FAST REACTORS UTILIZATION FOR LONG-LIVED NUCLEAR WASTES TRANSMUTATION

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ABSTRACT

The possibility calculation studies have been carried out for transmutation long-lived wastes of the nuclear power (minor actinides and fission products) in a fast reactor with the use of special devices, containing a large amount of moderator. Such devices can replace both radial blanket sub-assemblies and axial blanket. It has been shown that the implementation of these devices will allow the achievement long-lived waste burn-up at a level of 90-95%, decreasing essentially the radiotoxicity of wastes to be buried.

1. INTRODUCTION

A spent fuel of modern nuclear reactors contain a rather large quantity of long-lived high-active wastes (plutonium, minor actinides (MA) and fission products(FP)). These wastes quantity increases with increase in electricity production. If plutonium can be used as a fuel for reactors (both thermal and fast), the ways should be found for essential decrease of a danger from minor actinides and fission products. Therefore, the major problem of future nuclear system is a decrease in contamination of the environment by such wastes.

Nevertheless, the existing reactors are able to solve this type problems before development and putting into operation new promising nuclear systems. However, the implementation of thermal reactors for solving these problems is not efficient due to essential limitations in the core physics. The use of traditional fast reactors allows to solve the task partially. Really, a homogeneous wastes addition to the fuel makes it possible to utilize annually only 15% of all produced in this period MA and less than 10% of long-lived FP. However, a noticeable degradation of neutronic parameters (increase in sodium void reactivity effect (SVRE) and decrease in Doppler-effect) requires a search for other decisions, more acceptable from the safety standpoint.

One of these decisions can be implementation of special irradiation devices (ID), which can be located either in radial or in axial blanket. In this case the effect on the core physics will be

much less, as compared with homogeneous waste introduction to the fuel. But the efficiency of transmutation in these devices will be essentially lower.

The efficiency of these devices can be improved by introduction into them a rather large quantity of moderator. This is connected with that practically all FP have a maximum of cross-sections just in the thermal region and MA cross-sections increase as well at shifting of neutron spectrum to the thermal region. The possibilities for MA and FP efficient transmutation in ID containing moderator are considered in this report for a fast power reactor of BN-800 type as an example.

2. MA BURNING IN SPECIAL DEVICES LOCATED IN THE RADIAL BLANKET.

Americium oxide located in a magnesium oxide inert matrix is considered as a fuel composition for MA burning. A ratio between volume fractions of americium oxide and magnesium oxide can be varied in such a way that to conserve the total americium loading in the ID.

We note that fast reactors has an important advantage over thermal reactors, when burning minor actinides, because their neutron flux is two orders as higher. However, in a fast spectrum actinides have less cross-sections, as compared to a thermal spectrum.

On the basis of these two factors an idea appears to use moderated sub-assemblies (SAs) in fast reactors. In this case we conserve a rather high neutron flux and essentially increase the actinides cross-sections[1].

As an ID, we can consider a core SA, in which a part of fuel pins is replaced by pins containing americium oxide in an inert matrix, and others by pins containing a moderator. Varying the number of fuel pins with americium and moderator, one can change a moderator volume fraction in SA. For a more essential moderator fraction increase, on can use a promising fuel pin design, in which the central target material rod (of small diameter, with cladding or without it) is surrounded by a rather thick moderator layer. In this design it is very easy to vary the ratio of fuel and moderator volume fractions in a wide range, conserving the fuel pin external diameter and the developed for the core SA design. Just this type SA design was used in further studies.

One of the tasks of optimization studies was a search for moderator material, allowing a more efficient actinides transmutation, all other things being the same. Fig.1. presents the dependencies of the capture and fission cross- sections of some actinides on material type and Fig.2 - the dependencies of the same cross-sections on moderator volume fraction in the fuel pin for zirconium hydride as the most efficient moderator.

