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Nuclide Spectra of Activities of Thorium, Uranium Series and Application in Gamma-spectrometry of Point Technogenic Samples

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Abstract The radioactive nuclei decay of the nuclides in the ^{232}Th, ^{235}U and ^{238}U series is under consideration. The Bateman-Rubinson system of differential equations is applied. The activity of any n-th nuclide of the series has been calculated. The standard nuclide spectra are found and are compared with experimental nuclide spectra of activities. The results of the measurements are the time parameters, which determine the event date. The examples of the measurement of the experimental nuclide spectra in the nuclear gamma spectrometry of the point technogenic samples have been considered.

Keywords: *^{232}Th, ^{235}U and ^{238}U series, standard sets of nuclides, technogenic samples, nuclear gamma spectrometry, experimental nuclide spectra, standard nuclide spectra of activities, decomposition*

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1. Introduction

Investigated samples can be considered as a chemical systems, i. e. as an ordered set of nuclides (chemical elements). Chemical systems are the generalization of the geochemical, biochemical and other nuclides systems in the samples. The sets of nuclides of the series ^{232}Th, ^{235}U and ^{238}U belong to such systems as well. For such systems the methods of the nuclear gamma spectrometry of environment with semiconductor detectors are used.

The gamma spectrometry of the sample results in the table of activities A of the gamma-active nuclides. It can be presented graphically in the form of the experimental nuclide spectrum of the nuclide activity A [1]. A comparative geochemical analysis on the basis of nuclides spectra, in which the nuclides ^{238}U, ^{234}U, ^{230}Th, ^{226}Ra (series ^{238}U); ^{232}Th, ^{228}Ra, ^{228}Ac (series ^{232}Th) for different samples are presented, is enough informative. Nevertheless, the nuclide spectra were not used for the dating and other time parameters determining.

The methods of nuclear chronometers (the method of uranium series) for the dating and samples age determining are used. Such methods are based on the application of the equation

$$t = -\frac{1}{\lambda_D}\ln\left[1-\frac{A_D}{A_P}\right]. \qquad (1)$$

Note that such methods are valid only for the closed chemical systems of nuclides.

These methods relate the activities of the parent A_P and the daughter A_D nuclides; λ_D is the decay constant. It is, e. g., a pair of long-lived nuclides A_D ^{234}U and A_P ^{238}U; A_D ^{230}Th and A_P ^{234}U; A_D ^{231}Pa and A_P ^{235}U. The equation (1) for the nuclide couple's activities calculations is found after a number of assumptions on the basis of the Bateman equations [2,3].

Note that a detail derivation of the equation (1) is presented in [4,5]. Its analysis and mathematical backgrounds are beyond this brief article. The examples and problems of the nuclear chronometers methods application are considered in many investigations, see, e. g., [1,4,5,6,7,8].

The evident increasing and complication of problems, e. g. the identification of the technogenic materials with the radioactive nuclides of the ^{232}Th, ^{235}U and ^{238}U series, puts forward new demands to the existing methods.

Our investigation is the search for further development of the gamma spectrometric variant of the nuclear chronometers method. This study is based on the measurement of the experimental nuclide spectra of activities of any nuclides of the series ^{232}Th, ^{235}U and ^{238}U. The results of its measurement, in particular, are the time parameters that determine the date of the event in the samples. The event here is the registered changes of the nuclide content of the ^{232}Th, ^{235}U and ^{238}U series [9].

2. The Solutions of the Bateman-Rubinson Equations. The Standard Set of Nuclides

Consider briefly the algorithm of the standard set method. The activity $A_n(T_e)$ of any n-th nuclide of the series can be presented by the generalized for the case of

branched chains expression, which is the solution of the Bateman-Rubinson system of differential equations [9,10,11,12,13], and has the form:

$$A_{nB}(T_e) = \lambda_n N_{nB}(T_e) = N_{10} \sum_j \left(\prod b_{ij} \right) \sum_i \lambda_i C_{ij} e^{-\lambda_i T_e}, \quad (2)$$

where A_{10} are the initial values of the parent nuclei activity; b_{ij} are the branching coefficients («internal» branching ratios, which values are the nuclear constants); $b_{ij} \leq 1$; C_{ij} are the coefficients, which depends on decay constants λ_{ij}, T_e is the proper time, which is present in the activities calculations.

