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TWO REPORTS:

(i) CORRELATION PROPERTIES OF DELAYED NEUTRONS FROM FAST NEUTRON INDUCED FISSION

V.M. Piksaikin, S.G. Isaev
Institute of Physics and Power Engineering, Obninsk, Russia

(ii) METHOD AND SET-UP FOR MEASUREMENTS OF TRACE LEVEL CONTENT OF HEAVY FISSIONABLE ELEMENTS BASED ON DELAYED NEUTRON COUNTING

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Abstract

The experimental studies of the energy dependence of the delayed neutron parameters for various fissioning systems has shown 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.

(ii) METHOD AND SET-UP FOR MEASUREMENTS OF TRACE LEVEL CONTENT OF HEAVY FISSIONABLE ELEMENTS BASED ON DELAYED NEUTRON COUNTING

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Abstract

Methods and set-up for measurements of trace level content of fissionable nuclides based on the delayed neutron counting technique are presented. It is shown that the electrostatic accelerator based method using the ^{9}Be(d,n)^{10}B reaction as a neutron source allows to determine the content of fissionable nuclides in ultra trace level (nano gram). New method for the determination of isotopic content of the sample is proposed. This method is based on the new systematics of the average half life of delayed neutron precursors for different fissioning systems.

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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

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

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 $\mathbf{n}_d$.

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 ($\mathbf{n}_d$, 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 *<t>* for the isotopes of thorium, uranium, plutonium and americium can be presented by the following expression

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

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

$$\ln(\langle t_i \rangle) = a_{3i} + a_{2i}\left[-(A_c - 3Z) \cdot A_c / Z\right] \tag{2}$$
$$a_{3i} = \ln a_{1i}$$

and the appropriate delayed neutron data from the Table 1 were analyzed for obtaining the values $a_{3\,i}$ and $a_{2\,i}$ 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 $a_{3\,i}$ and $a_{2\,i}$ 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 $\mathbf{n}_d$ and the average half-life of delayed neutron precursors $< t >$ for the definite fissioning system, can be presented by the following expression

$$\mathbf{n}^i{}_d = c_{1i} \cdot \langle t_i \rangle^{c2i}, \tag{3}$$

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

$$\ln(\mathbf{n}_d^i) = c_{3i} + c_{2i} \cdot \ln(\langle t_i \rangle) \tag{4}$$
$$c_{3i} = \ln c_{1i}$$

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 $\mathbf{n}_d$ value corresponds to the $< t >$ value from the same work. But the analysis could be done using $< t >$ 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

$$n_d^i = d_{1i} \exp\left[d_{2i}(-(A_c - 3Z)A_c / Z)\right] \tag{5}$$

$$d_{1i} = c_{1i}a_{1i}^{c2i}, d_{2i} = a_{2i}c_{2i}$$

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

$$\ln n_d^i = d_{3i} + d_{2i}(-(A_c - 3Z)A_c / Z), \tag{6}$$

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

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

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

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 $< t >$ from A_c, Z and correlation between n_d and $< t >$ values derived from experimental data. The comparison of the total delayed neutron yield values obtained on the basis of the systematics $\langle t_i \rangle = a_{1i} \cdot \exp\left[(a_{2i}(-(A_c - 3Z) \cdot A_c / Z)\right]$ and correlations between the delayed neutron parameters $n_d^i = c_{1i} \cdot \langle t_i \rangle^{c2i}$ 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 $d_{3\,i}$ and $d_{2\,i}$ parameters are presented in Table 4.

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

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

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

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

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

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 n_d on parameter - $(A_c-3Z)A_c/Z$. The thorium and

uranium isotopes have a similar dependence. The dependence $\mathbf{n}_d$ 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 $\mathbf{n}_d$ 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 $< t >$ 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 $< t >$ 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 $\mathbf{n}_d$ 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 $\mathbf{n}_d$ 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 $\mathbf{n}_d$ 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.

Acknowledgment

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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	-(Ac-3*Z)*Ac/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 $\ln(\langle t_i \rangle) = a_{3i} + a_{2i}\left[-(A_c - 3Z) \cdot A_c / Z\right]$

