Annals of Nuclear Energy 53 (2013) 1–8

Contents lists available at SciVerse ScienceDirect

Annals of Nuclear Energy

journal homepage: www.elsevier.com/locate/anucene

Depletion analysis on long-term operation of the conceptual Molten Salt Actinide Recycler & Transmuter (MOSART) by using a special sequence based on SCALE6/TRITON

R.J. Sheu [a,b,*], C.H. Chang [a], C.C. Chao [a], Y.-W.H. Liu [a,b]

[a] Institute of Nuclear Engineering and Science, National Tsing-Hua University, Hsinchu 30013, Taiwan, ROC
[b] Department of Engineering and System Science, National Tsing-Hua University, Hsinchu 30013, Taiwan, ROC

ARTICLE INFO

Article history:
Received 12 July 2012
Received in revised form 12 October 2012
Accepted 17 October 2012
Available online 22 November 2012

Keywords:
Molten salt reactors (MSRs)
Molten Salt Actinide Recycler & Transmuter (MOSART)
SCALE6/TRITON
Depletion analysis
Radiotoxicity

ABSTRACT

A special sequence based on SCALE6/TRITON was developed to perform fuel cycle analysis of the Molten Salt Actinide Recycler & Transmuter (MOSART), with emphasis on the simulation of its dynamic refueling and salt reprocessing scheme during long-term operation. MOSART is one of conceptual designs in the molten salt reactor (MSR) category of the Generation-IV systems. This type of reactors is distinguished by the use of liquid fuel circulating in and out of the core, which offers many unique advantages but complicates the modeling and simulation of core behavior using conventional reactor physics codes. The TRITON control module in SCALE6 can perform reliable depletion and decay analysis for many reactor physics applications due to its problem-dependent cross-section processing and rigorous treatment of neutron transport. In order to accommodate a simulation of on-line refueling and reprocessing scenarios, several in-house programs together with a run script were developed to integrate a series of stepwise TRITON calculations; the result greatly facilitates the neutronics analyses of long-term MSR operation. Using this method, a detailed reexamination of the MOSART operation in 30 years was performed to investigate the neutronic characteristics of the core design, the change of fuel salt composition from start-up to equilibrium, the effects of various salt reprocessing scenarios, the performance of actinide transmutation, and the radiotoxicity reduction.

© 2012 Elsevier Ltd. All rights reserved.

1. Introduction

The Generation-IV International Forum for the future development of nuclear energy systems has identified six most promising reactor types that could most likely meet the primary goals, such as nuclear safety, proliferation resistance, waste minimization, resource utilization, and economics (GEN-IV, 2010). The molten salt reactor (MSR) concept is one of the selected systems, which can be designed to operate in a thermal or fast spectrum reactor that produces fission energy in a circulating molten salt fuel mixture with a full actinide recycling. The concept is gaining more and more renewed interest due to its distinctive features and advantages (Forsberg, 2007). The fluid nature of fuel in MSRs gives extra flexibility in reactor design, fuel fabrication and recycling. However, it also complicates the core modeling and simulation because of strong coupling between core neutronics and salt reprocessing strategies. Most of conventional reactor physics codes were developed for solid–fuel core design and therefore not capable of taking liquid fuel flow and online reprocessing into account. To partly address this issue, this study aims to develop an automatic computational procedure allowing user to incorporate online fuel addition and salt reprocessing into a reliable neutronics and depletion analysis for MSRs.

Literature on the subject of MSR neutronics studies is not abundant when compared with other conventional or experimental reactor types. A relevant literature review (Vieitez et al., 2002; Ignatiev et al., 2003; Nuttin et al., 2005; Nagy et al., 2008, 2011, 2012) indicates that, regarding a reliable burnup calculation, an iterative procedure combining the capabilities of at least a neutron transport code and a material evolution code is absolutely necessary. The transport calculation evaluates neutron fluxes in spatial and energy distributions and prepares weighted cross sections corresponding to the current core configuration including geometry model and material composition. The material evolution code solves the system of nuclide chain equations resulting from neutron interactions and radioactive decays, and predicts time-dependent concentrations of a large number of isotopes in the core, and then feeds back to the following transport calculations. In contrast to solid–fuel reactors, additional modeling of online refueling and

* Corresponding author at: Department of Engineering and System Science, National Tsing-Hua University, Hsinchu 30013, Taiwan, ROC. Tel.: +886 3 5715131x42377; fax: +886 3 5739522.
E-mail address: rjsheu@mx.nthu.edu.tw (R.J. Sheu).

0306-4549/$ - see front matter © 2012 Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.anucene.2012.10.017

salt reprocessing in a MSR operation is indispensable and important, which adds extra complications to the calculation of nuclide concentrations. As to the authors' awareness and channels to access necessary computational tools for this study, there are several matured computer codes capable of performing accurate depletion analysis for solid–fuel reactors or capable of doing transport calculation alone very well. However, only few codes available in literature can perform reliable burnup calculations for a MSR due to its nature of liquid fuel circulation.

