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Monte Carlo calculations of the incineration of plutonium and minor actinides of laser fusion inertial confinement fusion fission energy (LIFE) engine

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Abstract

In this paper, neutronic analysis in a laser fusion inertial confinement fusion fission energy (LIFE) engine fuelled plutonium and minor actinides using a MCNP codes was investigated. LIFE engine fuel zone contained 10 vol% TRISO particles and 90 vol% natural lithium coolant mixture. TRISO fuel compositions have Mod①: reactor grade plutonium (RG-Pu), Mod②: weapon grade plutonium (WG-Pu) and Mod③: minor actinides (MAs). Tritium breeding ratios (TBR) were computed as 1.52, 1.62 and 1.46 for Mod①, Mod② and Mod③, respectively. The operation period was computed as ∼21 years when the reference TBR $>$ 1.05 for a self-sustained reactor for all investigated cases. Blanket energy multiplication values (M) were calculated as 4.18, 4.95 and 3.75 for Mod①, Mod② and Mod③, respectively. The burnup (BU) values were obtained as ∼1230, ∼1550 and ∼1060 GWd tM^{-1}, respectively. As a result, the higher BU were provided with using TRISO particles for all cases in LIFE engine.

Keywords: fusion, nuclear, Monte Carlo

1. Introduction

A fusion energy research continues to be an attractive option for the future with respect to safety, inexhaustible energy and environmental advantages. In the worldwide, the fusion energy research carried out with magnetic fusion energy (MFE) and inertial confinement fusion (ICF). The MFE involved confining plasma with a low density deuterium and tritium (DT) in a magnetic field area to provide energy from fusion. The ICF uses lasers, heavy ion beams, or pulsed power systems. The ICF system is compressed as capsule shaped in mixture of DT ice and gas. The D–T fusion reactions generated fusion energy by both alpha particles and 14.1 MeV neutrons [1]. Laser fusion inertial confinement fusion fission energy (LIFE) engine system developed at Lawrence Livermore National Laboratory (LLNL) uses laser-driven ICF to generate energy and electricity [2–8]. In this study, LIFE engine concept developed at LLNL provides an attractive pathway to incineration/transmutation of nuclear waste fuels containing minor actinides, plutonium and fissionable fuels [6–10]. Nuclear waste fuels can be examined in two groups:

(I) Reactor/Weapon grade plutonium: Reactor grade-Pu [11] and weapon grade-Pu [12] are generally intended to reduce due to the accidental release of radiotoxic material to the environment, political concern, prevent misuse, and serious public. So, the plutonium must be kept under forceful security control. Recently, the main target is to reduce plutonium of nuclear waste as much as possible [11–14].

(II) Minor actinides (MAs): The initial nuclear fuel will be converted to other heavy metal species by various nuclear reactions such as, (n, γ), (n, 2n), (n, p), (n, α), etc. These are named as actinides (MAs). MAs have very long radioactive half-lives compared to geological times. Today's existing nuclear fission plants generate significant amounts of nuclear waste in the form of MAs, which has hazard for future generations. For example, ^{245}Cm and ^{242m}Am of MAs have very high thermal fission cross sections. These isotopes as an alternative of high quality nuclear fuel and prolific material has been considered [11, 13, 14].

Previous studies, LLNL scientists and the other author/s studied above solid fuel and molten salt concept of LIFE engine. Kramer *et al* [2] developed the baseline design of LIFE engine. The LIFE engine design as fuel was used depleted uranium with a flowing flibe coolant. Moses *et al* [3] studied a sustainable nuclear fuel recycle based on LIFE engine. Moses *et al* [3] studied to eliminate of the need for fuel enrichment, increasing energy content of the nuclear fuel, to determine subcritical blanket ($k_{\text{eff}} < 0.90$). Abbott *et al* [4] discussed physical properties as structural stresses, flow dynamics, temperatures, power cycle, and first wall behaviour. A systems model including the laser, inertial fusion target, fission and tritium and energy multiplication of LIFE engine was developed by Meier *et al* [5]. Kramer *et al* [6] utilized MCNP neutronic code, ORIGEN2 and Monteburns to determine the nuclear behaviour of LFHR. Farmer *et al* [7] performed neutronic analysis of LIFE engine to utilize of WG-Pu and highly enriched uranium with TRISO particles. They pointed an attractive pathway for burning excess weapons grade Pu with TRISO particles. Molten salt fuel version of LIFE engine was descripted by [8] and molten salt fuel type (UF_4 and ThF_4) physical properties were given. The LFHR having thin $Li_{17}Pb_{83}$ coolant and coolant zone consisting of Flibe for burning minor actinides and reactor grade plutonium were investigated by Şahin *et al* [9, 10]. The calculations were performed by one dimensional SCALE 5 neutronic code. The fissile burn up (BU) exceeded 400 000 GWd tM^{-1} over 8 years both minor actinides and reactor grade plutonium. Fissile fuel breeding and radiation damage behaviours of SS-304 first wall using various molten salt fuels (ThF_4 and $^{233}UF_4$) in modified LIFE engine with one dimensional SCALE 5 neutronic code were carried out by [15]. Şahin *et al* [16] investigated the main parameters of a laser driven thorium breeder with MCNPX-2.7.0 and the neutronic calculations were applicable to a static system.

