RADIATION CHARACTERISTICS OF PWR MOX SPENT FUEL AFTER LONG-TERM STORAGE BEFORE TRANSMUTATION IN ACCELERATOR DRIVEN SYSTEMS

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Abstract

Changes in a radiotoxicity and decay heat power of actinides from spent uranium and uranium-plutonium nuclear fuel for PWR-type reactors at long-term storage are investigated. The extraction of the most important nuclides for transmutation permits to reduce radiotoxic content of wastes remaining in storage. The decay heat power of actinides is determined by the same nuclides as the radiotoxicity is. The radiotoxicity and decay heat power of actinides of uranium-plutonium fuel is 2.5 times higher, than that of uranium fuel because of the greater accumulation of plutonium, americium, and curium isotopes.

1. Introduction

The problem of the management of long-lived radioactive waste from spent nuclear fuel is closely connected to the prospects for development of nuclear power. Some directions for decisions to this problem are now studied. One of the opportunities is the construction of a long-term controllable storage facility. Other ways are connected with the realisation of nuclear transmutation of long-lived waste. Apparently, the resolution of the problem will combine several approaches. To determine their correct combination, it is necessary to know how the major characteristics of radioactive waste vary during long-term storage.

One of the important radiation characteristics of radwaste is the decay heat power. It influences the design of a storage facility and the type of heat removal system. Other important characteristic of radwaste is the radiotoxicity. It is a more representative characteristic than activity because radiotoxicity includes the influence of radiation of separate nuclides on the human body. Radiotoxicity is a base to evaluate ecological danger of stored radwaste.

Time dependence of radwaste radiation characteristics during storage allows identifying most important nuclides in different stages of storage. Their separation and extraction from storage with subsequent transmutation permits to reduce the radiologic danger of wastes staying in storage. Removal of nuclides with a decrease of the remaining decay heat power in storage permits to ease requirements to the heat removal systems at long-term storage of wastes.

Changes in radiotoxicity and decay heat power of actinides from spent uranium – plutonium nuclear fuel of VVER-1000 type reactors at storage during 100 000 years are investigated in this paper.

The radiotoxicity RT_i of each nuclide *i* by air or by water is determined by the ratio:

$$RT_i = A_i/MPA_i$$

where A_i – activity of considered amount of a nuclide *i*, MPA_i – represents the maximum permissible activity of this nuclide by air or by water according to radiation safety standards. Total radiotoxicity is equal to a sum of radiotoxicities of all nuclides taken in those amounts in which they are contained in the considered mix of nuclides. For the calculations, data of MPA accepted in the Russian Federation [1] were used. For the calculations of a decay heat power, the contributions from alpha-, beta- and gamma – radiations [2] were taken into account.

2. Calculation results

The total radiotoxicity of actinides in air and in water and the contributions of the most important actinides in total radiotoxicity at storage of spent uranium-plutonium fuel of a VVER-1000 type reactor during 100 000 years are presented in Tables 1 and 2. The amount of actinides corresponded to their contents in 1 tonne of spent fuel with burn-up of 40 kg of fission products per 1 tonne and subsequent cooling during 3 years. The fresh fuel was a mix of depleted uranium with addition of 3.5% ^{239}Pu. Only isotopes of neptunium, plutonium, americium, and curium were considered as actinides.

The total decay heat power of actinides and the contributions of the most important actinides at storage of spent uranium-plutonium fuel during 100 000 years is given in Table 3. Decay heat power, as well as the radiotoxicity, corresponds to the content of actinides in 1 tonne of unloaded fuel.

Table 1**. Radiotoxicity of actinides from uranium-plutonium spent fuel in air, m^3 air**

T, year	1	10	100	1 000	10 000	100 000
^{238}Pu	4.39 + 16	4.09 + 16	2.02 + 16	2.35 + 13	–	–
^{239}Pu	9.57 + 15	9.57 + 15	9.55 + 15	9.34 + 15	7.38 + 15	5.68 + 14
^{240}Pu	2.62 + 16	2.63 + 16	2.63 + 16	2.40 + 16	9.23 + 15	–
^{241}Pu	8.78 + 16	5.69 + 16	7.49 + 14	1.19 + 12	–	–
^{242}Pu	1.01 + 14	1.01 + 14	1.01 + 14	1.01 + 14	9.92 + 13	8.41 + 13
^{241}Am	3.67 + 16	8.54 + 16	1.55 + 17	3.69 + 16	2.90 + 13	–
^{242m}Am	2.93 + 14	2.81 + 14	1.87 + 14	3.08 + 12	–	–
^{243}Am	1.03 + 15	1.03 + 15	1.03 + 15	9.42 + 14	4.05 + 14	–
^{243}Cm	8.21 + 14	6.59 + 14	7.39 + 13	–	–	–
^{244}Cm	9.15 + 16	6.48 + 16	2.07 + 15	–	–	–
Total	2.99 + 17	2.86 + 17	2.15 + 17	7.13 + 16	1.72 + 16	6.80 + 14

Table 2. **Radiotoxicity of actinides from uranium-plutonium spent fuel in water, kg water**

