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POSSIBILITY OF PLUTONIUM BURNING OUT AND MINOR ACTINIDES TRANSMUTATION IN CANDU TYPE REACTOR

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The possibility of power or weapon-grade plutonium use as nuclear fuel in CANDU type reactor with simultaneous minor actinides burn-out is studied. Total thermal power is 1900 MW. The fuel lifetime makes 0.24 years, neutron flux density 10^{14} neutr/cm^{2}s. About 40-45 % of plutonium is incinerated during fuel lifetime. If weapon-grade plutonium is used in fuel channels instead of power one, its consumption is 40% lower.

1. INTRODUCTION

The problem of incineration of plutonium and minor actinides accumulated in spent fuel of nuclear reactors is a part of total problem of radioactive waste management. Power plutonium is accumulated in spent fuel of power reactors. Weapon-grade plutonium is specially produced for militarian purposes. In perspective projects of plutonium utilization and minor actinides transmutation, the main role is usually assigned to fast reactors in which there is the effective fission of plutonium isotopes and minor actinides. However existing problems in development of fast reactors require to investigate other opportunities. It seems rather real to use plutonium as fuel in well approved and reliable reactors on thermal neutrons. Due to features of CANDU reactor ensuring small parasitic absorption of neutrons, the plutonium fuel can be effectively used in these reactors for energy production and burn-out of minor actinides.

Opportunity to use nuclear fuel on the basis of weapon-grade or power plutonium in reactor CANDU and simultaneously to burn-out minor actinides in separate channels is studied in the paper. Isotopic composition of power plutonium and minor actinides corresponds to unloading from pressure-water power reactors VVER operating on uranium fuel. The most favorable ratio between amount of plutonium and minor actinides loaded in the CANDU reactor is chosen. Fuel lifetime for various variants of a plutonium load is determined. It makes several months up to half-year. It is supposed that target assemblies with minor actinides are irradiated during long time. Continuous feed by new actinides and periodic (every 6 years) extraction of accumulated fission products is made. Productivity of a reactor from the view point of incineration of plutonium and minor actinides is determined. Opportunities of repeated use of plutonium are considered. The radiation characteristics - the activity and radiotoxicity of fuel assemblies are calculated.

2. FUEL ON BASE OF POWER PLUTONIUM

The basic data on reactor design are taken from [1]. Fuel assemblies contain 37 pin fuel elements in zirconium cladding surrounded by zirconium tubes. A diameter of pins with cladding is 6.75 mm. Target assemblies have the same structure with minor actinides in pins. Fuel and target assemblies are located in square lattice with pitch 15 cm. This value is about twice less in comparison with usual pitch of a lattices in CANDU reactor. When a fuel is natural uranium then large pitch of a lattice is required. At plutonium use, a pitch can be reduced. The minimal pitch is determined by a design of the refueling machine. A coolant and moderator is heavy water. The ratio of number of fuel assemblies to target assemblies n(fuel)/n(target) varied from 3:1 up to 1:1. Total number of assemblies is 1386. Height of an active core is 594 cm. Heat power of a reactor is 1930 MW.

Isotopic composition of power plutonium loaded in fuel assemblies: Pu-238 - 1.7 %, Pu-239 - 58.1 %, Pu-240 - 22.3 %, Pu-241 - 12.2 %, Pu-242 - 5.7 %. Two variants of plutonium load per 1 cm^3 of a pin are considered: P = 0.05 and 0.1 $gram/cm^3$. More highest values P result in undesirable growth of blocking effect and in reduction of a neutron flux.

For calculation of reaction rates and neutron balance in elementary cell containing fuel or target assembly, 37-pin assembly was presented as coaxial 4-ring assembly with preservation of volumes of all fuel and structural materials. The kinetics of nuclides conversion was calculated in homogeneous approximation. The fuel lifetime was determined with allowance for leakage of neutrons from reactor. It makes 0.24 years at average neutron flux density $1.3 \cdot 10^{14}$ $neutr/cm^2s$ for P = 0.05 $gram/cm^3$ and 0.53 years at $6.5 \cdot 10^{13}$ $neutr/cm^2s$ for P = 0.1 $gram/cm^3$

In table 1, some integrated characteristics for two values P = 0.05 and 0.1 $gram/cm^3$ and three values n(fuel)/n(target) - 3:1, 2:1, 1:1 are submitted. T is fuel lifetime. Φ - average neutron flux density. G is sum plutonium consumption over 60 years of operation. N(Pu) - number of VVER reactors giving plutonium for CANDU reactor (one VVER reactor produces 740 kg of plutonium in 4 years). N(MA) - number of VVER reactors giving minor actinides for CANDU reactor. There is also amount of americium and curium produced in plutonium fuel of CANDU reactor in the table.

