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DELAYED NEUTRONS AND SYMMETRIC FISSION

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DELAYED NEUTRONS AND SYMMETRIC FISSION

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ABSTRACT

It is shown that at $E_n = 15$ MeV, a significant contribution to the delayed neutron yields is made by fragments from the symmetric fission region, and in particular by ^{128}In. The distribution parameters of the fragment isotopes at this energy are calculated and the value of P_n for ^{128}In is determined from the decay of the delayed neutron activity. The ratios of the cumulative yields of seven pure precursors at this energy are calculated.

It was assumed until recently that the precursors of delayed neutrons were fragments belonging to the maximum yields in the doubly-peaked mass distribution curve. Rudstam [1] has now identified precursor fragments with masses of 123-130, belonging to the symmetric-fission region. However, the detection of such fragments against the background of others coming from the maximum yields by an analysis of the decay in activity is of course only possible when the neutrons causing the fission have a high energy and the contribution of the symmetric-fission fragments becomes significant.

Experimental data on the relative yields from fission by 15 MeV neutrons appeared a long time ago [2] and have since been repeated. Table 1 shows the experimental results from Refs [2-4] for ^{238}U. It is clear that there is good agreement. For our calculations here we chose the data from Ref. [4] since they also include the group of shortest-lived precursors.

From the relative yields and half-lives [4], we made a computer calculation of the decay in delayed neutron activity for ^{235}U ($E_n = 15$ MeV and thermal neutrons) and ^{238}U (at E_n = 15 MeV). We then determined the density distribution of the probability P(T) of delayed neutron yields as a function of the half-life T. The distributions for ^{235}U are shown in Fig. 1. The considerable discrepancy between the curves arises from the fact that in the range 4-24 s, the width of the distribution is greater for E_n = 15 MeV than it is for thermal-neutron fission and the maximum is shifted towards shorter half-lives. The distributions also differ markedly in the range 0-0.8 s (Fig. 2). The change in the probability density distribution between fission by thermal neutrons and fission by neutrons with E_n = 15 MeV becomes more obvious when we consider the ratio of the P(T) values for the two cases (see Fig. 3); the increase in the ratio at T = 11 s is particularly apparent in this plot. The arrows in Fig. 3 indicate the positions of the delayed neutron precursors and it can be seen that the increased yield can be ascribed to either ^{134}Sb or ^{128}In. However, the contribution from ^{134}Sb is extremely small because of the very low value (0.08%) of P_n (the probability of delayed neutron emission) and a change in the yield of the chain or a shift in the charge cannot lead to a significant increase in the ^{134}Sb yield (compared to that of precursors with similar half-lives). We can therefore only assume that the reason for the change in the distribution is the increase in the delayed-neutron yield from ^{128}In, i.e. a rise in the overall yield of mass 128.

The next step is therefore to expand the calculated delay curves into a greater number of exponential functions (with given half-lives) and to determine the contribution of ^{128}In and the other precursors in this range where they are comparatively few in number and the half-lives differ significantly from each other (long-lived precursors). We solved this problem by the method of least algebraic deviation [5], using a fast iteration process [6]. In view of the nearly equal half-lives of ^{138}I (6.3 s) and ^{93}Rb (5.9 s), we distinguished only a single group with T = 6.3 s. Half-lives shorter than 4.55 s (^{89}Br) were taken at equal intervals (2.5, 1.5, 1.0 and 0.5 s) since it is impossible to separate out the subsequent precursors owing to their large number and similar half-lives and anyway they are not required in the following analysis. The results of the expansion are given in Tables 2 and 3. Next to the results column, we show the order of magnitude (in %) of the errors obtained from the expansion of the decay curves to the appropriate statistical accuracy.

The further analysis consisted in using these data to get information on the parameters of the fission process and certain characteristics of the delayed neutron precursors and also in checking the consistency of these results.

The procedure was as follows:

1. From the relative yields y_i of three fragments (bromine isotopes with masses of 87, 88 and 89), we determined the parameters of the Gaussian distribution — the width σ_z and the position of the maximum A_p:

$$\sigma_z = (\ln Q_2^2 / Q_1 Q_3)^{-\frac{1}{2}} \qquad (1)$$

$$A_p = 87{,}5 + \sigma_z^2 \ln Q_2 / Q_1 , \qquad (2)$$

where Q_1, Q_2 and Q_3 are the cumulative yields of the corresponding bromine isotope precursors and $Q_i = y_i/P_{ni}$.

