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CALCULATIONS OF MINOR ACTINIDE TRANSMUTATION IN MOLTEN SALT REACTOR BASED ON MODEL SYSTEM

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Introduction

In Russia and in the whole world, various approaches to MA (Np, Am, and Cm) transmutation have been proposed to solve one of the most important challenges facing nuclear power engineering, namely, radioactive waste disposal.

Currently, Russian scientists have been considering the possibility of MA transmutation in energy reactor units with fast neutron spectrum like BREST [1] and in specific facilities called molten-salt reactors like GSR-S [2]. The second approach presents the following advantages:

- ✓ Elimination of fabrication of fuel pellets and elements which, on one hand, would reduce radiation load on the staff involved in fuel fabrication, and on the other hand, would allow one to use high-level nuclear fuel which, in turn, could be multiply recycled;
- ✓ Comparatively high MA content in fuel composition;
- ✓ Continuous/batch reprocessing of fuel composition would afford operating the reactor with minimum reactivity margin and using a wide range of isotope compositions as fuel loads;
- ✓ Centralized MA burning would become possible in a facility with in-site fuel cycle.

[1] – Ганев И.Х., Лопаткин А.В., Орлов В.В. Гомогенная трансмутация Am, Cm, Np в активной зоне реактора типа БРЕСТ. – Атомная энергия, 2000, т.89, вып.5, с. 355–361.

[2] – Дегтярев А.М., Мясников А.А., Коляскин О.Е. и др. Жидкосолевой подкритический реактор-сжигатель трансплутониевых изотопов. – Там же, 2013, т.114, вып.4, с. 225–232.

Requirements to MA Burner Reactor

- ✓ Elimination of actinide extraction from fuel composition during its reprocessing. In fact, this requirement means implementation of **closing nuclear cycle for all actinides**;
- ✓ Minimization of Pu and U consumption as the major fuel for energy reactor units.

Meeting all these requirements would provide highly efficient MA transmutation in terms of the mass of **nuclei of Np, Am, Cm and their derivatives which would be converted into fission products.** The necessary level of transmutation efficiency is determined by the performance of SNF recycling plant. For example, transmutation efficiency should constitute ~250 kg/year for MA post-combustion in SNF of WWER taking into account the projected performance of ~250 tons/year [3].

In addition to the above requirements, it is reasonable to:

- ✓ Minimize actinide mass in a fuel load using equilibrium operation mode; and
- ✓ Provide the stability of reactor characteristics to possible variations in fuel composition.

[3] – Пятый национальный доклад Российской Федерации о выполнении обязательств, вытекающих из Объединенной конвенции о безопасности обращения с отработавшим топливом и о безопасности обращения с радиоактивными отходами. – М., 2017

Objectives and Tasks

The key parameter when estimating the efficiency of Np, Am, and Cm transmutation in GSR-S involves the characteristics of **equilibrium transmutation mode.** The latter is reached when isotope composition and replenishment fuel mass remain almost constant.

The highest possible transmutation efficiency can be attained when the replenishment fuel will contain only MA salts. Meanwhile, MA mass in the replenishment fuel should be equivalent to the mass of Np, Am, and Cm fissioned nuclei (~320 kg/year for GSR-S thermal power of 1 GW).

The main objective of this work is to determine the main characteristics of equilibrium MA transmutation mode that meet the requirements to GSR-S using an ideal model system as an example.

The tasks are as follows***:***

- ✓ To select a basic model system which would meet the requirements to GSR-S and provide the highest possible MA transmutation efficiency;
- ✓ To study the stability of basic model to the composition of replenishment fuel; and
- ✓ To evaluate feasibility to provide criticality of model system when reaching equilibrium transmutation mode.

Task Formulation

For an ideal model we consider a sphere filled with fuel composition having neutron specular reflection as boundary conditions.

- ✓ Salt solvent: FLiNaK eutectic salt [4] having the following molar composition, %: 46,5LiF–11,5NaF–42KF;
- ✓ Fuel composition temperature: 650°C;
- ✓ Energy deposition density: 100 kW/l;
- ✓ Campaign duration: 300 effective days;
- ✓ After each campaign the whole fuel composition is subjected to reprocessing: all fission products are extracted, and Np, Am, and Cm fluorides are added as replenishment fuel .
- ✓ Isotope compositions of plutonium and minor actinides are obtained using PRIZMA+RISK code [8, 9] corresponding to SNF from WWER-1000 with burning rate of 35 GWt*days/tons and fuel cooling of 10 years.