In this paper, time dependent Monte Carlo calculations for neutronic performance using Mod①: RG-Pu, Mod②: WG-Pu and Mod③: MAs in the kernel of the TRISO particles in a new modified natural lithium cooled modified LIFE engine were performed. The variations of the tritium breeding ratios (TBR), *M*, BU, fissile and fissionable isotopes were obtained and compared among themselves. In addition, the obtained results were compared with literature.

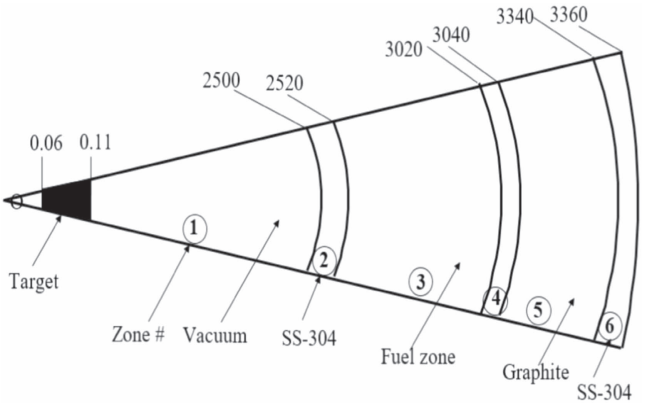


Figure 1. Cross sectional view in modified LIFE engine [15].

Table 1. Characteristics of layout of the Pu fuel spheres [17].

Properties	Values
Fuel kernels diameter	0.024 cm
PuC density	13.6 g cm^{-3}
Coating layers materials (inner to outer)	C/C/SiC/C
Layers thicknesses	95/49/35/40 μm
Layer material densities	1.05/1.9/3.18/1.90 g cm^{-3}

Note. Reference [13] has used PuO_2 as fuel.

2. Problem description

The LIFE engine concept idea has been studied at LLNL [2–8]. The LIFE engine design was 5 m diameter of spherical fusion chamber.

This concept has a thin $Li_{17}Pb_{83}$ coolant behind an oxide dispersion strengthened ferritic steel first wall, followed by a beryllium multiplier, coolant zone consisting of Flibe molten salt, and a carbon reflector. The LIFE engine concept details were given in [7, 8].

However, beryllium multiplier of LIFE engine design concept studies was ignored to avoid local neutron spectrum softening. A new concept design as difference of LLNL-LIFE engine was considered by [15].

The modified LIFE engine concept main components and cross sectional view in the present work was shown in figure 1. Also, the natural lithium in the present work was used as coolant material. A (D–T) fusion fuel pellet at LIFE is compressed to $\rho = 600$ gr cm^{-3} density for a fuel shell thickness of 0.005 cm.

The neutronic analysis in a modified LIFE engine using RG-Pu, WG-Pu and MAs were invesitaged. TRISO particle is a new type fuel structure, developed for high temperature reactor. The dimensions TRISO particles inspired from [17] were presented in table 1. The compositions (wt%) used in the calculations were taken as 4% ^{239}Pu and 6% ^{240}Pu for WG-Pu and 1% ^{238}Pu, 62% ^{239}Pu, 24% ^{240}Pu, 8% ^{241}Pu and 5% ^{242}Pu for RG-Pu [11]. The mass in kg/year per unit PWR for ^{237}Np, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{243}Am, ^{244}Cm and ^{245}Cm of minor actinide isotopes used in the calculations were performed as 15.1, 16.1, 205, 120, 72.7, 41.6, 6, 21.8,