The calculations are performed for:

- one mole of a substance: $N_{10} = N_A = 6{,}022 \cdot 10^{23}$ nuclei (Avogadro's number);

- the proper time T_e ^{232}Th (from 0 to $3{,}17 \cdot 10^{12}$ years); ^{235}U (from 0 to $3{,}17 \cdot 10^{12}$ years); ^{238}U (from 0 to $3{,}17 \cdot 10^{12}$ years).

The nuclear data are taken from [14]. Branching ratios b_i with the formation of clusters are not considered.

The following conditions are chosen for solving of the Bateman-Rubinson system of differential equations:

- the activities of all nuclides except parent (activity A_{10}) in the proper time moment T_e=0 are taken equal to zero;

- for an arbitrary time moment T_e any other possibilities of nuclei entrance and losses except the processes of decay and formation are absent.

A set of all these conditions, of the analytical expressions (2) and theirs solutions are called the standard conditions.

An ordered set of all radionuclides of one series, which activities $A(T_e)$ are determined by the expression (2), we call the standard set of this series. The standard nuclide set (SNS) is the ordered list of one series nuclides that are linked genetically by the mutual decay and formation [9]. The standard nuclide spectra of activities are the quantitative characteristic of the standard sets.

The result of event, as was indicated, is the registrated change of the nuclide content in the sample. It leads to the standard nuclide sets formation of the new daughter series [9]. We called these sets the standard daughter nuclide sets. The result of their formation will be the non-standard sets appearance. The non-standard set is the composition (sum) of two (and more) standard sets, which formation is caused by the event.

The experimental nuclide sets it is the nuclear gamma spectrometry registrated manifold of the parent and daughter ^{232}Th, ^{235}U and ^{238}U nuclide series in the samples. The standard and non-standard sets can be their models.

The experimental nuclide spectra of activities are the quantitative characteristics of the experimental nuclide sets. Indeed, the experimental nuclide spectra it is the ordered sequence of the experimental values of the gamma activities of the A nuclides of the ^{232}Th, ^{235}U and ^{238}U sets.

2.1. Tables of Standards

We have found the time dependence of the activities $A=f(T_e)$ for all nuclides of the ^{232}Th, ^{235}U and ^{238}U series. For these purposes the calculations of the values of the activities in (2) for the enough big number of points of the proper time T_e is applied. The table found is recalculated to the experimental values of the activities A. The table of standards is obtained as a result. Any row of this table is the standard nuclide spectrum of activities that is the standard for the concrete value of the proper time T_e. The values of activities for the corresponding nuclide series are given in the columns of the table.

2.2. The Measurement of the Experimental Nuclide Spectrum of Activities in the Table of Standards

The measurement is in comparison of the experimental nuclide spectrum ENS (T_m) with the corresponding standard that is the standard nuclide spectrum SNS (T_e):

$$\mathrm{ENS}(T_m) = \mathrm{SNS}(T_e). \quad (3)$$

The experimental nuclide spectrum corresponds to the standard conditions if the equality (3) is valid.

Measuring the gamma activities of scales we are working in the time scale T_m. It is the date (an ordinary calendar time). The calculations of the nuclide gamma activities are fulfilled in the scale of proper time T_e. The comparison (3) put into correspondence the proper time T_e to the date time T_m. The value of this proper time T_e is interpreted as the age (the duration of the existence) of the standard nuclide set, which is formed as a consequence of the event.

However, the nuclide activities, which form an experimental nuclide spectra, are the experimental values. These values are not determined in some sense. Moreover, the experimental set can be a nonstandard set. Therefore, the standard nuclide spectrum, which is equal exactly to the experimental nuclide spectrum, cannot be found. Thus, for the activity $A_n(T_m)$ of n–th nuclide the necessity of taking into account the uncertainties results in the inequality:

$$A(T_{e\min}) \leq A_n(T_m) \leq A(T_{e\max}), \quad (4)$$

where $A(T_{emin})$ belongs to the standard SNS $A(T_{emin})$; $A(T_{emax})$ belongs to the standard SNS $A(T_{emax})$; $A_n(T_m)$ belongs to the experimental nuclide spectrum ENS (T_m).

Since SNS $A(T_{emin})$ and SNS $A(T_{emax})$ are the standard spectra then the experimental nuclide spectrum ENS (T_m) satisfies the standard conditions as well.