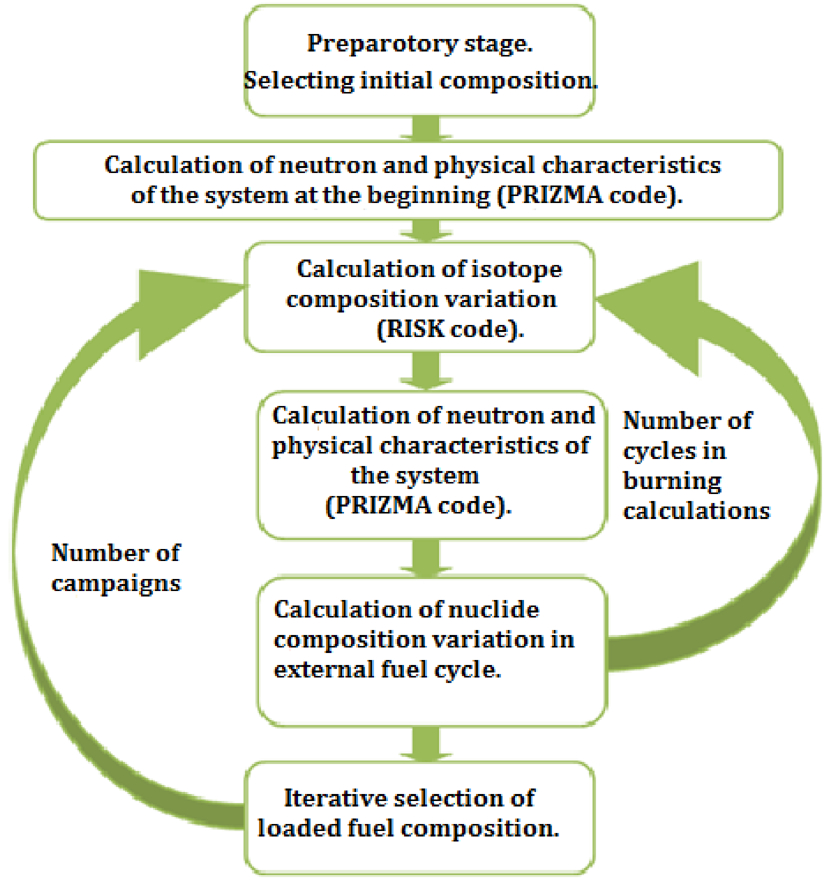
[4] – Лизин А.А., Томилин С.В, Гневашов О.Е., и др., Растворимость PuF_3, AmF_3, CeF_3, NdF_3 в расплаве LiF-NaF-KF // Атомная энергия. – 2013. – Т. 115. – Вып. 1. – С. 11-16.

[5] – Кандиев Я.З. Оценка эффектов малых возмущений в многовариантных расчётах по программе ПРИЗМА-Д // Атомная энергия. – 2005. – Т. 99. – Вып. 3, С. 203 – 210.

[6] – Модестов Д.Г. Программа решения задач ядерной кинетики РИСК-2014 // Препринт РФЯЦ–ВНИИТФ №243. – 2014.

Calculation Technique

Evolution of fuel isotope composition had been calculated during several campaigns with overloads being simulated at the end of each campaign. During overloading, fission products were extracted, and replenishment fuel was added. The amount of actinides in replenishment fuel was determined based on the condition of conserving the mole fraction of actinoids in fuel composition at the beginning of each campaign. The calculation had been performed up to the moment of reaching equilibrium mode of Np, Am, and Cm transmutation.



