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INTEGRAL EVALUATION OF ^{239}Pu AND ^{240}Pu MASSES IN THE CHERNOBYL FALLOUT ON THE TERRITORY OF BELARUS

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The activity ratio of the γ-radioactive fission fragments such as ^{95}Zr, ^{103,106}Ru, ^{141,144}Ce for the evaluation of the total α-activity of ^{239}Pu + ^{240}Pu isotopes in the soil of Gomel and Mogilev regions of Belarus is presented. We have taken into account a possible fluctuation of these ratios around their average values. Experimental data for several settlements in Gomel and Mogilev regions were also analysed, and a good correlation between theoretical evaluation and experimental data was obtained.

Keywords: radionuclide, isotope activity ratio, fission fragments, ^{239,240}Pu, activity and mass evaluation

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1. Introduction

As in any other thermal reactor, there is quite definite interrelation between specific activities of nuclides accumulated in the core of the RBMK-1000 reactor. It can be used for solving practical problems, in particular, for the analysis of the structure in the Chernobyl NPP accident radionuclide fallout. The estimation of radionuclide inventories and calculation of ratios between specific activities of radionuclides in the core of the reactor that are necessary for this analysis do not present any difficulties, and it has been done, for example, in [1, 2]. Therefore, they were widely used in the analysis of radioactive contamination of the Chernobyl origin in a number of scientific works, see [3]. Such ratios can especially be useful for indirect evaluation of levels of the territory contamination with γ-radiating fission fragments of transuranium elements such as ^{95}Zr, ^{103}Ru, ^{106}Ru, ^{141}Ce, ^{144}Ce, etc. For example, within the limits of the information system "Proba" such analysis has been made. Using data on the γ-radiation of ^{144}Ce, the map of the territories contaminated with ^{239}Pu + ^{240}Pu isotopes in the zone close to the Chernobyl NPP was made [3, 4]. It is known, however, that such an approach is not always correct during local measurements. Larger divergences between the results of radiochemical analysis and theoretical predictions about the ratios of the type $A(^{239,240}\mathrm{Pu}) \sim A(^{144}\mathrm{Ce})$, $A(^{239,240}\mathrm{Pu}) \sim A(^{103}\mathrm{Ru})$, etc., were regularly observed. It is clear, because the specified ratios between activities of the type $A(^{239,240}\mathrm{Pu})$ and $A(^{144}\mathrm{Ce})$ on the average were usually determined in the core and at a definite average depth of the nuclear fuel burnup W. Thus, we actually deal with the ratios of the type $\langle A(^{239,240}\mathrm{Pu})\rangle \sim \langle A(^{144}\mathrm{Ce})\rangle$. For general reasons, the use of such average evaluations should have a limited character. It is necessary to consider possible fluctuations of the nuclide specific activity ratios relative to their average.

We will proceed from the assumption that the burnup W in the nuclear fuel is described by the Poisson distribution $P_W = (\langle W\rangle)^W \mathrm{e}^{-\langle W\rangle}/W$. For the RBMK-1000 reactor of the Chernobyl NPP, the burnup W varies within the limits of 0–18.5 at the average depth of the burnup $\langle W\rangle = 11$, dispersions $D_W = \langle W\rangle = 11$, and a standard deviation $\Delta = \sqrt{D_W} \approx 3.32$. In some cases, it is more convenient to use the Gaussian distribution $P(W)\,\mathrm{d}W = (\sqrt{2\pi\langle W\rangle})^{-1} \exp[-(W-\langle W\rangle)^2/(2\langle W\rangle)]\,\mathrm{d}W$ which is connected with the Poisson distribution.