The values of P_{ni} were taken from Ref. [7].

2. From the value of $A_p(\mathrm{Br})$ found in this way, we determined $A_p(\mathrm{Rb})$:

$$A_p(\mathrm{Rb}) = A_p(\mathrm{Br}) + \frac{Z(\mathrm{Rb}) - Z(\mathrm{Br})}{K_\ell} \qquad (3)$$

where Z is the charge of the fragment and $k_\ell = \partial Z_p / \partial A$ was found from the data in [8].

3. This value of $A_p(\mathrm{Rb})$ was used to find $y(^{93}\mathrm{Rb})/y(^{87}\mathrm{Br})$ from the relationship:

$$2\sigma_z^2 \ln \frac{y(^{93}\mathrm{Rb})}{y(^{87}\mathrm{Br})} \, \frac{P_n(^{87}\mathrm{Br})}{P_n(^{93}\mathrm{Rb})} \, \frac{M(87)}{M(93)} = \qquad (4)$$

$$= [87 - A_p(\mathrm{Br})]^2 - [93 - A_p(\mathrm{Rb})]^2$$

Here, M (the yield of the chain as taken from Ref. [10]) appears as a normalizing factor.

4. We next determined $A_p(I)$ from a relationship similar to Eq. (4), using the ratio of the delayed neutron yields $y(^{137}I)/y(^{87}Br)$ and found the corresponding values of P_n and $M(A)$, taking equal distribution widths for the light and heavy fragments.

5. Using the value of $A_p(I)$, we found from Eq. (4) the delayed-neutron yield ratio $y(^{138}I)/y(^{137}I)$ and then $y(^{138}I/y(^{87}Br)$.

6. From relationship (3), with the appropriate values for the heavy fragments iodine and indium, we determined $A_p(In)$.

7. The value of P_n for ^{128}In was found from the yields of the eleven-second group by means of Eq. (4).

The values of the various physical quantities found from this analysis are listed in Table 4. We should add the following comments about the errors. The error in the value of P_n obtained by averaging two independent determinations includes all possible errors, both those which depend on the measurement of the decay in activity and those which arise from inaccuracies in the values of P_n used for other precursors. From this point of view, the average which we obtain,

$$P_n(^{128}In) = 2.60 \pm 0.07$$

has (though it may be fortuitous) an extremely small error (the value is usually 10-20%). The error in σ_z can be estimated as 20-30%.

We shall now consider the physical results obtained from the analysis.

The data show that the change in the relative (and hence in the absolute) delayed neutron yields with increase in the energy of the neutrons causing the fission is a result not only of the shift in the charge (mass) and the change in the width of the fragment distribution but also (as we pointed out earlier in Ref. [9]) of the change in the yield of the chains which occurs for the symmetric-fission region, whose contribution is extremely small at low energies but becomes significant and comparable to the asymmetric fission yields at high neutron energies. One of the contributing isotopes from this region is ^{128}In, with a half-life of 11 s.

This same effect can also be seen by comparing the probability density distributions (see Fig. 2) in the short-lived precursor region (0–0.8 s), although it is unfortunately not possible here to separate out the individual contributors. With the shortening of the beta-decay chains which takes place at higher neutron energies, the reduction in the fragment isotope yields (compared to that of ^{87}Br) is greater at higher mass numbers. This should lead to the almost total disappearance of the contributions to the 0–0.8 s group from the iodine, bromine and rubidium isotopes, which are the main contributors at low energies. When $E_n = 15$ MeV, we can expect the main contribution to this group to come from fragments from the symmetric fission region, namely ^{123}Ag with a half-life of 0.39 s. Thus, as we go from fission by low-energy neutrons to fission with $E_n = 15$ MeV, the composition of the short-lived precursors can alter completely. It follows from this that satisfactory agreement between the calculated and measured total absolute yields [7] at $E_n = 15$ MeV found on the assumption that the composition of the contributors remains constant and with no allowance made for the significant ^{128}In yield does not prove that the calculation has a correct physical basis.

