CALCULATION CHARACTERIZATION OF SPENT FUEL HAZARD RELATED TO PARTITIONING AND TRANSMUTATION OF MINOR ACTINIDES AND FISSION PRODUCTS

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ABSTRACT

Radiotoxicity is one of important characteristics of radwaste hazard. Radiotoxicity of actinides and fission products from spent fuel of VVER-1000 reactor for processes of burnup, long-term storage, and transmutation is discussed.

Key words: radiotoxicity, spent fuel, transmutation

1. Introduction

Minor actinides (MA) and long-lived fission products (FP) accumulating in the nuclear fuel during reactor's operation are the most dangerous waste of atomic power industry. Hazard of long-lived FP is due to gamma-radiation accompanying beta-decay. The most dangerous FP are ^{90}Sr and ^{137}Cs with half-life near 30 years. Hazard of long-lived actinides is related with the alpha-radiation. The most dangerous are isotopes of plutonium, accumulating in the nucleus fuel. In promising atomic power, plutonium is expected to be used as secondary nuclear fuel. After plutonium, ^{241}Am and ^{244}Cm are the most dangerous actinides.

Spent fuel management can include following stages:

- intermediate cooling for the decay of short-lived nuclides,
- long-term storage with delayed decision on spent fuel management,
- partitioning,
- radiochemical processing,
- transmutation,
- further storage in less strong conditions,
- ultimate disposal.

Current and perspective ways of spent fuel management are determined in Strategy of development of the nuclear power industry in Russia [1].

2. Prospect of development of the nuclear power industry in Russia

In May 2000, the government of Russia approved "Strategy of development of the nuclear power industry in Russia in the first half of the 21st century", which was developed and presented by Russian Federation Atomic Energy Ministry [1]. The development of nuclear power plants will proceed in two stages, with the total electric power of the nuclear plants brought up to 60GW by the year 2030:

- the first stage, which is being currently implemented, amounts to developing atomic power plants that use thermal neutron reactors and accumulating plutonium for start-up and parallel development of fast neutron reactors;

- the second stage will amount to the development of nuclear power plants on a broad scale, which will mostly be fast reactors. These will gradually replace the conventional power plants operating on fossil fuel.

Today Russia has in operation 30 nuclear power reactors. Among these are 14 power-generating units with VVER reactors, 11 power-generating units with RBMK reactors, 4 EGP power-generating units of the Bilibino nuclear central heating and power plant with water-graphite channel-type reactors, and 1 power-generating unit with BN-600 fast neutron reactor.
Among the reactors of the new generation are the BREST-300 and BREST-1200 fast neutron lead-cooled reactors.

A promising area in the strategy is the passage to closed fuel cycle, in which the natural nuclear fuel and the artificial fissionable materials produced as a result of the operation of reactors (plutonium and the like) can be used more fully.

One of possible modern way to reduce radiobiological hazard of nuclear power industry is related to transmutation of long-term radwaste, which could be a part of future nuclear fuel cycle.

3. Partitioning and transmutation of radwaste

The main task of transmutation is to reduce significantly an amount and radiobiological hazard of radwaste and to facilitate their ultimate disposal.

Transmutation can be realized in specialized reactors or in power reactors. In the first case development and building of special transmutation facilities is necessary. They should be based on promising nuclear technologies with own nuclear fuel cycle and processing. However construction of such facilities is a long way in the future. In the second case both economical parameters of power reactors and nuclear fuel cycle would be made worse. In any case application of transmutation requires high level of the development of nuclear power engineering with closed nuclear fuel cycle.

Transmutation assumes preliminary partitioning of actinides and FP as well as, possibly, more detail partitioning of FP and actinides into separate fractions. As for transmutation, necessity of partitioning is explained by the fact that processes of transmutation of FP and actinides are quite different. FP are transformed in stable nuclides by means of reactions (n, γ) while actinides are transformed in other actinides and finally in fission products. Because of this fact, it is necessary to transmute FP and actinides in different conditions, possibly, in different nuclear installations.

However partitioning of nuclear waste can be important even without transmutation. Profit of partitioning is explained by the fact that different fractions of radwaste have different half-life and

different nature of radiological hazard, and for this reason require different conditions of storage. So condition of storage of nuclear waste after partitioning can be less strong.