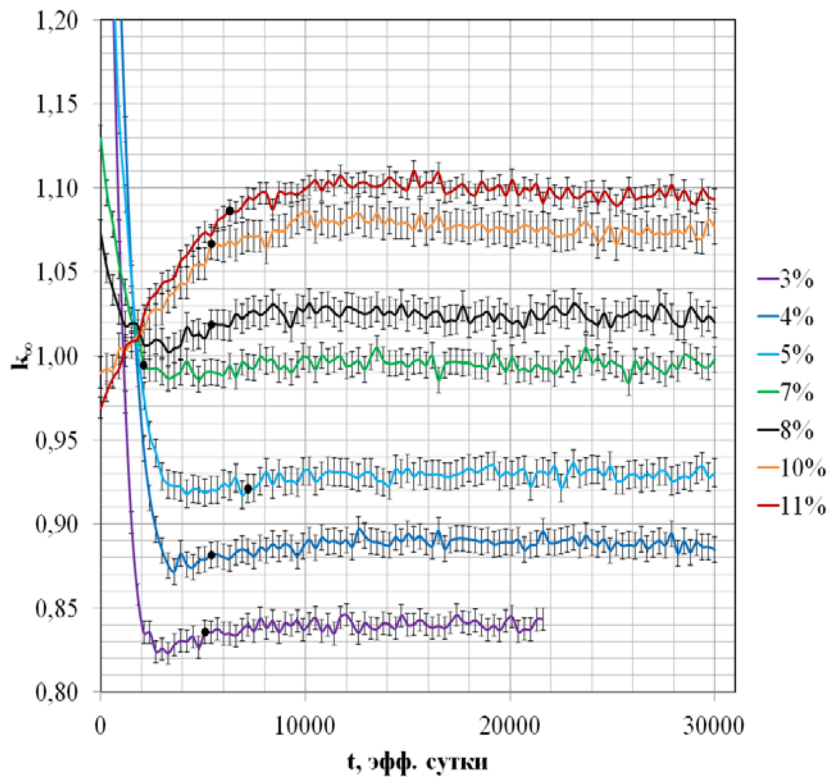
The results below were obtained from calculations using PRIZMA+RISK code and nuclear constants based on ENDF-B-VII library of estimated nuclear data.

Selection of Basic Model System

The parameters of equilibrium MA transmutation mode are independent on the initial composition of actinides in the core and are determined by their mole fraction in the fuel composition and nuclide composition of the replenished fuel.

System criticality at the beginning of the campaign in equilibrium MA transmutation mode.

Mole Fraction of Actinides, %	3	4	5	7	8	10	11
Кef	0.838	0.884	0.924	0.993	1.024	1.079	1.099
σ, 10^{-3}	3.3	3.8	4.1	3.8	4.4	4.7	3.1



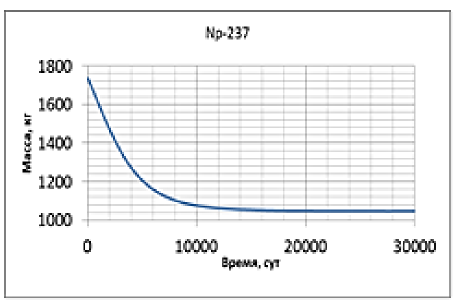
For basic model system we selected a model having **8% of actinide mole fraction in fuel composition**. In this case, **the highest possible transmutation efficiency** is attained, **the system criticality** being equal to **~1,02.**

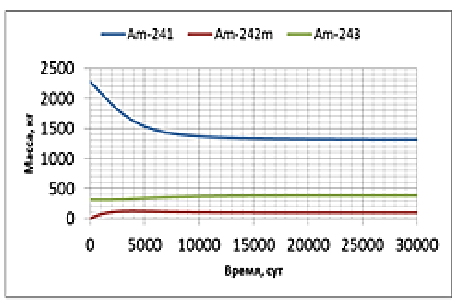
Characteristics of Equilibrium MA Transmutation Mode for Basic Model System

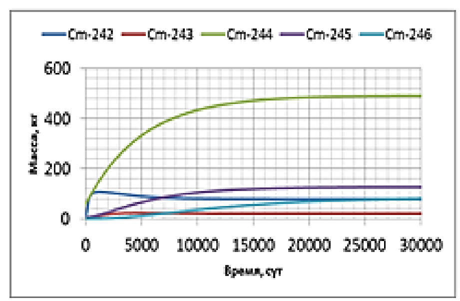
Equilibrium composition of the active core per 1 GW of thermal power

