TRANSMUTATION OF TRANSURANICS: NEUTRONICS, ACTINIDES BALANCE, SAFETY AND FUEL PROVISION ASPECTS

N.S.Rabotnov

State Research Center -A.I.Leipunskiy Institute of Physics and Power Engineering,
249020, Bondarenko Sq.1, Obninsk, Kaluga Region, Russia

Abstract

A review is given of the selected works on the subject in Russia with the emphasis on the latest results published or presented after GLOBAL95 Conference. The main attention is concentrated on following problems: evolution of the actinides isotopic mixtures during multiple recycling in thermal and fast neutron systems operating separately or in combinations; compatibility of the transmutation with the breeding of secondary fuel; influence of plutonium composition, including weapon-grade, on the recycling process; nuclear safety related problems of actinides recycling; nuclear data for transmutation; criteria of assessment of radiological and environmental consequences of actinides transmutation; excess neutrons available for transmutation in accelerator-driven facilities; activation of neutron producing accelerator targets.

Introduction

During last three years some new attitudes toward nuclear fuel cycle were developing fast as the new series of GLOBAL-93 and GLOBAL-95 [1,2] conferences clearly demonstrated. These new trends included: increased attention to transmutation of minor actinides and possibly of some fission products; fast disposition of the excess separated plutonium, especially weapon grade; extensive analyses of the transmutation and plutonium utilization potential of accelerator-driven facilities. These problems are closely interconnected and in Russia the interest in P&T research is mainly stimulated by the importance of plutonium problem.

In 1995 the IAEA has undertaken efforts to produce Status Report on Transmutation in Non-OECD Countries as an IAEA Technical Document. The text was compiled, corrected and approved last September and now is about to be published. It contains rather comprehensive general review of the progress in P&T covering the activity of about dozen largest Russian institutions during last few years. So present paper is focused on the results obtained quite recently and best known to the author which means mainly A.I.Leipunskiy IPPE publications. At first short general review is given and then a few points are outlined in more details.

The strategy of P&T research in Russia is determined by following considerations. Due to obvious slowing down of the development of nuclear power industry the balance of research efforts between evolutionary and innovative approaches shifted to the latter. In the domain of basically evolutionary designs, which means critical fission rectors with solid fuel and liquid coolant, an ideal facility included in closed fuel cycle with multiple recycling, aimed at as complete utilization of natural resources as possible, should have:

- all principal reactivity coefficients of proper signs and values;
- fuel burn-up not less than 100 GW days/thm;
- breeding ratio not less than 1.3;
- ability to burn, in equilibrium regime, all Np and Am of its own and, if needed, some quantity of MA from outside;
- proliferation resistant fuel cycle;
- transparent ability to withstand design basis accidents without catastrophic consequences;
- good economy.

If these requirements are conflicting or some of them just may not be realized then we should look into a broader domain of designs.

1. General review

Transmutation related research at IPPE connected with the topic of present paper consists of following interacting sectors:

- analysis of the status and prospects of closed fuel cycles technologies aimed at transmutation of minor actinides and probably selected fission products;
- extensive calculations of the properties and parameters of the plutonium fueled fast reactor cores used as actinide burners;
- experiments on critical facilities, imitating the cores of large fast rectors, coordinated with the calculations and analysis programs and used to verify both input data and calculation methods.
- measurements and evaluation of basic microscopic nuclear data;
- designing, related experiments and calculations on the neutron producing accelerator targets of accelerator-driven transmutation facilities.

Present paper deals mainly with the reactor side of the transmutation problem because in ADTT research in Russia the Institute of Theoretical and Experimental Physics (Moscow) is main driving and coordinating force as demonstrated by the latest ADTT Conference in Kalmar [3]. But some important work is done in IPPE also and those results are discussed briefly.

1.1 Closed nuclear fuel cycle as the way of developing long term sustainable nuclear power industry.

Mining of uranium does not satisfy present demand and situation will get worse in coming 15 years with fuel fabrication relying significantly on the stored uranium reserves. Spent fuel reprocessing rate is only a fraction of the discharge rate and does not prevent piling up of large masses of SNF consisting of almost untouched natural uranium with uncertain future. So a time will come, most probably in the second half of the next century, when nuclear power industry will face following situation in raw materials: millions of tons of depleted uranium and a limited quantity of assorted plutonium and minor actinides, mostly in the spent fuel

form. Let's call the time "Day X". Sustainable level of nuclear power after the Day X, P(t>X), depends on the quantity $M_{Pu}(X)$ and quality of plutonium available determined by the global scenario of plutonium utilization which is combination of national scenarios, formed **today** and varying drastically. $M_{Pu}(X)=0$ means $P(t>X)=0$ i.e. the end of the nuclear power industry based on critical fission reactors because no system critical on depleted uranium only is possible. We made an attempt to estimate X and P(X) for Russian national nuclear power industry and to investigate the sensitivity of these values to some features and parameters of the selected scheme of the nuclear fuel cycle within different concrete scenarios [4]. Basic assumptions are:

- thermal and fast reactors are used in varying proportion;
- regenerated uranium is recycled both in thermal and fast reactors;
- plutonium is recycled in MOX fuel of fast reactors;
- the possibility of utilization of excess weapon-grade plutonium is considered.

The results indicate that:

- in the model adopted X is close to 2080;
- P(X) is considerably lower than the maximum value of mainly uranium nuclear power capacity reached earlier;
- weapon-grade plutonium is essential; if added to reactor-grade plutonium as 20 per cent admixture and used in fast reactors with breeding ratio 1.3 it may add up to 30% to P(X);
- BR=1.3 increases P(X) by some 70 per cent as compared to the case with BR=1.0.

Delay of the introduction of fast breeders into national nuclear power industry obviously decreases P(X) considerably. From the point of view of nuclear power industry development in the next century there is no plutonium surplus in Russia. The same well may be true in global scale if present trend to develop primarily net plutonium burners prevails (warnings of this kind may be found elsewhere, see, for example, [5]). The best way to deal with weapon-grade plutonium is to convert it to "spent fuel standard" in fast breeders with as good breeding ratio as possible and keep the spent nuclear fuel for later recycling, thus solving non-proliferation problem and using plutonium's energy producing potential.

Pu recycling and breeding may be combined with MA burning neutronically but safety should be studied further; already nearest generation of new reactors with life-time expectancy of at least 50 years should be designed flexible to meet the requirements of steadily growing plutonium breeding when necessary. Recycling of U in thermal reactors increases total electricity production during the considered period by 14 per cent, recycling of Pu in thermal reactors - by further 4 per cent only. Introduction of fast reactors with BR=1.0 brings 21 per cent increase which rises to 43 per cent at BR=1.3.