4. Radiotoxicity as measure of radiobiological hazard of radwaste

Contents of different radioactive nuclides in spent fuel can be described in terms of mass, activity, Radiotoxicity. Radiotoxicity is more informative characteristics of radiobiological hazard of radionuclides than activity. It takes into account biological influence of radiation of concrete nuclides on the human body. Radiotoxicity RT_i for separate nuclide i is determined [2] by the 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 cubic meter of air or in one kilogram of drinking water assigned by the special normative document - Russian radiation safety standards [3] or corresponding documents of IAEA [4]. Total radiotoxicity is equal to the sum of radiotoxicities over all nuclides in considered mixture of nuclides. For such way of calculation, radiotoxicity is measured by an amount of air or water required for diluting of radionuclides to safe concentrations. Accordingly, it is called inhalation radiotoxicity or ingestion radiotoxicity.

In table 1 mass M, activity A, inhalation and ingestion radiotoxicity *RT inh* and *RT ing* of certain most important fission products and actinides ^{90}Sr, ^{99}Tc, ^{237}Np, ^{239}Pu, ^{241}Am, ^{244}Cm [5] from one ton of spent fuel of VVER-1000 type reactor are presented.

Table 1. Mass, activity, and radiotoxicity of ^{90}Sr, ^{99}Tc, ^{237}Np, ^{239}Pu, ^{241}Am, ^{244}Cm from 1 t of spent fuel of VVER-1000 type reactor

Characteristics	^{90}Sr	^{99}Tc	^{237}Np	^{239}Pu	^{241}Am	^{244}Cm
M, kg	0.63	0.95	0.63	5.5	0.24	0.042
A, 10^{12} Bq	$3.2\cdot10^{3}$	0.60	0.016	13	30	120
RT inh, 10^{15} m^3 air	1.2	$2.2\cdot10^{-5}$	$3.0\cdot10^{-3}$	5.1	10	27
RT ing, 10^{12} kg water	640	$2.7\cdot10^{-3}$	0.013	23	44	100

These data demonstrate that proportions between radiotoxicities of nuclides can differ significantly from proportions between masses of these nuclides. So, ^{244}Cm with the least mass gives the most inhalation radiotoxicity while ^{90}Sr gives the most ingestion radiotoxicity. At the same time long-lived nuclides ^{99}Tc and ^{237}Np give much lesser radiotoxicity.

Masses and radiotoxicities of basic actinides accumulated in one ton of fuel of VVER-1000 type reactor at different burnups W and subsequent 3-year cooling [6] are presented in fig.1 – 3. Increased burnup values are taken into consideration. These data demonstrate high increase of mass contents and radiotoxicities of ^{238}Pu and ^{244}Cm with the burnup increase.

