



CORRELATION PROPERTIES OF DELAYED NEUTRONS FROM FAST NEUTRON INDUCED FISSION

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Introduction

The knowledge of the time dependence of β - delayed neutron emission is of great importance for the development of reliable data base required both for reactor-physics analysis and for the investigation of nuclei which are far from the valley of stability. Since discovery of delayed neutrons the time dependence of delayed neutron emission from neutron induced fission was studied in more than 238 experiments [1]. In parallel to the experimental studies of aggregate DN emission from neutron induced fission the group parameters of delayed neutron emission were calculated using microscopic approach (summation technique) [2]. These calculations are based on the knowledge of the emission probabilities values P_n , half-life values and yields of individual precursors of delayed neutrons [3].

One of the main results of the aggregate DN decay curve measurements was that the group periods (or decay constants) for various fissioning systems for fast neutrons induced fission do not differ from each other [4]. But one should take into account that possible differences in the group decay constants are difficult to be observed because of the strong cross-correlation between DN group parameters and background intensity values in the process of least-squares fitting (LSF) analysis of composite decay curve [5,6]. The time dependence of DN emission is governed not only by half-life of DN precursors. The relative abundances of DN are also important in forming the DN decay curve of the concrete fissioning system. Until now there are difficulties in choosing the consistent set of DN parameters even for the most studied nuclides. Set of DN parameters in ENDF/B-VI file is based on the data obtained in the frame of the microscopic approach [2]. But reactor experiments [5,7] have shown inconsistency of this set and appropriate experimental data based on the aggregate DN decay data.

With the purpose to find the criteria for choosing the best set of DN parameters for various fissioning systems the work was done on the investigation of systematics and correlation properties of delayed neutrons from fast neutron induced fission.

Correlations between delayed neutron parameters for fast neutron induced fission of Th, U, Pu and Am

During our experimental studies of the energy dependence of the delayed neutron parameters for various fissioning systems it was found that the behavior of a some combination of delayed neutron parameters (group relative abundances a_i and half lives T_i) has a similar features. On the basis of this findings the systematics of delayed neutron experimental data for thorium, uranium, plutonium and americium isotopes have been investigated with the purpose to find a correlation of DN parameters with characteristics of fissioning system as well as a correlation between the delayed neutron parameters themselves. Below we will present the preliminary results which were obtained during this study omitting the physics interpretation of the results.

It is difficult to hope that comparison of DN group parameters a_i and T_i separately will be successful because of their strong cross-correlation resulting from the LSF procedure used in processing of the aggregate DN decay data and other reasons [5,6]. Therefore the delayed neutron parameters under investigations are the following

1) *the average half-life of delayed neutron precursors*

$$\langle t \rangle = \left(\sum_{i=1}^{i=6} a_i T_i \right),$$

where a_i and T_i - relative abundance and half-life of i -th group of delayed neutrons respectively in the six group presentations. (The average half life value for the exponential distribution of the precursors' life times fully determines the above distribution);

2) *total delayed neutron yield ν_d .*

For the purpose of easy comparison of the present results with existing systematics a parameter P which characterizes the fissioning nucleus was chosen as in the Tuttle's evaluation [8]

$$P = -(A_c - 3Z) \cdot A_c / Z,$$

where A_c and Z - mass number and atomic number of compound nucleus respectively. In general the Z^2/A parameter can be taken as well as having the same scaling factor.

In the analysis only experimental data (ν_d , a_i and T_i) were used (exception - the Waldo's delayed neutron data for ^{240}Pu [9]). In the first approximation the authors of the present work took the data which have been obtained preferably at one experimental set-up and included the wide range of investigated nuclides. The experimental data under investigations are presented in the Table 1. It should be noted that the fast neutron induced fission were considered in the analysis and only for the rare measured isotopes such as ^{229}Th (Gudkov's et al. data [10]) and ^{243}Am (the Saleh's et al. data [11]) the thermal (^{229}Th) or close to thermal (^{243}Am) data were taken into consideration. But according to our estimations this exception does not change the obtained results.

The uncertainties of the average half-life values are not accounted for the correlations which exist between the relative abundances and periods values obtained on the basis of the aggregate DN activities data and the LSF procedure.

Systematics of the average half-life of delayed neutron precursors for different fissioning systems

It was found that the average half-life of delayed neutron precursors $\langle t \rangle$ for the isotopes of thorium, uranium, plutonium and americium can be presented by the following expression

$$\langle t_i \rangle = a_{1i} \cdot \exp[(a_{2i}(- (A_c - 3Z) \cdot A_c / Z)], \quad (1)$$

where index i is related to the certain fissioning systems (thorium, uranium etc.). The above expression (1) was presented in the form

$$\begin{aligned} \ln(\langle t_i \rangle) &= a_{3i} + a_{2i}[- (A_c - 3Z) \cdot A_c / Z] \\ a_{3i} &= \ln a_{1i} \end{aligned} \quad (2)$$

and the appropriate delayed neutron data from the Table 1 were analyzed for obtaining the values a_{3i} and a_{2i} on the basis of the LSF procedure. The results of the LSF procedure (solid lines) and original experimental data from the table 1 are shown in Fig.1. The obtained a_{3i} and a_{2i} values for each considered element are presented in Table 2. It is seen from the Fig.1 that the average half life values for each considered element can be presented by the expression (2). The best approximation was obtained for uranium isotopes. This can be explained by the quality of the data for uranium isotopes as being the most investigated nuclides.