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 $\ln(n_d^i) = c_{3i} + c_{2i} \cdot \ln(\langle t_i \rangle)$ (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

$$\ln \boldsymbol{n}_d^i = d_{3i} + d_{2i}(-(A_c - 3Z)A_c / Z),$$
$$d_{3i} = c_{3i} + c_{2i}a_{3i}, d_{2i} = a_{2i}c_{2i}$$
(per 1000 fissions)
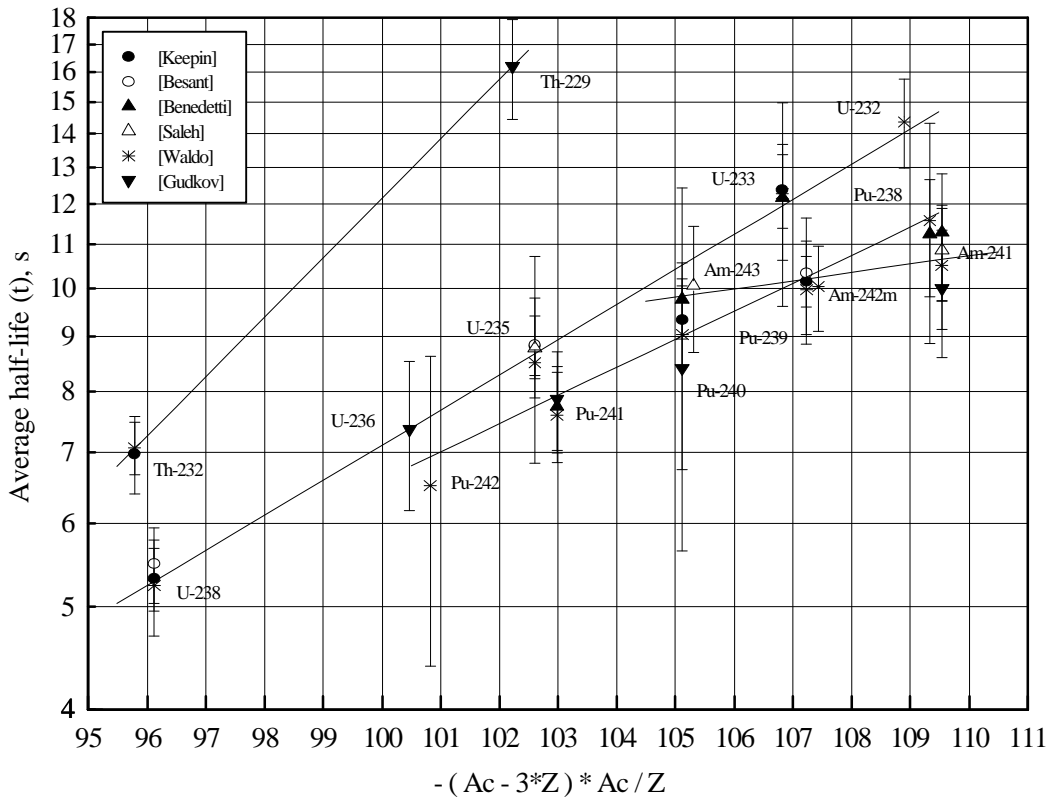
Fig. 1. Systematics of the average half life of DN precursors for
