EVALUATION OF ISOTOPE CONTENTS OF PLUTONIUM IN REACTOR EMISSIONS BY GAMMA-RADIATION OF ^{106}Ru, ^{111}Ag AND ^{125}Sb

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It is offered to use nuclides ^{106}Ru, ^{111}Ag and ^{125}Sb for evaluation of transuranium elements content in the spent fuel of HPOR-1000 reactor. It is found that ^{125}Sb is the most suitable for estimations of emergency reactor emissions. The developed formalism can be applied for reactors such as WWR.

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1. INTRODUCTION

For estimation of transuranium element contents in 30-km zone around the Chernobyl nuclear power plant (ChNPP) along with ^{144}Ce (see for example [1]) it is offered to use ^{106}Ru ($T_{1/2}$ = 371.2 d), ^{111}Ag (7.45 d) and ^{125}Sb (2.73 years) which have the relation of total yields on ^{239}Pu and ^{235}U $\rho = y(\mathrm{X}, ^{239}\mathrm{Pu})^c / y(\mathrm{X}, ^{235}\mathrm{U})^c$ equal to ~ 11, ~ 15 and ~ 4, accordingly [2]. Therefore there are bases to believe that ^{239}Pu gives the basic contribution in accumulation of mentioned nuclides at reactor operating time within about one year and longer. Hence ^{106}Ru, ^{111}Ag and ^{125}Sb should be basically located in the same place as ^{239}Pu. The presence of γ-lines of specified nuclides in emergency reactor emissions can accordingly indicate on presence of plutonium. Besides ^{106}Ru and ^{125}Sb have quite long half-life and keep the information within several years.

In this connection it is interesting to calculate theoretically relative contributions of isotopes ^{239}Pu and ^{241}Pu in accumulated activity of ^{106}Ru, ^{111}Ag and ^{125}Sb in HPOR-1000 depending on reactor operating time. The phenomenological model for estimation of fission fragments activities with half-life periods of about one day and longer in an active zone of the reactor on thermal neutrons was offered in [3, 4]. The fission of two basic components of fuel only (^{235}U and ^{239}Pu) was taken into account in the mentioned model. Thus the fission rate of nucleus ^{235}U depending on time of the reactor working in a constant capacity mode is found to be possible to present as $P_f(^{235}\mathrm{U}, t) = P_{0f} \cdot e^{-\lambda t}$ on the basis of numerical calculations [5], where P_{0f} and λ are characteristic constants for each type of reactors given in [5]. P_{0f} is defined by the reactor capacity, $\lambda \cong 1/\tau$, where τ is a campaign of the reactor. In particular, for emergency HPOR-1000 of ChNPP $\lambda \cong 0.35$ year^{-1}.

In this case the fission rate of nucleus ^{239}Pu $P_f(^{239}\mathrm{Pu}, t) = P_{0f}(1 - e^{-\lambda t})$ and there are analytical expressions for the accumulated weights and activities of fission fragments (see for example [6]). Dependences of specific weights and specific activities from operating time of HPOR-1000 for many fission fragments with half-life periods of about one day and longer ($^{89, 90}$Sr, ^{91}Y, $^{95, 97}$Zr, ^{99}Mo, $^{103, 106}$Ru, ^{125}Sb, ^{131}I, ^{137}Cs, ^{140}Ba, $^{141, 143, 144}$Ce etc.) were calculated within the framework of the given model. Thus the agreement of analytical calculations with numerical ones [7, 8] was within the limit of 1 … 3 %, except for ^{106}Ru (a divergence ~ 10 % for ^{106}Ru). As tentative estimations have shown [9, 10], in a case of nuclides which have the relation of total yields on ^{239}Pu and ^{235}U $\rho = y(\mathrm{X}, ^{239}\mathrm{Pu})^c / y(\mathrm{X}, ^{235}\mathrm{U})^c >> 1$ for receiving more exact theoretical estimations it is necessary except for ^{235}U and ^{239}Pu to take into account also the fission of ^{238}U and ^{239}Pu by fission neutrons and thermal neutrons, correspondingly. Realization of such calculations for ^{106}Ru, ^{111}Ag and ^{125}Sb is the basic purpose of the present work.