1.2 Recycling of plutonium and minor actinides: nuclear safety related consequences.

Plutonium and minor actinides may be recycled in both thermal and fast reactors. The efficiencies of these two options depend not only on the physics and technology of corresponding systems but on many varying and not always predictable external conditions like availability of competing energy sources, natural uranium prices, public acceptance, general national energy policies etc. But there is a set of much more definite safety related problems which should be considered first:

Core neutronics: worsening of Doppler and void reactivity effects, control rod worth, delayed neutrons parameters.

Power density distribution: sharper non-uniformities starting at the pellet level; strong and non-monotonous time-dependence; possible complications of loading and reloading schemes.

Accidents development: higher recriticality dangers in a case of melt down with high Pu content; worse kinetic parameters with resulting increase of the energy release during an excursion.

Proliferation issues: separated plutonium of practically any composition is most proliferation sensitive substance unlike plutonium in spent fuel.

All these aspects are closely investigated and some of the latest results are to be presented in a few papers at PHYSOR-96 Conference, September 16-20, 1996, Mito,Japan.[6-8].

Experience with liquid metal coolants in IPPE is not restricted by sodium cooled fast reactors but includes the reactors cooled by liquid heavy metal, lead-bismuth eutectics first of all. This concept is revisited nowadays in connection with the possibility to use fast reactors cooled by heavy metals for Pu utilization and MA transmutation (see, for example [9-10]).

1.3. Experiments on BFS facilities.

Actinide integral measurements were carried out on BFS similar to those on a set of FCA-9 assemblies to test the fission and capture cross sections of minor actinides (MA) described in [11]. The integral data measured are:

- the central fission rate ratio (FRR);
- the central sample worthies (CSR).

The core of BFS-67 assembly was composed with 96% enriched metal plutonium, depleted uranium dioxide, sodium and stainless steel. This composition was similar to the SUPERPHENIX core. About half of uranium dioxide in this composition was replaced with sodium for constructing of BFS-69 assembly core.

For both BFS assemblies the spectra were similar to the spectrum averaged over FCA assemblies. The integral data measured are:

- FRR of Np-237 and Pu-239 relative to fission in U-235 ;
- CSW of Np-237 ,Pu-239 and U-235.

All samples sizes were less than in FCA experiments. The Np and Pu samples were of three different size. The exact description of the assemblies, experimental devices and obtained results of measurements were given in [12].Some of the results provided by the authors are given in Tables 1-4.

The starting point of neutron data testing is homogeneous calculation of FRR and first order perturbation theory using ABBN approach [13] . Evaluation of experiments means taking in account the heterogeneous structure of core cells, finite sizes of samples and group constant correction at calculation of CSW.

Heterogeneous structure of critical assembly's cell is taken into account by using the integral-transport approximation. Undisturbed group fluxes and adjoint fluxes are obtained from solutions of corresponding integral-transport equations in the cell approximation. Criticality is attained by modification of a neutron leakage. Perturbation of collision probabilities are taking in account too by calculation results using perturbation theory (first type of correction). Consideration of a detailed energy structure of adjoint solution gives the additional contribution into the reactivity worth ratio (second type of correction). Taking in account the finite size of samples have consisted the third type of correction.

The results of measurements and evaluation are presented in Tables 1 and 3 for CSW ratios of Pu - 239 / U - 235 , in Table 2 for CSW ratios of Np-237/Pu-239 and in Table 4 for CSW ratios of Np-237/U-235.

Table 1. CSW ratio U-235 / Pu - 239

Assemb. FCA	EXP. virgin	Correction of 1 and 2 types	Correction of 3 type for Pu	Correction of 3 type for U	EXP. evaluated
9-1	1.476	-.023	-.146	+.109	+1.416
9-2	1.617	-.010	-.127	+.124	+1.604
9-3	1.713	-.009	-.008	+.016	+1.712
9-4	1.708	-.006	-.079	+.089	+1.712
9-5	1.750	0	-.075	+.081	+1.756
9.7	1.745	-.002	-.065	+.060	+1.738

Table 2. CSW ratio Np-237 / Pu - 239

Assemb. FCA	EXP. virgin	Correction of 1 and 2 types	Correction of 3 type for Pu	Correction of 3 type for U	EXP. evaluated
9-1	-.865	+.179	-.220	-.073	-.979
9-1	-.242	+.072	-.042	-.013	-.225
9-3	-.014	+.021	-.014	+.005	-.002
9-4	+.054	+.006	-.005	+.006	+.061
9-5	+.158	+.002	-.004	+.010	+.166
9-7	+.117	+.001	-.003	+.007	+.122

Table 3. CSW ratio Pu - 239/U - 235

Assembly	Type of sample	EXP VIRGIN	ZERO SIZE of samples	correction of 1 and 2 types	EXP evaluated
BFS 67	Pu - A	1.350 ± .020	1.327 ± .021		
	Pu - B	1.406 ± .008	1.365 ± .009		
	Pu - C	1.404 ± .005	1.352 ± .007		
	averaged value		1.353 ± .019	+.011	1.364 ± .019
BFS 69	Pu - A	1.591 ± .018	1.560 ± .020		
	Pu - B	1.607 ± .007	1.552 ± .008		
	Pu - C	1.592 ± .004	1.528 ± .006		
	averaged value		1.541 ± .019	-.002	1.539 ± .019

Table 4. CSW ratio Np-237 / U-235

Assembly		VIRGIN EXP	ZERO SIZE of samples	correction of 1 and 2 types	evaluated experiment
67-1	Np - A	-.240 ± .011	-.250 ± .011		
	Np - B	-.228 ± .006	-.240 ± .008		
	Np - C	-.228 ± .005	-.245 ± .007		
	averaged value		-.245 ± .010	+.023	-.222 ± .011
69-1	Np - A	-.120 ± .010	-.128 ± .010		
	Np - B	-.119 ± .006	-.130 ± .007		
	Np - C	-.114± .003	-.131 ± .004		
	averaged value		-.130 ± .005	+.021	-.109 ± .006

1.4. Nuclear data measurements and evaluations.

Improvement of nuclear data for minor actinides is important for transmutation projects using actinide burner reactors and needed for the nuclides ^{237,238}Np, ^{238,242}Pu, ^{241,242,242m,243}Am and ^{242,243,244,245,246}Cm. Some of the most important cases are now investigated in the framework of ISTC Project-304 "Measurement and Analysis of Basic Nuclear Data for Minor Actinides". The latest works include:

- preliminary results of precise measurements of the fission cross sections of ^{244,245,246,247}Cm and ^{242m}Am by 0.15÷7.0 energy neutrons;
- first runs of measurements of secondary neutron spectra, fission product yields, delayed neutron yields and inelastic scattering cross section for Np-237;
- improved evaluations of the most important cross sections for Np-237.