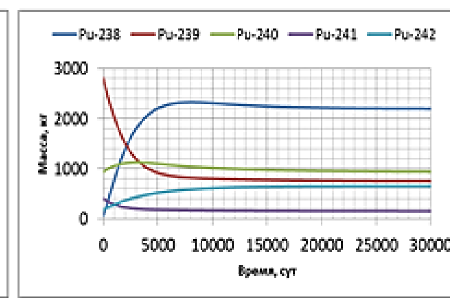
Isotope Composition in the Active Core, kg							
Isotope	Mass	Isotope	Mass	Isotope	Mass	Elements	Mass
^{237}Np	1047	^{244}Cm	491	^{241}Pu	152,	Pu	4693
^{241}Am	1312	^{245}Cm	127	^{242}Pu	652	U	437,
^{242m}Am	99	^{246}Cm	81	^{234}U	298	MA	3658
^{243}Am	379	^{238}Pu	2196	^{235}U	70	Actinides	8788
^{242}Cm	78	^{239}Pu	752	^{236}U	70	MA/Ac	0.4
^{243}Cm	20	^{240}Pu	941	^{238}U	0.1	Pu/MA	1.3

Chemical composition of the active core, % (in the approximation assuming that Pu, Am, and Cm are present in the fuel composition in the form of trifluorides, and U and Np - in the form of tetrafluoride)					
Compound	Mole Fraction	Compound	Mole Fraction	Compound	Mole Fraction
NpF_4	0.96	CmF_3	0.73	UF_4	0.41
AmF_3	1.62	PuF_3	4.28	FLiNaK	92









Study of Basic Model System Stability to the Composition of Replenishment Fuel

Five compositions of replenishment fuel were studied consisting of ^{241}Am and ^{237}Np with different mole ratio:
(1) AmF_3, (2) $25NpF_3$+$75AmF_3$
(3) $50NpF_3$+$50AmF_3$,
(4) $75NpF_3$+$25AmF_3$, and (5) NpF_3.
The criticality of the system changes within ~1% range that proves **the stability** of neutron and physical characteristics of the system to the replenishment fuel composition, the **criticality being mainly influenced by the mole fraction of actinides in the fuel composition**.

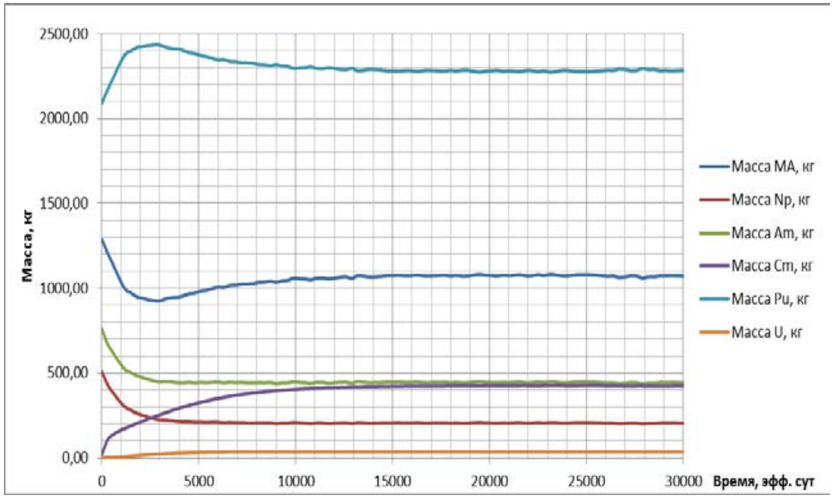
System criticality at the beginning of the campaign and actinide composition in the fuel at the end of the campaign under system thermal power of 1 GW in equilibrium transmutation mode.

