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DECAY HEAT POWER AND RADIOTOXICITY OF SPENT URANIUM, PLUTONIUM AND THORIUM FUEL AT LONG-TERM STORAGE

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ABSTRACT

Changes of decay heat power and radiotoxicity of spent nuclear fuel of VVER-1000 type reactors are calculated at storage during time up to 300 000 years. Decay heat power of long-lived radioactive wastes influences upon the heat removal system of the storage facility. Radiotoxicity is important from the viewpoint of ecological hazard of spent fuel. If the most dangerous nuclides are separated and extracted from storage with subsequent transmutation, radiological hazard of wastes staying in storage reduces.

For uranium spent fuel decay heat power and radiotoxicity are determined mainly by actinides. Alpha-radiation introduces the main contribution in power of actinides. The total radiotoxicity of actinides of U-Pu fuel is about 2.5 times higher, than that of U fuel. The radiotoxicity of actinides of Th-U fuel is 3.5 times less than that of U fuel.

Partitioning of actinides of U and U-Pu spent fuel should be made before long-term storage. The most important is transmutation of Am. In case of Th-U fuel, the overwhelming part of radiotoxicity is determined by isotope ^{232}U of the same chemical element as main fuel isotope ^{233}U. Therefore repeated use of Th-U fuel will be accompanied by accumulation of radiotoxicity.

Keywords: spent fuel, long-term storage, radiotoxicity, decay heat power.

1. INTRODUCTION

The problem of management of long-lived radioactive waste from spent nuclear fuel is closely connected to prospects of development of atomic power engineering. One of opportunities is construction of long-term controllable storage facility. Other ways are connected with realization of idea of nuclear transmutation of long-lived waste. For correct choice of strategy of long-term storage of spent nuclear fuel, it is necessary to know how the major characteristics of radioactive waste vary during long-term storage.

Development of atomic power industry inevitably leads to use of plutonium as a nuclear fuel alongside with the uranium. Accumulation of dangerous actinides in spent uranium-plutonium fuel is higher than in the usual uranium fuel. From the view point of reducing of radiation dangers of nuclear waste, thorium fuel cycle is prospective due to much lower formation of plutonium, americium, and curium in comparison with uranium or uranium-plutonium fuels. Separation of the most hazardous nuclides and extraction them from the storage facility with following transmutation allows to reduce radiobiological danger of the waste remaining in the storage. Removing of nuclides with increased decay heat power from the storage allows to decrease requirements to heat

removing systems of the storage facility. Quantitative comparison of radiobiological features of actinides forming in different fuel cycles is also of interest.

In the paper, results of researches of time dependence of decay heat power and radiotoxicity of 1 ton of spent fuel discharged from VVER-1000 reactor are presented. At calculations of power, contributions of alpha-, beta-, and gamma-radiation were taken into account. At calculations of radiotoxicity, maximum permissible activity of nuclides in water was used. We considered the total time of storage of spent fuel equal to 300 000 years, corresponding to the time of ultimate geological disposal.

The radiotoxicity of radioactive waste seems to be more descriptive characteristic of radiation danger than activity. The ingestion radiotoxicity RT_i of each nuclide i is determined as ratio

$$RT_i = A_i / G_i,$$

where A_i is activity of considered amount of nuclide i, G_i is limiting radioactive concentration guide for nuclide i in one kilogram of drinking water, assigned by the special normative document - Russian Radiation Safety Standards (1999) or corresponding documents of IAEA (International Basic Safety Standards, 1996). Total radiotoxicity is equal to the sum of radiotoxicities over all nuclides in considered mixture of nuclides. For such way of calculation, ingestion radiotoxicity is measured by an amount of water required for diluting of radionuclides to safe concentrations.

2. URANIUM SPENT FUEL

In Fig.1, values of calculated decay heat power of actinides (plutonium, americium, and curium) and fission products extracted from 1 ton of spent fuel unloaded from VVER-1000 reactor are submitted. T is storage time. The isotopic composition of spent fuel corresponds to a burnup of 40 MW·d/kg and subsequent cooling during 3 years. Nuclides giving the appreciable contribution to power are considered. The data on amount of nuclides in spent fuel are taken from the handbook (Kolobashkin, 1983).

