

Study of Uranium and Thorium Fuels in Breed-and-Burn Mode

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Abstract

The formation of fissile nuclei through breeding conversion is a hot topic in academic research, as it provides a continuous source of nuclear fuel for nuclear reactors. Fast neutron reactors, which have been extensively studied, use natural uranium or low-enriched uranium as the nuclear fuel, achieving burning after uranium-plutonium conversion. Thorium, as another potential fissile fuel, can theoretically be converted into nuclear reactor fuel through the thorium-uranium cycle. In this study, the physical evolution process of nuclear fuel in a specific core parameter is simulated using the Monte Carlo program, and the performance differences of the thorium-uranium cycle and uranium-plutonium cycle in achieving in situ breeding-burning (Breed-and-Burn, B&B) mode are analyzed using neutron balance analysis method. The optimal ratio conditions for achieving self-sustained B&B burning in a thorium-uranium fuel mixed core are investigated. The study shows that for traditional solid-state nuclear reactor cores with a lower fuel proportion, thorium-breeding fuel has poor neutron economy compared to uranium-based breeding fuel, making it more difficult to achieve the B&B mode.

Keywords

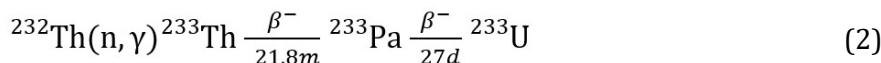
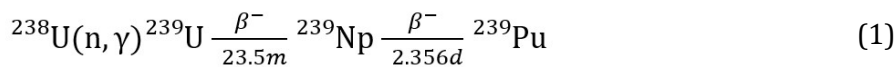
Uranium-Plutonium Cycle; Thorium-Uranium Cycle; Breed and Burn.

1. Introduction

The in-situ breed-and-burn (B&B) mode refers to how fissile nuclides such as thorium (^{232}Th) and uranium (^{238}U) in a nuclear reactor are proliferated and burnt without undergoing chemical processing [1, 2]. The nuclear reactor that achieves this mode is known as a B&B reactor, which belongs to a special type of fast neutron spectrum breeder reactor (Fast Breeder Reactor, FBR). Unlike the traditional commercial pressurized water reactor (PWR) loading method, the B&B reactor initially only requires a small amount of enriched uranium or super-uranium element to start the ignition, then the core can establish a power distribution and other equilibrium states that do not change with time after a certain burnup cycle. The geometric structure and composition of the newly loaded fuel in the nuclear reactor are the same, and each newly loaded nuclear fuel will undergo similar fuel proliferation and excess neutron ignition functions after a steady-state operation is achieved. The reactor can maintain stable operation by simply adding fissile nuclide new fuel to the core [3, 4]. The adoption of the in-situ breed-and-burn mode in a reactor can enhance the utilization efficiency of fissile fuel nuclides, reduce the total amount of spent fuel processing, and minimize the mining and enrichment of new fuel resources, while also enhancing nuclear non-proliferation capabilities [5, 6].

When a nuclear reactor operates in a B&B mode, the nuclear fuel in the active zone, which serves the ignition function, produces a large number of excess neutrons through chain fission. These neutrons may leak into the adjacent breeding zone, where fissile nuclides can absorb these neutrons and convert them into easily fissionable nuclear fuel. The conversion of

fissionable nuclides (^{238}U , ^{232}Th) to fissile nuclides (^{239}Pu , ^{233}U) is shown in equations 1 and 2 [7].



Equation (1) demonstrates the absorption of a fission neutron by the fissionable isotope ^{238}U , resulting in the conversion to ^{239}U and the emission of a γ -ray. Then ^{239}U undergoes two β -decay processes with half-lives of 23.5 minutes and 2.356 days, respectively, to transform into the fissile isotope ^{239}Pu . Equation (2) shows that the capture of a fission neutron by the fissionable isotope ^{232}Th , and the generation of the isotope ^{233}Th is accompanied by the emission of a γ -ray. ^{233}Th further undergoes two β -decay processes with half-lives of 21.8 minutes and 27 days, respectively, to convert into the fissile isotope ^{233}U . The fissile isotopes ^{239}Pu or ^{233}U generated from the conversion of fissile isotopes can, on average, produce more than two neutrons per fission in a wide neutron energy range. By employing a well-designed approach, one neutron can be utilized for the conversion of fissile isotopes, while the other neutron can sustain the chain fission reaction within the reactor. Theoretically, it is possible to experimentally breed and burn nuclear fuel in situ within the reactor, known as the B&B operational mode.

