

## CONCEPTUAL DEVELOPMENT OF MOLTEN-SALT REACTOR-BURNER OF MINOR ACTINIDES

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### ABSTRACT

Some neutronics aspects of a molten-salt reactor-burner concept are considered in the paper. This reactor is proposed for minimizing the equilibrium amounts of minor actinides in the future nuclear power sector. This reactor operation in equilibrium fuel cycle was studied for the fuel-salt composition based on 18%LiF–58%NaF–24%BeF<sub>2</sub> (mol.%) with specified nuclide composition of the feed fuel and definite parameters of system for removal of fission products from the fuel.

### 1. INTRODUCTION

Studies on development of the nuclear power structure with minimum amounts of long-lived radwastes are under way in RRC KI [1,2]. The results of these studies show that for the U-Pu closed fuel cycle the optimal neutron balance can be obtained in the system, consisting of thermal power reactors, fast breeder power reactors (FBR) and reactors-burners of minor actinides (MA). Critical or subcritical molten-salt reactors (MSR) are considered as such burners [2-4]. The purpose of the MSR-burner is to transform long-lived high-active MAs in stable, average-lived and short-lived fission products (FPs). In the chosen nuclear power structure all MAs and a part of plutonium is directed to MSR-burners, while the major part of plutonium circulates in fast and thermal reactors. The nuclide composition of transuranic elements (TRU), directed to the MSR-burner and corresponding to the optimal nuclear power structure, was obtained in a sequence of iterative studies and presented in the paper.

RRC KI in cooperation with a number of Russian organizations has developed a concept of nuclear power technological system with homogeneous MSR-burners [5] for burning of TRU from VVER spent fuel. A salt composition, based on Li, Be/F, which is one of the most studied, was chosen for this reactor. The TRU solubility in the chosen system was estimated about 1 mol.% at the fuel-salt inlet temperature of 620°C. The purpose of the study, presented in the paper, was optimization of operational parameters of the MSR-burner in frames of the studied nuclear power structure, in particular, by selection of the salt composition with increased TRU solubility. In the given study the fuel-molten-salt composition based on 18%LiF – 58%NaF – 24%BeF<sub>2</sub> (mol.%) is considered. The solubility of trivalent elements, including MAs and FPs, which compete each other, at the fuel-salt inlet temperature of 620°C is estimated by about 2 mol.%, melting temperature of the salt composition is close to 480°C. Thus, the application of this salt in the MSR-burner allows for optimizing the combination of operational ranges for temperature and TRU solubility. Side by side with critical MSR-burners, conceptual options of subcritical MSRs with external neutron sources are currently considered [6-9]. Problems and promises of subcritical accelerator-driven cascade MSR concept [7-9] are discussed in the paper. The work is performed in frames of ISTC #1486 Project.

## 2. INPUT DATA FOR THE MSR-BURNER MODEL

A number of model calculations of equilibrium nuclide flows, established in the proposed nuclear power structure, was conducted. This nuclear power structure consists of three components: LWRs, FBRs and MSRs. The U-Pu fuel cycle of this structure is assumed closed for all MAs. The nuclide flows are organized in such a way that total amounts of MAs in the nuclear power system become minimum [2]. Taking into account that the MSR-burner requires a high level of automation and remote control, this reactor can occur not profitable as a power reactor. Hence, amounts of plutonium to be loaded in the MSR-burner should be minimized in order to reduce the number of such reactors in the nuclear power structure. At the same time, a high level of plutonium separation from MAs, while reprocessing the spent fuel of power reactors, may occur not cost-effective and some amount of plutonium will be directed to MSR-burner along with MAs.

The conducted calculations allowed to find the nuclide composition of MAs, separated from the cooled and reprocessed fuel of power reactors and directed for utilization to the MSR-burners. This MA composition was accepted in the given study as a fixed feed fuel composition for the MSR-burner (see Fig. 3). The plutonium content in this feed fuel was chosen about 10%.

A quasi-static model of the equilibrium state of the MSR-burner core with periodic feed by the specified fuel composition was analyzed in the study. An infinite homogeneous medium was considered with composition of  $(18-x)\%LiF - 58\%NaF - 24\%BeF_2 - x(YF_3-ZF_3)$  (mol.%), where  $YF_3$ ,  $ZF_3$  – trifluorides of MAs and FPs, respectively. The solubility of trivalent elements, including MAs and FPs, which compete each other, is estimated  $x \approx 2$  mol.% at temperature of 620°C, melting temperature of the salt composition is close to 480°C.

