Computational investigation of nuclear waste incineration efficiency in a subcritical molten salt driven by 50-100 MeV protons

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Abstract. Molten salt reactors were designed and operated at 1960s. The subcritical accelerator driven MSRs are being considered recently. In the present work, accelerator driven homogeneous subcritical core configuration was Modelled using MCNPX code. The composition of NaF-$^{233}UF_4$-ThF_4-$TRUF_4$ and $^{233}UF_4$-ThF_4-$TRUF_4$ was selected as the fuel loaded inside a 58×60 cm cylindrical core respectively. NaBF was selected as coolant salt of the fuel salt circuit. Accelerated proton particles were used to induce fission in the transuranic nuclei. The projectiles energy was changed from 50 MeV up 100 MeV in five steps. TRU fission rate, deposited heat distribution and neutron flux distribution were determined inside the subcritical core. Neutron and proton flux distribution inside the subcritical molten salt core was compared with each other. Source multiplication factor and safety parameters were calculated for any different projectile energy. Optimized proton energy was suggested to be applied for nuclear waste incineration using such system. Burn-up calculations were carried out for the cores with different fuel loading.

Key Words: Molten salt, Subcritical reactor, Accelerator driven, TRU incineration

1. Introduction

After the Molten Salt Reactor Experiment (MSRE) at Oak Ridge National Laboratory (ORNL) in the 1960s, some molten salt reactor (MSR) designs have been proposed to produce power or incinerate transuranic waste. In addition, several proposed thorium-cycle configurations were designed to minimize waste production while producing considerable amounts of power. The U/Th fluoride salt proposed in all of these designs, which could allow operating temperatures around 700 ˚C [1].

Accelerator-driven type of these MSRs could be regarded because of their inherent safety, higher accessible burn-ups and higher manageable neutron flux by means of higher beam current application.

The Accelerator Research Laboratory at Texas A&M University is developing a design for accelerator-driven subcritical fission in a molten salt core (ADSMS) to destroy the transuranic (TRU) content in used nuclear fuel (UNF) using a chloride based fuel salt. The ADSMS core is designed to operate with keff = 0.96 and is driven by spallation occurred in the homogenous actinide-trichloride–NaCl molten salt from an 800 MeV proton beam. The results obtained from this work show a 12-core ADSMS system could destroy the transuranic elements produced by a conventional GWe power reactor and produce 300MWe of additional power [1].

Zuokang et al. (2013) illustrated to advance the safety of the molten salt reactor; an external neutron source has been used in the core center. In addition, they indicated preparation of a conceptual design for an electron accelerator-driven molten-salt subcritical system gives confidence that such a system is technically feasible. Their results showed 150MeV electron with power 150kW will induce about 280kW fission power in the system when the system works at Ks=0.9891, while the conversion ratio (CR) of 232Th to 233U is 0.3836 [2].

Degtyarev et al. (2005) investigated the molten salt ADS-burner of minor actinides (MA). Their obtained results showed the CSMSR-burner with power of 800 MWth and subcriticality

Δk=0.05 can incinerate ~50 kg of MA per year, i.e. MA produced by 5 thermal reactors of the same power [3].

MOSART developed in Kurchatov Institute is a pure actinide burner aiming to demonstrate the feasibility of MSRs in reducing long-lived waste radiotoxicity and producing electricity in fully closed fuel cycle. MOSART operated in 2400 MWt can steadily incinerate actinides in a capacity of 72 kg per 30 days. It is an effective actinide burner and in overall can achieve 80% of the radiotoxicity reduction [4].

Molten salt reactors have numerous operational and safety advantages over solid fuel designs which in the following some of them will be briefly reviewed. Fluid nature of the fuel means meltdown is an irrelevant term and allows the fuel salt to be automatically drained to passively cooled, critically safe dump tanks. Noble gases bubble out and are stored outside the reactor loop. Most MSR designs have very strong negative temperature and void coefficients, which act instantly, aiding safety and allowing automatic load following operation [5].

Hence, feasibility study of a subcritical molten salt reactor driven by low-energy protons was proposed in this work.