2. MODEL

The analysis of numerical data on accumulation of fission products of ^{235}U, ^{238}U, ^{239}Pu and ^{241}Pu for HPOR-1000 (for definiteness the model "Central zone" with water density $d_{\mathrm{H_2O}} = 0.41$ g·cm^{-3} [5]) shows that for approximation of partial fission rates of fuel nuclei P_f (^{235}U, t), P_f (^{238}U, t), P_f (^{239}Pu, t) and P_f (^{239}Pu, t) besides the exponential members $e^{-\lambda t}$ there should be necessarily the members proportional to t^n, where $n \geq 1$ – positive integers. The model developed in [3, 4] was based on a fundamental opportunity to receive in an analytical form the solution of known Beitman's problem on calculation of specific activities of members in isobar chains $A_1 \rightarrow A_2 \rightarrow \ldots \rightarrow A_j \rightarrow \ldots \rightarrow A_i$ of i elements if the fission rate of a nuclide $P(t)$ is constant or changed by the exponential law [6]:

$$\begin{aligned} A_1(t)' &= \lambda_1 A_1(t) - \lambda_1 y_1^{in} P(t); \\ A_2(t)' &= \lambda_2 A_2(t) - \lambda_2 y_2^{in} P(t) + \lambda_2 A_1(t); \\ &\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots \\ A_i(t)' &= \lambda_i A_i(t) - \lambda_i y_i^{in} P(t) + \lambda_i A_{i-1}(t), \end{aligned} \qquad (1)$$

where λ_j, y_j^i and $A_j(t)$ - the decay constant, the independent yield and the activity of j^{th} member in an isobar chain X_j.

Thereof taking into account members proportional to t^n in partial decay rates the analytical solutions of the system (1) do not formally exist. However it is possible to show that in case of fission fragments X_j with relatively long half-life periods (about day and more) the approached analytical solutions of the system (1) exist. Moreover, and in the model [3, 4] the approached, considerably more simple solution (it exists at performance of conditions $\lambda_i << \lambda_j$, $j < i$) and also for fission fragments with $T_{1/2}$ about a day and more, instead of the exact solution of the system of equations (1) with $P(t) \sim e^{-\lambda t}$ (generally it has very compound form) was used. Therefore there are bases to believe, that for the mentioned type of fission fragments the approached analytical solutions of the system (1) exist for decay rates $P(t)$, that are essentially different from a constant or with the exponential dependence from t. For receiving such approached analytical solution we shall present the system of equations (1) as:

$$\begin{aligned} &A_1(t)' + \lambda_1 A_1(t) = \lambda_1 y_1^{in} P(t); \\ &A_2(t)' + \lambda_2 A_2(t) = \lambda_2 y_2^{in} P(t) + \lambda_2 A_1(t); \\ &\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots \\ &A_i(t)' + \lambda_i A_i(t) = \lambda_i y_i^{in} P(t) + \lambda_i A_{i-1}(t). \end{aligned} \qquad (2)$$

It is possible to show, that at performance of conditions $\lambda_i << \lambda_j$, $j < i$ the system of equations (2) has the approached analytical solution for one nuclide decaying with rate $P(t) = \sum a_n t^n$, where $n = 0, 1, 2, \ldots$.

Essence of the approach we shall consider on an example of the first equation:

$$A_1(t)' + \lambda_1 A_1(t) = \lambda_1 y_1^{in} P(t), \qquad (3)$$

where let's assume that $P(t) = \pi t$, π - a constant. Its solution is trivial and reduced to calculation of tabulated integral, $\int e^{\alpha x} x dx = e^{\alpha x}(x/\alpha - 1/\alpha^2)$ and for $A_1(t)$ we have:

$$A_1(t) = \lambda_1 y_1^{u} \pi e^{-\lambda_1 t} \int_0^t e^{\lambda_1 \tau} \tau d\tau. \qquad (4)$$