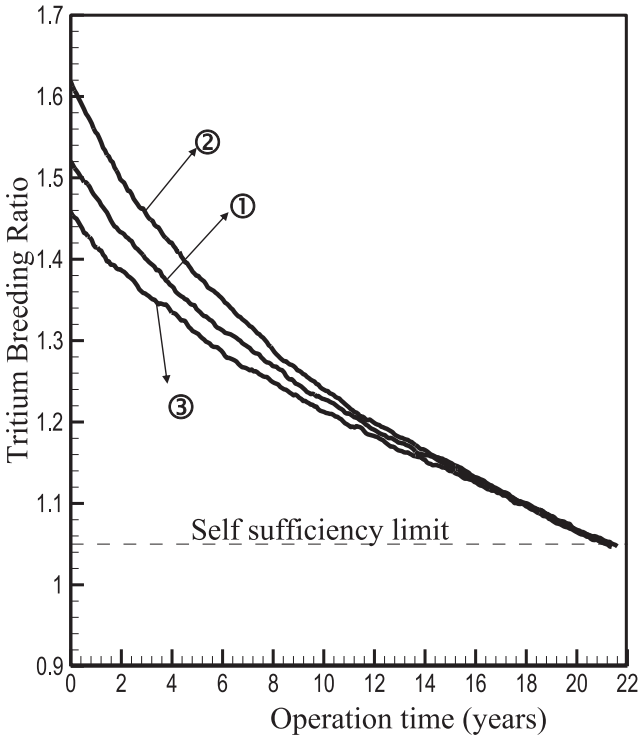


Figure 2. Change of tritium breeding ratio (TBR) in modified LIFE engine.

15.6 and 1.74, respectively [18]. In the present modified LIFE design, micro-size TRISO particles have been dispersed homogenously in the lithium coolant with 10 vol% fractions. In all investigations, the homogenous mix of the components in fuel zone was modelled and assumed, as in [9, 10, 16]. Calculations were carried out for the following cases:

Mod①: 10 vol% Reactor grade plutonium (RG-Pu) + 90 vol% natural lithium,

Mod②: 10 vol% Weapon grade plutonium (WG-Pu) + 90 vol% natural lithium,

Mod③: 10 vol% Minor actinides (MAs) + 90 vol% natural lithium.

3. Numerical calculations

The time dependent neutronic analysis has been performed with MCNP 4B [19] with ENDF/B.V library for continuous energy cross section. Calculations were conducted in spherical geometry to use the neutron spectrum with high precision with the compressed fusion neutron source up to the farthest external boundary of the geometry. The calculations were performed for a fusion driver power of 500 MW (thermal) corresponding to $1.774 \times 10^{+20}$ fusion neutrons per second with energy of 14 MeV for the sake of a consistent comparison with previous work [9, 10, 16]. Isotopes materials used in the present modified LIFE design are at 293.6 K temperature. The high precision estimation of neutronic analysis was carried out by 100 000 initial neutrons per run (cycles), i.e. 'nps', with MCNP. The auxiliary code ERDEMLI [20], which has already written for ANISN [21] results, was modified by author to evalute the output results in MCNP. The time dependent numerical output of the MCNP have further been processed with the help of the modified ERDEMLI auxiliary code for this work. Neutronic calculations were performed in time interval of 27 d and 100% plant factor. The TBR was computed at each time interval with MCNP F4 tallies. This tally was 205 (total tritium production) reactions in the coolant zone. Also, a source driven subcritical fissionable blanket should be kept by $k_{\text{eff}} < 0.8$–0.9 for the sake of reactor safety and energy control. Therefore, the initial criticality was performed with a nominal source size of 1000 particles and run a total of 115 cycles. The initial criticality values for Mod①, Mod② and Mod③ were computed as $0.3522 \pm 0.001\,87$, $0.423\,45 \pm 0.001\,78$ and $0.3295 \pm 0.001\,62$, respectively. Then, the time dependent neutronic analyses were performed of modified LIFE engine.

3.1. Tritium production (TBR)

TBR > 1.05 will be needed for a self-sufficiency fusion fuel supply [9, 10, 22–24]. ^{6}Li and ^{7}Li breeding reactions can be contributed to generate of TBR. The ^{6}Li and ^{7}Li isotope reactions were given as below:

$$^{6}\text{Li} + \text{n} \rightarrow \alpha + T + Q\ [4.784\ \text{MeV}], \tag{1}$$

$$^{7}\text{Li} + \text{n} \rightarrow \alpha + T + \text{n} + Q\ [-2.467\ \text{MeV}], \tag{2}$$

where the neutron, the alpha and the reaction energy were n, α and Q, respectively.

