

Proliferation protection of uranium due to the presence of U-232 decay products as intense sources of hard gamma radiation^{*}

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Abstract

The objectives of the article are (1) to show the nuclear and physical causes of hard γ -quanta in the U-232 decay chain, (2) to propose tactics for handling uranium containing U-232, and (3) to assess the efficiency of its protective γ -barrier against uncontrolled proliferation.

The authors show the general picture of the decay chains of U-232 nuclide transformations, on which the protection of uranium from its uncontrolled proliferation is based. During the decay of nuclei, their emission of α - or β -particles is only the first stage of the most complex process of rearrangement of both the internal structure of the nucleus itself, which consists in the rearrangement of the neutron and proton shells and the levels of its excitation, and in the rearrangement of the electron shells of the atom. As a rule, the daughter nucleus is in a highly excited state, which is removed by the emission of hard γ -quanta and internal conversion electrons. After the second case, the remaining excitation of the atom is removed by the emission of characteristic γ -quanta and Auger-electrons with characteristic γ -quanta.

In addition, explanations are given for the quantum-mechanical reasons for the hard γ -radiation of Tl-208 and Bi-212, which complete the U-232 decay chain.

The authors also proposed a tactic for handling uranium containing uranium-232. Since the hard γ -quanta of Tl-208 and Bi-212 appear only at the end of the U-232 decay chain, after its chemical purification from its decay products, U-232 itself does not pose a radiation hazard; therefore, at this time it is advisable to conduct all necessary operations for transporting the material to the plant, fabricating uranium-based fuel containing U-232, and transporting this fuel to the nuclear facility where it will be used.

Keywords

Hard γ -radiation, chain of U-232 nuclide transformations, uranium proliferation protection

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Introduction

Currently, the permissible content of U-232 in uranium from irradiated nuclear fuel, which must be handled using gloves, is limited to an extremely small value, which, according to various sources, is from 0.2 to 1 millionth of a percent due to hard gamma radiation in its decay chain. For this reason, it is desirable to determine the nuclear and physical causes of the occurrence of hard gamma radiation, as well as to propose tactics for handling uranium containing U-232, which, on the one hand, would reliably protect uranium from its uncontrolled proliferation, and, on the other hand, would not impose significant requirements for the protection of personnel when handling it.

Traditionally, nuclear reactor scientists have only used the characteristics of decay chains as input for their research. This article shows the nuclear and physical causes of these characteristics, “throwing a bridge” between nuclear physics and nuclear reactor physics, which is useful for specialists in nuclear reactors.

U-232 decay chain scheme and characteristics

Fig. 1 shows the decay chain of radioactive U-232 into stable Pb-208. The chain contains seven nuclides undergoing α -decay and three nuclides undergoing β -decay. All the decays (except for the α -decay of polonium-212) are accompanied by the emission of γ -rays as well as electrons of various nature.

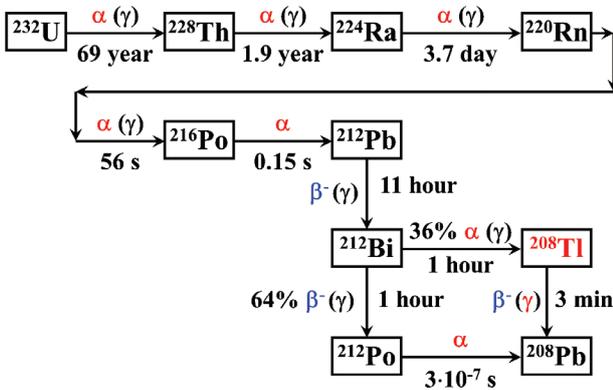


Figure 1. U-232 decay chain.

Table 1 shows the characteristics of the U-232 decay chain obtained from the National Nuclear Data Center of the Brookhaven National Laboratory, USA (National Nuclear Data Center), which specializes in nuclear physics. The table shows the most probable radiation. The radiation with the highest energy and probability of emission is shown in bold type.

Let us consider the physical processes leading to the emission of various radiation. The processes of α - or β -decay are often accompanied by additional radiation.

