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Investigation of the Causes of Violations of the Radioactive Balance between Radionuclides of the Uranium Decay Chain

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Abstract

Throughout the literature, it is mentioned that 15 radionuclides in the uranium decay chain have a constant radioactive equilibrium. Theoretical calculations give the value of the activity of each radionuclide in the uranium decay chain.This article examines various factors that affect the coefficient of radioactive equilibrium between radionuclides in the uranium decay chainThe concept of the coefficient of violations of nuclear equilibrium between radionuclides is adopted to determine the degree of violations in the uranium decay chain.Many nuclear-physical factors influence the radioactive balance between radionuclides. The most important of them is the recoil energy that the daughter nucleus receives when splitting from the mother nucleus.Another critical factor in the violation of the radioactive balance between radionuclides is the technological factor: leaching (acid, mini-reagent, bicarbonate, etc.) when leaching uranium by underground leaching of uranium.In addition, as a theoretical result of the study, the article presents a graphical relationship between the number of nuclear masses and the recoil energy of radionuclides in the uranium decay chain.

Keywords: *Coefficient of Radioactive Equilibrium; Decay Factors; Decay Law; Equilibrium Mass; Half-Life; Recoil Energy; Uranium Decay Chains; Radioactive Equilibrium*

1. Introduction

In the process of obtaining a uranium product - uranium mother liquor, uranium chemical concentrate, uranium nitrous oxide, etc., in addition to the content of the main element and impurities, requirements are imposed on their quantities. The actual values of the main element and impurities obtained in recent years show that there is a violation of the radioactive equilibrium coefficient between the isotopes of the uranium decay chain in most uranium products. This phenomenon will increase the natural background radiation in the premises where uranium products are stored. Taking into account this fact, in this article, we investigated the factors influencing the change in the values of the radioactive equilibrium coefficient.

In recent years, there has been a substantial concern over the electricity shortage all over the world. There are many ways and different ways to solve this problem. One of the most widely used methods of generating electricity is the use of nuclear energy. For the use of nuclear energy for peaceful purposes, it predetermines the implementation of several measures, including the construction of nuclear power plants and their provision with nuclear fuel. The primary fuel for nuclear power plants around the world is uranium, a radioactive element. It is known that natural uranium contains uranium radionuclides in the amount of ^{238}U-99,28%, ^{235}U-0,72% and ^{234}U-0,0056%. The specific activity of these radionuclides in mutual radioactive equilibrium is 1.23×10^4 Bq/g, 4.9×10^4Bq/g, and 2.3×10^8 Bq/g, respectively [1-5].It can be seen that the disruption of the radioactive equilibrium between radionuclides in the uranium decay chain will lead to a change in the radiation background around these uranium products and these objects.Quantitatively, the violation of the radioactive equilibrium between radionuclides - ^{234}U/^{238}U, ^{226}Ra/^{238}U, ^{222}Rn/^{226}Ra is characterized by the radioactive equilibrium coefficient – K_{re}.This value shows how many times more or less the daughter radionuclide to the parent in the decay series.Their values are expressed in dimensionless units (Table 1).While maintaining radioactive equilibrium between the radionuclides of the uranium decay chain, the coefficient (K_{re} =1) is equal to unity.If $K_{re}<1$, then the equilibrium is shifted towards the parent radionuclide, and if $K_{re}>1$, then towards the daughter radionuclide.So, for instance, if $K_{re}<1$, then the equilibrium is shifted towards the parent of- ^{238}U, if $K_{re}>1$, then towards the daughter of - ^{226}Ra, in the case of- ^{226}Ra/^{238}U.

