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Comparison of radiation hazard of HLW in several spent nuclear fuel reprocessing scenarios

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Abstract. Radiation hazard of radionuclide has been calculated as a product of $A\varepsilon$ where A is an activity of radionuclide and ε is a dose coefficient through ingestion. The values $A\varepsilon$ of 18 radionuclide in spent fuel of WWER-440 are calculated. Because the full division of americium and curium from HLW is very complicated a separation americium from curium is considered. It is shown that a separation of americium in a special fraction allows decreasing the radiation hazard of HLW by 97.6% after 1000 years.

Introduction

The modern fuel reprocessing is based on PUREX-process. Resulting HLW contains less than 0.01% of uranium, 0.025% of plutonium and 0.5% of neptunium [1]. Other radionuclides remaining in HLW are: americium, curium and fission products. Today's reality requires more safety. It is necessary to decrease contents of americium and curium in HLW making it significantly safer for future storage [2]. Separation of only americium can be considered as a partial measure due to significant impact of curium on radiation hazard in period around 700-800 years [2]. The aim of this work is comparison of radiation safety in those scenarios of HLW management.

Experimental data

Composition of radionuclide in spent fuel of WWER-440 [3] is given in Table 1. This data was taken as a base. Then activities of radionuclide are calculated for three periods: after stop, after 5 years storage and after reprocessing and dividing of uranium, plutonium and neptunium as in [1].

Table 1. Composition of spent fuel of WWER-440 [3] and activities of actinides

Radio-nuclide	$T_{1/2}$, years	Dose coefficient $\varepsilon\cdot10^7$, Sv/Bq	Content, kg/t U [3]	Activity A, Bq/t U		
				After stop	After 5 years storage	After repro-cessing
1	2	3	4	5	6	7
^{232}U	68.9	3.3	$9.00\cdot10^{-7}$	$7.44\cdot10^{8}$	$1.59\cdot10^{9}$	$1.59\cdot10^{5}$
^{234}U	$2.45\cdot10^{5}$	0.49	0.215	$4.96\cdot10^{10}$	$5.09\cdot10^{10}$	$5.09\cdot10^{6}$
^{235}U	$7.04\cdot10^{8}$	0.47	11.2	$8.95\cdot10^{8}$	$8.96\cdot10^{8}$	$8.96\cdot10^{4}$
^{236}U	$2.34\cdot10^{7}$	0.47	4.50	$1.08\cdot10^{10}$	$1.08\cdot10^{11}$	$1.08\cdot10^{7}$
^{238}U	$4.47\cdot10^{9}$	0.45	941	$1.17\cdot10^{10}$	$1.17\cdot10^{10}$	$1.17\cdot10^{6}$
^{237}Np	$2.14\cdot10^{6}$	1.1	0.445	$1.16\cdot10^{10}$	$4.71\cdot10^{9}$	$2.35\cdot10^{7}$
^{236}Pu	2.85	2.0	$1.60\cdot10^{-6}$	$3.15\cdot10^{10}$	$9.41\cdot10^{9}$	$2.35\cdot10^{6}$
^{238}Pu	87.7	2.3	0.137	$8.67\cdot10^{13}$	$9.16\cdot10^{13}$	$2.29\cdot10^{10}$