We note that the transmutation of americium in such ID would be worthwhile only if the radiotoxicity of wastes remained after irradiation is much less than the radiotoxicity of non-irradiated americium. Fig.3. presents a change in waste radiotixicity for different ID burn-up in relation to storage of non-irradiated americium

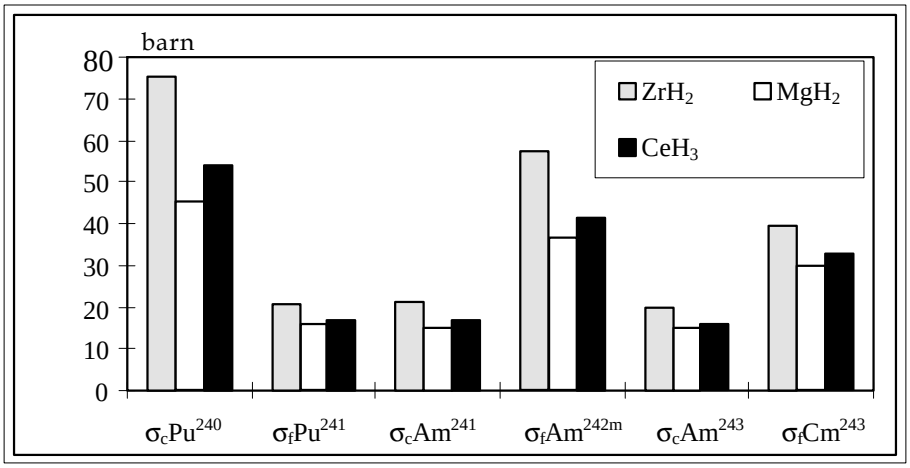

Fig.1. Dependencies of actinide cross-sections on a moderator type

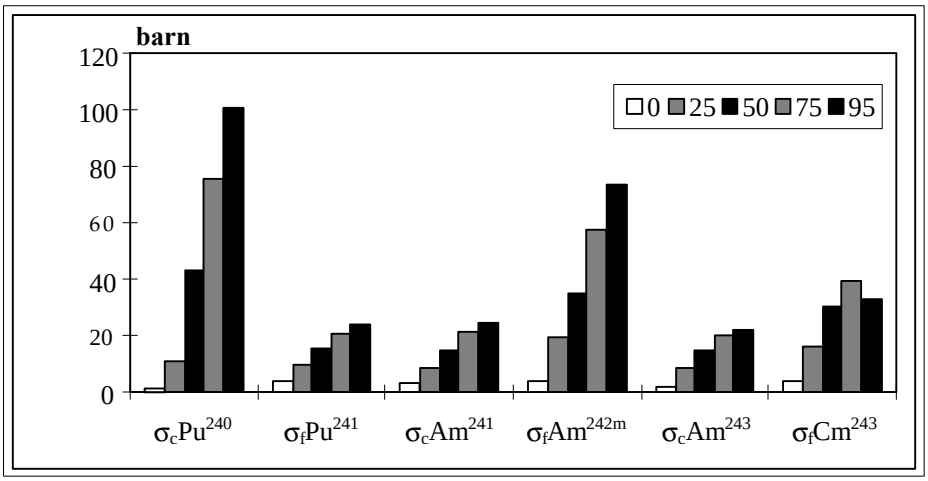

Fig.2. Dependencies of actinide cross-sections on a moderator volume fraction

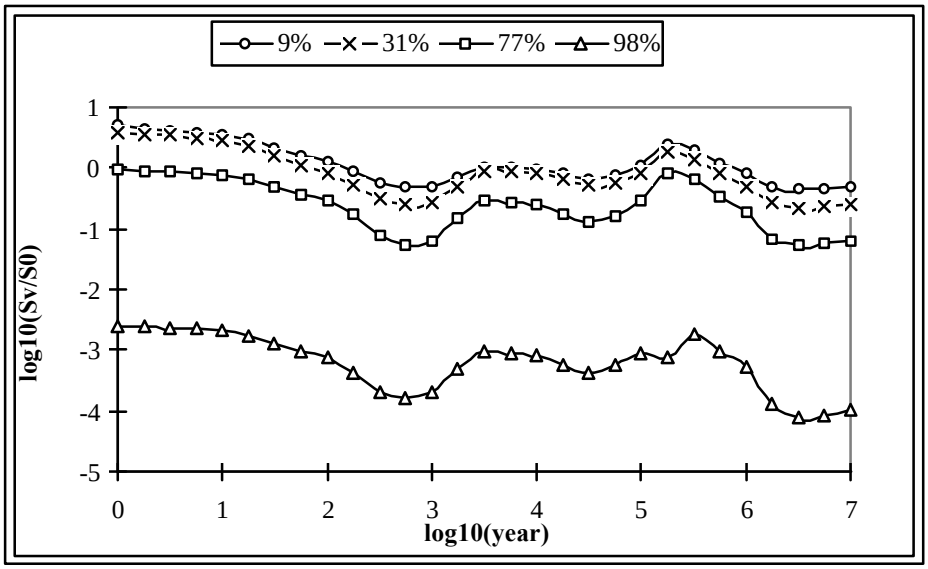

Fig.3. A change in radiotoxicity of ID wastes for different burn-up relating to storage of non-irradiated americium

The dependencies presented show that for decrease in the waste radiotoxocity at least two orders it is necessary to reach 93-95% h.a. americium burn-up. Thus, when designing an ID for americium burning, we should proceed from a necessity to reach just this high burn-up with maximum possible americium loading, and at the same time not to fall outside the existing limitations for basic structure materials of fast reactor cores.

Fig.4. presents the dependencies of americium burning value on irradiation time interval for different moderator volume fraction (zirconium hydride).

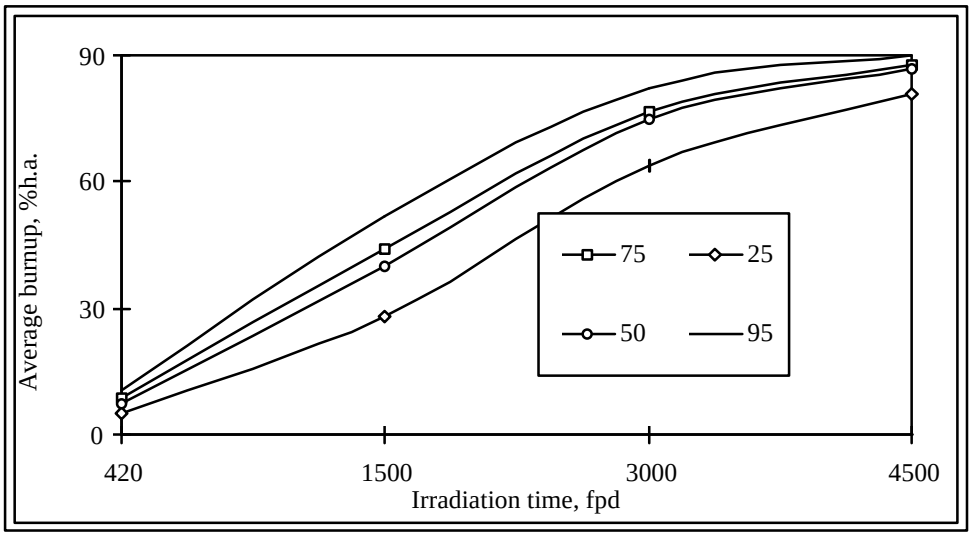

Fig.4. Average americium burnup as a function of irradiation time

It is easy to see that a handling the problem of reaching ~90% h.a. burning requires an essential irradiation time (~10-15 years) and a rather high volume moderator fraction in fuel pins (>90%). We note that so long irradiation of SAs with americium will require the development of a special reloading regime. For example, in order to eliminate an essential burn-up irregularity over SA cross section, it is necessary to turn it 180° during irradiation cycle. In this case the maximum damage dose will be ≈200 dpa, which will require the performance substantiation for structure materials that will be used in these ID. A large changes in ID power with americium burnup will require the development of a special regime for their cooling. Besides, the introduction of the ID with moderator in the first row of radial blanket will lead to increase in power of adjacent core SAs. The changing in power of these SAs can reach 20-30%. However, in our opinion, this power increase is not critical and will not require special measures.

Thus, in order that the americium transmutation in ID is appropriate from standpoint of essential decrease in actinide radiotoxicity, average americium burnup should be not less than 90%h.à. This requires the irradiation time not less than 10 years, which, in its turn, will require the structure material performance substantiation for high damage doses.