The fuel clean-up scheme, in general, is assumed in periodical replacement of a part of the primary fuel-salt composition by the same volume, which is free of FPs. Addition of feed fuel for compensation of MA incineration and accumulation of FPs between fuel clean-up can be made constantly. The feed fuel supply may be accommodated in intervals between fuel clean-up for compensation of reactivity drop. The following parameters of the MSR-burner fuel cycle were assumed in the study:

- *fuel feed*: feed fuel is added each day to restore the total amount of heavy nuclides in the fuel-salt composition;
- *fuel clean-up*: 10% of FPs is instantly removed each 10 days; contribution of fission gas release and precipitation of FPs in neutron absorption is assumed negligible.

The establishment of the equilibrium nuclide composition of infinite medium was calculated for four values of neutron flux density:  $1 \cdot 10^{14}$ ,  $5 \cdot 10^{14}$ ,  $1 \cdot 10^{15}$  and  $5 \cdot 10^{15}$  n/cm<sup>2</sup>s. An equilibrium state is assumed a regime with established nuclide composition in reactor, i.e. the time-average rates of change in nuclide concentrations are zero. The equilibrium composition was found by multi-recycling calculations, i.e. isotopic kinetics is simulated with recalculation of neutron spectrum and cross sections at each time step and with account for fuel feed and clean-up for a time period necessary for establishment of the reactor own composition and corresponding  $k_{\infty}$ . The problems of reactivity compensation in the interval between clean-up were not considered.

The nuclide flows in the simulated nuclear power system were calculated with the use of the ISTAR system [2] and nuclear data in the ENDF format. The equilibrium state of the MSR-burner with specified feed fuel was calculated with the JAR-FR code [10], using CONSYST/ABBN nuclear data system [11]. The parameters of the critical MSR-burner were estimated with the use of MCNP, SCALE, LOOP2 [12], JAR-FR codes.

### 3. NEUTRONICS ANALYSIS OF THE MSR-BURNER FUEL-SALT COMPOSITIONS

A feature of the MSR-burner of MAs is a complex nuclide composition of fuel, which establishes in reactor after long-term operation with constant feeding by nuclides, extracted from spent fuel of power reactors. An equilibrium composition of fuel, fission products and neutron balance in MSR-burner will be established, according to schemes of fuel feeding and clean up, neutron spectrum and neutron flux density. If neutron flux in the MSR-burner is low, the rates of radiative decay of fuel nuclides exceed the rates of transmutation of fuel nuclides due to the (n, $\gamma$ ) reaction and the neutron multiplication properties of the fuel composition deteriorate.

The value of infinite multiplication factor  $k_{\infty}$  of the fuel-salt composition in the equilibrium cycle versus neutron flux density with and without account for FPs is given on Fig. 1. When neutron absorption by FPs is not taken into account, the  $k_{\infty}$  value grows with increase of neutron flux density. This is provided by improvement of neutron balance in the equilibrium fuel-salt composition, when neutron flux density increases, first of all, due to accumulation of  $^{245}\text{Cm}$ , making the greatest contribution in  $k_{\infty}$  of the equilibrium fuel composition (see Fig. 4). Accumulation of FPs grows in high neutron flux densities along with “shift” of the nuclide composition to Cm. When the fuel clean-up rate and efficiency are constant, the dependence of  $k_{\infty}$  on neutron flux density has a maximum (see Fig. 1). Therefore, there are optimal combinations of neutron flux density and clean-up intensity, which provide the best neutron balance in the reactor.

Time history of  $k_{\infty}$  in equilibrium fuel cycle for various neutron flux densities for the accepted fuel cycle scheme is presented on Fig. 2. The drop of  $k_{\infty}$  in intervals between clean-up, especially noticeable at high neutron flux density, is provided, first of all, by fuel feed, rather than by FP accumulation. The value of  $k_{\infty}$  of the feed fuel composition is close to 0.34, i.e. this composition is a strong neutron absorber. The higher MA incineration rate is, the more feed fuel is added and the deeper the drop of  $k_{\infty}$  is in intervals between the clean-up. Each 10 days 10% of accumulated FPs are removed and  $k_{\infty}$  grows.