Table 1. Characteristics of CANDU reactor operation with power plutonium fuel

Characteristics	P, $gram/cm^3$					
	0.05			0.10		
n(fuel)/n(target)	3:1	2:1	1:1	3:1	2:1	1:1
Φ, 10^{13} $neutr/cm^2s$	12.7	12.7	12.7	6.5	6.5	6.5
T, year	0.24	0.24	0.24	0.53	0.53	0.53
G, ton	82.1	73.0	57.4	74.3	66.0	49.3
N(Pu)	7.4	6.6	4.9	6.7	5.9	4.4
N(MA)	19	14	7	27	22	14
Am, kg/year	5.9	5.2	3.9	8.9	7.9	5.9
Cm, kg/year	0.40	0.36	0.27	2.2	2.0	1.5

The analysis of submitted data shows that variants with n(fuel)/n(target) = 1:1 should be excluded from further consideration because of appreciable loss of reactors number N(MA). From other variants having close characteristics, variant with n(fuel)/n(target) = 3:1 and P = 0.05 gram per 1 cm^3 of a fuel pin is preferable. It ensures more deep burn-out of minor actinides.

In table 2, the radiation characteristics of loaded and unloaded over 60 years fuel assemblies are submitted. In loaded assemblies, they are determined by isotopes of plutonium, in unloaded assemblies - also by accumulated isotopes of americium and curium. Masses G_i, activities A_i, and radiotoxicities by water RT_i are presented. The radiotoxicities $RT_i = A_i/MPA_i$ were calculated with allowance for maximum permissible activity of nuclides in water MPA_i determined by the radiation safety standards [2]. A unit of a radiotoxicity by water is litter of water.

Table 2. Radiation characteristics of loaded and unloaded over 60 years fuel assemblies with power plutonium

Nuclid	Load			î Unload		
	G, kg	A, Curi	RT, litter	G, kg	A, Curi	RT, litter
Pu-238	1380	2.38+7	1.63+47	860	1.49+7	1.02+17
Pu-239	47560	2.95+6	2.19+16	13000	8.07+5	5.98+15
Pu-240	18330	4.16+6	3.08+16	21550	4.89+6	3.62+16
Pu-241	10080	1.04+9	1.42+17	5460	4.86+8	6.66+16
Pu-242	4730	1.86+4	1.33+14	7310	2.87+4	2.05+14
Total Pu	82080	1.07+9	3.58+17	48180	5.01+8	2.11+17
Am-241	-	-	-	917	3.16+6	1.86+16
Am-242m	-	-	-	0.43	4.50+3	2.53+13
Am-243	-	-	-	285	5.73+4	3.37+14
Total Am	-	-	-	1202	3.22+6	1.90+16
Cm-242	-	-	-	0.02	6.5+4	2.50+13
Cm-243	-	-	-	0.02	1.0+3	5.58+12
Cm-244	-	-	-	17.8	1.44+6	6.86+15
Cm-245	-	-	-	0.15	25	2.21+11
Cm-246	-	-	-	0.016	5	4.31+10
Cm-247	-	-	-	1.8-5	1.7-6	-
Total Cm	-	-	-	18	1.5+6	6.89+15
Total	82080	1.07+9	2.22+13	49400	5.11+8	2.37+17

The data presented show that 1.5 % of plutonium is transformed to isotopes of americium and curium, about 40 % of plutonium turns to fission products. The radiotoxicity of plutonium falls 1.7 time over a fuel lifetime, however the total radiotoxicity in result of an irradiation falls by 34 %, that is explained by accumulation of Am-241.

Unloaded from fuel assembly plutonium has the following isotopic composition: Pu-238 - 0.2 %, Pu-239 - 27 %, Pu-240 - 45 %, Pu-241 - 13 %, Pu-242 - 15 %. It has rather low multiplicating properties and cannot be used repeatedly as fuel. But it could be irradiated in targets.

3. FUEL ON BASE OF WEAPON-GRADE PLUTONIUM

Weapon-grade plutonium is practically pure Pu-239. One variant of plutonium load in 1 cm^3 of fuel pin is considered here: P = 0.035 $gram/cm^3$. The reduced load of weapon-grade plutonium in comparison with load of power plutonium P = 0.05 $gram/cm^3$ is chosen to maintain the same flux density as for power plutonium. The ratio of fuel assemblies to target assemblies n(fuel)/n(target) is 3: 1.