Correlation of the total delayed neutron yields and the average half-life of delayed neutron precursors

It was found that the total delayed neutron yields ν_d and the average half-life of delayed neutron precursors $\langle t \rangle$ for the definite fissioning system, can be presented by the following expression

$$\nu_d^i = c_{1i} \cdot \langle t_i \rangle^{c_{2i}}, \quad (3)$$

where index i is related to the definite fissioning systems (thorium, uranium etc.). The expression (3) was presented in the following form

$$\begin{aligned} \ln(\nu_d^i) &= c_{3i} + c_{2i} \cdot \ln(\langle t_i \rangle) \\ c_{3i} &= \ln c_{1i} \end{aligned} \quad (4)$$

with the purpose to obtain the c_{2i} and c_{3i} values for each considered fissioning systems (thorium, uranium etc.). In the LSF analysis of the equation (4) the uncertainty of the average half life value for the definite fissioning system were taken as it was equal for all authors. The magnitude of this uncertainty was equal to the average value over all data related to the given fissioning system. The obtained results of the LSF procedure (solid lines) and the original experimental data from the Table 1 are shown in Figs.2, 3, 4 and 5. The appropriate c_{2i} and c_{3i} values for each fissioning system under consideration are presented in Table 3. It is seen from the Figs. 2, 3, 4, 5 that the data on the total delayed yields follow the dependence determined by the expression (4).

In the present analysis the each ν_d value corresponds to the $\langle t \rangle$ value from the same work. But the analysis could be done using $\langle t \rangle$ values which are obtained by preliminary averaging over appropriate experimental data or can be taken from the evaluation according to expression (1).

Total delayed neutron yields from fast neutron induced fission of isotopes of Th, U, Pu, and Am

The expressions (1) and (3) can be used for obtaining the values of total delayed neutron yields for any isotope of Th, U, Pu and Am elements on the basis of the following expression obtained by simple derivations

$$\begin{aligned} \nu_d^i &= d_{1i} \exp[d_{2i}(-(A_c - 3Z)A_c / Z)] \\ d_{1i} &= c_{1i}a_{1i}^{c_{2i}}, d_{2i} = a_{2i}c_{2i} \end{aligned} \quad (5)$$

The obtained dependence of the total delayed neutron yields on the parameter $-(A_c - 3Z)A_c/Z$ was presented in the logarithmic form for each fissioning system I

$$\begin{aligned} \ln \nu_d^i &= d_{3i} + d_{2i}(-(A_c - 3Z)A_c / Z), \\ d_{3i} &= c_{3i} + c_{2i}a_{3i}, d_{2i} = a_{2i}c_{2i} \end{aligned} \quad (6)$$

with the purpose of easy comparison with the Tuttle's equation [8]

$$\ln \nu_d = 13.81 + 0.1754(A_c - 3Z)(A_c / Z) \text{ (per 1 fission)}$$

It must be noted that the expression (6) obtained in the present work is *independent* from the Tuttle's one and based on the systematics $\langle t \rangle$ from A_c , Z and correlation between ν_d and $\langle t \rangle$ values derived from experimental data. The comparison of the total delayed neutron yield values obtained on the basis of the systematics $\langle t_i \rangle = a_{1i} \cdot \exp[(a_{2i}(-(A_c - 3Z) \cdot A_c / Z)]$ and correlations between the delayed neutron parameters $\nu_d^i = c_{1i} \cdot \langle t_i \rangle^{c_{2i}}$ in the present studies with the appropriate data of the Tuttle's evaluation is presented in Fig. 6 where the Tuttle's data are presented by a dash line. Numerical values of d_{3i} and d_{2i} parameters are presented in Table 4.