The technique of average calculation of power and exponential functions $f(W)$ by the Poisson and Gaussian distributions is described in [5]. Therefore, for the most simple function $f(W) = W$ the average of $f(W)$ is equal to $\langle W\rangle$, dispersion is $D\,[f(W)] = \langle W\rangle$, and the standard deviation is $\Delta\,[f(W)] = \sqrt{\langle W\rangle}$. The Gaussian function $P\,(|y|) = 2\Phi(|y|/\Delta)$ defines probability that abmodality of the burnup W module

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Table 1. Dependence of functions $v(\mathrm{Ru}, W)$, $v(\mathrm{Pu}, W)$, $\alpha v(\mathrm{Ru}, W)$, and $\alpha v(\mathrm{Pu}, W)$ on the burnup W.

t, year	W	$v(\mathrm{Ru}, W)$	$\alpha v(\mathrm{Ru}, W)$	$\alpha v(\mathrm{Ru}, W)/W$	$v(\mathrm{Pu}, W)$	$\alpha v(\mathrm{Pu}, W)$	$\alpha v(\mathrm{Pu}, W)/W$
0.15	1.06	$2.64{\cdot}10^{-2}$	1.31	1.24	$1.41{\cdot}10^{-1}$	1.12	1.06
0.30	2.12	$4.31{\cdot}10^{-2}$	2.12	1.00	$2.85{\cdot}10^{-1}$	2.27	1.07
0.45	3.18	$6.25{\cdot}10^{-2}$	3.08	0.97	$4.25{\cdot}10^{-1}$	3.39	107
0.60	4.24	$8.33{\cdot}10^{-2}$	4.14	0.97	$5.61{\cdot}10^{-1}$	4.47	1.05
0.75	5.30	$1.06{\cdot}10^{-1}$	5.25	0.99	$6.95{\cdot}10^{-1}$	5.54	1.04
0.90	6.36	$1.28{\cdot}10^{-1}$	6.34	1.00	$8.25{\cdot}10^{-1}$	6.58	1.03
1.05	7.42	$1.51{\cdot}10^{-1}$	7.48	1.01	$9.45{\cdot}10^{-1}$	7.53	1.01
1.20	8.48	$1.72{\cdot}10^{-1}$	8.52	1.00	1.07	8.53	1.01
1.35	9.54	$1.94{\cdot}10^{-1}$	9.61	1.01	1.20	9.56	1.00
1.50	10.6	$2.14{\cdot}10^{-1}$	10.6	1.00	1.33	10.6	1.00
1.65	11.7	$2.33{\cdot}10^{-1}$	11.5	0.99	1.47	11.7	1.01
1.80	12.7	$2.53{\cdot}10^{-1}$	12.5	0.99	1.61	12.8	1.01
1.95	13.8	$2.74{\cdot}10^{-1}$	13.6	0.98	1.77	14.1	1.02
2.10	14.8	$2.99{\cdot}10^{-1}$	14.6	0.98	1.92	15.5	1.04
2.25	15.9	$3.12{\cdot}10^{-1}$	15.4	0.97	2.07	16.5	1.04
2.40	17.0	$3.27{\cdot}10^{-1}$	16.2	0.96	2.23	17.8	1.05
2.55	18.0	$3.40{\cdot}10^{-1}$	16.9	0.93	2.40	19.1	1.06
2.70	19.1	$3.56{\cdot}10^{-1}$	17.6	0.92	2.56	20.4	1.07

from the average is within the limits of $|y| = |W - \langle W\rangle|$.

It is obvious that the knowledge of the average burnup $\langle W\rangle$ per one tonne of fuel does not mean that we will determine this value of the burnup per one tonne of casually chosen fuel.

The burnup W will be in the interval $\langle W\rangle \pm \Delta$ with the probability of ∼68%, which means $W \sim \langle W\rangle \pm \sqrt{\langle W\rangle}$. Abmodality of the burnup W from the average does not exceed 30%. Similarly, the burnup W is in the interval $\langle W\rangle \pm 2\Delta$ with the probability of 95%, and $W \sim \langle W\rangle \pm 2\sqrt{\langle W\rangle}$. The burnup $W \sim 11{\pm}6.6$ is also in the interval 4.4–17.6 and the abmodality from the average $\langle W\rangle = 11$ is within the limits of 60%. If we consider a case when W slightly differs from $\langle W\rangle$, the probability of such event will be considerably lower. For example, the probability will be $P(|y|) \approx 0.08$ if we have $|y|/\Delta \leq 0.1$.

