# COMPARATIVE ANALYSIS OF MINOR ACTINIDES TRANSMUTATION IN A MSR-BURNER BASED ON

# LiF–NaF–KF AND LiF–BeF2 SALTS

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**Abstract**

In Russia, research is actively underway to develop a specialized molten-salt burner reactor (MSR-burner) of minor actinides (MA) from spent nuclear fuel of power reactors. Two candidate fluoride salts, LiF–BeF2 and LiF–NaF–KF, are considered as the solvent of the reactor fuel components. The purpose of the present paper is to study MA transmutation in the MSR-burner based on selected salts in the equilibrium mode of reactor operation at different volumes of the core. The calculations were performed using the PRISMA+RISK software package developed at the «Russian Federal Nuclear Center – Zababakhin All-Russian Research Institute of Technical Physics ». The LiF–BeF2 salt has a low solubility limit of actinide fluorides, which leads to the need to feed the reactor with a significant amount of Pu and, consequently, to a low efficiency of MA transmutation. By reducing of volume of the active zone increases the consumption of Pu and reduces the efficiency of transmutation. In contrast to LiF–BeF2, LiF–NaF–KF eutectic is characterized by a relatively high solubility of actinide fluorides. For a MSR-burner based on this salt, plutonium is needed mainly for starting; in the equilibrium mode reactor consumes only MA. In this case, the maximum efficiency of MA transmutation can be achieved in a wide range of core volume: from 2 m3 to 30 m3 with a concentration of actinide fluorides from 17 to 10%, mol., respectively.

## INTRODUCTION

Long-lived isotopes of minor actinides (Np, Am, and Cm isotopes) are known to seriously contribute to the long-lived activity and energy release of the spent fuel in light water reactors. IAEA estimations demonstrate that spent fuel in the amount of about 370 thousand tons, including more than 300 tons of minor actinides, was accumulated by 2013 in the world [1]. Many countries (for example, Russia, the USA) are planning to develop nuclear waste storage facilities. However, the half-life for certain isotopes of minor actinides (MA) involves thousands and millions years. Therefore, currently there is no scientific justification that would guarantee safe and non-hazardous geological disposal of these isotopes for the time period comparable to their lifetime. Moreover, such storage facilities are expensive to operate and not all nuclear-powered countries can build these repositories for geographical, political, or other reasons. The conversion of MA into fission products in the neutron flux will significantly reduce the amount of radioactive waste intended for disposal and in the future will allow us to do without deep disposal. Currently in Russia transmutation of minor actinides in a specialized molten salt burner reactor (MSR-burner) is proposed as a possible approach to address this challenge [2 – 4]. Absence of necessity to fabricate fuel pellets, possibility to organize continuous (batch) reprocessing of a fuel composition, capability to achieve high-efficiency transmutation of MA can be considered as obvious advantages of this approach [2].

The purpose of the calculations – is to study the basic regularities of MA transmutation and to determine characteristics of the MSR-burner operational modes in which: (1) reprocessing of the fuel composition extracts only the fission products and (2) feed fuel contains mostly MA, and, therefore, the mass of their isotopes transmuted in the reactor is close to that of fission products extracted from the reactor. In our understanding, MA transmutation is transformation of MA isotope nuclei into fission products and also actinides formed therefrom due to decays and interaction with neutrons. In the article, these reactor operational modes are called optimal

##  CALCULATION PROCEDURE

PRIZMA+RISK code and ENDF/B-VII neutron constants were used to determine basic regularities of MA transmutation in the MSR-burner based on the results of multivariate calculations of neutron-physical characteristics and nuclide kinetics [5 – 7]. In order to modeling reprocessing fuel composition, all fission products were extracted therefrom and the equivalent amount of feed fuel consisting, in the general case, of MA and Pu fluorides was added thereto (reprocessing time was neglected). In this case, concentration of actinides in the fuel composition was maintained the same at every campaign startup and by campaign we shall understand the system operation period needed to reprocessing the fuel composition volume that is equal to the fuel circuit.

If fuel reprocessing is the same for each campaign, then the system asymptotically reaches the equilibrium operational conditions wherein reactor characteristics, including nuclide composition of the fuel, are similarly changing with time during any two campaigns. Since continuous operation presupposes close-to-equilibrium conditions, the paper is mainly focused on reactor characteristics in the equilibrium operational conditions.