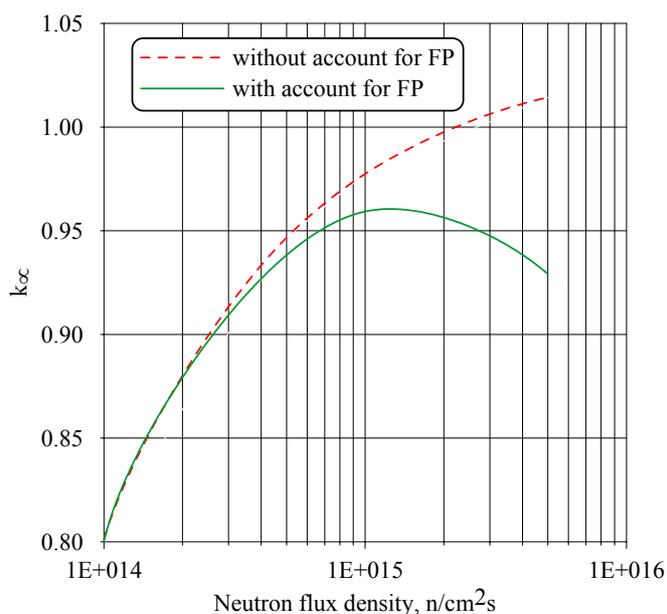


Fig. 1. Dependence of  $k_{\infty}$  of the equilibrium fuel-salt composition on neutron flux density after fuel clean-up

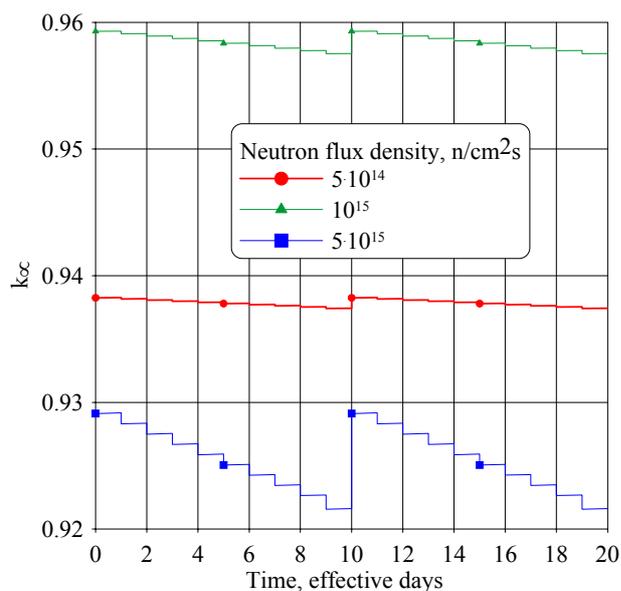


Fig. 2. Time history of  $k_{\infty}$  in equilibrium fuel cycle for various neutron flux densities

The feed fuel nuclide composition and the equilibrium fuel composition for various neutron flux densities are given on Fig. 3. Contribution of heavy nuclides (with account for their concentrations) in  $k_{\infty}$  of the fuel-salt composition relative to contribution of  $^{239}\text{Pu}$  is given on Fig. 4.

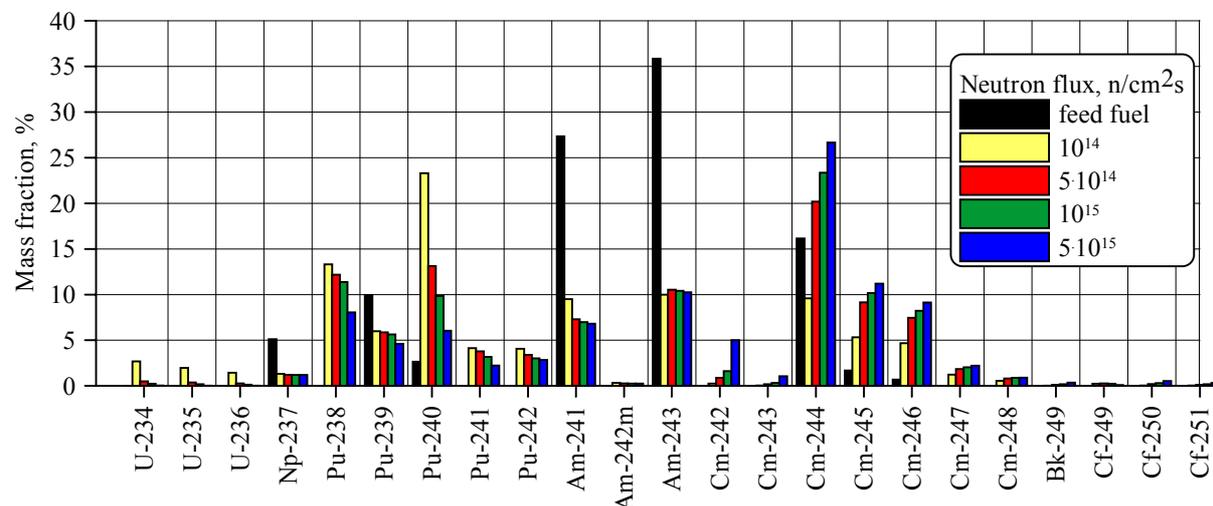


Fig. 3. Feed fuel nuclide composition and equilibrium fuel composition for various neutron flux densities