T, year	1	10	100	1 000	10 000	100 000
^{238}Pu	1.98 + 14	1.84 + 14	9.10 + 13	1.06 + 11	–	–
^{239}Pu	4.27 + 13	4.27 + 13	4.26 + 13	4.17 + 13	3.29 + 13	2.54 + 12
^{240}Pu	1.17 + 14	1.17 + 14	1.18 + 14	1.07 + 14	4.12 + 13	–
^{241}Pu	4.24 + 14	2.75 + 14	3.62 + 12	–	–	–
^{242}Pu	4.52 + 11	4.52 + 11	4.52 + 11	4.51 + 11	4.45 + 11	3.77 + 11
^{241}Am	1.54 + 14	3.59 + 14	6.50 + 14	1.55 + 14	1.22 + 11	–
^{242m}Am	1.32 + 12	1.27 + 12	8.44 + 11	–	–	–
^{243}Am	4.50 + 12	4.50 + 12	4.46 + 12	4.10 + 12	1.76 + 12	–
^{243}Cm	3.53 + 12	2.84 + 12	3.18 + 11	–	–	–
^{244}Cm	3.51 + 14	2.49 + 14	7.93 + 12	–	–	–
Total	1.30 + 15	1.24 + 15	9.19 + 14	3.09 + 14	7.67 + 13	3.11 + 12

Table 3. **Decay heat power of actinides from uranium-plutonium spent fuel, W**

T, year	1	10	100	1000	10 000	100 000
^{226}Ra	–	–	–	–	0.015	0.114
^{238}Pu	106	99.0	48.9	0.057	–	–
^{239}Pu	20.1	20.1	20.1	19.6	15.5	1.19
^{240}Pu	55.0	55.3	55.4	50.4	19.4	0.0014
^{241}Pu	10.6	6.85	0.090	–	–	–
^{242}Pu	0.209	0.209	0.209	0.209	0.206	0.174
^{241}Am	96.5	225	407	96.9	0.076	–
^{243}Am	2.70	2.70	2.68	2.46	1.06	–
^{242}Cm	15.5	0.763	0.506	–	–	–
^{243}Cm	3.24	2.60	0.292	–	–	–
^{244}Cm	398	282	9.00	–	–	–
^{245}Cm	0.162	0.162	0.160	0.149	0.071	–
Total	708	694	544	170	36.5	1.66

The data presented show that the radiotoxicity of actinides of spent uranium-plutonium fuel in air in an initial period of storage is determined by nuclides ^{244}Cm, ^{241}Pu and ^{238}Pu. Their contribution in beginning of the storage is about 75%. All isotopes of plutonium give 56%, ^{244}Cm – 30%. In addition, ^{241}Am creates 12% of radiotoxicity. At storage there is the conversion ^{241}Pu into ^{241}Am. After 100 years of storage, total radiotoxicity of actinides decreases 1.4 times. The main contribution (72%) comes from ^{241}Am. The contribution of plutonium isotopes makes 26%. The amount of ^{244}Cm decreases essentially because of the decay. Its radiotoxicity falls 44 times and makes 1% of total radiotoxicity at the end of 100-year storage. After 1 000 years of storage, radiotoxicity in air falls 4.2 times, after 10 000 years – 17 times, after 100 000 years it falls 440 times.

The radiotoxicity in water is determined by the same nuclides. In initial period of storage, all plutonium isotopes give the contribution in total radiotoxicity 60%, ^{244}Cm – 27%, ^{241}Am – 12%. After 100 years of storage, total radiotoxicity of actinides decreases 1.4 times. The main contribution 71% gives ^{241}Am. The contribution of plutonium isotopes makes 28%, ^{244}Cm – 0.9%. The ^{241}Am gives maximal respective contribution 72% after 300 years. After 1 000 years its share decreases quickly. After 1 000 years, total radiotoxicity in water reduces 4.2 times, after 10 000 years – 17 years, after 100 000 years – 420 times.

The decay heat power of actinides of spent uranium-plutonium fuel in initial period of storage is determined by a nuclide ^{244}Cm, which creates 56% of power. The contribution of plutonium isotopes makes 27%, ^{241}Am – 13%. After 100 years of storage, total power of actinides decreases 1.3 times. The main contribution 75% gives ^{241}Am, plutonium isotopes – 23%, ^{244}Cm – 1.6%. After 10 000 years, power reduces 20 times, after 100 000 years – 460 times.

The radiotoxicity of actinides of uranium-plutonium fuel appears 2.5 times more and the decay heat power appears 2.7 times more than that of usual uranium fuel because of greater (by 2-3 times) accumulation of ^{239}Pu, ^{240}Pu, ^{241}Pu, and ^{244}Cm.

3. Conclusion

Recommendations could be done to perform chemical separation of plutonium, americium, curium before long-term storage. Americium should be separated after 50-70 year of storage sufficient for conversion ^{241}Pu in ^{241}Am. Curium can be separated in the beginning of storage. This will allow reducing radiotoxicity of the remaining actinides by 20-30%. If we abandon a separation of curium then it decays in 100 years almost fully. Extracted americium (possibly, with long-lived curium isotopes) should be directed to transmutation and plutonium – to repeated use. The separation of actinides is expedient also to reduce decay heat power. So, extraction of americium after ^{241}Pu decay and decay of greater part of ^{238}Pu-permits to reduce essentially decay heat power of the plutonium fraction.

REFERENCES

[1] *Radiation Safety Standards (NRB-99),* Minzdrav of Russia, Moscow, 1999.

[2] *Schemes of Decay of Radionuclides. Energy and Intensity of Irradiation,* Publication 38 ICRS, Moscow, Energoatomizdat, 1987.