ON POSSIBILITY OF SEPARATION OF ^{239}Pu AND ^{240}Pu CONTRIBUTIONS IN EMERGENCY PLUTONIUM FALLOUT OF CHERNOBYL ACCIDENT ORIGIN

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The experiment-calculated method for separation of total α-activities of plutonium isotopes $A(^{239}\mathrm{Pu}+^{240}\mathrm{Pu})$ on partial activities $A(^{239}\mathrm{Pu})$ and $A(^{240}\mathrm{Pu})$ in emergency fallout of Chernobyl accident origin is developed.

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1. INTRODUCTION

It is known that ^{238}Pu, ^{239}Pu and ^{240}Pu give the main contribution to the total α-activity of plutonium isotopes in emergency fallout of Chernobyl accident origin. Thus, for technical reasons, only the total α-activity of ^{239}Pu and ^{240}Pu isotopes

$$A(^{239,240}\mathrm{Pu}) = A(^{239}\mathrm{Pu}) + A(^{240}\mathrm{Pu})$$

is usually measured, and that reduces the usefulness of laborious and costly α-spectrometric measurements.

It was previously shown [1], [2], that fission fragments ^{103}Ru and ^{106}Ru can be used for indirect definition of $A(^{239,240}\mathrm{Pu})$ and the activity ratio

$$\rho(\mathrm{Ru}) = A(^{106}\mathrm{Ru})/A(^{103}\mathrm{Ru})$$

can have independent value for the estimation of fuel burnout W. According to theoretical calculations [3], [4], the relevant activity ratio is characterized by a linear dependence on W:

$$\rho(\mathrm{Ru}, W) \approx W/49.5.$$

And since the burnout W comply with the Poisson distribution

$$P_W = (< W >)^W \cdot \frac{e^{-<W>}}{W!}$$

or the related Gaussian distribution

$$P(W)dW = \sqrt{2\pi < W >} \cdot e^{\frac{-(W-<W>)^2}{2<W>}} dW,$$

the experimental yields of the activity ratio

$$\rho(\mathrm{Ru})_{exp} = A(^{106}\mathrm{Ru})/A(^{103}\mathrm{Ru})$$

must also comply with the Poisson or Gaussian distribution.

In [3] by using the experimental data on soil contamination of Mogilev region (the distance from Chernobyl nuclear power plant (ChNPP) is about 200 km) and Bragin region (the distance from ChNPP is about 50 km) it was shown that this is really true. Since the ruthenium fell out mostly in aerosol form in Mogilev region, and both in fuel and aerosol form in Bragin region, it is clear that the activity ratio of ^{103}Ru and ^{106}Ru does not depend on the form of ruthenium fallout.

According to [1], [2], a similar situation should occur for ^{239}Pu and ^{240}Pu since the activity ratio of these isotopes is also characterized by a linear dependence on W:

$$\rho(\mathrm{Pu}, W) = A(^{240}\mathrm{Pu}, W)/A(^{239}\mathrm{Pu}, W) \approx W/8.$$

This fact can be used to separate the contributions of ^{239}Pu and ^{240}Pu isotopes in the total α-activity $A(^{239,240}\mathrm{Pu})$. And that is the main purpose of this work.

2. ANALYSIS OF EXPERIMENTAL DATA ON SOIL CONTAMINATION SOUTH OF GOMEL REGION

Since the activity ratios

$$\rho(\mathrm{Ru}, W) = A(^{106}\mathrm{Ru}, W)/A(^{103}\mathrm{Ru}, W)$$

and

$$\rho(\mathrm{Pu}, W) = A(^{240}\mathrm{Pu}, W)/A(^{239}\mathrm{Pu}, W)$$

similarly depend on W it is possible to eliminate an explicit dependence of W and obtain the activity ratio:

$$A(^{240}\mathrm{Pu})/A(^{239}\mathrm{Pu}) = 6.2 \cdot \rho(\mathrm{Ru}), \qquad (1)$$

where $\rho(\mathrm{Ru})$=$A(^{106}\mathrm{Ru})/A(^{103}\mathrm{Ru})$.

