PROSPECTS OF SUBCRITICAL MOLTEN SALT REACTOR FOR MINOR ACTINIDES INCINERATION IN CLOSED FUEL CYCLE

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Abstract

Paper presents an attempt to carry out synthesis of the approaches based on use of subcritical reactors for minor actinides (MA) recycling and incineration, and gas-fluorides methods of reprocessing of spent nuclear fuel.

The subcritical molten salt reactor ($K_{eff} = 0.95$) with fuel composition based on molten FLiNaK salt and fluorides of MA (separated from spent fuel VVER-1000 light water reactor) and external neutron source, based on 1 GeV proton accelerator with 6 mA protons current and molten salt cooled tungsten target is considered.

Paper presents the results of parametrical analysis of equilibrium isotopic composition of molten salt reactor (MSR) with MA feed in dependence of core dimensions, average neutron flux and external neutron source intensity.

Introduction

One of the requirements on the nuclear power wide-scale development is minimization of long-lived dangerous radioactive wastes (MA and fission products) volume in the fuel cycle. As investigations performed in NRC KI show this goal (with some others) may be achieved through the progressive going to multi-component nuclear power structure with fuel cycle closed by heavy nuclides. Two components will form the base of this multi-component system: light-water thermal reactors, producing electricity and/or high-potential heat for energy-technological applications, and also fast reactors producing excess plutonium and closing fuel cycle. Fast reactor hard neutron spectrum permits, in principle, transmute and incinerate MA from the spent fuel of thermal and fast reactors. But this implementation results in decreasing deleted neutron effective part (which itself in fast neutron reactors using plutonium fuel is lesser essentially then in thermal neutron reactors using uranium fuel) and in degrading Doppler-effect part of reactivity feedback (reactivity feedback may remain negative but lesser in absolute value or even, depending on loading and MA content, become positive). Thus reactor safety deteriorates significantly with relation to reactivity accidents. These factors have inspired at the end of 90-th in Europe, USA and China renewed investigations of electro-nuclear units with sub-critical blanket ($K_{eff} = 0.95 \div 0.99$) and with nearby zero probability of reactivity accident realization [1, 2].

A further problem of MA transmutation in fast neutron reactors is development of special fuel or irradiation facilities containing MA. The process equipment for such fuel fabrication must be remotely operating and requires very strong radiation shield for maintenance personnel radiation exposure decreasing. In addition, the coating of MA fuel (or elements of MA irradiation facilities design) itself are high-level radioactive wastes and demand respective managing. Design of MSR with dissolved MA may be alternative way. In this kind of reactor fuel pin coating and pin assembly covering are absent.

The development of radioactive waste processing is another necessary component on MA transmutation work. Present-day processing aqueous technologies based on one or another of PUREX-process modifications result in rather large liquid radioactive wastes what require special utilization or large storage volumes. Innovative radioactive wastes processing technologies based on dry gas-fluoride methods may be alternative way [3].

In a number of investigations performed in NRC KI, for example [4], an effort was made to combine approaches based on subcritical reactors and gas-fluoride RAW processing technologies. For this purpose reactor has been proposed with molten salt as coolant and fuel with MA dissolved in molten fluorides. There is some experience of pilot MSR operation in the world, but such kind reactor presents sufficiently complicated and innovative facility, which justification nowadays invites further investigations.

The design of reactor with molten salt and dissolved MA involves some difficulties and one of problem is lowered effective part of delayed neutrons. The reason is MA using (this part is small for most of MA) and delayed neutron precursors removing the core in circulation (some precursors are decayed in outer circuit).

These molten-salt reactor design technical problems can be solved using subcritical mode of MA incineration in MSR with external neutron source.

For the first time the conception of subcritical MSR was proposed in NRC KI in 1995 [4]. Over a long period conception of subcritical MSR was elaborated on the base of cascade principle of neutrons from target source multiplication due to special designed multiplication parameters of subcritical blanket and zone of cascade multiplication posed between target and subcritical blanket [5-7].