We take time t in years. For the first members of isobar chain $\lambda_j >> 1$ [2]. Therefore $\lambda_1 >> 1$, and $A_1(t) \cong y_1^{u} \pi t$. Substituting the given expression for $A_1(t)$ in the second equation we received:

$$A_2(t)' + \lambda_2 A_2(t) \cong \lambda_2 y_2^{in} \pi t + \lambda_2 y_1^{in} \pi t. \qquad (5)$$

As $\lambda_2 >> 1$ the approached analytical solution of given equation $A_2(t) \cong y_2^c \pi t$ is similar with $A_1(t) \cong y_1^{in} \pi t$ with the only difference, that the total yield y_1^c appears instead of an independent yield y_2^{in}. Other activities $A_j(t)$ with $j < i$ will also have the similar form. Finally for the last i^{th} member of the isobar chain with $\lambda_i << \lambda_j$ we received:

$$A_i(t) = \lambda_i y_i^c \pi e^{-\lambda_i t} \int_0^t e^{\lambda_i \tau} \tau d\tau. \qquad (6)$$

In general case at $P(t) = \rho t^n$ the solution of the first equation in system (2) is also reduced to calculation of tabulated integral:

$$\int e^{\alpha x} x^n dx = e^{\alpha x}\left[x^n/\alpha + \sum_{k=1}^{n} (-1)^k n \times \right. \\ \left. \times (n-1)\ldots(n-k+1) x^{n-k}/\alpha^{k+1}\right]. \qquad (7)$$

Repeating the same operations, as for $P(t) = \pi t$, we received for $A_i(t)$:

$$A_i(t) = \lambda_i y_i^c \pi e^{-\lambda_i t} \int_0^t e^{\lambda_i \tau} \tau^n d\tau. \qquad (8)$$

In general case $P(t)$ can be a polynomial and we shall receive by analogy

$$A_i(t) = \lambda_i y_i^c \pi e^{-\lambda_i t} \int_0^t e^{\lambda_i \tau} P(\tau) d\tau. \qquad (9)$$

As polynomial approximations exist for many functions representing physical interest it is possible to consider, that practically for any reasonable rates of the decaying nuclide $P(t)$ the approached analytical expressions of $A_i(t)$ for i^{th} member in isobar chain with $\lambda_i << \lambda_j$, $j < i$ can be received. Including for a problem considered in the present work.

In the model offered here the total decay rate for fuel nuclei is constant and consists of four partial rates:

$$P_{0f} = P(^{235}\mathrm{U},t)_f + P(^{238}\mathrm{U},t)_f + \\ + P(^{239}\mathrm{Pu},t)_f + P(^{239}\mathrm{Pu},t)_f. \qquad (10)$$

For receiving the partial decay rates $P(^{235}\mathrm{U}, t)_f$, $P(^{238}\mathrm{U}, t)_f$, $P(^{239}\mathrm{Pu}, t)_f$ and $P(^{241}\mathrm{Pu}, t)_f$ in an analytical form we use results of numerical calculations for dependences of appropriate fission products from time for HPOR-1000 [5] (the model "Central zone" with water density $d_{H_2O} = 0.41$ g·cm^{-3}, a campaign of the reactor $\tau = 2.7$ years also is broken on 18 time intervals with duration $\Delta t = 0.15$ years).

According to [5], for specified working mode of HPOR-1000 the total weight of fission products ^{235}U, ^{238}U, ^{239}Pu and ^{241}Pu $m_f(t)^c \cong 7.88\ t$ g·kg^{-1}, where t is expressed in years. Accordingly the total decay rate of nuclear fuel $P_{0f} \cong 7.88$ g·(kg·years)$^{-1}$.

