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Studies of Physical Features of Cascade Subcritical Molten Salt Reactor with External Neutron Source

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Paper presents the results of computational analysis of cascade schemes of a molten salt subcritical reactor (CSMSR) – burner of long-lived radioactive wastes with the feed isotopic composition formed in the closed fuel cycle of the multicomponent nuclear power system. In the considered equilibrium model of the nuclear power with the closed fuel cycle, minimization of quantities of minor actinides serves as the optimization criterion in determination of shares of installed capacity of reactor of different types.

In this paper we consider the CSMSR (with the proton accelerator and target as a neutron source) as an element of the multicomponent nuclear power system, which is responsible for minor actinides utilization.

In the present moment one can hardly propose a technical solution for reaching the high level of neutron flux in the transmutation zone in a subcritical reactor with subcriticality in the range of 1-5% K_{eff} (K_{eff} is in the range of 0.99-0.95).

The paper presents studies of the possibility to use special intermediate zone with high importance of fast neutrons that allow to achieve neutron flux level, sufficient for effective transmutation under condition of fixed accelerator power. Several CSMSR reactor designs are considered with molten salt and solid fuel zones of cascade amplification.

KEYWORDS: long-lived radioactive wastes, transmutation, accelerator driven systems, molten salt, reactor, cascade amplification.

1. Introduction

Minimization of amounts of dangerous long-lived radioactive wastes can make nuclear power more acceptable for society. Investigations show that this problem can be solved by the multi-component structure of Nuclear Power system [1]. Molten salt reactors can be used as components of such system for effective incineration of minor actinides (MA). In spite of the high value of prompt negative density effect of reactivity, safety justification of this type of reactor is a new and rather complex problem due to low value of Doppler reactivity effect and low value of the efficient fraction of delayed neutrons, which is caused by minor actinides loading and fuel circulation.

In this paper we consider a cascade subcritical molten salt reactor (CSMSR) with a proton accelerator as an external source of neutrons. For effective transmutation, high neutron flux density is necessary. At the present moment one can hardly reach the high level of neutron flux density in the transmutation zone in a subcritical reactor with subcriticality in the range of 1-5% k_{eff} (k_{eff} is in the range of 0.99-0.95) because of low power of existing accelerators. To decrease the necessary accelerator power, we investigate the effect of cascade amplification of neutrons [2].

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2. Cascade Molten Salt Reactor

2.1 Principle of Cascade Reactor

The main feature of the cascade scheme is an intermediate zone around the target, which amplifies effectively the neutrons emerging from the target. In this case, appropriate neutron spectra must be chosen to provide the one-way coupling of central and peripheral zones. The neutrons of peripheral zone (transmutation zone) must not affect on the processes in the central zone of cascade amplification. The cascade amplification factor can be determined as a ratio of external neutron source intensities providing the same power level for non-cascade and cascade accelerator driving. Neutron flux, in its turn, is the main characteristic for transmutation features, so we can express the cascade amplification factor A_F in terms of ratio of averaged by volume and integrated by energy neutron fluxes in CSMSR transmutation zone and in a homogeneous MSR core:

$$A_F = \frac{1}{V_{transmut}} \int_{V_{transmutation}} \Phi_{geter}^{transmut} dV \Bigg/ \frac{1}{V_{\text{hom}}} \int_{V_{\text{hom}}} \Phi_{\text{hom}} dV \qquad (1)$$

2.2 The Required CSMSR Features

In previous works [3, 4] the preliminary requirements for the selection of a CSMSR scheme with chosen fuel and salt compositions and structure materials were determined:

1. Average neutron flux in the transmutation zone must not be lower than $2{\cdot}10^{15}$ neutron/($cm^2{\cdot}s$) for effective incineration of minor actinides (see Figure 1);
2. Molten salt circulation velocity must not be greater then 5 m/s for acceptable erosion of structure materials;
3. Limiting temperature for Hastelloy-H (870°C) and melting temperature for salt composition determine temperature range in the central zone from 650°C to 850°C;
4. Fuel content in molten salt compositions must be smaller than the solubility limit to provide additional solution of fission products and opportunity to correct the molten salt composition.