Table 1. U-232 decay chain characteristics

Nuclide	Decay T1/2	Decay energy, MeV	Most probable radiation, MeV (%)		
			Particles (%)	γ -radiation*)	Electrons**)
U-232	α , 68.9 year	5.414	5.263 (31.55)	0.013 (10.9)	0.009 (12.0)
			5.320 (68.15)	0.058 (0.20)	0.037 (22.79)
Th-228	α , 19.1 year	5.521	5.340 (26.0)	0.012 (8.4)	0.009 (10.2)
			5.423 (73.4)	0.084 (1.19)	0.065 (18.5)
					0.080 (5.04)
				0.083 (1.65)	
Ra-224	α , 3.66 day	5.789	5.449 (5.06)	0.012 (0.37)	0.009 (0.5)
			5.685 (94.92)	0.241 (4.1)	0.143 (0.46)
				0.223 (0.5)	
Rn-220	α , 55.6 s	6.405	6.288 (99.89)	0.550 (0.11)	
Po-216	α , 0.114 s	6.906	6.778 (99.998)	0.805 (0.002)	0.717 (2E-5)
Pb-212	β , 10.62 s	0.569	0.041 (5.01)	0.011 (13.7)	0.008 (21.3)
			0.093 (83.5)	0.075 (9.9)	0.148 (31.0)
			0.171 (13.7)	0.077 (16.4)	0.210 (1.25)
				0.239 (43.6)	0.222 (7.02)
Bi-212	α , 60.55 min	6.207	6.051 (25.13)	0.010 (7.0)	0.008 (12.1)
			6.090 (9.75)	0.040 (1.06)	0.025 (18.9)
				0.036 (4.4)	
Bi-212	β , 60.55 min	2.252	0.834 (55.37)	0.727 (6.67)	0.634 (0.13)
				0.785 (1.10)	
				1.079 (0.56)	
				1.513 (0.29)	
			1.621 (1.47)		
Po-212	α , 0.299 μ s	8.954	8.785 (100)		
TL-208	β , 3.053 min	4.999	0.343 (3.18)	0.511 (22.6)	0.423 (1.90)
			0.442 (24.2)	0.583 (85.0)	0.495 (1.70)
			0.535 (22.2)	0.861 (12.5)	0.567 (0.46)
			0.649 (49.1)	2.615 (99.754)	2.527 (0.21)

*) Characteristic and hard γ -radiation;

**) Internal conversion electrons and Auger electrons.

In particular, after decay, the formed nucleus often is in an excited state, which can be removed by high-energy γ -quanta emitted by this nucleus.

However, this is not the only process of removing excitation of the nucleus. The nucleus can transfer the excitation energy to one of the atomic electrons through quantum mechanical processes. As a result, the nucleus passes into the ground (unexcited) state, and an excited electron is released from the atomic shell; this electron is called the “internal conversion electron”. Thus, the removal of excitation of the nucleus through the process of internal conversion occurs without the emission of γ -quanta. Since the excitation energies of the nucleus and the binding energies of electrons in the atomic shells are discrete quantities, the kinetic energy of internal conversion electrons is also a discrete quantity, which is determined by the formula:

$$E(\text{internal conversion electron}) = E(\text{nuclear excitation}) - E(\text{electron bond}).$$

As a result of the emission of an internal conversion electron, the atomic nucleus passes into the ground (unexcited) state, but the atom remains excited, since one of its electron shells lacks the electron emitted during internal conversion. A vacancy is filled with an electron from a more distant (outer) shell almost instantly (in a time of about $1 \cdot 10^{-15}$ s). In this case, the so-called “characteristic γ -quantum” is emitted. Its energy is discrete and equal to

the difference between the binding energies of the vacant and outer electron shells:

$$E(\text{characteristic } \gamma\text{-quantum}) = E(\text{vacancy}) - E(\text{outer shell}).$$

This γ -radiation is called “characteristic”, since the spectrum of emitted γ -quanta is discrete and corresponds to the structure of the atomic shells, i.e., is completely defined by this structure; therefore, it can serve to determine the element by which it was emitted.