Th, U, Pu, Am elements
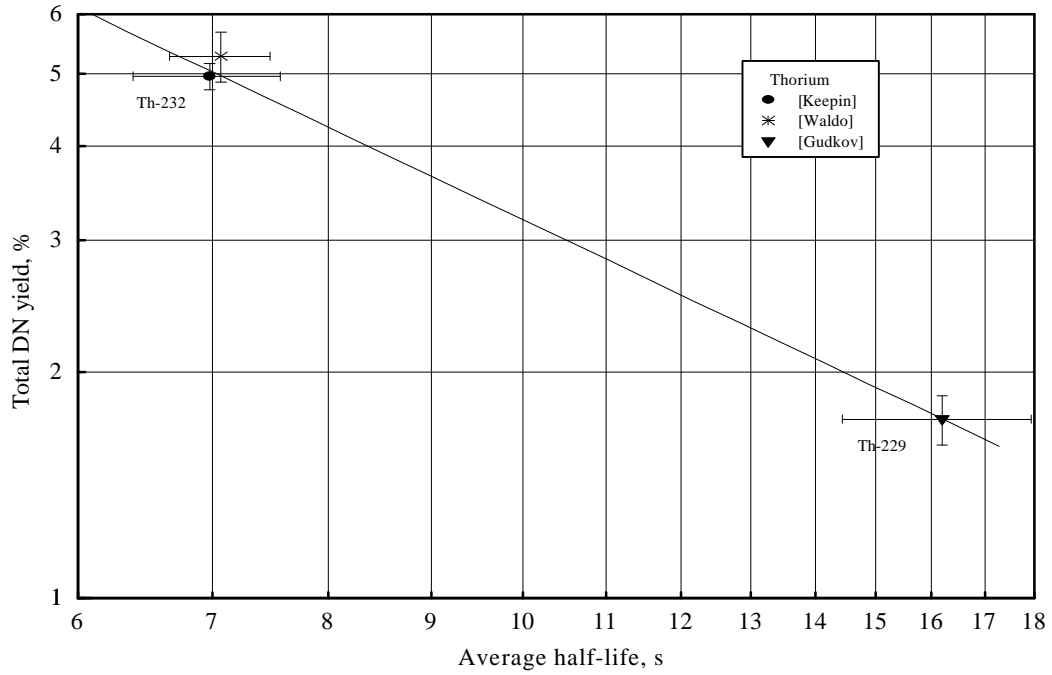
Fig. 2. Total DN yield as a function of average half-life of DN precursors for Th
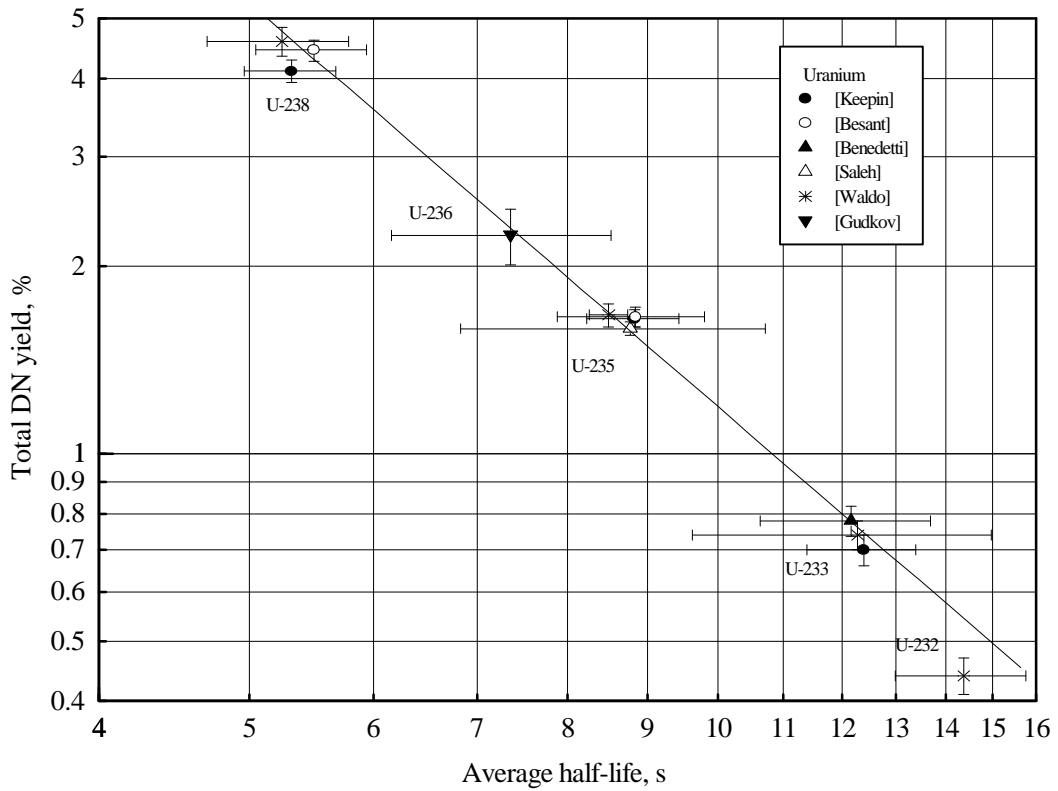
Fig. 3. Total DN yield as a function of average half life of DN precursors for U
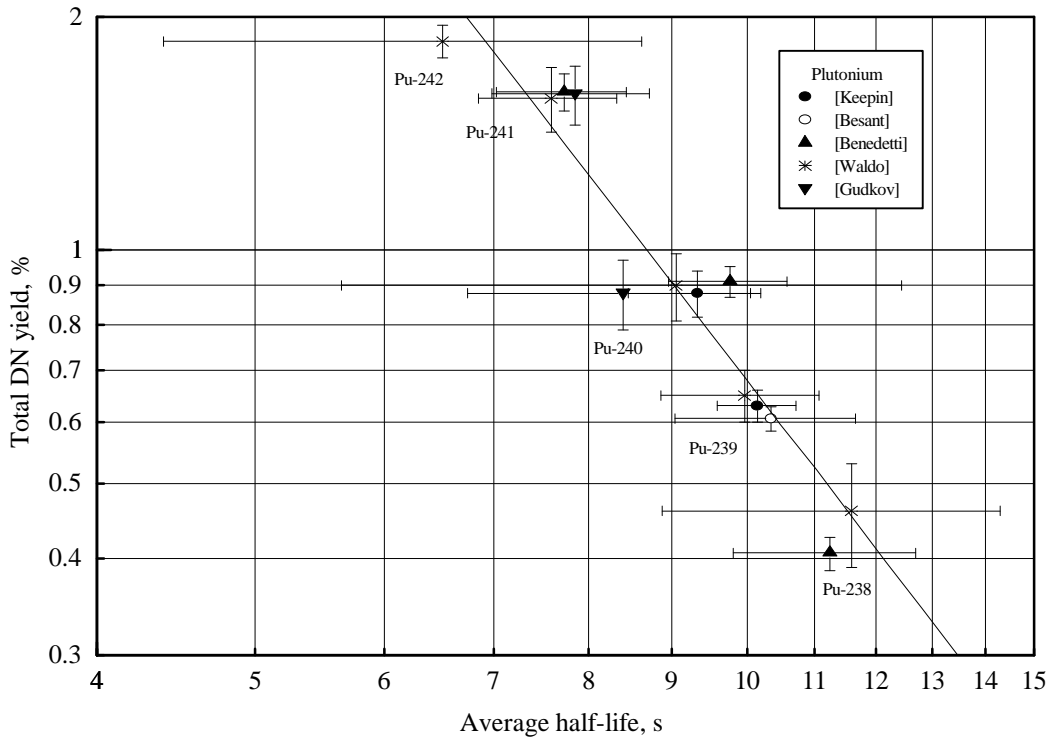
Fig. 4. Total DN yield as a function of average half life of DN precursors for Pu
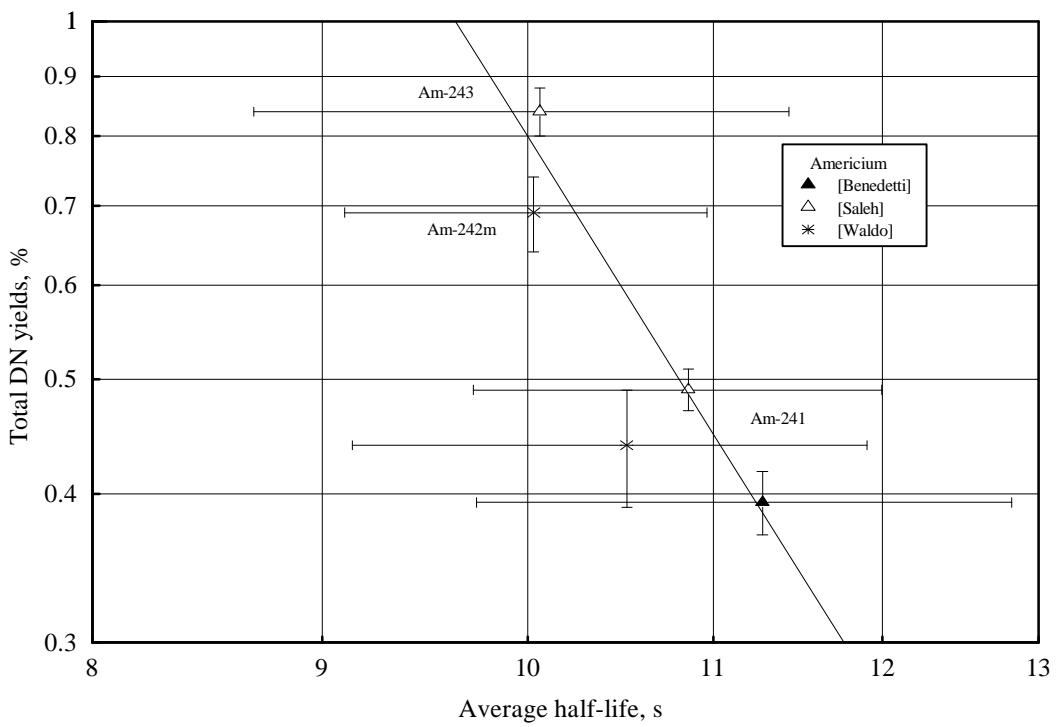