Extensive calculations and evaluations of the nuclear data are now done for higher energy range to satisfy the needs of ADTT research and development. The latest results:

- special library is developed (MENDL-2, Medium Energy Nuclear Data Library) for the investigations of the activation and transmutation of the materials irradiated by the nucleons of intermediate energies (neutrons up to 100 MeV, protons up to 200 MeV, more than 100 000 reactions). First, neutronic, part is described in [14], proton part is to be converted to an agreed format and then released;
- systematics were developed of the cross sections of the threshold reactions on 14 MeV neutrons based on a new approach taking into account both equilibrium and non-equilibrium mechanisms of nuclear reactions [15-16].

1.5. Research on the accelerator targets for ADTT.

Efforts are undertaken in collaboration with Design Bureau "Gidropress" on the development of neutron producing liquid heavy metal targets irradiated by very powerful proton beams in GeV energy range. Both beam-window and windowless targets are investigated. This research includes:

- calculations of thermophysical and hydrodynamic properties of the targets and optimization of the designs;
- material studies;
- calculations of the diaphragm behavior, energy release, activation, including the accumulation of long-lived radionuclides, and gas production in liquid Pb-Bi and Pb targets;
- release of radionuclides from liquid and solid targets.

A few papers on these subjects were presented at Kalmar Conference [17-21].

2. Selected results on the actinides balance in various scenarios of Pu and MA recycling

2.1. Comparison of thermal and fast reactors in actinides recycling.

Transmutation of MA is closely linked with Pu utilization and would influence the actinide balance. The problem is whether MA burning is compatible with Pu breeding needed to support stable to say nothing of developing nuclear power industry. Another problem is whether nuclear safety requirements to Pu and MA containing cores may be met in fuel cycles involving Pu breeding. It's very wide domain so here we consider only some most prominent differences between thermal and fast reactors used for multiple long-term recycling of transuranics. A simple calculational model based on KARE code [22] was chosen to make the comparison (the results were presented in [4]). Basic features of the model are:

- the fuel of the first cycle is MOX-fuel, mixture of depleted uranium with reactor plutonium (isotopic composition of the latter is given in Table 5); enrichment by plutonium is sufficient to support chain reaction for planned campaign period and to reach planned burnup (parameters of the first cycle are given in Table 6);
- neutron spectra, fluxes and one-group constants coincide with those of BN-800 for fast reactor and with the parameters of Pu fueled VVER-1000 for thermal reactor;
- after each cycle the spent fuel is cooled for three years, fission products are eliminated completely, plutonium of initial composition and depleted uranium are added in the quantities necessary to complete the next cycle;
- all the actinides are left in refabricated fuel.

Table 5. Isotopic composition of fresh plutonium used in calculational model.

Isotope	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242
Concentration, per cent	0.5	60.0	24.5	10.9	4.1

Table 6. Parameters of the first cycle.

Parameter of the cycle	Reactor type	
	Fast	Thermal
Time of irradiation, days	420	920
Pu enrichment, per cent	18.9	5.96
Neutron flux, $cm^{-2} c^{-1}$	$6.0\ 10^{15}$	$4.0\ 10^{14}$
Burn up, per cent of heavy atoms	6.6	3.6
Number of fissions/number of captures	0.98	0.65

Final actinide composition was calculated for every cycle and then the quantities of plutonium and depleted uranium to be added to the fuel were determined. The behavior of fast and thermal reactors was compared by following parameters: plutonium enrichment of the fuel; average value of neutron flux; share of the fissioning isotopes in plutonium; concentration of minor actinides. The results are presented in Figs. 1-6 as a function of total burnup for all cycles (which may be more than 100 per cent if the number of cycles is large enough - more than 15 for fast reactors and more than 25 for thermal reactors which refers to rather long periods of time).

These results demonstrate that:
- enrichment grows slower in fast reactor, almost stabilizing asymptotically; it's important because enrichment in thermal reactors is restricted by safety requirements at about 10 per cent;
- average flux is practically stable in fast reactor but it's almost halved in thermal reactors and this decrease is to be compensated by enrichment;
- degradation of Pu isotope composition is much faster in thermal reactors;
- accumulation of neptunium is almost identical in both reactors up to 50 per cent burn up and then americium decay will increase it in TR (burn up is four times faster in fast reactors);
- accumulation of americium and especially of curium is more intense in TR.

These calculations were done for FR without breeding blankets.

2.2. The effects of plutonium composition and breeding ratio value on recycling of the actinides in fast sodium cooled reactors of BN type.

Multiple recycling of plutonium and MA in sodium cooled fast reactors BN-800 with highly Pu-enriched MOX-fuel was considered for the cases of civil Pu (BR=1.0 and 1.3) and weapon-grade Pu (BR=1.0) with irradiation cycle of 420 days. Following values were calculated:
- initial enrichment and final burn up in all three sub-cores for every cycle;
- plutonium isotopic composition at the beginning of every cycle;
- actinides concentrations at the end of every cycle.

Some of the results are illustrated by Figs.7-12 . Following conclusions may be made:

1. There is prominent difference in the evolution of Pu isotopic composition during the recycling in converter and breeder modes: plutonium quality (the concentration of fissionable isotopes Pu-239 and Pu-241) is degrading significantly in converter mode while it is even improving slightly in the breeder mode, first of all due to accumulation of Pu-239 (see Fig.7) because very high grade plutonium is produced in the radial blanket (more than 96 per cent of Pu-239).
2. Minor actinides pile up slower in the breeder mode as may be seen in Figs.9-10 for the case of the most important nuclides Am-241 and Cm-244.
3. Switching to weapon-grade plutonium reduces accumulation of MA in converter mode drastically (see also Figs.9-10).
4. Tendencies mentioned in the points 2-3 are consequences of the reduced concentration of the key nuclide Pu-241 in the fuel (see Fig.8).
5. Practically all accumulation of Am-241 in plutonium fuel is due to the decay of Pu-241 in the load which may be seen in Fig.11 where the mass of Am-241 Vs Pu-241 mass in the load is shown. These masses are practically directly proportional both in for civil and weapon-grade plutonium.
6. Strong correlation exists also between the accumulated mass of most important curium isotope Cm-244 and Pu-242 concentration in the fuel load (Fig.12).

2.3. On combining thermal and fast reactors in Pu utilization and MA transmutation.

Various possibilities to create a combined system of fast and thermal reactors capable of recycling all the plutonium and MA are analytically explored now. Some of the latest results [23] are outlined below. The system considered includes two types of reactors: VVER-1000 with either uranium or 30 per cent MOX fuel and BN-800 with various cores. Basic parameters of the systems and some results of the calculations are given in Tables 7-9.

Table 7.Parameters of BN-800 cores.

Model number	Parameters of the model	Volume share of the fuel	Average Pu enrichment, per cent	Actinides burn up,kg/GWe year
1.	Uranium fuel with high Pu enrichment	0.29	37.0	57
2.	Inert matrix fuel without U-238	0.10	100	110

Table 8. Equilibrium fuel composition in VVER-BN system (kg/t).