## OPTIMAL OPERATIONAL CONDITIONS FOR MINOR ACTINIDES TRANSMUTATION IN THE MSR-burner

Basic regularities of MA transmutation were determined for the model system representing the infinite medium [8]. This system prevents effect of the geometry of the reactor core and its design elements on the neutron-physical characteristics. This allows identification of common regularities conditioned by the character of neutron-physical and nuclear-physical processes in the core as these regularities are characteristic for different versions of the MSR-burner.

Our computations assumed that temperature of the fuel composition is 6500С [9], specific thermal power in the system is 100 kW/l [10], and campaign duration is 300 effective days. Isotopic composition of Pu and MA in the startup loading is just the same as their composition in the spent fuel from WWER-1000 reactor with the 35 GW∙day/t burnup and 10 years cooling. This composition was calculated with the help of PIZMA+RISK code. Eutectics LiF–NaF–KF (molar composition, %: 46.5LiF–11.5NaF–42KF) stated in [11, 12] to have a rather high solubility of actinides (more than 20 %, mole PuF3 at 6500C) is considered as the salt solvent. Density of the fuel composition was calculated as density of the mechanically mixed salt carrier and actinide fluorides. Statistical calculation error $K\_{eff}$is less than 0.4%.

Reactor operational conditions are optimal when: (1) only fission products are extracted from the fuel composition due to reprocessing (idealized variant of reprocessing); (2) feed fuel contains only MA; (3) reactor criticality during the campaign $K\_{eff}=1.01…1.03$. Optimal operational conditions require a specific concentration of actinides in the fuel composition [8]. A number of initial concentrations of actinide fluorides $ν\_{A}$ were tried until the optimal concentration was determined (Fig. 1, statistical calculation error $K\_{eff}$ is given to be 2σ.). From the figure, it is obvious that the optimal equilibrium conditions of MA transmutation take place at $ν\_{A}$ ~8 %, mole. In this case, we have the maximum possible efficiency of MA transmutation, i.e. ~0.31 tons at thermal power of 1 GW per the 300 effective days campaign (Table 1).



*FIG. 1.* $K\_{eff}$ *of the system at the campaign startup versus time at different concentrations of actinides in the fuel composition (%, mole)*

TABLE 1. EQUILIBRIUM COMPOSITION OF FUEL AT THE CAMPAIGN STARTUP FOR THE MODEL SYSTEM WITH ACTINIDE CONCENTRATION 8 %, MOLE

|  |  |  |  |
| --- | --- | --- | --- |
| Element (fluoride) | $ν\_{i}$, %, mole | Isotopic composition, % wt | $m\_{i}$, tons |
| Pu (PuF3)  | 4.28 | 238Pu46.8239Pu16240Pu20.1241Pu3.2242Pu13.9 | 4.69 |
| U (UF4)  | 0.41 | 234U68.1235U15.9236U16 | 0.44 |
| Np (NpF4)  | 0.96 | 237Np | 1.05 |
| Am (AmF3)  | 1.62 | 241Am73.3242mAm5.5243Am21.2 | 1.79 |
| Cm (CmF3)  | 0.73 | 242Cm9.6243Cm2.4244Cm59.9245Cm15.5246Cm9.8247Cm1.7248Cm1.1 | 0.82 |
| MA  | 3.31 | Np28.6Am49Cm22.4 | 3.66 |
| Actinides  | 8.0 | U5Pu53.4MA41.6 | 8.79 |

In the table: $m\_{i}$– is mass of the appropriate element, $ν\_{i}$ – is concentrations of actinide fluorides.

## criticalITY maintenance when it reaches equilibrium conditions of MA transmutation

Under actual operating conditions, the MSR-burner can go critical at the stage when it reaches the equilibrium conditions if plutonium salts are added into the fuel composition. In order to estimate how MA transmutation characteristics are changing with the account of time needed to reach the equilibrium conditions, we performed calculations for several variants with different concentrations of actinides $ν\_{A}$(both higher and lower than the optimal one) needed for the system to reach criticality $K\_{eff}$~1.01 (Table 2, Fig. 2 and 3).