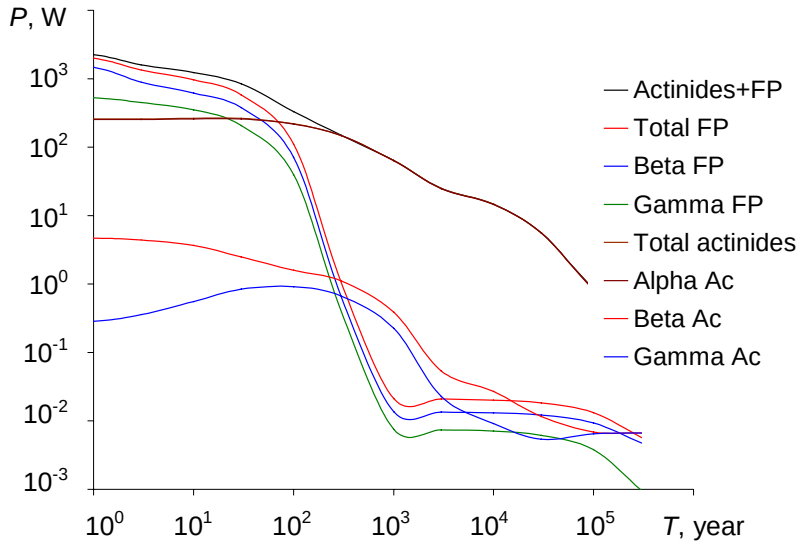

Fig.1. Decay heat power of actinides and fission products at long-term storage of 1 ton of spent fuel of VVER-1000 reactor

Values of decay heat power of most important actinides from 1 ton of spent fuel are submitted in Fig.2. The same data for fission products are given in Fig.3.

The data presented demonstrate the following. At storage less than 50 years, fission products give the main contribution to decay heat power, while at large times of storage the contribution of fission products falls, and decay heat power is determined by actinides. At time of storage 100 years, the contribution of fission products to total power is 33 %, at time 300 years - 0.7 %. Alpha-radiation introduces the main contribution in power of actinides, the part of beta- and gamma-radiation does not exceed several percents.

