MA Transmutation Performance Simulation and Accompanied Burning-up Analysis for C-ADS✩

Jilang Miaoa,*, Zichen Zhaoa, Zhenqi Changa,∗∗

✩School of Nuclear Science and Technology, University of Science and Technology of China, Hefei, Anhui, China 230027

Abstract

An accelerator-driven subcritical reactor functions well in incinerating high-level radiotoxic waste (HLW) as well as providing energy. China is on his way to establish such a facility to transmute the annual 1000 tons of HLW. A neutronic analysis has been performed for a reference core with a special task of burning minor actinides 237Np, 241Am, 243Am and 244Cm. Instant operation parameters are determined, including neutron energy spectra, thermal power distribution and transmutation validation. Burning-up analysis is carried out to further confirm the incineration efficiency. The core parameters optimized in this work will be applied to simulate in-core fuel behavior in future research.

Keywords: C-ADS, MA Transmutation, Neutronics, Burn-up

1. Introduction

Nuclear Power is one of the major sources of energy in the world. The installed capacity of the 436 nuclear power reactors operating in the world is 370 GWe and these provide nearly 18% of the worlds electricity [1]. However, along with the energy, a large amount of high level radioactive waste is discharged from the spent fuel every year. Reduction of burden caused by radioactive waste management is one of the most critical issues for the sustainable utilization of nuclear power. Without efficient transmutation, time evolution of the radiotoxic waste is shown in Fig 1. The toxicity depletion time is in the scale of million years [2]. An accelerator driven subcritical reactor (ADS) works well as a solution in that it destroys heavy isotopes contained in the used fuel from a conventional nuclear reactor, while at the same time producing electricity. The controllable neutron source generated by an accelerator also grants the system inherent safety.

In this work, an elementary core layout is es-

Figure 1: Time evolution of radiotoxic inventory in high-level waste (normalized by 1 ton of enriched uranium)
tablished for the to-be-constructed China ADS (C-ADS). Based on the core, runtime neutron behavior, thermal power distribution and transmutation performance after long operation time are worked out. MCNP5 is applied to simulate neutron behavior. And a self-developed BUMA code is applied to carry out burn-up calculations. The results are required for future irradiation damage investigation and thermal hydraulics and safety analysis.

2. Reference core and fuel description

2.1. Core structure, neutron source and structural materials composition

C-ADS consist of a high-energy proton accelerator, a Lead-Bismuth Eutectic spallation target and a subcritical core. The reactor core, a cylindrical assembly system, consists of 168 hexagonal prismatic fuel assemblies (FA), each of which contains 61 fuel elements (FE). Liquid lead-bismuth eutectic alloy is used as coolant for its low melting point, high boiling point and low neutron absorption cross section. Fuel assemblies and coolant in the core are radially and axially covered by a layer of lead reflector. The cross-sectional views of the core, a fuel assembly and a fuel element are given in Fig 2.

How a neutron generated from the spallation reaction behaves and its dependence on incident proton parameters is beyond current investigation. Isotropically-emitted neutrons generated from the target lateral surface along with those produced from fission function as neutron source in the model. Energy distribution of neutrons from the spallation target is approximated by a Maxwell energy spectrum with the temperature parameter 10 MeV. The FE cladding is made of ferritic martensitic steel (FMS) T91 [3], whose typical simplified composition is presented in Table 1.

| Element | wt% |
|---------|-----|
| Cr      | 9.0 |
| Mo      | 1.0 |
| V       | 0.2 |
| Nb      | 0.1 |
| Mn      | 0.5 |
| Si      | 0.4 |
| Ni      | 0.4 |
| C       | 0.1 |
| Fe      | 88.3 |