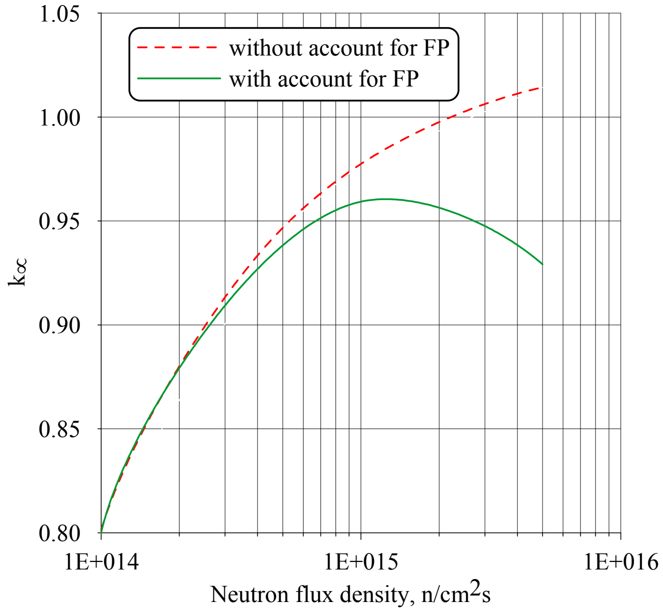

Fig. 1 Dependence of k_∞ of the equilibrium fuel-salt composition on neutron flux density after fuel clean-up [3].

In terms of the neutron balance the first requirement is crucial but hardly feasible for subcritical reactor. Therefore to achieve the required level of neutron flux in a subcritical molten salt reactor we consider the cascade principle of neutron amplification.

Figure 1 shows the dependence of infinite-medium multiplication factor of fuel-salt composition on neutron flux density in the equilibrium state [3]. 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 minor actinides and fission products (FP), respectively. As shown in Figure 1, the level of neutron flux in the order of $(1\text{-}2)10^{15}$ is the optimal level in terms of the lower subcriticality and consequently, lower required accelerator power.

2.3 Cascade Molten Salt Subcritial Reactor Designs

2.3.1 CSMSR with Molten Salt Cascade Amplification Zone

In paper [4] the optimized molten salt scheme of CSMSR was considered with molten salt compositions both in cascade neutron amplification zone and transmutation zone. On the next investigation stage the possibility to reach the necessary intensity of neutron source was analysed [5]. It had been shown that under proton bombardment the spallation neutrons yield from the molten salt target is essentially lower than from lead or tungsten one. For the molten salt target 20 cm diameter and 60 cm height the necessary current of proton beam is about from 15.5mA to 43mA, in dependence on energy of incident protons. These values of beam currents significantly exceed the value of typical accelerator driven system (ADS) current, which is equal 10 mA [6]. The increasing of target dimensions makes it possible to decrease the required beam current to 11.4 mA. Further increase of target does not result in neutron yield increase. That is why it should be admitted that it is difficult to reach the intensity of neutron source on the level of $6.5\cdot10^{17}$n/s for the considered design of a molten salt subcritical reactor. Additional analysis showed that the insertion of tungsten discs into salt composition significantly increases the yield of spallation neutrons from the target and this value approaches to the neutron yield from a lead or tungsten target.

Results of reactor calculations showed that application of molten salt cascade multiplication zone does not permit to obtain sufficient increase (with a factor greater than 3) of neutron flux. Concept of the CSMSR as a burner of minor actinides [7, 8] requires different neutron spectra in the central and peripheral zones of reactor. Neutron spectrum must be as hard as possible in the central zone and close to thermal in the peripheral zone. If the central zone contains a nuclide with threshold fission, then the sub-threshold neutron spectrum must be formed in the peripheral zone for realization of the cascade principle. The considered molten salt systems form mainly thermal-resonance neutron spectrum [4].

To produce the hard neutron spectrum in the cascade amplification zone, we have to consider CSMSR schemes with the cascade amplification zone containing neptunium [5].

2.3.2 Cascade Amplification Zone, Containing Neptunium

On the next stage of research we investigated the possibility to enhance the cascade amplification effect by implementation of neptunium-237 (in the cascade amplification zone) as the main fission element. A number of schemes with various designs and composition of the cascade-target unit were considered. There were variants with both liquid and solid fuel in cascade amplification zone.

The transmutation zone for all variants contains fuel salt composition 16.5·LiF-58·NaF-24·BeF_2-1.5·MAF_3 % molar. The 30 cm thick graphite reflector surrounds the transmutation zone, with the exception of fuel salt elevation section. The thermal power of a unit was assumed to be about 2500 MW.

Six variants of the cascade-target unit design were considered:

1. Neptunium nitride fuel rods containing 5% of plutonium separated from the spent fuel of a LWR-type reactor VVER-1000 in the Hastelloy cladding surround target. This zone is cooled by fuel salt composition 16.5·LiF-58·NaF-24·BeF_2-1.5·MAF_3 % molar.