However, it is possible to use correlated ratios $A(^{239,240}\mathrm{Pu}) \sim A(^{144}\mathrm{Ce})$, $A(^{239,240}\mathrm{Pu}) \sim A(^{103}\mathrm{Ru})$, etc. for the evaluation of activities and masses of plutonium isotopes on large territories. In this case they average naturally. Thus, referring to $^{95}\mathrm{Zr}$, $^{103}\mathrm{Ru}$, $^{106}\mathrm{Ru}$, $^{141}\mathrm{Ce}$, $^{144}\mathrm{Ce}$ nuclide amounts it is possible to evaluate the amount of the fallen out plutonium isotopes $^{239}\mathrm{Pu}$, $^{240}\mathrm{Pu}$ and other isotopes of transuranium elements. It is easy to calculate the activity ratios $\langle A(^{239,240}\mathrm{Pu})\rangle/\langle A(^{144}\mathrm{Ce})\rangle$, $\langle A(^{239,240}\mathrm{Pu})\rangle/\langle A(^{103}\mathrm{Ru})\rangle$, $\langle A(^{239,240}\mathrm{Pu})\rangle/\langle A(^{106}\mathrm{Ru})\rangle$ for the core of Unit 4 of the RBMK-1000 reactor of the Chernobyl NPP.

The aim of the present study is to calculate such ratios for Unit 4 of the RBMK-1000 reactor of the Chernobyl NPP and using this ratio to evaluate the amount of $^{239}\mathrm{Pu}$ and $^{240}\mathrm{Pu}$ isotopes fallen out on the territory of Belarus after the accident.

2. Sampling and methods

Let us assume that instead of the burnup W we take the function $v(W) \approx W/\alpha$, where a is a constant. In this case the average of $v(W)$ is equal to $\langle W\rangle/\alpha$, dispersion is $D[v(W)] = \langle W\rangle/\alpha^2$, and the standard deviation is $\Delta[v(W)] = \sqrt{\langle W\rangle}/\alpha$. Therefore, the $v(W)$ function value should be in the interval $(4.4\text{–}17.6)/\alpha$ with the probability of about 95%. The abmodality from the average $11/\alpha$ is within the limits of 60%.

The indirect definition of the α-radiating plutonium isotope concentration in the Chernobyl NPP accident fallout by γ-radiation of some accompanying fission fragments is related to the correct choice of $v(W) \approx W/\alpha$ type functions.

Function $v(W)$ should be approximately linear according to the burnup W. Thus, the constant α can be calculated theoretically in some cases, and in other cases it is defined experimentally. Further we will deal with functions of the $v(W) = A(W)_1/A(W)_2$ type, where $A(W)_1$ and $A(W)_2$ are calculated specific activities of X_1 and X_2 nuclides in the active zone of the RBMK-1000 reactor depending on the burnup W or experimental density of soil contamination levels.

Function $v(W)$ should be proportional to the burnup W. Thus, it is simple to use $A(W)_1$ as specific activi-

Table 2. Parameters of specific activity ratios $v(\mathrm{Ru}) = A(^{106}\mathrm{Ru})/A(^{103}\mathrm{Ru})$, $v(\mathrm{Pu}) = A(^{240}\mathrm{Pu})/A(^{239}\mathrm{Pu})$ for the RBMK-1000 of the Chernobyl NPP.