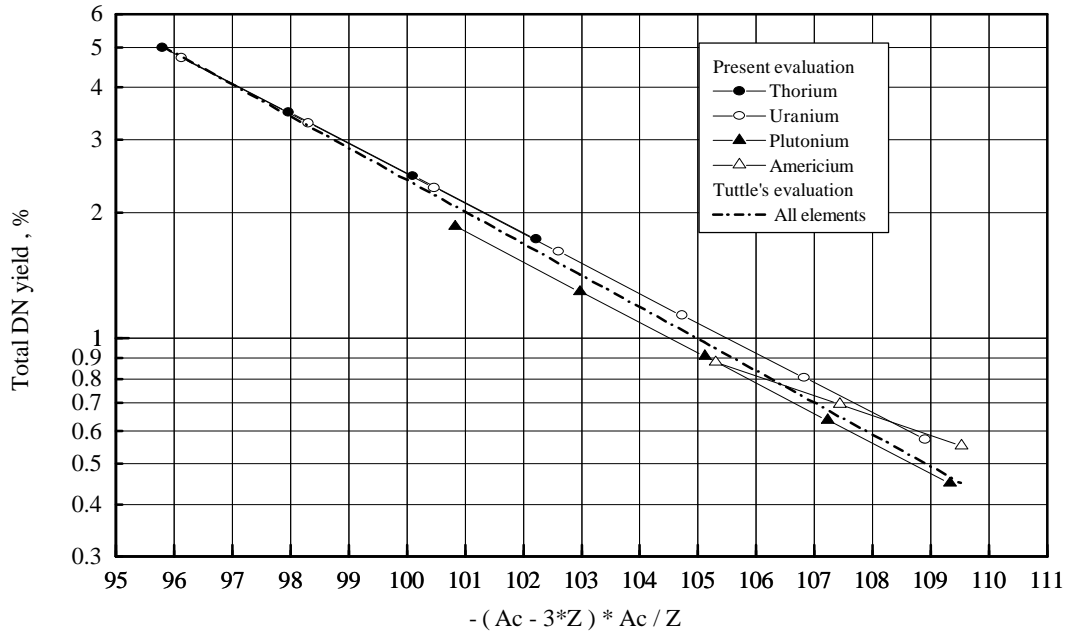Fig. 5. Total DN yield as a function of average half life of DN precursors for Am