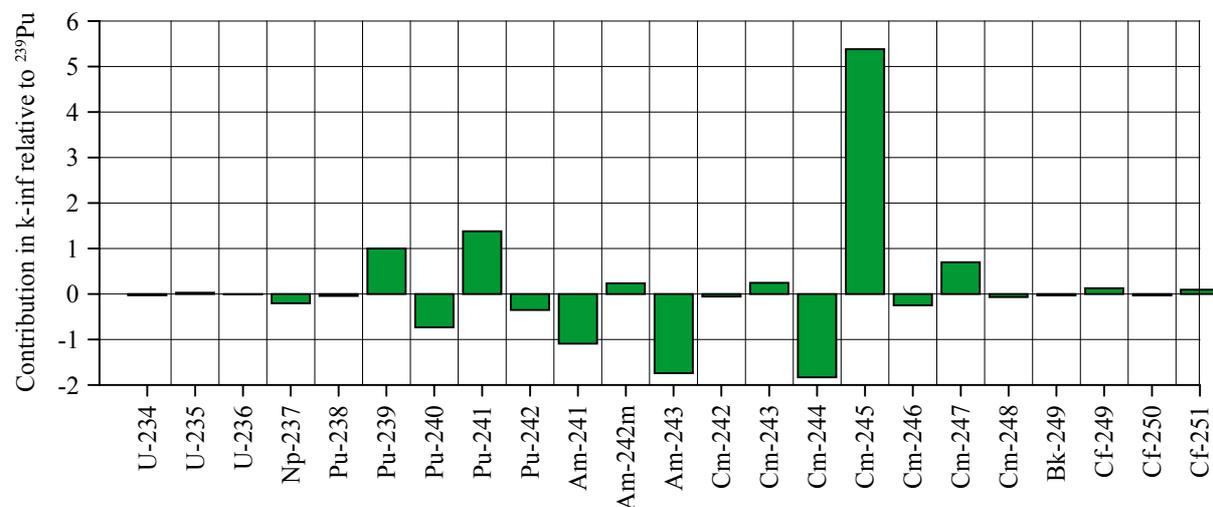


Fig. 4. Contribution of heavy nuclides in  $k_{\infty}$  of fuel-salt composition relative to contribution of  $^{239}\text{Pu}$

The efficiency of the MSR-burner of MA can be estimated by the fuel incineration rate. According to preliminary estimates, about 25% of FPs for the considered fuel compositions are trivalent (first of all, lanthanides) and compete by solubility with fuel. The calculational estimates of the incineration rates and equilibrium content of FPs in fuel-salt composition are given in Table 1.

Table 1. Parameters of fuel incineration in different neutron fluxes for the specified fuel cycle scheme

Neutron flux density, $\text{n/cm}^2\text{s}$	$1 \cdot 10^{14}$	$5 \cdot 10^{14}$	$1 \cdot 10^{15}$	$5 \cdot 10^{15}$
MA incineration rate, % of heavy atoms (h.a.) per year	1.1	5.9	12.3	58.4
Equilibrium amount of FPs before clean-up, % of h.a.	0.6	3.3	6.8	32.0

#### 4. PROBLEMS OF THE MSR-BURNER, REQUIRING COMPREHENSIVE ANALYSIS

The following factors should be taken into account, when developing the MSR-burner design and choosing operational parameters:

- the  $k_{\infty}$  value of the equilibrium fuel-salt composition should be high enough for development of the critical or near-to-critical MSR-burner with account for equilibrium amounts of FPs, established in the fuel-salt composition for the chosen neutron flux density and the clean-up system efficiency;
- heat should be removed at acceptable temperatures for the established neutron spectrum, chosen neutron flux density and fuel concentration;
- long-term operation of structural materials should be provided in high neutron fluxes at high temperatures;
- the rates of MA transmutation by  $(n,\gamma)$  and incineration by  $(n,f)$  should be optimized.