In order to examine the characteristics and performance of a MSR, one has to develop an appropriate computation method first. The methodologies presented in the literature are not described very clearly or in detail due to limited space (Vieitez et al., 2002; Ignatiev et al., 2003; Nuttin et al., 2005; Nagy et al., 2008, 2011, 2012). Most of them either resort to modifying or rewriting subroutines of an existing neutronics/depletion package or using a matured transport code coupled with an in-house material depletion code. Number of nuclides in consideration and proper treatment of problem-dependent cross sections are important to the accuracy of material depletion calculations. In this study, we proposed a relatively simple but reliable approach for performing MSR neutronics and fuel depletion analyses. The TRITON control module in the SCALE6 code system (ORNL, 2009, 2011) is considered as an accurate nuclide depletion engine. Taking advantage of its functionality, a coupling procedure was developed for controlling a series of stepwise TRITON calculations that can be used to approximate the operation of a MSR. The following section first introduces important features of SCALE6/TRITON relevant to this work and further adaptation for MSR applications. A brief introduction to the Molten Salt Actinide Recycler & Transmuter (MOSART), which is the target of this study, is also given in Section 2. Section 3 presents the result, an automatic calculation sequence developed for MSR fuel cycle analyses, and its application to MOSART. The purpose of the present study is twofold: to test and demonstrate the capabilities of the developed method and to reexamine the neutronic characteristics and performance of long-term MOSART operation. In addition, the coupled neutronics and fuel depletion tool will be useful in the investigations of the MSR systems of different type, such as various core designs to meet different goals, choice of fuel and molten salt solvent, effects of reprocessing schemes on reactor operation and performance.

2. Materials and methods

2.1. SCALE6/TRITON

The TRITON control module in SCALE6 has been designed to provide the combined capabilities of problem-dependent cross-section processing, rigorous treatment of neutron transport, and coupled with the ORIGEN-S depletion calculations to predict isotopic concentrations of material burnup. It presents a powerful and high-fidelity approach for many reactor physics analyses and has greatly enhanced the SCALE capabilities to handle increasing complexity in advanced fuel and core designs. Two major releases of the SCALE6 code system, versions 6.0 and 6.1 (ORNL, 2009, 2011), have been tested in this study and both versions can give very consistent results.

Currently, TRITON only supports multigroup depletion calculations. For the multigroup cross-section processing, BONAMI performs Bondarenko calculations for resonance self-shielding correction in the unresolved–resonance energy range. On the other hand, we chose CENTRM/PMC to process the cross sections in the resolved–resonance region, which based on rigorous discrete-ordinates spatial solution of neutron flux is more accurate than traditional options. TRITON uses a predictor–corrector algorithm (DeHart, 2009) to perform accurate fuel burnup. The T6-DEPL calculation sequence in TRITON was selected to do the depletion analysis in this study, where the three-dimensional Monte Carlo transport code KENO-VI is used in tandem with point-depletion and decay analysis module ORIGEN-S. All calculations were based on the most updated cross-section library v7-238 (238-group ENDF/B-VII) in the package.

To preserve maximal accuracy during a depletion calculation, the option PARM = (addnux = 3) in SCALE6.0 (ORNL, 2009) or PARM = (addnux = 4) in SCALE6.1 (ORNL, 2011) should be specified for inclusion of most additional nuclides in transport updates for depletion. In SCALE6.0, the default ORIGEN-S library is not suitable for fast reactors, because the library for cross sections not updated by the transport calculation was mainly developed for LWR applications and also it uses a cross section normalized to the thermal flux. Since effective actinide burners usually prefer spectra in fast neutron energies, the only fast reactor ORIGEN-S library named "abtrx" should be used in the TRITON depletion calculations (DeHart, 2009). This library applies cross section normalization to the total flux instead of thermal flux. It also contains fast fission product yields and the cross sections are weighted according to a typical fast reactor spectrum. In this regard, SCALE6.1 shows a significant improvement. The selection of an ORIGEN-S library with the appropriate neutron spectrum and fission yields for the application has been eliminated since their effects have been taken into account and automatically applied in the depletion calculation (Gauld, 2011).

2.2. Adaptation for MSR applications

As described above, SCALE6/TRITON built with state-of-the-art methodologies and most updated cross-section libraries is a great tool for performing reactor physics analyses on most solid–fuel core designs. Using it to investigate the neutronic properties of a start-up or static MSR core is straightforward; however, it cannot be directly applied to tracking the subsequent core behavior without proper modeling of the online fuel feeding and salt reprocessing, which is the most distinct feature of a MSR operation.

To reasonably simulate this time-dependent scenario, it is intuitive to divide the entire period of reactor operation into a series of consecutive operation steps. At the dummy gap between two steps, core compositions are to be adjusted to reflect the operation of attached salt reprocessing units. This part of manipulation was decided to be done by a small external program. Also we have to build a run script to control the execution logics and automate the process of each calculation step. In this study, the automation has been designed and realized based on three convenient features of SCALE6/TRITON, they are "Standard Composition Restart Files", "External Input", and "SHELL script". At the end of the last ORIGEN-S calculation in TRITON, standard composition files listing the nuclides and their atom densities are created for each mixture being depleted in the problem. Although ORIGEN-S is able to track more than 2000 isotopes in its depletion and decay analysis, a restart using the standard composition file is limited to maximum 388 isotopes by specifying PARM = (addnux = 4) in SCALE6.1. It is not a big concern to this study partly because these 388 isotopes already include nuclides whose cross sections significantly impact the transport solution and partly because most of the missing isotopes between the two sets are short half-lives. The output composition files will be automatically altered or updated by an auxiliary program following a user-defined refueling and reprocessing recipe. Using SCALE's external file reading capability, these modified composition files can then be fed into a follow-on calculation that relies on the previously calculated results and user requested intervention. In addition, the SHELL script that can be embedded in a TRITON input is also essential for our implementation especially

in data transfer and file management. In the end, a run script in DOS/UNIX environment is necessary to activate the auto-run sequence by repeatedly linking our material adjustment program with standard TRITON depletion calculations.

2.3. MOSART

Researchers in Kurchatov Institute of Russia have devoted to the study of a conceptual design of molten salt reactor called Molten Salt Actinide Recycler & Transmuter (MOSART) (Ignatiev et al., 2003, 2005, 2007). The MOSART concept uses a single-fluid system fueled with transuranic trifluorides from LWR spent fuels. It emphasizes on the performance of actinide burning without U/Th support that offers several advantages, such as maximizing actinide transmutation efficiency, simplified core configuration and easier fuel salt reprocessing. These features are quite attracting for an effective nuclear waste burner. Several combinations of core and reflector design, fuel and solvent compositions, operation and reprocessing schemes have been explored and reported in references (Ignatiev et al., 2003, 2005, 2007). In this work, as a demonstration and crosscheck of our developed method, some studies on neutronic characteristics of a preferred MOSART design were repeated and examined first. Furthermore, a series of depletion analyses on the long-term operation of MOSART was conducted with a focus on the effects of different fission product removal scenarios.