The weight of decayed ^{238}U is possible to present as $m(^{238}\mathrm{U}, t)_f \cong 0.2556\ t + 0.0123\ t^2$ g·kg^{-1}, i.e. the decay rate of nuclei ^{238}U $P(^{238}\mathrm{U}, t)_f \cong 0.2556 + 0.0246\ t$ g·(kg·years)$^{-1}$. The weight of decayed ^{241}Pu accurate to several percents is described by the cubic dependence on time - $m(^{241}\mathrm{Pu}, t)_f \cong 0.0437\ t^3$ g·kg^{-1}, i.e. $P(^{241}\mathrm{Pu}, t)_f \cong 0.1311 \times t^2$ g·(kg·years)$^{-1}$. The weight of decayed ^{235}U accurate to ~ 1 % is described by the function $m(^{235}\mathrm{U}, t)_f \cong 13.27e^{0.081t}(1 - e^{-0.575t})$ g·kg^{-1}, and $P(^{235}\mathrm{U}, t)_f \cong 1.075 \times e^{0.081t} + 6.556e^{-0.494t}$ g·(kg·years)$^{-1}$.

The weight of decayed ^{239}Pu is as a difference between the total weight of the decayed fission products $m_f(t)^c$ and weights of decayed ^{235}U, ^{238}U and ^{241}Pu. The decay rate of nuclei ^{239}Pu is well approximated by the function $P(^{239}\mathrm{Pu}, t)_f \cong 5.122(1 - e^{-0.612t}) - 0.0302t^3$ g·(kg× years)$^{-1}$. Using an obvious form of above mentioned partial decay rates of nuclei ^{235}U, ^{238}U, ^{239}Pu and ^{241}Pu (4), on the basis of formulas such as (9) and received in [3, 4] the approached analytical solution for $P(t) \sim$ const and $P(t) \sim e^{-\beta t}$ we received finally for the partial activity of a i^{th} member in isobar chain X_i with $\lambda_i << \lambda_j\, j < i$:

$$A_i(\mathrm{X}_i, ^{235}\mathrm{U}, t) = 0.1364 P_{0f}\, y(\mathrm{X}_i, ^{235}\mathrm{U})^c \left[\lambda_i/(\lambda_i + 0.081)\right]\left[e^{0.081t} - e^{\lambda_i t}\right] + 0.8319 P_{0f}\, y(\mathrm{X}_i, ^{235}\mathrm{U})^c \times \left[\lambda_i/(\lambda_i + 0.494)\right]\left[e^{0.494t} - e^{\lambda_i t}\right],$$

$$A_i(\mathrm{X}_i, ^{238}\mathrm{U}, t) = 0.3244 P_{0f}\, y(\mathrm{X}_i, ^{238}\mathrm{U})^c \left[1 - e^{\lambda_i t}\right] + 0.00313 P_{0f}\, y(\mathrm{X}_i, ^{238}\mathrm{U})^c \left[e^{\lambda_i t} - 1 + \lambda_i\right]/\lambda_i, \quad (11)$$

$$A_i(\mathrm{X}_i, ^{239}\mathrm{Pu}, t) = 0.65 P_{0f}\, y(\mathrm{X}_i, ^{239}\mathrm{Pu})^c \left[1 - e^{-\lambda_i t}\right] - 0.65 P_{0f}\, y(\mathrm{X}_i, ^{239}\mathrm{Pu})^c \left[1 - e^{-\lambda_i t}\right]\left[\lambda_i/(\lambda_i - 0.612)\right] \times \left[e^{-0.612t} - e^{-\lambda_i t}\right] - 0.02298 P_{0f}\, y(\mathrm{X}_i, ^{239}\mathrm{Pu})^c \times \left[e^{-\lambda_i t} - 1 + \lambda_i - (\lambda_i t)^2/2 + (\lambda_i t)^3/6)\right]/\lambda_i^3,$$

$$A_i(\mathrm{X}_i, ^{241}\mathrm{Pu}, t) = 0.3328 P_{0f}\, y(\mathrm{X}_i, ^{241}\mathrm{Pu})^c \times \left[1 - e^{-\lambda_i t} - \lambda_i t + (\lambda_i t)^2/2\right]/\lambda_i^2.$$

Here at $P_{0f} = 6.062 \cdot 10^{14}$ (s·kg)$^{-1}$ the specific activities are expressed in 10^{-3}·Bq·kg^{-1}. It is obvious, that the total specific activity is $A(\mathrm{X}_i, t) = A_i(\mathrm{X}_i, ^{235}\mathrm{U}, t) + A_i(\mathrm{X}_i, ^{238}\mathrm{U}, t) + A_i(\mathrm{X}_i, ^{239}\mathrm{Pu}, t) + A_i(\mathrm{X}_i, ^{241}\mathrm{Pu}, t)$. The specified formalism was also used at the analysis of the total specific activity structure $A(\mathrm{X}_i, t)$ for nuclei ^{106}Ru, ^{111}Ag and ^{125}Sb.