$f(W)$	W	$v(\mathrm{Ru})$	$v(\mathrm{Pu})$
α	1	49.5	7.97
$\langle v\rangle$	11	0.222	1.38
$D(v)$	11	$4.49{\cdot}10^{-3}$	$1.73{\cdot}10^{-1}$
$\Delta(v)$	$\sqrt{11}$	$6.7{\cdot}10^{-2}$	$4.16{\cdot}10^{-1}$

ties of nuclides with close to linear dependence according to the burnup W, and $A(W)_2$ as specific activities of nuclides with slight dependence on the burnup.

Specific activities of ^{90}Sr and ^{137}Cs practically linearly depend on the burnup W. Specific activity of ^{106}Ru, total activity of plutonium isotopes ^{239}Pu + ^{240}Pu $A(^{239,240}\mathrm{Pu})$ and ^{238}Pu + ^{239}Pu + ^{240}Pu $A(^{238,239,240}\mathrm{Pu})$ show close to linear dependence on the burnup W. The dependence of specific activity of nuclides with rather short half-life periods (approximately up to 60 days) on the burnup W is slight, and independent yields of nuclides during fission of ^{235}U and ^{239}Pu (^{89}Sr, ^{95}Zr, ^{99}Mo, ^{103}Ru, ^{131}I, ^{141}Ce, etc.) are comparable by their value. Functions $v(W)$ with more complex dependences of $A(W)_1$ and $A(W)_2$ specific activities on the burnup W than those considered above can also be close to linear. For example, the function $A(^{240}\mathrm{Pu},W)/A(^{239}\mathrm{Pu},W)$ is important for practical purposes.

The results of numerical calculations of specific activities of nuclides in the RBMK-1000 reactor [6–8] and analytical calculations [9–11] were used for the calculation of $v(W)$ functions. Experimental data on soil contamination with γ-radiating fission fragments are taken from [12].

We will restrict ourselves to the determination of the ^{239}Pu + ^{240}Pu isotope total α-activity $A(^{239,240}\mathrm{Pu})$ using ^{95}Zr, ^{103}Ru, ^{106}Ru, ^{141}Ce, ^{144}Ce γ-radiation. Using ^{103}Ru and ^{106}Ru nuclides as an example we will show the method of calculations. We will consider two functions $v(\mathrm{Ru}) = A(^{106}\mathrm{Ru})/A(^{103}\mathrm{Ru})$ and $v(\mathrm{Pu}) = A(^{240}\mathrm{Pu})/A(^{239}\mathrm{Pu})$. For calculation, the results of [6, 7] are used. Corresponding functions $\alpha v(\mathrm{Ru})$, $\alpha v(\mathrm{Pu})$ are obtained by the normalization of the $v(\mathrm{Ru})$, $v(\mathrm{Pu})$ functions with the use of the factor α when $W = 10.6$: $\alpha(\mathrm{Ru}) = 49.5$, $\alpha(\mathrm{Pu}) = 7.97$. Values of these functions at different burnup W are presented in Table 1. As shown in Table 1, ratios $\alpha v(\mathrm{Ru},W)$, $\alpha v(\mathrm{Pu},W)$ are very close to the function of burnup $f(W) = W$. Therefore, ratios $v(\mathrm{Ru},W)$, $v(\mathrm{Pu},W)$ within the accuracy of several percent are equal to $W/\alpha(\mathrm{Ru})$, $W/\alpha(\mathrm{Pu})$, respectively. All these functions should be described by the Gaussian distribution. The calculation of their averages, dispersions, and standard deviations (Table 2) is possible. Averages of considered specific activities are the following: $\langle A(^{103}\mathrm{Ru})\rangle = 2.74{\cdot}10^{16}$, $\langle A(^{106}\mathrm{Ru})\rangle = 6.13{\cdot}10^{15}$, $\langle A(^{239}\mathrm{Pu})\rangle = 5.43{\cdot}10^{12}$, $\langle A(^{240}\mathrm{Pu})\rangle = 7.83{\cdot}10^{12}$ Bq/t. Ratios of $\langle A(^{106}\mathrm{Ru})\rangle/\langle A(^{103}\mathrm{Ru})\rangle = 0.224$, $\langle A(^{240}\mathrm{Pu})\rangle/\langle A(^{239}\mathrm{Pu})\rangle = 1.44$ practically coincide with the average of the corresponding functions $\langle v\rangle$.