The total tritium production has been given as follows:

$$\text{TBR} = T_6 + T_7, \tag{3}$$

where, $T_6 = \iint \Phi \cdot \Sigma_{(\text{n},\alpha)T} \cdot \text{d}E \cdot \text{d}V$, and $T_7 = \iint \Phi \cdot \Sigma_{(\text{n},\text{n}'\alpha)T} \cdot \text{d}E \cdot \text{d}V$.

The variation of the time dependent TBR for Mod①, Mod② and Mod③ cases were presented in figure 2. The startup TBR values were calculated as 1.52, 1.62 and 1.46 for the cited cases, respectively. The highest TBR values with Mod② were obtained, followed by Mod① and Mod③. The plutonium isotopes have greatly contributed to the neutron production with (n, $2n$) reactions and fission process. It was considered that the higher fissionable ^{239}Pu isotope ratio caused to higher TBR values at startup. As shown in figure 2, TBR was gradually decreased during operation time for the cited cases. It can be explained that neutron population was decreased with depletion of fissionable isotopes in the cited cases. Therefore, TBR decreased along with the depletion of fissionable isotopes for the cited cases. The self-sustaining tritium production of modified LIFE engine for the cited cases provided as ~21 years for TBR = 1.05.

3.2. Energy multiplication factor

The energy production of modified LIFE engine can be defined as energy multiplication factor (M). The nuclear fission reaction in the blanket can be contributed of the amplifications of energy of the fusion source neutrons. The average energy released of the blanket with fission reaction is 185 MeV. And also, the ^{6}Li(n, α)T reaction average of energy release is 4.784 MeV, while the ^{7}Li(n, nα)T average energy

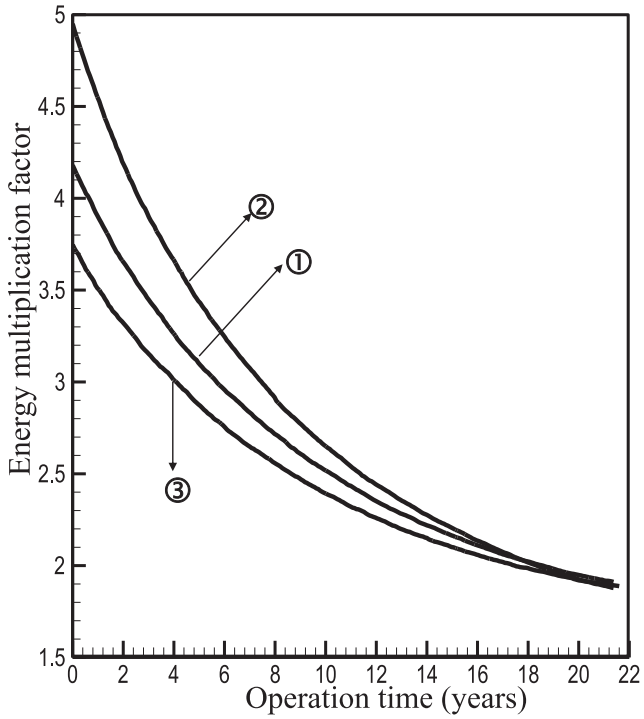


Figure 3. Change of energy multiplication factor (M) in modified LIFE engine.