According to the present systematics the following ν_d dependencies were obtained for the considered elements

$$\ln \nu_d = 12.7479 + 0.1644(A_c - 3Z)(A_c / Z) \text{ (per 1 fission) for thorium isotopes,}$$

$$\ln \nu_d = 12.7887 + 0.1648(A_c - 3Z)(A_c / Z) \text{ (per 1 fission) for uranium isotopes,}$$

$$\ln \nu_d = 12.8389 + 0.1669(A_c - 3Z)(A_c / Z) \text{ (per 1 fission) for plutonium isotopes,}$$

$$\ln \nu_d = 6.8201 + 0.1097(A_c - 3Z)(A_c / Z) \text{ (per 1 fission) for americium isotopes.}$$

It is seen from Fig. 6 that the whole set of the total delayed neutron yields data cannot be presented by only one equation as it was done before and each element (isotopes of definite element) has its own dependence of ν_d on parameter $-(A_c - 3Z)A_c/Z$. The thorium and

uranium isotopes have a similar dependence. The dependence ν_d for Pu has the same slope as in the case of U and Th elements but has a parallel shift relative to U and Th data. The dependence of ν_d on parameter $-(A_c-3Z)A_c/Z$ for the americium isotopes has significantly different character. Therefore according to these preliminary studies the attempts to introduce more complicated parameters with the purpose to get better agreement with all experimental data is doubtful.

Analysis of ENDF/B-VI delayed neutron data

The ENDF/B-VI data was presented in the form of equation (2) and the comparison was made with the appropriate systematics data. The results of comparison were presented in fig.7. The systematics data are presented by solid lines. The ENDF/B-VI data are presented by the separate points. It is seen from the fig.7 that the average half life values for uranium has approximately the same overall dependence as it is in the systematics data. But it is important to note that the most studied isotope ^{235}U (having the best known fission yield data) is outside of the systematics dependence. The discrepancy in the time dependence of DN emission for ^{235}U were observed in the reactor based experiments. The dependencies of $\langle t \rangle$ values for Pu and Am elements obtained from ENDF/B-VI data base are significantly defer from the appropriate systematics dependencies obtained in the present work. And as in the case of ^{235}U isotope the $\langle t \rangle$ value for well studied ^{239}Pu isotope is significantly deferent from the systematics value. The reason of the observed discrepancies must be carefully investigated.

Conclusion

On the basis of the analysis of DN experimental data it was shown that the average half-life of DN precursors for Th, U, Pu, Am elements can be presented by the exponential dependence on the fissioning nucleus parameter $-(A_c-3Z)A_c/Z$ within experimental uncertainties. It was shown that the total delayed neutron yield values for each considered elements correlate with the appropriate values of the average half life of DN precursors. These properties of delayed neutrons allowed to obtain the independent systematics of the total DN yields for Th, U, Pu and Am elements. According to this systematics it was shown that the well known dependence of ν_d on $-(A_c-3Z)A_c/Z$ parameter has a more complex structure than it was assumed until now [8] - each of the considered element has its own ν_d dependence on this parameter.

Preliminary analysis of ENDF/B-VI delayed neutron data on the basis of the present systematics showed that the DN parameters a_i and T_i obtained by the summation method must be carefully checked with the purpose to find the source of their disagreement with experimental data.

As a result of the present findings some of the possible application of the reported results are the following. The obtained systematics of DN parameters can be used

- for testing of DN measurement techniques and the LSF procedures;
- for testing the existing DN parameters data base;

- as a criteria for testing the summation calculation procedure and appropriate input data (P_n and fission yields as well as the distribution of precursors between the DN groups).

Preliminary analysis of ENDF/B-VI data showed that the relative abundances and half lives data for the most studied in respect to fission yield data (fission yield is believed the main source of uncertainties in the summation method) ^{239}Pu and ^{235}U isotopes according to the present systematics are not correct;

- for the prediction of the DN parameters for elements for which there is no experimental data;
- for the evaluation of the DN parameters. The total delayed yields can be calculated on the basis of expression (5) and numerical values of the coefficients from the table 4 for the thorium, uranium, plutonium and americium isotopes. Appropriate data for neptunium and protactinium isotopes can be obtained in the first approximation by the linear interpolation of the dependence of d_{3i} and d_{2i} coefficients (see table 4) on the atomic number of fissioning nucleus.

The present systematics gives the possibility to improve the total delayed neutron data ν_d through performing the measurements of the aggregate DN decay curves only.

It should be noted that in the present analysis only the restricted part of the available experimental information on the DN parameters were used. The uncertainties of the average half life values must be calculated using the information on the correlation between the DN group parameters [6]. Therefore the obtained results (the coefficients in the tables) should be considered as preliminary data. The authors understand that the work must be continued with analysis of all available experimental information on the DN parameters.

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Table 1

Experimental delayed neutron parameters

Isotope	$-(Ac-3*Z)*Ac/Z$	Average half-life,s	Total DN yield, %	Reference
²²⁹ Th	102.2222	16.1872 ± 1.7429	1.73 ± 0.13	Gudkov(th.)
²³² Th	95.7889	7.0721 ± 0.4071	5.27 ± 0.40	Waldo
²³² Th	95.7889	6.9779 ± 0.5897	4.96 ± 0.20	Keepin