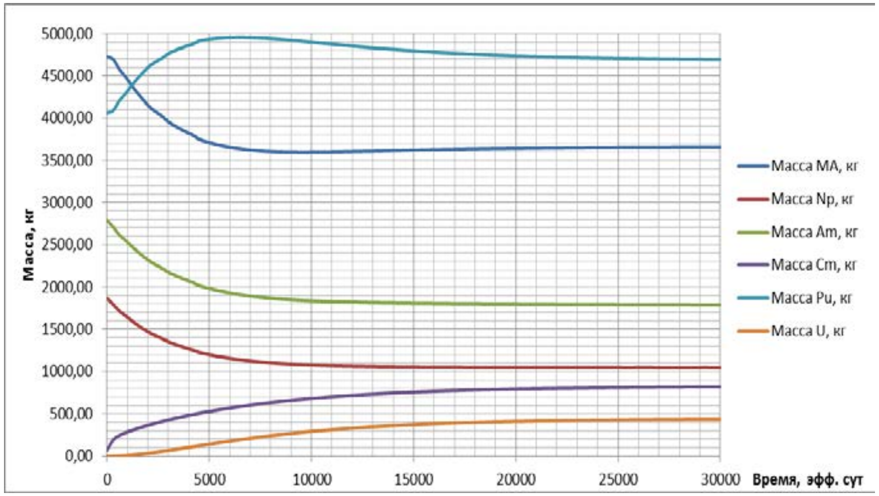
Variant of MA Composition	Basic Scenario	AmF_3	$25NpF_3$+ $75AmF_3$	$50NpF_3$+ $50AmF_3$	$75NpF_3$ + $25AmF_3$	NpF_3
Кэф	1.024	1.012	1.014	1.017	1.019	1.02
U	438	388	443	500	559	620
^{234}U	298	264	302	342	384	427
^{235}U	70	62	70	79	89	98
^{236}U	70	63	71	79	87	95
Pu	4694	4682	4816	4954	5094	5237
^{238}Pu	2197	1958	2220	2490	2768	3053
^{239}Pu	752	673	759	848	938	1032
^{240}Pu	941	807	828	848	867	884
^{241}Pu	152	132	134	137	139	142
^{242}Pu	652	1112	874	631	381	126
MA	3349	3749	3536	3314	3090	2860
^{237}Np	925	38	671	1328	2010	2716
^{241}Am	1152	2451	1882	1291	678	43
^{242m}Am	99	184	142	97	51	3
^{243}Am	357	321	253	183	109	34
^{242}Cm	79	147	112	76	40	3
^{243}Cm	20	37	28	19	10	1
^{244}Cm	487	393	308	220	131	40
^{245}Cm	126	101	79	57	34	11
^{246}Cm	81	61	48	34	21	7
^{247}Cm	14	10	8	6	4	1

Feasibility Study to Provide Criticality when Reaching Equilibrium MA Transmutation Mode -1

In real GSR-S operating conditions its criticality at the stage of reaching equilibrium mode of MA transmutation could be ensured by adding Pu salts in the fuel composition.

Calculations involving selection of fuel composition were performed to study the feasibility to provide GSR-S criticality. The initial composition and replenishment fuel composition were selected after each campaign by identifying the mole fraction of Pu fluoride in actinide mixture ensuring system criticality of ~1,01. At that, the mole fraction of all actinide fluorides in the fuel salt was maintained constant. The scenarios with actinide mole fraction of 3% (left) … to 8% (right) in the fuel composition were considered.





Feasibility Study to Provide Criticality when Reaching Equilibrium MA Transmutation Mode – 2

Mole Fraction of Actinide Fluorides in the Fuel Composition, %	3	4	5	7	8
Initial/Equilibrium Loading (at the End of the Campaign), kg					
U	0/34	0/76	0/138	0/327	0/438
^{234}U	0/20	0/46	0/87	0/217	0/298
^{235}U	0/6	0/13	0/24	0/53	0/70
^{236}U	0/7	0/16	0/27	0/56	0/70
Pu	2089/2132	2604/2796	3057/3361	3758/4264	4060/4694
^{238}Pu	28/462	35/750	42/1086	51/1854	55/2197
^{239}Pu	1337/382	1666/511	1956/602	2404/711	2598/752
^{240}Pu	446/736	557/897	653/974	803/938	868/941
^{241}Pu	191/194	238/208	280/205	344/165	371/152
^{242}Pu	86/358	108/431	127/493	155/595	168/652
MA	1290/916	1878/1316	2518/1778	3970/2833	4728/3349
^{237}Np	509/143	742/250	995/390	1568/743	1868/925
^{241}Am	669/204	974/342	1305/517	2058/937	2451/1152
^{242m}Am	0/20	0/32	0/47	0/82	0/99
^{243}Am	93/128	135/169	181/213	285/308	340/357
^{242}Cm	0/40	0/50	0/59	0/74	0/79
^{243}Cm	0,2/13	0,3/16	0,4/17	0,7/20	0,8/20
^{244}Cm	17/231	25/289	34/344	53/446	64/487
^{245}Cm	1/72	2/87	3/99	4/120	5/126
^{246}Cm	0,08/46	0,1/57	0,2/67	0,3/78	0,3/81
^{247}Cm	0/11	0/13	0/14	0/14	0/14
^{248}Cm	0/9	0/11	0/11	0/10	0/9
Consumption (Transmutation) of MA and Pu per Campaign in Equilibrium Operation Mode, kg					
MA	156	192	229	292	310
Pu	151	116	80	18	0