Figure 2: Radial schematics of the core
- Core Diameter: 2.21m, Height 2.20m
- Reflector thickness: 0.5m
- FA diagonal: 84.9mm
- FE \(R_i:6.00\text{mm}, R_o:6.96\text{mm}, \text{Height}:90\text{mm}
- Duty Ratio: 49.4%
Fuel pins loaded in a FE are assumed to be CERMET pellets. They are composite materials consisting of \((Pu_y,MA_{1-y})O_{2-x}\) particles dispersed in a metallic molybdenum matrix (called thereafter CERMET) [4]. Though in this calculation model, plutonium, minor actinides, oxygen and molybdenum are homogeneously distributed in fuel pins, their atomic fraction and fuel pin density are determined from the metal oxides and molybdenum densities and their weight fraction. Simulation of dispersed particles will be investigated in further research concerning fuel element and \((Pu_y,MA_{1-y})O_{2-x}\) particle tests.

2.2. Controlled fuel composition parameters

Fuel composition optimization is based on the isotopic compositions of LWR spent fuels in [5] [6] [7] since it is assumed that contemporary technology cycles spent fuels without distinguishing isotopes of an element. For the fixed isotopic composition (given in Table 2), weight fractions of the metal dioxides including PuO\(_2\), AmO\(_2\), and CmO\(_2\) are adjusted for better neutron performance. For a fuel pellet in the loaded CERMET fuel elements, metal dioxide microspheres and the molybdenum matrix account for nearly 40 vol% and 60 vol% respectively. Weight fractions among the metal oxides and the matrix are optimized for required neutron performance including subcritical multiplication factor \(k_s\), effective neutron multiplication factor \(k_{eff}\), flat power distribution, high MA transmutation efficiency and low fuel irradiation damage.

3. Computation tools

The calculations in this work are performed applying the probabilistic neutronic code MCNP5 and a coupled code system BUMA for rough burning-up analysis. MCNP5 yields instant results including effective neutron multiplication factor, neutron flux and various reaction rates averaged over specified number of transportation histories. The developed burning-up code with nuclear data compiled from evaluated libraries [8] provides an elementary view over time-dependent transmutation performance. In the calculation progress, MCNP5 gives spatial reaction rate distribution and BUMA uses MCNP5 output file to evolve fuel compositions in MCNP5 input files. A batch file calls the above two steps alternatively till the end of the transmutation cycle.

4. Instant transmutation efficiency and power distribution

The life time of a neutron history is found to be not greater than the scale of \(10^3\) shakes. And the current, flux, collision reactions accumulated over the history can be esteemed as instant values if divided by the time period or timed by the source generating rate. System performance under the above equilibrium conditions is believed to be a good basis for optimization analysis. And therefore in this work, instant transmutation efficiency and power distribution are analyzed first and then burning-up calculation is performed on the base of equilibrium cycles.

4.1. Determination of initial fuel composition

Initial fuel composition aims to result in sufficient future MA transmutation as well as maintain safe effective multiplication factor \(k_{eff}\), subcritical multiplication factor \(k_s\) and external neutron source efficiency \(\varphi^*\). \(k_s\), an important parameter for a subcritical system is defined as below [9]. From the neutron balance equation describing a subcritical system with an external source (Eq(1)), total fission neutrons \(F\) (Eq(2)), total source neutrons \(S\) (Eq(3)) can be determined, and \(k_s\) defined with \(F\)
Table 2: Initial isotopic composition

| Isotopic Vector | Atom fraction (at%) |
|-----------------|---------------------|
| Pu/Np/Am/Cm/Mo/O | 4.34/1.84/6.47/1.07/58.91/27.45 |
| $^{234}\text{U}/^{235}\text{U}/^{236}\text{U}/^{238}\text{U}$ | 0.0132/0.5793/0.3560/99.0516 |
| $^{238}\text{Pu}/^{239}\text{Pu}/^{240}\text{Pu}/^{241}\text{Pu}/^{242}\text{Pu}$ | 1.28/62.0/23.5/8.90/4.36 |
| $^{237}\text{Np}$ | 100 |
| $^{241}\text{Am}/^{242}\text{Am}/^{243}\text{Am}$ | 84.61/0.246/15.14 |
| $^{243}\text{Cm}/^{244}\text{Cm}/^{245}\text{Cm}$ | 2.01/90.01/7.97 |

and S (Eq(4)). And the relationship between $k_s$ and $k_{eff}$ is obtained as:

$$1 - \frac{1}{k_{eff}} = \varphi^* (1 - \frac{1}{k_s})$$  \hspace{1cm} (5) \hspace{1cm} [9]