The experts estimated that the accelerator proton beam current must not exceed 10 mA [8]. It was expected that such design implementation permit decrease the requirements to accelerator proton beam by a factor equal 10 to provide needed for MA transmutation and incineration neutron flux at level $1 \cdot 10^{15}$ $n \cdot cm^{-2} \cdot sec^{-1}$. However careful investigation of cascade subcritical reactor physical features [9-11], shows that needed proton beam current can be decreased only by a factor 2, and in some situations it is necessary to provide solid-fuel or liquid-metal insertions around the target. Solid-fuel insertion materials (fuel and structural) in the process are situated in practically extreme conditions, so their long-time functioning cannot be guaranteed.

The possible ways from this situation were to replace molten-salt compositions LiF-BeF_2-NaF or LiF-BeF_2 to another with more MA solubility, and to decrease unit thermal power (in the cited papers a unit with thermal power 2500 MW was considered).

Demonstration subcritical reactor

In present work an analysis of possibilities to design molten-salt blanket for demonstration subcritical reactor was performed. Also the possibility was considered to use electro-nuclear neutron source for transmutation and incineration of MA from spent fuel of light-water VVER-type reactor, and for conversion Th-232 to U-233 and Pa-231 in thorium molten-salt blanket. The nuclide composition of MA feed is shown in Table 1.

Table 1 – Minor actinides feed to SMSR

Nuclide	Mass fraction	Element fraction
Np-237	3.18E-01	32%
Am-241	6.15E-01	67%
Am-242m	4.43E-04	
Am-243	5.96E-02	
Cm-243	9.17E-05	1%
Cm-244	5.80E-03	
Cm-245	1.13E-03	
Cm-246	9.43E-05	
Cm-247	1.02E-06	
Cm-248	7.80E-08	

Parametrical investigations were performed to choose subcritical molten-salt reactor (SMSR) acceptable design.

Fuel salt circuit in MSR consists of blanket (subcritical core with neutron flux) and outer loop (without neutron flux). There was adopted that during 30% of cycle time fuel salt is situated in blanket and during 70% of cycle time – in outer loop without neutron flux. Equilibrium fuel cycle parameters were investigated at 3 levels of average neutron flux in circuit:

- $1\cdot10^{14}$ $n\cdot sm^{-2}\cdot sec^{-1}$ ($3.33\cdot10^{14}$ in blanket),
- $5\cdot10^{14}$ $n\cdot sm^{-2}\cdot sec^{-1}$ ($1.67\cdot10^{15}$ in blanket),
- $1\cdot10^{15}$ $n\cdot sm^{-2}\cdot sec^{-1}$ ($3.33\cdot10^{15}$ in blanket).

External neutron source (proton accelerator target unit) was proposed to supply neutron flux in blanket. The results are shown in Table 2 and on Figures 1 and 2.

Table 2 – Parameters of SMSR Model-1 with external neutron source

Blanker diameter, m	2		
Blanket height, m	2		
Blanket volume, m^3	6.28		
Average neutron flux in circuit, $n\cdot sm^{-2}\cdot sec^{-1}$	$1\cdot10^{14}$	$5\cdot10^{14}$	$1\cdot10^{15}$
Heavy metals fraction in salt, % mol.	21.8%	13.8%	12.4%
Heavy metals mass in circuit, ton	44.4	30.5	27.8
Heavy metals mass in blanket, ton	13.3	9.2	8.4
MA consumption, ton/year	0.1	0.4	0.8
Thermal power, GW	0.3	1.2	2.3