*FIG. 2. Variation of plutonium mass in the feed fuel for the 1 GW (th) of the system*

TABLE 2. CHARACTERISTICS OF MODEL SYSTEM IN THE EQUILIBRIUM OPERATIONAL CONDITIONS AT THE CAMPAIGN END WITH DIFFERENT CONCENTRATIONS OF ACTINIDE FLUORIDES

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| Characteristics | 3%, mole | 5%, mole | 8%, mole | 15%, mole | 30%, mole |
| $<E\_{n}>$, MeV | 0.29 | 0.34 | 0.40 | 0.52 | 0.68 |
| $m\_{U}$, | 0.03 | 0.14 | 0.44 | 0.68 | 0.45 |
| $$m\_{Pu}$$ | 2.13 | 3.36 | 4.69 | 5.16 | 2.45 |
| $$m\_{MA}$$ | 0.92 | 1.78 | 3.36 | 9.78 | 26.54 |
| $$m\_{Np}$$ | 0.14 | 0.39 | 0.93 | 3.49 | 10.52 |
| $$m\_{Am}$$ | 0.35 | 0.78 | 1.61 | 5.21 | 14.58 |
| $$m\_{Cm}$$ | 0.42 | 0.61 | 0.82 | 1.08 | 1.44 |

In the table: $<E\_{n}>$ – is neutron mean energy averaged over the neutron flux density, m – is mass of the appropriate element, tons.



*FIG. 3. Variation of actinides mass at the 1 GW (th) of the system at the campaign startup for* $ν\_{A}$ *= 8 %, mole*

The above results allow us to estimate the time needed to reach the equilibrium conditions if this time is assumed to be the period between the reactor startup and the instant after which the reactor consumes and produces the same amount of actinides. For the considered variants, the time needed to reach the equilibrium conditions falls within 5…10 year (Fig. 2).

When the actinides concentration is below the optimal one, it is necessary to add reactor-grade plutonium into the feed fuel and this leads to decrease in the mass of MA undergoing transmutation (Table 3). Plutonium consumption decreases with the increase of actinides concentration and when this concentration equals the optimal one, the reactor consumes only MA. When the actinides concentration is above the optimal one, the mass of MA undergoing transmutation is maximal. At the same time part of fuel with a high content of 238Pu isotope must be extracted. As the MSR-burner functions as a specialized facility for radioactive wastes conversion through MA transmutation, accumulation of significant amounts of highly active isotopes of actinides while in operation seems unacceptable.

TABLE 3. MASS OF LOADED AND TRANSMUTED ACTINIDES (TONS) IN THE EQUILIBRIUM MODE AT THE 1 GW (TH) FOR CAMPAIGN DURATION 300 EFFECTIVE DAYS

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| $ν\_{A}$, %, mole | 3 | 5 | 8 | 15 | 30 |
| $$m\_{MA}$$ | 0.16 | 0.23 | 0.31 | 0.54 | 0.80 |
| $$m\_{Pu}^{load}$$ | 0.15 | 0.08 | 0 | 0 | 0 |
| $$m\_{Pu}^{unload}$$ | 0 | 0 | 0 | 0.23 | 0.49 |
| $$Δm\_{TMA}$$ | 0.16 | 0.23 | 0.31 | 0.31 | 0.31 |

In the table: $m\_{MA}$ и $m\_{Pu}^{load}$ – MA and Pu mass in the feed fuel, respectively; $m\_{Pu}^{unload}$ – mass of unloading Pu; $Δm\_{TMA}$ – mass of transmuted MA.

If the reactor operation amounts to 50 campaigns, then for the variant with $ν\_{A}$= 3 %, mole, the total loading, including the startup one, is ~8.9 t of MA, ~9.5 t of reactor-grade plutonium. In this case, ~8 t of MA undergoes transmutation. In the case of the optimal concentration $ν\_{A}$= 8%, twice as large MA undergo transmutation, i.e. ~16 t when their total loading is 20 t and plutonium consumption is ~4.3 t. For the variant with $ν\_{A}$= 15%, during the same operation time with the same amount of transmuted MA as in conditions with the optimal concentration of actinides (~16 t), it will be necessary to load ~5 t of plutonium and ~36 t of MA. In this case, ~10 t of plutonium containing ~6 t of 238Pu will be accumulated as a result of fuel composition recycling. Even greater amount of this plutonium will be accumulated in the variant with $ν\_{A}$= 30%.