This paragraph briefly summarizes a simplified version of the MOSART core that was adopted in this study. The preferred design is a homogeneous core without graphite moderator because of its great performance on actinide incineration and simple core configuration. It operates at a power level of 2400 MWt with an effective core volume of 40.4 m^3 (equivalent to a cylindrical core with both diameter and height of 3.7 m) (Ignatiev et al., 2007). The radial thickness of graphite reflector is 20 cm. The average salt temperature in core is assumed to be 930.5 K. Fuel carrier is a molten salt mixture of 15LiF-27BeF_2-58NaF in mole percent (mol%). The content of feeding fuel has a typical composition of plutonium and minor actinides reprocessed from spent fuels of a commercial PWR (60 GWd/MTU, 4.9% enrichment, and after 1 year cooling) (Ignatiev et al., 2003). Table 1 lists the compositions of initial fuel and molten salt used in this study. Total mass of initial transuranic loading is about 2409 kg. The start-up concentration of transuranics in the fuel salt is about 0.5 mol%. In order to keep the core critical with low fuel inventory and minimum excess reactivity, fission products must be removed continuously or batch-wise during operation. Depending on the chemical processes involved, their removal efficiencies are actually element dependent. For simplicity in calculations, we assumed three categories for fission product clean-up: 100% removal efficiency for gas fission products by helium purge; 100% removal efficiency for non-soluble noble metals by plating out on surfaces; and about 300 EFPD (Effective Full Power Days) removal cycle time for soluble fission products (Ignatiev et al., 2003). Table 2 details the assumed reprocessing scenario for fission product removal during the MOSART operation.

3. Results and discussion

3.1. An automatic calculation sequence for MSR neutronics analyses

The operation of a MSR with online refueling and salt reprocessing was approximated by a series of stepwise TRITON calculations in this study. This approach should be a good approximation if the duration of each step, which is controlled by user, is short enough. To link these stepwise TRITON calculations as illustrated in Fig. 1, we have developed three auxiliary programs and a run script to facilitate the building of a simulation sequence. Between two consecutive steps, a FORTRAN program called *REFRESH* will be first invoked to make a step change of core composition reflecting the results of fuel feeding and fission product removal. This program basically performs three operations: after browsing the restart composition file from the calculation of previous step, it modifies atom densities on a nuclide-by-nuclide basis following a prescription defined by user; it also re-calculates total mass of actinides in the core and gives a correct specific power in MW per metric ton of heavy metal; then it edits and creates a new TRITON input file with updated content for the next-step calculation. After TRITON completes its iteration of transport and depletion calculations at this step, another program named *KEFF* is responsible for collecting useful data from the TRITON outputs including effective multiplication factors (k_{eff}) and isotopic concentrations for each sub-interval of the step. The last k_{eff} is used to determine whether the amount of fuel added at the beginning of this step is proper or not. The goal is to keep the core critical with reasonably minimum excess reactivity. The third program called *CRITICAL* is designed to test a criticality criterion, which is also controlled by user, and if

Table 1
Compositions of initial fuel and molten salt adopted in the study of MOSART.

Fuel	Plutonium and minor actinides								
Isotope	^{237}Np	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am	^{243}Am	^{244}Cm
wt.%	6.42	3.18	43.90	21.27	13.52	7.88	0.55	2.33	0.92
Salt	LiF-NaF-BeF_2								
Element	Li	Na	Be	F					
wt.%	2.54	32.57	5.94	58.94					

Table 2
Assumed reprocessing scenario for fission product (FP) removal during the MOSART operation.

FP category	Element										Removal
Fission gas	He	Kr	Xe								All
Noble metal	Ag	As	Cd	Ga	Ge	In	Mo	Nb	Pd	Rh	All
	Ru	Sb	Se	Sn	Tc	Te	Zn				
Soluble FPs	Ce	Cr	Dy	Er	Eu	Fe	Gd	Ho	La	Nd	300 EFPD[a] cycle
	Ni	Pm	Pr	Sm	Tb	Y	Zr				
	Ba	Cs	I	Rb	Sr						

[a] EFPD stands for Effective Full Power Days.

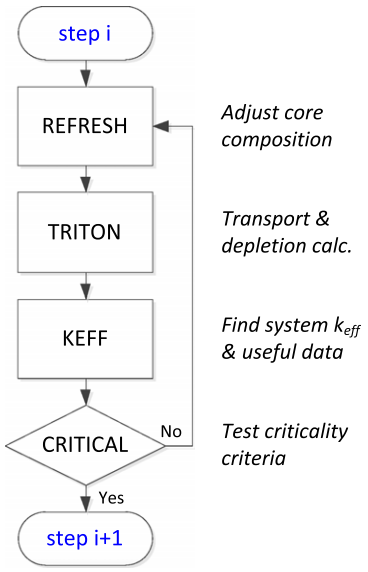

Fig. 1. Program flow chart of simulating the operation of a MSR with salt reprocessing.

necessary to give a revised amount of fuel addition for this step and go through the loop again.