Nuclide	Model 1				Model 2	
	Burnup 10%		Burnup 20%		Burnup 50%	
	Tc=1year	Tc=3 years	Tc=1 year	Tc=3years	Tc=1 year	Tc=3 year
U-235	2.4	2.3	2.1	2.1	-	-
U-238	564.1	575.9	521.4	516.4	-	-
Pu-238	12.3	14.7	13.5	15.0	46.6	53.5
Pu-239	151.4	153.8	170.1	171.6	249.0	250.1
Pu-240	137.6	140.3	153.6	155.5	358.7	358.3
Pu-241	30.0	26.6	38.1	35.6	80.2	67.9
Pu-242	38.6	38.4	47.3	47.4	118.6	119.0
Np-237	6.0	6.3	8.0	8.2	17.1	17.8
Am-241	12.4	17.9	13.8	17.6	47.2	62.1
Am-242	0.7	0.9	0.7	0.8	3.0	3.5
Am-243	14.3	14.3	18.0	18.2	46.6	43.7
Cm-242	0.2	0.02	0.2	0.02	0.6	0.06
Cm-244	7.8	6.8	10.4	9.1	26.3	20.1
Cm-245	2.1	1.8	2.7	2.5	5.3	4.0

Table 9. The number of VVER-1000 reactors with the actinide output utilized by one BN-800.

BN model	BN burnup		
	10%	20%	50%
Model 1	1.5 (1.0)*	1.7 (~ 1.05)	
Model 2			2.6 (2.6)

* Figures in brackets refer to 30 per cent MOX fueled VVER-1000.

3. The neutrons available for transmutation of fission products.

The price and value of the neutrons used for transmutation may be estimated by considering the income provided by nuclear electricity production. The approach [24] based on such considerations is used below in a simplified form. These results were presented in [25]

Modifications in energy producing system decreasing sales of electricity is the price of achieving environmental goals. To get one neutron as a result of interaction of high energy proton with specially designed target one needs not less than 20 MeV of beam energy. Taking thermal efficiency of electricity production equal to 30 per cent and accelerator beam efficiency 40 per cent we conclude that production of one " external" neutron takes the energy almost equal to that released in an act of fission. Thus the production of 4.2 kg of neutrons in the proton target means the loss of income equivalent to selling electricity generated by 1 t of fissioned heavy metal, i.e. approximately 1 GWt(e) year. If every neutron transmutes 1 nucleus of Tc-99 then that's the price of transmuting 420 kg of Tc-99.

Not only " external" neutrons may be used for transmutation but also reactor excess neutrons not used for energy production. Their alternative use may be breeding of Pu, so their price is not less than the value of lost Pu.

In the case of actinides transmutation in equilibrium mode when every nucleus of fresh fuel is sooner or later fissioned after a chain of nuclear transformations consuming neutrons the price of the neutrons is not easy to determine.

Hybrid transmutation concept when accelerator-driven blanket both produces energy and multiplicates neutrons is most popular now. In this case both " external" and " multiplied' neutrons are used to transmute FP. But it does not a priori mean that one " external" neutron provides transmutation of more than one fission fragment.

Let's consider the neutronics of FP transmutation in accelerator-driven blanket. Notations used: ν - the number of fission neutrons, Σ_f - macroscopic fission cross section of the fuel; Σ_T - macroscopic total absorption cross section; Σ_{tr} - macroscopic cross section of FP placed in the blanket to be transmuted; $k = \nu\Sigma_f/(\Sigma_T + \Sigma_{tr})$ - blanket multiplication; $k_o = \nu\Sigma_f/\Sigma_T$ - initial multiplication of the blanket free from FP. The maximum number of fissions initiated by one external neutron in the blanket is $\varphi = k/\nu(1 - k)$. The share of neutrons absorbed by FP being transmuted is

$$\eta = \Sigma_{tr}/(\Sigma_T + \Sigma_{tr}) = 1 - k/k_o \quad (1)$$

This definition differs from that of Takahashi [26] by a factor $(1 - k/\nu)$ because in [26] the share of absorptions used is $\eta_{FP} = \Sigma_{tr}/(\Sigma_T - \Sigma_f + \Sigma_{tr})$ which does not provide direct information on the share of neutrons used for transmutation.

The number of FP transmuted per one "external" neutron (the transmutation "value" of external neutron) is the product of η and the number of multiplied neutrons:

$$\tau = \eta(1 + \varphi\ \nu) = (1 - k/k_o)/(1 - k) = 1 - \rho_o/\rho \quad (2)$$

where $\rho = (k - 1/k)$ is blanket reactivity. Let's now consider different reactivities of initial blanket (without FP added). If it were critical, $k_o = 1$, $\rho_o = 0$, we get $\tau = 1$, i.e. each "external' neutron provides transmutation of one FP just compensating added absorption. If initial blanket is subcritical ($k_o < 1$) then $\tau < 1$, i.e. "external" neutron transmutes less than one fragment in spite the multiplication in the blanket. And, finally, $k_o > 1$ results in $\tau > 1$.

If some quantity of FP is placed in initially supercritical blanket, making it precisely critical, the blanket will operate in reactor mode transmuting $1 - 1/k_o$ of fragments per every "internal" neutron,. $\phi = k/\nu(1 - k)$ fissions will transmute $\tau^* = k(k_o-1)/k_o(1-k)$ fragments.

If the power of such a blanket, i.e. the rate of fissions, is equal to the power of subcritical accelerator-driven blanket, "combined" transmutation of FP in such a critical blanket plus direct transmutation by "external" neutrons will bring the same results as hybrid transmutation in subcritical accelerator-driven blanket with the same number of external neutrons: $\tau = \tau^* + 1$.

4. Criterion of radiation equivalence in environmental assessment of nuclear fuel cycles

Accumulation of highly radiotoxic nuclear wastes created a problem unique in the history of science - it's necessary to choose the direction of the development of nuclear technologies optimized both to ensure safety of our descendants in dozens and hundreds of thousands of years and to minimize radiological risks and economic losses for the present and nearest generations of nuclear electricity consumers who are not only to pay for those long-term safety measures but to learn how to handle and actually handle large volumes of radioactive materials taking risks and damages.

Now hazards of the nuclear power radwaste are estimated by comparison of its radiotoxicity with that of the excavated uranium ore (see, for example, [27,28]). This approach is typical for the use of traditional mineral resources - their volume reflects directly both their value and its environmental price. Nuclear power falls out of this picture in both aspects. Useful effect, i.e. the quantity of electricity produced, may be increased by almost two orders if we switch from today's open nuclear fuel cycle (ONFC) to closed one (CNFC).