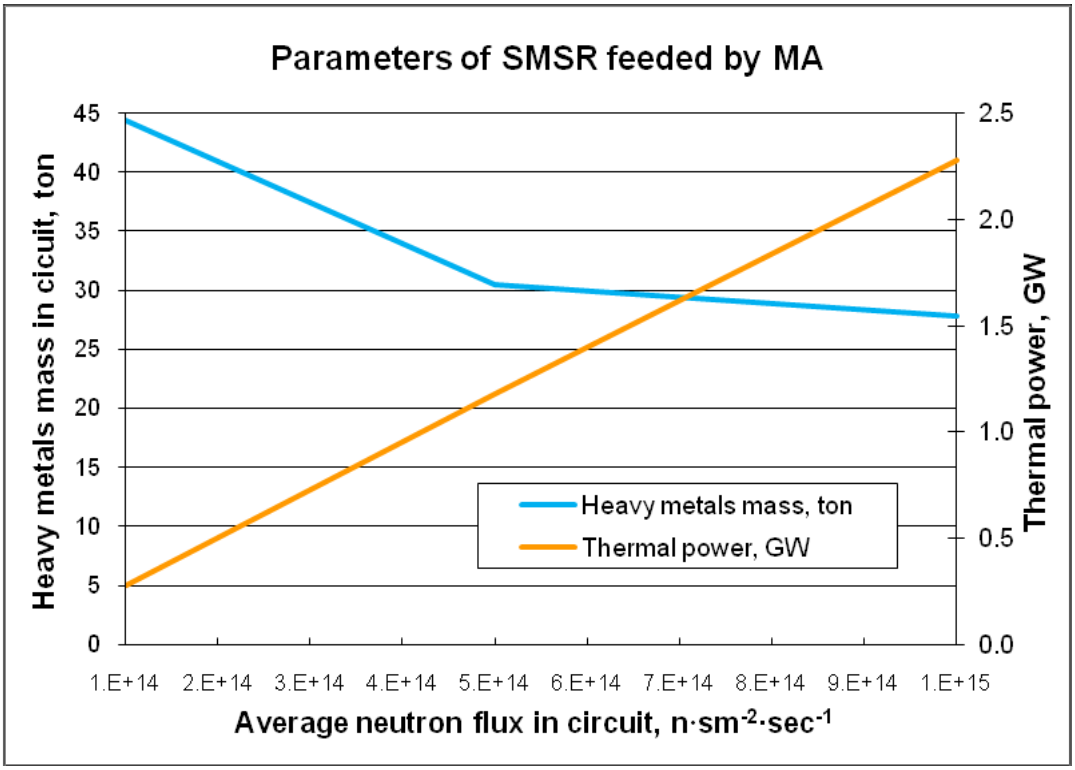

Figure 1 – HM mass and thermal power in dependence of average neutron flux in circuit

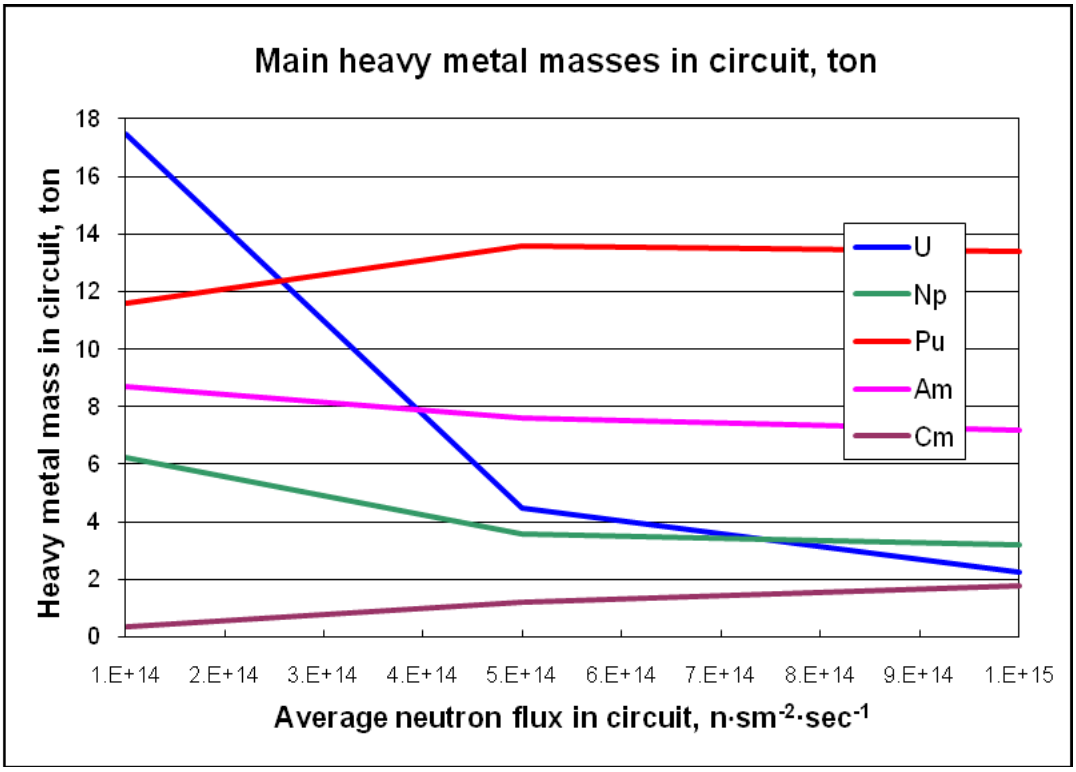

Figure 2 – HM masses in dependence of average neutron flux in circuit

Reactor VVER-1000 produces about 40 kg/year MA. In equilibrium station reactor model-1 can contain about 26÷44 tons MA. There is little likelihood that such amount of spent fuel can be processed in a needed time. So Model-1 project cannot be realized.