By choosing the SNS $A(T_{emin})$ and the SNS $A(T_{emax})$ from the table of standards, we choose the meaning of the times T_{emin} and T_{emax}. Thereat, the time interval ΔT_e and its mean value ΔT_{mid}.

$$\Delta T_e = T_{e\max} - T_{e\min}, \quad (5)$$

$$\Delta T_{mid} = \frac{\left(T_{e\max} + T_{e\min} \right)}{2} \quad (6)$$

are determined. The value ΔT can be considered not only as an uncertainty of time T_e, but as some “interval of standardization” of the experimental nuclide spectrum ENS (T_m) as well. Inside this interval ΔT the experimental set is considered as the standard set.

2.3. Decomposition of the Experimental Nuclide Spectra

In order to determine the parent nuclide spectrum PNS or daughter nuclide spectrum DNS it is necessary to fulfill

the decomposition of the experimental nuclide spectrum ENS. The nuclide spectra peculiarity is as follows. It is the result of term by term subtraction of the corresponding eponymous activities of the experimental and standard spectra.

In this case one can write:

$$\text{ENS} = \text{PNS} + \text{DNS};$$
$$\text{PNS} = \text{ENS} - \text{DNS};$$
$$\text{DNS} = \text{ENS} - \text{PNS}.$$

Procedure of decomposition allows to find the standard sets on the basis of the experimental spectrum. Moreover, it allows to determine their standard nuclide spectra and theirs time coordinates T_e in the tables of standards. After the decomposition one of the known method of nuclear chronometers can be used for the determination of the proper time T_e.

Therefore, the pairs of nuclides, which are used in the methods of nuclear chronometers (1), contains in the standard set of all nuclides of the series. Thus, the standard nuclide set as a whole can be considered independently in the role of nuclear chronometer.

3. The Experimental Part

In this article the data for a few point technogenic samples are presented. These samples have a low but enhanced (in comparison with natural) activity. (The samples with higher, in comparison with natural, activity.) The radioactive source is called point if its size parameters (e. g. height h and radius r) are essentially lower (in 5-10 times) from the corresponding parameters of the detector (h = 60 mm; r = 50 mm). Such point-like conditions were used in the selection of the samples. Choosing the point samples we eliminated the problems of self-absorption, standard selection etc., during the samples gamma-spectroscopy.

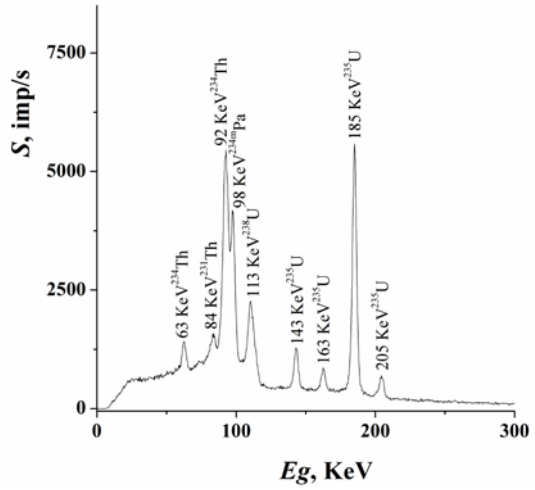

Figure 1. The characteristic apparatus gamma-spectrum of the technogenic sample

Apparatus gamma-spectra of technogenic samples found at HPGe-detector are 150 sm^3 (energy resolution – 2 KeV for the line 1332 KeV ^{60}Co), and at Ge(Li)-detector are 100 sm^3 (energy resolution – 3,9 KeV for the line 1332 KeV ^{60}Co). The passive multilayered low background detector defense is used. The samples are measured without any special preparation. The samples impermeability was guaranteed. The length of the measurement is 2–4 hours. The characteristic apparatus gamma-spectrum of the technogenic sample is given on the Figure 1.

4. Measurement Errors

Program package of SEG-40-Ge complex determines the absolute ($\Delta A(T_e)$) and the relative ($\delta_{\Delta A(Te)}$) measurement errors of the activities $A(T_e)$. The values of these errors are influenced at the determination of such important quantities as the time interval ΔT values errors and errors due to decomposition.