After an internal conversion electron is emitted, the atom can remove its excitation even without the emission of a γ -quantum. When the electron vacancy is filled, the released energy can be transferred to an electron of another higher level, which will bring it to an excited state. The process of energy transfer has not yet been determined. If the transferred energy is greater than its binding energy, then the excited electron will leave the atom, i.e., the atom will emit an Auger electron. Since all the energy levels of atomic electrons are discrete, then the kinetic energy of the Auger electron is also discrete and is calculated by the formula:

$$E(\text{Auger electron}) = E(\text{vacancy}) - E(\text{outer shell}) - E(\text{bonds}),$$

where $E(\text{bonds})$ is the binding energy of an electron on the shell from which the Auger electron flew out. As a result, instead of one primary electronic vacancy, two new vacancies appear in the atom, but, at higher energy levels, i.e., the atom is doubly ionized, which means that it is still in an excited state. For the final removal of excitation in the atom, electronic transitions occur with the emission of characteristic γ -quanta and the addition of the missing electrons to the atom. Table 1 lists the energies and outputs of all the types of radiation considered.

Thus, using the example of the U-232 decay chain, we can see that, during the decay of nuclei, their emission of α - or β -particles is only the first stage of the most complex process of rearrangement of both the internal structure of the nucleus itself, which consists in the rearrangement of the neutron and proton shells and the levels of its excitation, and in the rearrangement of the electron shells of the atom. As a rule, the daughter nucleus is in a highly excited state, which is removed by the emission of hard γ -quanta and internal conversion electrons. After the second case, the remaining excitation of the atom is removed by the emission of characteristic γ -quanta and Auger-electrons with characteristic γ -quanta. The scheme of emission of various radiations is shown in Fig. 2.

We can see that the energy of the accompanying γ -radiation varies from several tens of keV to several MeV. Note that the hardest γ -quanta with an energy of 2.615 MeV are emitted by thallium-208, and the yield of this radiation is high, amounting to almost 100%. The latter means that almost every Tl-208 β -decay is accompanied by the emission of a high-energy γ -quantum of 2.615 MeV. In addition, the decay of thallium-208 is accompanied by the

emission of albeit less hard but still relatively high-energy γ -radiation with an energy of 0.583 MeV with a significant yield of 85%.

Since Tl-208, in turn, is formed in 36% of the cases of α -decay of Bi-212 (Fig. 1), which, in turn, is a daughter product of the U-232 decay, it can be argued that 36% of the U-232 decay leads to the emission of approximately a pair of hard γ -quanta with energies of 2.615 and 0.583 MeV (the probability of the latter is slightly less than 100%, amounting to 85%). We can also note the hard γ -quanta accompanying the Bi-212 β -decay (1.513 and 1.621 MeV), but their yields are much lower, 0.29 and 1.47%, respectively.

Quantum-mechanical causes of hard gamma-radiation of Thallium-208

Let us now find out the reasons for such an anomalously hard (2.615 MeV) γ -radiation with a high (almost 100%) probability. Fig. 3 shows a simplified scheme of the Tl-208 parent nucleus β -decay.

The Tl-208 nucleus decay occurs from the ground, i.e., unexcited state (indicated as “0.0” in the figure) characterized by spin 5 and an even wave function (indicated as “5+”) into the ground state of the Pb-208 stable daughter nucleus, which is described by an even wave function (indicated as “0.0 0+”). For brevity, the wave function is often called the psi-function or ψ -function. The Tl-208 nucleus is odd-odd; it has an odd number of protons (81) and neutrons (127). Such nuclei are fragile, i.e., characterized by a reduced binding energy per nucleon (proton and neutron) that make up the nucleus. At the same time, the Pb-208 nucleus has not only an even number of protons (82) and neutrons (126), but all nucleon shells are closed. This is the so-called “double-magic” core. There are only five such nuclei in nature, namely: ${}^2_2\text{He-4}$, ${}^8_8\text{O-16}$, ${}^{20}_{20}\text{Ca-40}$, ${}^{28}_{20}\text{Ca-48}$, ${}^{126}_{82}\text{Pb-208}$. They are extremely durable, i.e., they have an increased binding energy of each nucleon. That is why the β -decay energy, i.e., the difference between the ground states of Tl-208 and Pb-208 is a huge value of ~ 5 MeV.