3. FISSION PRODUCTS TRANSMUTATION IN IRRADIATION DEVISES.

Among different FPs, greatly contributing to a long-lived activity, it is the practice to separate a group of several isotopes: Tc^{99}, Pd^{107}, Zr^{93}, Cs^{135}, I^{129}, Sn^{126} and Se^{79}. The problems of

these isotopes transmutation both in thermal and fast reactors have been considered in detail in many publications[2,3,4]. The fact is no longer doubted that a more efficient transmutation of these nuclides is reached in a thermal or close to thermal neutron spectra, since the basic resonances of these FPs are just in this energy range.

Two aspects are considered in this report, connected with FP transmutation in fast reactors with special ID implementation, containing a large moderator quantity:

– an effect of different moderators on the FP transmutation efficiency;
– an effect of irradiation devices on major neutronic core characteristics.

Let us consider two possible ways to locate the IDs: in first row of the radial blanket and instead of the lower blanket.

3.1 TECHECIUM-99 TRANSMUTATION

3.1.1. Transmutation efficiency.

Tc^{99} half-life period is $2.13*10^5$ years, and its production in spent fuel of modern power reactors comprises 3.0 kg/TW*h for fast reactors and 3.2 kg/TW*h for thermal reactors.

Table I presents a comparison of Tc^{99} transmutation when using different moderators.

Table I. Comparison of Tc^{99} transmutation efficiency when using different moderators

Moderator	Radial blanket		Axial blanket	
	%/cycle	kg/TW*h	%/cycle	kg/TW*h
CaH_2	77.3	2.29	35.7	5.33
MgH_2	77.3	2.29	35.7	5.33
TiH_2	71.5	2.12	31.2	4.66
CeH_3	74.6	2.21	33.5	5.01
ZrH_2	72.1	2.14	31.7	4.73
Be	64.0	1.90	26.2	3.92
C	60.1	1.78	24.0	3.58
Be_2C	65.3	1.94	27.1	4.04
$NbBe_{17}$	63.5	1.88	25.9	3.87

It should be noted that the introduction of hydrated moderators makes it possible to obtain the most efficient transmutation. The effect of volume moderator fraction on the transmutation rate and absolute value of Tc^{99} transmutation is presented in table II.

Table II. Effect of moderator volume fraction on Tc^{99} transmutation efficiency (radial blanket/axial blanket).

	Moderator volume fraction (CaH_2), %			
	0	22	80	95
%/cycle	11.2/3.4	15.4/4.8	44.0/15.6	77.3/35.7
kg/TW*h	6.0/9.6	7.3/11.1	5.9/10.2	2.3/5.3

When increasing the moderator fraction up to 95%, it is possible to transmute up to 80% of initial loading of the ID during once-through irradiation, thus providing ~8% per year. More transmutation rate (up to 25%/year) can be provided by an ID located in the axial blanket.

However, a small irradiation time (~1.5 year) allows to burn during one cycle only 35% of loaded technetium.

Thus, the introduction of a moderator in ID allows an essential increase in the transmutation rate. However, a decrease in these case of total FP loading leads to decrease in absolute value of transmuted technetium.

Notice that even an essential decrease in the quantity of long-lived FPs as a result of irradiation doesn't still assure a solving the task of activity decrease, since after irradiation the nuclides can be produced, the radioactivity of which can exceed that of the target nuclide. In the case with Tc^{99} this problem doesn't appear, since during the irradiation a short-lived Tc^{100} isotope and two stable ruthenium isotopes are produced. The calculation results are presented in table III.

Table III. Different isotope contribution into activity after irradiation of Tc^{99}, Cu/kg

Isotope	Loading	Discharge	After cooling, year		
			3	100	1000
Tc^{99}	17,05	3,5	3,5	3,5	3,5
Tc^{100}	-	5,26+5	-	-	-
Total	17,05	5,26+5	3,5	3,5	3,5

Thus, the ID activity will be defined by Tc^{99} both before and after irradiation.

The results presented indicate a rather high efficiency of technetium transmutation in ID with a moderator. However the required transmutation efficiency should be determined from economical standpoint. Small technetium transmuted quantity in an irradiation cycle, even for a rather high transmutation rate, will require much more cycles of irradiated target processing, which increases irretrievable losses. Larger transmutation volumes, but with low transmutation rate, require either a larger number of IDs in the reactor or an increase in reactor number, which should be loaded by IDs.

3.1.2. The effect moderator in the blanket on core neutronic parameters.

We will consider the effect of a moderator in the axial blanket on the core parameters for Tc^{99} transmutation in axial blanket.