The mass of required weapon-grade plutonium makes 70 % of power plutonium, that is 58 ton for 60 years of operation. Minor actinides from 19 VVER reactors are transmuted.

In table 3, radiation characteristics of loaded and unloaded in 60 years fuel assemblies such as in table 2 are submitted.

Table 3. Radiation characteristics of loaded and unloaded over 60 years fuel assemblies with weapon-grade plutonium

Nuclid	Load			Unload		
	G, kg	A, Curi	RT, litter	G, kg	A, Curi	RT, litter
Pu-238	-	-	-	.04	725	4.97+12
Pu-239	54540	3.39+6	2.51+16	9707	6.03+5	4.85+15
Pu-240	2584	5.87+5	4.35+15	12190	2.77+6	2.05+16
Pu-241	287	2.96+7	4.05+15	1972	2.04+8	2.79+16
Pu-242	-	-	-	681.5	2.68+3	1.91+13
Total Pu	57411	3.36+7	3.35+16	24551	2.07+8	5.29+16
Am-241	-	-	-	10.8	3.69+4	2.17+14
Am-242m	-	-	-	0.1	1.49+3	8.37+12
Am-243	-	-	-	14.1	2.83+3	1.66+13
Total Am	-	-	-	25	4.12+4	2.42+14
Cm-242	-	-	-	0.005	1.70+4	6.54+12
Cm-243	-	-	-	0.003	158	9.29+11
Cm-244	-	-	-	0.6	4.72+4	2.25+14
Cm-245	-	-	-	0.003	0.61	5.40+9
Total Cm	-	-	-	15	6.42+4	2.32+14
Total	57411	3.36+7	2.08+12	24591	2.08+8	5.34+16

The data submitted show that 57 % of plutonium turns to fission products. Production of americium and curium is negligible. The radiotoxicity of plutonium grows 1.6 times because of accumulation of Pu-241.

Burnt-up weapon-grade plutonium unloaded from fuel assemblies after extraction of fission products and minor actinides has the following isotopic composition: Pu-239 - 40 %, Pu-240 - 49 %, Pu-241 - 8 %, Pu-242 - 3 %. Multiplicating property of this plutonium is lower than that of power plutonium but it is higher than unloaded power plutonium. Calculation of the neutron balance shows that unloaded weapon-grade plutonium can be used repeatedly as a fuel. In this case the amount of incinerated minor actinides in target assemblies should be reduced by 40 % in comparison with operation on base of fresh weapon-grade plutonium.

4. CONCLUSION

The research performed permits to make a conclusion about reality of CANDU reactor modes with fuel on basis of power or weapon-grade plutonium with location minor actinides in separate assemblies for their burn-out. To maintain effective heat removal, the diameter of pins in fuel assembly is accepted about 6 mm. At the expense of intensive fission of actinides, heat power in target assemblies is close to that of in fuel assemblies. Multiplicating property of target assemblies is about 1.5 times lower than that of fuel assemblies. The most preferable is the ratio between number of fuel and target assemblies 3:1 and plutonium load ensuring a neutron flux density of the order of 10^{14} neutr/cm^{2}s. It makes 0.05 gram of power plutonium or 0.035 gram of weapon-grade plutonium per 1 cm^3 of a fuel pin.

The fuel lifetime makes 0.24 years. 40 % of a power plutonium break into fission products over a lifetime.

The CANDU reactor could consume a power plutonium extracted from spent fuel of 7 VVER reactors and minor actinides from 19 VVER reactors (VVER is like PWR reactor by thermal power 3.2 GW).

The radiotoxicity of unloaded plutonium with accumulated actinides is 34 % lower than loaded power plutonium. The rather small decrease of radiotoxicity is explained by production of Am-241. The radiotoxicity of actinides irradiating in target assemblies is also reduced insignificantly as Am-241 essentially burns up but Cm-244 is accumulated at irradiation

When weapon-grade plutonium is used in fuel assemblies instead of power plutonium, its annual consumption is 30 % lower. Over a lifetime, 57 % of weapon-grade plutonium. breaks into fission products. The radiotoxicity of plutonium at irradiation grows 1.6 times because of accumulation of Pu-241.

Weapon-grade plutonium used in CANDU reactor can be used repeatedly. The amount of loaded actinides in target assemblies decreases by 40 %. The once used power plutonium could be loaded in target assemblies.

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