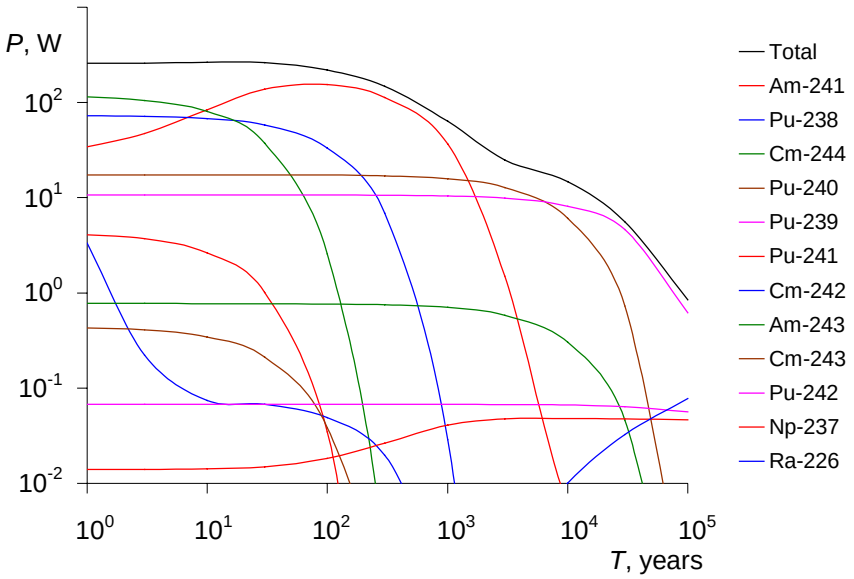

Fig.2. Decay heat power of actinides from 1 ton of spent fuel, W

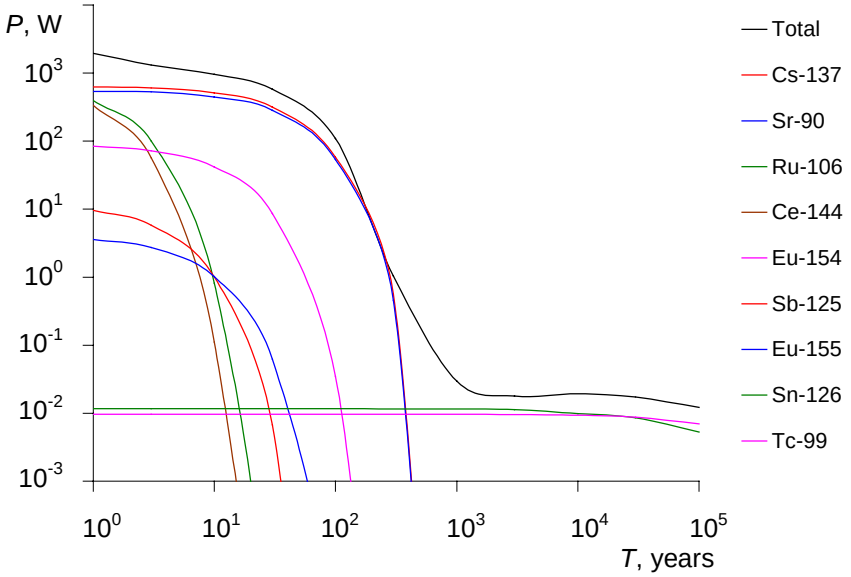

Fig.3. Decay heat power of fission products from 1 ton of spent fuel, W

Decay heat power of actinides in different periods of storage is determined by the following nuclides. At time of storage to 10 years the main nuclides are ^{244}Cm (60%) and ^{238}Pu (30%), at time 100 years - ^{241}Am, at 3 000 years - ^{240}Pu, at 30 000-100 000 years - ^{239}Pu. Power of fission products during first 300 years is determined about in equal degree by nuclides ^{90}Sr, ^{137}Cs, and in more than 1000 years - by ^{99}Tc and ^{126}Sn.

Values of ingestion radiotoxiciy of actinides and fission products extracted from 1 ton of spent fuel unloaded from VVER-1000 reactor are submitted in table 1. Values of ingestion radiotoxicity of most important actinides from 1 ton of spent fuel are submitted in table 2. The same data for fission products are given in table 3.

Table 1. Ingestion radiotoxicity of actinides and fission products at long-term storage of 1 ton of spent fuel of VVER-1000 reactor

T, year	Ingestion radiotoxicity, 10^{14} kg water		
	Actinides	FP	Total
1	5.16	11.6	16.8
10	4.99	8.25	13.2
100	3.74	0.988	4.73
300	2.54	0.00903	2.55
1000	1.16	$5.22 \cdot 10^{-5}$	1.16
10000	0.309	$5.10 \cdot 10^{-5}$	0.309
100000	0.0155	$4.17 \cdot 10^{-5}$	0.0155

Table 2. Ingestion radiotoxicity of actinides from 1 ton of spent fuel, 10^{14} kg water

Nuclide	*T*, year					
	1	10	100	1000	10 000	100 000
^{238}Pu	1.4	1.3	0.6231	$5.4 \cdot 10^{-4}$	-	-
^{239}Pu	0.23	0.23	0.23	0.22	0.17	0.013
^{240}Pu	0.37	0.37	0.37	0.33	0.13	$9.2 \cdot 10^{-6}$
^{241}Pu	1.6	1.1	0.014	$4.6 \cdot 10^{-6}$	$2.2 \cdot 10^{-6}$	-
^{241}Am	0.55	1.3	2.5	0.59	$9.7 \cdot 10^{-5}$	-
^{244}Cm	1.0	0.71	0.023	-	-	-
Total	5.2	5.0	3.7	1.2	0.31	0.016