This study investigates the B&B performance of nuclear fuels composed of ^{238}U and ^{232}Th in a nuclear reactor. Section 2 provides a brief overview of the physical properties and characteristics of the uranium-plutonium cycle and the thorium-uranium cycle. Section 3 introduces the neutron balance method, core evolution parameters, and B&B characteristics employed in the study.

2. Uranium and Thorium Fuel Cycle

2.1. Uranium-Plutonium Fuel Cycle Chain

Natural uranium or low-enriched uranium fuel contains the fissionable isotope ^{238}U . When ^{238}U absorbs neutrons with energies above 1.1 MeV in a reactor, fast fission reactions are triggered. On the other hand, when ^{238}U absorbs neutrons with energies below 1.1 MeV, mainly capture reactions occur, followed by β -decay and isotope conversion. This process is of great importance as a potential nuclear fuel. The primary changes in nuclear isotopes in the uranium fuel during the uranium-plutonium cycle are illustrated in Figure 1.

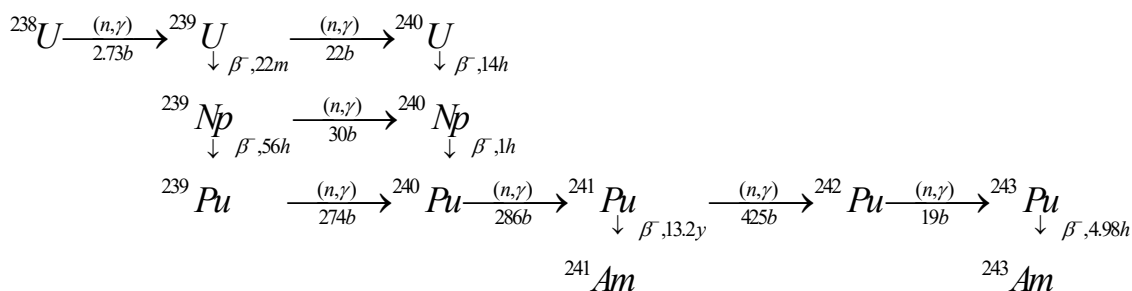


Figure 1. Simplified uranium-plutonium fuel cycle chain

After capturing a neutron, ^{238}U is converted into ^{239}U and emits a β -particle. ^{239}U is unstable and undergoes β -decay with a half-life of 22 minutes to become ^{239}Np , which further undergoes β -

decay with a half-life of 56 hours to produce ^{239}Pu . In a fast reactor, for every consumed ^{239}Pu , approximately 1.4 new ^{239}Pu isotopes can be generated, indicating that, in principle, uranium-based breeder fuels can achieve the B&B mode [8].

2.2. Thorium-Uranium Fuel Cycle Chain

Similar to the uranium-plutonium cycle chain of ^{238}U , ^{232}Th can be converted through irradiation to generate ^{233}U , which is another potential nuclear fuel. The primary nuclear chain of the thorium-uranium cycle with ^{232}Th is illustrated in Figure 2 [9].

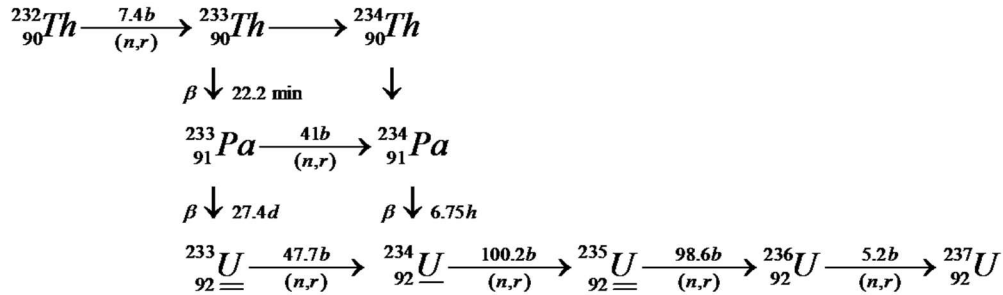


Figure 2. Simplified thorium-uranium fuel cycle chain

After capturing a neutron, ^{232}Th generates ^{233}Th and emits a γ -ray. ^{233}Th is unstable and undergoes β -decay with a half-life of 22.2 minutes to produce ^{233}Pa , which further undergoes β -decay with a half-life of 27.4 days to generate the fissile isotope ^{233}U .