2. Similar to variant 1, but with 6% plutonium content in fuel rods and, consequently, with two times decreased subcriticality depth and required intensity of external neutron source.
3. Neptunium nitride fuel rods in Hastelloy cladding surround target. This zone is cooled by Pb-Bi eutectic (Pb-Bi alloy with 44.5%Pb-55.5%Bi composition). In this case the given level of criticality is achieved without adding of plutonium.
4. The cascade amplification zone is filled with the homogeneous mixture of lead-bismuth alloy and neptunium taken with volumetric fractions 0.7 and 0.3, correspondingly.
5. After analysis of variants 1 and 2 it was found that in order to provide reliable cooling of fuel rods it is necessary to increase coolant flow cross-section by two times. This necessity leads to consideration of a model with fuel rod pitch increased from 8 mm to 9.1 mm.
6. Similar to variant 4, but the intensity of external neutron source is decreased by two times.

Some parameters of these CSMSR variants are shown in Table 1. A typical CSMSR calculational model [5] for variants 1, 2 and 5 is given in Figure 2.

Table 1 The Main Parameters of CSMSR variants

Parameter	Variant 1	Variant 2	Variant 3	Variant 4	Variant 5	Variant 6
Plutonium content, %	5	6	0	0	7	0
Central zone outer diameter, cm	43.5	43.5	43.5	24.6	51	24.6
Core outer radius, cm	150	150	150	124	150	124
A_F	1.95	2.78	2.13	4.05	1.31	3.96
Average volumetric heat generation density, MW/m^3	1563	1544	1611	4414	1070	2090
Volumetric share of solid fuel	0.510	0.510	0.510	-	0.394	-
Neutron source intensity, neutron/s	$1.74 \cdot 10^{18}$	$8.22 \cdot 10^{17}$	$1.74 \cdot 10^{18}$	$1.74 \cdot 10^{18}$	$1.74 \cdot 10^{18}$	$8.22 \cdot 10^{17}$
Target unit power, MW	8.53	4.03	8.53	8.53	8.53	4.03
Criticality level	0.98	0.99	0.98	0.98	0.98	0.98
Reactor thermal power, MW	2500	2500	2580	2868	1915	1357

The target design (variants 1, 2 and 5) is a bundle of vertically located tungsten rods, cooled by the fuel-salt composition. Starting with the upper part of the active core, there is a beam pipe in the Hastelloy cladding leading to the target. Below this beam pipe, also in Hastelloy cladding, the salt composition for cooling the tungsten rods is supplied. The fuel rods from the mixture of nitrides of plutonium and neptunium in the Hastelloy cladding are located around the target. Under the lower edge of the fuel stack, a gas plenum for accumulation of gaseous fission products is located. Space between fuel elements and rods of target is filled with fuel-salt composition. Cooling of fuel rods is carried out by the same fuel-salt composition. This decision eliminates the necessity to have two circuits with salts having different physical and chemical properties and, thus, simplifies the design of an installation.

The target-cascade unit is located in the center of the molten salt reactor core in such a way that the middle plane of the target coincides with the middle plane of the cascade amplification zone. The active core is surrounded by the graphite reflector of 30 cm thick from all sides (except for the elevation section of fuel salt).

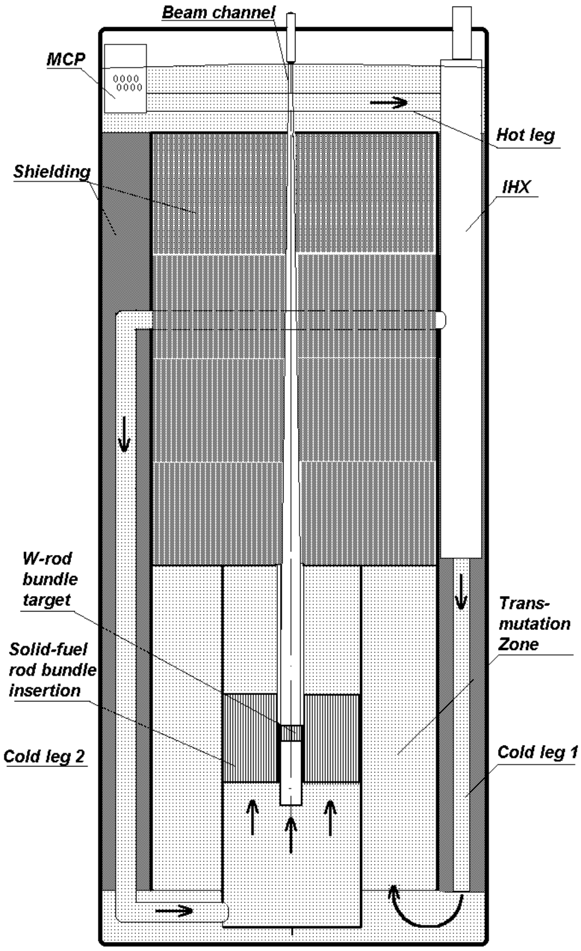

Fig. 2 CSMSR model with the target unit and solid fuel zone of cascade amplification.