To integrate the programs shown in Fig. 1, a run script named *simMSR.bat* has been developed to control the whole simulation sequence by iteratively calling *REFRESH* for composition adjustment and calling *batch6.bat* or *batch6.1.bat* for executing a TRITON depletion calculation as well as calling *KEFF/CRITICAL* for determining whether the current calculation step is acceptable. The calculation sequence can also be operated in manual mode where the automatic criticality search is turned off. The amount of actinides added at each step and the removal efficiencies of fission products are isotope-dependent and time-dependent, and can be altered by user for the upcoming calculation steps even after starting the simulation sequence. The method presented here is straightforward, accurate and easy to implement without any modification of the existing codes in SCALE6. Therefore, this approach can conveniently extend the applicable range of TRITON to MSR studies, while retaining all the original features and capabilities of SCALE6.

3.2. Neutronic characteristics of the MOSART core

Modeling the MOSART core configuration in TRITON is simple since it is essentially a tank of molten salt homogeneously mixed with certain amount of plutonium and minor actinides. By using the developed calculation sequence, we have successfully simulated the MOSART operation in 30 years with emphasis on tracking the changes of core composition as a function of time. At startup, the effective multiplication factor of the fresh core was estimated to be 1.03789 ± 0.00065 without considering the loss of delayed neutron fraction outside the core. The delayed neutron fraction of ^{239}Pu with fast neutrons is about 0.0020 (Stacey, 2007). During routine operation, there is always 28% of the fuel inventory that exists out of the MOSART core and in the primary loop, which means as a rough approximation the calculated eigenvalue should be subtracted by a small amount of ~0.00056 to account for the loss of delayed neutrons emitted in the primary loop.

To reach the goal of low fuel inventory and minimum excess reactivity in the core, the fuel salt must be reprocessed in either continuous or batch-wise mode to limit the neutron absorption by fission products to an acceptable level and to feed the core with fresh fuel to compensate the burnup effect. The time step of the calculation sequence in Fig. 1 was arbitrary set to 30 days for the first trial. Thus, 300 EFPD removal cycle for soluble fission products in Table 2 can be implemented in the calculation sequence by removing ~10% of fission products in the fuel salt at each step (assuming 300-day operation in a year). Under this condition, Fig. 2 gives an example of the MOSART operation showing the stepwise fuel addition and consumption for each 30-day calculation step. Since all the reactor power is coming from fission events, gross fuel consumption actually depends almost on the thermal power produced in the core. A fission event generates ~200 MeV of energy, which corresponds to roughly 1 GWd per kg of actinides being fissioned. The 2400 MWt MOSART core namely should transmute ~72 kg actinides in any 30-day operation step, which is quite close to that predicted by sophisticated TRITON calculations. Fig. 2 also shows the change of actinide concentration in the core and indicates the time needed for transition from non-equilibrium to near-equilibrium operation, which is about 20 years under the current model. The near-equilibrium state is defined here as reaching 98% of the maximum actinide concentration in the fuel salt. To compensate negative reactivities resulting from a buildup of fission product poisons, necessary amount of fuel fed into the core during the first few years is obviously larger than that required at equilibrium state. The concentration of actinides in the fuel salt therefore gradually increases from 0.5 mol% in the start-up core to ~1.04 mol% near the equilibrium state, which is well below the solubility limit of the fuel solvent used in this study (Ignatiev et al., 2003). The actinide and fission product concentrations in the fuel salt were also evaluated by using different cross-section libraries. The resultant concentrations of actinides near equilibrium were estimated to be 1.04, 1.10, 1.21 mol%, respectively, when using v7-238 (ENDF/B-VII), v6-238 (ENDF/B-VI), and v5-238 (ENDF/B-V) libraries. Meanwhile, the concentrations of soluble fission products in the fuel salt do not show obvious change in these calculations. Corresponding to the MOSART operation illustrated in Figs. 2 and 3 shows a good control of the resultant k_{eff} during the entire period of 30-year operation. Through automatic and careful control of fuel addition in the calculation sequence, effective multiplication factors are kept within a relative small range of 1.001–1.005 except for the initial k_{eff} that represents the start-up core configuration.

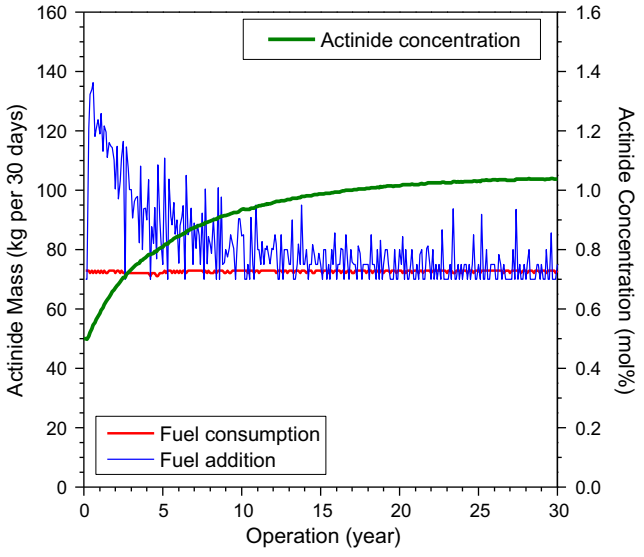

Fig. 2. An example of the MOSART operation showing the fuel consumption and addition for each 30-day step, as well as the actinide concentration in the fuel salt.

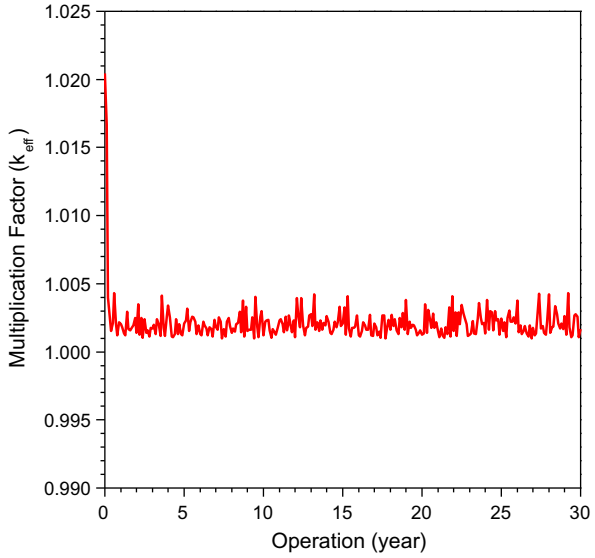

Fig. 3. Control of effective multiplication factors (or excess reactivity) within a user-defined range during a simulation of MOSART operation.