Conclusions – 1

- ***The system criticality in equilibrium MA transmutation mode is determined by the content of actinides in the fuel composition***.
- The system criticality weakly depends on MA composition in the replenishment fuel that proves the stability of neutron and physical characteristics of the system.
- As for ***the basic scenario*** characterized by ***8%***-mole fraction of actinides in the fuel composition, it was found out that:
  - ✓ Model system reaches the equilibrium mode ensuring ***the highest possible transmutation efficiency***: it will be equal to ~320 kg/year for the thermal power of 1 GW. In equilibrium operating mode, the mass of actinides in the fuel composition will constitute ~ 8788 kg: ~4693 kg of Pu, 437 kg of U, 1047 kg of Np, 1790 kg of Am, and 797 kg of Cm. Meanwhile, ***the system converts ~122 kg of Np, 183 kg of Am, and 5 kg of Cm into fission products*** per 300 efficient days***;***
  - ✓ System criticality at the beginning of the campaign equals to ~1,02, and reactivity overshoot does not exceed 1%;
  - ✓ ***Comparatively low plutonium consumption*** is attained when reaching equilibrium operating mode (initial loading of ~4060 kg, and additional replenishment of ~ 220 during 50 years of operation per 1 GW of thermal power). The time to reach this mode will constitute ~7 years, and during this period of time **the system will consume ~24 kg of Pu, 112 kg of Np, 168 kg of Am, and 4 kg of Cm on average per *300 efficient days* per 1 GW of thermal power*.***

Conclusions – 2

- To ensure transmutation efficiency of ~250 kg/year in equilibrium operating mode, the thermal power of the system with 8%-mole fraction of actinides in the fuel composition should constitute ~ 800 MW for active core volume of 8 m^3 and average fuel rating of 100 kW/l.
- With ~3%-mole fraction of actinides in the fuel composition that corresponds to their solubility limit in 73LiF-27BeF_2 and 15LiF-58NaF-27BeF_2 salts, the efficiency of ***Np, Am, and Cm transmutation*** in equilibrium mode ***will constitute ~156 kg (62 kg of Np, 92 kg of Am, and 2 kg of Cm) per 300 efficient days and per 1 GW of thermal power with additional replenishment with plutonium of ~151 кг as appropriate***. In this case, equilibrium transmutation mode will be reached in ~5 years. ***During this period of time the system will consume 161 kg of Pu, 58 kg of Np, 86 kg of Am, and 2 kg of Cm on average per 300 efficient days and per 1 GW of thermal power***.

In the future, it is planned to develop a reactor basic circuit arrangement, to determine the most efficient operation modes, to estimate reactivity coefficients, and to formulate the proposals on CPS design.

The main challenges involve the lack of full-scope information on the solubility of actinide salts in the carrier, and the uncertainty in the selection of structural materials. It is supposed that such data will be obtained under the efforts on GSR-S design.

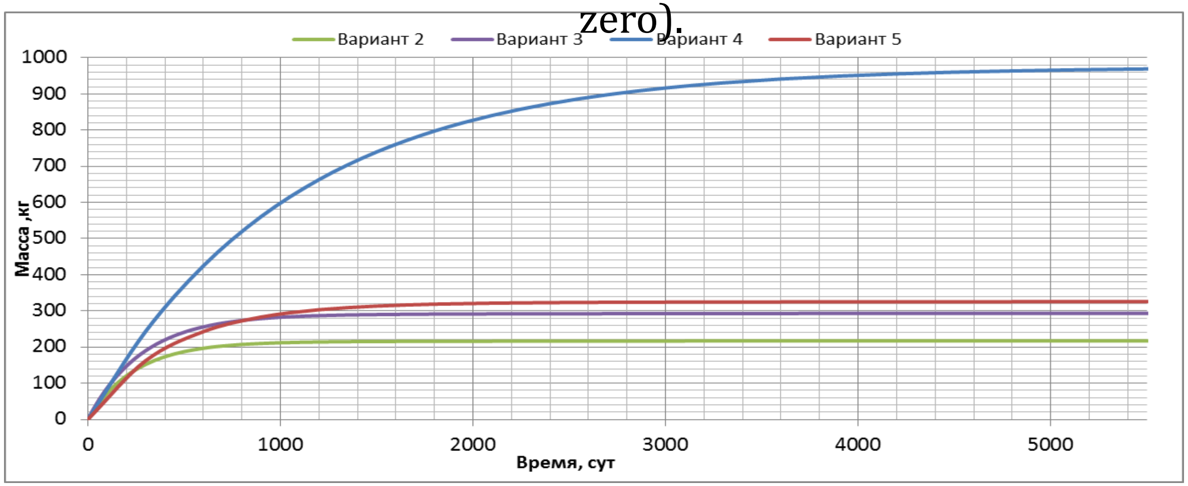
Thank you for your attention!