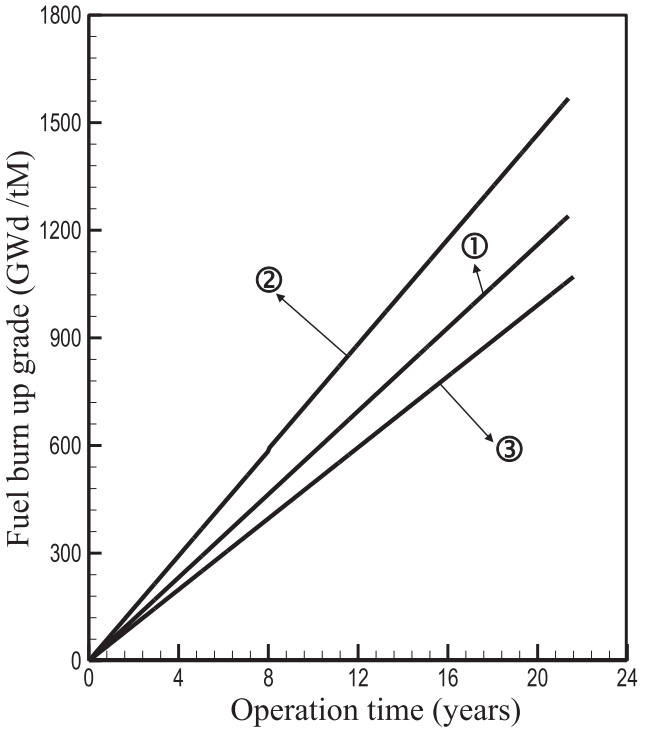


Figure 4. Change of fuel burnup grade in modified LIFE engine.

absorber is 2.467 MeV. M can be calculated as below [9, 10, 22–24]:

$$M = 1 + \frac{\text{Blanket energy release (MeV)}}{14.1\ \text{MeV}}. \quad (4)$$

Energy release of fusion blanket (MeV)

$$= 185\ \text{MeV} \cdot \langle \Phi \cdot \Sigma_f \rangle + 4.784\ \text{MeV} \cdot T_6 - 2.467\ \text{MeV} \cdot T_7$$

$$\langle \Phi \cdot \Sigma_f \rangle = \iint \Phi \cdot \Sigma_f \mathrm{d}E\mathrm{d}V: \text{ Total integral fission rate.} \quad (5)$$

The variations energy multiplication factors for Mod①, Mod② and Mod③ at TRISO fuel particles were illustrated in figure 3. At startup M calculated as 4.18, 4.95 and 3.75 for the cited cases, respectively. TRISO fuels given of Mod①, Mod② and Mod③ comprised the higher fissionable fuel content. The highest M values with Mod② were computed, followed by Mod① and Mod③. M can be increased with the increasing fissionable fuel content in the coolant. The highest M values obtained with Mod② related with fissionable ^{239}Pu isotope ratio. Fission reaction will be increased with high quality fissile fuel of TRISO fuel particles. However, M decreased gradually with increasing operation time with the depletion of fission fuel for the cited cases, as shown in figure 3.

3.3. Fissile fuel BU

The time dependent BU behaviours for Mod①, Mod② and Mod③ in a source driven subcritical LIFE engine were computed under 14 MeV neutron source. The fuel BU has been given as follows [10]:

$$\mathrm{BU} = \mathrm{PF} \cdot \Delta t \cdot E_\mathrm{f} \cdot F \cdot C_\mathrm{f} \iint \sum\nolimits_f (E) \cdot \Phi(E) \cdot \mathrm{d}E \cdot \mathrm{d}V / m_\mathrm{fu}, \quad (6)$$

where PF is the plant factor, 1.0; E_f is energy per fission, 180 MeV; C_f is conversion factor, 1.6021×10^{-19} MW s/ 1 MeV; F is fusion neutron source, $1.774 \times 10^{+20}$ (neutrons per second with energy of 14 MeV); and m_fu is the fissionable fuel charge in the fissile zone.

The variations of BU values for Mod①, Mod② and Mod③ were shown in figure 4. The values of BU compared among themselves for TRISO particles fuelled with Mod①, Mod② and Mod③. As shown in figure 4, the very high BU values were calculated as ~1230, ~1550 and ~1060 GWd tM^{-1}, respectively, for the cited cases. The highest BU values with Mod② were obtained, followed by Mod①. The operation time for the cited cases was obtained as ~21 years in the natural lithium coolant. Evaluation of at the present level of technology, the very high BU values were obtained with TRISO particles, and illustrated in figure 4. The obtained high BU values will greatly contributed to the reduction of residual nuclear waste.

3.4. Variation of the fissile fuel density

A series fissionable isotope will be continuously produced in the TRISO particles with blanket operation time. Mod①, Mod② and Mod③ in TRISO particles will be transformed with radioactive decay and neutron interaction. The variation of fissile fuel density for Mod① was given in figures 5 and 6. As shown in figure 5(a), ^{239}Pu decreases rapidly due to fission and neutron capture from ~0.115 to ~0.01 gr cm^{-3} during blanket operation time. ^{240}Pu was slowly increased during the first 4 year of decay and after that; ^{240}Pu was slowly decreased during blanket operation time.