As seen from the presented results, development of the critical or near-to-critical MSR-burner is complicated by the following restrictions related to the fuel cycle:

1. High neutron flux density is required for effective transmutation and incineration of MAs as well as for necessary neutron balance of the equilibrium fuel-salt composition. The analysis of the presented results shows that for the studied fuel-salt composition and for the chosen parameters of the fuel feed and clean-up the highest value of  $k_{\infty}$  is obtained for neutron flux density of about  $1 \cdot 10^{15}$  n/cm<sup>2</sup>s. Higher neutron flux density requires more intensive fuel clean-up from FPs and corresponds to higher neutron fluences on the reactor structural materials and higher power density. Note, that the infinite medium was analyzed. If fuel in the MSR-burner circulates in the primary circuit, then the ratio of fuel volume in core to fuel volume out of core decreases and the effective neutron flux density reduces. It is also important that amount of FPs will also correspond not to the neutron flux density in the core, but to the effective neutron flux density in the reactor.
2. The high fuel concentration and high neutron flux density can result in high power density in the MSR-burner core. Therefore, the volume of the MSR-burner core is restricted by allowable reactor thermal power.
3. Rather low volume of the MSR-burner core can provide high neutron leakage, which can be significantly reduced, for example, with graphite reflector. However, in this case, the problems arise with the growth of neutron flux and power density near the reflector.
4. To provide necessary neutron balance in the reactor, the FP concentration in the fuel-salt composition should be restricted. Qualitative comparison of average neutron spectrum in the reactors, considered in the nuclear power structure, is given on Fig. 5. As seen, the developed MSR-burner could be qualified neither as fast reactor nor as thermal reactor. Considerable part of neutrons in the MSR-burner is in the resonance-thermal region, where the cross sections of FPs are high. The dependence of equilibrium amount of FPs before clean-up on time intervals between partial removal of FPs and fraction of removed FPs is shown on Fig. 6 for the chosen neutron flux density of  $1 \cdot 10^{15}$  n/cm<sup>2</sup>s. This plot provides basis for analyzing the relation between clean-up parameters and FP content in the fuel-salt composition.

If reflector is used, channels can be made in the reflector for cooling as well as for circulation of the fuel-salt composition. This design may allow to improve the MA incineration rate, because fission of many nuclides, transmuted from the feed fuel, is more effective in more thermal neutron spectrum, provided in the graphite channels. Thus, the MSR-burner can consist of two zones: main central cavity, in which neutron spectrum and neutron flux density are optimal for transmutation, and thermal zone for more effective MA fission, which also serves as a neutron reflector for the central zone. The difficulty of this design is increase in the volume of graphite, used for the reflector, which activation

and swelling under the given conditions can require frequent replacement and utilization in large amounts. In general, the MSR-burner operation will inevitably be associated with high power density and neutron fluences on the structural materials. The lifetime of the structural materials could occur lower than lifetime typical for power reactors. In this case, periodical replacement of spent materials should be envisaged. These problems are planned to be studied at the next stages of the study.

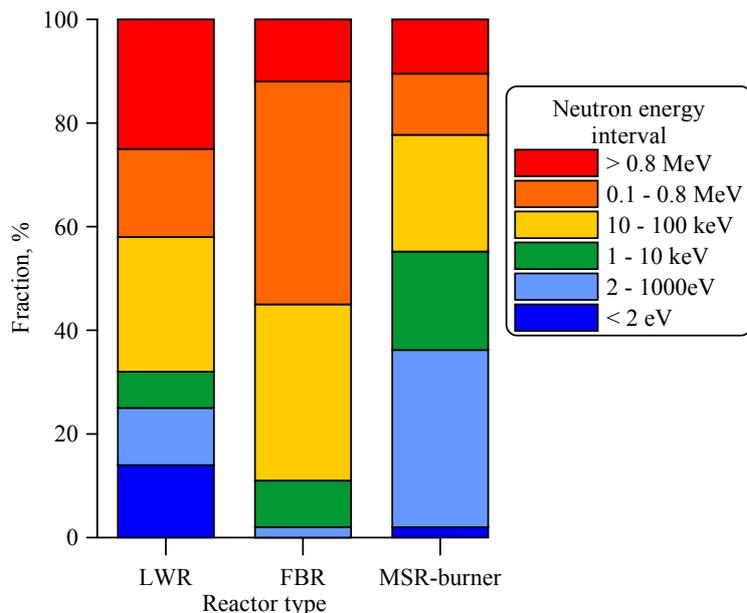


Fig. 5. Distribution of neutrons in energy intervals in reactors of various types