Amount of Am and Cm is raised for more incineration and that of Pu is decreased to maintain $k_{eff}$ between 0.95 and 1 which helps reduce accelerator requirements. As mentioned above, weight fraction of the metal oxides and the molybdenum matrix in a fuel element pin is controlled as independent variables. And the initial fuel composition program is presented in Table 3. Volume fraction is estimated from the theoretical densities of the metal dioxides and molybdenum matrix. And taking into account the isotopic composition assumed in Sec.2.2, the fuel compositions in term of individual nuclides are obtained for MCNP input file.

Table 3: Initial materials composition of fuel element pins

| Materials | wt%  | vol%  |
|-----------|------|-------|
| $UO_2$   | 13.94| 13.49 |
| $NpO_2$  | 1.39 | 1.33  |
| $PuO_2$  | 14   | 12.92 |
| $AmO_2$  | 3.35 | 3.05  |
| $CmO_2$  | 2.24 | 2.03  |
| Mo       | 65.08| 67.19 |

Initial $k_{eff}$ for the core loaded above is estimated to be $0.963 \pm 0.001$. $k_s$, the multiplication factor with external neutron source is 0.992. And from Eq (5), external neutron efficiency $\varphi^*$ is 4.7. In fact, since the amount of Am and Cm accounts for less than 20% of the total amount of Pu and U, Am and Cm accumulation cannot compensate for the reactivity decrease resulting from U and Pu depletion. Therefore this initial loading program maintains stable $k_{eff}$ over operation time and is suitable for energy production.

In comparison, if the amount of Am and Cm accounts for too much of the fuel, neutron multiplication factor increases over time due to breeding of fissile nuclides like $^{242}\text{Am}$. Burn-up analysis suggests that $k_{eff}$ of the system approaches 1.00 within a month and such a program is not suitable for energy production. On the other hand, for an initial isotopic composition in which Am and Cm accounts for less than 10% of the amount of Pu and U, the duty ratio (fuel volume fraction) of the FAs cannot exceed 15% and reactor core efficiency will be decreased greatly.

4.2. Runtime neutron energy and spatial distributions

Incineration performance in the core mainly depends on flux level and neutron spectrum. In order to investigate the neutron fluxes and currents as time derivatives, the MCNP-yielding neutron influence and current, which are integrated over the cutoff time period and are averaged over all source histories, are timed by the product of the neutron yield of the spallation source per incident proton, $Y_{sp}$, and the proton beam current $I_p$. They are assumed to me $27.4 \text{n/p}$ and $8.1 \text{mA}$ respectively and the multiplier is $1.385 \times 10^{13} \text{n/s}$. With the multiplier, the results given by MCNP such as tally per particle are converted to current and reaction rates.

Neutron current over the target lateral surface, the reflector inner and external surfaces, the supposed surfaces surrounding 7 zones (The 168 FAs in Fig 2 are assumed to be arranged in 7 circles.) are given in Fig3. The neutron spectrum shifts toward high energy from the source surface to the inner reflector surface. Moderation of the lead-bismuth coolant and energy distribution of fission neutrons are responsible to the movement. In comparison with the spectra of source neutrons and those through the inner reflector surface, spectrum
of the neutron moving out shifts down apparently and it is caused by neutron leakage.

The moderation effect is exhibited more directly in Fig 3(b), spectrum shifts gradually from source to zone I, II, VII and finally contributes to the apparent shift of spectrum of neutrons entering the reflector or leaving the active zones.