We propose a modification of the assessment which is outlined below (see also [24]). In the estimates we shall assume the fission energy release equal to 200 MeV per act for all the actinides, differences in individual fission products (FP) yields for various actinides will be also neglected. In this approximation accumulation of 1 t of fission products corresponds to a little more than 1 GWt(e) year for standard thermal reactors. Consumption of uranium components and pile-up of the actinides are close for all major types of thermal reactors. Normalization of waste to 1 t of FP allows to compare hazards of various nuclear installations and cycles.

We shall use following mass notations: $M_{ore,i}$ - ore components, M_S - all spent fuel, $M_{F,i}$ -components of the fuel load, $M_{S,i}$ -components of the discharged spent fuel, M_{FP} - fission products, $\Delta_i = m_{S,i} - m_{F,i}$ where m_i are normalized to 1 t of FP. If specific "danger" of i-th nuclide is d_i then total danger of nuclides mixture is

$$D = \Sigma\ M_i d_i \quad (3)$$

What follows is true no matter what concept of "danger" is used, it may be mass, volume, activity, radiotoxicity, masses of diluting substances etc. Criterion of radiation equivalence of the spent fuel and excavated uranium usually used is the ratio of their dangers:

$$K_{S/R} = \Sigma M_{S,i}\, d_i / \Sigma M_{ore,i}\, d_i \qquad (4)$$

Only about 0.7 per cent of natural uranium nuclei react in ONFC. More than 99 per cent of them were dislocated but their radiotoxicity was unchanged. Whether such "displaced" nuclei are taken into account or not almost does not influence the numerator value in Eq(4) but is very essential for the denominator - the mass of uranium excavated is by two orders larger than the mass of uranium transmuted, the ratio of their activities being about 40. If one applies the criterion (4) to ideal CNFC where all the uranium excavated is turned into FPs with some 200 times more electricity produced we get a paradox - most efficient use of natural uranium formally results in increased environmental danger because the accumulation of FPs per ton of natural U increases correspondingly. In practice lower specific consumption of natural uranium in CNFC is not taken into account in comparisons of the waste radiotoxicities of various NFCs. Such a "default" refusal to use the criterion (4) needs justification.

Let's now formulate a criterion of radiation equivalence which takes the specific ways of using raw materials in nuclear power into account. The idea is to compare the accumulated danger with the danger of transmuted (not only fissioned) uranium excluding "displaced" nuclides. The ratio of the dangers summed up over all the power and transmutation plants (index k) is

$$K_{W/U} = \Sigma_k M^k_{S,FP} \Sigma_i \Delta^k_i d_i \,/\, \Sigma_k M^k_{S,FP} \Sigma_i \Delta^k_{ore,i} d_i \qquad (5)$$

$\Delta^k_{ore,i}$ includes not only U isotopes but co-extracted products of their decay chains. Total energy produced is proportional to $\Sigma_k M^k_{S,FP} \Sigma_i \Delta^k_{FP,i}$. The numerator includes total accumulation of FPs and accumulation or burning of actinides, the denominator - total quantity of the transmuted uranium. In CNFC without FP transmutation only FP and irretrievable losses of actinides in reprocessing enter the numerator, and denominator includes 1 ton of U per every ton in the numerator. This criterion makes obvious the advantages of CNFC compared to ONFC in a very long run (thousands of years) when the main contribution to radiotoxicity is made by actinides, burnt in CNFC and accumulated in ONFC. This criterion is valid for the case of FP transmutation as well but it is not without deficiencies also - according to it burning of less active ^{238}U is 6 times more dangerous (due to small destroyed radioactivity) then burning of U-235.

The use of Eq (5) (accumulated danger/annihilated danger) instead of (4) (danger of spent fuel/danger of excavated uranium) increases the relative danger of ONFC correcting the traditional approach. It should be stressed that absolute indices of waste dangers do not depend on the choice of any of the two criteria.

Conclusions

Research and development efforts on transmutation should be aimed at the methods of closing nuclear fuel cycle efficient in a few senses:

- saving natural uranium by breeding secondary fuel;
- producing and directing to the radwaste less long-lived alpha-emitters;
- ensuring nuclear and radiological safety of all power producing plants and reprocessing installations;
- reducing proliferation dangers by keeping most of plutonium in spent fuel and strictly regulating quantity of separated plutonium.
- prompt introduction of fast reactors using Pu is vital for solving these problems;
- already nearest generation of new reactors with life-time expectancy of at least 50 years should be designed flexible to meet the requirements of steadily growing plutonium breeding when necessary;
- using of high-grade Pu in CNFC reduces the accumulation of minor actinides;
- useful energy produced should enter the denominator in any numerical criteria describing radiological hazards of NFC.