1	2	3	4	5	6	7
^{239}Pu	$2.41\cdot10^{4}$	2.5	5.37	$1.23\cdot10^{13}$	$1.23\cdot10^{13}$	$3.08\cdot10^{9}$
^{240}Pu	$6.56\cdot10^{3}$	2.5	2.17	$1.82\cdot10^{13}$	$1.82\cdot10^{13}$	$4.56\cdot10^{9}$
^{241}Pu	14.4	0.048	1.06	$4.04\cdot10^{15}$	$3.18\cdot10^{15}$	$7.94\cdot10^{11}$
^{242}Pu	$3.76\cdot10^{5}$	2.4	0.430	$6.25\cdot10^{10}$	$6.25\cdot10^{10}$	$1.56\cdot10^{7}$
^{241}Am	432	2.0	0.0410	$5.20\cdot10^{12}$	$3.38\cdot10^{13}$	$3.38\cdot10^{13}$
^{242m}Am	152	1.9	$6.40\cdot10^{-4}$	$2.30\cdot10^{11}$	$2.25\cdot10^{11}$	$2.25\cdot10^{11}$
^{243}Am	$7.38\cdot10^{3}$	2.0	0.0490	$3.61\cdot10^{11}$	$3.61\cdot10^{11}$	$3.61\cdot10^{11}$
^{242}Cm	0.446	0.12	0.0136	$1.67\cdot10^{15}$	$9.29\cdot10^{11}$	$9.29\cdot10^{11}$
^{243}Cm	28.5	1.5	$3.80\cdot10^{-4}$	$7.26\cdot10^{11}$	$6.45\cdot10^{11}$	$6.45\cdot10^{11}$
^{244}Cm	18.1	1.2	0.0197	$5.89\cdot10^{13}$	$4.87\cdot10^{13}$	$4.87\cdot10^{13}$
^{245}Cm	$8.50\cdot10^{3}$	2.1	$1.03\cdot10^{-5}$	$6.54\cdot10^{9}$	$6.54\cdot10^{9}$	$6.54\cdot10^{9}$
^{246}Cm	$4.73\cdot10^{3}$	2.1	$6.90\cdot10^{-5}$	$7.84\cdot10^{5}$	$7.84\cdot10^{5}$	$7.84\cdot10^{5}$
		$\sum$	**967**	$\mathbf{5.89\cdot10^{15}}$	$\mathbf{3.38\cdot10^{15}}$	$\mathbf{8.57\cdot10^{13}}$

After 5 years storage activities of some radionuclide became less and it was seen from Table 2.

Table 2. Decay of some radionuclide after 5 years storage.

Radio-nuclide	$T_{1/2}$, year	Content g/t U		Decay products		
		Initial	Final	Nuclide	Mass g/t U	Bq/t U
^{241}Pu	14.4	1060	832	^{241}Am	228	$2.86\cdot10^{13}$
^{242}Cm	0.446	13.6	0.006	^{238}Pu	13.5	$8.49\cdot10^{12}$
^{243}Cm	28.5	0.38	0.34	^{239}Pu	0.04	$0.8\cdot10^{8}$
^{244}Cm	18.1	19.7	16.3	^{240}Pu	3.4	$2.87\cdot10^{10}$

Radiation hazard

The activities of original radionuclide together with their decay products were calculated as function of time. The radiation hazard of radionuclide is calculated as a product of $A\varepsilon$ where A is an activity of radionuclide and ε is a dose coefficient through ingestion [4]. The results of the calculation are given in Table 3 and they only include dose coefficients of radionuclide with $T_{1/2} > 15$ days. There are 18 nuclide presented in Table 1 after reprocessing (column 7). The data in Table 3 (column 2) presented $A\varepsilon$ at 0. The data at other columns were calculated from the data at column 2 and decay results for column 2. Therefore it is possible to divide the data $A\varepsilon$ into 18 lines each corresponding to the radiation hazard in various time periods.