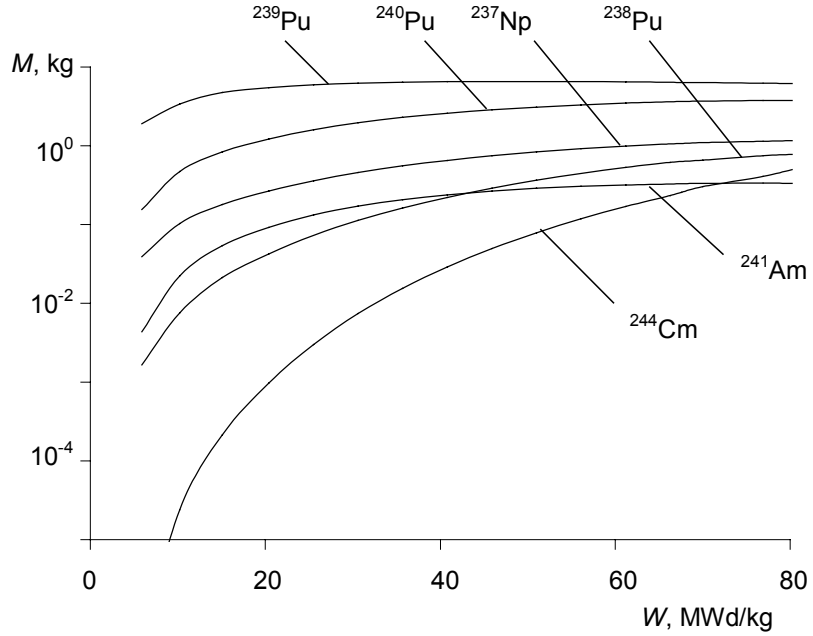

Fig.1. *Masses of base actinides in fuel of VVER-1000 type reactor*

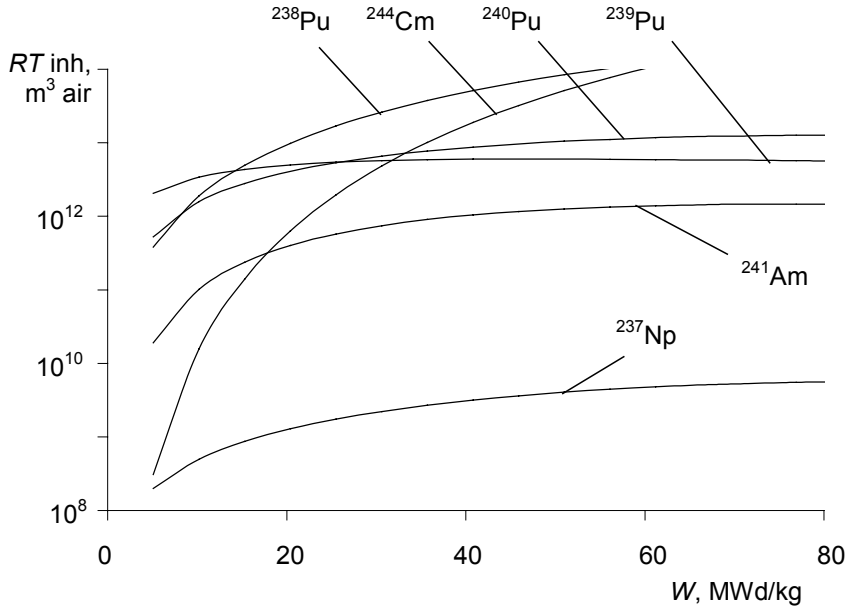

Fig.2. *Inhalation radiotoxicities of base actinides in fuel of VVER-1000 type reactor*

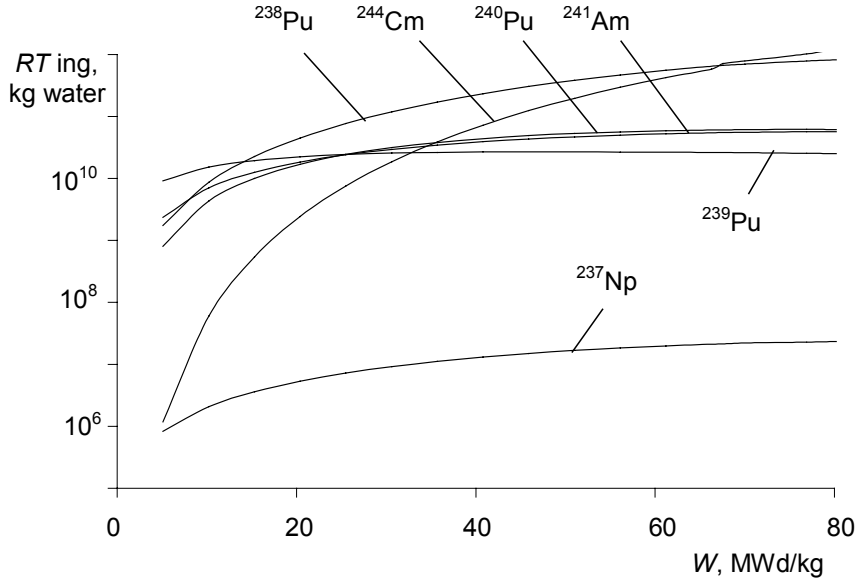

Fig.3. *Ingestion radiotoxicities of base actinides in fuel of VVER-1000 type reactor*

In recent years some authors calculate radiotoxicity by another way using dose factors ε_i presented also in radiation safety standards [3, 4]:

$$RT_i = A_i \, \varepsilon_i \, .$$

This radiotoxicity is measured in Sievert. Calculation is based on the fact that dose factor for given nuclide ε_i multiplied by the activity A_i gives an effective dose got by the person under inhalation or ingestion consumption of given amount of radionuclide *i*. Paper [7] illustrates calculation of radiotoxicity in Sievert for estimation of potential biological hazard.