3. CORRELATION RATIOS

For calculation of partial specific activities of ^{106}Ru, ^{111}Ag and ^{125}Sb the necessary total yields of nuclides like $y(\mathrm{X}_i, ^{235}\mathrm{U})^c$ and decay constants λ_i were took from [2]. Formulas (11) became:

$$A(^{106}\underline{\mathrm{Ru}}, ^{235}\mathrm{U}, t) \cong 7.4173 \cdot 10^{12} \times \left[e^{-0.494t} + 0.0401 e^{-0.081t} - 1.0401 e^{-0.68t}\right] (\mathrm{Bq \cdot kg^{-1}}),$$

$$A(^{106}\underline{\mathrm{Ru}}, ^{238}\mathrm{U}, t) \cong 4.2711 \cdot 10^{11} \times \left[1 - e^{-0.68t}\right] + 4.8030(+13)t\ (\mathrm{Bq \cdot kg^{-1}}),$$

$$A(^{106}\underline{\mathrm{Ru}}, ^{241}\mathrm{Pu}, t) \cong 2.7270 \cdot 10^{12} \times \left[1 - e^{-0.68t} - 0.68t + 0.2312t^2\right] (\mathrm{Bq \cdot kg^{-1}}), \quad (12)$$

$$A(^{106}\underline{\mathrm{Ru}}, ^{239}\mathrm{Pu}, t) \cong 1.6885 \cdot 10^{13} \times \left[1 - 10 e^{-0.612} + 9 e^{-0.68t}\right] - 1.8985 \cdot 10^{12} \times \left[e^{-0.68t} - 10.68t - 0.2312t^2 + 0.05241t^3\right] (\mathrm{Bq \cdot kg^{-1}}).$$

$$A(^{111}\underline{\mathrm{Ag}}, ^{235}\mathrm{U}, t) \cong 1.0215 \cdot 10^{11} \times \left[e^{-0.494t} + 0.161 e^{-0.081t} - 1.161 e^{-34t}\right] (\mathrm{Bq \cdot kg^{-1}}),$$

$$A(^{111}\underline{\mathrm{Ag}}, ^{238}\mathrm{U}, t) \cong 1.5681 \cdot 10^{10} \times \left[1 - e^{-34t}\right] + 1.5172 \cdot 10^{9} t\ (\mathrm{Bq \cdot kg^{-1}}),$$

$$A(^{111}\underline{\mathrm{Ag}}, ^{241}\mathrm{Pu}, t) \cong 9.9161 \cdot 10^{7} \times \left[1 - e^{-34t} - 34t + 578t^2\right] (\mathrm{Bq \cdot kg^{-1}}). \quad (13)$$

$$A(^{111}\underline{\mathrm{Ag}}, ^{239}\mathrm{Pu}, t) \cong 1.1927 \cdot 10^{12} \times \left[1 - 10183 e^{-0.612} + 0.0183 e^{-34t}\right] - 1.0729 \cdot 10^{6} \times \left[e^{-34t} - 1 + 34t - 578t^2 + 6551t^3\right] (\mathrm{Bq \cdot kg^{-1}}).$$

$$A(^{125}\underline{\mathrm{Sb}}, ^{235}\mathrm{U}, t) \cong 1.8347 \cdot 10^{10} \times \left[e^{-0.081t} - 8.274 e^{-0.494t} + 7.274 e^{-0.25t}\right] (\mathrm{Bq \cdot kg^{-1}}),$$