## Characteristics of MA TRANSMUTATION IN A MSR-BURNER BASED ON LiF–NaF–KF AND LiF–BeF2 SALTS

At the comparative analysis of a transmutation MA in MSR-burner based on LiF-NaF-KF and LiF-BeF2 salts the system representing a fuel composition enclosed in a metal casing made of the in the metal case from alloy HN80MTY [13] was considered. The core volume varied from that of the research (experimental) molten-salt burner reactor, i.e. 0.5 m3 up to 30 m3, i.e. MOSART reactor core volume [3]. Our calculations used the following input data: (1) thickness of the reactor vessel walls is 5.5 cm, (2) vessel density is 8000 kg/m3, (3) the vessel diameter – height ratio is 1, (4) salt solvent is eutectic LiF–NaF–KF, (5) nuclide compositions of actinides correspond to the calculated compositions of spent fuel from VVER-1000 with the 50 GW∙day/t burnup and the 9 years cooling [12], (6) temperature of the fuel composition and the vessel is 6500С, (7) specific thermal power is 100 kW/l.

For a quantitative assessment of transmutation МА in MSR-burner expedient to introduce concept of efficiency of transmutation МА as the coefficient characterizing balance between burning МА, Pu consumption and accumulation of a radioactive waste (except fission products):

$K\_{MA}=\frac{M\_{MA}\left(load\right)-M\_{A}(unload)}{M\_{A}(load)} ,$ (1)

where $M\_{A}(load)$ – mass of loading actinides, $M\_{MA}\left(load\right)$ – mass of loading MA, $M\_{A}(unload)$ – mass of unloading actinides, not returned subsequently in a fuel circuit of the reactor, including losses at reprocessing and a fabrication. The numerator (1) represents a decrease actinides, not suitable to use in solid-fuel power reactors. In the absence of an unloading actinides $K\_{MA}$ defines fraction МА from total loaded actinides, including plutonium. In an optimum mode of transmutation МА and operation in this mode long time efficiency of a transmutation $K\_{MA} (t\rightarrow \infty )$ asymptotically converge to one.

Concentration of actinide fluorides in fuel composition for the chosen volume of an active zone ($V\_{az}$) was defined in view of their limiting solubility such that to provide maximum asymptotically value of efficiency of transmutation МА. Variants with unloading actinides from the reactor were not considered ($M\_{A}\left(unload\right)=0$). Calculation data are given in Table 4 and Fig. 4.

TABLE 4. Characteristics of MA TRANSMUTATION IN A MSR-BURNER BASED ON LiF–NaF–KF AND LiF–BeF2 SALTS

|  |  |
| --- | --- |
| Characteristics | Variant |
| $V\_{az}$, m3 | 30 | 15 | 8 | 4 | 2 | 1 |
| $M\_{A}(load)$, kg | 935 | 468 | 250 | 126 | 63 | 32 |
| LiF–NaF–KF |
| $ν\_{A}$, %, mole | 10 | 11 | 12 | 14 | 17 | 20 |
| $K\_{MA}$(1) | 0.53 | 0.53 | 0.53 | 0.56 | 0.6 | 0.6 |
| $K\_{MA}$(10) | 0.59 | 0.59 | 0.59 | 0.60 | 0.63 | 0.63 |
| $K\_{MA}$ (20) | 0.66 | 0.65 | 0.65 | 0.65 | 0.67 | 0.66 |
| $K\_{MA}$ (50) | 0.78 | 0.77 | 0.75 | 0.75 | 0.76 | 0.74 |
| $K\_{MA}$ (100) | 0.86 | 0.85 | 0.83 | 0.83 | 0.84 | 0.81 |
| $$K\_{MA} (t\rightarrow \infty )$$ | 1 | 1 | 1 | 1 | 1 | 1 |
| LiF–BeF2 |
| $ν\_{A}$, %, mole | 3 | 3 | 3 | 3 | 3 | 3 |
| $K\_{MA}$ (1) | 0.35 | 0.34 | 0.32 | 0.29 | 0.24 | 0.16 |
| $K\_{MA}$ (10) | 0.37 | 0.34 | 0.31 | 0.25 | 0.16 | –\* |
| $K\_{MA}$ (20) | 0.39 | 0.36 | 0.31 | 0.23 | 0.13 | –\* |
| $K\_{MA}$ (50) | 0.42 | 0.37 | 0.30 | 0.20 | 0.07 | –\* |
| $K\_{MA}$ (100) | 0.43 | 0.37 | 0.30 | 0.18 | 0.04 | –\* |
| $$K\_{MA} (t\rightarrow \infty )$$ | 0.45 | 0.38 | 0.30 | 0.17 | 0 | –\* |
| \*at $ν\_{A}$ = 3%, mole in the fuel composition based on LiF–BeF2, $V\_{az}$ = 1 м3 and specific thermal power 100 kW/l after the first campaign at feed only Pu $K\_{eff}<1$. |