In addition, the double-magic nuclei are characterized by an extremely high first excitation level and a large energy distance between subsequent excitation levels. Thus, in Pb-208, the first excitation level is significant, about 2.6 MeV, and the next two levels are about 3.2 and 3.5 MeV. Then they begin to thicken, i.e., are located relatively close to each other. The theory of β -decay was developed on the basis of quantum mechanics. According to this theory, β -decay is a weak interaction: it is weaker than the nuclear and electromagnetic interactions by about 13 and 11 orders of magnitude, respectively, but 25 orders of magnitude stronger than the gravitational one. The range of the weak interaction is about 3 orders of magnitude smaller than even the short-range nuclear interaction, amounting to about $1 \cdot 10^{-18}$ m. The probability

of the decay of the parent nucleus into a specific state of the daughter nucleus, which is often called “transition”, is calculated on the basis of the wave functions of the nuclei in these states, taking into account their spins and the parity of the ψ -functions. Moreover, the greater the difference in spins between the parent and daughter nuclei, the lower the transition probability. Changing the parity of the wave function during the transition also reduces the probability of the transition. As a result, Table 2 can be compiled, describing the probabilities of various transitions. In the table, the magnitude of spin changes during the transition is calculated as the modulus of their difference in the initial and final states, i.e., the “+” sign means that the parity of the ψ -function has not changed, and the “-” sign means that the parity of the ψ -function has changed. Unfortunately, the theory does not give an exact value of the probability of a specific transition, but only an approximate estimate of it, i.e., a trend in the change in probabilities when different transitions are compared, which in practice can be violated.

Table 2. Transition probabilities in β -decay

The value of the change in the spin and parity of the ψ -function during the transition	Transition denotation	Transition probability
0+	Superallowed	Utmost
1+	Allowed	Very large
0- and 1-	Non-unique, of first exclusion	Large
2-	Unique, of first exclusion	Little
2+	Non-unique, of second exclusion	Mean
3-	Unique, of second exclusion	Low
3+	Non-unique, of third exclusion	Very low

Fig. 2 shows that the transition from the ground state of Tl-208 to the ground state of Pb-208 corresponds to a very large spin change ($|5 - 0| = 5$), although without a change in the parity of the ψ -function; therefore, such a transition is so unlikely that practically never implemented. The transition of Tl-208 to the first excitation level of Pb-208 also corresponds to a significant change in the spin ($|5 - 3| = 2$) and a change in the parity of the ψ -function from even (+) to odd (-). According to Tab. 2, this is the case “2-”, i.e., the unique transition of first exclusion, the probability of which is little; therefore, it is not implemented either. Finally, the transition of Tl-208 to the second excitation level of Pb-208 occurs without a change in the spin ($|5 - 5| = 0$) but with a change in the parity of the ψ -function from even (+) to odd (-), i.e., according to Tab. 2 is denoted as “0-” and is called the non-unique transition of first exclusion, the probability of which is large; therefore, it occurs in approximately half of the cases of β -decay (49.1%). After that, two γ -quanta are emitted with high energies ~ 0.6 and ~ 2.6 MeV. In general, the β -decay energy (5 MeV) is released as a result of the emission of a single β -particle with an energy of about 1 to 1.8 MeV and the subsequent emission of a series of γ -quanta with energies from 0.3 to 2.6 MeV as well as internal conversion electrons and Auger electrons.

Thus, superhard penetrating γ -radiation is the result of a huge energy of β -decay (5 MeV), which, according to

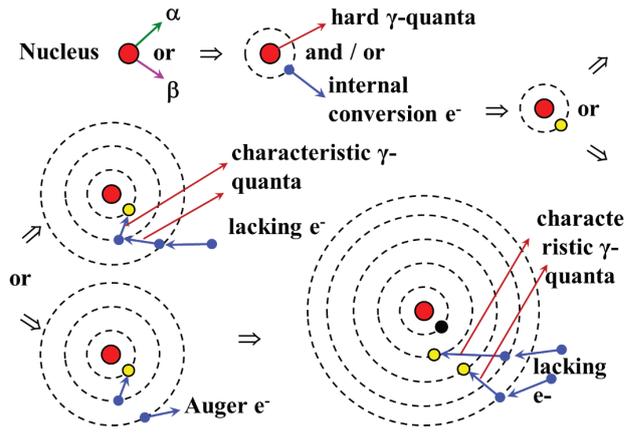


Figure 2. Nuclear decay scheme.