### 4.3. Energy deposition and thermal power distribution

Power distribution can be determined from the fuel element heat-depositing reaction rates. Taking into account the core symmetry, radial power distribution is characterized with the line power density along one radius. As given in Fig 2(a), the 7 fuel assemblies located in the same radius are selected with 5 fuel elements along the same line to determine the line power density, which is presented in Fig 4. The radial power peaking factor is determined to be 2.18. Summing up the power density of the unique fuel elements, multiplied by repeating numbers and FE volume yields the total power, 451 MW with the assumed $Y_{sp}$ and $I_p$.

Besides the radial power distribution and peaking power factor, axial heat-depositing rate is required for further thermal hydraulics and safety analysis. One of the 7 fuel elements in one assembly in the 4th zone is taken as an example to show the axial power distribution. The thermal power profile in Fig 5 is based on a fuel element divided into 30 equal-volume horizontal sectors.

### 4.4. Minor Actinides transmutation validation

Energy-dependent neutron current or flux through separating surfaces illustrated in Sec 4.2 describes neutrons traversing the fuel elements and structural materials. And their spectra are responsible for reaction rates spatial distribution. As the detected surface moves outward the core, the spectra shift to low energy while at the same time, reaction rates of most concerned types decrease outward (Fig 7). The opposite shift direction of neutron spectrum and reaction rate can be accounted to the fact that neutrons at high energy contribute mainly to the concerned types of reaction, namely the transmutation of MA elements.

In addition, microscope fission, absorption and inelastic cross-section of MA elements decrease over the minimum value and reach a high plain above about 3MeV (Fig 6). The critical energy is consistent with the turning point in Fig 3. In conclusion,
though energy spectra shift to low energy in the core from center to reflector, reactions induced by high energy neutrons account for more weight and thus are responsible to the radial reaction decrease.

Figure 4: Radial line power density for the core

Figure 5: Axial power profile in a fuel element

(a) $\sigma_f$ of Am, Cm isotopes

(b) $\sigma_f$ of Pu isotopes

Figure 6: Fission cross-section of MA elements

MA transmutation can be further validated with the neutron spectrum (Fig 8) for the same core layout but with a thermal neutron source. Radial reaction rate decrease is also observed in the reference core. However, the energy profiles of neutron current through the target lateral surface, zone separating surfaces and reflector surfaces differ greatly.
from those presented in Fig 3. The spectra shift toward high energy especially after a similar turning point at about 1eV. Therefore, it can be concluded that reactions resulting from neutron at low energy predominate in the thermal core and MA transmutation performance is not promising.
5. Time-dependent and Burn-up analysis

5.1. Calculation approach

Instant transmutation results present nothing about how the fuel composition and transmutation rate vary with time. Burn-up analysis is necessary to confirm whether the obtained results are conserved over time and to optimize fuel composition and realize sufficient incineration of MA nuclides.

Evaluated nuclear data file libraries and MCNP are applied to obtain transmutation relationships (Fig. 9). Burn-up analysis in this work concentrates on MA, their mother and daughter isotopes in neutron reaction or decay, and fission products that have high neutron absorption cross section. Main MA isotopes remaining in spent fuel and need incinerating are $^{237}Np$, $^{241}Pu$, $^{242}Pu$, $^{243}Am$ and $^{244}Cm$. With neutron reaction cross section and decay half-life from IAEA nuclear data service [8] and reaction rates from MCNP pre-calculation, other nuclides like $^{239}Np$ and $^{238}Pu$ are added to the transmutation relationship. Fission products that are out of Fig. 9 but responsible for neutron absorption are $^{135}I$, $^{135}Xe$ and $^{149}Nd$. The resulted neutron poisons are $^{135}Xe$ and $^{149}Sm$. And radioactive transformation of the above light nuclides is also considered in calculation.