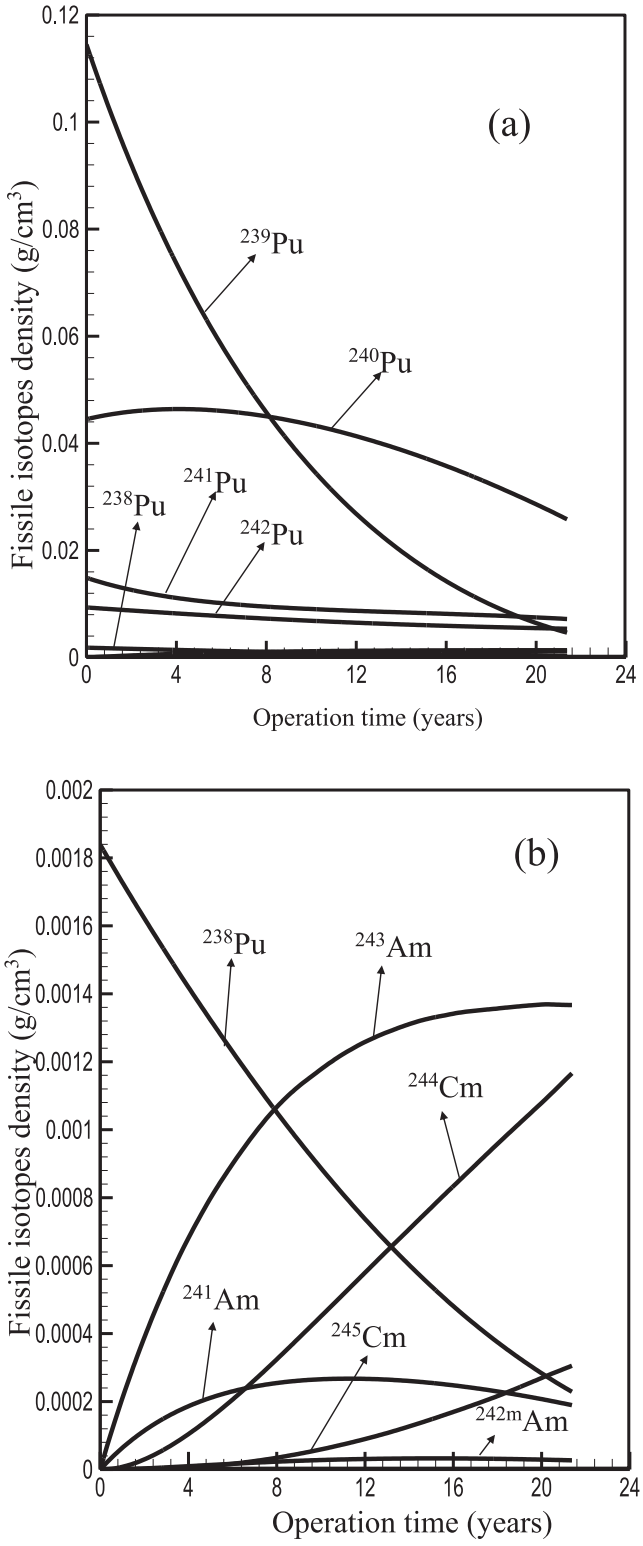


Figure 5. (a) Change of fissile isotopes density for 10 vol% RG-Pu in fuel zone. (b) Change of fissile isotopes density for 10 vol% RG-Pu of fuel zone.