First of all, we note that moderator location in the immediate vicinity of the core leads to that thermal neutrons produced in the moderator reentering the core, increase sharply the fission number and, therefore, the power in the nearest core layers. It is evident from Fig.5, which shows an axial power field in the most fuel pin of the core.

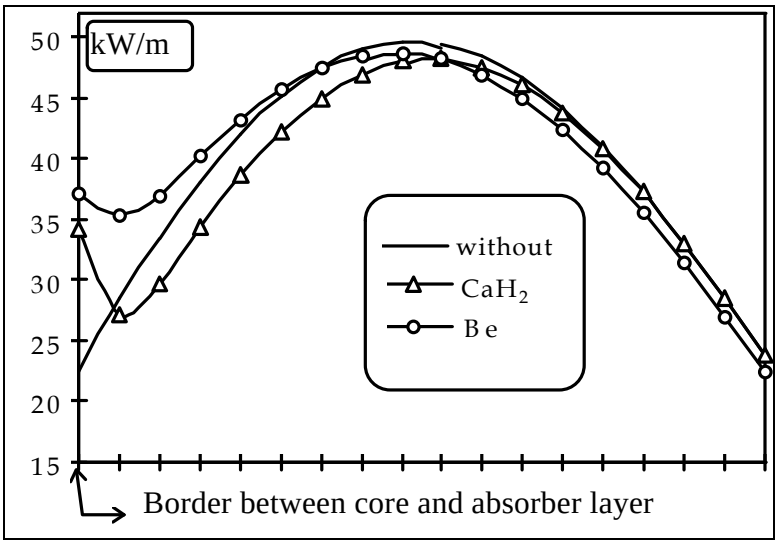

Fig.5. Axial power distribution for various moderators in axial blanket.

It is necessary in this case to pay attention to the fact that the use of hydrated moderators leads to a very sharp power increase in the region adjacent the blanket. The power at the boundary between the core and the blanket increases on 80%.

Even greater power increase is observed when using a beryllium moderator, although in this case the power increase takes place smoothly along a height of the lower core half. The question which one of these cases is more dangerous from the standpoint of reactor safety requires the performance of detailed thermalhydraulic calculations.

However, the effect of moderator on the power distribution can be essentially decreased by introduction of an absorber layer between the core and the blanket with a moderator. Figs. 5 and 6 present axial power distributions when introducing a layer of different type absorber.

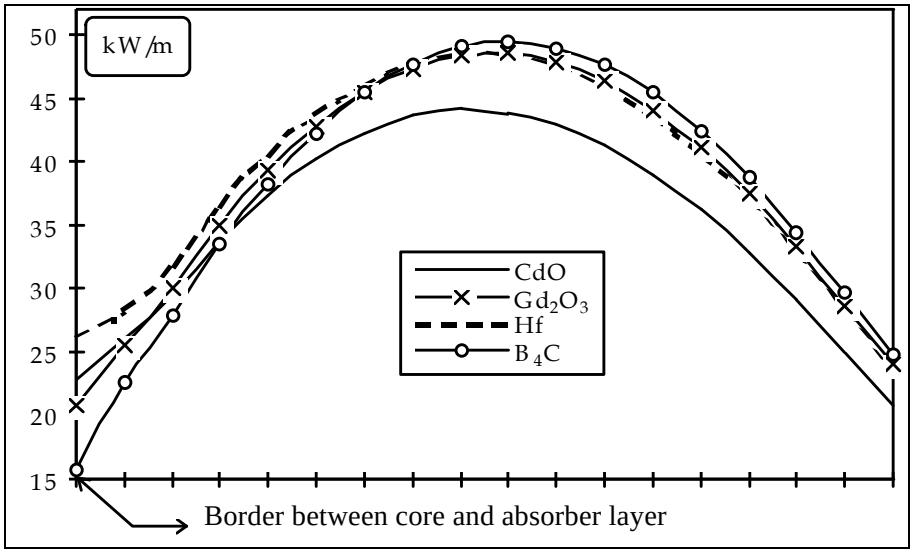

Fig.6. Axial power distribution for various absorber in layer between core and axial blanket (moderator CaH_2)

These distributions show that the best results is achieved when using an absorbing layer with cadmium oxide. In this case not only the power at the boundary decreases, but a maximum power value decreases as well. We note that when using beryllium as a moderator, the effect of absorbers on the power fields is observed only in the immediate vicinity of the blanket.