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The ratio (1) should also be confirmed by experimental data. Withal the right side of this ratio can be reliably measured. Therefore the activity ratio of $A(^{240}\mathrm{Pu})$ and $A(^{239}\mathrm{Pu})$

$$A(^{240}\mathrm{Pu}) = 6.2 \cdot \rho(\mathrm{Ru})_{exp} \cdot A(^{239}\mathrm{Pu}) \qquad (2)$$

can be experimentally established. Based on (1) and (2) the expressions for the partial activities $A(^{239}\mathrm{Pu})$ and $A(^{240}\mathrm{Pu})$ can be obtained through the total activity $A(^{239,240}\mathrm{Pu})$:

$$A(^{239}\mathrm{Pu}) = A(^{239,240}\mathrm{Pu})\frac{1}{[1 + 6.2 \cdot \rho(\mathrm{Ru})_{exp}]}, \quad (3)$$

$$A(^{240}\mathrm{Pu}) = A(^{239,240}\mathrm{Pu})\frac{6.2 \cdot \rho(\mathrm{Ru})_{exp}}{[1 + 6.2 \cdot \rho(\mathrm{Ru})_{exp}]}. \quad (4)$$

By using (3) and (4) the division of experimental data on plutonium ^{239}Pu+^{240}Pu contamination of soil south of Gomel region can be performed.

The main experimental data on density of soil contamination in Belarus by γ-emitting fission fragments were obtained in [5]-[7]. Later they were systematized and formed the basis of the national database.

The most complete experimental data on plutonium ^{239}Pu+^{240}Pu contamination levels are contained in [8], [9]. For the analysis 168 populated areas were selected for which the data both for plutonium and fission fragments were available.

As a result the experimental activity ratios $A(^{240}\mathrm{Pu})/A(^{239}\mathrm{Pu})$ were received. The diagram of experimental yields of $A(^{240}\mathrm{Pu})/A(^{239}\mathrm{Pu})$ is shown in Fig.1.

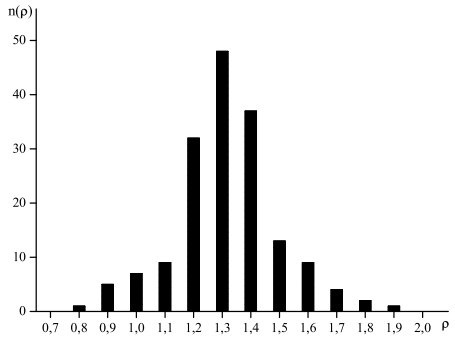

Fig.1. *Yields* n(ρ) *of experimental ratios* $A(^{240}\mathrm{Pu})/A(^{239}\mathrm{Pu})$

Qualitatively this diagram is well conformed to theoretical predictions for the ratio $A(^{240}\mathrm{Pu})/A(^{239}\mathrm{Pu})$. The average experimental value is equal to $< \rho_{exp} > \approx 1.37$. Theoretical ratios are well described by a function:

$$\rho_{theor} = 0.122 + 23.7 \cdot (e^{0.00483W} - 1).$$

The average theoretical value at $< W >= 11$ is equal to $< \rho_{theor} > \approx 1.4$, i.e. practically coincides with the experimental one.

It is obvious that the division of experimental values of α-activities $A(^{239,240}\mathrm{Pu})$ with help of the ratio $\rho_{exp} = A(^{106}\mathrm{Ru})/A(^{103}\mathrm{Ru})$ leads to reasonable results.

The partial experimental α-activities $A(^{239}\mathrm{Pu})$ and $A(^{240}\mathrm{Pu})$ received can be used to analyze the ratios like

$$\rho = A(^{239}\mathrm{Pu})/A(X)$$

and

$$\rho = A(^{240}\mathrm{Pu})/A(X),$$

where X - fission fragments associated with the fuel matrix (e.g., ^{95}Zr, ^{141}Ce, ^{144}Ce) or fission fragments with variable volatility (e.g., ^{103}Ru, ^{106}Ru).

3. EXPERIMENTAL ACTIVITY RATIOS $\rho = \mathbf{A_1/A_2}$

In relation to dependence of isotope production on fuel burnout W the activities $A(^{103}\mathrm{Ru})$ and $A(^{106}\mathrm{Ru})$ are analogues of $A(^{239}\mathrm{Pu})$ and $A(^{240}\mathrm{Pu})$, respectively. According to theoretical calculations [3], [4], [10]:

$$A(^{239}\mathrm{Pu}) \approx 2 \cdot 10^{-4} \cdot A(^{103}\mathrm{Ru})$$

and

$$A(^{240}\mathrm{Pu}) \approx 1.36 \cdot 10^{-3} \cdot A(^{106}\mathrm{Ru}).$$

Therefore, this fact can be used to establish the nature of plutonium isotopes in Chernobyl fallout.