REFERENCES

1. GLOBAL'93. International Conference and Technology Exhibition. Future Nuclear Systems: Emerging Fuel Cycles & Waste Disposal Options. Proceedings. September 12-17, 1993, Seattle,Washington. Volumes 1-2.
2. GLOBAL 1995, International Conference on Evaluation of Emerging Nuclear Fuel Cycle Systems. September 11-14,1995, Versailles, France. Volumes 1-2.
3. Second International Conference on Accelerator-Driven Transmutation Technologies and Applications. 3-7 June 1966, Kalmar, Sweden. Abstracts.
4. E.V.Gai, B.K.Gordeev, A.L.Kochetkov, T.S.Milovidova, N.S.Rabotnov. The Closure of Nuclear Fuel Cycle: Actinides Balance, Safety and Fuel Provision Problems. The Eighth International Conference on Emerging Nuclear Energy Systems, ICENES'96, Obninsk, Russia, 24-28 June 1996. Book of Abstracts., p.4.
5. Suzuki T. Long-Term Logistic Analysis of FBR Introduction Strategy: Avoiding Both Uranium and Plutonium Shortage. In [2], v.2, p.1971.
6. V.I.Matveev. Choice and Preliminary Justification of BN-600 and BN-800 Reactors for Actinides burning and Its Fuel Cycle. PHYSOR-96 Paper M-290.
7. A.P.Ivanov, E.M.Efimenko, A.G.Tsykunov, G.G.Buiburin. Analytical Study on Optimization of Fast Reactors for U-233 Production and Plutonium and Minor Actinides Burning.. PHYSOR-96 Paper SM-200.
8. A.P.Ivanov, E.M.Efimenko, A.G.Tsykunov. Fast Reactor Application for the Fission Products Burning. PHYSOR-96 Paper SM-150.
9. A.G.Kalashnikov, N.N.Novikova, Yu.G.Pashkin, V.V.Chekunov. Some Investigations of the Lead-Bismuth Cooled Fast Power Reactor for the Weapon-Grade Plutonium Utilization and the Actinide Burn-Up. The Eighth International Conference on Emerging Nuclear Energy Systems, ICENES'96, Obninsk, Russia, 24-28 June 1996. Book of Abstracts, p.132.
10.G.I.Toshinskiy. The Concept of Nuclear Power Development on the Basis of LMFBR Operating in the Open Nuclear Fuel Cycle. The Eighth International Conference on Emerging Nuclear Energy Systems, ICENES'96, Obninsk, Russia, 24-28 June 1996. Book of Abstracts, p.36.
11.T. Mukaiyama et. all, Actinide Integral Measurements on FCA for Evaluating and Improving Their Cross Section Data, 1985, NEACRP - A - 684.
12. S.P.Belov, J.C.Cabrillat et al.. Investigations of MA Transmutation Problem in Bench-Mark Experiments at BFS Facility with Neptunium in Fuel Composition. PHYSOR-96 Paper M-296.
13. L.P.Abagyan, N.O.Bazazyants, M.N.Nikolaev, A.M.Tsybulya. Group Constants for Reactor and Shielding Calculations. Energoizdat, Moscow, 1981(In Russian).
14. Yu.N.Shubin, V.P.Lunev, A.Yu.Konobeev, A.I.Dityuk. Cross Section Data Library MENDL-2 to study Activation and Transmutation of Materials Irradiated by Nucleons of Intermediate energies. INDC(CCP)-385, May 1995, IAEA, Vienna.
15.Yu.N.Shubin, V.P.Lunev, A.Yu.Konobeev. Semi-Empirical Systematics for (n,α)-reaction cross section. Nuclear Instruments and Methods in Physical Research. (1996), v.B108, pp.233-242.
16. A.Yu.Konobeev, V.P.Lunev, Yu.N.Shubin. Semiempirical systematics of (n,d), (n,t) and (n,np) reactions cross sections at 14.5-15 MeV. IPPE Preprints 2491,2492,2494 (1996).
17. A.A.Veremeev, V.T.Gorshkov, B.F.Gromov, E.I.Efimov, M.P.Leonchuk. Preconceptual Design of Liquid Metal Targets with Diaphragm for Accelerator-Based Systems.[3], p.T/B7.
18. B.F.Gromov, Yu.I.Orlov, M.I.Sotov, V.A.Gulevsky, Yu.A.Musikhin. Studies of Windowless Liquid Metal Target with Lead-Bismuth and the Coolant Technology. [3], p.T/B25.
19. E.I.Efimov, D.V.Pankratov. Polonium and Volatile Radionuclides Output from Liquid Metal Target into Ion Guide and Gas System. [3],p.S/C3.
20. Yu.N.Shubin, A.V.Ignatyuk, A.Yu.Konobeev, V.P.Lunev, E.V.Kulikov. The Analysis of Energy Release, Beam Attenuation, Radiation Damage, Gas Production and Accumulation of Long-Lived Activity in Pb an Pb-Bi Targets. [3], p.M4.
21. E.I.Efimov, D.V.Pankratov, S.I.Ignatiev. Secondary Neutrons Yields by 0.8- and 1.6-GeV Proton Interaction with Pb and W thick Targets. [3], p.D47.
22. A.L.Kochetkov KARE code for the calculations of isotopes kinetics, radiational and environmental properties of nuclear fuel during its irradiation and cooling. IPPE preprint-2431, Obninsk, 1995.
23. V.M.Poplavskiy, V.I.Matveev, N.S.Rabotnov. Paper, presented to the Conference of Russian National Nuclear Society "New Power Technologies and the Role of the Nuclear Power, Fission and Fusion, September 14-18, 1996, Moscow.

24. H.Kuesters, H.W.Wiese, M.Salvatores. Transmutation of Actinides and Fission Products and Corresponding Fundamental Safety Aspects. In: Safety and environmental aspects of partitioning and transmutation of actinides and fission products. Proc. of a Technical Committee meeting held in Vienna, 29 November - 2 December 1993. IAEA-TECDOC-783. IAEA, January 1995., p.53.
25.E.V.Gai, A.L.Kochetkov, N.S.Rabotnov. Radiological Criteria, Potential and Limitations of ADTT at Closing Nuclear Fuel Cycle. [3], p.S35.
26 Takahashi, H., "Transmutation of High-Level Radioactive Waste by a Charged Particle Accelerator", in Proceedings of Global'93, International Conference on Future Nuclear Systems: Emerging Fuel Cycles and Waste Disposal Options, Seattle, WA, September 12-17, 1993, pp1293-1299.
27. Koch, L., "Minor Actinide Transmutation - a Waste Management Option", Journal of Less-common Metals, 122 (1986), pp371-382.
28. Adamov, E.O., Ganev I.C., Orlov, V.V., Attainment of Radiation Equivalence in Nuclear Power Radioactive Product Management, Nuclear Technology, v.104 (1993), N2, pp 233-241.

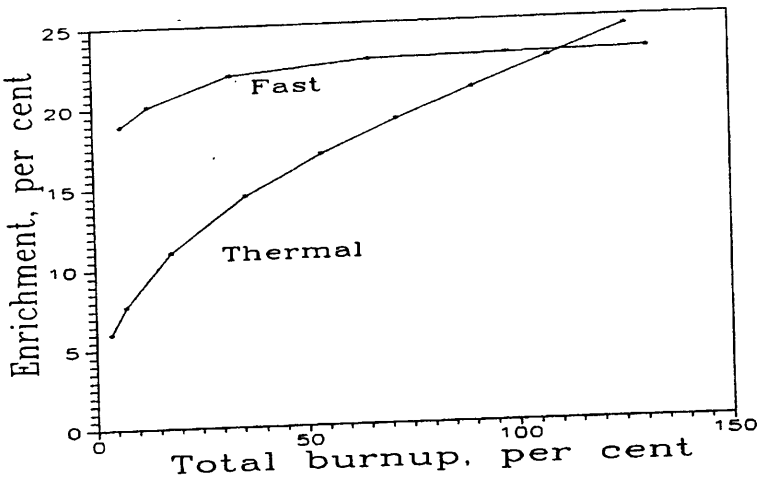

Fig.1. Increasing of enrichment in multiple recycling of actinides.

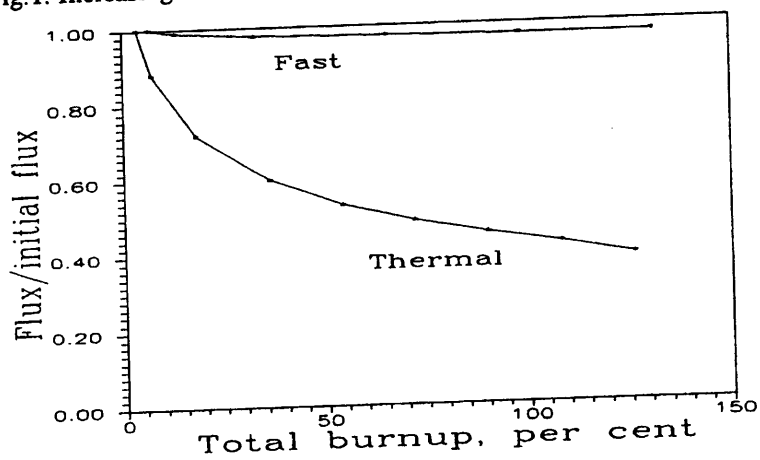

Fig.2. Flux degradation in multiple recycling of actinides.

