Neutronic Analysis and Transmutation Performance of Th-based Molten Salt Fuels

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1. Introduction

Thorium fuel and thorium fuel cycles are attractive for the long-term nuclear energy production with low radioactive waste. The utilization of thorium in various reactors ranging from thermal to fast neutron energies has been successfully demonstrated, but substantial R&D is required before commercialization becomes possible. The molten-salt reactor (MSR) systems present the very special feature of a liquid fuel. MSR concepts, which can be used as efficient burners of TRU from spent LWR fuel, have also a breeding capability in any kind of neutron spectrum (from thermal to fast), when using the thorium fuel cycle. It has a very interesting potential for the minimization of radiotoxic nuclear waste.

2. Methods

This study is on a subcritical Accelerator Driven System with molten salt fuels. We considered three types of fuels based on thorium and compared the results on neutronic characteristics, transmutation of minor-actinides, breeding ratios, etc.

We evaluated the transmutation potential of different molten salt fuels (pure minor actinide + additionally Pu and Th), the breeding potential for Th-based fuels, and the safety characteristic which is temperature coefficient of such systems.



Fig.1. Concept of JAERI molten salt ADS (left) and a simplified geometry for the simulation (right)

2.1 Configuration of Geometry

JAERI did conceptual design studies on an 800 MWth molten salt target/blanket, accelerator driven system (ADS) for the nuclear waste transmutation.[1]

The present work adopted JAERI's conceptual design. Figure 1 shows the concept geometry and the simplified one for the simulation.

2.2 Molten Salt Fuels

We choose different molten salt fuels based on Li-Be fluoride, Na chloride and Pb chloride for analysis of the systems which have neutron spectrum from thermal to fast. We have changed the composition of Th, Pu, U, and minor actinides in the fuels to compare their neutronic characteristics, breeding, effective critical factors and transmutations of minor actinides. On the other hand, their chemical properties are not considered in this work. The Pu isotopes ratio is chosen 239 Pu : 240 Pu = 80 : 20 and the ratios of minor actinides considered are 237 Np : 241 Am : 243 Am : 244 Cm = 12.5 : 50 : 25 : 12.5.

2.3 Monte Carlo Simulations

For the simulation work, we used two Monte Carlo simulation codes, which are Energy Amplifier Monte Carlo (EAMC) and FLUKA. EAMC uses a point data analysis with JAR nuclear data library[3]. It simulates the system with neutrons produced by deuteron-deuteron fusion assumed to be in the center of the reactor, and calculates K_{eff} , the neutron flux and the number of reactions. FLUKA has the 260 group data library below 20 MeV neutron energy, based on ENDF.B-VII. FLUKA simulates the systems with 1 GeV 1mA proton beam impinging on the fuels directly. The calculated quantities with FLUKA are the distribution of the particles (neutron, photon, and charged particles), the energy distribution, and the neutron flux.

3. Results

The simulations have been done with the different molten salt fuel systems, and we compared neutron spectrum, fuel breeding, energy production ($K_{eff} \sim 0.98$), and transmutation of minor actinides.

The system of the thorium based fuel needs the thermal/epithermal neutrons to breed fissile fuel ²³³U. However, the transmutation of nuclear wastes which are minor actinides needs fast neutrons. The fission cross sections of minor actinides are usually bigger than the

capture cross section when the neutron energy is above several hundred keV. The aim of the effective criticality factor is 0.98.

3.1 Neutronic characterization of Molten Salt Fuels

We first used only the thorium based molten salt fuel without any seed fuels like U and Pu. Fig. 2 shows the neutron spectrum in the core region, Li-Be fluoride fuel has higher flux in thermal/epithermal regions than other fuels. Na- and Pb-chloride fuels have hard neutron spectrum. Because of the heavy mass of Pb so that the Pb chloride fuel has a higher neutron flux than Na chloride fuel in the fast neutron region. The neutron spectra show sharp decreases in the epithermal region where the neutron capture resonances of thorium exist.



Fig.2. Neutron spectra in the core region with different molten salt fuels

3.2 Molten salt fuels with 232 Th and ^{233}U for energy production

We changed the enrichment of 233 U for the study of the molten salt fuels for energy production. The enrichment has been changed till K_{eff} reaches at 0.98. Table I shows the enrichment and the neutron escape from the system. Li-Be fluoride fuels can get K_{eff} ~ 0.98 with much lower enrichment than Na- or Pb- chloride fuels. Because the Na- or Pb- chloride fuels have fast neutron spectrum so that these fuels' neutron escape values are 4 or 5 times much bigger than Li-Be fuels'.

Table I: ²⁵⁵ U enrichment at K _{eff} is 0	9.9	9)))	ļ))))))		,	-	5		1					•	•	•	•	•		•	•	•	•	•	•	•	•	•		•	•	•	•	•	•	•	•	•	•	•	•	•	•	•	•	ł,	I))]	ί	((5	5	2		Ĺ	i	1				¥	9	f	2	0	,			ζ	5]						t	t	t	t	1	ľ	l	1	3	8	2	έ	ć	ŝ	i	;					t	1	1	1	1	1	•	2	e	(l	1		ľ	1	1	ľ	I	1		l	1]	ŀ	ł]		;				(((
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Fuels	Enrich- ment	Neutron Escape	Tempe Coeff (PCN	erature icient M/K)
(% moles)	(%)	(%)	900K→ 800K	900K→ 973K
⁷ LiF+BeF ₂ +ThF ₄ +UF ₄ 64:18:15.6:1.74	13.3	0.54	-2.6	-3.2
⁷ LiF+BeF ₂ +ThF ₄ +UF ₄ 72.76:15:10.5:1.74	14.2	0.53	-3.2	-3.3
NaCl+ThCl ₃ +UCl ₃ 64:27.8: 8.2	22.8	2.15	0.3	-0.5
PbCl ₂ +ThCl ₃ +UCl ₃ 64:25:11	30.6	2.58	-0.1	-0.2

The temperature coefficient is one of the most important parameters of the nuclear reactor safety. The negative coefficient value is suitable for reactor safety. Most of resulting values are negative but Na-chloride has a positive value. This value cannot be serious for the safety because these systems are the accelerator driven subcritical systems so that they have a margin of the criticality for the safety.

3.3 Transmutation potential of Molten Salt systems

Fig 3 shows the fission per capture ratio of the different minor actinides in different molten salt fuels. This shows the transmutation of the minor actinides is higher in the fast neutron region than in the thermal/epithermal region so that Na-chloride or Pb-chloride fuels can be better fuels than Li-Be fuels, since the fission reaction is dominating over capture in the fast neutron region.



Fig.3. Fission per capture ratio of the minor actinides

3.3 Th-based fuels with Pu and Minor Actinides seeds

When the fuels have mixed Pu and MA, the thorium based fast molten salt fuels which are Na-chloride or Pb-chloride, have similar breeding capability as thermal/epithermal systems. The other characteristics depend on the spectrum those are similar behavior as above.

3. Conclusion

For a pure Th-U fuel, thermal/epithermal molten salt is better than a fast one. The system has better breeding ratio and can reach K_{eff} with smaller enrichment than fast system, because of the neutron leakage of the fast system. However the fast fuel salts is preferred for the transmutation of Minor Actinides. When mixed with MA and Pu, thorium based fast molten salts have similar breeding capability as thermal/epithermal salts.

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