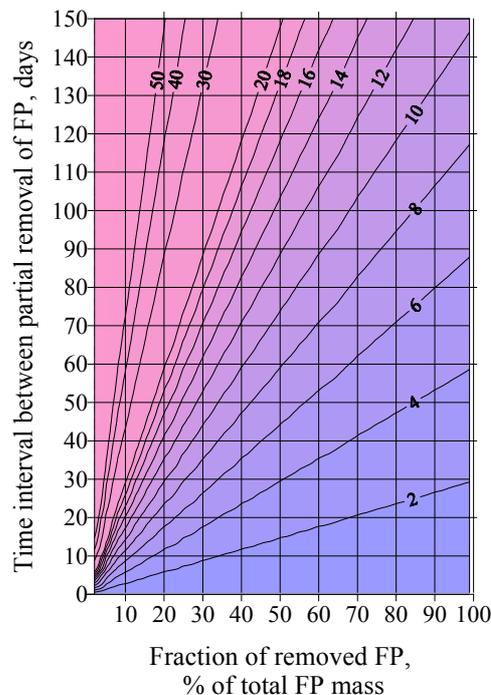


Fig. 6. Equilibrium amount of FPs before clean-up, % of h.a., vs the clean-up system parameters

## 5. MODEL OF MSR-BURNER OF MINOR ACTINIDES

RRC KI in cooperation with a number of Russian organizations has developed a concept of nuclear power technological system with homogeneous MSR-burners [5] for burning of TRU from VVER spent fuel. A salt composition, based on Li, Be/F, which is one of the most studied and used in MSRB and DMSR projects, was chosen for this reactor. The TRU solubility in the chosen system was estimated about 1 mol.% at the fuel-salt inlet temperature of 620°C.

The purpose of the conducted study was optimization of operational parameters of the MSR-burner in frames of the studied nuclear power structure, in particular, by selection of the salt composition with increased TRU solubility. Preliminary estimates, made with account for the presented data for the fuel-salt composition based on 18%LiF – 58%NaF – 24%BeF<sub>2</sub> (mol.%), show that the model of the reactor facility with the homogeneous MSR-burner can be developed on the basis of design, presented in [5] (Fig. 7). In this case, the plutonium content in the feed fuel is assumed about 10%, while total content of MAs and lanthanides in the fuel-salt composition is about 2 mol.% and the clean-up intensity is about 1% of the volume of the fuel-salt composition per day. In the MSR-burner project [5] the ratio of fuel volume in core to fuel volume out of core is close to 1:1. At the current stage of the study the effective neutron flux density is assumed, in this case, about two times lower

than the neutron flux density in the core. Then, the optimal neutron flux density in the MSR-burner core is estimated close to  $2 \cdot 10^{15}$  n/cm<sup>2</sup>s (see Fig. 1). The 30-cm-thick graphite reflector was assumed around the core. The preliminary parameters of the MSR-burner are given in Table 2. Other parameters of this reactor project are presented in [5].

Table 2. Preliminary parameters of the MSR-burner of MAs

Salt composition, mol.%	18%LiF–58%NaF–24%BeF <sub>2</sub>
Content of heavy atoms in the salt composition, mol.%	1.8
Reactor thermal power, MW	2500
Core height, m	3
Core diameter, m	3
Average power density, MW/m <sup>3</sup>	118
Average neutron flux density in the core, n/cm <sup>2</sup> s	$2.3 \cdot 10^{15}$
Effective neutron flux density in the reactor, n/cm <sup>2</sup> s	$1 \cdot 10^{15}$
Equilibrium amount of FPs in the fuel, % of h.a.	7
Peak fluence of fast neutrons ( $E > 50$ keV) on the core boundary, n/cm <sup>2</sup>	$5 \cdot 10^{22}$
Effective MA incineration rate*, % of h.a. per year	12.3

\*calculated for infinite medium with neutron flux density of  $1 \cdot 10^{15}$  n/cm<sup>2</sup>s

In the proposed option of the MSR-burner the MA incineration rates are 0.05 t/year of Np, 0.13 t/year of Pu, 0.67 t/year of Am and 0.2 t/year of Cm.