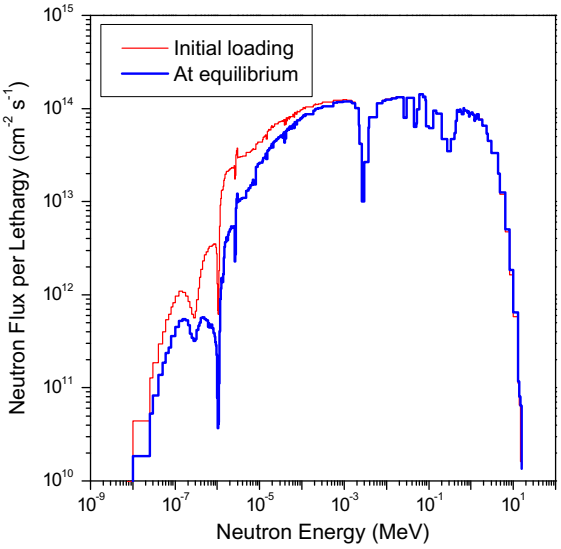

Fig. 5. Average neutron spectra of the MOSART core calculated with initial fuel loading and at equilibrium, respectively.

The composition of the fuel salt in MOSART is expected to change with time as a result of continuous fuel consumption and feeding during operation. The predicted changes in composition are given in Fig. 4 where shows a comparison of two core compositions in mass fraction, i.e. initial fuel loading and near equilibrium. Consequently, the spectrum shape of neutrons in the core will gradually change during its lifetime operation due to the evolution of core composition. Neutron spectra at each step can be extracted directly from the TRITON output. Fig. 5 compares two neutron spectra of the MOSART core with initial fuel loading and near the equilibrium state, respectively. MOSART is obviously a fast-spectrum reactor and the effective neutron flux at full power operation is about 1.2×10^{15} n/cm^2 s. As can be expected, the neutron spectrum in the core slightly shifts to higher energies as the actinide concentration in fuel salt increases with operating time.

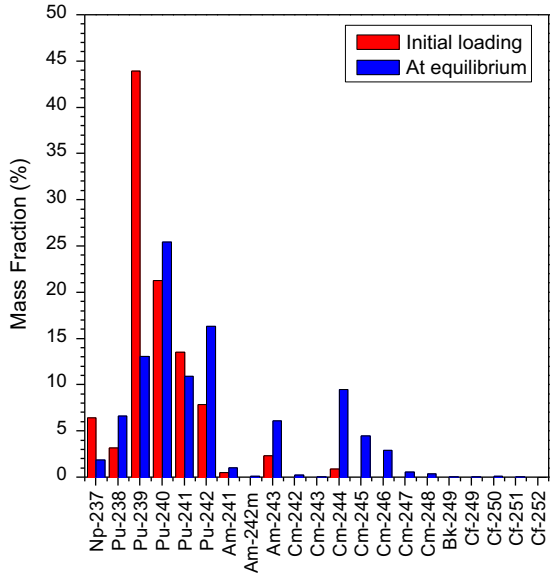

Fig. 4. Mass fraction of core compositions with initial fuel loading and at equilibrium, respectively.

3.3. Performance of actinide incineration and radiotoxicity reduction

The MOSART design conceived in Kurchatov Institute represents a promising option in response to the goals of future nuclear systems, especially in terms of effective transmutation of plutonium and minor actinides from LWR spent fuels. It aims at reducing the long-lived radiotoxicity of spent fuels discharged from the operation of conventional nuclear power plants. In terms of burning actinides, fast-spectrum reactor is more efficient. Therefore, the optimal design of MOSART is a core without graphite moderator such as the model in this work. Summing up the fuel addition and consumption of the 30-year MOSART operation in Fig. 2, a total of 24.4 metric tons of actinides are to be loaded and 89% of them (21.8 tons) are to be incinerated in the core. If the reactor operation is prolonged to 50 years, total actinide transmutation efficiency could be increased to 93%, within which the transmutation efficiency of plutonium is 96% and that of minor actinides is 68%.

Simple analysis of fuel burnup confirms that MOSART can effectively transmute actinides in a capacity of ~72 kg per 30 days. TRITON calculations enable us to look into the details. At the initial phase, the operation of MOSART almost solely relies on the consumption of ^{239}Pu. Following the accumulation of other actinides in the core, the burning of these isotopes is expected to become a notable source of power. Fig. 6 shows the details of actinide consumption during the operation of MOSART in 30 years. Although total consumption rate keeps nearly constant, the contribution of each nuclide varies with time. The consumption of ^{239}Pu drops quickly in the first few years and finally reaches an equilibrium consumption rate of ~34 kg per 30 days, which is about one half of its initial consumption rate. This difference is mainly compensated by burning of ^{240}Pu, ^{241}Pu, ^{237}Np, and ^{242}Pu. At equilibrium operation of MOSART, their respective consumption rates are estimated to be 15.4, 10.1, 5.0, and 4.3 kg per 30 days. Note that, during the first few years, net changes of some minor actinides such as ^{238}Pu, ^{243}Am, ^{244}Cm, and ^{245}Cm are actually accumulating in amount (with negative consumption rates in Fig. 6); however they only account for a small portion of actinide inventory and will quickly reach balance between production and destruction.