$$A(^{125}\underline{\mathrm{Sb}}, ^{238}\mathrm{U}, t) \cong 6.3447 \cdot 10^{9} \times \left[1 - e^{-0.25t}\right] + 9.9694 \cdot 10^{8} t\ (\mathrm{Bq \cdot kg^{-1}}),$$

$$A(^{125}\underline{\mathrm{Sb}}, ^{241}\mathrm{Pu}, t) \cong 1.3686 \cdot 10^{11} \times \left[1 - e^{-0.25t} - 0.25t + 0.03125t^2\right] (\mathrm{Bq \cdot kg^{-1}}). \quad (14)$$

$$A(^{125}\underline{\mathrm{Sb}}, ^{239}\mathrm{Pu}, t) \cong 4.3749 \cdot 10^{11} \times \left[1 + 0.691 e^{-0.612t} - 1.691 e^{-0.25t}\right] - 9.8989 \cdot 10^{11}\left[e^{-0.25t} - 1 + 0.25t - 0.03125t^2 + 0.0026041t^3\right] (\mathrm{Bq \cdot kg^{-1}}).$$

As an example, the calculation of total and partial activities for ^{106}Ru is shown in Fig. 1. Results of calculations of the total specific activity $A(^{106}\mathrm{Ru}, t)$ with taking into account four decaying nuclides ^{235}U, ^{238}U, ^{239}Pu and ^{241}Pu (the ■-curve) are coincided with results of numerical calculations [7, 8] (the ○-curve) much better, than calculations under formulas [3] without taking into account ^{238}U and ^{239}Pu (the □-curve).

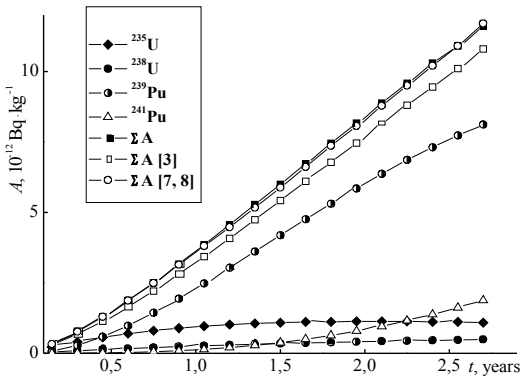

Fig. 1. *Results of calculations of partial and total activity of ^{106}Ru and comparison with results of analytical* [3] *and numerical calculations* [7, 8]

The same situation takes place and for the total specific activities $A(^{111}\mathrm{Ag}, t)$ and $A(^{125}\mathrm{Sb}, t)$. In the compact form the total specific activities of specified nuclides can be presented as [9, 10]:

$$
\begin{aligned}
A(^{106}\mathrm{Ru},t) &= 2.796\cdot 10^{13}[1-1.851\mathrm{e}^{-0.35t}+ \\
&+ 0.851\mathrm{e}^{-0.35t}]\,(\mathrm{Bq}\cdot\mathrm{kg}^{-1}), \\
A(^{111}\mathrm{Ag},t) &= 2.014\cdot 10^{12}[1-0.944\mathrm{e}^{-0.35t}- \\
&- 0.056\mathrm{e}^{-0.34t}]\,(\mathrm{Bq}\cdot\mathrm{kg}^{-1}), \\
A(^{125}\mathrm{Sb},t) &= 6.561\cdot 10^{11}[1+1.804\mathrm{e}^{-0.35t}- \\
&- 0.804\mathrm{e}^{-0.25t}]\,(\mathrm{Bq}\cdot\mathrm{kg}^{-1}).
\end{aligned}
\qquad (15)
$$

The results of present calculations can be used for estimation of the relative contribution of isotopes ^{239}Pu and ^{241}Pu in total accumulation of specific activities $A(^{106}\mathrm{Ru}, t)$, $A(^{111}\mathrm{Ag}, t)$ and $A(^{125}\mathrm{Sb}, t)$ (see Fig. 2).