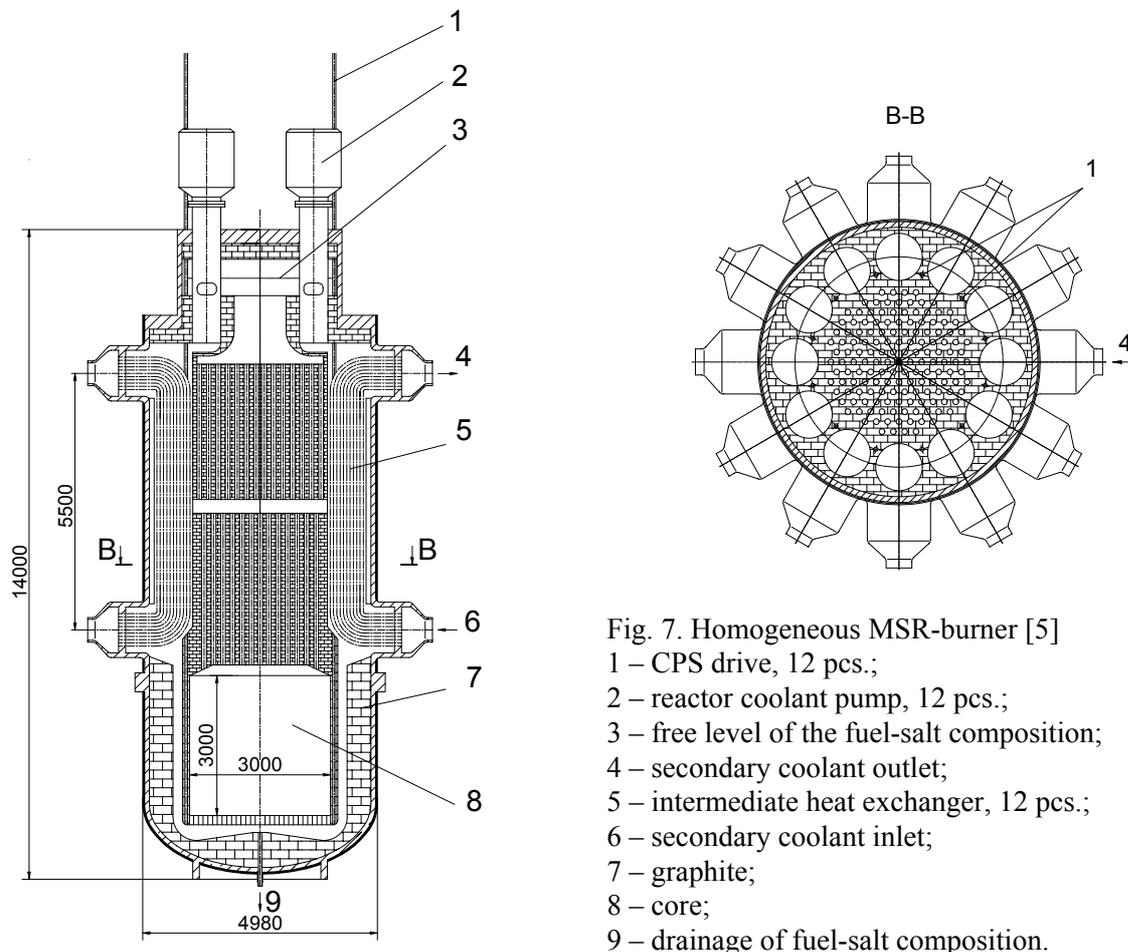


Fig. 7. Homogeneous MSR-burner [5]  
 1 – CPS drive, 12 pcs.;  
 2 – reactor coolant pump, 12 pcs.;  
 3 – free level of the fuel-salt composition;  
 4 – secondary coolant outlet;  
 5 – intermediate heat exchanger, 12 pcs.;  
 6 – secondary coolant inlet;  
 7 – graphite;  
 8 – core;  
 9 – drainage of fuel-salt composition.

## 6. ASPECTS OF SUBCRITICAL MSR-BURNER CONCEPT

Side by side with critical MSR-burners conceptual options of subcritical MSRs with external neutron sources are currently considered [6-9]. However, provision of neutron flux density necessary for effective MA burning at significant reactor subcriticality can occur difficult in view of high requirements to power and reliability of external neutron source. Therefore, although a subcritical reactor with external neutron source is more flexible in both the feed fuel nuclide composition and the own equilibrium fuel composition, the schemes of fuel feeding and clean-up for this reactor and the reactor operational parameters should also be optimized.

Besides, the estimates of MA solubility in the salt systems, proposed for the MSR-burners, as well as nuclear data for MAs are, mainly, calculational and theoretical results, which require experimental verification. The development of the critical MSR-burner can be complicated by low solubility of MAs in the salt composition at acceptable temperatures as well as by high requirements to the system of fuel clean-up from FPs. The reactor criticality can be obtained by adding the plutonium isotopes (or  $^{235}\text{U}$ ,  $^{233}\text{U}$ ) to the feed fuel. However, this measure would deteriorate the MA incineration efficiency and increase the equilibrium amount of MA in the whole nuclear power system. Maintenance of the reactor criticality, i.e. compensation of reactivity changes (see Fig. 2) can occur a problem, which is technologically more difficult, than the maintenance of reactivity in the given interval for the subcritical reactor. Besides, the subcritical accelerator-driven reactor-burner of MAs has evident advantages for safety licensing over the critical reactor.