From the above, it can be seen that the specific activity of the ^{234}U radionuclide equals 2.3×10^8 Bq/g. Studies [6,7] prove that radionuclides – ^{238}U, ^{235}U, and ^{234}U – are not always in mutually radioactive equilibrium with the final uranium element – U_3O_8.This is the main factor affecting the quality of U_3O_8under the requirements of international standards [6-8].Thus, the study of the factors of the radioactive equilibrium between the radionuclides of the uranium decay chain — ^{234}Th, ^{234}Pa, ^{234}U, ^{230}Th, ^{226}Ra, ^{222}Rn, ^{218}Po, ^{218}Pb, ^{214}Bi, ^{214}Po, ^{210}Tl, ^{210}Pb, ^{210}Bi, ^{210}Po in samples of environmental objects and uranium products is a crucial issue in the field of nuclear physics, radiochemistry and radioecology. The primary purpose of the current work is to study the behavior of radionuclides and determine the radioactive equilibrium coefficients between the radionuclides of the uranium decay chain — ^{234}Th, ^{234}Pa, ^{234}U, ^{230}Th, ^{226}Ra, ^{222}Rn, ^{218}Po, ^{218}Pb, ^{214}Bi, ^{214}Po, ^{210}Tl, ^{210}Pb, ^{210}Bi, ^{210}Po in samples of environmental objects and uranium products.

2. Research Methods and Techniques

Before studying the nuclear-physical factors that affect the violation of the mutual radioactive balance between the radionuclides of uranium ^{238}U, ^{235}U, and ^{234}U, we will get acquainted with the regularities of the chain of radioactive decay of uranium (Figure 1).

For the theoretical calculation of the coefficient of radioactive equilibrium between radionuclides in the uranium decay chain, the following formula is used:

$$K_{re} = \frac{Ra^{226}}{U^{238}} = \frac{U^{234}}{U^{238}} = \frac{Rn^{222}}{U^{238}} = \frac{Th^{234}}{U^{238}} = 1 \qquad (1)$$

here K_{re} -is the radioactive equilibrium coefficient,A (**^{238}U**) is the activity of the parent**^{238}U**, A (^{226}Ra) is the activity of the daughter^{226}Ra and so on.

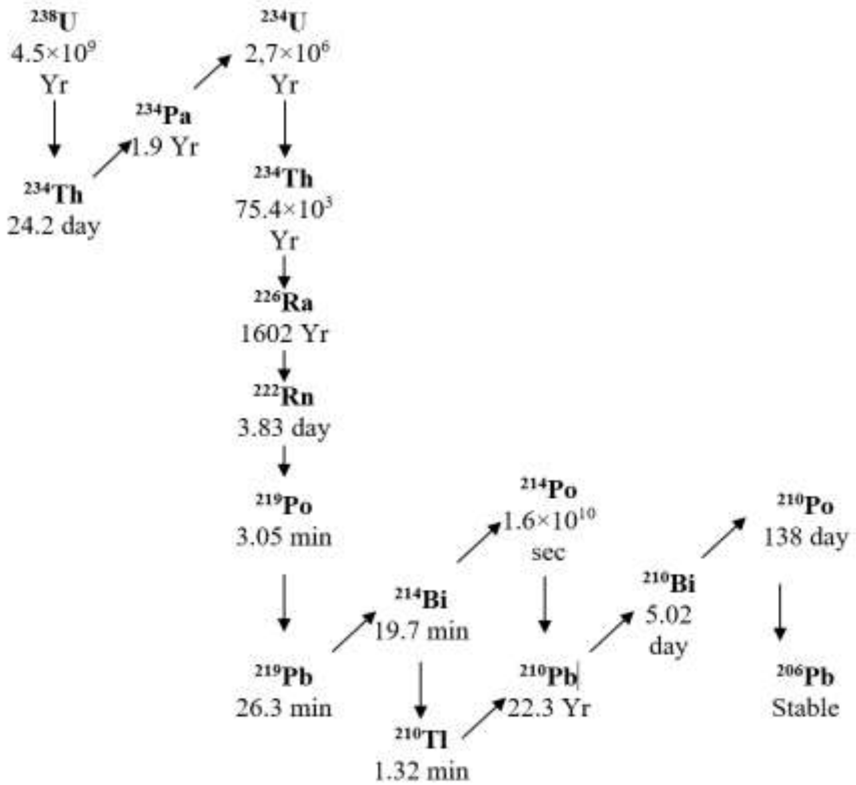

Figure 1. The chain of radioactive decay of the uranium nucleus (OpenStax: 2016).

Figure 1 illustrates the uranium decay series consisting of 14 separate steps before producing stable lead-206.