Table 3. Ingestion radiotoxicity of fission products from 1 ton of spent fuel, 10^{14} kg water

Nuclide	*T*, year					
	1	10	100	1000	10 000	100 000
^{90}Sr	6.2	5.0	0.59	-	-	-
^{93}Zr	$6.5 \cdot 10^{-6}$	$6.5 \cdot 10^{-6}$	$6.5 \cdot 10^{-6}$	$6.5 \cdot 10^{-6}$	$6.4 \cdot 10^{-6}$	$6.2 \cdot 10^{-6}$
^{99}Tc	$2.7 \cdot 10^{-5}$	$2.7 \cdot 10^{-5}$	$2.7 \cdot 10^{-5}$	$2.7 \cdot 10^{-5}$	$2.6 \cdot 10^{-5}$	$1.9 \cdot 10^{-5}$
^{106}Ru	0.76	1.6-3	-	-	-	-
^{129}I	$1.1 \cdot 10^{-5}$	$1.1 \cdot 10^{-5}$	$1.1 \cdot 10^{-5}$	$1.1 \cdot 10^{-5}$	$1.1 \cdot 10^{-5}$	$1.1 \cdot 10^{-5}$
^{137}Cs	3.9	3.2	0.40	-	-	-
Total	11.6	8.2	1.0	$5.9 \cdot 10^{-5}$	$5.7 \cdot 10^{-5}$	$4.5 \cdot 10^{-5}$

The ingestion radiotoxicity of fission products at storage less than 20 years appears some higher than radiotoxicity of actinides. At further storage, the relative contribution of fission products falls while radiotoxicity of actinides reduces slowly. At time of storage 100 years, fission products give the contribution in total radiotoxicity 20 %, and at time 300 years - about 0.35 %.

The radiotoxicity of actinides in different periods of storage is determined basically by the same nuclides, which determine power. At storage time to 10 years, the main nuclides are ^{244}Cm, ^{241}Pu, and ^{238}Pu, at time more than 100 years - ^{241}Am, at 3 000 years - ^{240}Pu, and at 30 000 - 100 000 years - ^{239}Pu.

The ingestion radiotoxicity of fission products is determined by ^{90}Sr and ^{137}Cs, and at times more than 1 000 years - by ^{99}Tc, ^{129}I, ^{93}Zr, and ^{135}Cs gives a little contribution.

3. URANIUM-PLUTONIUM AND THORIUM FUEL

Data on actinides only are presented in this section, because formation of fission products is practically the same as for uranium fuel. It basically depends on fuel burnup. Data on the content of actinides in spent uranium-plutonium and thorium fuel were obtained as result of model calculations in the neutron spectrum typical for uranium fuel in the active core of VVER-1000 reactor. It was supposed for calculations for uranium-plutonium fuel that uranium was extracted from spent fuel, and only isotopes of neptunium, plutonium, americium, curium were presented in stored spent fuel as actinides. At calculation of decay heat power of actinides from the uranium-thorium fuel, contribution of decay of ^{232}U was taken into account separately, while contribution of ^{228}Th was summarized with contributions of all its daughter short-lived nuclides.

Total decay heat power of actinides from spent uranium-plutonium fuel and contributions of the most important actinides in total power are shown in Fig.4. Analogous data on radiotoxicity are given in Fig.5. The data are referred to 1 ton of spent fuel.