Radiotoxicity calculated in cubic meters of air or in kg of water for given nuclide could be transformed to radiotoxicity in Sievert by multiplying by constant factors. These factors are determined in radiation safety standards [3]. Annual air consumption depends on age groups. For the age group of 12-17 years annual air consumption equals 7300 m^3, and for the age group upward of 17 years it is 8100 m^3. Annual dose for inhalation consumption of radionuclide diluted to limiting concentrations equals 1 mSv. Analogous data are available for ingestion consumption.

Fig.4 and 5 illustrate inhalation radiotoxicity of actinides and FP during long-term storage calculated both in cubic meters of air and in Sieverts (with corresponding scales on left and right ordinate axis).

As far as inhalation radiotoxicity of all important actinides is determined for the age group upward of 17 years, radiotoxicity of actinides in Sieverts is equal to the radiotoxicity in m^3 air multiplied by 1 mSv and divided by 8100 m^3 air. Analogous simple calculation for FP provides a mistake of approximately 10%, as far as two reference age groups are applied for most important nuclides: group of 12-17 years and group upward of 17 years. Data of fig.4 and 5 are referred to nuclides extracted from 1 ton of spent fuel of power reactor VVER-1000 with increased burnup of 70 MWd/kg with subsequent cooling during 3 years. Analogous data for common-type burnup of 40 MWd/kg were presented in the YUNSC-2000 conference proceedings [5].