Other useful ratios of the $A_1(W)/A_2(W) \approx W/\alpha$ type can be obtained analogously. Some of them are presented in Table 3.

Calculation of averages for ratios of the $A_1(W)/A_2(W) \approx W/\alpha$ type is reduced to replacement of the burnup W by its average value $\langle W\rangle = 11$. Therefore, if we know experimental activities of γ-radiating fission fragments of ^{95}Zr, ^{103}Ru, ^{106}Ru, ^{141}Ce, ^{144}Ce, it is possible to evaluate the amount of α-radiating plutonium isotopes ^{239}Pu, ^{240}Pu and to compare the theoretical evaluation with the data of radiochemical measurements [13].

3. Results and discussion

According to [13], due to the accident in the Chernobyl NNP, 4.2 kg of ^{239}Pu or $A(^{239,240}\mathrm{Pu})$ = $23{\cdot}10^{12}$ Bq total activity of ^{239}Pu, ^{240}Pu isotopes fell out on the territory of Belarus. About 1.9 kg of ^{239}Pu fell out on the Belarus territory of the 30-km zone. The rest of plutonium was dispersed over larger distances (see, e. g. [14]).

The most correct experimental density of ^{95}Zr, ^{103}Ru, ^{106}Ru, ^{141}Ce, ^{144}Ce radionuclide contamination in Gomel and Mogilyov regions is provided in [15]. Data on the average density of soil contamination $\langle$A(X)$\rangle$ (Ci/km^2) of some areas (area S is given in km^2 in brackets) of the specified regions (* for Mogilyov) according to [12, 15] are presented in Table 4.

As shown in Table 4, average densities of $\langle A(^{95}\mathrm{Zr})\rangle$, $\langle A(^{141}\mathrm{Ce})\rangle$, $\langle A(^{144}\mathrm{Ce})\rangle$ contamination are lower by about an order than the average density of $\langle A(^{103}\mathrm{Ru})\rangle$, $\langle A(^{106}\mathrm{Ru})\rangle$ contamination in all districts of the Mogilyov region at a 200 km distance from the Chernobyl NPP. The same situation, except for southern areas (Braginsky, Narovlyansky, Choyniksky), is observed in other northern districts of the Gomel region.

In the γ-spectrum of fuel fragments calculated for the average burnup $\langle W\rangle \approx 11$ for relative activities

Table 3. Some useful ratios of the $A_1(W)/A_2(W) \approx W/\alpha$ type.

$A_1(W)$	$A_2(W)$	W/α	$A_1(W)$	$A_2(W)$	W/α
$A(^{240}\mathrm{Pu})$	$A(^{103}\mathrm{Ru})$	$W/(3.9{\cdot}10^4)$	$A(^{103}\mathrm{Ru})$	$A(^{239}\mathrm{Pu})$	$W/(1.02{\cdot}10^{-2})$
$A(^{240}\mathrm{Pu})$	$A(^{141}\mathrm{Ce})$	$W/(4.82{\cdot}10^4)$	$A(^{240}\mathrm{Pu})$	$A(^{144}\mathrm{Ce})$	$W/(3.15{\cdot}10^4)$
$A(^{239,240}\mathrm{Pu})$	$A(^{141}\mathrm{Ce})$	$W/(2.74{\cdot}10^4)$	$A(^{239,240}\mathrm{Pu})$	$A(^{103}\mathrm{Ru})$	$W/(2.2{\cdot}10^4)$
$A(^{239,240}\mathrm{Pu})$	$A(^{95}\mathrm{Zr})$	$W/(2.88{\cdot}10^4)$	$A(^{238,239,240}\mathrm{Pu})$	$A(^{103}\mathrm{Ru})$	$W/(1.76{\cdot}10^4)$
$A(^{238,239,240}\mathrm{Pu})$	$A(^{95}\mathrm{Zr})$	$W/(2.29{\cdot}10^4)$	$A(^{238,239,240}\mathrm{Pu})$	$A(^{144}\mathrm{Ce})$	$W/(1.43{\cdot}10^4)$