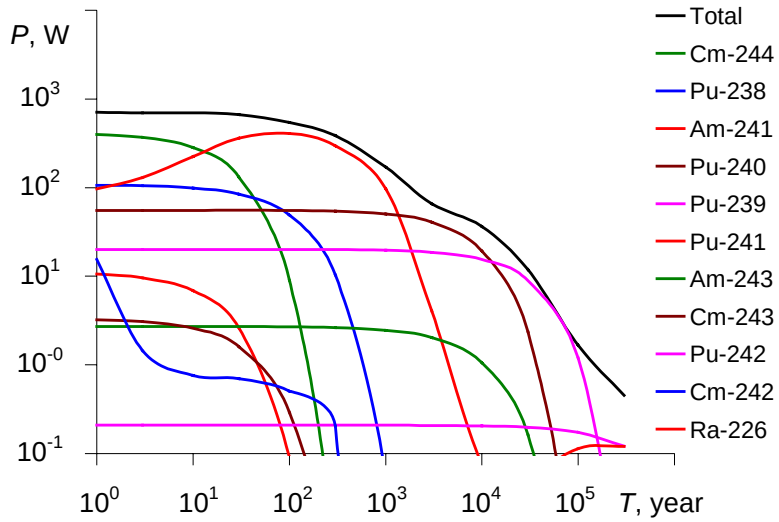

Fig.4. Decay heat power of actinides from 1 ton of spent uranium-plutonium fuel

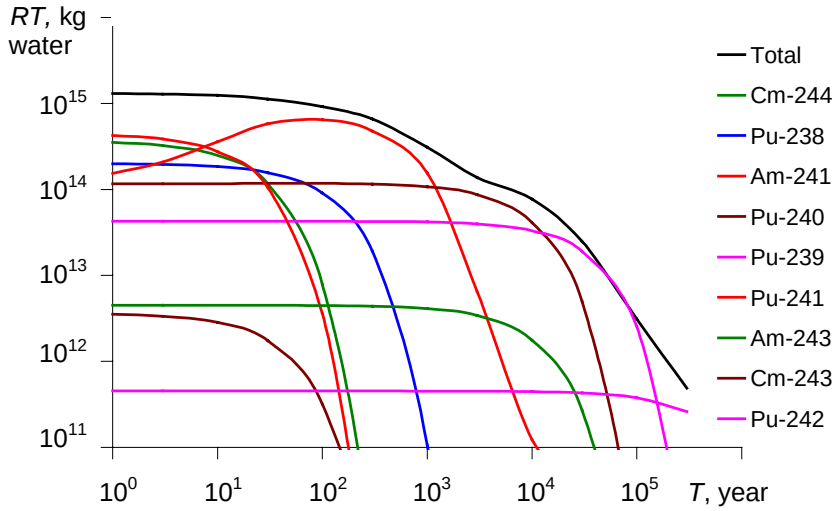

Fig.5. Radiotoxicity of actinides from 1 ton of spent uranium-plutonium fuel

Total decay heat power of actinides from spent thorium-uranium fuel and contributions of the most important actinides in total power are given in Fig.6. Analogous data on radiotoxicity are shown in Fig.7. All actinides including thorium, uranium, neptunium, plutonium, americium, curium were considered. Fresh fuel was accepted for calculations as mixture of ^{232}Th with the admixture of 3.3% of ^{233}U.