2. Material and methods

In this work, MCNPX 2.6.0 has been used as a powerful particle transport code with the ability to calculate steady-state reaction rates, normalization parameters, neutronic parameters, as well as fuel burn up using CINDER90 to calculate the time-dependent parameters [6-7].

A cylindrical molten salt reactor was Modelled using the MCNPX 2.6.0 code. NaBF4-NaF was selected as a coolant for the fuel solution. A 3D neutronic model was set up using the MCNPX 2.6.0 code in cold zero power situations by means of ENDF/B-VI continuous-energy cross section. KCODE card was used for neutronic parameter calculations. The modelled core specifications are presented in Table 1.

Table 1 Core material and dimensions modelled using MCNPX 2.6.0

Core specifications	value	unit
Molten salt fuel 1: $^{233}UF_4$ (20%), ThF_4 (12%), $TRUF_4$ (68%)	4.5	g/cm^3
Molten salt fuel 2: $^{233}UF_4$ (21%), ThF_4 (2%), NaF (57%), $TRUF_4$ (20%)	4.5	g/cm^3
Hastalloy c-22	8.69	g/cm^3
Dimension	60×58	cm

A fixed dimension (60×58 cm) was selected for the Modelled cylindrical core, which the core volume is about 170 lit for the integrated core (Fig.1).

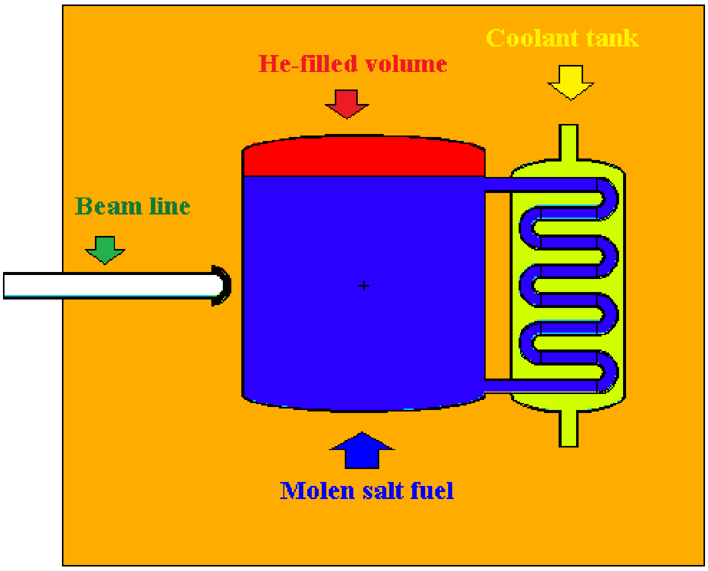

FIG. 1. Axial view of the Modelled subcritical accelerator driven core.

Accelerated proton particle was used to induce fission in the molten salt solution. Protons of 50-100 MeV was used to induce fission process inside the homogeneous core. ^{233}U content was selected so that produces an effective multiplication factor about 0.90. Radial and axial neutron flux distributions were calculated using the mesh tally card of the computational code. Deposited power distributions were calculated using the mesh tally card for the molten salt fuel loading. Reactivity coefficients of fuel, coolant, and moderator were calculated using the TMP card and temperature-related cross section library of .71c from endf70 in MCNPX. Void reactivity effect of the solution fuel was calculated for the different fuel loading in the Modelled core. Delayed neutron fraction and effective delayed neutron fraction were calculated.

Burn-up calculations were carried out for 5 years for the Modelled core exposed to I_{acc}(mA)×Deposited heat$_{fiss}$ (MeV) which is the subcritical core power (MW).

3. Results and discussion

The subcritical core driven by proton accelerator was considered for HLW transmutation. Clearly higher proton currents could produce higher fission rate and thereby deposited power. Proton flux leakage trough the subcritical core was avoided. The calculations showed proton current enhancement from 50 MeV to 100 MeV increases the fission produced deposited heat exponentially because of higher neutrons produced inside the tungsten target placed at the end of the proton beam line. Figure 2 shows the fission deposited power variation on the proton energy.