2.3. Equilibrium Self-Stabilizing Characteristic

Figure 1 illustrates the burnup chain of the uranium-plutonium fuel cycle with ^{238}U . The variation in the concentration of ^{239}Pu with time is equal to the rate of disappearance of ^{238}U minus the rates of disappearance of ^{239}Pu , ^{239}U , and ^{239}Np . The burnup equation can be expressed as:

$$\frac{dN_{239\text{Pu}}(t)}{dt} = \varphi \left(\sigma_{\gamma}^{238\text{U}} N_{238\text{U}} - \sigma_{\gamma}^{239\text{U}} N_{239\text{U}} - \sigma_{\gamma}^{239\text{Np}} N_{239\text{Np}} - \sigma_a^{239\text{Pu}} N_{239\text{Pu}} \right) \tag{3}$$

Where $N_{239\text{Pu}}(t)$, $N_{238\text{U}}$, $N_{239\text{U}}$, $N_{239\text{Np}}$ represent the nuclear densities (concentrations) of ^{239}Pu , ^{238}U , ^{239}U , and ^{239}Np isotopes, respectively. φ is the neutron flux. t is the core evolution time, $\sigma_{\gamma}^{238\text{U}}$, $\sigma_{\gamma}^{239\text{U}}$, $\sigma_{\gamma}^{239\text{Np}}$ represent the neutron capture cross-sections of ^{238}U , ^{239}U , and ^{239}Np isotopes, respectively. $\sigma_a^{239\text{Pu}}$ represents the neutron absorption cross-section of the ^{239}Pu isotope. When the concentration of ^{239}Pu in the fuel chain does not change with time, i.e., the left side of the equation is zero, the core reaches an equilibrium state.

When the equilibrium concentration of ^{239}Pu is higher than its critical concentration, the amount of ^{239}Pu produced by breeding is greater than the amount consumed by burning, resulting in the stable and sustained propagation of breeding waves following the burn wave. Conversely, if the generated ^{239}Pu from the conversion is less than the consumed amount, it exhibits typical ignition zone behavior. By optimizing the core design, the breeding region of the nuclear fuel can be maintained in a neutron balance state, where the equilibrium concentration of ^{239}Pu is equal to the critical concentration. The effective breeding factor of the B&B reactor core can be maintained near $k_{\text{eff}} = 1$.

The equilibrium state of the B&B core exhibits self-stabilizing operational characteristics, where the conversion-generated ^{239}Pu and the consumed ^{239}Pu automatically maintain

dynamic balance. When a small positive reactivity perturbation is introduced to the B&B core, it consumes much more ^{239}Pu , leading to a decrease in its concentration and consumption rate. After a period of decay, ^{239}Np accumulates and produces a large amount of ^{239}Pu . As a result, the concentration of ^{239}Pu increases, establishing a new equilibrium and maintaining the self-stable operation of the core.

2.4. Characteristics of Uranium-Plutonium and Thorium-Uranium Cycles

Research on the uranium-plutonium and thorium-uranium cycles began with the development of the first atomic bomb [11]. In the following sections, we will compare the conversion efficiency, breeding performance, and the radiotoxicity of nuclear waste between these two cycles.

(1) Conversion Efficiency: In the fast neutron region, the neutron capture cross-sections of ^{233}U generated from the conversion of ^{232}Th , and the neutron capture cross-sections of ^{239}Pu generated from the conversion of ^{238}U , are similar. However, the neutron capture cross-section of ^{232}Th is slightly higher than that of ^{238}U , indicating comparable conversion capabilities. However, the presence of ^{233}Pa , a precursor to ^{233}U , in the thorium-uranium cycle significantly affects the operation of the core. With a half-life of 27 days, ^{233}Pa undergoes neutron capture to become ^{234}Pa , which further decays into ^{234}U . ^{234}U can then absorb one neutron to produce fissile nuclide ^{235}U . This process consumes two neutrons, reducing the neutron utilization efficiency and competing with the ^{233}U breeding process, resulting in a decrease in ^{233}U production. The neutron capture cross-section of ^{233}Pa is smaller in the thermal neutron region and the fast neutron region. Therefore, in a thorium-based fuel core operating in the fast neutron spectrum, the influence of ^{233}Pa is relatively small. This study will compare and analyze the differences between the uranium-plutonium and thorium-uranium cycles in fast-spectrum cores.