Isotope	$-(Ac-3*Z)*Ac/Z$	Average half-life	Total DN yield	Reference
²³² U	108.9022	14.3717 ± 1.3821	0.44 ± 0.03	Waldo
²³³ U	106.8261	12.2852 ± 2.6682	0.74 ± 0.04	Waldo
²³³ U	106.8261	12.3962 ± 0.9936	0.70 ± 0.04	Keepin
²³³ U	106.8261	12.1575 ± 1.5285	0.78 ± 0.04	Benedetti
²³⁵ U	102.6087	8.5012 ± 0.2417	1.67 ± 0.07	Waldo
²³⁵ U	102.6087	8.8271 ± 0.6001	1.65 ± 0.05	Keepin
²³⁵ U	102.6087	8.8378 ± 0.9575	1.64 ± 0.06	Besant
²³⁵ U	102.6087	8.7763 ± 1.9440	1.59 ± 0.04	Saleh
²³⁶ U	100.4674	7.3477 ± 1.1848	2.24 ± 0.23	Gudkov
²³⁸ U	96.1196	5.2409 ± 0.5470	4.60 ± 0.25	Waldo
²³⁸ U	96.1196	5.3194 ± 0.3599	4.12 ± 0.17	Keepin
²³⁸ U	96.1196	5.4918 ± 0.4496	4.39 ± 0.17	Besant

Isotope	$-(Ac-3*Z)*Ac/Z$	Average half-life	Total DN yield	Reference
²³⁸ Pu	109.3298	11.5871 ± 2.7142	0.46 ± 0.07	Waldo
²³⁸ Pu	109.3298	11.2468 ± 1.4339	0.41 ± 0.02	Benedetti
²³⁹ Pu	107.2340	9.9626 ± 1.1105	0.65 ± 0.05	Waldo
²³⁹ Pu	107.2340	10.1544 ± 0.5655	0.63 ± 0.03	Keepin
²³⁹ Pu	107.2340	10.3506 ± 1.3112	0.60 ± 0.02	Besant
²⁴⁰ Pu	105.1170	9.0461 ± 3.3965	0.88 ± 0.06	Waldo(rec.)
²⁴⁰ Pu	105.1170	9.3292 ± 0.8691	0.88 ± 0.06	Keepin
²⁴⁰ Pu	105.1170	9.7678 ± 0.8152	0.91 ± 0.04	Benedetti
²⁴⁰ Pu	105.1170	8.3947 ± 1.6539	0.88 ± 0.09	Gudkov
²⁴¹ Pu	102.9787	7.5900 ± 0.7403	1.57 ± 0.15	Waldo
²⁴¹ Pu	102.9787	7.7299 ± 0.7081	1.60 ± 0.09	Benedetti
²⁴¹ Pu	102.9787	7.8503 ± 0.8640	1.59 ± 0.14	Gudkov
²⁴² Pu	100.8192	6.5097 ± 2.1127	1.86 ± 0.09	Waldo

Isotope	$-(A_c - 3Z) \cdot A_c / Z$	Average half-life	Total DN yield	Reference
²⁴¹ Am	109.5368	10.5224 ± 1.3794	0.44 ± 0.05	Waldo
²⁴¹ Am	109.5368	11.2833 ± 1.5397	0.39 ± 0.02	Benedetti
²⁴¹ Am	109.5368	10.8596 ± 1.1322	0.49 ± 0.02	Saleh
²⁴¹ Am	109.5368	9.9757 ± 1.3831	-----	Gudkov
^{242m} Am	107.4316	10.0342 ± 0.9304	0.69 ± 0.05	Waldo
²⁴³ Am	105.3053	10.0646 ± 1.3730	0.84 ± 0.04	Saleh

Table 2

Results of approximation of experimental average DN half life dependence on parameter

$$P = -(A_c - 3Z) \cdot A_c / Z$$

Isotopes	Atomic number	a_{3i}^*	a_{2i}^* ($\times 10^2$)
Th	90	-10.440920 ± 0.232768	12.93765 ± 0.24030
U	92	-5.686208 ± 0.206631	7.645697 ± 0.202394
Pu	94	-4.210423 ± 0.597558	6.095660 ± 0.565445
Am	95	0.378623 ± 1.264624	1.812617 ± 1.166219

*) the coefficient of the equation $\ln(\langle t_i \rangle) = a_{3i} + a_{2i} [-(A_c - 3Z) \cdot A_c / Z]$

Table 3

Results of approximation of total DN yield dependence on average DN half life parameter

Isotopes	Atomic number	c_{3i}^*	c_{2i}^*
Th	90	6.38930 ± 0.19035	-1.27061 ± 0.08962
U	92	7.43714 ± 0.15498	-2.15597 ± 0.07258
Pu	94	8.21954 ± 0.55917	-2.73775 ± 0.25073
Am	95	16.01973 ± 2.77435	-6.05317 ± 1.17572

*) the coefficients of the equation $\ln(v_d^i) = c_{3i} + c_{2i} \cdot \ln(\langle t_i \rangle)$ (per 1000 fissions)