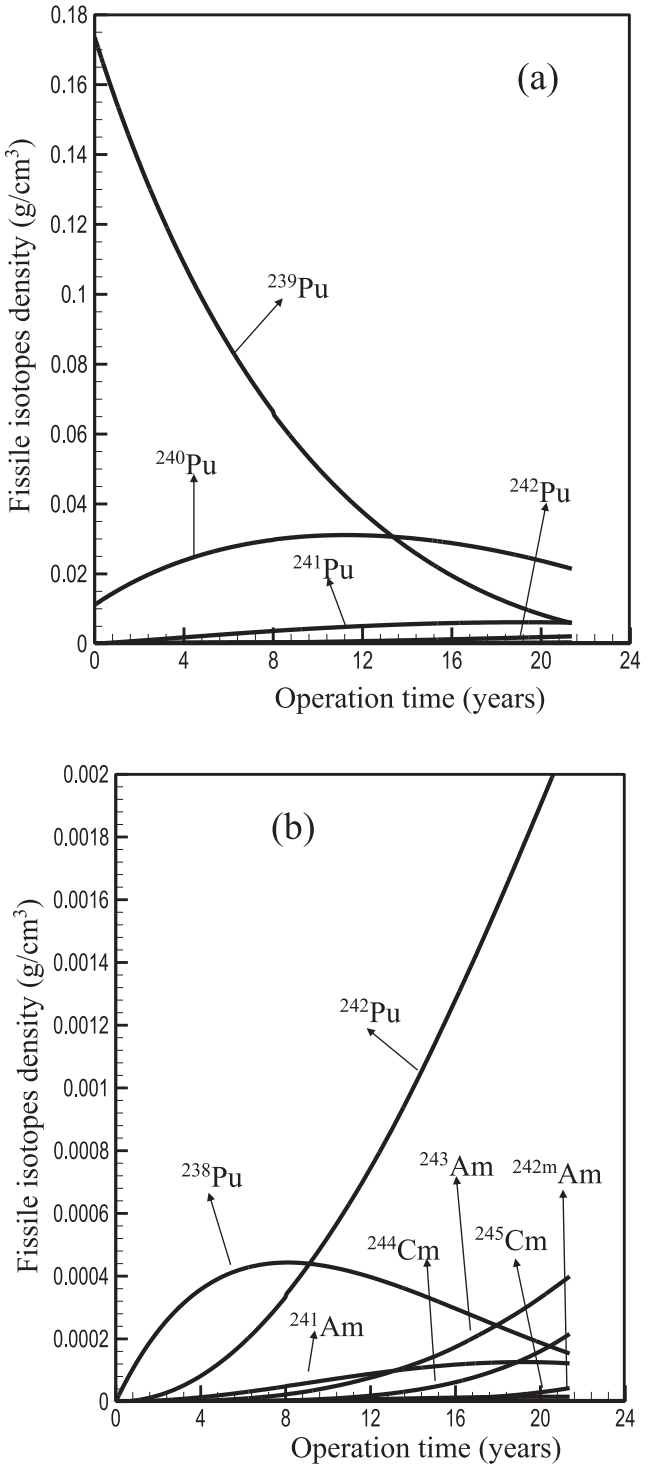


Figure 6. (a) Change of fissile isotopes density for 10 vol% WG-Pu of fuel zone. (b) Change of fissile isotopes density for 10 vol% WG-Pu of fuel zone.

The variations of initial ^{238}Pu, ^{241}Pu and ^{242}Pu isotopes were decreased of minor level during burning stage as shown in figure 5(a). ^{241}Am, ^{243}Am, ^{244}Cm and ^{245}Cm were produced by radioactive decay and neutron interaction whereas; ^{242m}Am remained constant as illustrated in figure 5(b). The variation of the main fuel isotopes for Mod② during burning stage was given in figures 6(a) and (b). ^{239}Pu decreased rapidly due to fission and neutron capture from ∼0.174 to ∼0.01 gr cm^{-3} during blanket operation time as shown in figure 6(b). ^{240}Pu was increased during the first 12 year whereas; the isotope was decreased during burning stage.