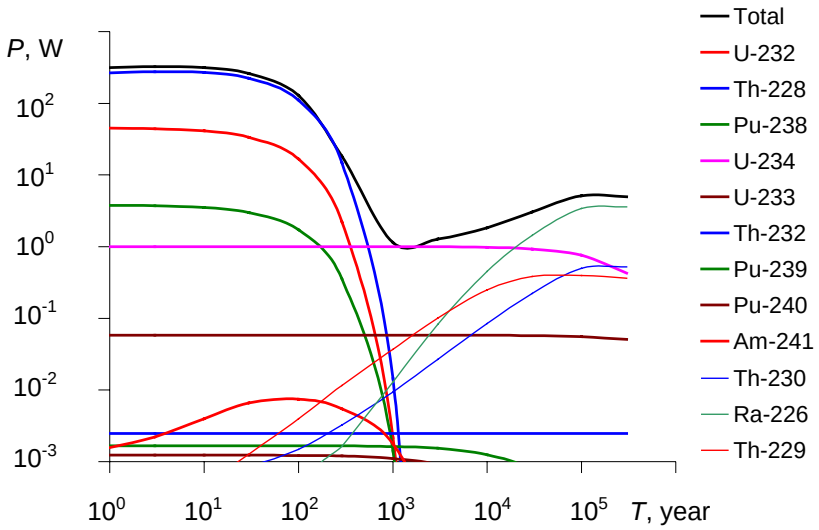

Fig.6. Decay heat power of actinides from 1 ton of spent thorium-uranium fuel

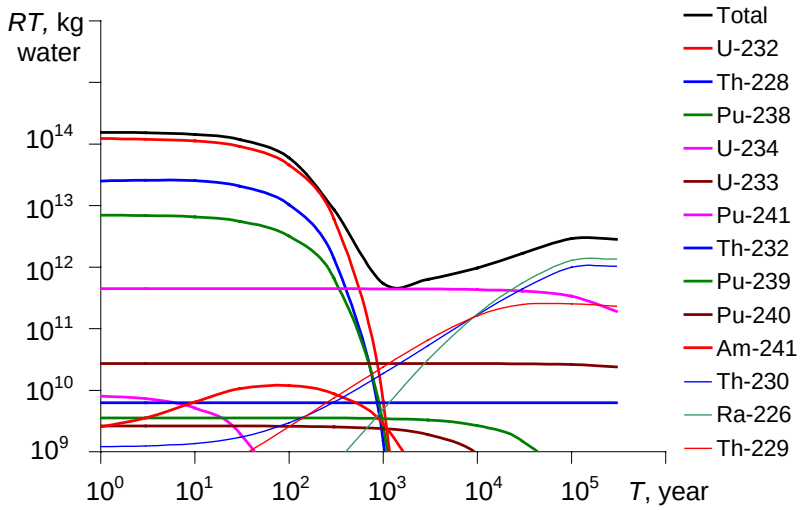

Fig.7. Radiotoxicity of actinides from 1 ton of spent thorium-uranium fuel

The results presented demonstrate the following.

Decay heat power of actinides from spent uranium-plutonium fuel during initial period of storage is due to ^{241}Am, ^{244}Cm, and ^{238}Pu. After 30 years, isotopes of plutonium give 24%, ^{241}Am - 55%, ^{244}Cm - 20% to total power. After 100 years, total power of actinides decreases by 1.4 times. Main contribution 75% gives ^{241}Am, isotopes of plutonium - 23%, ^{244}Cm – 1.6%. After 10 000 years of storage, decay heat power decreases by 20 times, after 100 000 years - by 460 times.

Radiotoxicity of actinides of spent uranium-plutonium fuel during an initial period of storage is determined by the nuclides ^{241}Am, ^{238}Pu, ^{241}Pu, and ^{244}Cm. For the time of storage of 30 years, contribution of isotopes of plutonium is 37%, ^{241}Am - 51%, ^{244}Cm - 10%. Conversion of ^{241}Pu in ^{241}Am occurs during storage. After 100 years of storage, total radiotoxicity of actinides decreases by 1.4 times. Main contribution 71% is given by ^{241}Am. Contribution of isotopes of plutonium is 28%. Amount of ^{244}Cm decreases greatly because of the decay, its radiotoxicity decreases by 46 times and makes 0,86% with respect to total radiotoxicity by the end of 100-year storage. ^{241}Am gives maximum relative contribution 72% at 300 years, after 1000 years it decreases quickly. After 10 000 years of storage, total radiotoxicity decreases by 17 times, after 100 000 years total radiotoxicity decreases by 420 times. In this period of time, main nuclides are ^{239}Pu and ^{242}Pu.

Decay heat power of actinides from spent thorium-uranium fuel during first 300 years of storage is determined by ^{228}Th together with short-lived daughter nuclides, as well as by ^{232}U, which is mother nuclide for ^{228}Th. Among the rest actinides, ^{238}Pu and ^{234}U are the most important nuclides. Their contribution is 1-2 orders of magnitude less. Certain increase of the decay heat power during first several years is explained by the fact that ^{228}Th in the discharged fuel and during subsequent cooling is far from equilibrium with his mother nuclide ^{232}U. Therefore, the amount of ^{228}Th increases during first several years of storage. For 100 years of storage, decay heat power decreases by 2.4 times. After 1000 years of storage, power is determined by the nuclides ^{234}U, ^{230}Th, in the period of storage more than 10000 years – by ^{226}Ra. The curve corresponding to ^{226}Ra in Fig.6 presents total power of ^{226}Ra together with its daughter short-lived nuclides. This total power 6.1 times exceeds the power of decay of ^{226}Ra. During the period of time after 3000 years, decay heat power increases because of accumulation of ^{226}Ra, ^{229}Th, and ^{230}Th. At storage of 10000 years, decay heat power decreases by 170 times.

