CALCULATION AND EXPERIMENTAL STUDIES ON MINOR ACTINIDES REACTOR TRANSMUTATION

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Abstract

Brief survey of the activity being carried out in IPPE in the field of calculation and experimental studies on MA transmutation is given. Results of recent developments on calculation code and cross-sections supply for MA transmutation works are presented. Calculation investigations on incineration of Am and Cm with shares of rare-earth elements in BOR-60 reactor are described for the forthcoming experiment. Comparative analysis on MA transmutation in fast and thermal reactors is given.

Introduction

Nowadays in Russia the program on transmutation of MA and long-lived fission products is developed only on each next year (due to lack of financing supplied by the Government for development of the whole nuclear power). Therefore, the schedule of works is corrected each year to provide fulfilment of the most valuable tasks. Nevertheless, the large volume of works has been done, in particular, thanks to the assistance of our foreign colleagues supporting realisation of our plans in the joint projects.

General review

As it was in previous years, the main directions of the activity in the field of MA and long-slived fission products transmutation in Russia are as follows:

1. Modernisation of nuclear constants data and software codes needed for calculation studies on MA and long-lived fission products transmutation.
2. Development and calculation-experimental grounding of the specialised fast reactor cores for incineration of Plutonium, MA and some fission products.
3. Investigation of closed fuel cycle with Plutonium and MA recycling.
4. Carrying out integral experiments on the critical assemblies simulating power reactors to correct the data on MA nuclear reactions cross-sections.
5. Preparation and carrying out experiments in power reactors on MA sample fuel irradiation.

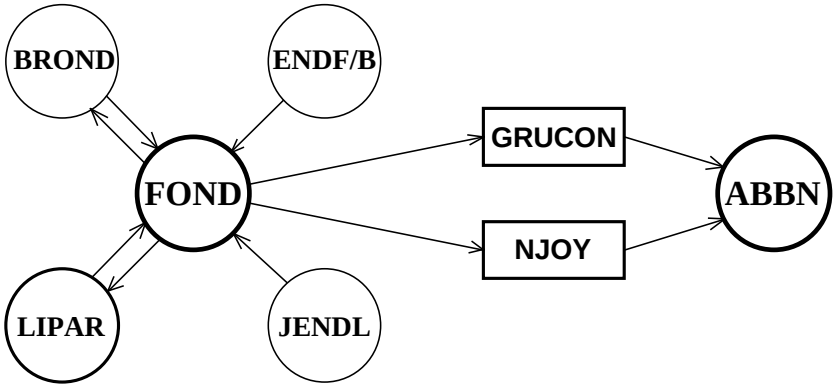
Nuclear constants data and software for minor actinides transmutation calculations

Calculations of MA transmutation in fast reactors base on the TRIGEX [1] code performing diffusion 3D reactor calculation in HEX-Z geometry with ABBN-93 [2] nuclear constants. Software system CONSYST2/ABBN (see DLC-182/ABBN-90 in the RSICC Data Library Collection) provides ABBN-93 constants binding with TRIGEX code by means of PRECON1 subroutine. TRIGEX code performs calculations of fuel composition variation with burn-up using CARE [3] code.

Calculations of MA transmutation in thermal reactors at present are carried out by means of Monte-Carlo method codes MCU-RFFI (RRC KI, Moscow), KENO (being the part of the American calculation complex SCALE 4.3 [4]) and MCNP. For cells and subassemblies calculations, TBC-M code (RRC KI, Moscow) is used. Constants preparation for the KENO code is realised through WIMS-ABBN code as well as by CONSYST code being the part of the CONSYST2/ABBN nuclear constant supply system with ABBN-93 nuclear constants. Constants binding is performed using ANISN-like formats. CONSYST2/ABBN system application supports carrying out of calculations performed by both precise codes implementing Monte-Carlo method, and kinetic transport reactor calculations in DSN-PN approximation, such as ANISN, TWODANT and DOT.

ABBN-93 constant system is the most modern version of the ABBN group constants. It is created on the base of evaluated neutron data library files FOND-2 [5], which integrates files from Russian library BROND-2, foreign libraries ENDF/B-VI, JENDL-3, and some other sources (Figure 1).

Figure 1. **ABBN-93 constact system**



Calculation of the nuclide transformation chains during MA transmutation calculations is performed by both CARE code and ORIGEN code which is the component of the American calculation complex SCALE 4.3.