Table 4. Average density of soil contamination with ^{95}Zr, ^{103}Ru, ^{106}Ru, ^{141}Ce, ^{144}Ce in some areas of Gomel and Mogilyov (*) regions.

		$\langle A$ (X)$\rangle$, Ci/km^2				
No	Site (km^2)	^{95}Zr	^{141}Ce	^{144}Ce	^{103}Ru	^{106}Ru
1	Braginsky (1900)	43.0	38.0	24.0	54.0	11.0
2	B. Koshelevsky (1600)	3.1	4.6	2.4	18.0	4.9
3	Vetkovsky (1600)	3.7	5.3	2.7	47.0	11.0
4	Gomelsky (2100)	2.4	2.9	1.8	3.8	0.84
5	Dobrushsky (1500)	5.6	7.7	4.0	40.0	8.9
6	Elsky (1400)	5.4	5.9	3.7	19.0	3.6
7	Zhlobinsky (2100)	3.1	3.1	2.2	5.6	1.3
8	Kalinkovichsky (2800)	10.0	11.0	6.6	9.8	2.1
9	Kormiansky (900)	1.7	2.8	1.1	28.0	5.7
10	Kostiukovichsky* (1500)	0.71	1.3	0.58	35.0	7.3
11	Krasnopolsky* (1200)	1.1	1.7	0.76	36.0	6.9
12	Lelchitsky (3200)	2.6	3.1	2.3	8.6	1.8
13	Loevsky(1000)	7.7	7.7	5.6	11.0	2.5
14	Mozyrsky (1600)	8.1	7.2	4.6	8.9	1.4
15	Narovliansky (1600)	21.0	24.0	13.0	54.0	10.0
16	Rechitsky (2100)	10.0	10.0	6.2	11.0	2.4
17	Rogachevsky (2100)	1.5	1.4	1.1	6.3	1.5
18	Slavgorodsky* (1300)	0.96	1.5	0.56	23.0	4.6
19	Choiniksky (2000)	54.0	49.0	31.0	69.0	14.0
20	Cherikovsky* (2000)	1.0	1.8	0.80	30.0	6.3
21	Chechersky (2100)	1.6	3.5	1.8	32.0	7.8

Table 5. Experimental data on $\sigma(X)$ ^{95}Zr, ^{103}Ru, ^{106}Ru, ^{141}Ce, ^{144}Ce amounts, the calculated amounts of plutonium $\sigma(\mathrm{Pu}, X)$ $^{239}\mathrm{Pu} + {}^{240}\mathrm{Pu}$ (Bq) and masses $m(^{239}\mathrm{Pu}, X)$ (kg).

Nuclide	$\sigma(X)$	$\langle A(^{239,240}\mathrm{Pu})\rangle/\langle A(X)\rangle$	$\sigma(\mathrm{Pu}, X)$ on $A(X)$	$m(^{239}\mathrm{Pu},X)$
^{95}Zr	$1.30{\cdot}10^{16}$	$3.8{\cdot}10^{-4}$	$4.94{\cdot}10^{12}$	0.90
^{141}Ce	$1.32{\cdot}10^{16}$	$4.0{\cdot}10^{-4}$	$5.28{\cdot}10^{12}$	0.96
^{144}Ce	$8.05{\cdot}10^{15}$	$6.2{\cdot}10^{-4}$	$5.00{\cdot}10^{12}$	0.91
^{103}Ru	$3.34{\cdot}10^{16}$	$5.0{\cdot}10^{-4}$	$1.67{\cdot}10^{13}$	3.00
^{106}Ru	$7.03{\cdot}10^{15}$	$2.2{\cdot}10^{-3}$	$1.55{\cdot}10^{13}$	2.80