Study of GSR-S Stability. Reprocessing Mode – 1

The following modes of fuel composition reprocessing were considered:

- ✓ During a campaign 1/k- part of a fuel composition was reprocessed k times with equal time intervals, T/k (T = 300 efficient days). For the basic scenario k=1, scenarios with k = 3 (scenario 2) and k=10 (scenario 3) were additionally studied. In this case with equilibrium mode, the mass of fission products in the active core will constitute m_{MAmax} at the end of the campaign, and $m_{MA}*(1-k^{-1})$ at the beginning of the campaign.
- ✓ Having a constant campaign duration, T = 300 efficiency days, 1/k- part of a fuel composition is reprocessed at the end of the campaign. For the basic scenario k=1, scenarios with k = 4 (scenario 4) and k=2 (scenario 5) were additionally studied. Under the given conditions and in equilibrium mode, the mass of fission products in the active core will constitute $m_{MAmax}*k$ at the end of the campaign, and $m_{MAmax}*(k-1)$ at the beginning of the campaign.

Mass of fission products in the active core at the beginning of the campaign in equilibrium MA transmutation mode vs. time (the mass of fission products in the basic scenario is equal to zero).



Study of GSR-S Stability. Reprocessing Mode – 2

System criticality in equilibrium MA transmutation mode for various reprocessing modes.

Режим переработки	1	2	3	4	5
K_{ef}	1.024	1.026	1.029	0.993	1.013
σ, 10^{-3}	3.7	3.4	3.2	2.6	3.3

Reducing a part of the reprocessed fuel and correspondingly increasing reprocessing rate do not have any substantial influence on the nuclide composition and system criticality in equilibrium MA transmutation mode (scenarios 1, 2, and 3). This means that the rate of fuel reprocessing with a fixed annual volume of reprocessed fuel does not almost have any impact on neutron and physical characteristics of the system and can be selected, in particular, based on technological capabilities.

Reducing the fraction of reprocessed fuel at the fixed reprocessing rate results in accumulation of fission products in the active core and in degradation of K_{ef} of the system in equilibrium MA transmutation mode that place lower limits on the fraction of reprocessed fuel at the given campaign duration (it is equivalent to placing upper limits on the campaign duration with the given volume of reprocessed fuel).

Study of GSR-S Stability. Energy Deposition Density and Salt Solvent Composition.

Energy depositions densities in GSR-S active core:

- ✓ 50 kW/l;
- ✓ 100 kW/l (basic scenario);
- ✓ 200 kW/l .

Energy Deposition Density	50 kW/l	100 kW/l	200 kW/l
Kef	1.004	1.024	1.031
σ, 10^{-3}	3.1	3.7	3.1
Φ, $\cdot 10^{15}$ sm^{-2} s^{-1}	0.82	1.62	3.2
E_{ϕ}, keV	395	401	404

Salt Solvent	FLiNaK	FLiBe (73LiF-27BeF_2)	FLIBeNa (15LiF-58NaF-27BeF_2)	67KCl-33$MgCl_2$
Kef	1.024	1.018	1.015	1.023
σ, 10^{-3}	3.7	4.2	3.7	3.6
E_{ϕ}, keV	401	389	420	446

There is only a slight difference both in K_{ef} (~1%) and in average neutron energy (~10%) for the given salt carriers in equilibrium MA transmutation mode. The mole fraction of actinides in the active core for these salts required to reach the criticality will be almost the same. The selection of salt carrier does not have any substantial impact on the neutron and physical characteristics of the system.

Feasibility Study to Provide Criticality when Reaching Equilibrium MA Transmutation Mode – 3

Mass of replenished plutonium for various mole fractions of actinides in the fuel composition vs. campaign.