Calculating the nuclide transformation, resonance shielding of neutron cross-sections is taken into account for the isotopes being members of the media composition (from TRIGEX code). For all the rest isotopes needed for transformation chains calculation, cross-section data is being taken from the ABBN external libraries of the group neutron cross-sections: FP - library of fission product nuclei, ACT - library of actinides. Library of masses is also used additionally.

Multi-group library of fission product nuclei FP contains data on the radiation capture cross-sections for 169 nuclides:

^{75}As, ^{76}Ge, ^{77,78,79,80,82}Se, ^{81,82}Br, ^{82,83,84,85,86}Kr, ^{85,86,87}Rb, ^{86,88,89,90}Sr, ^{89,90,91}Y, ^{90,91,92,93,94,95,96}Zr, ^{95}Nb, ^{95,96,97,98,99,100}Mo, ^{99}Tc, ^{100,101,102,103,104,106}Ru, ^{103,105}Rh, ^{104,105,106,107,108,110}Pd, ^{109,110m,111}Ag, ^{100,110,112,113,114,115m,115n,116}Cd, ^{115}In, ^{115,116,117,118,119,120,121,122,123,124,125,126}Sn, ^{121,122,123,124,125,126,127}Sb, ^{122,123,124,125,126,127m,128,129m,130,132}Te, ^{127,129,130,131}I, ^{128,129,129m,130,131,131m,132,133,133m,134,135,136}Xe, ^{133,134,135,136,137}Cs, ^{134,135,136,137,138,139,140}Ba, ^{139,140}La, ^{140,141,142,143,144}Ce, ^{141,143}Pr, ^{142,143,144,145,146,147,148,150}Nd, ^{147,148,148m,149}Pm, ^{147,148,149,150,151,152,153,154}Sm, ^{153,154,155,156}Eu, ^{154,155,156,157,158,160}Gd, ^{159,160,161}Tb, ^{160,161,162,163,164}Dy.

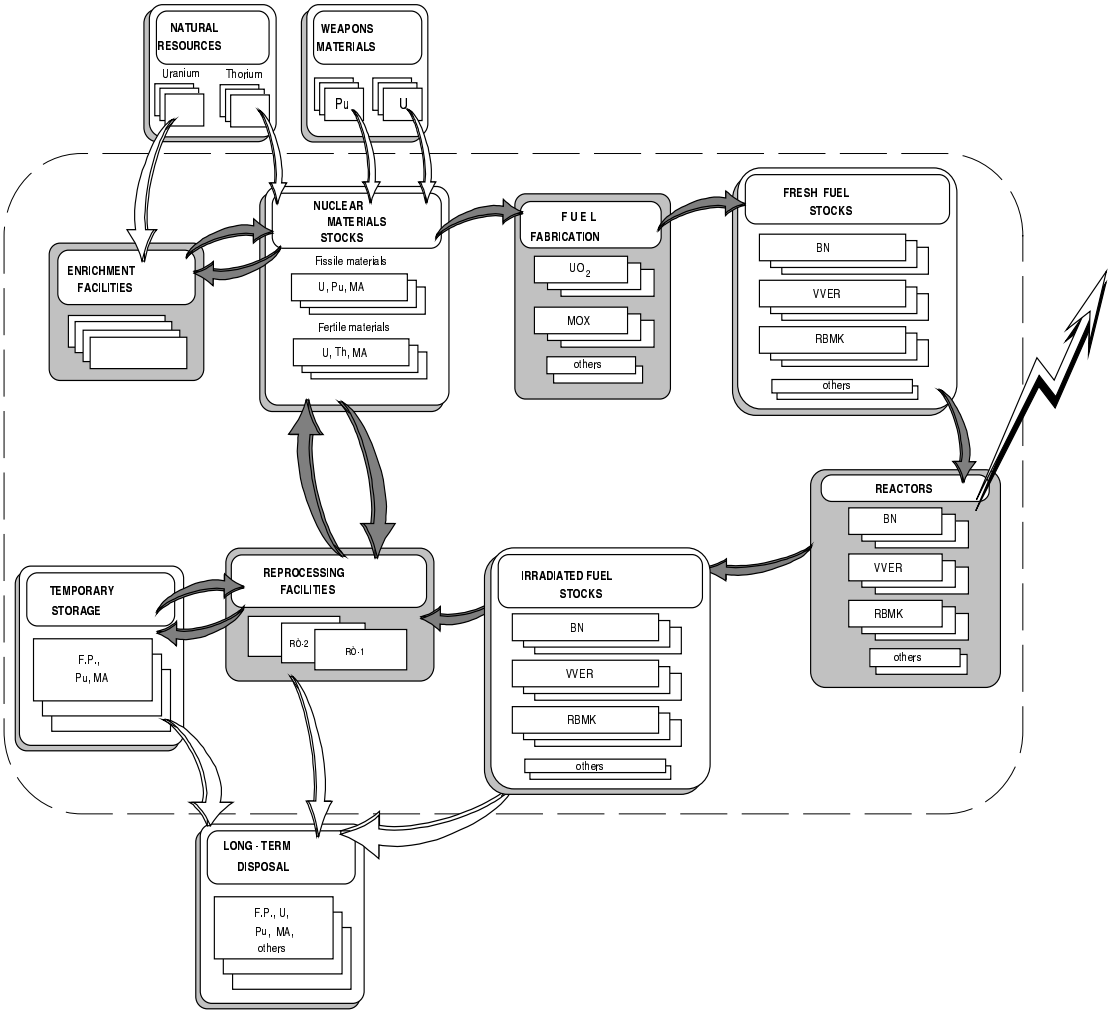
Actinides library ACT contains group cross-sections of (n,2n), (n,3n), (n,γ) and (n,f) reactions for the following nuclei:

^{223,224,225,226}Ra, ^{225,226,227}Ac, ^{227,228,229,230,232,233,234}Th, ^{231,232,233}Pa, ^{232,233,234,235,236,237,238}U, ^{236,237,238,239}Np, ^{236,238,239,240,241,242}Pu, ^{241,242,242m,243,244,244m}Am, ^{241,242,243,244,245,246,247,248,249,250}Cm, ^{249,250}Bk, ^{249,250,251,252,254}Cf, ^{254,255}Es, ^{255}Fm.

For the nuclear fuel cycle simulation, CANFU [6] code is developed. The main purpose of the CANFU code is concluded in automation of physical, radiation and ecological characteristics of the nuclear fuel cycle. To perform the above mentioned task, the code is designed and developed as a kind of meccano, allowing to create and arrange various schemes of a fuel cycle using predefined fuel cycle components (reactors, fuel management facilities, storages, etc.) with standard or user-defined parameters. The code takes use of the program modules for physical characteristics calculation (TRIGEX, CARE) and nuclear data library (ABBN-93). Figure 3 represents sample scheme of nuclear fuel cycle being simulated in the CANFU code.

Application of CANFU code allows to perform calculation studies of closed fuel cycles with both Plutonium and minor actinides recycling.

Figure 2. **Sample fuel cycle model implemented in CANFU code**



Development of specialised fast reactors for Plutonium and minor actinides incineration

Fast reactors are capable not only to effectively incinerate Plutonium of any isotopic composition, they can be used also for efficient utilisation of minor actinides. The main physical problem being arisen is connected with the degradation of void reactivity effect. However, cores with increased fuel enrichment have some resource in SVRE value, allowing to introduce (homogeneously) up to 5-7% of minor actinides in fuel. Transmutation efficiency in this case can reach up to 50 kg annually.

As the most effective incinerators of both Plutonium and minor actinides, cores should be considered with the fuel not containing ^{238}U. The main tasks to be solved on the way of such cores creation are increase of safety due to Doppler-effect growth and fuel reloading schedule optimisation. Numerous investigations carried out recently in IPPE permitted to optimise such a core using new

fuel material (PuO_2+MgO+Fe with MA addition up to 15% vol.), which has been also certified. Utilisation of such kind of fuel allows to increase MA incineration efficiency up to 100-150 kg annually. Table 1 presents calculated values of MA incineration efficiency [7] for the various cores of fast reactor with capacity 800 MWt.

Table 1. **Comparison of calculated values of MA incineration efficiency for the various cores**

	Cores with the MOX fuel of increased enrichment			Cores with the fuel not containing ^{238}U		
MA Location	Homogeneously in the fuel	Heterogeneously in the core	In the radial blanket	Homogeneously in the fuel		In the radial blanket
Kind of fuel with MA	MOX	MOX	MAO_2 in inert matrix	PuO_2 +MAO_2	$^{235}UO_2$+MAO_2	MAO_2 in inert matrix
Efficiency of minor actinides incineration, kg/year	**50**	**50**	**50**	**120**	**260**	**80**

Further increase of MA incineration efficiency is possible with substitution of Plutonium as a fuel material by ^{235}U.