The appearance of the time interval ΔT and its value are the consequences of the different types measurement errors and uncertainties. The values T_{emin}, T_{emax} from the formula (2) cannot be determine analytically. Therefore, the assertion is accepted. For the monotonically increasing (decreasing) time dependences $A(T_e)$ the uncertainties of the times ΔT_{emin}, ΔT_{emax} are linearly proportional for the uncertainties $\Delta A(T_{emin})$, $\Delta A(T_{emax})$. Further, it is accepted that relative errors of the activities $A(T_{emin})$ and times T_e determination are equal, i. e. $\delta_{\Delta A(Te)} = \delta_{\Delta T}$. Therefore, if the relative errors $\delta A(T_{emin})$ and $\delta A(T_{emax})$ are known, then the absolute errors can be determined in terms of these relative errors:

$$\Delta A(T_{e\min}) = \delta A(T_{e\min}) \cdot A(T_{e\min});$$
$$\Delta A(T_{e\max}) = \delta A(T_{e\max}) \cdot A(T_{e\max}).$$

The relative error $\delta_{\Delta T}$ of the time interval ΔT value is given by

$$\delta_{\Delta T} = \begin{bmatrix} (\delta A(T_{emin}) \cdot A(T_{emin}))^2 \\ + (\delta A(T_{emax}) \cdot A(T_{emax}))^2 \end{bmatrix}^{1/2} / \begin{pmatrix} A(T_{e\max}) \\ -A(T_{e\min}) \end{pmatrix}.$$

The decomposition of spectra is the term by term subtraction of the experimental nuclide spectrum activities $A_n(T_m)$ from the eponymous activity of some standard $A_{ne}(T_m)$:

$$\Delta A_n(T_m) = A_n(T_m) - A_{ne}(T_m).$$

The value $A_n(T_m)$ is the experimental quantity. The value $A_{ne}(T_m)$ is the calculating quantity, which is determined on the basis of its parent nuclide activity. The relative error of the value $\Delta A_n(T_m)$ determination is given by

$$\delta\Delta A_{\text{n}}(T_{\text{m}}) = \begin{bmatrix} (\delta A_n(T_m) \cdot A(T_m))^2 \\ +(\delta A_{ne}(T_m) \cdot A_{ne}(T_m))^2 \end{bmatrix}^{1/2} / \begin{pmatrix} A_n(T_m) \\ -A_{ne}(T_m) \end{pmatrix}.$$

For the problems, where the decomposition is necessary, the found value of the relative error $\delta\Delta A_n(T_m)$ will be taken into account in the formulae for the error $\delta_{\Delta T}$ of time interval ΔT determination.

In the method of the nuclear chronometers the formula

$$\delta_t = \sqrt{\delta^2 \cdot \delta A_M + \delta^2 \cdot \delta A_D}$$

is used for the relative time error δ_t evaluation. Here δA_M, δA_D are the relative errors of the corresponding activities determination.

5. The Results of the Investigation

The standard sets determination includes the determination of the experimental nuclide spectra of activities ENS $A(T_m)$ in the table of standards. It finds a graphic mapping, respectively.

Figure 1 gives experimental nuclide spectrum ENS of A sample 1th and standards of comparison SNS $A(T_{emin})$ and SNS $A(T_{emax})$, which satisfy the condition (4). Experimental nuclide spectrum of the sample represents the standard nuclide set of the series ^{232}Th. Fig. 3 shows the positions of the experimental nuclide spectrum ENS A and the standards of comparison SNS $A(T_{emin})$ and SNS $A(T_{emax})$ at the nuclide activities time dependence of the series ^{232}Th. Imaginary they look like the points of intersection of the vertical line with these dependences and are presented as the rectangle. The width of «the rectangle» is given by the value of the time interval $\Delta T = T_{emax} - T_{emin}$. This interval represents the uncertainty of the position of the experimental nuclide spectrum ENS A given by the formula (4). The given measurement errors $\delta_{\Delta A(Te)}=\delta_{\Delta T}$ are found in condition that the activity determination error is equal to 5%.

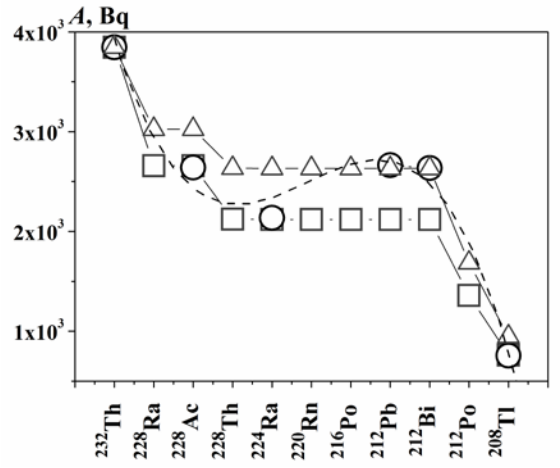

Figure 2. The experimental nuclide spectrum of activities of the sample 1th (circles – dashed line), and the standard spectra SNS $A(T_{emin})$ (four squares) and SNS $A(T_{emax})$ ^{232}Th