Calculations show that neutron flux increase from $1 \cdot 10^{14}$ to $5 \cdot 10^{14}$ $n \cdot sm^{-2} \cdot sec^{-1}$ results in significant decrease of HM mass in circuit, at the cost of uranium mainly; consumption of MA and thermal power increase. Further increase of neutron flux is not so profitable. However HM mass in circuit (several tens of tons) seems to be rather large. Only blanket volume decrease permits to keep the necessary level of neutron flux and to decrease loading mass. Therefore SMSR Model-2 with lesser blanket dimensions was considered. The preliminary results are shown in Table 3.

Resulting SMSR with accelerator proton beam target unit as external neutron source can utilize about 120 kg MA per year (the annual production of 3 reactors VVER-1000 type). It is consistent with industrial scale and opens up possibilities to design, construction and using this type SMSR as a component of spent fuel processing plants.

Comparison with parameters of SMSRs projecting and constructing now in other countries [12-15], in particular with reactor MYRRHA (Table 4) designing in European Community for investigation of materials, medical isotopes production and transmutation researches, shows that SMSR demonstration model (Table 3) provides the close neutron flux level, so this reactor will be suitable not only for transmutation and incineration but as neutron source for investigation of materials.

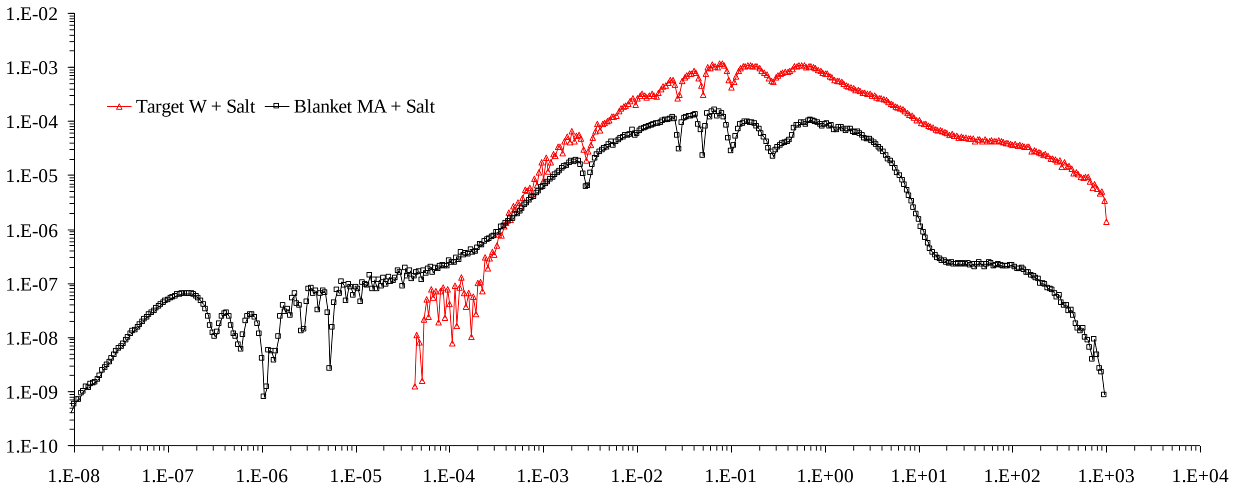

Figure 3 – Neutron spectra in molten salt blanket and target (with molten salt cooling)

Computational analysis shown that power of the target can be increased in case of sodium coolant.