In the table: $K\_{MA}(N)$, where N – is campaign number



*FIG. 4. Variation of* $K\_{MA}$ *for MSR-burner based on LiF-NaF-KF with volume of an active zone 30 m3 (1) and LiF-BeF2 with volumes 30 (2), 8 (3) and 2 м3 (4)*

The above results allow us to draw following conclusions:

1. realization of an optimum mode of transmutation МА in MSR-burner based on of LiF-NaF-KF is possible at concentration of actinide fluorides in a fuel composition in a range 10 … 17%, mole. This concentration below their solubility limit in LiF-NaF-KF at temperatures above 6500С;
2. for MSR-burner based on LiF-NaF-KF efficiency of transmutation МА weakly depends on the volume of an active zone. During operation of the reactor within 50 years fraction МА reaches 74 … 78 % from loaded actinides. Thus possibility of further use of equilibrium fuel composition after replacement of the reactor vessel on the expiration of its resource will provide efficiency of transmutation МА close to one. It practically excludes plutonium consumption;
3. at low concentration actinide fluorides in fuel composition (~3%, mole), corresponding solubility limit in LiF-BeF2, efficiency of transmutation МА is less than 50 %. It means primary burning plutonium in such reactor. Reduction of volume of an active zone MSR-burner based on LiF-BeF2 leads to decrease $K\_{MA}$. At small volume of an active zone (less than 2 m3 at specific thermal power 100 kw/l) reactor operation can appear impossible because of degradation of fuel composition (accumulation of even isotopes Pu) and impossibility of achievement of a criticality (even at feed by only plutonium).

**CONCLUSION**

Our effort is focused on studying the main regularities of MA transmutation in the MSR-burner. The optimal equilibrium conditions of MA transmutation in the MSR-burner were demonstrated to be possible in principle when the makeup fuel containing only MA is added and fission products with the mass being equivalent to the amount of loaded MA are extracted. A certain concentration of actinides is required to implement these conditions of MA transmutation. When concentration of actinides is below that one taken for the reactor to go critical in the equilibrium conditions, instead of a certain amount of MA, plutonium shall be added to the makeup fuel thus reducing transmutation efficiency. When, vice versa, the required concentration of actinides is above that one taken for the reactor to go critical, a part of fuel with a high content of 238Pu isotope shall be extracted in order to support criticality in the equilibrium conditions.

The obtained results demonstrate that optimal concentration for the MSR-burner with the core volume ranging from 2 to 30 m3 is within 17…10%, mole. According to the published experimental results (for example [12]), this range can be expected to fall within solubility of actinide fluorides in the eutectics LiF–NaF–KF. To confirm this, the joint solubility of actinide fluorides in the eutectics LiF–NaF–KF is planned to be experimentally investigated at FSUE «RFNC – VNIITF named after Academ. E.I. Zababakhin» (Russia).

For MSR-burner based on of LiF-BeF2 because of low solubility of actinide fluorides (no more than ~3 %, mole) the greatest efficiency of transmutation МА ~40 % can be reached only at big (more than 15 m3) the volume of an active zone. However, in this case such reactor is plutonium burner therefore Pu consumption in 1.5 times exceeds МА consumption. Thus the increase in the volume of an active zone does not allow to reach an optimum mode of a transmutation (for infinite idealized system $K\_{MA}\~0.63$ [8]).

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