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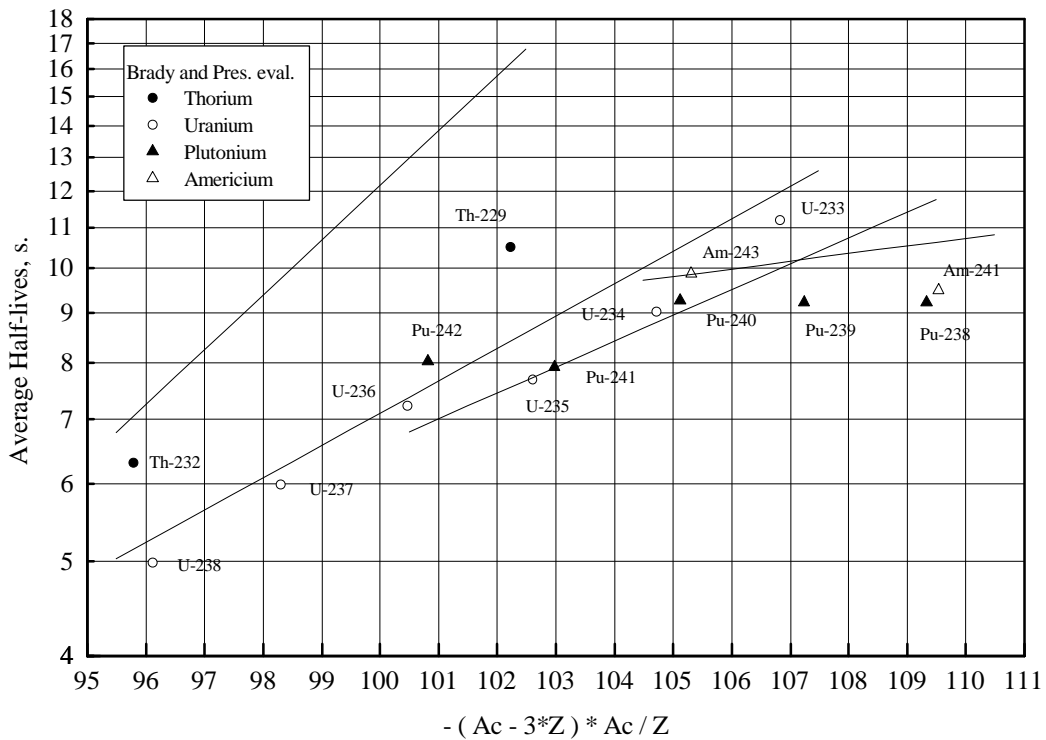Points connected by solid lines - present systematics, dach line - Tuttle's equation [8]

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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METHOD AND SET-UP FOR MEASUREMENTS OF TRACE LEVEL CONTENT OF HEAVY FISSIONABLE ELEMENTS BASED ON DELAYED NEUTRON COUNTING

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Abstract

Methods and set-up for measurements of trace level content of fissionable nuclides based on the delayed neutron counting technique are presented. It is shown that the electrostatic accelerator based method using the $^9Be(d,n)^{10}B$ reaction as a neutron source allows to determine the content of fissionable nuclides in ultra trace level (nano gram). New method for the determination of isotopic content of the sample is proposed. This method is based on the new systematics of the average half life of delayed neutron precursors for different fissioning systems.