2.3.3 Results of Computational Analysis

Analysis of performed calculations has shown that for the achievement of high neutron flux level in the transmutation zone it is necessary to increase the neutron flux level in the cascade amplification zone that causes unacceptable heat generation.

From this point of view all considered variants have practically the same disadvantages. There are no structure materials that can withstand such heat generation level. For example, in the case of cooling by fuel salt composition the heat generation density significantly exceeds the utmost value 1000 MW/m^3. In case of cooling by Pb-Bi alloy (both with or without neptunium) the heat generation density exceeds the utmost value 200-300 MW/m^3. Thus, the results show serious engineering problems connected with reactor subcriticality and insufficiency of effect of neutron cascade amplification. At the same time we cannot supply the neutron flux level necessary for transmutation and satisfy technical restrictions connected with performance of structure materials.

Presented results show that without reduction of the total power of a reactor unit, decrease of subcriticality level and optimization of dimensions of cascade amplification and transmutation zones it is impossible to provide reliable cooling of fuel rods in cascade amplification zone.

It should be noted that none of variants has significant advantages in neutron cascade amplification excluding variants 4 and 6 with homogeneous mixture of neptunium and Pb-Bi coolant, but these variants appear to be unlikely technically feasible.

2.4 Investigation of Parameters of Molten Salt Cooled Tungsten Spallation Target

Target unit for a solid fuel neptunium cascade amplification zone was a vertical bundle of tungsten rods cooled by fuel salt composition 16.5·LiF-58·NaF-24·BeF_2-1.5·MAF_3 % mol. Two targets were calculated with equivalent radii 9.5 cm and 17 cm.

2.4.1 Target thermal physics

Some questions concerning target thermal physics were considered. The result of parametric investigation gave the idea that tight W-rod bundle packed in hexagonal lattice had the best ability of heat removal from the target. The diameter of the rods and the lattice pitch are 6 and 7 mm, respectively. Spacing of the rods is assumed to be the same as in a typical LMFBR fuel assembly (wire wrapping around the rod of 'wire-to-rod' kind). The utmost value of the heat generation volumetric density in the tungsten rods, which is equal to 2 GW/m^3, was determined by thermal physics and thermal mechanics numerical analysis. At such high values of heat generation rate the temperature difference between rod surface and coolant can be 90 - 100 K; azimuth temperature deviation at the rod surface can reach 40 K. The utmost power released in tungsten rod must not exceed 4.2 MW for the 9.5 cm target and 11.5 MW for the 17 cm target.

2.4.2 Neutron Physical Parameters of Tungsten Molten Salt Cooled Target

When we direct the narrow proton beam to the target center, the non-uniformity factor of the radial heat generation distribution in the target can reach value k_r=16 and even higher. In this case, the target will be used ineffectively from the viewpoint of thermal mechanical limitations. In order to enhance the effectiveness of the target, we considered a target scanning by the beam oscillating in radial and azimuth directions according to the following law:

$$J_p(r) = \frac{I_0}{2\pi\sigma}\exp\left(-\frac{(r-r_0)^2}{\sigma^2}\right) \qquad (2)$$

where:

r_0 - coordinate of the maximum of the beam intensity:

$$r_0 = [R_{max}\, sin(2\pi v_1 t) + R_{min}\, cos(2\pi v_1 t)] sin(2\pi v_2 t) + [R_{max}\, sin(2\pi v_1 t) + R_{min}\, cos(2\pi v_1 t)] cos(2\pi v_2 t)$$

R_{max} and R_{min} - radial coordinates of the maximum and minimum deviation of the beam intensity peak from the target center, respectively;

v_1 and v_2 - frequency of the radial and azimuth oscillations, respectively;

$\sigma = 1.6$ cm - standard deviation of normal distribution for beam intensity.

It was assumed that the proton beam fall down normally on the target top.