The obtained practical results show that this equation is not always fulfilled: the value of the radioactive equilibrium $K_{re} \neq 1$.We investigated the following factors to identify the actual cause of disturbances in the radioactive balance between radionuclides.

One of them is the nuclear-physical characteristics of the main radionuclides in the decay chain of uranium (Tab. 1).

Table 1. Nuclear-physical characteristics of the significant radionuclides in the uranium nuclear decay chain (Muzafarov: 2019).

Radionuclide	$T_{1/2}$(half-life decay time)	The amount of mass in equilibrium (kg)	Specificactivityofradionuclide
U-238	$4,5 \cdot 10^9$year	10^{-3}	12500
Th-234	24,1 day	$2,55 \cdot 10^{-19}$	$49 \cdot 10^{24}$
Pa-234	1,2 min	$5 \cdot 10^{-19}$	$25 \cdot 10^{18}$
U-234	$2,5 \cdot 10^5$year	$53,41 \cdot 10^{-9}$	$230,22 \cdot 10^6$
Th-230	$8,0 \cdot 10^4$year	$173 \cdot 10^{-10}$	$7.2 \cdot 10^7$
Ra-226	1602 year	$34 \cdot 10^{-11}$	$3,7 \cdot 10^{10}$

Rn-222	3,8 day	$0{,}22\cdot10^{-14}$	$5{,}7\cdot10^{15}$
Po-218	3,1 min	$12\cdot10^{-16}$	10^{19}
Pb-214	26,8 min	10^{-17}	$1{,}2\cdot10^{18}$
Bi-214	19,8 min	$76\cdot10^{-19}$	$1{,}64\cdot10^{18}$
Po-214	$1{,}6\cdot10^{-4}$ s	10^{-20}	$1{,}22\cdot10^{25}$
Po-210	22 year	$4{,}3\cdot10^{-12}$	$3\cdot10^{12}$
Bi-210	5,0 day	$27\cdot10^{-16}$	$4{,}6\cdot10^{15}$
Po-210	138,4 day	$75\cdot10^{-15}$	$1{,}7\cdot10^{14}$
Pb-206	Stable	Stable	Stable

The specific activity of any 1 gram of radionuclide given in this table is calculated using the following formula:

$$A_x = \frac{m_x \cdot ln2 \cdot N_A}{T_x \cdot \mu_x} \quad (2)$$

As shown above, the energy of the alpha particles emitted by radionuclides in the uranium chain and their reaction energies have different values. We can consider the difference in the energies of reactions produced by different radionuclides as one factor of the radioactive imbalance. The calculated values of the α-particle energy and the reaction energy for the chain elements under consideration are given in Table 2.

Table 2. Calculated values for the α-particle energy and reaction energy (National Nuclear Data; Thoennessen: 2016).

Nuclide	Historicdesignation	Historname	Typemode	Half-life	Energyreleased, MeV	Decay product
^{238}U	UI	Uranium I	α	$4{,}468\cdot10^{9}$year	4,270	^{234}Th
^{234}Th	UX$_1$	Uranium X$_1$	β^-	24,10 day	0,273	^{234}Pa^m
^{234}Pa^m	UX$_2$	Uranium X$_2$, Brevium	β^- 99,84% isomerictransition0,16%	1,16 min	2,271 0,074	^{234}U ^{234}Pa
^{234}Pa	UZ	UraniumZ	β^-	6,70 h	2,197	^{234}U
^{234}U	U$_{II}$	UraniumII	α	245500 year	4,859	^{230}Th
^{230}Th	Io	Ionium	α	75380 year	4,770	^{226}Ra
^{226}Ra	Ra	Radium	α	1602 year	4,871	^{222}Rn
^{222}Rn	Rn(RaEm)	Radon, RadiumEmanation	α	3,8235 day	5,590	^{218}Po
^{218}Po	RaA	RadiumA	α -99,98% β^- 0,02%	3,10 min	6,115 0,265	^{214}Pb ^{218}At
^{218}At	RaAt	Astatine	α 99,90 % β^-0,10 %	1,5 sec	6,874 2,883	^{214}Bi ^{218}Rn
^{218}Rn	AtEm	AstatineEmanation	α	35 msec	7,263	^{214}Po
^{214}Pb	RaB	RadiumB	β^-	26,8 min	1,024	^{214}Bi
^{214}Bi	RaC	RadiumC	β^- -99,98% α - 0,02%	19,9 min	3,272 5,617	^{214}Po ^{210}Tl