Table 3. Alteration of radiation hazard of actinides during HLW storage

Radio-nuclide	Radiation hazard $A\cdot\varepsilon$, Sv/t U after storage during						
	0 years	10 years	1000 years	10000 years	50000 years	10^5 years	10^6 years
1	2	3	4	5	6	7	8
^{232}U	$5.23\cdot10^{-2}$	0.0577	0*)	0*)	0*)	0*)	0*)
^{234}U	0.249	0.249	0.276	1.07	4.18	6.27	1.06
^{235}U	$4.21\cdot10^{-3}$	$4.23\cdot10^{-3}$	$7.73\cdot10^{-3}$	0.0369	0.116	0.155	0.176
^{236}U	$5.06\cdot10^{-2}$	0.0506	0.0506	0.0506	0.0505	0.0505	0.0492
^{238}U	$5.27\cdot10^{-2}$	0.0566	0.0568	0.0605	0.132	0.304	2.62
^{237}Np	6.4	6.45	6.47	7.07	12.3	18.1	33.3
^{236}Pu	0.47	0.0743	0*)	0*)	0*)	0*)	0*)
^{238}Pu	$5.27\cdot10^{3}$	$5.27\cdot10^{3}$	2.38	1.69	6.66	10.0	16.9
^{239}Pu	770	770	749	578	183	43.6	0.207
^{240}Pu	$1.14\cdot10^{3}$	$1.14\cdot10^{3}$	$1.03\cdot10^{3}$	39.6	5.85	0.0893	0.0584
^{241}Pu	$3.81\cdot10^{3}$	$4.36\cdot10^{3}$	$1.10\cdot10^{3}$	0.644	1.12	1.66	3.06
^{242}Pu	3.75	3.75	3.74	3.68	3.42	3.12	0.592
1	2	3	4	5	6	7	8
(1) $\sum$U+Pu	$\mathbf{1.10\cdot10^{4}}$	$\mathbf{1.11\cdot10^{4}}$	$\mathbf{2.89\cdot10^{3}}$	**631**	**217**	**83.3**	**58.0**

^{242}Cm	$1.11\cdot10^{4}$	$1.00\cdot10^{3}$	0.490	0.346	1.37	2.05	0.348
^{243}Cm	$9.66\cdot10^{4}$	$7.62\cdot10^{4}$	189	146	46.2	11.0	0.0523
^{244}Cm	$5.84\cdot10^{6}$	$3.99\cdot10^{6}$	$3.03\cdot10^{4}$	$1.17\cdot10^{4}$	173	2.63	1.72
^{245}Cm	$1.37\cdot10^{3}$	$1.39\cdot10^{3}$	$2.28\cdot10^{3}$	$1.23\cdot10^{3}$	51.9	8.26	14.9
^{246}Cm	0.165	0.164	0.143	0.0402	0.0023	0.0020	0 *)
(2) ∑Cm	**$5.95\cdot10^{6}$**	**$4.08\cdot10^{6}$**	**$3.57\cdot10^{4}$**	**$1.31\cdot10^{4}$**	**272**	**23.9**	**17.0**
^{241}Am	$6.76\cdot10^{6}$	$6.65\cdot10^{6}$	$1.36\cdot10^{6}$	824	$1.43\cdot10^{3}$	$2.12\cdot10^{3}$	$3.91\cdot10^{3}$
^{242m}Am	$4.26\cdot10^{4}$	$4.68\cdot10^{4}$	$1.29\cdot10^{3}$	26.1	105	158	26.7
^{243}Am	$7.22\cdot10^{4}$	$7.22\cdot10^{4}$	$6.82\cdot10^{4}$	$4.25\cdot10^{4}$	$9.74\cdot10^{3}$	$2.25\cdot10^{3}$	7.43
(3) ∑Am	**$6.87\cdot10^{6}$**	**$1.09\cdot10^{7}$**	**$1.47\cdot10^{6}$**	**$4.34\cdot10^{4}$**	**$1.13\cdot10^{4}$**	**$4.53\cdot10^{3}$**	**$3.94\cdot10^{3}$**

Table 4. The data for some fission products.