of nuclides (with respect to ^{95}Zr activity) the following condition should be satisfied: $A(^{103}\mathrm{Ru})/A(^{95}\mathrm{Zr}) : A(^{106}\mathrm{Ru})/A(^{95}\mathrm{Zr}) : A(^{141}\mathrm{Ce})/A(^{95}\mathrm{Zr}) : A(^{144}\mathrm{Ce})/A(^{95}\mathrm{Zr}) = 0.79 : 0.18 : 0.97 : 0.62$.

Experimental data for ratios of $A(^{141}\mathrm{Ce})/A(^{95}\mathrm{Zr})$, $A(^{144}\mathrm{Ce})/A(^{95}\mathrm{Zr})$ do not contradict the theoretical evaluation for fuel. However, ratios of $A(^{103}\mathrm{Ru})/A(^{95}\mathrm{Zr})$, $A(^{106}\mathrm{Ru})/A(^{95}\mathrm{Zr})$ are obviously overestimated. In general, there is nothing surprising in it. Indeed, according to [3] and references therein, in some cases the isotopes of ruthenium (together with plutonium) behave as volatile ones.

In this connection, it is interesting to evaluate the ^{95}Zr, ^{103}Ru, ^{106}Ru, ^{141}Ce, ^{144}Ce radionuclide amounts $\sigma(X) = \sum\langle A(X)\rangle_i \cdot S_i$ (i is the number of the district) according to the data presented in Table 4 and estimate the amount of plutonium nuclides $^{239}\mathrm{Pu}+{}^{240}\mathrm{Pu}$, $\sigma(\mathrm{Pu}, X) = \sum\langle A(^{239,240}\mathrm{Pu}, X)\rangle_i \cdot S_i$. Factors of recalculation of $\langle A(^{239,240}\mathrm{Pu})\rangle/\langle A(^{95}\mathrm{Zr})\rangle$, etc., are calculated theoretically.

For example, we calculated the amount $\sigma(\mathrm{Pu}, {}^{95}\mathrm{Zr})$ of plutonium isotopes ${}^{239}\mathrm{Pu}+{}^{240}\mathrm{Pu}$ according to ${}^{95}\mathrm{Zr}$. The amount of ${}^{95}\mathrm{Zr}$ is $\sigma({}^{95}\mathrm{Zr}) \approx 1.3{\cdot}10^{16}$ Bq and the amount of plutonium isotopes ${}^{239}\mathrm{Pu}+{}^{240}\mathrm{Pu}$ according to ${}^{95}\mathrm{Zr}$ is $\sigma(\mathrm{Pu}, {}^{95}\mathrm{Zr}) \approx 4.94{\cdot}10^{12}$ Bq, respectively. The part of ${}^{239}\mathrm{Pu}$ activity makes up about 0.42 of the total activity: $\sigma({}^{239}\mathrm{Pu}, {}^{95}\mathrm{Zr}) \approx 2.1{\cdot}10^{12}$ Bq. The specific activity of ${}^{239}\mathrm{Pu}$ is $A({}^{239}\mathrm{Pu})_{\mathrm{sa}} \approx 2.3{\cdot}10^{12}$ Bq/kg, i. e., the mass of fallen out ${}^{239}\mathrm{Pu}$ is $m({}^{239}\mathrm{Pu}, {}^{95}\mathrm{Zr}) \approx$ 0.9 kg. Results of calculations of all nuclides are shown in Table 5. The evaluation of the amount of activities $A({}^{239,240}\mathrm{Pu}, X)$ and mass $m({}^{239}\mathrm{Pu}, X)$ presented in Table 5 is approximate. However, the results of calculation of the ${}^{239}\mathrm{Pu}$ amount according to ${}^{95}\mathrm{Zr}$, ${}^{141}\mathrm{Ce}$, and ${}^{144}\mathrm{Ce}$ nuclides, which are rigidly connected with a fuel matrix, are several times lower than the experimental values of $m({}^{239}\mathrm{Pu}) \approx 4.2$ kg [13]. The evaluation results of ${}^{239}\mathrm{Pu}$ masses according to ${}^{103}\mathrm{Ru}$ and ${}^{106}\mathrm{Ru}$ (nuclides with a variable volatility) correlate better with the data of the experiment.