Table 4

Results of approximation of the dependence of total DN yields on parameter

$$P = -(A_c - 3Z) \cdot A_c / Z$$

Isotopes	Atomic number	d_{3i} *	d_{2i} *
Th	90	19.65563	-0.16438
U	92	19.69643	-0.16483
Pu	94	19.74662	-0.16688
Am	95	13.72786	-0.10972

*) the coefficients of the equation

$$\ln v_d^i = d_{3i} + d_{2i}(- (A_c - 3Z) A_c / Z),$$

$$d_{3i} = c_{3i} + c_{2i} a_{3i}, d_{2i} = a_{2i} c_{2i}$$

(per 1000 fissions)

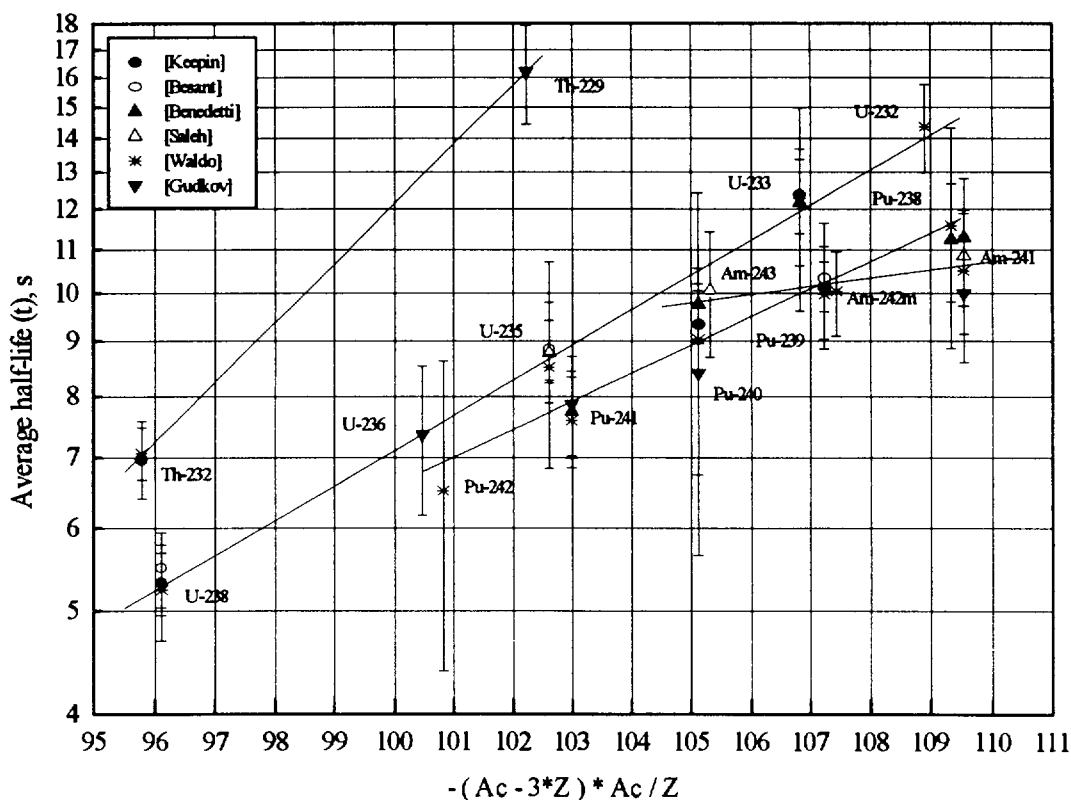


Fig. 1. Systematics of the average half life of DN precursors for Th, U, Pu, Am elements