Besides focusing on actinide transmutation, it is also interesting and necessary to evaluate the radiotoxicity of the remaining fuel salt in the MOSART core after reaching its lifetime operation. The estimated result using ORIGEN-S is shown in Fig. 7, where an

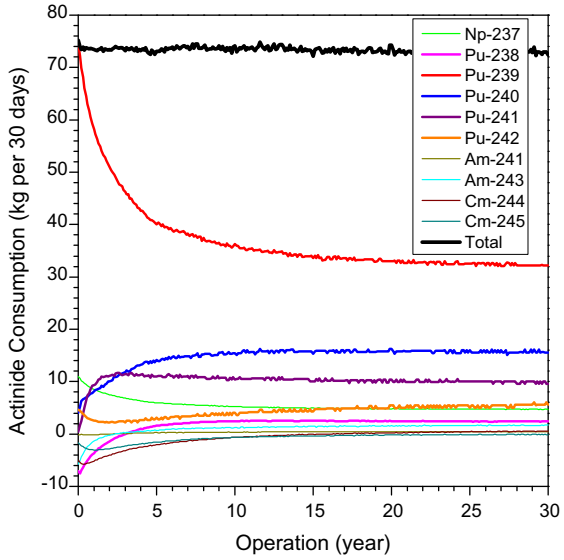

Fig. 6. Actinide consumption rates during the operation of MOSART in 30 years.

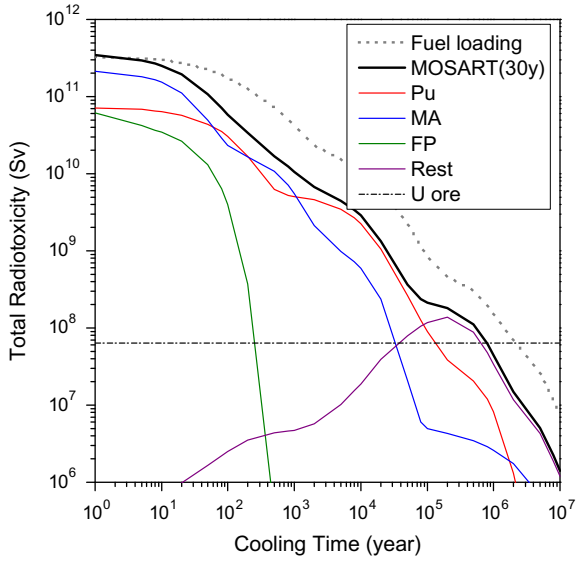

Fig. 7. Time evolution of the radiotoxicity of the remaining fuel salt in the MOSART core after 30-year operation.

equivalent radiotoxicity of the total fuel loaded in 30 years is also plotted for more objective comparison. The radiotoxicity (*Sv*) of a nuclide is defined as the product of its activity (*Bq*) and the corresponding dose coefficient per unit intake (*Sv*/*Bq*) adopted from ICRP-72 (1996). The line of radiotoxicity of uranium ore required to produce the MOSART fuel in 30-year operation is added in Fig. 7 for reference. The origins of total radiotoxicity can be conventionally split up into several parts: for example, plutonium, minor actinides, fission products, and the rest of nuclides. Minor actinides dominate initial radiotoxicity within 100 years. The plutonium will take over the primary contributor of radiotoxicity during 10^3–10^5 - years. After that, the rest of nuclides, mainly thorium, dominate radiotoxicity in most long-term perspective. In overall, approximately 80% of the radiotoxicity reduction can be achieved due to the consumption of a large amount of actinides in MOSART, and those incinerated actinides have been converted into enormous energy output and electricity.

3.4. Effects of reprocessing schemes on the MOSART operation

Automation of the calculation sequence in Fig. 1 is an important feature of the method. Repeated simulations to evaluate new designs or to test different parameters can be prepared and conducted with minimum user effort. For example, a selection of the step-length in Fig. 1 is artificial but it may affect the accuracy of the results. To evaluate the effect of step-length selection, we have carried out a series of similar simulations but with different step-length settings. The results obtained using step-lengths of 15, 30, and 60 days are presented in Fig. 8. Without changing the power level, the fuel consumption rate in MOSART keeps unchanged, for example ~36 kg per 15 days, ~72 kg per 30 days, or ~144 kg per 60 days. The amount of fuel added to the core also changes accordingly. The resultant actinide concentration in the fuel salt only shows small difference between these step-lengths, actinide concentrations at equilibrium are estimated to be 1.006, 1.038, and 1.043 mol% for 15, 30, and 60-day step-lengths, respectively. Shortening the step-length for a simulation has a penalty on the computing time. Its selection is therefore a trade-off between calculation effort and accuracy requirement. The 30-day step-length used in this study is considered to be appropriate to our purpose, as suggested by the comparison in Fig. 8.

Many complicated chemical processes have to be applied in salt reprocessing for a successful MSR operation (Uhlir, 2007). It is therefore interesting to explore different strategies for fission product removal from neutronics point of view. The method presented here is suitable for performing this kind of sensitivity analyses. Taking the reprocessing scenario in Table 2 as a template (called scenario A), we defined another scenario B which is mostly the same as scenario A except for eliminating the last row of soluble fission products in the table, i.e. excluding Ba, Cs, I, Rb, and Sr from the removal list. This modification was motivated by a paper of Ignatiev et al. (2003), in which the removal time for these elements was assumed longer than 30 years. Although this modification seems a small change, it turns out to be having a significant impact on the neutronics of MOSART. Fig. 9 shows the resultant actinide concentrations in the fuel salt as a function of operating time for two fission product removal scenarios (A and B). The result suggests that scenario B may not be workable for the MOSART