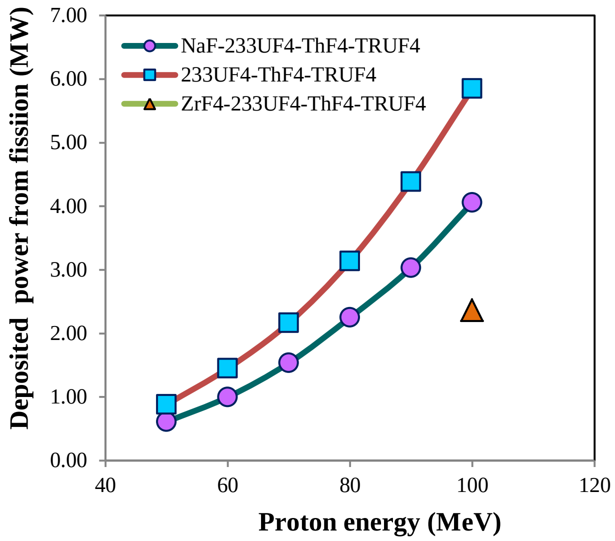

FIG.2. Deposited heat inside the Modelled core, 100 mA proton current.

Proton current of 100 mA and proton energy of 100 MeV was selected for driving the subcritical system.

Deposited heat inside the modelled subcritical cores was calculated using pedep mesh tally card. The proton beam line was positioned in front of the central plane of the core (Fig.3).

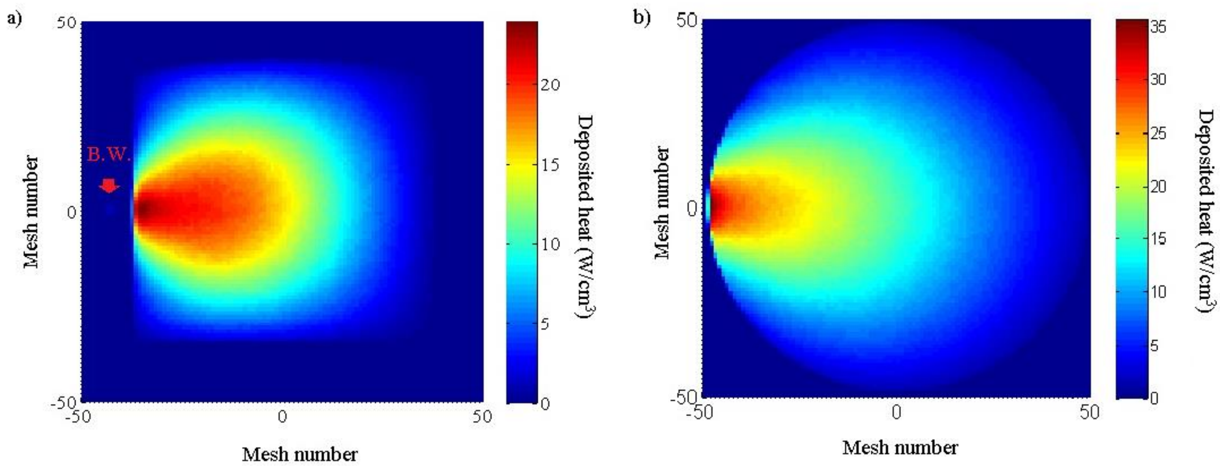

FIG.3. Deposited heat inside the modelled core, 100 mA proton current a) axial b) radial.

Neutronic parameters were calculated for the Modelled core fuelled with the different salts. According to data in Table 1, the core fuelled with the different salts experiences approximately identical delayed and effective delayed neutron fractions but neutron generation time is higher in the case of the core fed with ^{233}UF4-ThF$_4$-NaF-TRUF$_4$.

TABLE 1: NEUTRONIC PARAMETERS OF THE MODELLED SUBCRITICAL CORES

Neutronic parameters	$^{233}UF_4$ - ThF_4-$TRUF_4$	$^{233}UF_4$-ThF_4-NaF-$TRUF_4$
Effective multiplication factor	0.89940	0.90537
Neutron generation time (ns)	42	269
Beta (pcm)	318	361
Beta$_{eff}$ (pcm)	195	211
Fuel void reactivity coefficient (mk/K)	-6.6	-5.5
Fuel temperature reactivity coefficient (mk/K)	+0.0014	+0.0019
Fission per non-fission absorption ratio	1.54	1.53
Neutron per fission number	2.95	2.65

The carried out calculations showed a thermal neutron flux in order of 10^{13} n/s.cm^2 is available outside the Modelled cores. The thermal neutron flux intensity is higher in the edge; near to the beam line (Fig.4).