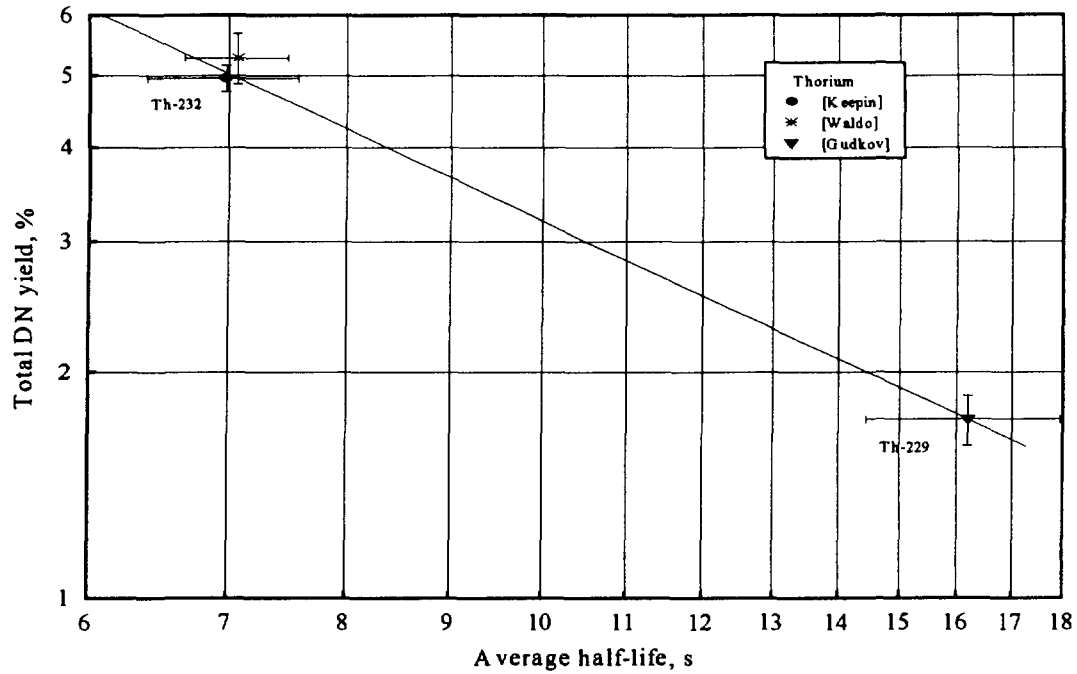


Fig. 2. Total DN yield as a function of average half-life of DN precursors for Th