First let us consider the behavior of ruthenium isotopes $A(^{103}\mathrm{Ru})$ and $A(^{106}\mathrm{Ru})$. It is known that they are fission fragments with variable «volatile» (in the terminology of [11]), i.e. they can fall out as nuclides associated with the fuel matrix (at relatively small distances from ChNPP), and as aerosol compounds (at large distances from ChNPP) (e.g., see [11] and references therein). This fact should affect the spectrum of experimental ratio yields, e.g.

$$\rho(^{103}\mathrm{Ru},^{95}\,\mathrm{Zr}) = A(^{103}\mathrm{Ru})/A(^{95}\mathrm{Zr})$$

and

$$\rho(^{106}\mathrm{Ru},^{95}\,\mathrm{Zr}) = A(^{106}\mathrm{Ru})/A(^{95}\mathrm{Zr}).$$

Diagrams of experimental yields of $\rho(^{103}\mathrm{Ru},^{95}\mathrm{Zr})$ and $\rho(^{106}\mathrm{Ru},\ ^{95}\mathrm{Zr})$ are shown in Fig.2 and Fig.3.

According to theoretical calculations, the average values of the ratios at $< W >= 11$ are equal to

$$< \rho(^{103}\mathrm{Ru},^{95}\,\mathrm{Zr})_{theor} > \approx 0.75$$

and

$$< \rho(^{106}\mathrm{Ru},^{95}\,\mathrm{Zr})_{theor} > \approx 0.16.$$

That is 1.6 and 1.7 times less than the experimental values

$$< \rho(^{103}\mathrm{Ru},^{95}\,\mathrm{Zr})_{exp} > \approx 1.2$$

and

$$< \rho(^{106}\mathrm{Ru},^{95}\,\mathrm{Zr})_{exp} > \approx 0.27,$$

respectively. While the peaks in distributions are approximately located at the points where the theory predicts. The average values of ratios increase because of the «trail» smoothly decreasing towards the large values.

