66	5.98
^{232}U	0.44(3)	0.45		0.75
^{233}U	0.74(4)	0.79	0.83	1.11
^{235}U	1.67(7)	1.67	1.72	2.02
^{238}U	4.60(25)	4.43	3.31	4.06
^{237}Np	1.07(10)	1.04	1.22	1.29
^{238}Pu	0.46(7)	0.43		0.55
^{239}Pu	0.65(5)	0.68	0.74	0.72
^{240}Pu	0.90(9)	1.05	0.86	1.11
^{241}Pu	1.57(15)	1.57	1.51	1.43
^{242}Pu	1.86(9)	2.46	1.33	1.84
^{241}Am	0.44(5)	0.45		0.48
^{242}Am^m	0.69(5)	0.69		0.62
^{245}Cm	0.59(4)	0.75		0.56
^{249}Cf	0.27(2)	0.36		0.20
^{252}Cf(*sf*)	0.86(10)	0.86	0.63	0.67
^{238}U(γ,*f*)	2.91(9)	3.25		
^{235}U(γ,*f*)	1.04(4)	1.16		

[a] Number in parentheses is error $\times 10^2$, e.g., $5.27(40) = 5.27 \pm 0.40$.

solute yields which should be used are those reported in the Evaluated Nuclear Data Files B, version IV (ENDF/B-IV).[6] Thus, absolute group yields can be obtained by multiplying relative yields by the ENDF/B-IV absolute yields. Using these criteria the group parameters listed in Table IX are recommended values.

V. CONCLUSION

We conclude that total delayed-neutron yields can be expressed in terms of a simple model that is accurate for a large variety of nuclides from ^{232}Th to ^{252}Cf. The model does hold in the previously unmeasured region between ^{241}Pu and ^{252}Cf. Secondly, the time dependent decay of delayed neutrons can also be expressed. Nuclides with similar A_c/Z_c ratios have similar relative decay patterns. Thus, the relative decay pattern for one nuclide may be estimated by another measured nuclide with a similar mass to charge ratio.

Most important, we find that it is possible to accurately reproduce the observed yield and decay characteristics of delayed neutrons using a simple fission-yield model and known precursor characteristics. Cumulative fission yields for ^{87}Br and ^{137}I have been measured directly by studying the group-wise decay of delayed neutron emission for a large number of exotic nuclides in a nondestructive fashion. In addition, we have compared all reported delayed neutron data and have presented a recommended set. For work where short times or short irradiations are unimportant, we find our results to be the most applicable.

Finally, delayed-neutron studies indicate that the even-odd effect is not yet well understood. Nuclides such as ^{232}Th and ^{238}U were supposed to have large even-odd effects and seem instead to have very small effects. Estimates of the size of the even-odd effect have been made for a large variety of nuclides.

The delayed neutrons from neutron-induced fission in ^{232}U, ^{237}Np, ^{238}Pu, ^{241}Am, ^{242m}Am, ^{245}Cm, and ^{249}Cf were studied for the first time; those from ^{232}Th, ^{233}U, ^{235}U, ^{238}U, ^{239}Pu, ^{241}Pu, and ^{242}Pu were measured again. The data were used to develop an expression for the prediction of the absolute delayed neutron yield, and the prediction of delayed neutron emission with time.

TABLE IX. Recommended delayed neutron group parameters.