Rardio-nuclide	$T_{1/2}$, years	Yield at ^{235}U/^{239}Pu,%	Dose coeffi-cient $\varepsilon\cdot10^{7}$, Sv/Bq	Activity A_0, Bq/t	Radiation hazard $A\cdot\varepsilon$, Sv/t U			
					0 years	500 years	1000 years	50000 years
^{79}Se	$6.5\cdot10^{4}$	0.0453/ 0.0470	2.9	$1.4\cdot10^{10}$	28.0	28.0	28.0	26.0
^{90}Sr/^{90}Y	29.1	5.90/2.10	28+2.7	$2.73\cdot10^{15}$	$9.2\cdot10^{7}$	621	0	0
^{93}Zr	$1.53\cdot10^{6}$	6.39/3.90	1.1	$6.7\cdot10^{10}$	88.0	88.0	88.0	86.0
^{99}Tc	$2.13\cdot10^{5}$	6.11/6.14	0.64	$5.7\cdot10^{11}$	375	375	375	318
^{107}Pd	$6.5\cdot10^{6}$	0.139/3.36	0.037	$5.3\cdot10^{9}$	0.0295	0.0295	0.0295	0.0293
^{126}Sn	$1.0\cdot10^{5}$	0.055/0.266	4.7	$3.2\cdot10^{10}$	33.4	33.4	33.4	28.8
^{129}I	$1.57\cdot10^{7}$	0.353/0.749	110	$7.0\cdot10^{8}$	92.0	92.0	92.0	91.8
^{135}Cs	$2.3\cdot10^{6}$	6.53/7.62	2.0	$1.50\cdot10^{10}$	30.0	30.0	30.0	29.6
^{137}Cs	30.0	6.27/6.73	13	$4.30\cdot10^{15}$	$5.2\cdot10^{7}$	515	0	0

Possible scenarios of fuel reprocessing

Then radionuclide from Table 3 can be divided in three groups. The first group (I) included the radionuclide of uranium, neptunium and plutonium. The second group (II) included the radionuclide of curium and the thirst group (III) included the radionuclide of americium. Besides data for some important fission products are included in Table 4. The data $A\varepsilon$ for fission products is decreased fast from 0 to 500÷1000 years due to decay of strontion-90 and cesium-137. The data $A\varepsilon$ for fission products after 1000 years is lightly more than $A\varepsilon$ for actinides.

Table 5. Possible scenarios of fuel reprocessing.

Radio-nuclide	Radiation hazard $A\cdot\varepsilon$, Sv/t U after storage during					
	0 years	10 years	1000 years	50000 years	10^5 years	10^6 years
Scenario I. ∑ (U+Np+Pu)	11000	11100	2890	217	83.3	58.0
∑ Cm	$5.95\cdot10^{6}$	$4.07\cdot10^{6}$	$3.28\cdot10^{4}$	272	23.9	17.0
Scenario II. ∑(U+Np+Pu+Cm)	$5.96\cdot10^{6}$	$4.08\cdot10^{6}$	$3.57\cdot10^{4}$	489	107	75.0
∑Am	$6.87\cdot10^{6}$	$6.77\cdot10^{6}$	$1.43\cdot10^{6}$	11300	4530	3940
Scenario III. ∑(U+Np+Pu+Cm+Am)	$1.28\cdot10^{7}$	$1.08\cdot10^{7}$	$1.47\cdot10^{6}$	11800	4640	4020
∑Am/∑(U+Np+Pu+Cm+Am)	0.536	0.624	0.976	0.958	0.977	0.981
Fission products	$14.0\cdot10^{7}$	-	646	580	-	-

The data Table 5 can be used to divide three possible scenarios:

Scenario I. Americium and curium are separated in other residue. HLW contain uranium, plutonium and neptunium. Concentrations of americium and curium are known. This scenario can be considered as ideal.

Scenario II. Americium is separated solely while curium remains in HLW [2]. The effect of americium separation was calculated as relation of ∑Am/∑(U+Np+Pu+Cm+Am).
Scenario III. The values of Aε are maximal as americium and curium together with uranium, neptunium and plutonium are all part of nuclear waste. This process is similar to currently used.

Conclusion.

Obviously the scenario I (first) is optimal. A separation of americium in the scenario II allows decreasing a part of problem. But it is possible that some part of americium or curium can be remained in HLW. Then the relation ∑Am/∑(U+Np+Pu+Cm+Am) can be used to estimate the danger posed by the americium remaining in HLW. It is necessary to note that remains of americium in HLW pose much more danger than remains of curium in HLW.

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