the laws of quantum mechanics, cannot be carried away by a β -particle, and the first two excitation levels of the double-magic nucleus of Pb-208 are located very high. These high-energy excitations are removed by the emission of high-energy γ -quanta (~ 0.6 and ~ 2.6 MeV) with a large probability (~ 85 and $\sim 100\%$, respectively). All of the above considerations were conveniently carried out using the simplified Tl-208 β -decay scheme (Fig. 3). To obtain a more complete picture of the entire complexity of this process, one can refer to the complete Tl-208 β -decay scheme (National Nuclear Data Center).

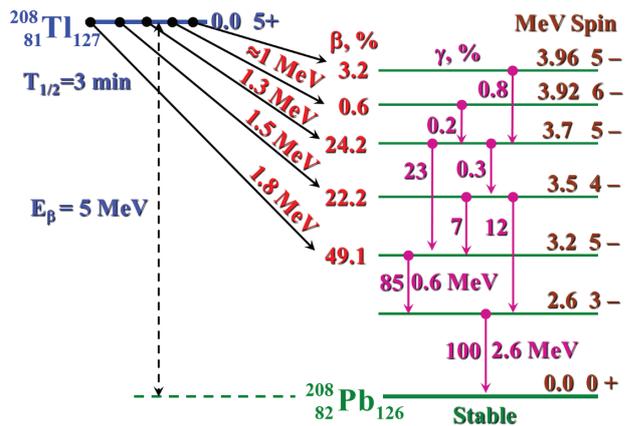


Figure 3. Simplified Tl-208 β -decay scheme.

Bismuth-212 decay

As a result of the Bi-212 β -decay (National Nuclear Data Center), a pair of hard γ -rays with energies of 1.621 and 1.513 MeV is emitted. However, the probability of their emission is significantly less than that of Tl-208, amounting to only 1.47 and 0.29%, respectively.

This is explained by the fact that the spins of the initial Bi-212 nucleus and the final Po-212 nucleus coincide, amounting to 0, and the β -decay energy (2.252 MeV) is not so high; therefore, it can be carried away by a β -particle. As a result, most of the β -decays proceed to the ground state of Po-212 without γ -radiation at all. However, in Po-212, the first three excitation levels are characterized by spins that differ slightly from the spin of Bi-212

(0), amounting to 1+ (third level) and 2+ (first and second levels) and a rather high excitation energy. Therefore, the Bi-212 β -decay is still accompanied by the emission of γ -quanta with an energy of 1.621 MeV and a probability of 1.47%, which corresponds to a change in the nuclear spin by 1 and the emission of γ -quanta with an energy of 1.513 MeV and a lower probability of 0.29%, because this corresponds to a larger change in the nuclear spin by 2 (Varlamov et al. 2010).

Tactics for handling uranium containing U-232

Keeping in mind the sequence and half-lives of the nuclides included in the U-232 decay chain, we can see that hard γ -quanta of Tl-208 and Bi-212 appear with a significant yield only at the end of the decay chain. This means that, after chemical purification of U-232 from the products of its decay chain, U-232 itself does not pose a danger in terms of penetrating hard γ -radiation (De Volpi 1982, Kang and Von Hippel 2001).

It is at this time that it is advisable to carry out all the necessary operations to transport the material to the plant, manufacture fuel based on uranium containing U-232, and also transport fuel to a nuclear facility where it will be used. The sooner this is done, the safer the handling of uranium will be. Based on the data in the next section, the period of safe handling of uranium-232 can be about one year.

Creating a protective gamma-barrier for uranium containing U-232

To create a radiation barrier in the form of hard γ -radiation, it is desirable to store fresh uranium fuel containing U-232 for several years, during which the U-232 decay chain will come into equilibrium with the γ -emitting Tl-208 and Bi-212 accumulated in it. This is confirmed by the dependence of the dose rate of dose U-232 and its daughter nuclides with the U-232 storage time, which is shown in Fig. 4.