A subcritical accelerator-driven reactor as an option of the MSR-burner of MAs is proposed in studies [6-9]. The MSR-burner operation under subcritical conditions can provide a definite freedom both for the reactor operational regimes and for the feed fuel composition, formed in the nuclear power structure with the fuel cycle, closed for all MAs. A concept of the cascade subcritical reactor is aimed at improvement of external neutron source efficiency in accelerator-driven reactor [6,7]. The studies are now under way on selection of appropriate salts, reactor design and on estimation of the MA transmutation and incineration efficiency in the cascade subcritical MSR (CSMSR).

A feature of the CSMSR project is increase in the number of external neutrons due to their multiplication in the reactor central zone with high  $k_{\infty}$  value. The CSMSR core consists of two zones: 1) the central zone, inserted in the reactor center and designed for multiplication of external neutrons, with its own fuel molten-salt composition and circulation circuit; 2) the main peripheral core, designed for the MA incineration. The difficulties in application of the cascade amplification principle to the subcritical MSR-burner concept relate, first of all, to the requirement to difference in neutron spectrum of the central and peripheral zones. The central zone should have the neutron spectrum as fast as possible, while the peripheral zone should have the neutron spectrum as thermal as possible. As shown in [8,9], in the CSMSR concept the  $k_{\infty}$  value and power density of the central zone are rather high, while volume is rather small. Therefore, the important problem is heat removal at acceptable temperatures. One of the problems for further studies is a search for ways to decrease power density in the CSMSR with preservation of the MA transmutation and incineration efficiency.

The model of the homogeneous MSR-burner, presented in the paper, can be considered as one of the stages of the CSMSR development, aimed at estimation of transmutation zone parameters. The central zone design, neutronics and thermal-hydraulics parameters are assumed to be analyzed at the next stage of the study. When developing the CSMSR, it could be expedient to compare the efficiency of including the MSR and CSMSR in the nuclear power structure with estimation of technical and economic indexes of the system as a whole.

## CONCLUSIONS

Some neutronics aspects of a molten-salt reactor-burner concept are considered in the paper. This reactor is proposed for minimizing the equilibrium amounts of minor actinides (MAs) in the future nuclear power sector.

Fluoride salts were chosen for the use in the MSR-burners due to their satisfactory neutronics, thermal and chemical properties. One of the key reasons of their consideration is an assumption about the excellent compatibility of MSR feed fuel preparation with technologies for fluoride reprocessing of spent fuel from power reactors. Such compatibility allows to minimize losses of high-active nuclides and number of technological processes, while utilizing MAs in MSR.

The reactor-burner operation in equilibrium fuel cycle was studied for molten-salt fuel composition based on 18%LiF – 58%NaF – 24%BeF<sub>2</sub> (mol.%) with specified nuclide composition of feed fuel and definite parameters of system for clean-up from fission products.

A sequence of iterative studies was conducted on modeling of nuclide flows in the considered three-component structure of the nuclear power, consisting of light-water thermal reactors, fast reactors and molten-salt reactors (MSRs) with the U-Pu closed fuel cycle. As a result of these studies, the nuclide composition of transuranic elements, directed to the MSR-burner and corresponding to the optimal nuclear power structure, was obtained.

Preliminary parameters of the homogeneous critical MSR-burner are presented in the paper. According to the obtained results, the neutron flux density of about  $2 \cdot 10^{15}$  n/cm<sup>2</sup>s is close to optimal for the MSR-burner core with the chosen parameters of the fuel clean-up system. It was shown that the neutron flux density increase in the reactor is indeed profitable for improvement of the MA transmutation and incineration. The main factors, restricting the neutron flux density in the MSR-burner, are reliability of the structural materials under impact of high temperatures and high irradiation doses, as well as necessity of intensive fuel clean-up from FPs. The estimate of optimal parameters of the fuel clean-up system for the MSR-burner of MAs should be based on special comprehensive neutronics, chemical, technological and economic studies for both the MSR-burner and the nuclear power system as a whole with account for MA losses, when separating FPs. Problems and promises of subcritical cascade MSR concept, driven by accelerator, were discussed in the paper.

Taking into account significant uncertainties in nuclear data for MAs, in properties fuel-salt compositions, in data on reliability of structural materials under conditions of high temperatures and neutron fluences, as well as the methodical assumptions, accepted at the current stage of the study, the obtained results require further comprehensive qualification.

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