Experimental studies

Estimations of requirements have been performed in SSC RF IPPE on actinides neutron cross-sections correction for the calculation of recycling parameters for the purpose of transmutation. The estimates show that with the increase of actinides share in the core, contribution to the whole K_{eff} uncertainty is stipulated for the uncertainties in fission and elastic scattering cross-sections of ^{237}Np, ^{241}Am, ^{243}Am isotopes. With the share of actinides in fuel equal to 25%, the K_{eff} uncertainty makes up 2.6%, and due to correction of actinides neutron cross-sections it can be reduced up to 1.5%. In this connection, carrying out of experiments on BFS facilities and power reactors is being continued.

During several years in SSC RF IPPE experimental program is being intensively realised aimed at detailisation of data on MA in integral experiments on BFS critical facilities. Experiments of various types have been carried out:

- measurement of fission cross-section ratios of MA isotopes (there are fission chambers with layers of Am, Np, Cm, Pu, Th, U on the stands) to the standard ones (^{239}Pu or ^{235}U) on the series of fast critical assemblies with various neutron spectrums;
- measurement of central reactivity worths of MA sample, which gives an opportunity to obtain data on capture cross-sections taking into account previously mentioned information;
- estimation of the influence of introduction of considerable quantity of ^{237}Np in fuel on the main neutron-physical characteristics of cores (void effect of coolant, Doppler-effect, spectral indices, etc.).

To carry out the experiments with introduction of Neptunium into the core, 250 BFS pellets (about 11 kg) with Neptunium dioxide have been produced. Then the blocks with Neptunium dioxide have been placed in central parts of the cores of Superphenix and CAPRA type, simulating 13% and 6.5% Neptunium content in fuel.

According to the program being planned of the MOX fuel studying in light-water reactors on SUPR facility being under construction, investigations are intended on MA burning in thermal spectra as well.

The series of experiments is being completed on irradiation of actinide samples in BN-350 reactor. At present the samples of ^{238}Pu, ^{237}Np, ^{241}Am, ^{243}Am irradiated in 80-90th are delivered to the hot cell in IPPE.

Preparation of works in on the way on irradiation of experimental AMOX fuel elements in BOR-60 reactor. Calculations of isotopic composition evolution and decay heat of AMOX fuel have been performed as a part of works on the branch program “RECYCLE”.

The calculations took into account the following assumptions on the composition of AMOX fuel for BOR-60 reactor:

1. the basis of the fuel consists of Uranium with the enrichment 75%.
2. Americium-Curium fraction obtained during reprocessing of VVER-1000 reactor spent fuel with starting enrichment 4.4% and burnup 40 GWt d/t is then added in proportion 20% or 50% mass.
3. this fraction, in its turn, contains impurity of 10% rare-earth elements not separated from the Americium and Curium during chemical reprocessing.
4. The chemical form of actinides in fresh fuel has been adopted as follows: Uranium - UO_2, Americium and Curium - Am_2O_3 and Cm_2O_3.

Calculated values of the isotopic composition of source Americium-Curium mixture is given in Table 2.

Table 2. **Isotopic composition of Am-Cm fraction in VVER-1000 reactor spent fuel with 10 year cooling.**

Isotope	Content, %
^{241}Am	83.746
^{242}Am	1.22E-06
^{242m}Am	0.101
^{243}Am	13.163
^{242}Cm	2.46E-04
^{243}Cm	0.029
^{244}Cm	2.731
^{245}Cm	0.227
^{246}Cm	3.59E-03
^{247}Cm	6.31E-05
^{248}Cm	6.25E-06

Some important numerical results are presented in Table 3.

Table 3. **The most important characteristics of the transmutation process of Americium-Curium mixture being a part of AMOX fuel of BOR-60 reactor.**

Parameter	20% Am-Cm	50% Am-Cm
Transmutation of Americium, g/t h.a.	32500	81300
Build-up of Curium, g/t h.a.	4448	10940
Build-up of Plutonium, g/t h.a.	14700	28370
"Incineration of Americium" (mass of transmutated Americium excluding sum mass of Plutonium and Curium built-up), g/t h.a.	13352	41630
Build-up of helium, g/t h.a.	151	378
Specific Am transmutation, g/GWt hour (t)	10500	30100
Specific Am incineration, g/GWt hour (t)	4300	15400

In course of transmutation of Americium added to the BOR-60 conventional fuel during the refuelling interval about 21% ^{241}Am and 13% ^{243}Am is transmuted; accumulation of Curium isotopes in spent fuel increases in thousand times as compared to the initial conventional fuel; accumulation of ^{238}Pu and ^{242}Pu increases in dozen and hundred times; mass of helium released under irradiation increases in hundred times, during the storing period mass of ^{237}Np raises in dozen times as compared to MOX fuel.