Table 3 – Main technical parameters of demonstration SMSR

Blanket with minor actinides (BMA)	
BMA volume, m^3	1.53
Salt composition	0.465·LiF- 0.115·NaF- 0.42·KF
Heavy metals fraction in salt, % mol.	17.8%
Calculated K_{eff}	0.96
Heavy metals mass in BMA circuit, ton	9.2
Heavy metals mass in BMA, ton	2.8
MA consumption, ton/year	0.12
Thermal power, GW	0.36
Average neutron flux in BMA circuit, $n \cdot sm^{-2} \cdot sec^{-1}$	$5 \cdot 10^{14}$
Average neutron flux in BMA, $n \cdot sm^{-2} \cdot sec^{-1}$	$1.67 \cdot 10^{15}$
Average neutron flux in BMA, per 1 source $neutron \cdot sec^{-1}$	$2.47 \cdot 10^{-3}$
Accelerator target unit	
External source intensity needed, $neutron \cdot sec^{-1}$	$7.9 \cdot 10^{17}$
Proton beam current needed, mA	7
Proton energy in beam, MeV	1000
Power released in target unit, MW	3
Thorium (second) blanket (TB)	
TB volume, m^3	0.569
Average neutron flux in TB circuit, $n \cdot sm^{-2} \cdot sec^{-1}$	$5.27 \cdot 10^{13}$
Thorium fraction in salt (TB), % mol.	30%
Heavy metals mass in TB circuit, ton	5.3
Heavy metals mass in TB, ton	1.6
Thorium consumption, kg/year	13.5
Thermal power in TB, MW	5.54
Average neutron flux in TB, $n \cdot sm^{-2} \cdot sec^{-1}$	$1.76 \cdot 10^{14}$
Average neutron flux in TB, per 1 source $neutron \cdot sec^{-1}$	$2.52 \cdot 10^{-4}$

Table 4 – reactor MYRRAH parameters

Fast neutrons flux (in fission products transmutation mode)	$\sim 10^{15}$ $n \cdot sm^{-2} \cdot sec^{-1}$
Thermal neutrons flux (in fission products transmutation mode) and in medical isotopes production mode	$\sim 2 \cdot 10^{14}$ $n \cdot sm^{-2} \cdot sec^{-1}$
Fast neutron flux in neutron source for materials investigation mode	$\sim 5 \cdot 10^{14}$ $n \cdot sm^{-2} \cdot sec^{-1}$
Full neutron flux for fuel researches	$\sim 10^{14} \div 10^{15}$ $n \cdot sm^{-2} \cdot sec^{-1}$
Proton beam current	5 mA
External source intensity	10^{17} $n \cdot sec^{-1}$

It should be noted that demonstration SMSR at thermal power 360 MW can produce about 100 MW electricity for accelerator supply. Also in thorium blanket it is possible to produce reasonable amounts of U-233 (10 kg/year) and Pa-233 (1.3 kg/year) for technological researches on their separation from molten salt.

Conclusions

Investigations performed show that multipurpose demonstration subcritical reactor with molten salt blanket can be realized [16]. This type reactors can be used in frame of MAYAK enterprise (Chelyabinsk region) for incineration of MA obtained when spent fuel is processed at RT-1 plant; and at GHK (Krasnoyarsk region) for radiochemical production wastes processing. In the present time Development and Demonstration Center is under construction, in future it is supposed to build RT-2 plant for spent fuel processing.

It should be noted that considered SMSR calculated parameters are rather realistic: for annual feed it is necessary to process 3-year full-power irradiated loading of VVER-1000 reactor. Heavy metals mass in circuit is 9.2 ton and it is equivalent to annual volume of MA separated from spent fuel from 23 reactors VVER type. It is comparable with MA amount that is stored in irradiated fuel, so it is possible to get after irradiated fuel processing enough MA to fill SMSR circuit. Thus SMSR considered is quite suited for introducing in fuel cycle.