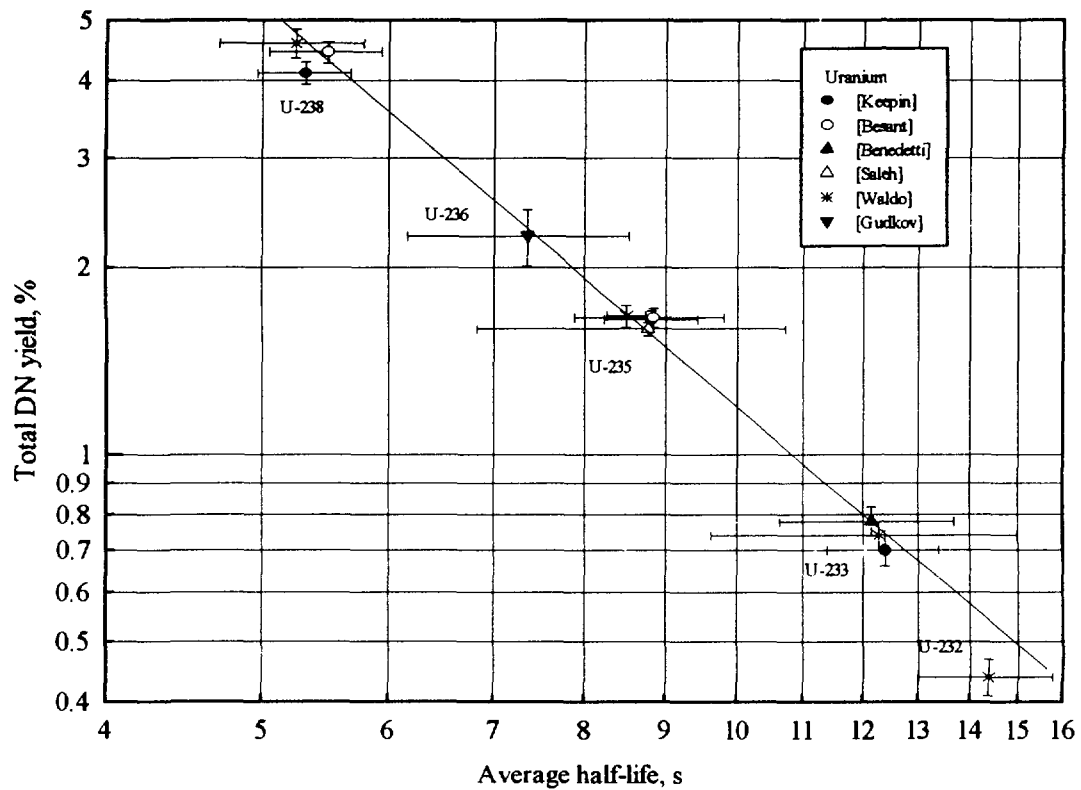


Fig. 3. Total DN yield as a function of average half life of DN precursors for U

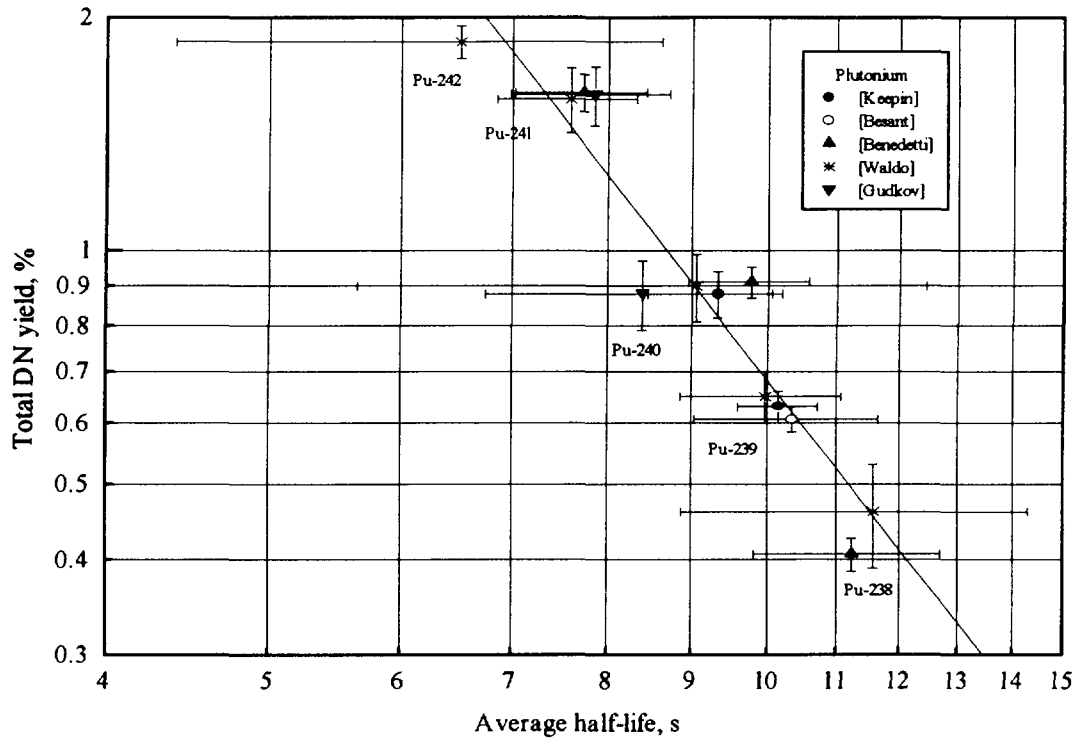


Fig. 4. Total DN yield as a function of average half life of DN precursors for Pu

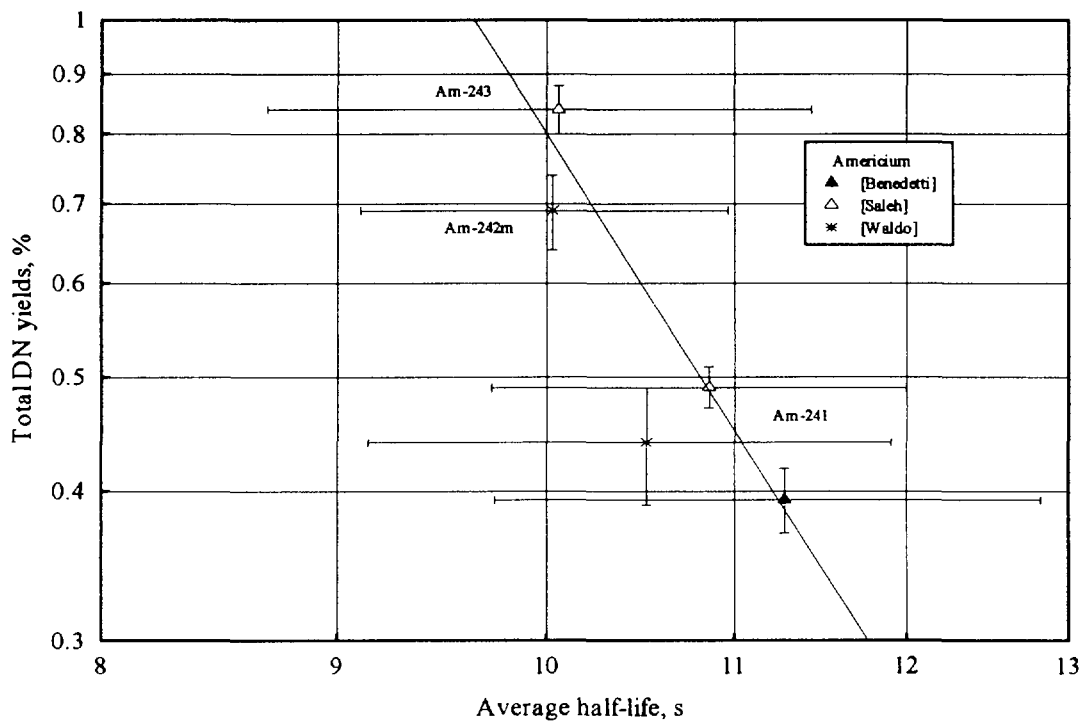


Fig. 5. Total DN yield as a function of average half life of DN precursors for Am