We shall now discuss the results for the second group of data derived in this study, namely the mass distribution parameters of the fragment isotopes (see Table 4). The increase in the width of the distribution at the higher neutron energy is natural because already at $E_n = 15$ MeV, three nuclei are undergoing fission and we therefore get the sum of three Gaussian curves. As can be seen for the case of ^{235}U, the shift that we have found in the maximum of the mass distribution curve (15 MeV from the thermal-neutron position) is such that the fragment approaches the stability line at the higher energy. The data in Ref. [10] show a shift in the charge of the light fragments from ^{235}U fission (this is given in mass units in Table 4, $\Delta A_p = \Delta Z_p/k_\ell$) in the opposite direction; this seems to be incorrect. The discrepancy in this parameter between our data and those in [10] amounts to as much as ~ 1 amu. The total shift in the distribution maximum for light and heavy fragments from our results is (in the case of ^{235}U)

$$A_{pl} + A_{ph} = -2.51 \text{ amu}$$

which (in view of the accuracy of the initial data) is quite close to the total change

$$\Delta\nu = 2.11 \text{ amu.}$$

This once again confirms the correctness of our results.

We may further note that the shift in the distribution maximum for heavy fragments ($\Delta A_{ph} = -1.90$) is considerably greater than that for light fragments ($\Delta A_{pl} = -0.61$) and this means that the $Z(A_p)$ lines for the light and heavy fragments move closer together. The correlation between the increase in the symmetric-fission yield and the convergence of these lines has already been noted in Ref. [12].

Finally, we turn to the third group of data obtained from our analysis, namely the ratios of the cumulative yields of the fission fragments. The method used to find these ratios (dividing the delayed neutron yields from each precursor by the corresponding value of P_n) has the advantage that the relative yields of a whole group of fragments are obtained in a single experiment which does not involve monitoring of the flux, chemical separation of the short-lived fragments and so on (processes which lead to additional errors). An improvement in the accuracy of the P_n values leads to even greater accuracy in these results. We may also note that it is difficult to obtain such data for fast-neutron fission by other methods because the yields of the fission products in question are small. This means that these quantities can be calculated only if various hypotheses are resorted to (that the beta-decay chains are of equal length, for example) and additional relationships are introduced (values are assumed for the partial contributions from the members of the fissionable-nuclei chain and for the width of the distribution). In our present calculations, we have used only two rather simple assumptions: (a) the cumulative yields of the fragment isotopes have a Gaussian distribution; (b) the width of this distribution is the same for the light and heavy fragments.

Since Meek and Rider [10] give the cumulative fragment yields and our results represent the yields of the delayed neutrons from these fragments, we have (for the purposes of comparison) multiplied the fragment yields by the corresponding values of P_n. The final values are compared in Tables 2 and 3 where the difference between Ref. [10] and our results are illustrated by the y_i/y_i' column. In many cases, the difference exceeds the attributed limits of error. There is, however, a simple way of checking whether the results are correct. The total integrated delayed neutron yield from the precursors ^{137}I, ^{88}Br and ^{128}In represents the yield of the second group in a six-group description of the decay in activity and the yield of the third group is determined by the precursors ^{138}I, ^{93}Rb and ^{89}Br. In Table 5, we compare the integrated yields of these groups for the data from Refs [4] and [10] and the present paper. It can be seen that our data are much closer than those of Meek and Rider [10] to the original values in Ref. [4].

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Table 1

Relative delayed neutron yields of ^{238}U at E_n = 15 MeV

Group No.	Ref. [2]	Ref. [3]	Ref. [4]
I	I	I	I
2	7,64 ± 0,47	7,4 ± I,5	7,6 ± I,4
3	7,8 ± I,0	8,9 ± I,5	7,4 ± I,4
4	I8,7 ± I,I	I8,2 ± 3,I	23,6 ± 4,4
5	I0,4 ± I,0	8,4 ± 4,0	II,4 ± 2,I
6	-	-	7,2 ± I,4