References

1. A.Gandini, M. Salvatores. The Physics of Subcritical Multiplying Systems. Journal of Nuclear Science and Technology, vol.39, No.6, pp. 673-686 (June 2002).
2. M. Salvatores. Physics features comparison of TRU burners: Fusion/Fission Hybrids, Accelerator-Driven Systems and low conversion ratio critical fast reactors. Jour: Annals of Nuclear Energy, Vol. 36, 2009, p. 1653.
3. Molten salt Technology and decision of nuclear power poroblems. P.N. Alekseev et. al.. Preprint IAE--5906/2, Moscow, 1995 (In Russian).
4. P.N. Alekseev et. al. Conception of molten salt subcritical reactor with engreased safety. Preprint IAE-5857/2, Moscow 1995 (In Russian).
5. P.N. Alekseev, V.V. Ignatiev, O.E. Kolyaskin, L.I. Men'shikov, L.I. Ponomarev, N.N.Ponomarev-Stepnoy, S.A. Subbotin, A.V. Vasiliev, R.Ya. Zakirov. ‘Concept of the Cascade Subcritical Molten Salt Reactor (CSMSR) for Harmonization of the Nuclear Fuel Cycle’. In Proc. of GLOBAL' 99, Wyoming, USA, log#216, 1999.
6. A.V. Vasiliev, P.N. Alekseev, A.A. Dudnikov, V.V. Ignatiev, K.O. Mikityuk, S.A. Subbotin, R.J. Zakirov. Features of Cascade Subcritical Molten Salt Reactor – Burner of Long-Lived Radioactive Wastes, In Proc. of GLOBAL’2001, log#218, September 9-13, Paris, France.
7. Vasiliev A., Alekseev P., Dudnikov A., Fomichenko P., Mikityuk K., Subbotin S. Optimization of Conceptual Design of Cascade Subcritical Molten-Salt Reactor. Proc. of Int. Conf. “Advanced Reactors and Innovative Fuels”, ARWIF-2001, pp. 433-443. Chester, UK, October 22-24, 2001.
8. S. Buono. “Beam Target Design in Accelerator Driven Systems”. Proceeding of FJ/OH Summer School in Reactor Physiscs’2002, Aix-en Provence, Cadarache, France, August 21-30, 2002, on CD-ROM.
9. P. Alekseev, A. Dudnikov, P. Fomichenko, V. Nevinitsa, A. Sedov, S. Subbotin, A. Vasiliev, A. Voloschenko. V. Gagin, K. Mikityuk. Computational investigation of cascade scheme of subcritical molten salt reactor Preprint IAE-6280/4, Moscow 2003.

10. P. Alekseev, A. Dudnikov, P. Fomichenko, V. Nevinitsa, A. Sedov, S. Subbotin, A. Vasiliev, A. Voloshchenko. “Computational optimization of efficiency and safety of cascade scheme of the subcritical molten salt reactor”, Proceedings of “Nuclear Energy for New Europe ’2003”, Portoroz, Slovenia, September 8-11, 2003, pp 106.1-106.7.
11. P. Alekseev, A. Dudnikov, P. Fomichenko, V. Nevinitsa, A. Sedov, S. Subbotin, A. Vasiliev, A. Voloschenko. Studies of Physical Features of Cascade Subcritical Molten Salt Reactor with External Neutron Source. Proceeding of *PHYSOR 2004 -The Physics of Fuel Cycles and Advanced Nuclear Systems: Global Developments*. Chicago, Illinois, April 25-29, 2004, on CD-ROM, American Nuclear Society, Lagrange Park, IL. (2004), ID95513.
12. H. Aït Abderrahim, P. D'hondt. "MYRRHA: A European Experimental ADS for R&D Applications. Status at Mid-2005 and Prospective towards Implementation", Jour: Journal of Nuclear science and technology, Vol. 44, No. 3, p. 491, 2007.
13. H. Abderrahim. MYRRHA, A Multipurpose Accelerator Driven System for Research & Development. Proceeding of FJOH Summer School 21-30 August 2002, Cadarache, France.
14. Independent Evaluation of the MYRRHA project. Report by an international team of experts. OECD 2009, ISBN 978-92-6499114-9.
15. D. Bowman, R.P. Johnson. Accelerator for Subcritical Molten Salt Reactor. Proceeding of 2011 Particle Accelerator conference, New York, NY, USA, p2181-2183.
16. V.A. Nevinitsa, A.A.Dudnikov, V.Yu. Blandinskiy, A.L. Balanin, P.N. Alekseev**,** Yu.E. Titarenko, V.F. Batyaev, K.V. Pavlov, A.Yu. Titarenko Calculation Investigations of isotope equilibrium in demonstration subcritical molten salt reactor: procedure and results. VANT issue 1-2, p.44-50 2014 (in Russian).