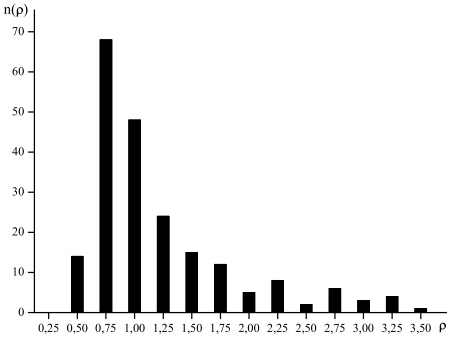

Fig.2. *Yields of the ratio* $\rho(^{103}\mathrm{Ru},^{95}\mathrm{Zr})$

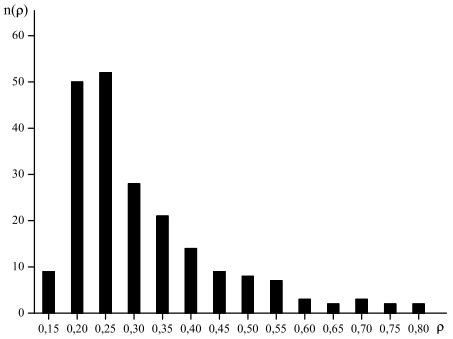

Fig.3. *Yields of the ratio* $\rho(^{106}\mathrm{Ru},^{95}\mathrm{Zr})$

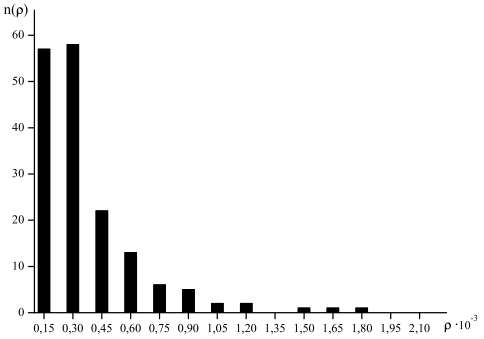

Fig.4. *Yields of the ratio* $\rho(^{239}\mathrm{Pu},^{95}\mathrm{Zr})$

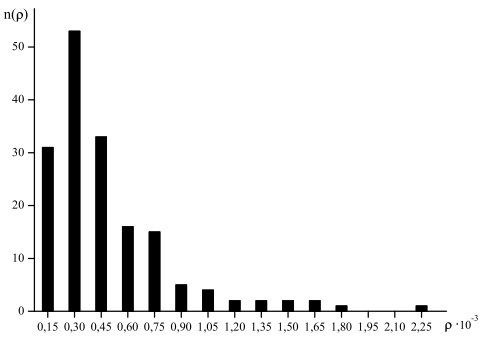

Fig.5. *Yields of the ratio* $\rho(^{240}\mathrm{Pu},^{95}\mathrm{Zr})$

Ruthenium isotopes ^{103}Ru and ^{106}Ru are nuclides with variable ≪volatility≫, and ^{95}Zr is associated with the fuel matrix. Therefore, the increase in ratios $<\rho(^{103}\mathrm{Ru},^{95}\mathrm{Zr})_{exp}>$ and $<\rho(^{106}\mathrm{Ru},^{95}\mathrm{Zr})_{exp}>$ compared with the theoretical estimation can be explained only by ≪volatile≫ of the ruthenium.

It is obvious that if the ruthenium in the form of aerosols and other species not associated with the fuel matrix can be added to the fuel ruthenium, the ratios $<\rho(^{103}\mathrm{Ru},^{95}\mathrm{Zr})_{exp}>$ and $<\rho(^{106}\mathrm{Ru},^{95}\mathrm{Zr})_{exp}>$ only increase as compared to $<\rho(^{103}\mathrm{Ru},^{95}\mathrm{Zr})_{theor}>$ and $<\rho(^{106}\mathrm{Ru},^{95}\mathrm{Zr})_{theor}>$.

By analogy we can consider the experimental ratios

$$\rho(^{239}\mathrm{Pu},^{95}\,\mathrm{Zr}) = A(^{239}\mathrm{Pu})/A(^{95}\mathrm{Zr})$$

and

$$\rho(^{240}\mathrm{Pu},^{95}\,\mathrm{Zr}) = A(^{240}\mathrm{Pu})/A(^{95}\mathrm{Zr}).$$

Diagrams of experimental yields of $\rho(^{239}\mathrm{Pu},^{95}\mathrm{Zr})$ and $\rho(^{240}\mathrm{Pu},^{95}\mathrm{Zr})$ are shown in Fig.4 and Fig.5.

According to theoretical calculations, the average values of these ratios at $<W>=11$ are equal to

$$<\rho(^{239}\mathrm{Pu},^{95}\,\mathrm{Zr})_{theor}>\approx 1.5\cdot 10^{-4}$$

and

$$<\rho(^{240}\mathrm{Pu},^{95}\,\mathrm{Zr})_{theor}>\approx 2.1\cdot 10^{-4}.$$

That is 2 times less than the corresponding experimental values

$$<\rho(^{239}\mathrm{Pu},^{95}\,\mathrm{Zr})_{exp}>\approx 3\cdot 10^{-4}$$

and

$$<\rho(^{240}\mathrm{Pu},^{95}\,\mathrm{Zr})_{exp}>\approx 4.1\cdot 10^{-4}.$$

The situation is the same as in the case of ratios $<\rho(^{103}\mathrm{Ru},^{95}\mathrm{Zr})_{exp}>$ and $<\rho(^{106}\mathrm{Ru},^{95}\mathrm{Zr})_{exp}>$ discussed earlier. The peaks in distributions are approximately located at the points where the theory predicts. The average values of ratios increase because of the ≪trail≫ smoothly decreasing towards the large values too.

Therefore, we can assume that the ruthenium and plutonium isotopes in Chernobyl fallout in the south of Gomel region behave similarly. Thus, for the consideration set of experimental data this confirms a similar conclusion of [12],[13].

It is interesting to consider the ratios $\rho(^{239}\mathrm{Pu},^{103}\mathrm{Ru})$ and $\rho(^{240}\mathrm{Pu},^{106}\mathrm{Ru})$ under the assumption that the ruthenium and plutonium isotopes could fall out both as fuel and not associated with the fuel forms. Diagrams of experimental yields of $\rho(^{239}\mathrm{Pu},^{103}\mathrm{Ru})$ and $\rho(^{240}\mathrm{Pu},^{106}\mathrm{Ru})$ are shown in Fig.6 and Fig.7.