(2) Breeding Performance: In the fast neutron region, the fission cross-section of ^{233}U is slightly larger than that of ^{239}Pu , while the fission cross-section of ^{232}Th is only about 25% of that of ^{238}U . The neutron capture-to-fission ratio is similar between thorium and uranium fuels. However, in fast spectrum cores, the η value of ^{233}U is significantly smaller than that of ^{239}Pu , indicating that thorium-based fuels are more favorable for achieving self-sustained burning. The η value of ^{233}U remains above 2.3 throughout the fast spectrum region, with one neutron used for fissile fuel breeding and the other neutron maintaining the chain reaction in the core. This leaves only about 0.3 neutrons, meaning that thorium-based fuels require a reduction in neutron leakage and parasitic absorption by structural materials and coolant fission products to achieve the B&B self-sustained burning mode. This implies a large core volume, minimal parasitic absorption by structural materials and coolant, and minimal accumulation of fission products' neutron absorption.

(3) Radiotoxicity of Nuclear Waste: The production of transuranic elements requires the absorption of six additional neutrons for low-Z isotopes (^{232}Th and ^{233}U) compared to high-Z isotopes (^{238}U and ^{239}Pu). Therefore, the thorium-uranium cycle generates fewer long-lived actinide nuclides, resulting in lower toxicity of the spent fuel components. Generally, the radiotoxicity of thorium-based fuel cores is one to two orders of magnitude lower than that of uranium-based fuel cores, and the disposal time for nuclear waste is also reduced [12].

3. Comparison of Breeding Performance in Uranium and Thorium Fuel Cores

3.1. Neutron Balance Method

The neutron balance method calculates the evolution of the excess neutron count with fuel burnup, providing information on the minimum required burnup and maximum required

burnup of the fuel [13]. The excess neutron count is defined as the total number of neutrons produced per unit volume per unit time minus the total number of neutrons lost, and it varies with burnup [14].

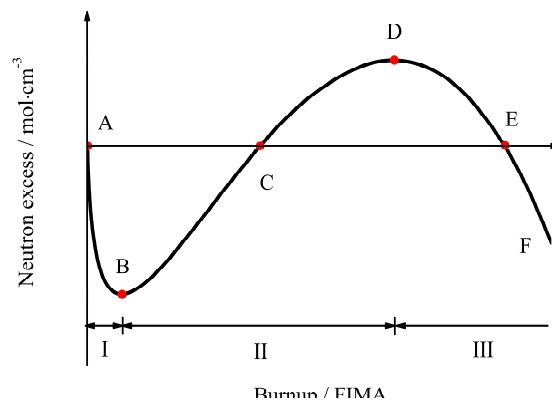


Figure 3. Schematic diagram of neutron balance of breeder fuel

Figure 3 illustrates the neutron balance of breeding fuel as a function of burnup [15]. In Region I (segments A-B), the breeding fuel acts as a net neutron absorber. During this phase, a significant amount of fission neutrons is consumed for the conversion of fertile isotopes, but the concentration of fissile isotopes produced from the conversion is still low, resulting in a smaller neutron contribution. The total number of neutrons produced in the fuel is less than the total number of neutrons absorbed, and the excess neutron count reaches a minimum at point B, representing the total number of neutrons required for the conversion of breeding fuel (fissile isotopes).

In Region II (segments B-D), the breeding fuel acts as a net neutron producer. During this phase, the breeding process continues to consume fission neutrons, while the neutrons produced from the fission of the generated fissile isotopes can compensate for (or even exceed) the neutron consumption by the breeding process. The total number of neutrons produced in the fuel is greater than the total number of neutrons absorbed. Point C represents the minimum required burnup of the breeding fuel, indicating that the excess neutron count is sufficient to repay the total number of neutrons borrowed by the breeding process. This is when the breeding fuel first reaches a neutron balance state.