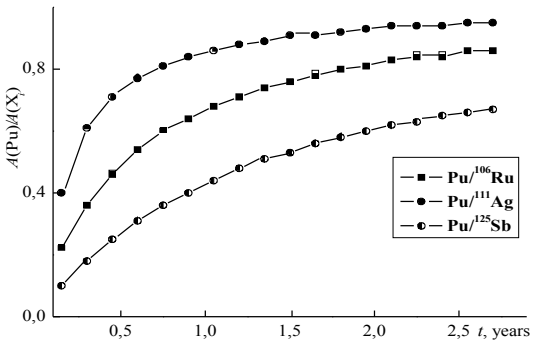

Fig. 2. *Relative contributions of ^{239}Pu and ^{241}Pu in accumulation of activities $A(^{106}Ru, t)$, $A(^{111}Ag, t)$ and $A(^{125}Sb, t)$ in HPOR-1000.*

It is obvious, that all three nuclides can be used at indirect definition of the ^{239}Pu and ^{240}Pu contents in Chernobyl emergency emissions. Necessary correlation ratios can be received from the formulas given below:

$$
\begin{aligned}
&\langle A(^{238}\mathrm{Pu},t)\rangle \cong 1.47\cdot 10^{9}\ t^{2.6}\,(\mathrm{Bq}\cdot\mathrm{kg}^{-1}), \\
&\langle m(^{238}\mathrm{Pu},t)\rangle \cong 1.47\ t^{2.6}\,(\mathrm{g}\cdot\mathrm{kg}^{-1}), \\
&\langle A(^{239}\mathrm{Pu},t)\rangle \cong 5.21\cdot 10^{9}\ t^{0.13}\,(\mathrm{Bq}\cdot\mathrm{kg}^{-1}), \\
&\langle m(^{239}\mathrm{Pu},t)\rangle \cong 2.27\ t^{0.13}\,(\mathrm{g}\cdot\mathrm{kg}^{-1}), \\
&\langle A(^{240}\mathrm{Pu},t)\rangle \cong 4.63\cdot 10^{9}\ t^{1.19}\,(\mathrm{Bq}\cdot\mathrm{kg}^{-1}), \\
&\langle m(^{240}\mathrm{Pu},t)\rangle \cong 0.55\ t^{1.19}\,(\mathrm{g}\cdot\mathrm{kg}^{-1}), \\
&\langle A(^{241}\mathrm{Pu},t)\rangle \cong 6.09\cdot 10^{11}\ t^{1.21}\,(\mathrm{Bq}\cdot\mathrm{kg}^{-1}), \\
&\langle m(^{241}\mathrm{Pu},t)\rangle \cong 0.55\ t^{1.21}\,(\mathrm{g}\cdot\mathrm{kg}^{-1}), \\
&\langle A(^{106}\mathrm{Ru},t)\rangle \cong 3.81\cdot 10^{12}\ t^{1.13}\,(\mathrm{Bq}\cdot\mathrm{kg}^{-1}), \\
&\langle A(^{111}\mathrm{Ag},t)\rangle \cong 6.50\cdot 10^{11}\ t^{0.78}\,(\mathrm{Bq}\cdot\mathrm{kg}^{-1}), \\
&\langle A(^{125}\mathrm{Sb},t)\rangle \cong 5.94\cdot 10^{10}\ t^{1.16}\,(\mathrm{Bq}\cdot\mathrm{kg}^{-1}),
\end{aligned}
\qquad (16)
$$

The given formulas are suitable for a time interval of $1.35 \le t \le 2.7$ years and take into account averaging on burning out of fuel [11].

With reference to spent reactor fuel ^{111}Ag can be very useful. With its γ-lines 0.3420 MeV (8 %) and 0.2459 MeV (1.2 %) [12]) it is possible to estimate weights of accumulated isotopes of plutonium. As to Chernobyl emergency emissions the experimental data on presence of ^{111}Ag in it practically is not present because of a small half-life period and rather low intensity of γ-lines. Anyway, in the known monography [13] the data on it is not present. Basically ^{106}Ru can be used, for example, for estimation of weights and activities dropped out in the near zone around ChNPP. On the data [13], activity of ^{106}Ru is $A(^{106}\mathrm{Ru}) \cong 5{,}92\cdot 10^{16}$ Bq. At $t \cong 1.8$ years it gives $m(^{239}\mathrm{Pu}) \cong 5.96\cdot 10^{-16} t^{1} A$ (^{106}Ru) kg and $m(^{240}\mathrm{Pu}) \cong 1.44\cdot 10^{-16} t^{0.06} A(^{106}\mathrm{Ru})$ kg, i.e. ~ 20 kg and ~ 9 kg accordingly. It approximately in 2 times exceeds the estimations made of the assumption that ~ 3.5 % of fuel [13] has dropped out in near zone around ChNPP.