Introduction

Availability of the method for the identification of ultra trace levels of fissionable elements (actinides) in samples of varied origins is of great importance for many area of applications. The commonly used methods of elemental analysis are the neutron activation analysis (NAA), neutron-induced prompt gamma-ray analysis (PGA), proton-induced X-ray analysis. The minimum detectable amounts are a function of many factors such as the source strengths, detector efficiency, geometry, sample quality, interfering reactions and other factors related to specific experiments. For instance reactor-based PGA method has the highest sensitivity that allows to obtain for thorium and uranium elements the detection limit approximately of ~ 1 mg/g [1]. At the present time the alpha spectrometry method with preliminary chemical separation of actinides is routinely used for the identification of these elements in trace levels but it is time consuming.

The method of elemental analysis based on the delayed neutron (DN) activity counting was estimated as having for thorium and uranium elements the minimal detectable amount ~50 μg [2]. However the developments of more reliable data base for DN parameters and utilization of high strength neutron sources make it possible to extend the DN counting technique to quantitative and qualitative analysis of fissionable elements in ultra trace level.

Experimental method and set-up for content determination of fissionable elements in samples

Experimental set-up primarily was designed and successfully used for the investigations of the delayed neutron yields from neutron induced fission of heavy nuclei [3]. The set-up was installed at the electrostatic accelerator KG-2.5 and has the following main parameters: ion (proton and deuteron) current - up to 500 μA, pneumatic sample delivery system - 150 ms and 1 s for 'fall down' sample delivery system, high voltage - up to 2 MV, neutron flux monitor - calibrated fission chamber, neutron detector - 30 boron counters embedded in the polyethylene moderator. Neutron detector efficiency is 0.084 with very low sensitivity to gamma-ray

background of the sample under investigation. The intensity of the neutron background during delayed neutron counting period is about 0.008 counts/s per 1 µA of deuteron current in case of the (d, n) neutron production reactions.

The general equation for elemental analysis on the basis of the delayed neutron counting can be expressed as the following:

$$N(t_k) = A \cdot \sum_{i=1}^{i=m} F_i \cdot \frac{a_i}{\boldsymbol{l}_i} \cdot (1 - \exp(-\boldsymbol{l}_i \Delta t_k)) \cdot \exp(-\boldsymbol{l}_i t_k) + B \cdot \Delta t_k , \qquad (1)$$

$$F_i = (1 - \exp(-\boldsymbol{l}_i t_{ir})) \left(\frac{n}{1 - \exp(-\boldsymbol{l}_i T)} - \exp(-\boldsymbol{l}_i T) \left(\frac{1 - \exp(-n\boldsymbol{l}_i T)}{((1 - \exp(-\boldsymbol{l}_i T))^2} \right) \right),$$

$$A = \boldsymbol{e}\, \boldsymbol{s}_f \boldsymbol{j}\; N_f \boldsymbol{n}_d ,$$

where $N(t_k)$ - the number of counts registered by the neutron detector in the time-channel t_k with time-channel width Δt_k, $\boldsymbol{n}_d$ - the total delayed neutron yield per one fission, B- the intensity of neutron background, $\boldsymbol{l}_i$ è a_i - the decay constant and relative abundance of i -th group of DN, n - the number of cycles, m- the number of DN groups, T - the duration of one cycle of measurements, which includes the irradiation and the delayed neutron counting time, t_{ir} - irradiation time, $\boldsymbol{e}$ - efficiency of neutron detector, $\boldsymbol{j}$ - the neutron flux, $\boldsymbol{s}_f$ - fission cross section, N_f - the number of atoms of fissionable element (nuclide) under investigation.

Equation (1) and the value of parameters of the set-up allow to estimate the (detectable concentration) detection limit of fissionable elements (as well as minimal detectable amount) in the samples for the neutron source based on the ^{9}Be(d,n)^{10}B reaction and deuteron ion current of 500 µA [4]. It was assumed that for the reliable analysis one needs to register one hundred delayed neutron counts above the background. The result of the estimation for thorium, uranium and plutonium elements are presented in the Table 1.

Table 1

Sensitivity of delayed neutron counting technique in the analysis
of the content of fissionable elements in the samples.

Nuclide	Minimal detectable amount *, g		Detectable concentration *, g/g	
	Fast neutron flux	Thermal neutron flux**	Fast neutron flux	Thermal neutron flux**
^{235}U	$6.3 \bullet 10^{-6}$	$1.5 \bullet 10^{-6}$	$1.3 \bullet 10^{-8}$	$3 \bullet 10^{-9}$
^{238}U	$1 \bullet 10^{-5}$		$2 \bullet 10^{-8}$	
^{239}Pu	$1 \bullet 10^{-5}$	$2.6 \bullet 10^{-6}$	$1.9 \bullet 10^{-8}$	$5 \bullet 10^{-9}$
^{232}Th	$1.7 \bullet 10^{-5}$		$3.3 \bullet 10^{-8}$	

*)   Amounts which were obtained at the experimental conditions indicated in the text
**)   Degradation of the neutron flux in the neutron slowing down process was taken into account.