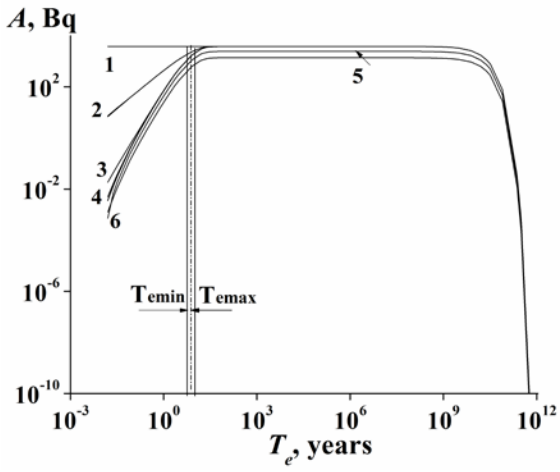

Figure 3. Time dependence of the activities of the standard nuclide set of the series ^{232}Th (1 – ^{232}Th, 2 – ^{228}Ra, ^{228}Ac, 3 – ^{228}Th, ^{224}Ra, 4 – ^{220}Rn, ^{216}Po, ^{212}Pb, ^{212}Bi, 5 – ^{212}Po, 6 – ^{208}Tl) of the sample 1th, time interval ΔT (in the form of the rectangle) and T_{mid} (dashed line in the rectangle)

Figure 4 shows experimental nuclide spectrum ENS A of the sample 10s and the comparison standards SNS $A(T_{emin})$ and SNS $A(T_{emax})$. Given sample contains the material, which contains the noticeable activities of the daughter nuclides ^{238}U – ^{230}Th, ^{226}Ra, ^{214}Pb, ^{214}Bi. Activities of the parent ^{238}U and daughter ^{234}Th are relatively smaller. Therefore, the sample contains a nonstandard set. It is a composition of two standard sets: the standard set ^{238}U and the standard set ^{230}Th. We fulfill the decomposition, by selecting at first the standard set ^{238}U, and after that the standard set ^{230}Th: DNS ^{230}Th = ENS $A(T_e)$ – PNS ^{238}U. The result after decomposition is given by the Figure 5.

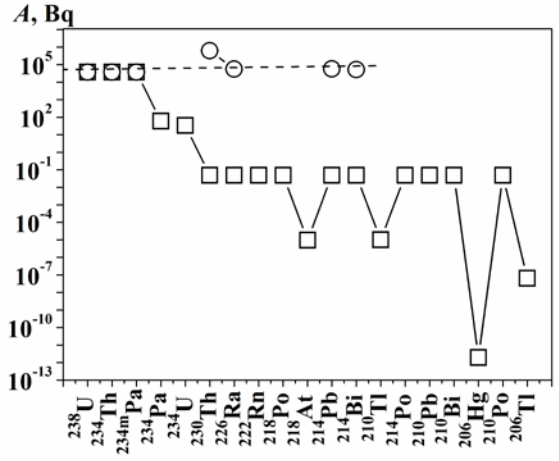

Figure 4. The experimental nuclide spectrum of activities of the sample 10s (circles – dashed line) and the standard spectrum SNS (four squares) ^{238}U before the decomposition

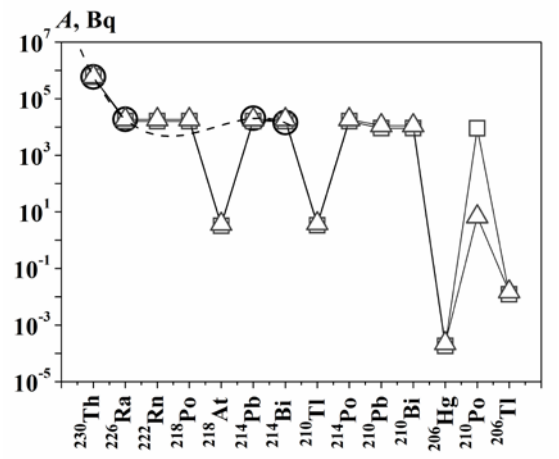

Figure 5. The experimental nuclide spectrum of activities of the sample 10s (circles – dashed line), and the standard spectrum SNS $A(T_{emin})$ (four squares) and SNS $A(T_{emax})$ (rectangles) ^{230}Th after the decomposition

The results of the measurement are given in the table. Moreover, the data for the nuclear chronometers method are given in the table as well. We determine, as it was already considered above, the time interval ΔT_{mid} (the formula (6)) of experimental set of the nuclides of the series. The time t (formula (1)) is determined in the method of nuclear chronometers. The conclusion about the satisfactory comparability between the values found with the help of two data methods can be suggested by the comparison of the values ΔT_{mid} and t.