^{214}Po	RaC’	RadiumC’	α	0,1643 msec	7,883	^{210}Pb
^{210}Tl	RaC’’	RadiumC’’	β⁻	1,30 min	5,484	^{210}Pb
^{210}Pb	RaD	RadiumD	β⁻	22,3 year	0,064	^{210}Bi
^{210}Bi	RaE	RadiumE	β⁻99,99987% α - 0,00013 %	5,013 day	1,426 5,982	^{210}Po ^{206}Tl
^{210}Po	RaF	RadiumF	α	138,376 day	5,407	^{206}Pb
^{206}Tl	RaE’’	RadiumE’’	β⁻	4,199 min	1,533	^{206}Pb
^{206}Pb	RaG	RadiumG, UraniumLead	—	Stable		

As can be seen from Table 2, the amount of energy generated during alpha decay is expressed in MeV, and column 2 of the table shows the amount of reactive energy calculated from the alpha decay energy. It is assumed that the reaction energy obtained by radionuclides during radioactive decay is about the average value of 1 MeV. It can be noted that the amount of energy generated by the alpha decay of radionuclides is related to their half-life.

The recoil energy of radionuclides during alpha decay is calculated as follows:

$$(\mathrm{M}_{\alpha} + \mathrm{M}_{\mathrm{rec.en}})\mathrm{E}_{\mathrm{total}} = \mathrm{M}_{\mathrm{rec.en}}\mathrm{E}_{\mathrm{rec.en}} \qquad (3)$$

where $M_{\alpha}$and E_{α} -are the mass and energy of the alpha particle, respectively, and $\mathrm{M}_{\mathrm{rec.en}}$ and $\mathrm{E}_{\mathrm{rec,en}}$-are the mass and recoil energy of the regenerating radionuclide. In order for alpha decay to be energetically possible, the following inequality should be performed:

$$E_{total} = E_k + E_{\alpha} \qquad (4)$$

The mass (energy) of the parent nucleus must be greater than the sum of the masses (energies) of the formed nucleus and the α-particle. The excess energy of the parent nucleus during α-decay is released in the form of kinetic energies of the particles:

$$E_{\alpha} = [\mathrm{M(A,Z)} - \mathrm{M(A-4,Z-2)} - \mathrm{M}({}^{4}_{2}He)]c^2 = E_{\alpha} + \mathrm{E_K} \qquad (5)$$

here $\mathrm{E_K}$ -is the kinetic energy of the pulsed nucleus, and E_{α} -is the binding energy of the particle.

If the nucleus is in a stationary state, then $\mathrm{P}_{\alpha}=\mathrm{P}_{\mathrm{rec.nucleus,}}$ consequently the kinetic energy of the daughter nucleus is determined from the momentum conservation equation (3).

Taking into account (3), formula (5) can be denoted as follows:

$$\mathrm{E}_{\mathrm{rec.}} = \left(1 + \frac{\mathrm{M}_{\alpha}}{\mathrm{M}_{\mathrm{rec.}}}\right)\mathrm{E}_{\mathrm{total}} \qquad (6)$$

$$\mathrm{E}_{\mathrm{total}} = \mathrm{E}_{\mathrm{rec.}} = \left(\frac{\mathrm{M}_{\mathrm{rec.nud}}}{\mathrm{M}_{\alpha}+\mathrm{M}_{\mathrm{rec.}}}\right) \qquad (7)$$

here $\mathrm{M}_{\mathrm{rec.nucl.}}$-is the mass of the nucleus in a stable state.

Based on analytical formulas (3-7), it is possible to calculate the recoil energies of each radionuclide in the uranium decay chain.Based on the obtained values of the recoil energy of nuclei, it is possible to find their relationship with the number of atomic masses of nuclei in the uranium decay chain.