We consider further the effect of absorber introduction on transmutation efficiency and SVRE value. The calculation results are presented in table IV.

Table IV. The effect of absorber on transmutation efficiency and SVRE value

	moderator CaH_2				
absorber	–	CdO	Gd_2O_3	Hf	B_4C
kg/TW*h	5.29	4.59	3.36	3.79	1.90
SVRE, %Δk/k	-0.169	+0.145	-0.100	-0.102	-0.311
	moderator Be				
absorber	–	CdO	Gd_2O_3	Hf	B_4C
kg/TW*h	3.76	3.56	2.13	2.38	1.79
SVRE, %Δk/k	+0.339	+0.650	+0.171	+0.188	+0.109

The results presented show that the introduction CaH_2 as a moderator decreases SVRE value ~ 0.5%Δk/k as compared with the use of beryllium. The implementation of different absorbers changes the SVRE value in the limits 0.5 %Δk/k.

Thus, the introduction of an absorber solves the power distribution problem and at the same time leads to a decrease in the FP transmutation efficiency. But in case of the CdO use as a absorber the decrease of the transmutation efficiency doesn't exceed 15%.

3.2. IODINE-129 TRANSMUTATION.

I^{129} has the largest among all long-lived wastes half-life period, equal $1.57*10^7$ years, and its production amounts: 0.7 kg/TW*h for fast reactors and 0.66 kg/TW*h for thermal reactors.

When considering I^{129} transmutation, it is necessary to choose the best chemical form for a material containing large quantities of I^{129}. Among different chemical compositions, three compositions are considered as a target materials: BeI_2, NaI and CeI_3, the latter having the largest number of I^{129} nuclei[5]. Table V presents a comparison of I^{129} transmutation efficiency for different moderators and three chemical composition stabilizing iodine

Table V. A comparison of I^{129} transmutation for different chemical compositions.

	BeI_2		NaI		CeI_3	
	kg/TW*h	%/cycle	kg/TW*h	%/cycle	kg/TW*h	%/cycle
MgH_2	0.71	29.3	1.17	28.6	1.25	28.8
ZrH_2	0.71	29.2	1.17	28.6	1.25	28.8

It is necessary to pay attention to that the BeI_2 introduction allows the assurance of maximum transmutation rate, whereas the CeI_3 introduction provides the largest absolute transmutation efficiency. Further the results for BeI_2 composition are presented.

The comparison of I^{129} transmutation efficiency for implementation of different moderators is given in table VI.

Table VI. I^{129} transmutation efficiency when using different type moderators

Moderator	Radial blanket		Axial blanket	
	%/cycle	kg/TW*h	%/cycle	kg/TW*h
CaH_2	65.2	0.32	27.0	0.66
MgH_2	68.7	0.33	29.3	0.71
TiH_2	52.2	0.25	19.8	0.48
CeH_3	67.8	0.33	28.7	0.70
ZrH_2	68.6	0.33	29.2	0.71
Be	50.3	0.24	18.7	0.46
C	41.6	0.20	14.8	0.36
Be_2C	51.3	0.25	19.3	0.47
$NbBe_{17}$	47.7	0.23	17.6	0.43

Because the I^{129} capture cross-section has a clearly defined maximum in the thermal range, the best results are obtained when using hydrated moderators (MgH_2, CeH_3, ZrH_2).

The effect of a moderator volume fraction on the transmutation rate and absolute quantity of iodine transmuted is presented in table VII.

Table VII. The effect of moderator volume fraction on I^{129} transmutation efficiency (radial blanket/axial blanket).

	Moderator volume fraction (ZrH_2), %			
	0	25	70	95
%/cycle	18.2/5.7	35.2/12.0	62.1/25.1	68.6/29.2
kg/TW*h	1.8/2.3	2.7/4.6	1.3/2.6	0.33/0.71

An analysis of activity changing for I^{129} and the products of its irradiation is presented in table VIII.