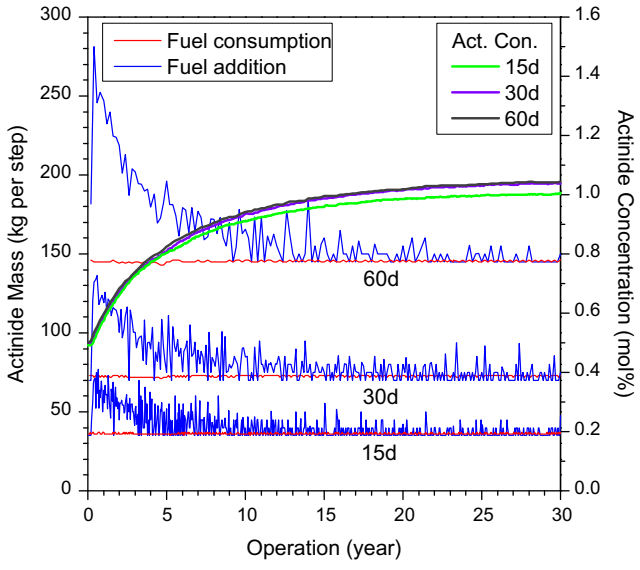

Fig. 8. Step-length effects on simulating the fuel consumption and addition, and the resultant actinide concentration in the fuel salt during the operation of MOSART in 30 years.

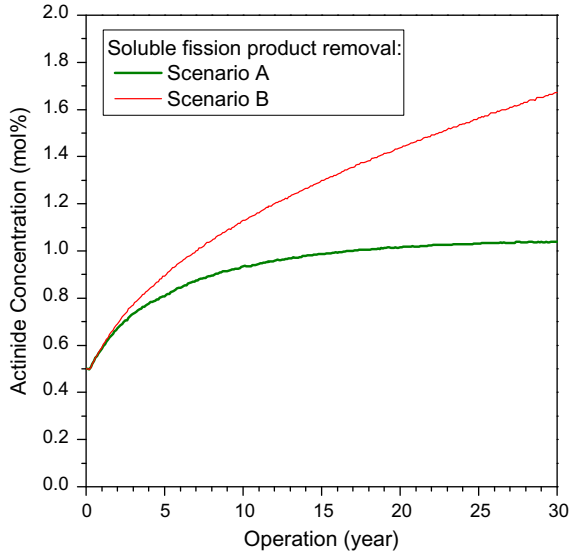

Fig. 9. Actinide concentration in the fuel salt as a function of operating time for two scenarios of fission product removal (A and B).

operation because the actinide concentration in the core continues to rise with time and may exceed the 2% solubility limit of the solvent in a long run (Ignatiev et al., 2003). Further sensitivity study indicates that, among these five elements, cesium is responsible for the difference between scenarios A and B as shown in Fig. 9. Without regular extraction of cesium isotopes in the salt, more fuel is necessary for compensating the negative reactivity caused by neutron absorption of cesium, especially ^{134}Cs and ^{133}Cs because of their relatively high fission yields and capture cross sections.

Removal of soluble fission products plays an essential part in a MSR operation. It helps reduce neutron absorption by fission product poisons and hold the concentrations of both actinides and fission products in the salt below the solubility limit. The effects of fission product removal rates on the MOSART operation were evaluated. Fig. 10 shows the actinide and fission product concentrations in the fuel salt as a function of operating time for various fission product removal rates, from 5% to 20% per 30-day step.

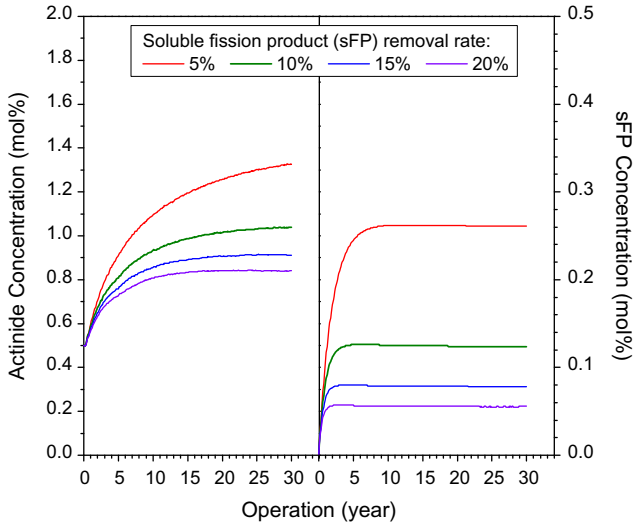

Fig. 10. Actinide and fission product concentrations in the fuel salt as a function of operating time for various fission product removal rates (5%, 10%, 15%, and 20% per 30-day step).

Faster removal rates result in a direct decrease of the concentration of fission products in the fuel salt. It improves neutron economics, reduces necessary amount of fuel fed into the core and therefore lowers actinide concentrations as well. Another influence worth noting is that it helps shorten the operating time needed for the core composition to reach equilibrium state. In reality, these advantages in core neutronics have to be weighted in balance with the increasing burden of salt reprocessing units and the operation costs.

4. Conclusions

Based on a series of stepwise SCALE6/TRITON calculations, an automatic computation and control sequence has been developed for MSR neutronics and depletion analyses. Its concept, implementation, and application were described and demonstrated in this paper. Simulations of long-term MSR operation with online fuel salt reprocessing can be easily achieved in accuracy and the automation of this calculation sequence is an important feature for its use and applications.