The temporal change of the atomic densities of a related nuclide $dN_j$ for a discrete time interval $\Delta t$ is evaluated in Eq. (6), where $N_{m,i}$ indicates atomic density of nuclide $m$ during time interval $i$, $\sigma_{m,b}^{(j)}$ indicates micro cross section breeding nuclide $j$ from reaction $b$ of nuclide $m$, $\sigma_{m,b}^{(j)}$ indicates micro cross section depleting nuclide $j$ from production of nuclide $n$ in reaction $d$, $\lambda_{i,b}^{(j)}$ indicates type $b$ decay rate of radioactive transformation from nuclide $i$ to $j$, and $\lambda_{j,d}^{(k)}$ is type $d$ decay rate of radioactive transformation from nuclide $j$ to $k$.

Take $^{244}Cm$ for example, \[
\frac{N_{Cm4,i+1} - N_{Cm4,i}}{\Delta t} = \int N_{Cm3,i} \sigma_{Cm3,(n,2n)}^{(Cm4)}(E) \Phi(E) dE - \int N_{Cm4,i} \sigma_{Cm4,(n,\gamma)}^{(Cm5)}(E) \Phi(E) dE - \lambda_{Am4,0}^{(Cm4)} N_{Am4,i} \lambda_{Cm4,0}^{(Cm4)} N_{Cm4,i}.
\]

Traditional burn-up calculation works out cross-section as group constants first and then the constants are applied for reaction rates determination. In this research, reaction rates of several concerned types are obtained from MCNP output files and group constant calculation is limited to fission yields. Therefore, calculation amount may be increased since Monte-Carlo reaction counts calculation is substituted for group constants determination but burn-up analysis precision can be obtained from Monte-Carlo results. Burning-up analysis is executed by alternative calling MCNP5 and the coupled codes system BUMA. Initial fuel isotopic composition and reaction rates along with the transmutation flow (Fig. 9) suffice to determine the isotope composition after the burn-up time step. MCNP5 works out reaction rates averaged over all the neutron histories of concerned types described above for the initial core composition. BUMA converts the MCNP5 results to time derivatives with special $Y_p$ and $I_p$ values and performs fuel composition variation (Eq. (6)). Continuing the above two steps will yield fuel compositions, neutron energy profile through concerned surfaces and cells, spatial thermal power distribution and MA transmutation rates as time-dependent variables.

5.2. Burning-up results

For the core with initial fuel composition as given in Sec. 4.1, the above burning-up analysis is performed for 365 days of stable operation. The temporal evolution of atom density for concerned MA isotopes is presented in Fig. 10. And based on the evolved FE composition, $k_{eff}$ and $k_s$ is calculated to confirm reactor core safety. Over the 365d operation period, $k_{eff}$ decreases gradually from 0.963 to 0.939 and $k_s$ remains approximately 0.994.

Two fuel elements from Zone II and Zone VII respectively are selected for evoluton data. Fig. 10(a)10(b).10(c) describe the evolution in the two FE of HLW isotopes given in Fig. 1. As expected from decreased reaction rates toward peripher-
\[
\frac{N_{j,i+1} - N_{j,i}}{\Delta t} = \sum_{m,b} N_{m,i} \sigma_{m,b}^{(j)}(E) \Phi(E) dE - \sum_{n,d} N_{j,i} \sigma_{n,d}^{(j)}(E) \Phi(E) dE + \sum_{b,l} \lambda_{j,b}^{(j)} N_{l,i} - \sum_{d,k} \lambda_{j,d}^{(j)} N_{j,i}
\] (6)