The experimental nuclide spectra of the samples 1th, 2th satisfy the condition (4). It means that the main

component of their experimental sets are the standard sets ^{232}Th.

The experimental nuclide spectra of the samples 3u – 10u satisfy the condition (4). The standard sets method data and the nuclear chronometers data testify about the satisfactory comparability between each other. The standard sets of ^{238}U are the main parts of their experimental sets. Existence duration large values of the determined standard set of the ^{238}U (10^7 – 10^8 years) allowed to suggest that they were formed enough long ago (maybe in the epoch of the intensive geologic processes) and were not changed from these times.

The experimental nuclide spectra of the samples 10s – 13s do not satisfy the condition (4). After the decomposition of the experimental sets the standard sets and nuclear chronometers methods data are comparable within the errors. Therefore, the result of the decomposition demonstrates that the experimental nuclide set of the samples 10s – 13s can be represented as the composition of the standard set of ^{238}U and the standard nuclide set of ^{230}Th.

Relatively large values of the measurement errors can be localized by the decreasing of the measurement duration.

Table 1. Comparison of the Standard Sets and the Nuclear Chronometers Methods

№	Samples	Standard set (SS)	ΔT_{mid}, years	Error $\delta_{\Delta T}$, %	Nuclear chronometer	t, years	Error, δ_t, %
1	1th	^{232}Th	8	11	^{228}Th/^{232}Th	0,12	10
2	2th	^{232}Th	8	11	^{228}Th/^{232}Th	0,43	10
3	3u	^{238}U	$3\cdot10^7$	12	^{234}U/^{238}U	$3\cdot10^7$	14
4	4u	^{238}U	$3\cdot10^7$	14	^{234}U/^{238}U	$3\cdot10^7$	14
5	5u	^{238}U	$1{,}2\cdot10^7$	14	^{234}U/^{238}U	$1\cdot10^7$	16
6	6u	^{238}U	$2\cdot10^8$	18	^{234}U/^{238}U	$2\cdot10^8$	11
7	7u	^{238}U	$8\cdot10^7$	12	^{234}U/^{238}U	$8\cdot10^7$	12
8	8u	^{238}U	$5\cdot10^8$	10	^{234}U/^{238}U	$3{,}8\cdot10^8$	20
9	9u	^{238}U	$2{,}8\cdot10^8$	18	^{234}U/^{238}U	$2{,}8\cdot10^8$	19
10	10u	^{238}U	$1{,}2\cdot10^8$	21	^{234}U/^{238}U	$1{,}4\cdot10^8$	15
11	10s b*	^{238}U	$3{,}2\cdot10^8$	13	^{226}Ra/^{230}Th	117	14
	10s a*	^{230}Th	76	15	^{226}Ra/^{230}Th	74	14
12	11s b*	^{238}U	$1{,}6\cdot10^8$	17	^{226}Ra/^{230}Th	156	11
	11s a*	^{230}Th	73	14	^{226}Ra/^{230}Th	73	11
13	12s b*	^{238}U	$8\cdot10^8$	17	^{226}Ra/^{230}Th	209	12
	12s a*	^{230}Th	82	10	^{226}Ra/^{230}Th	85	12
14	13s b*	^{238}U	$2{,}4\cdot10^8$	20	^{226}Ra/^{230}Th	102	15
	13s a*	^{230}Th	71	23	^{226}Ra/^{230}Th	77	15

*b –before the decomposition
*a –after the decomposition
*c – a satisfactory pair of the nuclear chronometers is not found.

6. Conclusions

The standard nuclide spectra of the nuclide activities A of series ^{232}Th, ^{235}U, ^{238}U have been calculated. The table of standards by the nuclide activities A time dependence is created. Each spectrum from the table of standards is shown to be the standard itself. The experimental nuclide spectrum of activities A, by the way of its comparison with the standards of the standard nuclide spectrum A, has been measured. The measurement result is the time coordinate T_e, which is the position from the experimental nuclide spectrum of activities A in the table of standards. The found value can be interpreted as the duration of the standard set existence. Here the duration of the existence is considered as the time interval started from the moment of the event, in which this event appeared, and finished in the moment of the measurement.

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