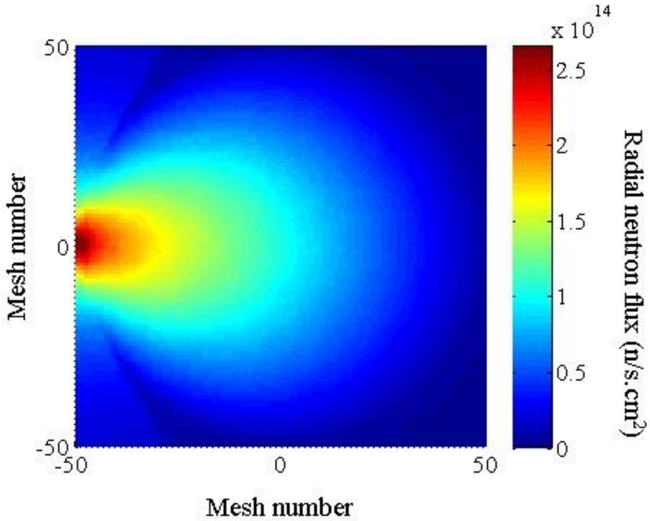

FIG.4. Thermal neutron flux distribution, 100 mA proton current.

Neutron spectra were determined in the core volume. The calculations were done per source particle per cm3 unit. As it is observed in Fig.5, the core fed with $^{233}UF_4$-ThF_4-$TRUF_4$ experiences harder neutron spectra than the other investigated one; which is more suitable for pure nuclear waste transmutation as a result of TRU fission process.

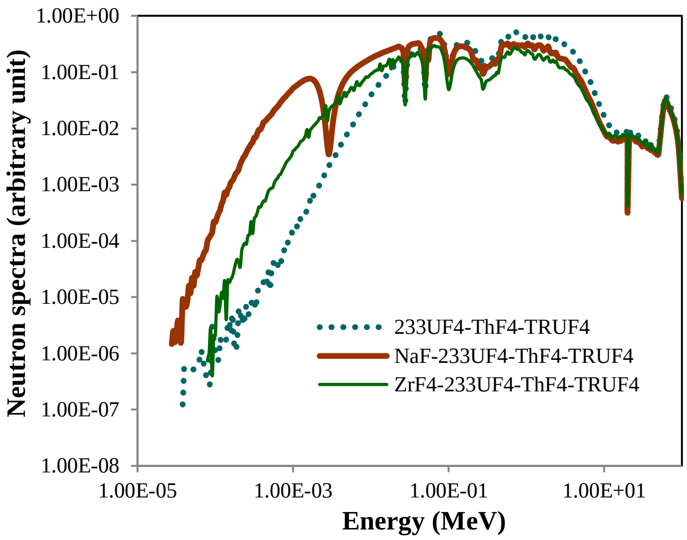

FIG.5. Comparison of neutron spectra for different core geometries.

Burn-up calculation was performed for 5 years using 4.05 MW power ($I_{accelerator}$ (mA)×$E_{deposited}$(MeV)) for ^{233}UF4-ThF_4-NaF-$TRUF_4$ fuel composition and 5.84 MW power for ^{233}UF4-ThF_4-$TRUF_4$ fuel composition. After 5-years burn-up, the core effective multiplication drops about 2000 pcm in the case of ^{233}UF4-ThF_4-NaF-$TRUF_4$ fuel loading and the value is 1400 pcm in the case of the second investigated fuel (Fig.6).

As is seen in Fig.7, $^{233}UF_4$-ThF_4-NaF-$TRUF_4$ loading achieves higher fuel burn-pus than the other investigated fuel.