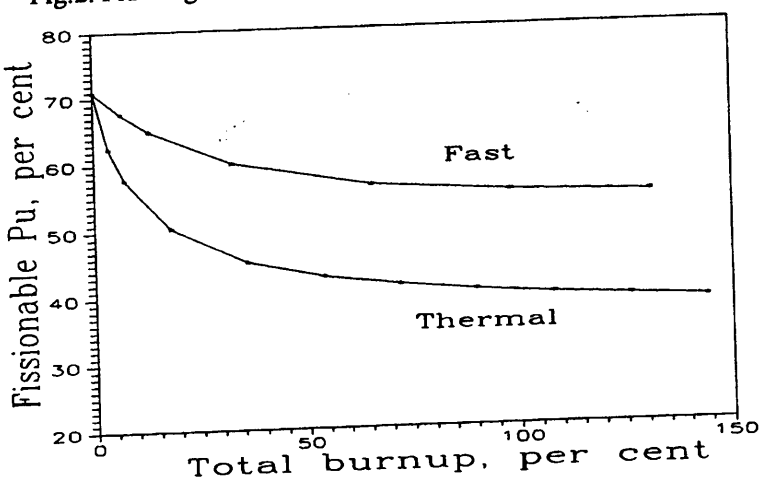

Fig.3. Degradation of Pu isotopic composition in multiple recycling.

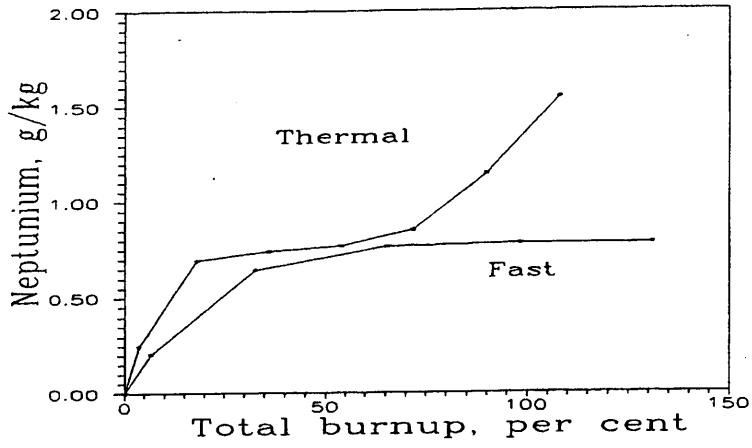

Fig.4. Accumulation of neptunium in actinides recycling.

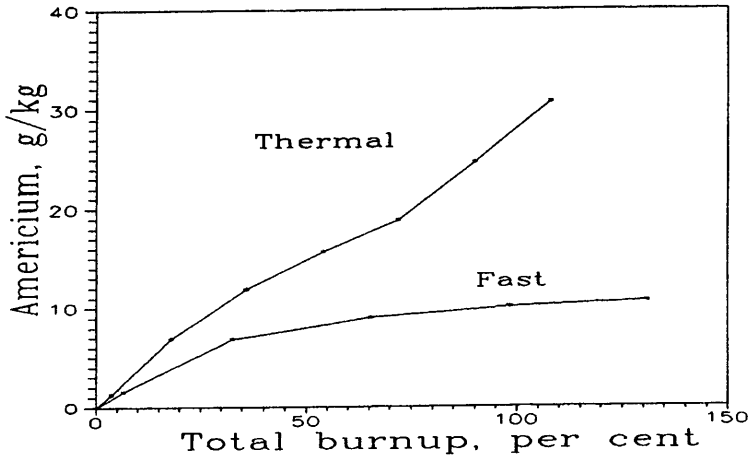

Fig.5. Accumulation of americium in actinides recycling.

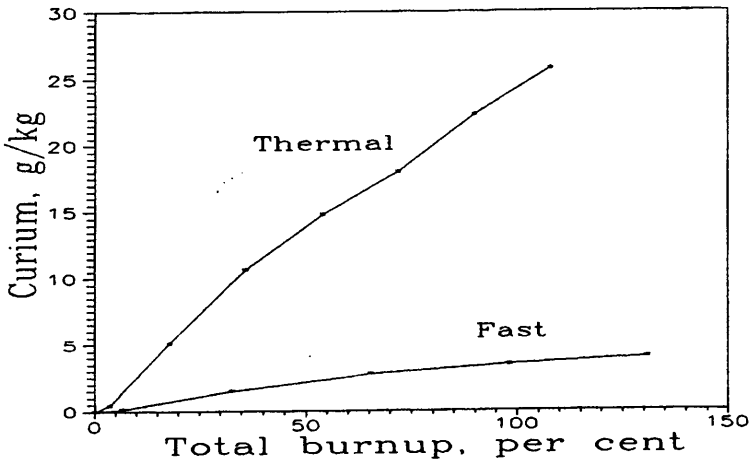

Fig.6. Accumulation of curium in actinides recycling.

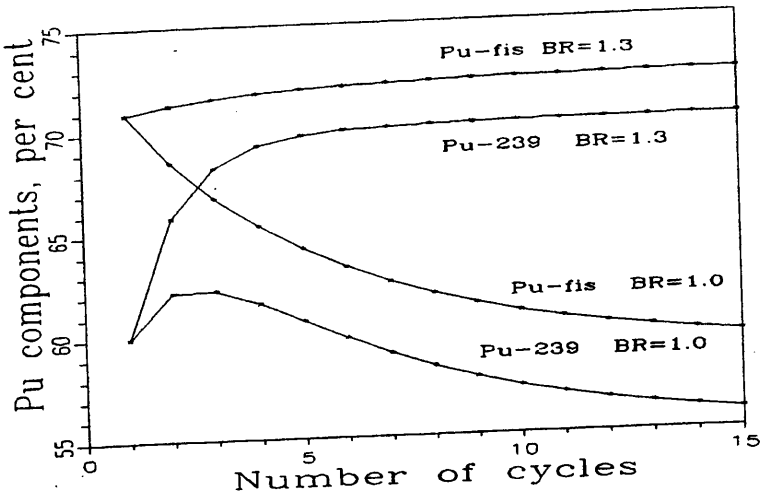

Fig.7. Evolution of Pu isotopic components in multiple recycling in BN-800 fast reactor at different values of breeding ratio.