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. By using the developed tool, this study conducted a detailed reexamination of long-term MOSART operation including the neutronic characteristics of the core design, the change of fuel salt composition from start-up to equilibrium, the effects of various salt reprocessing scenarios, the performance of actinide incineration and radiotoxicity reduction. 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. TRITON analyses allow us to examine the details of actinide transmutation in the core. Depending on a prescribed fission product removal scenario, an appropriate amount of fuel added at each step can be determined though automatic criticality search. The resultant actinide concentration in the fuel salt as a function of time is one of the most important characteristics corresponding to a specific operation strategy of MOSART. It shows the transition of core composition from start-up to equilibrium and checks if the actinide concentration below the solubility limit of the selected solvent. Clean-up scenarios of soluble fission products in the fuel salt including species of elements and their removal rates affect neutron economics in the core and can have a significant impact on the long-term operation, in particular for those isotopes having high neutron absorption cross sections.

From neutronics and depletion perspectives, SCALE6/TRITON possesses the state-of-the-art methodologies and most updated and comprehensive cross-section libraries when compared with other approaches in similar studies. With the help of several in-house developed programs and a run script, we are able to use a sequence of stepwise TRITON calculations to approximate the operation of a MSR with continuous or batch-wise fuel feeding and salt reprocessing. Taking the analysis of MOSART operation as an example, the method developed here proves to be a useful and flexible tool that allows user to prescribe and control a simulation sequence for reactor physics analysis on a MSR operation, especially in terms of optimizing neutronics parameters and testing various reprocessing schemes.

Acknowledgements

This work is a follow-up of our previous preliminary study presented at the International Conference on Mathematics and Computational Methods Applied to Nuclear Science and Engineering

(M&C 2011). It was supported by National Science Council of Taiwan under contract No. NSC100-2923-E-007-002-MY3. The authors would like to thank Dr. Ignatiev and Dr. Feynberg of Kurchatov Institute for the introduction of the MOSART concept during their stay in National Tsing Hua University in Taiwan.

References

DeHart, M.D., 2009. TRITON: A Two Dimensional Transport and Depletion Module for Characterization of Spent Nuclear Fuel. ORNL/TM-2005/39, Version 6, vol. I, Sect. T1. Oak Ridge National Laboratory, Oak Ridge, TN, USA.

Forsberg, C.W., 2007. Thermal- and fast-spectrum molten salt reactors for actinide burning and fuel production. In: Proceeding of Global 2007, Boise, Idaho, USA.

Gauld, I.C., Radulescu, G., Ilas, G., Murphy, B.D., Williams, M.L., Wiarda, D., 2011. Isotopic depletion and decay methods and analysis capabilities in SCALE. Nucl. Technol. 174, 169–195.

Gen-IV, 2010. The Generation IV International Forum. <http://www.gen-4.org/>.

ICRP, 1996. Publication 72, Annals of the ICRP 26. Pergamon Press.

Ignatiev, V., Feynberg, O., Myasnikov, A., Zakirov, R., 2003. Neutronic properties and possible fuel cycle of a molten salt transmuter, 1873–1880. In: Proceeding of Global 2003, New Orleans, USA.

Ignatiev, V., Feynberg, O., Merzlyakov, A., Smirnov, V., Surenkov, A., Tataurov, A., Vanukova, G., Zakirov, R., Subbotin, V., Toropov, A., Panov, A., Afonichkin, V., Khokhlov, V., 2005. Integrated study of molten Na, Li, Be/F salts for LWR waste burning in accelerator driven and critical systems, Paper No. 027. In: Proceeding of Global 2005, Tsukuba, Japan.

Ignatiev, V., Feynberg, O., Gnidoi, I., Merzlyakov, A., Smirnov, V., Surenkov, A., Tretiakov, I., Zakirov, R., Afonichkin, V., Bovet, A., Subbotin, V., Panov, A., Toropov, A., Zherebtsov, A., 2007. Progress in development of Li, Be, Na/F Molten Salt Actinide Recycler & Transmuter concept, Paper 7548. In: Proceeding of ICAPP 2007, Nice, France.

Nagy, K., Kloosterman, J.L., Lathouwers, D., Van der Hagen, T.H.J.J., 2008. Parametric studies on the fuel salt composition in thermal molten salt breeder reactors. In: Proceeding of International Conference on the Physics of Reactors, Interlaken, Switzerland.

Nagy, K., Kloosterman, J.L., Lathouwers, D., Van der Hagen, T.H.J.J., 2011. New breeding gain definitions and their application to the optimization of a molten salt reactor design. Ann. Nucl. Energy 38, 601–609.

Nagy, K., Kloosterman, J.L., Lathouwers, D., Van der Hagen, T.H.J.J., 2012. The effects of core zoning on the graphite lifespan and breeding gain of a moderated molten salt reactor. Ann. Nucl. Energy 43, 19–25.

Nuttin, A., Heuer, D., Billebaud, A., Brissot, R., Le Brun, C., Liatard, E., Loiseaux, J.M., Mathieu, L., Meplan, O., Merle-Lucotte, E., Nifenecker, H., Perdu, F., David, S., 2005. Potential of thorium molten salt reactors: detailed calculations and concept evolution with a view to large scale energy production. Prog. Nucl. Energy 46, 77–99.

ORNL, 2009. SCALE: A Modular Code System for Performing Standardized Computer Analyses for Licensing Evaluations. ORNL/TM-2005/39, Version 6, vols. I–III. Oak Ridge National Laboratory, Oak Ridge, TN, USA.

ORNL, 2011. SCALE: A Comprehensive Modeling and Simulation Suite for Nuclear Safety Analysis and Design. ORNL/TM-2005/39, Version 6.1. Oak Ridge National Laboratory, Oak Ridge, TN, USA.

Stacey, W.M., 2007. Nuclear Reactor Physics, second ed. WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim, Germany.

Uhlir, J., 2007. Chemistry and technology of molten salt reactors – history and perspectives. J. Nucl. Mater. 360, 6–11.

Vieitez, E.R., Lowenthal, M.D., Greenspan, E., Ahn, J., 2002. Optimization of a molten salt transmuting reactor. In: Proceeding of PHYSOR 2002, Seoul, Korea.