Ten cycles of irradiation and delayed neutron counting were taken into consideration. The sample irradiation time was 100 s and the delayed neutron counting time was 25 s starting at 1 s after the end of irradiation. The total time spent for analysis was 1260 s. The estimation was made both for the fast neutron flux from the ^{9}Be(d,n)^{10}B reaction at 2 MeV deuteron energy and for the thermal neutron flux which can be easily obtained by slowing down the neutrons from the neutron target. Degradation of the neutron flux during the neutron slowing down process was accounted for.

It is seen from the Table 1 that the set-up under discussion affords to determine the fissionable elements containing in the sample in trace level. Utilization of many cycles measurements leads to increasing the sensitivities of the analysis. Moreover in contrast to the gamma rays and alpha particles analysis methods the delayed neutron counting method has no restriction on the weight of the sample under investigation that also leads to increasing the sensitivity of the analysis based on the delayed neutron counting. The detectable concentration of fissionable nuclides was estimated for 500 g sample.

The combination of the fast neutron flux and the thermal neutron flux analysis allows to make the identification of the isotopic content of the sample.

Method of identification of isotopic content of sample

Until now the identification of isotopic content of the sample in the frame of the DN counting technique was based on the difference between the values of relative abundances of the definite DN group for different nuclides [5]. This method requires a high statistical accuracy of DN decay curve [6] and reliable data base for the DN group parameters (decay constants and relative abundances). The first condition is difficult to reach because of small amount of fissionable elements in the sample.

We propose another approach for the identification of isotopic content of sample which is based on the new systematic of the delayed neutron parameters [7]. According to this systematic the average half-life of the delayed neutron precursors for the isotopes of thorium, uranium, plutonium and americium elements can be presented by the following expression

$$\langle T_i \rangle = a_{1i} \cdot \exp\left[(a_{2i}(-(A_c - 3Z) \cdot A_c / Z)\right], \tag{2}$$

where index i is related to the certain fissioning systems (thorium, uranium, etc.), A_c and Z - the mass number and atomic number of the fissioning nuclei respectively. The experimental data on the average half life parameters were obtained using the formula

$$\langle T \rangle = \sum_{i=1}^{6} a_i t_i,$$

where a_i and t_i are the relative abundance and period of the i-th DN group. The above expression (2) was presented in the form

$$\ln\langle T_i \rangle = a_{3i} + a_{2i}\left[-(A_c - 3Z) \cdot A_c / Z\right],$$
$$a_{3i} = \ln a_{1i} \tag{3}$$

and the appropriate delayed neutron data were analyzed for obtaining the values $a_{3\,i}$ and $a_{2\,i}$ on the basis of the least-square procedure. The results of the fitting procedure (solid lines) are shown in Fig.1. The obtained $a_{3\,i}$ and $a_{2\,i}$ values for each of the considered element are presented in Table 2. Thus all of the known isotopes can uniquely be identified by only one parameter - the average half life of the delayed neutron precursors. Therefore for the identification of the isotopic content of the sample one needs to make measurements and the least squares analysis of DN decay curve with the purpose to obtain the value of average half life parameter (for the mixture of nuclides). As compared with the six group parameters analysis [6] such analysis can be done using decay curves with much less statistical accuracy.