Table 2

Relative delayed neutron yields of ^{238}U at E_n = 15 MeV

T (s)	Precursor	y_i (this paper)		Error (%)	y_i' [IO]	y_i/y_i'	Q_i [IO]	P_n(%)	M(A) [IO]
54,59	^{87}Br	I		I	I	I	I	2,34	I,733
24,62	^{I37}I (^{I4I}Cs, ^{I34}Sn, ^{I36}Te)$^{+}$	4,60		I	2,76	I,67	I,45	4,46	5,I46
I5,88	^{88}Br	2,64		I	I,48	I,78	0,765	4,54	
II,0	^{I28}In(^{I34}Sb)$^{+}$	I,44		2	(0,68I)^{x}	2,II	0,256	(2,66)§	I,9I5
6,3	^{I38}I		I,2I	5	I,48	0,79			5,I03
	^{93}Rb	2,I5	$\underline{2,I4}$		$\underline{I,25}$				4,4I8
			3,35		2,73				
4,55	^{89}Br	4,52		5-I0	2,60	I,74	0,762	8,0	-

\+ Secondary (not-dominant) contributors are given in brackets;

x Assuming that P_n = 2.66%;

§ Calculated in this paper.

Table 3

Relative delayed neutron yields of ^{235}U at E_n = 15 MeV

T (s)	Precursor	y_i (this paper)	Error (%)	y_i' [IO]	y_i/y_i'	Q_i [IO]	P_n(%)	M(A) [IO]
54,59	^{87}Br	I	I	I	I	I	2,34	2,689
24,62	^{I37}I	I,65	I	I,02	I,62	0,537	4,46	4,9I2
	(^{I4I}Cs, ^{I34}Sn, ^{I36}Te)$^{+}$							
I5,88	^{88}Br	I,92	I	I,98	0,97	I,02	4,54	
II,0	^{I28}In(^{I34}Sb)$^{+}$	0,442	2	(0,I60)^{x}	2,8	0,I48	(2,53)§	2,388
6,3	^{I38}I	0,39	5	0,324		0,296	2,56	4,58I
	^{93}Rb	0,546 $\underline{0,94}$		$\underline{0,936}$	0,43	I,50	I,46	5,2II
		I,33		I,260				
4,55	^{89}Br	2,5I	5-I0	3,24	0,77	0,949	8,0	

\+ Secondary (not-dominant) contributors are given in brackets;

x Assuming that P_n = 2.53%;

§ Calculated in this paper.

Table 4

Fragment distribution parameters

Parameter	Parameter value		
	^{238}U E_n = 15 MeV	^{235}U E_n = 15 MeV	 Thermal^{+} neutrons
σ_z (amu)	1,723	1,863	1,51
A_p(Br)	88,41	87,48	88,09
ΔA_{pl} (amu)	-	- 0,61	-
ΔZ_{pl} (amu)^{x}	-	+ 0,39	-
A_p(I)	135,20	134,67	136,57
ΔA_{ph} (amu)^{x}	-	- 1,90	
ΔZ_{ph} (amu)	-	- 0,42	
$P_n(^{128}In)$	2,66	2,53	
* k_1	0,4860 ± 0,0013		
* k_1	0,4426 ± 0,064		

+ From Ref. [11]

x From Ref. [10]

P_n = 2.60 ± 0.07 (^{128}In)

* From data in Ref. [8]

Table 5

Comparison of total yields according to groups

Group No.	This paper	Ref. [4]	Ref. [10]
	^{238}U (15 MeV)		
I	I	I	I
2	3,I3	2,95	I,80
3	0,623	0,79	0,532
$\sum^{*}_{2,3}$	3,75	3,74	2,33
	^{235}U (15 MeV)		
I	I	I	I
2	I,39	I,25	I,I2
3	0,273	0,33	0,4I7
$\sum^{*}_{2,3}$	I,66	I,58	I,54

* Overall ratio of the yields of the second and third groups to that of the first.