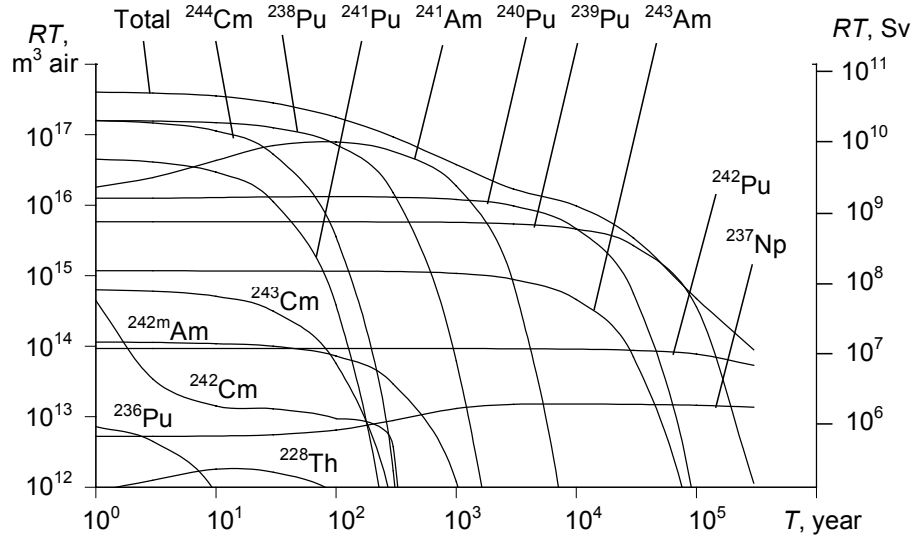

Fig.4. *Inhalation radiotoxicity of actinides*

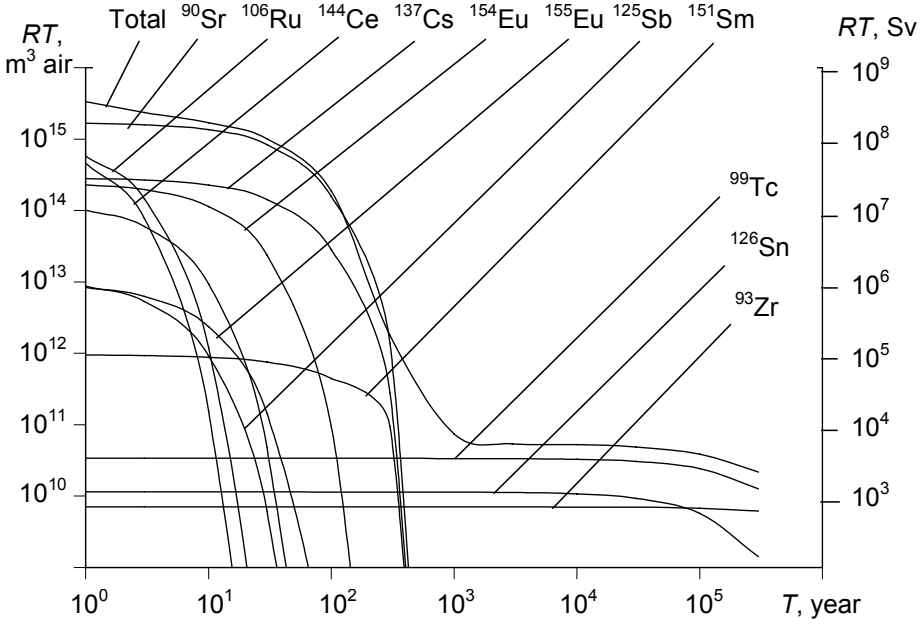

Fig.5. *Inhalation radiotoxicity of fission products*

Fig.6 illustrates final stage of nuclear power when power reactors cease operation, and it is necessary to transmute actinides accumulated in transmutation reactors [8]. It was assumed for calculations that transmutation was performed in thermal neutron reactors with neutron flux of 10^{14} $cm^{-2}s^{-1}$. The data presented demonstrate that isotopes of plutonium, americium, light isotopes of curium are transmuted firstly. At the same time heavy isotopes of curium, isotopes of berkelium, californium, and einsteinium are formed. Rather long time or high neutron flux are required for their transmutation.

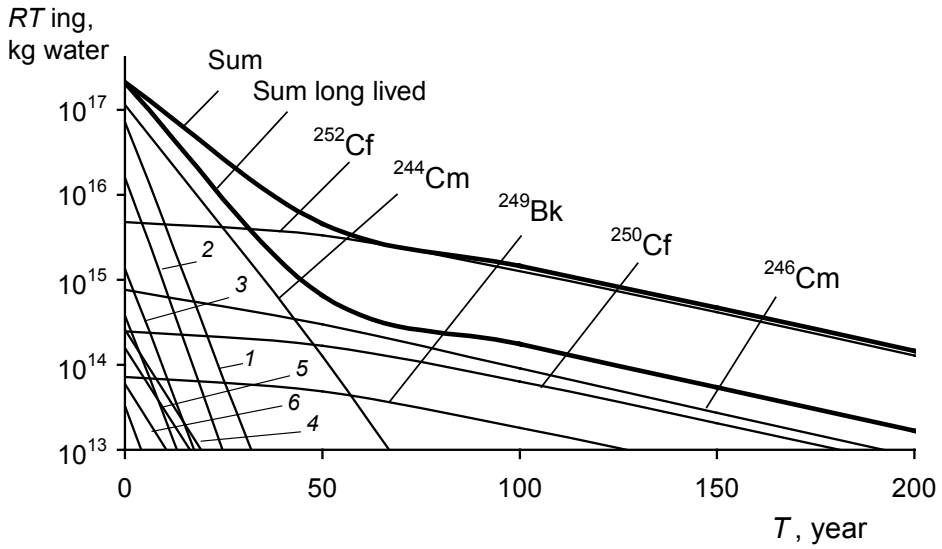

Fig.6. *Transmutation of actinides in final stage of nuclear power*.
1 – ^{242}Cm, *2* – ^{238}Pu, *3* – ^{241}Am, *4* – ^{241}Pu, *5* – ^{243}Am, ^{243}Cm, *6* – ^{239}Pu, ^{240}Pu

5. Conclusions

The concept of radiotoxicity is useful for description of radiobiological hazard of radwaste in processes of waste formation in nuclear fuel, partitioning, storage, and transmutation. The results of calculations presented can be used for the development of recommendations on partitioning, storage, and transmutation of high active waste.

Increase of fuel burnup from 40 to70 MWd/kg leads to the increase of the mass of ^{238}Pu discharged from 1 ton of spent fuel by 3 times, ^{244}Cm – by 10 times. Annual discharge of ^{238}Pu from the reactor increases by 1.8 times, ^{244}Cm – by 6 times.

Americium and curium should be separated from other actinides for subsequent storage during 100 years with the decay of ^{244}Cm and transmutation of americium. Cs and Sr should be separated from other FP for subsequent storage during 100 years for the decay of ^{90}Sr, ^{137}Cs

6. References

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