As noted above, according to theoretical calculations

$$A(^{239}\mathrm{Pu}) \approx 2\cdot 10^{-4}\cdot A(^{103}\mathrm{Ru})$$

and

$$A(^{240}\mathrm{Pu}) \approx 1.36\cdot 10^{-3}\cdot A(^{106}\mathrm{Ru}).$$

Therefore the average experimental values

$$<\rho(^{239}\mathrm{Pu},^{103}\,\mathrm{Ru})_{exp}>\approx 2.2\cdot 10^{-4}$$

and

$$<\rho(^{240}\mathrm{Pu},^{106}\,\mathrm{Ru})_{exp}>\approx 1.4\cdot 10^{-3}$$

practically coincide with theoretical estimates. Naturally, this can only be if the ruthenium and plutonium isotopes in Chernobyl fallout behave similarly.

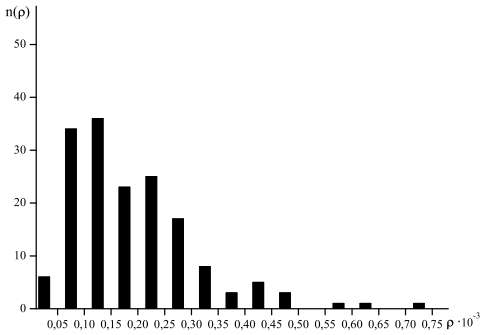

Fig.6. *Yields of the ratio* $\rho(^{239}\mathrm{Pu},^{103}\mathrm{Ru})$

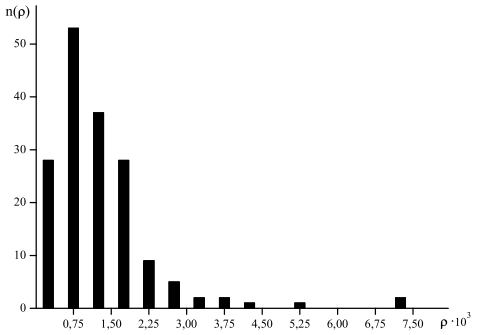

Fig.7. *Yields of the ratio* $\rho(^{240}\mathrm{Pu},^{106}\mathrm{Ru})$

However, the experimental values of $\rho(^{239}\mathrm{Pu},^{103}\mathrm{Ru})$ and $\rho(^{240}\mathrm{Pu},^{106}\mathrm{Ru})$ are both left and right of the average theoretical values

$$< \rho(^{239}\mathrm{Pu},^{103}\,\mathrm{Ru})_{theor} > \approx 2 \cdot 10^{-4}$$

and

$$< \rho(^{240}\mathrm{Pu},^{106}\,\mathrm{Ru})_{theor} > \approx 1.4 \cdot 10^{-3}.$$

It is easy to explain with the sorption of ruthenium or plutonium isotopes in aerosol form by the fuel particles with the «normal» average activity ratios $A(^{239}\mathrm{Pu})/A(^{103}\mathrm{Ru})$ and $A(^{240}\mathrm{Pu})/A(^{106}\mathrm{Ru})$.

4. CONCLUSIONS

The main purpose of this work is the development of the experiment-calculated method for separation of total α-activities of plutonium isotopes $A(^{239}\mathrm{Pu}+^{240}\mathrm{Pu})$ on partial activities $A(^{239}\mathrm{Pu})$ and $A(^{240}\mathrm{Pu})$ in emergency fallout of Chernobyl accident origin. The method is based on the fact that radionuclide activities $A(^{106}\mathrm{Ru})$, $A(239\mathrm{Pu})$ and $A(^{240}\mathrm{Pu})$ in the active zone of the thermal reactor are closely associated with each other.

The ^{106}Ru nuclide is generated mainly in the reaction of fission of ^{239}Pu by thermal neutrons. Its production is approximately linear on the burnout W and that is confirmed experimentally [14].

The ^{240}Pu nuclide is also generated primarily on ^{239}Pu but in the reaction of radioactive neutron capture. Its production is also proportional to W. Consequently, the activity ratio $A(^{240}\mathrm{Pu})/A(^{106}\mathrm{Ru})$ practically does not depend on W.

The productions of ^{239}Pu and ^{103}Ru in thermal reactors do not have a direct genetic association. However, the dependence of its activities on W in the thermal reactor is similar.