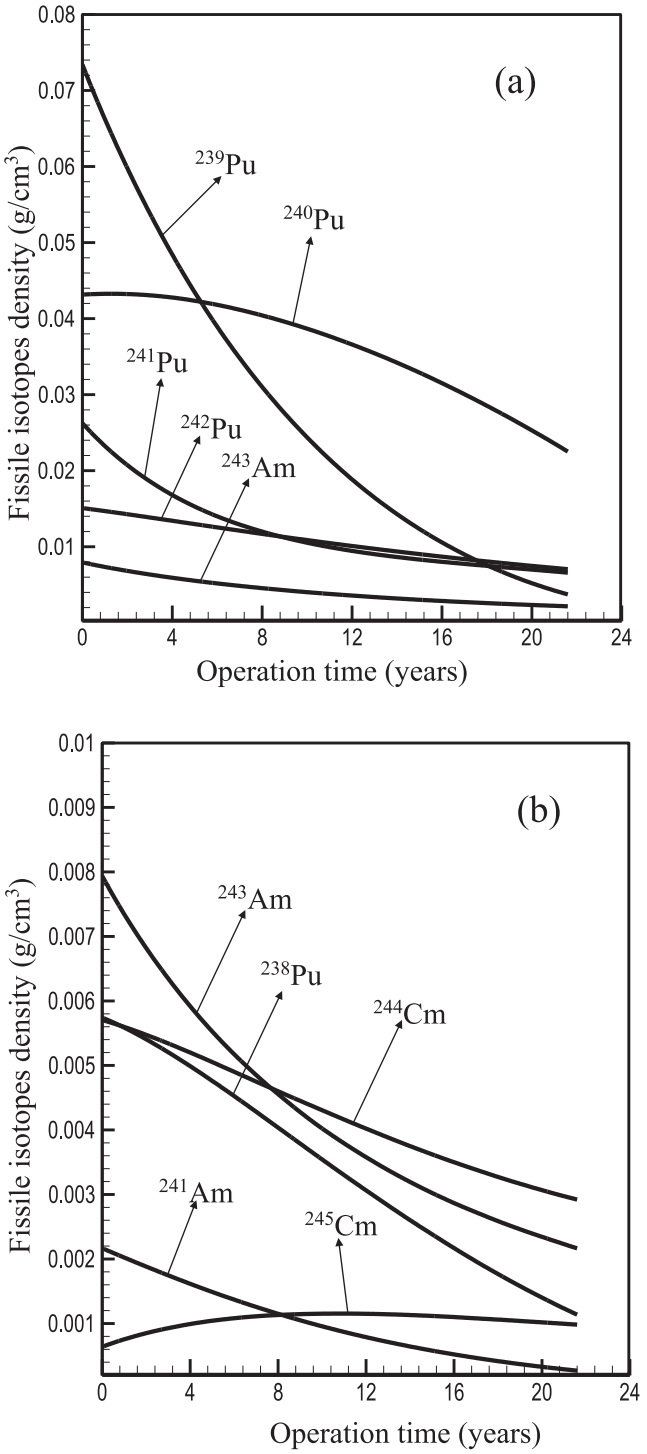


Figure 7. (a) Change of fissile isotopes density for 10 vol% MAs of fuel zone. (b) Change of fissile isotopes density for 10 vol% MAs of fuel zone.

On the other hand; ^{238}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{242m}Am, ^{243}Am, ^{244}Cm and ^{245}Cm were formed with radioactive transformation of the initial fissile isotope as plotted in figures 6(a) and (b). These isotopes generated of blanket were related to the increase of the BU values.

These isotopes will be contributed of the additional amount of energy production. Although the ^{239}Pu decreased rapidly, there was an increase produced in the new isotopes during blanket operation time as shown in figures 6(a) and (b). However, initial ^{238}Pu was increased during the first 8 year whereas; the isotope was decreased after 8 years during burning stage.

The variation of the fissile fuel isotopes for Mod③ during blanket operation time was shown in figures 7(a) and (b). High quality fissile fuel ^{239}Pu decreased rapidly due to fission and neutron capture from ∼0.075 to ∼0.01 gr cm^{-3} during burning stage as shown in figure 7(a). In figures 7(a) and (b), the initial ^{238}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{243}Am and ^{244}Cm decreased with blanket operation time. These isotopes were main fuel isotopes in the Mod③. In figure 7(b) was observed an increase in the accumulation rate of ^{245}Cm.

4. Conclusion

The study was performed to incinerate of RG-Pu, WG-Pu and MAs in the kernel of the TRISO particles of the natural lithium cooled LIFE engine using the MCNP code. Initial TBR for Mod①, Mod② and Mod③ were calculated as 1.52, 1.62 and 1.46. The TBR for the cited cases was decreased during operation time due to depletion of fissionable isotopes. The reactor operation period was computed as ∼21 years when the reference TBR $>$ 1.05 for a self-sustained reactor. The M was computed as 4.18, 4.95 and 3.75 for Mod①, Mod② and Mod③, respectively for the cited cases. The very high BU values with TRISO particles were achieved as ∼1230, ∼1550 and ∼1060 GWd tM^{-1}, respectively, for the cited cases. The calculation results shown that the most hazardous nuclear waste will be incinerated with TRISO fuel particles. Hence, the modified natural lithium cooled LIFE engine presented in this paper was exhibited the very higher BU in the incineration of RG-Pu, WG-Pu and MAs in the kernel of the TRISO particles when compared to previous studies [9, 10].

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