The main feature of spent AMOX fuel is extremely high decay energy release and intensity of neutron emission, in comparison with Uranium and even MOX fuel. The absolute value of the sum decay energy release remains at the level exceeding 1 MWt/t during approximately 1 month. This circumstance is of importance with respect to one of the main problems of reactor safety - cooldown at the emergency shutdown with heat removal cessation - and should be studied in detail.

Some results on actinide balance in various scenarios of Pu and MA Recycling

The present chapter presents calculation studies of physical characteristics of fuel cycles based on thermal reactors exclusively as well as of fuel cycles with thermal reactors and specialised fast reactors with high enrichment on Plutonium. Material indices on Plutonium consumption and radioactive waste accumulation have been calculated. Equilibrium isotopic compositions of fuel being reached in closed systems of reactors have been estimated. Basing on the same power production level, waste radiotoxicity values are given for the scenarios considering different types of reactors. The main goal of the work was to show how the accumulation of MA changes in the wastes of fuel cycle with introduction of specialised fast reactor.

Description of the considered scenarios

Three different scenarios of fuel cycle have been considered:

1. Open fuel cycle with VVER-1000 type reactors operating on civil Plutonium.
2. Closed fuel cycle with Pu recycling in VVER-1000 reactor.
3. Closed fuel cycle with Pu recycling in VVER-1000 reactor and MA recycling in specialised fast reactor FR. with capacity 1300 MWt for MA incineration.

Calculations of long-lived radioactive wastes accumulation have been performed under condition of equal power production level (100 GWt(e) year) for each scenario.

In the open fuel cycle at the beginning of the irradiation stage fresh fuel had the same isotopic composition, and after irradiation all the spent fuel has been transported to waste storing facility. During the period of reactor operation, isotopic composition of heavy radioactive nuclei in the storage change due to radioactive decay and receipt of next portion of spent fuel from cycle to cycle.

In the calculation model of the closed fuel cycle, irradiated fuel of VVER-1000 reactor has been reprocessed extracting Plutonium and Uranium, which has been used for the production of fuel for the next cycle. The wastes in this case were formed by fission products and MA from the spent fuel. The new fuel has been enriched by the initial civil Plutonium, diluted by depleted Uranium and sent again to the irradiation in reactor. For the FR reactor, all heavy isotopes have been extracted from the spent fuel, MA from VVER spent fuel have been added and after enrichment this new fuel has been sent to the next cycle of irradiation. The extraction extent for all heavy isotopes during chemical reprocessing has been adopted as 0.999.

Isotopic composition of the feed Plutonium for all scenarios was equal to the isotopic composition of civil Plutonium (%):

$$^{238}Pu/\ ^{239}Pu/\ ^{240}Pu/\ ^{241}Pu/\ ^{242}Pu = 0.9/61.0/22.0/12.0/4.1$$

The main characteristics of VVER-1000 and FR reactors operating in open fuel cycle are presented in Table 4.

Table 4 **The main characteristics of VVER-1000 and FR reactors**

Reactor Parameter	VVER-1000	FR
Electric power (MWt)	1000	1300
Type of fuel	MOX	MOX
Number of subassemblies in core	163	349
Enrichment, %	6.6	25.3/29.34
Burnup, GWt days/t	40	70
Irradiation interval, eff.days	876	1200
Number of reloads	3	3
Load factor	0.8	0.8
Start loading, t (h.a.)	65.25	34.51
Loading, t/year(h.a.)	21.75	8.4
Pu loading, kg/year	1435.5	2277.5
Pu discharge, kg/year	1040.5	1840.6
Pu balance, kg/year	-395	432
Pu isotopic composition in spent fuel (%)		
^{238}Pu	1.84	0.94
^{239}Pu	42.55	56.01
^{240}Pu	28.05	29.79
^{241}Pu	18.76	7.90
^{242}Pu	8.79	5.36

Calculation results

Figures 3, 4, 5 illustrate the balance of fuel nuclides for 3 different scenarios under the same power production rate 100 GWt·year.