	λ_1	A_1	λ_2	A_2	λ_3	A_3	λ_4	A_4	λ_5	A_5	λ_6	A_6	A Total
^{232}Th	0.012 4	0.019 0	0.033 4	0.836	0.121	0.864	0.321	2.49	1.21	0.958	3.29	0.239	5.27
	±0.002	±0.013	±0.001 1	±0.042	±0.005	±0.121	±0.011	±0.12	±0.090	±0.082	±0.30	±0.035	±0.40
^{232}U	0.012 76	0.052 4	0.035 02	0.131	0.143 9	0.134	0.396	0.113	1.35[a]	0.007[a]			0.437
	±0.000 04	±0.004 0	±0.000 29	±0.010	±0.005 9	±0.014	±0.045	±0.012		±0.039			±0.033
^{233}U	0.012 6	0.064	0.033 7	0.221	0.139	0.186	0.325	0.206	1.13	0.038	2.50	0.025	0.74
	±0.000 3	±0.003	±0.000 6	±0.010	±0.006	±0.030	±0.030	± 0.018	±0.40	±0.018	±0.42	±0.010	±0.04
^{235}U	0.012 7	0.055	0.031 7	0.366	0.115	0.328	0.311	0.660	1.40	0.192	3.87	0.070	1.67
	±0.000 3	±0.005	±0.001 2	±0.038	±0.004	±0.038	±0.012	±0.027	±0.12	±0.016	±0.55	±0.008	±0.07
^{238}U	0.013 2	0.057 7	0.032 1	0.608	0.139	0.719	0.358	1.72	1.41	1.00	4.02	0.33	4.44
	±0.000 3	±0.004	±0.006	±0.009	±0.005	±0.089	±0.014	±0.05	±0.07	±0.06	±0.21	±0.02	±0.23
^{237}Np	0.012 58	0.036 83	0.030 6	0.244	0.065 3	0.070	0.139	0.153	0.328	0.424	1.62	0.132	1.068
	±0.000 04	±0.003 4	±0.000 34	±0.024	±0.016	±0.033	±0.019	±0.065	±0.030	±0.053	±0.69	±0.031	±0.098
^{238}Pu	0.012 62	0.019 7	0.030 26	0.142	0.085 1	0.052 8	0.197	0.081 5	0.356	0.151	1.35[a]	0.015[a]	0.461
	±0.000 13	±0.003 1	±0.000 35	±0.022	±0.012	±0.031	±0.023	±0.013	±0.051	±0.024		±0.087	±0.073
^{239}Pu	0.012 8	0.022	0.030 1	0.192	0.124	0.136	0.325	0.210	1.12	0.055	2.69	0.029	0.645
	±0.000 5	±0.006	±0.002 2	±0.024	±0.009	±0.032	±0.036	±0.023	±0.39	±0.019	±0.48	±0.011	±0.05
^{240}Pu	0.012 9	0.022	0.031 3	0.238	0.135	0.162	0.333	0.315	1.36	0.119	4.03	0.029	0.88
	±0.000 3	±0.003	±0.000 5	±0.016	±0.11	±0.044	±0.030	±0.027	±0.20	±0.018	±0.77	±0.006	±0.06
^{241}Pu	0.012 8	0.015 6	0.029 9	0.357	0.124	0.279	0.352	0.608	1.61	0.284	3.47±	0.025	1.57
	±0.000 2	±0.004 7	±0.000 6	±0.009	±0.013	±0.039	±0.018	±0.078	±0.15	±0.030	1.70	±0.008	±0.15
^{242}Pu	0.013 4	0.022 1	0.029 5	0.316	0.040 9	0.061 6	0.127	0.322	0.397	0.721	2.22	0.523	1.97
	±0.000 27	±0.002 7	±0.001 5	±0.104	±0.014	±0.097	±0.005 6	±0.030	±0.033	±0.071	±0.87	±0.169	±0.23
^{241}Am	0.012 71	0.018 5	0.029 85	0.146	0.152	0.154	0.446	0.154	2.63±	0.036			0.509
	±0.000 03	±0.002 2	±0.000 04	±0.018	±0.003	±0.019	±0.022	±0.020	2.11	±0.048			±0.060
^{242}Am^m	0.012 73	0.017 6	0.030 0	0.195	0.093	0.082 2	0.246 2	0.244	0.656	0.119	1.35[a]	0.030[a]	0.688
	±0.000 5	±0.001 2	±0.000 11	±0.013	±0.005 4	±0.009 2	±0.006 7	±0.026	±0.083	±0.013		±0.045	±0.045
^{245}Cm	0.013 35	0.013 97	0.030 31	0.179 3	0.104	0.054	0.211	1 0.174	0.537	0.136	1.35[a]	0.035[a]	0.592
	±0.000 09	±0.009	±0.000 14	±0.012	±0.014	±0.017	±0.011	±0.031	±0.073	±0.016		±0.056	±0.039
^{249}Cf	0.012 851	0.007 65	0.030 37	0.094 35	0.167 8	0.102	0.541	0.062 8					0.267
	±0.000 021	±0.000 56	±0.000 04	±0.006 9	±0.003 7	±0.008 6	±0.063	±0.006 9					±0.019
^{252}Cf	0.034 7	0.22	0.35	0.29	1.4±	0.35							0.86
	±0.000 9	±0.01	±0.07	±0.04	1.1	±0.10							±0.10

[a] Decay constant of 1.35 assumed.

This approach accurately predicts observed delayed neutron yields and decay characteristics. A fission product yield model was used in conjunction with delayed neutron emission probability to analytically predict delayed neutron characteristics. The results of this analysis are in excellent agreement with experimental values.

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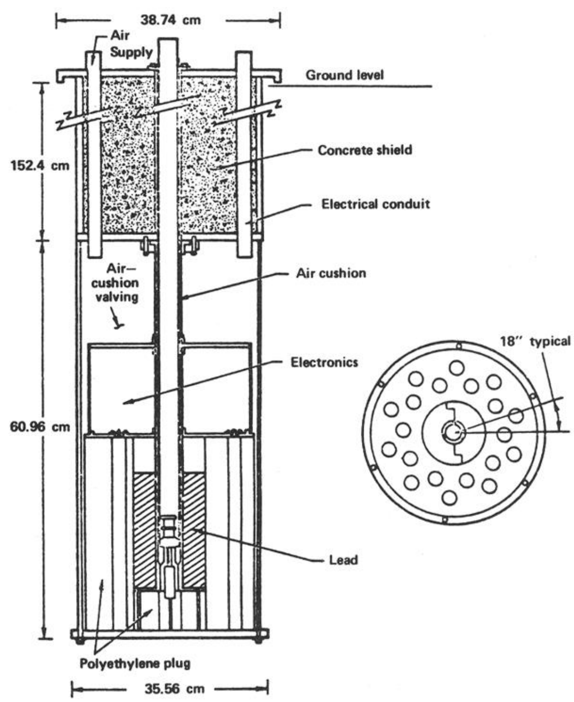

FIG. 2. Diagram of the delayed-neutron detector (taken from Lawrence Livermore Lab Report No. UCID 16911-76-3).