Table VIII. Different isotope contribution to activity after I^{129} irradiation, Cu/kg

Isotope	Loading	Discharge	After cooling, year		
			3	100	1000
I^{129}	0,18	0,05	0,05	0,05	0,05
I^{130m}	-	62272,73	-	-	-
I^{130}	-	1,15+5	-	-	-
I^{131}	-	68,18	-	-	-
Xe^{131m}	-	595,45	-	-	-
Xe^{133m}	-	1,68	-	-	-
Xe^{133}	-	23,55	-	-	-
Cs^{134m}	-	0,8	0,64	-	-
Cs^{134}	-	1,75	-	-	-
Total	0,18	1,77+5	0,69	0,05	0,05

Contrary to Tc^{99}, an irradiation of I^{129} leads to creation of such (Cs^{134m}), the activity of which during some time (~30 years) will define the activity of irradiated targets. Nevertheless, the irradiation of iodine is also a rather efficient method for decreasing FP radioactivity. However, it is necessary to pay attention to that the irradiation products are also gaseous xenon isotopes (Xe^{130}, Xe^{131} and Xe^{132}), creating a rather high pressure in the targets used.

3.3. PALLADIUM-107 TRANSMUTATION.

Pd^{107} has a half-life period of $6.5*10^6$ years, and its production in the power reactor fuel amounts: 1.54 kg/TW*h for fast reactors and 0.78 kg/TW*h for thermal reactors.

The effect of different moderators on Pd^{107} transmutation efficiency is presented in table IX.

Table IX. Pd^{107} transmutation efficiency when using different moderators

Moderator	Radial blanket		Axial blanket	
	%/cycle	kg/TW*h	%/cycle	kg/TW*h
CaH_2	63.4	1.89	25.9	3.90
MgH_2	64.0	1.91	26.2	3.95
TiH_2	52.9	1.58	20.1	3.02
CeH_3	58.1	1.73	22.8	3.43
ZrH_2	57.1	1.71	22.3	3.36
Be	77.0	2.30	35.4	5.33
C	67.8	2.03	28.7	4.31
Be_2C	77.9	2.32	36.2	5.45
$NbBe_{17}$	74.6	2.23	33.5	5.05

Contrary to the technetium and iodine, the highest efficiency of palladium transmutation is provided by using beryllium containing moderators.

The effect of moderator volume fraction on the transmutation rate and absolute quantity of transmuted palladium is presented in table X.

Table X. The effect of moderator volume fraction onPd^{107} transmutation efficiency.

	Moderator volume fraction (Be_2C), %			
	0	25	70	95
%/cycle	9.6/2.9	22.7/7.2	49.7/18.2	77.9/36.2
kg/TW*h	5.8/8.7	10.6/16.9	6.5/12.0	2.32/5.45

The analysis results for palladium irradiation product activity are presented in table XI.

The results presented show that the irradiated palladium activity increases several orders due to creation of short-lived nuclides (Ag^{110m}, Ag^{110}), which then falls quickly, and in approximately 30 years these targets activity will be again defined by palladium only.

Table XI. Different isotopes contribution to activity after Pd^{107} irradiation, Cu/kg

Isotope	Loading	Discharge	After cooling, year		
			3	100	1000
Pd^{107}	0,51	0,11	0,11	0,11	0,11
Ag^{108}	-	0,33	-	-	-
Pd^{108m}	-	5641,60	-	-	-
Pd^{108}	-	77589,41	-	-	-
Ag^{109m}	-	3,53+5	-	-	-
Ag^{110m}	-	1,31+5	632,00	-	-
Ag^{110}	-	3,30+5	8,85	-	-
Pd^{111m}	-	5,0	-	-	-
Pd^{111}	-	31,76	-	-	-
Ag^{111}	-	32,4	-	-	-
Cd^{111m}	-	102,8	-	-	-
Cd^{113m}	-	0,73	0,63	-	-
Cd^{115}	-	2,87	-	0,01	-
Total	0,51	1,06+6	642,1	0,12	0,11

3.4. CESIUM-135 TRANSMUTATION.

Cs^{135} has a half-life time of $2.3*10^6$ ëåò, and its production in the power reactor fuel amounts: 3.7 kg/TW*h for fast reactors and 1.4 kg/TW*h for thermal reactors.