The activity ratio $A(^{239}\mathrm{Pu})/A(^{103}\mathrm{Ru})$, as in the case of $A(^{240}\mathrm{Pu})/A(^{106}\mathrm{Ru})$, practically does not depend on W. In other words, a combination of activities

$$\rho(\mathrm{Ru,Pu}) = \frac{A(^{240}\mathrm{Pu},W)\cdot A(^{103}\mathrm{Ru},W)}{A(^{239}\mathrm{Pu},W)\cdot A(^{106}\mathrm{Ru},W)}$$

both in the active zone of a thermal reactor, and in the emergency fallout does not depend on W, in particular, for the emergency high power channel reactor HPChR-1000 ρ(Ru,Pu)≈0,16. This ratio was used to analyze the experimental data on soil contamination south of Gomel region by ^{239}Pu+^{240}Pu isotopes and ^{95}Zr, ^{103}Ru, ^{106}Ru, ^{137}Cs, ^{141}Ce and ^{144}Ce fission fragments.

At the same time the data both on the local burnout W values and on the partial α-activities A(^{239}Pu) and A(^{240}Pu), which are more informative than the initial total α-activity A(^{239,240}Pu), were received. As a result, this allowed to analyze the experimental data on pairs of activity ratios like $A(^{239}\mathrm{Pu})/A(XX)$ and $A(^{240}\mathrm{Pu})/A(XX)$, $A(^{103}\mathrm{Ru})/A(XX)$ and $A(^{106}\mathrm{Ru})/A(XX)$.

The main conclusions are:

- Spectra of the experimental outputs of $A(^{239}\mathrm{Pu})/A(XX)$ and $A(^{103}\mathrm{Ru})/A(XX)$, $A(^{240}\mathrm{Pu})/A(XX)$ and $A(^{106}\mathrm{Ru})/A(XX)$ have the same structure, where X - fission fragments associated the fuel matrix ^{95}Zr, ^{141}Ce, ^{144}Ce.
- The average experimental values of this ratios is about 1.5 times more than the theoretical ones for the fuel at the average value $< W >= 11$.

It should be noted that besides the ratios

$$\rho(\mathrm{Ru,Pu}) = \frac{A(^{240}\mathrm{Pu},W)\cdot A(^{103}\mathrm{Ru},W)}{A(^{239}\mathrm{Pu},W)\cdot A(^{106}\mathrm{Ru},W)},$$

there are other four activities with similar structure, which can be useful in the analysis of soil contamination with plutonium isotopes. In particular,

$$\frac{A(^{239}\mathrm{Pu},W)\cdot A(^{240}\mathrm{Pu},W)}{A(^{103}\mathrm{Ru},W)\cdot A(^{106}\mathrm{Ru},W)} \approx 2.6 \cdot 10^{-7}$$

also does not depend on the fuel burnout.

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О ВОЗМОЖНОСТИ РАЗДЕЛЕНИЯ ВКЛАДОВ ^{239}Pu И ^{240}Pu В АВАРИЙНЫХ ВЫПАДЕНИЯХ ПЛУТОНИЯ ЧЕРНОБЫЛЬСКОГО ПРОИСХОЖДЕНИЯ

А.Н. Водин, С.Н. Олейник, Э.А. Рудак, И.В. Ушаков, О.И. Ячник

Разработан расчетно-экспериментальный метод разделения суммарных α-активностей изотопов плутония $A(^{239}\mathrm{Pu}+^{240}\mathrm{Pu})$ на парциальные α-активности $A(^{239}\mathrm{Pu})$ и $A(^{240}\mathrm{Pu})$ в аварийных выпадениях Чернобыльского происхождения.

ПРО МОЖЛИВІСТЬ РОЗДІЛЕННЯ ВНЕСКІВ ^{239}Pu ТА ^{240}Pu В АВАРІЙНИХ ВИПАДАННЯХ ПЛУТОНІЮ ЧОРНОБИЛЬСЬКОГО ПОХОДЖЕННЯ

О.М. Водін, С.М Олійник, Е.А. Рудак, І.В. Ушаков, О.І. Ячник

Розроблено розрахунково-експериментальний метод розділення сумарних α-активностей ізотопів плутонію $A(^{239}\mathrm{Pu}+^{240}\mathrm{Pu})$ на парціальні α-активності $A(^{239}\mathrm{Pu})$ та $A(^{240}\mathrm{Pu})$ в аварійних випаданнях Чорнобильського походження.