Table 2

Results of LSF analysis of DN experimental data

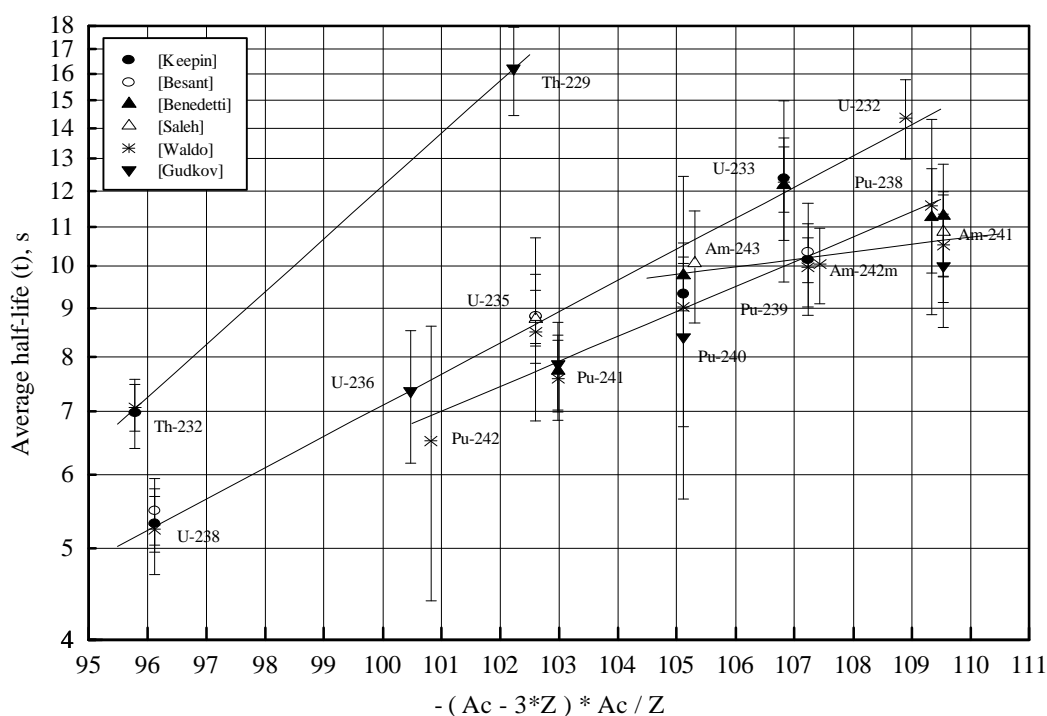| Element	a_3	a_2, ($\times 10^2$)
Th	-10.44 ± 0.23	12.94 ± 0.24
U	-5.69 ± 0.21	7.65 ± 0.20
Pu	-4.21 ± 0.60	6.10 ± 0.57
Am	0.38 ± 1.26	1.81 ± 1.17

Fig.1. Systematics of the average half life of delayed neutron precursors

In case of presence of two nuclides in the sample the obtained value $<T_{1,2}>$ is connected to unknown value of the fractional amount of the number of atoms of nuclides 1 and 2 by the following expressions

$$<T_{1,2}> = (n_1\, s_1\, j\, m_1 <T_1> + n_2\, s_2\, j\, m_2 <T_2>)/(n_1\, s_1\, j\, m_1 + n_2\, s_2\, j\, m_2\,),$$

$$m_1 + m_2 = 1,$$

where n_1, n_2 - the total delayed neutron yields related to nuclide 1 and 2, s_1, s_2 - the fission cross section of nuclides 1 and 2, $<T_1>$, $<T_2>$ - the average half life of DN precursors of nuclide 1 and 2, m_1, m_2 - the fractional amount of the number of atoms of nuclide 1 and 2 respectively, j - the neutron flux through the sample.

In case of tree nuclides in the sample with two of them which are fissionable by thermal neutrons (for example ^{235}U, ^{238}U, ^{239}Pu) the combination of the fast neutron and thermal neutron flux analysis will give respectively the average half life values $<T_{12}>$ and $<T_{123}>$ for the mixture of nuclides which are connected to the fractional amount of the number of atoms of nuclides m_1, m_2, and m_3 in the sample by the following expression

$$<T_{1,2}> = (n_1\, s_{1\,,th}\, j_{\,th}\, m_1 <T_1> + n_2\, s_{2,\,th}\, j_{\,th}\, m_2 <T_2>)/$$
$$.../(n_1\, s_{1\,,th}\, j_{\,th}\, m_1 + n_2\, s_{2,th}\, j_{\,th}\, m_2\,),$$

$$<T_{1,2,3}> = (n_1\, s_{1f}\, j_{\,f}\, m_1 <T_1> + n_2\, s_{2f}\, j_{\,f}\, m_2 <T_2> + n_3\, s_{3f}\, j_{\,f}\, m_3 <T_3>)/$$
$$.../(n_1\, s_{1f}\, j_{\,f}\, m_1 + n_2\, s_{2f}\, j_{\,f}\, m_2 + n_3\, s_{3f}\, j_{\,f}\, m_3),$$

$$m_1 + m_2 + m_3 = 1,$$

where $\sigma_{1\,,th}$, σ_{1f} and φ_{th}, φ_f - are the fission cross sections and the neutron fluxes for thermal and fast neutrons respectively. $<T>$ values for thermal and fast neutron induced fission of all fissioning system were assumed to be equal.

Conclusion

The DN counting technique coupled with the electrostatic accelerator based neutron source ^{9}Be(d,n)^{10}B is a powerful instrument in performing the analysis of trace level content of fissionable elements in the samples of varied origins. The combination of thermal and fast neutron measurements and the analysis of the appropriate aggregate decay curves with the purpose to obtain the average half life parameters affords to extend the possibilities of the techniques to the identification of the isotopic abundances in the sample under investigation.

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