Figure 2illustrates the relationship between the recoil energies of nuclei with the number of atomic masses of nuclei in the uranium decay chain.

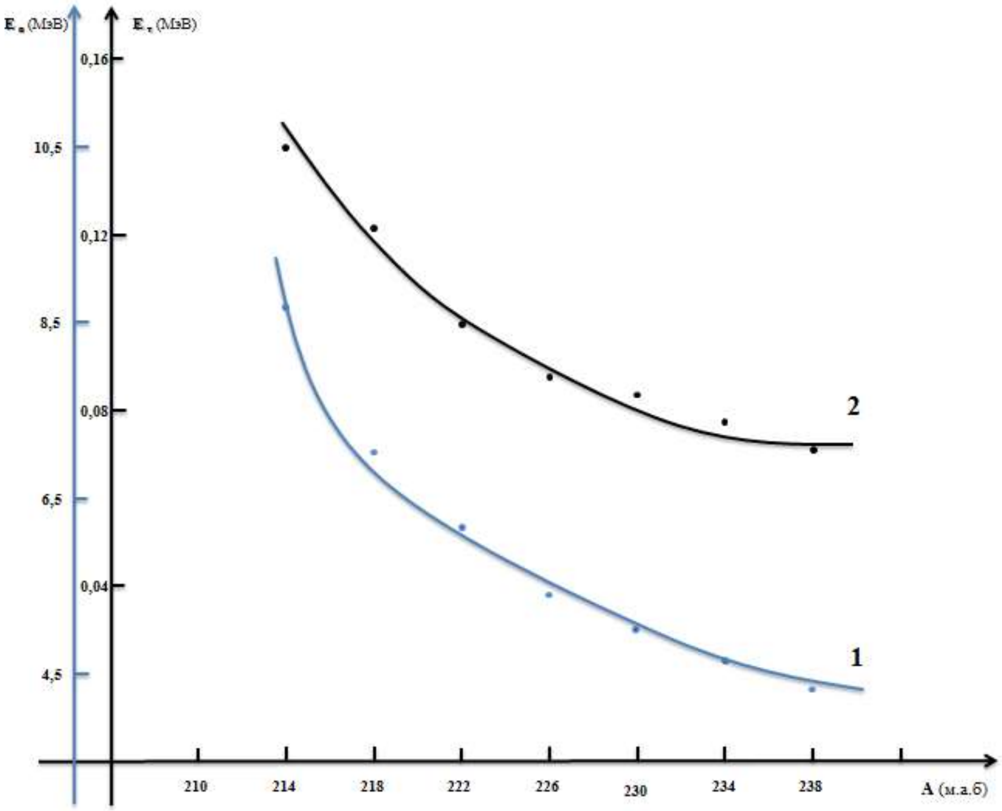

Figure 2. The relationship between the recoil energies of nuclei with the number of atomic masses of nuclei in the decay chain of uranium (Muzafarov: 2020).

As can be seen from Figure 2, with an increase in the number of atomic masses of nuclei, it is observed that the alpha-decay energy (1-curve) and recoil energy (2-curve) of daughter nuclei decrease. Consequently, the recoil energy and the energy of alpha decay of nuclei are directly proportional. This means that the electric field of the uranium nucleus, the last element found in nature, is the largest and deepest potential field in nature.

Conclusion

The main conclusion drawn from this work is that the specific activity of the radionuclide is interrelated with the half-life, that is, with an increase in the half-life, the specific activity of the radionuclide decreases. The formula for calculating the output energy of the alpha decay time of radionuclides in the uranium decay chain shows that it is related to its half-life. It was found that the reaction energy produced by the radionuclides is related to the energy during their half-life.It is found that to calculate the recoil energy of radionuclides in the uranium decay chain, and it is enough to know their half-lives. Based on the conducted research, it can be concluded that the development of a methodology for determining the age of various environmental objects and uranium deposits requires the determination of the coefficient of radioactive equilibrium between radionuclides in the uranium decay chain. We believe that reported results are of significance forsolving one of the most critical problems of dating the ages of various objects, including uranium deposits.

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