Radiotoxicity of actinides from spent thorium-uranium fuel during first 300 years is determined by the nuclide ^{232}U along with daughter nuclides. Among the rest actinides, ^{238}Pu and ^{234}U are the most important. Their contribution is 1-2 orders of magnitude less. Contribution of ^{239}Pu, ^{240}Pu, ^{241}Am, and ^{232}Th is 4 orders of magnitude lower in comparison with ^{232}U. For 100 years of storage, total radiotoxicity decreases by 2.6 times. At storage during more than 1000 years, ^{232}U with the daughter nuclides decays completely. In this period of time, radiotoxicity is determined by ^{234}U, ^{230}Th. At storage during more than 10 000 years, main nuclide is ^{226}Ra, and also ^{229}Th and ^{233}U give certain contribution. During period of time after 3000 years, radiotoxicity increases due to accumulation of ^{226}Ra, ^{229}Th, and ^{230}Th. At storage time of 10 000 years, total radiotoxicity decreases by 160 times.

Total calculated radiotoxicity at the beginning of storage for actinides from uranium, uranium-plutonium, and thorium-uranium spent fuel equals accordingly $5.2 \cdot 10^{14}$, $1.3 \cdot 10^{15}$, and $1.5 \cdot 10^{14}$ kg of water per 1 ton of discharged fuel. Radiotoxicity of actinides of uranium-plutonium fuel seems to be 2.5 times higher than for usual uranium fuel due to higher (by 2-3 times) accumulations of ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{244}Cm. Radiotoxicity of actinides from thorium-uranium fuel calculated with account of ^{232}U, turns out to be 3.5 times lower than for the uranium fuel.

4. CONCLUSIONS

The data presented allow to conclude, what actinides should firstly be extracted from the storage facility for transmutation. It should be noted that we consider chemical separation of different chemical elements, rather then isotopic separation of high-active nuclides.

In case of the uranium-plutonium fuel, it is expedient to perform chemical separation of actinides before long-term storage with the allocation of plutonium, americium, and curium. Americium should be extracted after a period of 50-70 years sufficient for the conversion of ^{241}Pu in ^{241}Am. Curium can be extracted at the beginning of storage. This will allow to reduce radiotoxicity of the rest actinides on 20-30%. However, if abandon of extraction of curium, ^{244}Cm will decay almost fully during 100 years. It is possible to use fractions of plutonium and americium (may be, with long-lived isotopes of curium) for transmutation. For plutonium, this transmutation would better be identified as using as secondary fuel. Separation of actinides on fractions is also reasonable from the point of view of reducing of decay heat power. So, removing of americium after the decay of ^{241}Pu and decay of most part of ^{238}Pu allows to reduce greatly decay heat power of the plutonium fraction.

In case of the uranium-thorium fuel, major part of radiotoxicity is determined by the isotope of the same chemical element as the main fuel isotope. In this case, repeated use of thorium-uranium fuel will be accompanied by the accumulation of radiotoxicity. However, if we consider one-fold use of thorium-uranium fuel with deep burnup of ^{233}U, it should be recommended to perform additional deep burning (transmutation) of the uranium fraction containing both ^{233}U and ^{232}U. The following reduction of the radiotoxicity by several times can be related with removing and transmutation of the plutonium fraction (^{238}Pu). It is not needed to transmute ^{228}Th as far as this nuclide is not long-lived, it practically completely decays during 10 years along with their own short-lived daughter nuclides. Extraction of the uranium fractions will lead also to reducing of the decay heat power, which will be determined by ^{238}Pu after the decay of ^{228}Th.

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