Figure 3. **The balance of fuel nuclides in open fuel cycle (Scenario 1)**

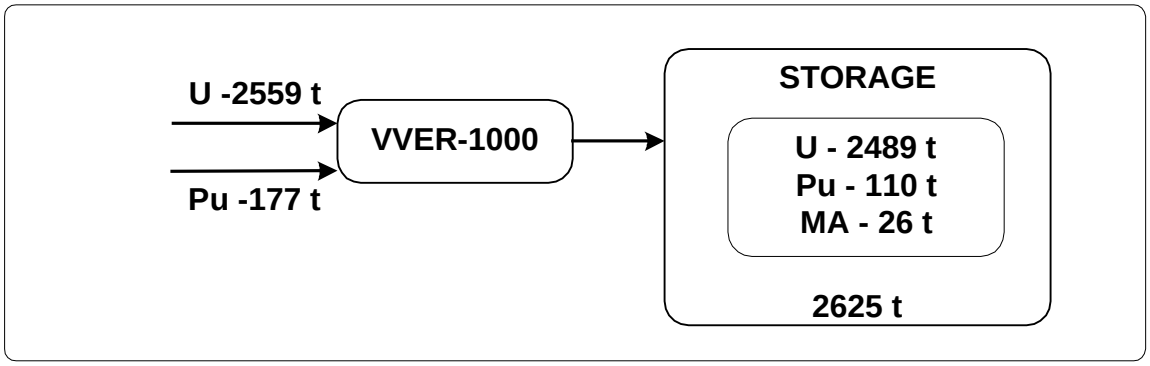


Figure 4. **The balance of fuel nuclides in closed fuel cycle with U and Pu recycling in VVER reactor (Scenario 2)**

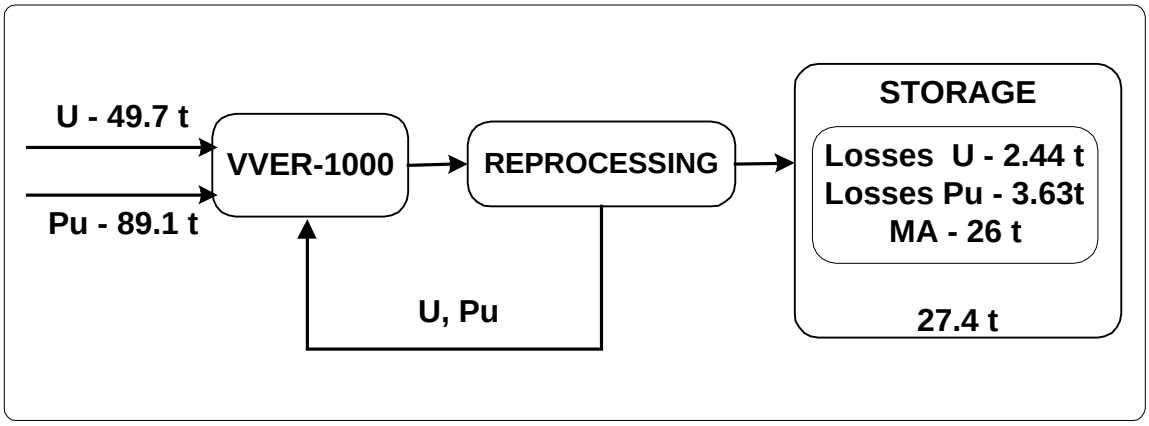
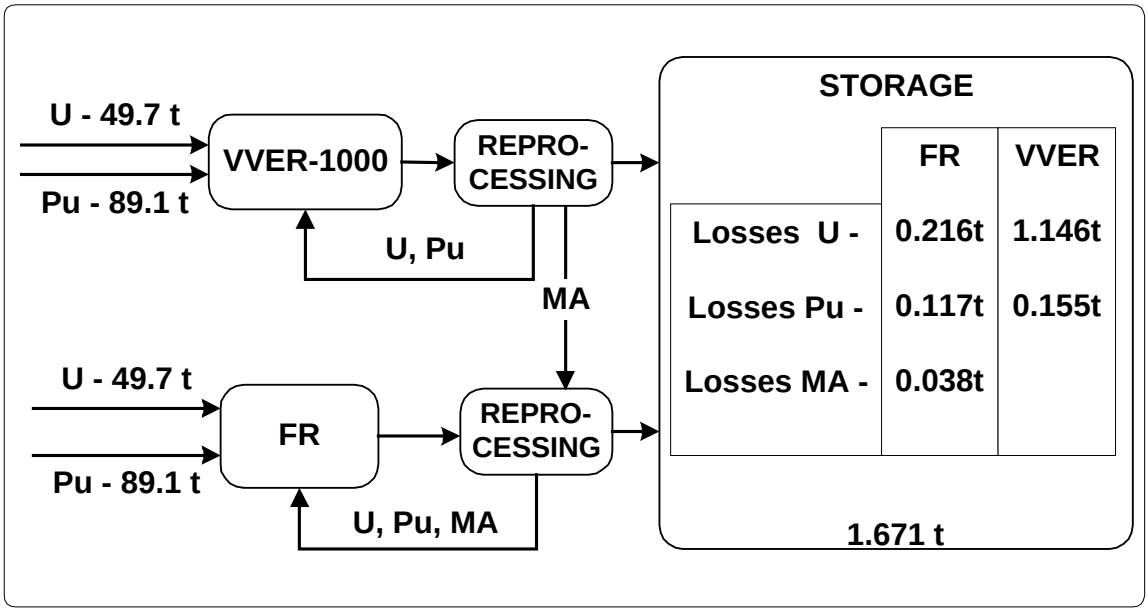