The results of neutron yield and spectrum calculations (normalized for 1 proton of source) as well as the total power release are presented in Tables 2 and 3.

Table 2 Neutron yields and total power release for different targets dimensions and radial distributions of the 1 GeV proton beam.

Target	Neutron yield	Number of source "spallation" neutrons with E<15.02 MeV	Power release
R=9.5 cm, L=40.0 cm	18.48	17.26	454.5 MeV as per particles: p/π/n/γ - 388.1/16.8/13.0/36.6
R=17.0 cm, L=40.0 cm	19.53	20.67	519.5 MeV as per particles: p/π/n/γ - 408.6/18.8/19.9/72.2

Table 3 Energy group distribution of the number of source "spallation" neutrons with E<15.02 MeV for "homogeneous" targets

Group number, q	Upper boundary, MeV	Lower boundary, MeV	Number of neutrons	Number of neutrons
			Target R=9.5 cm, L=40 cm	Target R=17.0 cm, L=40 cm
1	15.02	13.98	0.2112	0.2588
2	13.98	10.5	1.021	1.242
3	10.5	6.5	2.544	3.064
4	6.5	4.0	3.551	4.247
5	4.0	2.5	3.686	4.400
6	2.5	1.4	3.568	4.256
7	1.4	0.8	1.819	2.170
8	0.8	0.4	0.7324	0.8761
9	0.4	0.2	0.1293	0.156
		Total:	**17.262**	**20.670**

The calculated energy release for the 9.5 cm target was about 450 MeV per proton. Therefore, at the source intensity $8.22\cdot10^{17}$ neutron/s in the reactor with criticality level 0.99 (variant 2) the target power can achieve 4 MW that corresponds to accelerator current of 7.7 mA. This heat is generated in the entire target, not only in tungsten rods. It is obvious that generated heat does not exceed utmost level. On the contrary, for a reactor with criticality level 0.98 the source intensity $1.74\cdot10^{18}$ neutron/s is required, and this intensity is connected with such heat generation that cannot be removed from the target with 9.5 cm radius. But it is possible to remove about 11.5 MW from target with radius 17 cm. For energy release which is about 520 MeV per proton in this target it is possible to increase source intensity up to $2.82\cdot10^{18}$ neutron/s that corresponds to accelerator current of 22.1 mA. This value of beam current significantly exceeds the value of typical ADS current, which is equal 10 mA [6]. To apply the accelerator with the current, which is not higher then value mentioned above, it is necessary to reduce the reactor subriticality level. In this case, as shown in Table 1 (variant 2 with the 9.5 cm target), there is a possibility to supply the necessary value of neutron source and reliable target cooling.

3. Conclusion

In this paper we considered various approaches for implementation of the neutron cascade amplification in a cascade subcritical molten salt reactor (CSMSR) intended for incineration of minor actinides.

It is shown that special destination of such reactor unit puts several contradicting requirements to its neutron-physical characteristics and technical parameters:

1. Reactor safety with respect to prompt criticality is supposed to be provided by associated subcriticality, but in this case it is difficult to obtain neutron flux sufficient for transmutation.
2. The low solubility of minor actinides in molten salt (fluorides) and requirement to support given subcriticality level call for high neutron flux level in the transmutation zone (about $2\cdot10^{15}$neuron/(cm$^2\cdot$s)). In this case neutron flux level in the cascade amplification zone must be significantly higher then in the transmutation zone. And this involves either inadmissibly high levels of heat generation in solid fuel rods or impossibility to provide neutron spectrum hard enough to achieve high cascade amplification factor in the molten salt cascade amplification zone (due to low solubility minor actinide in fluorides). Technical possibilities to create molten metal cascade amplification zones are nowadays even lower than for schemes with molten salt and solid fuel.
3. Small dimensions of cascade amplification zone and subcriticality result in strong

non-uniformity in heat generation and high local heat production. This fact involves problems with provision of safe operation of solid fuel rods. The expansion of the cascade amplification zone makes it possible to decrease heat generation non-uniformity but simultaneously involves the reduction of transmutation zone and decrease of transmutation efficiency.

4. Reducing the reactor subcriticality depth makes possible both increase of neutron cascade amplification factor and solution of problems connected with non-uniformity of heat generation.
5. Main problems in reactor facility with solid fuel rods are connected generally with fuel rods cooling. In case of small subcriticality depth it is possible to provide reliable operation of target unit elements.

The set of problems mentioned above demonstrates that nowadays the concept of a critical molten salt reactor [9] seems more feasible for incineration of compositions based on minor actinides.

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