It can be seen that this value is reached its maximum after ~ 10 years of U-232 storage and then slowly decreases over several decades. Thus, this proliferation protection is long-term, which is easily explained given that the half-life of U-232 is almost 70 years.

The uranium loading of VVER-1000 is approximately 66 tons in 163 fuel assemblies, i.e., about 405 kg of uranium in one assembly. The promising fuel assumes 5% enrichment in uranium-235. For ease of evaluation, we will accept the equivalence of the breeding properties of U-235 and U-233, although the breeding properties of U-233 in the thermal spectrum are better than those of uranium-235. Then the content of U-233 per assembly will

be about 20 kg. It was previously estimated that the content of U-232 could reach $\sim 1\%$ in U-233 produced in the thorium blanket of a thermonuclear facility by irradiating thorium for ~ 500 days (Orlov et al. 1979, Kulikov et al. 2016, 2020a, 2020b, Andrianov et al. 2019). In this case, the assembly will contain about 0.2 kg of U-232 (Shieff et al. 1977, Krumbein et al. 1980).

Fig. 4 shows the dose rate from 1 kg of U-232. It can be seen from the figure that in order to estimate the dose from the assembly, it is necessary to reduce by a factor of 5 the values shown in Fig. 4. If attackers want to manually extract uranium from the assembly, which contains 5% U-233 with 1% U-232 in it, stored for 10 years, then the dose rate from the assembly will be 2400 rem/h (rem is the biological equivalent of X-ray). Note that the lethal dose is 450 rem. Consequently, the attackers will receive a lethal dose in about 11 minutes. This is an essential radiation protection against unauthorized proliferation.

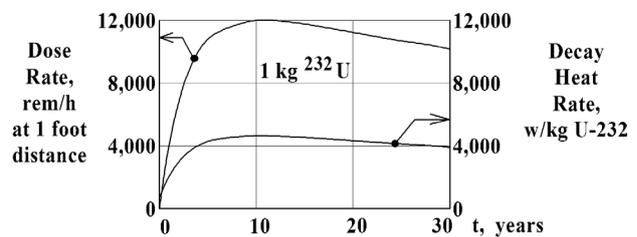


Figure 4. Dose rate and heat release of U-232 depending on its storage time.

Conclusion

The authors show the general picture of the decays of the chain of nuclide transformations of uranium-232, on which the protection of uranium from its uncontrolled use is based. During the decay of nuclei, their emission of α - or β -particles is only the first stage of the most complex process of rearrangement of both the internal structure of the nucleus itself, which consists in the rearrangement of the neutron and proton shells and the levels of its excitation, and in the rearrangement of the electron shells of the atom. As a rule, the daughter nucleus is in a highly excited state, which is removed by the emission of hard γ -quanta and internal conversion electrons. After the second case, the remaining excitation of the atom is removed by the emission of characteristic γ -quanta and Auger-electrons with characteristic γ -quanta.

In addition, explanations are given for the quantum-mechanical reasons for the hard γ -radiation of thallium-208 and bismuth-212, which complete the decay chain of uranium-232. It is estimated that 36% of U-232 decays lead to the emission of a pair of hard γ -quanta accompanying the Tl-208 β -decay with energies of 2.615 and 0.583 MeV and probabilities of 100 and 85%, respectively, whereas in the remaining 64% of U-232 decays, one more pair of hard γ -quanta accompanying the Bi-212 β -decay with energies of 1.621 and 1.513 MeV and probabilities of 1.47 and 0.29%, respectively.

The authors also proposed a tactic for handling uranium containing uranium-232. Since the hard γ -quanta of thallium-208 and bismuth-212 appear only at the end of the decay chain of uranium-232, after its chemical purification from its decay products, uranium-232 itself does not pose a radiation hazard; therefore, at this time it is advisable to conduct all necessary operations for transporting the material to the plant, fabricating uranium-based fuel containing uranium-232, and transporting this fuel to the nuclear facility where it will be used. The sooner this is done, the safer the handling of uranium will be. According to the estimates obtained, the period of safe handling

of U-232 can be about one year after its purification from decay products.

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