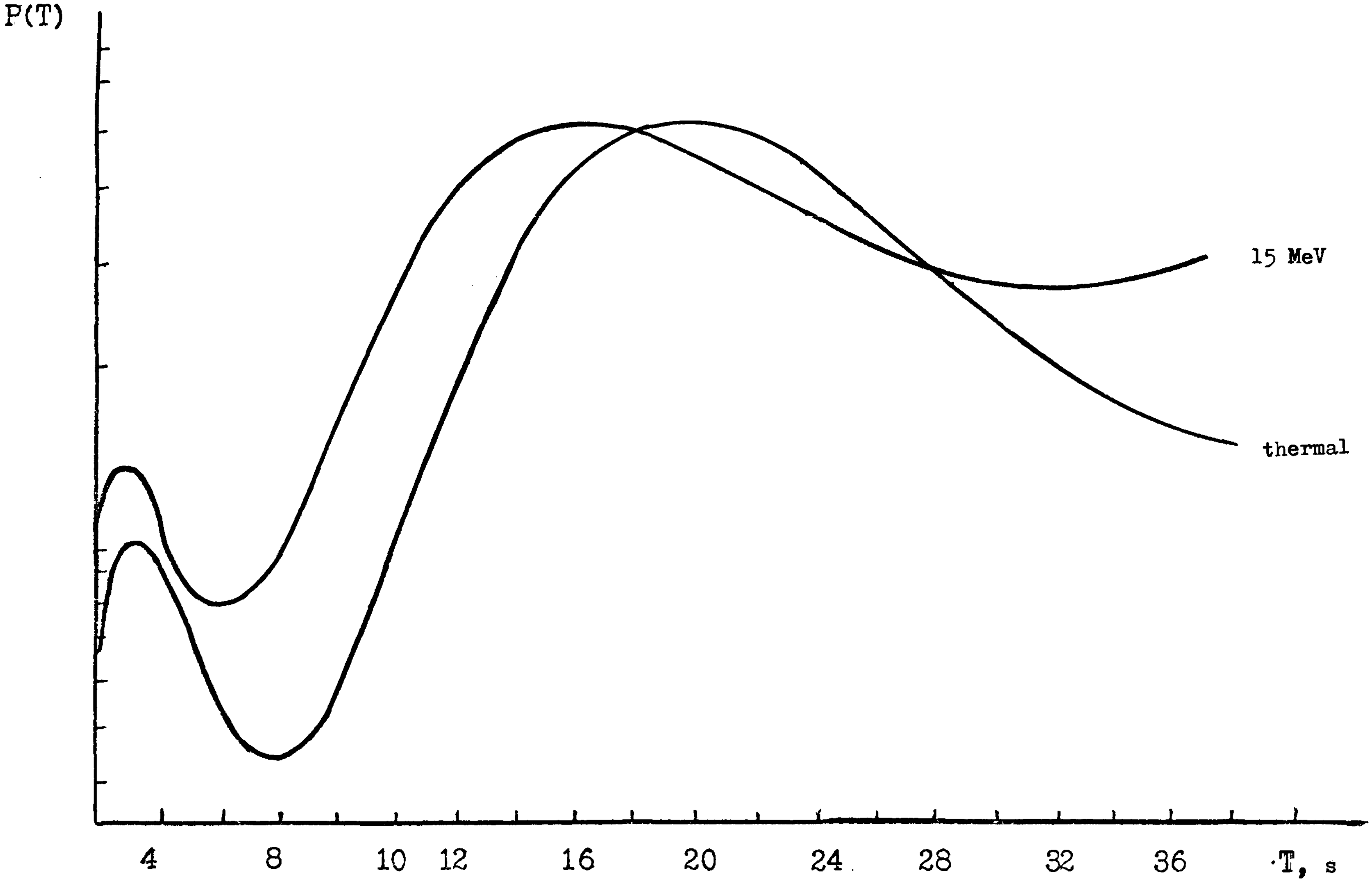

Fig. 1

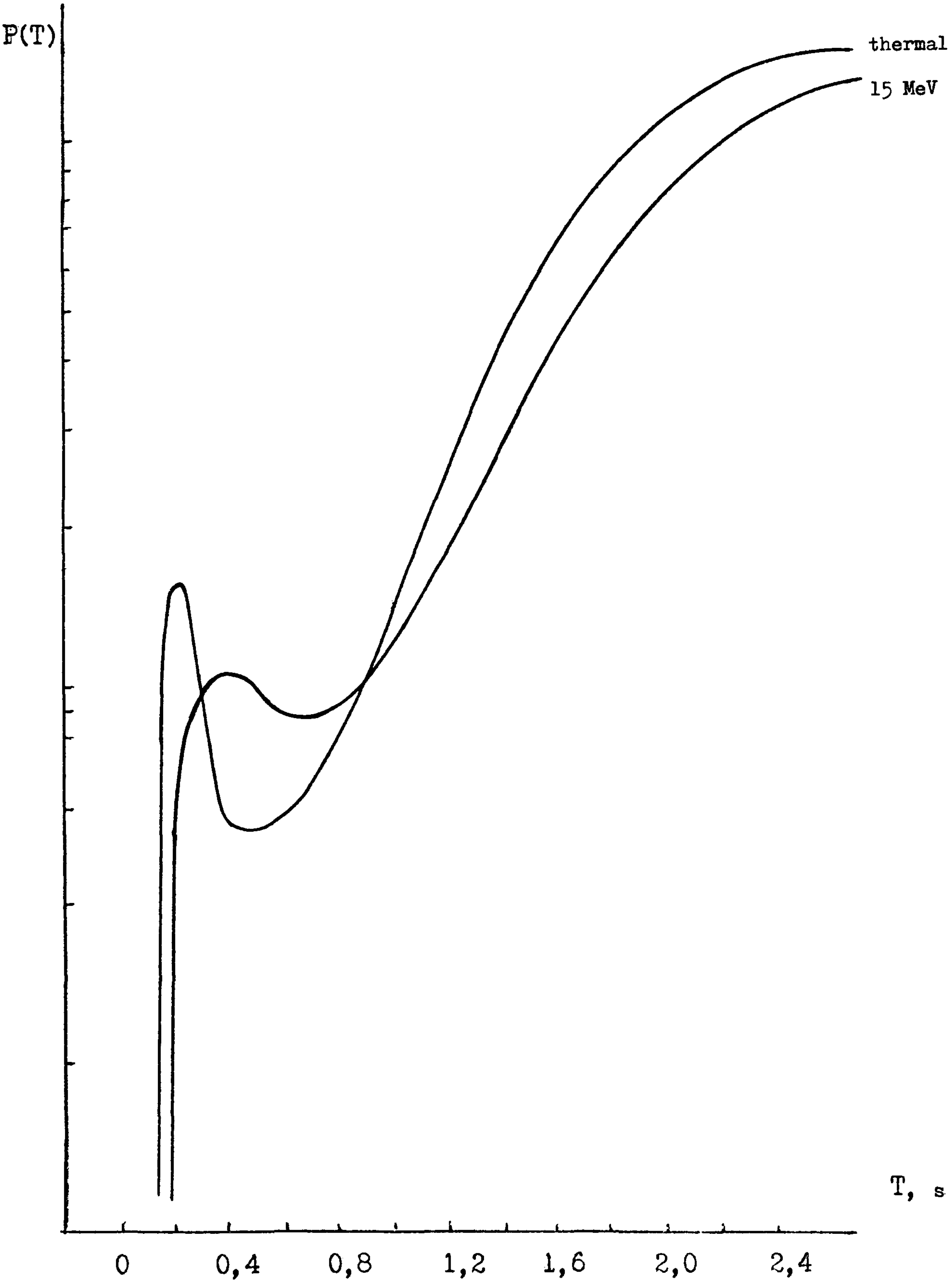

Fig. 2

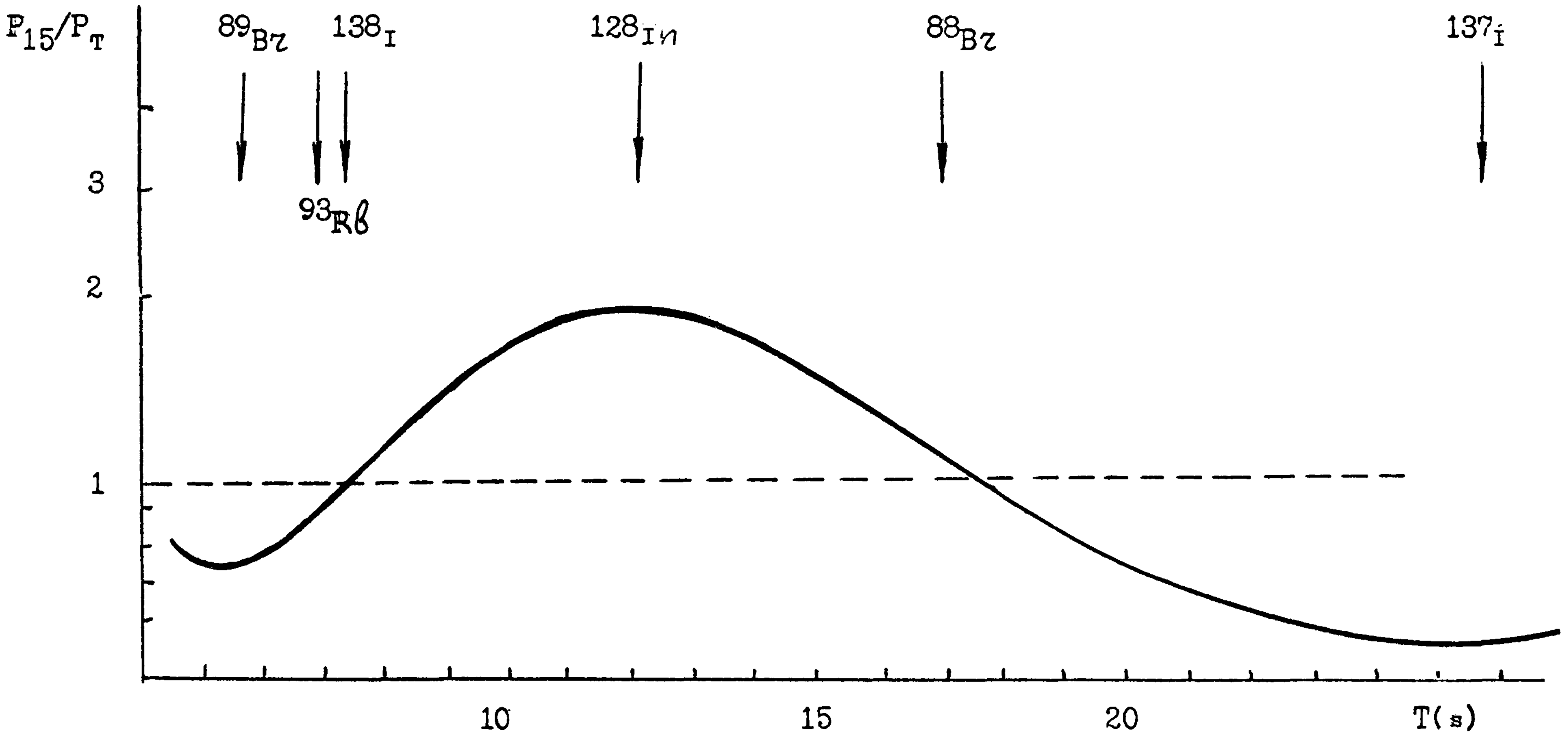

Fig. 3. Ratio of the probability density distributions P(T) for E_n = 15 MeV and for fission by thermal neutrons.

Appendix B

B1

Contents

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Subject index

1. Fission product yields[*]

Fissionable isotope	Page
	Fast neutrons E_n = 15 MeV
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^{238}U	9

[*] Note: The values given are the relative cumulative yields multiplied by P_n (the probability of delayed neutron yields from the given fragment).

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2. Fragment isotope distribution parameters

Fissionable isotope	Page
	Fast neutrons E_n = 15 MeV
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^{238}U	11

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3. Delayed neutron data (relative yields)

Precursor	Page
^{87}Br, ^{88}Br, ^{89}Br, ^{137}I, ^{93}Rb, ^{128}In	9,10

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4. Value of P_n for ^{128}In pp. 9,10

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Data evaluation

Type of data: input data; parameters of the experimentally measured decay in activity with E_n = 15 MeV for ^{235}U and ^{238}U (see Ref. [4]). These are used to give:

(a) Relative delayed neutron yields from seven pure precursors;

(b) Relative cumulative fragment yields;

(c) Value of P_n for ^{128}In;

(d) Distribution parameters for the fragment isotopes.

Purpose: for physical interpretation; nuclear physical constants for reactors.

Method: analysis of decay in activity by the mathematical method of least algebraic deviation; subsequent calculations based on the parameters for a Gaussian distribution of the fragment isotopes.

Main sources of information: see Ref. [4].

Most recent work taken into account: see Ref. [7].

Status: work completed.

Co-operation: with other groups - none.

Computer file: not relevant.

Evaluated data computer file: not relevant.

Discrepancies found: with Ref. [10].

Date of completion: February 1978.

Publication: intended.