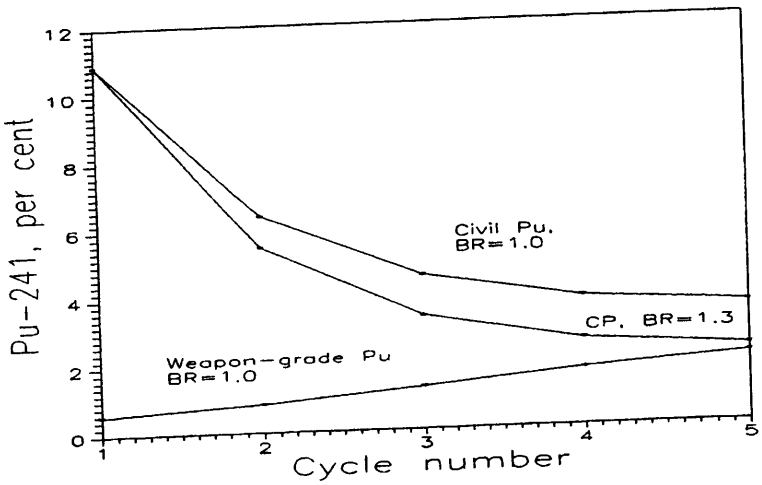

Fig.8. Total accumulation of Pu-241 in the core of BN-800 operating in various recycling modes.

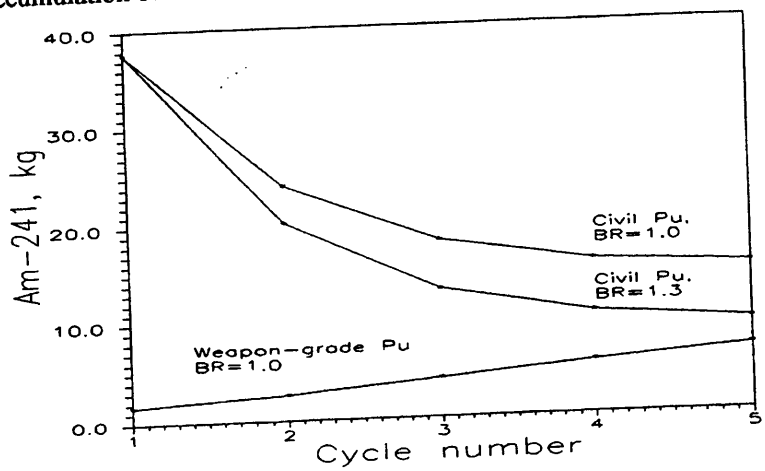

Fig.9. Total accumulation of Am-241 in the core of BN-800 operating in various recycling modes.

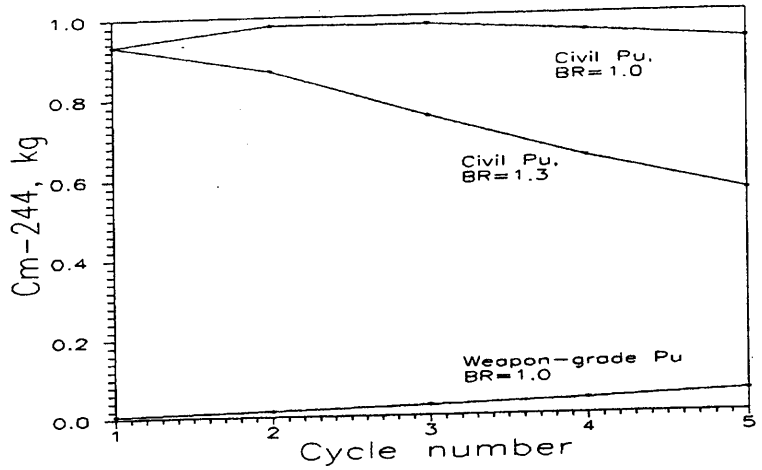

Fig.10. Total accumulation of Cm-244 in the core of BN-800 operating in various recycling modes.

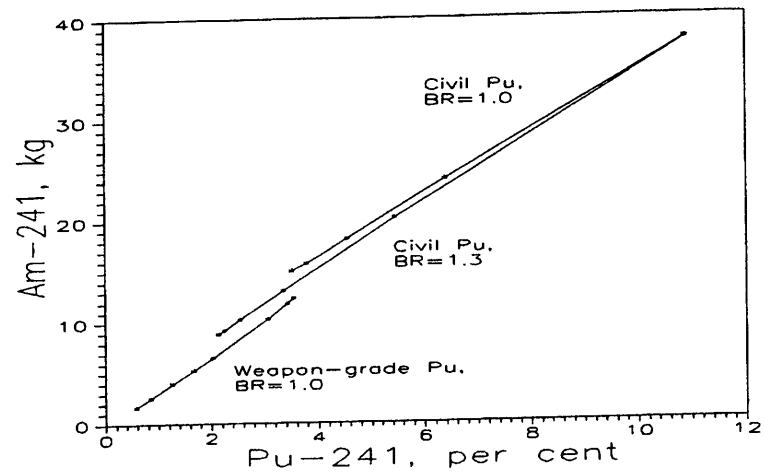

Fig.11 . Accumulation of Am-241 as a function of Pu-241 concentration in the fuel load of BN-800 during recycling of civil and weapon-grade plutonium in the converter mode (BR=1) and of civil Pu in breeder mode (BR=1.3).

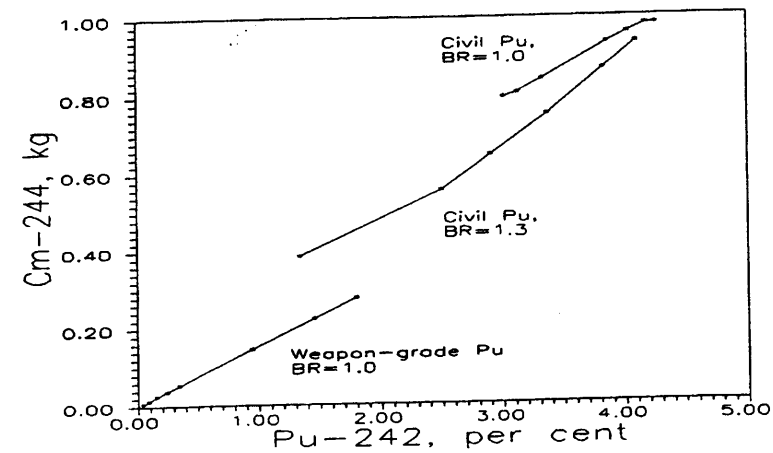

Fig.12. Accumulation of Cm-244 as a function of Pu-242 concentration in the fuel load of BN-800 during recycling of civil and weapon-grade plutonium in the converter mode (BR=1) and of civil Pu in breeder mode (BR=1.3).