4. Conclusion

It is obvious that a part of plutonium as well as ruthenium have fallen out not in the form of nuclear fuel particles. According to [3] and to the references presented in it, oxides of ruthenium and plutonium isotopes, formed as intermediate compounds in the reaction of fuel carbonization, could be sorbed from a gas phase on submicron inert particles and be transferred over large distances from the Chernobyl NPP. The data analysis of the Mogilyov region [16, 17] has shown that the fallout of plutonium basically was in the form of aerosols and well correlated with fallouts of ${}^{103}\mathrm{Ru}$, ${}^{106}\mathrm{Ru}$, and ${}^{137}\mathrm{Cs}$ radionuclides that are similar by their nature. The analysis of the experimental data obtained in the present work has shown that a similar situation is observed as a whole in the Gomel region as well.

It is plausible that on the territory of Belarus α-radiating isotopes of transuranium elements have fallen out mainly not in the form of a fuel matrix. Therefore, the territory self-cleaning from α-radiating pollutants should be faster than if they were in the form of fuel particles. It is known that at large distances from the Chernobyl NPP ($\sim$250 km) plutonium migrates deep into soil while in the area of the 30-km zone it remains in the top layer (see, e. g. [18, 19]).

Moreover, the isotope of ${}^{241}\mathrm{Pu}$, the basic source of ${}^{241}\mathrm{Am}$, will faster diffuse deep into soil than if it all were in the form of fuel matrix elements. In this connection, long-term forecasts about levels of the territory contamination with α-radiating radionuclides taking into consideration nuclear decay of ${}^{241}\mathrm{Pu}$ into ${}^{241}\mathrm{Am}$ require an essential correction.

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^{239}Pu IR ^{240}Pu MASIŲ KOMPLEKSINIS ĮVERTINIMAS ČERNOBYLIO AE AVARIJOS IŠKRITOSE BALTARUSIJOJE

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Santrauka

RBMK-1000 reaktoriaus branduolinio kuro izotopinės sudėties bei tik jam būdingų skilimo produktų aktyvumų santykių duomenys taikyti radionuklidų, patekusių po Černobylio AE avarijos į dirvožemį Gomelio ir Mogiliovo rajonuose (Baltarusija), sudėties analizei. ^{239}Pu + ^{240}Pu suminio aktyvumo įvertinimui buvo naudoti gama skilimo produktų, tokių kaip ^{95}Zr, ^{103}Ru, ^{106}Ru, ^{141}Ce ir ^{144}Ce, aktyvumų santykiai. Dėl galimų santykių fliuktuacijų naudotos vidutinės jų vertės. Gauta, kad plutonis Černobylio AE avarijos metu pagrindinai iškrito aerozolių pavidalu, analogiškai kaip ^{103}Ru, ^{106}Ru ir ^{137}Cs, vykstant sublimacijai ant submikroninių inertinių dalelių, kai plutonis buvo tarpinėje dujinėje fazėje, vykstant branduolinio kuro karbonizacijai. Todėl Baltarusijos teritorijos savivala nuo transuraninių elementų alfa spinduolių turėtų vykti greičiau nei tuo atveju, jei plutonis būtų iškritęs branduolinio kuro matricos sudėtyje. ^{241}Pu, pagrindinis ^{141}Am atsiradimo šaltinis, iškritęs aerozolių pavidalu, dirvoje difunduos gilyn taip pat greičiau.