When analyzing the calculation results for possibility to transmute Cs^{135} in the targets with a moderator, presented in table XII, a conclusion can be made that in case of Cs^{135} irradiation the choice of moderator doesn't play important role, since all moderators give approximately the same results.

Table XII. Cs^{135} transmutation efficiency when using different moderators

Moderator	Radial blanket		Axial blanket	
	%/cycle	kg/TW*h	%/cycle	kg/TW*h
$ÑàÍ_2$	44.7	0.40	16.2	0.72
MgH_2	44.8	0.40	16.2	0.72
TiH_2	33.6	0.30	11.5	0.51
CeH_3	39.0	0.35	13.7	0.61
ZrH_2	40.2	0.36	14.2	0.64
Be	40.3	0.36	14.2	0.64
C	41.3	0.37	14.7	0.65
Be_2C	47.3	0.42	17.4	0.78
$NbBe_{17}$	42.7	0.39	15.3	0.68

Notice that the cesium transmutation rate is somewhat lower as compared with that of the nuclides considered above, which is explained by a lower capture cross-section.

Besides, other cesium isotopes (Cs^{133}, Cs^{137} etc.) are accumulated in power reactor spent fuel and the Cs^{135} fraction is ~10% only. When transmuting cesium without chemical isotope separation, the transmutation efficiency decreases one order due to creation secondary Cs^{135}.

Thus, the results presented show that the issue on advisability Cs^{135} transmutation in reactor conditions remains open.

The transmutation of Zr^{93} ($T_{1/2}=1.53*10^6$ years, production 1.74 kg/TW*h for fast reactors 2.8 kg/TW*h for thermal reactors) was not considered in detail in this report due to large uncertainties in its nuclear data.

The transmutation of such elements Se^{79} ($T_{1/2}=65000$ years) and Sn^{126} ($T_{1/2}=10^5$ years) is not considered because of their low transmutation rate.

The analysis results allows to form the final table XIII.

Table XIII. A comparison of transmutation rate for different FP and their production in power reactors.

Isotope	$T_{1/2}$	Production in reactors, kg/TW*h	Transmutation efficiency, kg/TW*h	
			without moderator	with moderator
Tc^{99}	$2.13*10^5$	3.0/3.2	10.6	5.3
Pd^{107}	$6.5*10^6$	1.54/0.78	5.5	3.0
Cs^{135}	$2.3*10^6$	3.70/1.4	5.1	0.8
I^{129}	$1.57*10^7$	0.70/0.66	5.3	0.7
Se^{79}	$6.5*10^4$	0.03/0.02	0.09	0.01
Sn^{126}	10^5	0.15/0.08	0.2	0.04
Zr^{93}	$1.53*10^6$	1.74/2.8	2.3	1.4

Thus the transmutation of such FPs Tc^{99}, Pd^{107} and I^{129} with the use of moderators in IDs is rather proved, since their transmutation efficiency exceeds their production. The transmutation of Cs^{135}, Zr^{93} in such blanket can turn out to be not useful, and a further optimization of the moderator quantity is necessary. Moreover, Cs^{135} will require its separation from other Cs isotopes produced in
spent fuel.

CONCLUSIONS

The considered in this report method for radioactive wastes (actinides and fission products) transmutation in special irradiation devices containing large moderator quantity, has essential advantages over the homogeneous method foe these wastes recycling.

The results presented have shown that to essentially decrease the americium radiotoxicity, a very high actinide burnup (up to 95% h.a.) should be achieved in these irradiation devices. In this case up to 60 kg of americium per year will be destroyed. Such quantity is accumulated presently in all VVER-100 reactors in Russia.

Majority of long-lived fission products can be also transmuted in such irradiation devices. However, the isotope separation will be needed to increase the transmutation efficiency of Cs^{135} è Zr^{93} isotopes, for example. The major advantage of this fission product utilization concept

consists in decrease of total losses in each step of waste reprocessing. Nevertheless, a serious contradiction should be pointed out between the transmutation rate and absolute quantity of utilized fission products.

The influence of irradiation devices with a moderator on some core neutronic parameters has been considered. And it has been shown that the implementation of absorbing blankets, made from cadmium oxide, will allow an essential decrease in the effect of such devices on the core parameters, decreasing slightly the transmutation efficiency.

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