In this connection it would be desirable to note, that according to [13] the ratio of activities $A(^{103}\mathrm{Ru})/A(^{106}\mathrm{Ru}) \cong 2.4$ in the near zone around ChNPP, that is not coincident with a number of theoretical estimations for emergency HPOR of ChNPP at the moment of emergency and also given in [13] (~ 4). Most probably, this contradiction speaks about higher "volatility" of ^{106}Ru in comparison with ^{103}Ru, which in turn is possible to explain by primary localization of ^{106}Ru on a surface of heat-generating element while such effect is not present for ^{103}Ru.

Considerably best results are received in a case of ^{125}Sb, which total activity in near zone around ChNPP according to [13] is $A(^{125}\mathrm{Sb}) \cong 6{,}07\cdot 10^{14}$ Bq. By analogy to estimations for ^{106}Ru we have $m(^{239}\mathrm{Pu}) \cong 3.82\cdot 10^{-14}\cdot t^{1.03} A(^{125}\mathrm{Sb})$ kg and $m(^{240}\mathrm{Pu}) \cong 9.26\cdot 10^{-16}\cdot t^{0.06} A(^{106}\mathrm{Ru})$ kg, i.e. ~ 13 kg and ~ 6 kg accordingly. It is very well coincident with estimation [13] on ~ 3.5 % transuranium elements dropped out in the near zone around ChNPP: ~ 13 kg of ^{239}Pu and ~ 5.0 kg of ^{240}Pu. And it should be expected, as according to [13] the fractionating factor for ^{125}Sb is close to unit.

4. CONCLUSION

According to received estimations, at working time of HPOR-1000 about 1.8 years the contribution of ^{239}Pu and ^{241}Pu in accumulation of ^{106}Ru is ~ 80 %, ^{111}Ag - ~ 90 % and ^{125}Sb - ~ 60 %. Thereof all three nuclides can be used at indirect definition of the transuranium element contents in the spent reactor fuel. In case of realization of the same estimations in Chernobyl emergency reactor emissions ^{125}Sb is the most suitable as its fractionating factor is close to unit and its half-life period is rather great (~ 2.7 years). In the long term the formalism developed in the present work can be used for realization of similar estimations for reactors such as WWR [5].

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ОПРЕДЕЛЕНИЕ СОДЕРЖАНИЯ ИЗОТОПОВ ПЛУТОНИЯ В РЕАКТОРНЫХ ВЫБРОСАХ ПО γ-ИЗЛУЧЕНИЮ ^{106}Ru, ^{111}Ag И ^{125}Sb

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Предлагается использовать нуклиды ^{106}Ru, ^{111}Ag и ^{125}Sb для определения содержания трансурановых элементов в отработанном топливе реактора РБМК-1000. Для оценок аварийных реакторных выбросов наиболее подходящим является ^{125}Sb. Разработанный формализм оценок может быть применен для реакторов типа ВВЭР.

ВИЗНАЧЕННЯ ЗМІСТУ ІЗОТОПІВ ПЛУТОНІЮ В РЕАКТОРНИХ ВИКИДАХ ПО γ-ВИПРОМІНЮВАННЮ ^{106}Ru, ^{111}Ag ТА ^{125}Sb

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Пропонується використовувати нукліди ^{106}Ru, ^{111}Ag і ^{125}Sb для визначення змісту трансуранових елементів у відпрацьованому паливі реактора РБМК-1000. Для оцінок аварійних реакторних викидів найбільш підходящим є ^{125}Sb. Розроблений формалізм оцінок може бути застосований для реакторів типу ВВЕР.