Figure 9: Transmutation flow of significant isotopes in burning-up calculation

5.3. Refueling of the core after incineration

Due to the decreasing radial reaction rate profile, fuel assemblies in peripheral zones of the core fail to experience sufficient transmutation and do not deserve subsequent procedure to separate nuclides. Therefore, the peripheral assemblies are moved inward and fresh assemblies are refueled from outside to realize deep incineration. After 365 days of evolution, 36 assemblies in the first 3 zones are replaced with 36 ones from the 7th zone. The other assemblies from the last 4 zones can be installed in a new core. Besides, the refueling program provides another approach for flattening thermal power profile in addition to applying more than one spallation source in the system [10], adjusting inert matrix ratio [11] and loading different types of fuel for different reactor core zones [12]. Thermal power for the refueled reactor is 415MW if proton beam remains constant but the power peaking factor fails to decrease obviously because the fuel difference caused by annual transmutation cannot make up for the spatial neutron spectrum difference. Effective neutron multiplication factor \(k_{\text{eff}}\) for the refueled system is determined to be 0.96, which is approximately equal to that of the initial core and the reactor is equivalently restarted for a new cycle. Without the refueling program, \(k_{\text{eff}}\) decrease can also be compensated by adding burnable poisons when starting the reactor [13] and burnable poisons utility will be confirmed for C-ADS after initial fuel composition and refueling programs are determined.
6. Summary and conclusion

A neutronic analysis of the simulated C-ADS reactor core with the composite CERMET fuel with Mo matrix, selected for MA transmutation, was performed aimed at optimizing the transmutation and neutronic performances. Runtime neutron energy profile and spatial distribution is presented. Thermal power of the core is determined to be 451MW with the radial power peaking factor 2.18. Neutron spectrum shift along with reaction rate variation validates the transmutation efficiency of MA elements. With an approximate burning-up analysis method, fuel composition evolution is presented for the 365d operation period.

7. Acknowledgement

Authors are grateful to National Nature Science Foundation of China for having funded this work through the grand No. 91226109.

References
[1] IAEA website. http://www.iaea.org.
[2] Hiroyuki Oigawa, Kazufumi Tsujimoto, Kenji Nishihara, Takanori Sugawara, Yuji Kurata, Hayanori Takei, Shigeru Saito, Toshinobu Sasa, and Hironari Obayashi. Journal of Nuclear Materials, 415(229-236), 2011.
[3] K. Haarmann et al. The T91/P91 Book. Vallourec & Mannesmann Tubes, 1999.
[4] F. Delage et al. Design, development and qualification of advanced fuels for an industrial ADS prototype, in: Proceedings of the Tenth OECD/NEA Information Exchange Meeting on Actinide nd Fission Product Partitioning and Transmutation, Mito (Japan), OECD/NEA, 2010, 352(pp 169-178), 6-10 October 2008.
[5] Wim Haeck, Edouard Malambu Mbala, Vitaly Sobolev, and Hamid At Abderrahim. Journal of Nuclear Materials, 352(285-290), 2006.
[6] V. Sobolev, S. Lemechov, N. Messaoudi, P. Van Uffelen, and H. At Abderrahim. Journal of Nuclear Materials, 319(131-141), 2003.
[7] Masayoshi KUROSAWA, Yoshitaka NAITO, Hiroki SAKAMOTO, and Toshiyuki KANEKO. JAERI-Data/Code, 1996.
[8] http://www-nds.iaea.org.
[9] Hesham Shahbunde, Cheol Ho Pyeon, Tsuyoshi Misawa, Jae-Yong Li, and Seiji Shiroya. Annals of Nuclear Energy, 37(1214-1222), 2010.
[10] C.H.M Broeders and I. Broeders. Nuclear Engineering and Design, 202(209-218), 2000.
[11] Kenji NISHIHARA, Kohei IWANAGA, Kazufumi TSUJIMOTO, Yuji KURATA, Hiroyuki OIGAWA, and Tomohiko IWASAKI. Journal of NUCLEAR SCIENCE and TECHNOLOGY, 45(8,p.812-822), 2008.
[12] Hiroshi SEKIMOTO, Sinsuke NAKAYAMA, Hiroshi TAGUCHI, and Tsuyoshi OHKAWA. Proceedings of ICAPP 10, Paper 10277.
[13] J.L. Kloosterman, H. van Dam, and T.H.J.J. van der Hagen. Nuclear Engineering and Design, 222(105-115), 2003.