Figure 5. **The balance of fuel nuclides in closed fuel cycle with U and Pu recycling in VVER reactor and U, Pu, MA recycling in FR reactor (Scenario 3)**



In course of recycling equilibrium compositions of loaded and discharged fuel are set. Tables 5 and 6 show the compositions of loaded and discharged fuel for VVER-1000 and FR reactors. It can be seen from the tables that in the system consisting of VVER-FR reactors 4 kg of Np, 207 kg of Am and 47 kg of Cm are transmuted per year.

Table 5. **Equilibrium compositions of loaded and discharged fuel set in VVER-1000 reactor in course of Pu recycling**

	Loading, kg/cycle	Discharge, kg/cycle	Balance, kg/cycle	Balance, kg/year
^{235}U	31	28	2	1
^{238}U	57941	56753	1188	495
^{237}Np	0	10	-10	-4
^{238}Pu	254	235	19	8
^{239}Pu	2884	1561	1323	551
^{240}Pu	1805	1329	477	199
^{241}Pu	1147	887	260	108
^{242}Pu	1121	1033	88	37
^{241}Am	0	230	-230	-96
^{242m}Am	0	3	-3	-1
^{243}Am	0	267	-267	-111
^{242}Cm	0	0	0	0
^{243}Cm	0	1	-1	0
^{244}Cm	0	103	-103	-43
^{245}Cm	0	10	-10	-4
^{246}Cm	0	0	0	0

Table 6. **Equilibrium compositions of loaded and discharged fuel set in FR reactor in course of Pu and MA recycling**

	Loading, kg/cycle	Discharge, kg/cycle	Balance, kg/cycle	Balance, kg/year
^{235}U	42	39	3	1
^{238}U	17108	15460	1647	501
^{237}Np	71	58	13	4
^{238}Pu	727	709	18	5
^{239}Pu	4722	3460	1262	384
^{240}Pu	5248	4795	453	138
^{241}Pu	982	732	250	76
^{242}Pu	1292	1207	84	26
^{241}Am	1183	870	313	96
^{242m}Am	61	57	3	1
^{243}Am	1510	1145	364	111
^{242}Cm	1	1	0	0
^{243}Cm	10	9	1	0
^{244}Cm	1059	918	140	43
^{245}Cm	219	205	14	4
^{246}Cm	100	100	0	0

For the Scenario 3, equilibrium compositions of Pu being loaded in VVER reactor and MA being loaded in FR reactor are of interest. Figure 6 indicates variation of Pu isotopic composition in the loading of VVER-1000 reactor. Fuel enrichment in this case increases from 6.6% up to 10%. Figure 7 illustrates variation of mass of actinides being loaded in FR reactor in recycling of MA built-up by both FR and VVER reactors. Fuel enrichment of FR reactor in equilibrium increases up to 34% and 40% for inner and outer cores accordingly.

Figure 6. **Variation of Pu isotopic composition in the loading in course of its recycling in VVER-1000 reactor**