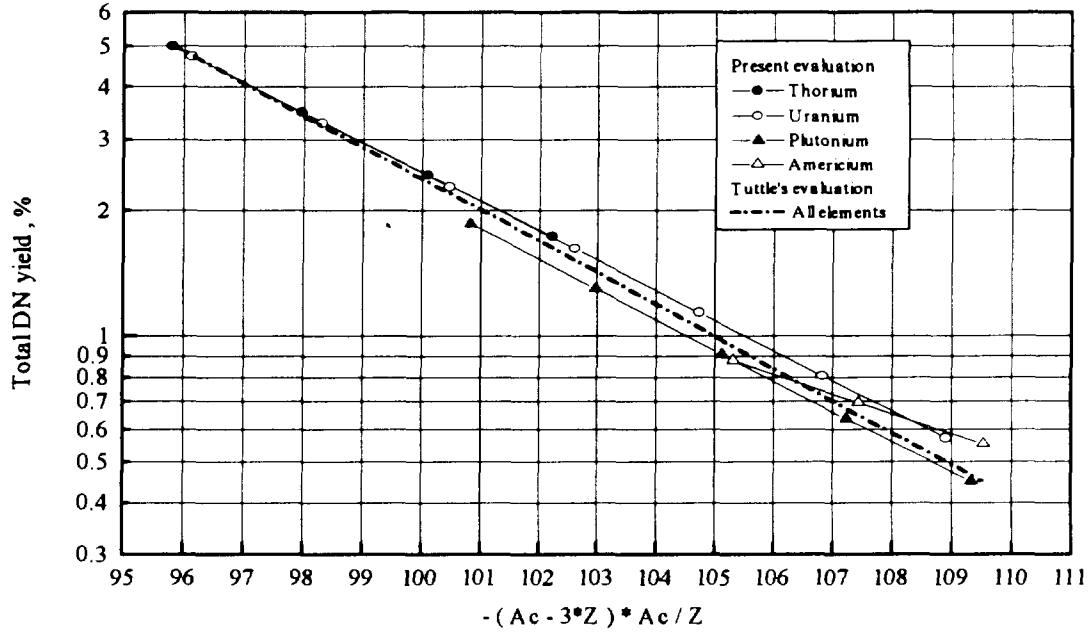


Fig. 6. Dependence of the total DN yields on parameter $P = -(A_c - 3Z) \cdot A_c / Z$
 Points connected by solid lines - present systematics, dashed line - Tuttle's equation [8]

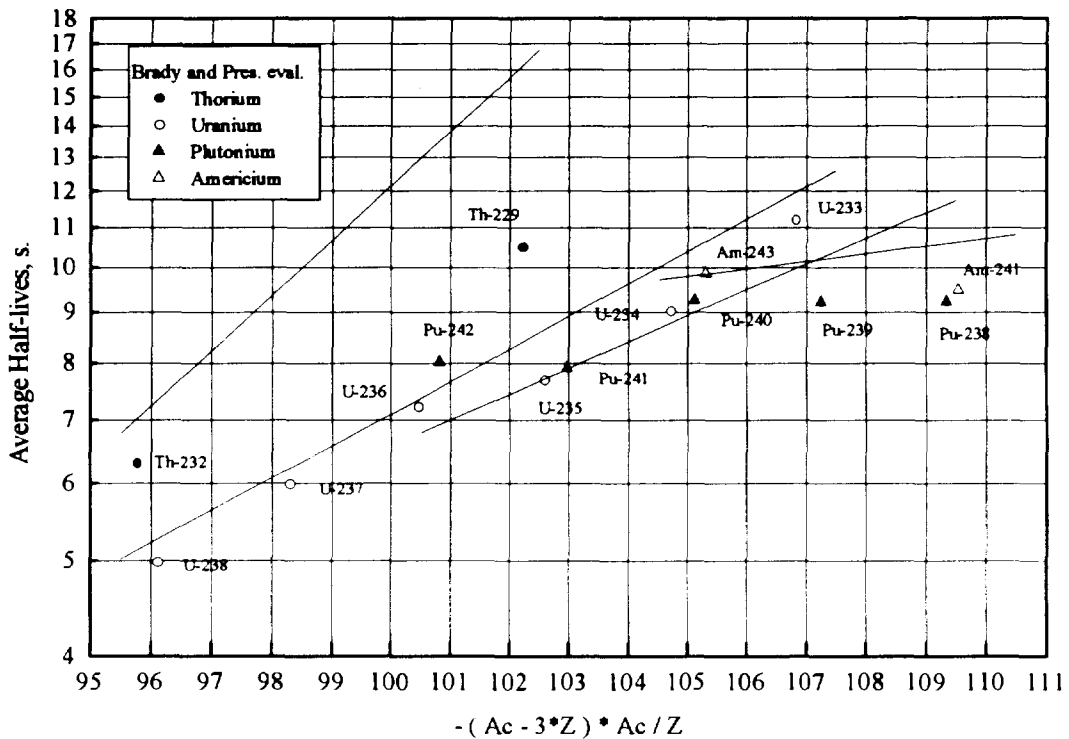


Fig. 7. Dependence of the average half life of DN precursors on parameter $P = -(A_c - 3Z) \cdot A_c / Z$,
 Solid lines - present systematics (see Fig. 1), points - the ENDF/B-VI data.

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