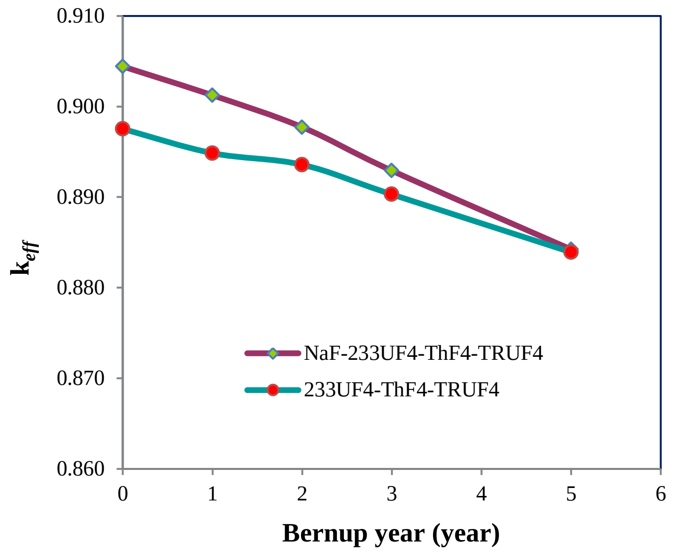

FIG.6. Comparison of the Modelled core effective multiplication factor variation during burnup.

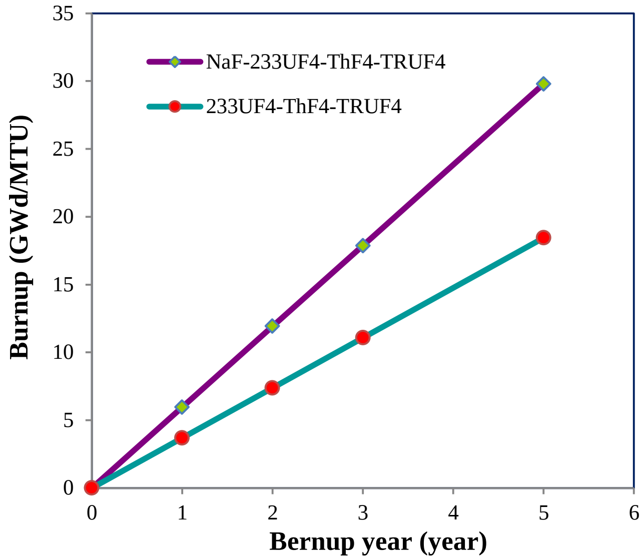

FIG.7. Comparison of fuel burn-ups of the Modelled core.

Table 2 presents first loading and the consumed values of the fissile and fertile isotopes. Utilization of $^{233}UF_4$-ThF_4-NaF-$TRUF_4$-based molten salt homogeneous core resulted in about 0.6 kg higher ^{233}U requirements to keep the same effective multiplication of the subcritical core fuelled with $^{233}UF_4$-ThF_4-$TRUF_4$ molten salt. Otherwise, the TRU transmutation efficiency is higher in the case of $^{233}UF_4$-ThF_4 -$TRUF_4$-based molten salt core.

TABLE 2: COMPARISON OF FIRST FUEL LOADING AND CONSUMPTION IN THE DIFFERENT CORES

Fuel type	Consumption (g)							First load of ^{233}U (g)
	^{232}Th	^{233}U	^{237}Np	^{241}Am	^{243}Am	^{244}Cm	Total transmuted TRU	
$^{233}UF_4$-ThF_4-NaF-$TRUF_4$	100	7300	2370	1710	700	2430	7210	12100
$^{233}UF_4$-ThF_4-$TRUF_4$	360	5200	5400	4310	1480	8990	20180	11500

4. Conclusion

The molten salt reactors are being taken in attention again because some of their vital characteristics such as higher efficiency due to higher operational temperature of them, low-pressure operation which remove the pressure vessel requirement and ^{233}U storage when they use in open cycle. The present study investigates neutronic performance of the subcritical accelerator driven molten salt reactor. According to the obtained results, the system has good efficiency for incineration of TRU. Geometry and thermo-hydraulic optimization could drive the system toward higher efficiencies for nuclear waste incineration.

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