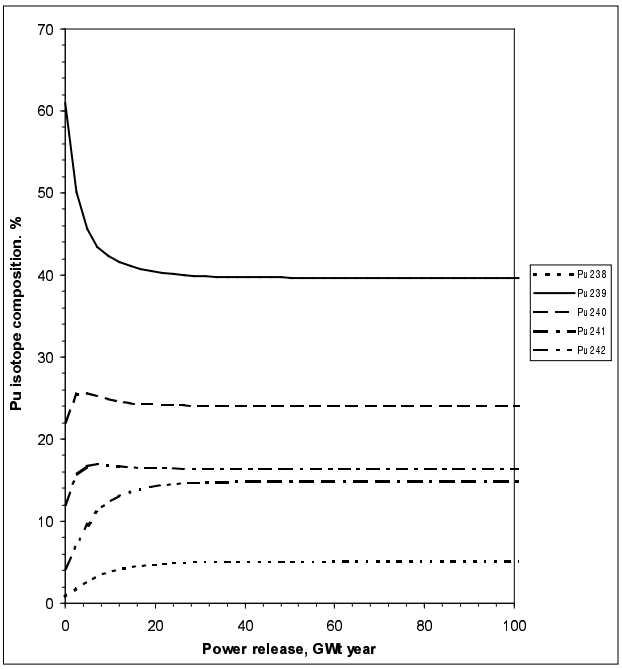


Figure 7. **Variation of MA content in the loading in course of its recycling in FR reactor**

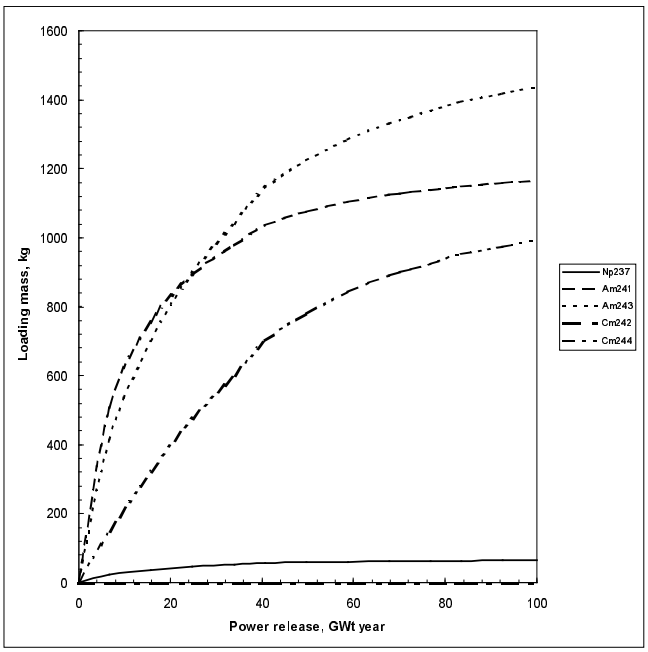
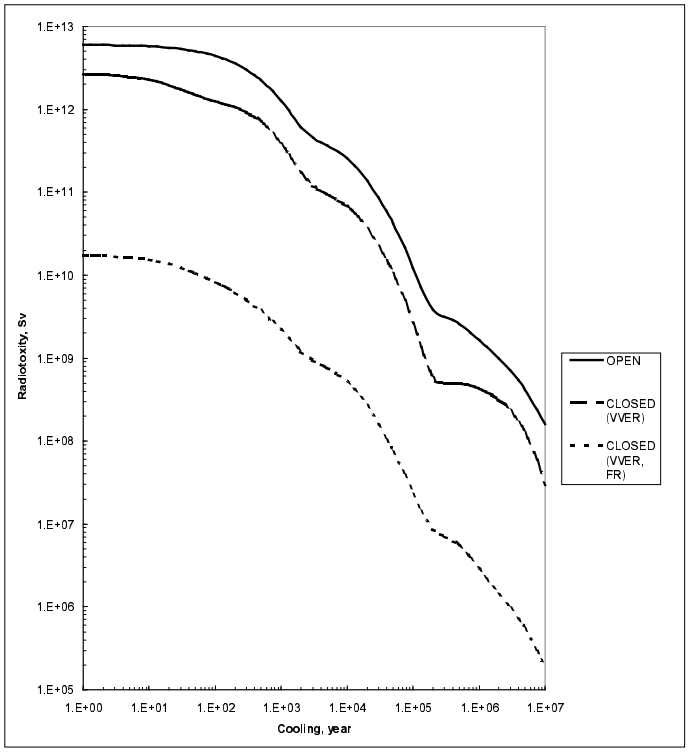


Figure 8 presents potential sources of radiotoxicity of long-lived nuclides formed in storage for three scenarios under consideration. The curves are obtained by means of simple multiplication of activity of radionuclides formed in storage during cooling by their hazard factor. It follows from the figure that closure of fuel cycle only on Plutonium and Uranium does not affect dramatically on the change of potential radiotoxicity. Decrease of radiotoxicity in this case constitutes merely 2-3 times. Recycling of all actinides, as it can be seen from the curve corresponding to Scenario 3, decreases radiotoxicity in 100-150 times.

Figure 8. **The sources of potential radiotoxicity in the waste storage for three considered scenarios**



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