In Region III (segments D-F), the breeding fuel acts as a net neutron absorber. During this phase, the accumulated fissile isotopes from the breeding process are gradually consumed, and the neutron consumption by fission products increases, resulting in the total number of neutrons produced is less than the total number of neutrons absorbed. Point E represents the maximum theoretical burnup of the breeding fuel, where the total number of neutrons produced is equal to the total number of neutrons absorbed. The breeding fuel once again reaches a neutron balance state. If the breeding fuel is discharged at the maximum theoretical burnup, the core can operate steadily with $k_{eff} = 1$ [16].

3.2. Core Parameters

In this study, a thorium-based core with metallic thorium as fuel was used, with a nuclear fuel density of 11.65 g/cm^3 . On the other hand, a uranium-based core with U-Zr alloy as fuel was used, with a nuclear fuel density of 15.85 g/cm^3 . Both thorium-based and uranium-based cores employed HT-9 steel as cladding material. The thorium-based and uranium-based cores are cooled by sodium and helium, respectively. The composition ratios for both cores were the same, as shown in Table 1.

Table 1. Volume fractions of core components

Th, U core	
Fuel	37.5%
Gap	12.5%
Coolant	21%
Cladding	29%

The Monte Carlo program MCNPX version 2.6 was utilized to simulate the burnup evolution process of the core [17]. The ENDF/B-VII database was used for calculations [18].

3.3. Comparison of Breeding Performance between Thorium and Uranium Fuel Cores

After the MCNPX evolution, the average number of neutrons released per fission (ν) for the two breeding nuclear fuels, depleted uranium (U) and metallic thorium (Th), was extracted as a function of their burnup, as shown in Figure 4. For the Th fuel, the initial value of ν (corresponding to 0% FIMA, Fission per Initial heavy Metal Atom) was approximately 2.378. As the burnup depth (Burnup) gradually increased, the accumulation of ^{233}U generated from breeding conversion led to an increase in ν , stabilizing at around 2.525.

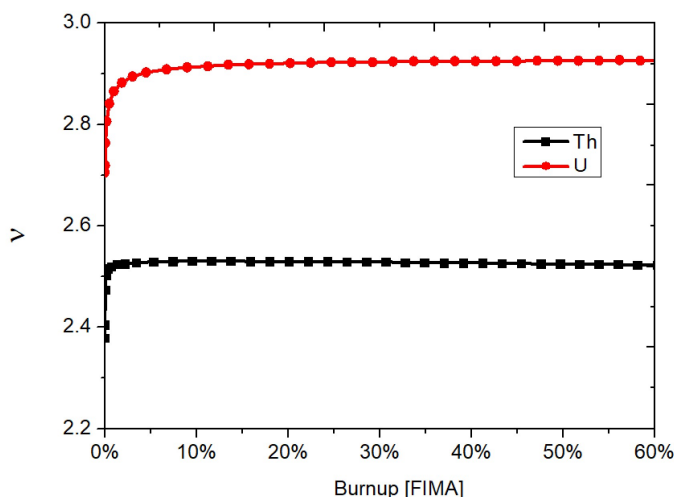


Figure 4. Evolution of ν with burnup

For the depleted uranium fuel containing only a small amount of ^{235}U and mostly ^{238}U , at the beginning of life (0% FIMA), the value of ν is approximately 2.705, which is about 14% higher compared to the Th fuel. As the burnup increases, the accumulation of ^{239}Pu from breeding and conversion gradually increases, leading to an average ν of around 2.925, which is approximately 16% higher compared to the Th fuel.

The total neutron absorption ΔA can be obtained by using the effective multiplication factor k_{eff} of the fuel as a bridge, calculated from the total neutron production ΔP , as $\Delta A = \frac{\Delta P}{k_{eff}}$. When

simulating the evolution using the infinite medium model (0D), the effective multiplication factor k_{eff} of the core is equivalent to the infinite multiplication factor k_{∞} . The evolution of the infinite multiplication factor k_{∞} for both Th and U fuels with their burnup is shown in Figure 5. At the beginning of life (0% FIMA), the value of k_{∞} for the helium-cooled depleted uranium fuel (U-Zr alloy) is approximately 0.24, indicating a subcritical state where consuming 4 neutrons only produces about 1 neutron, as shown in Zone I of Figure 3. For the sodium-cooled metallic thorium fuel at the beginning of life (0% FIMA), the value of k_{∞} is approximately 0.05, which is

lower compared to the metallic uranium fuel. This is because ^{232}Th has a smaller fraction of fast neutron fission compared to ^{238}U .

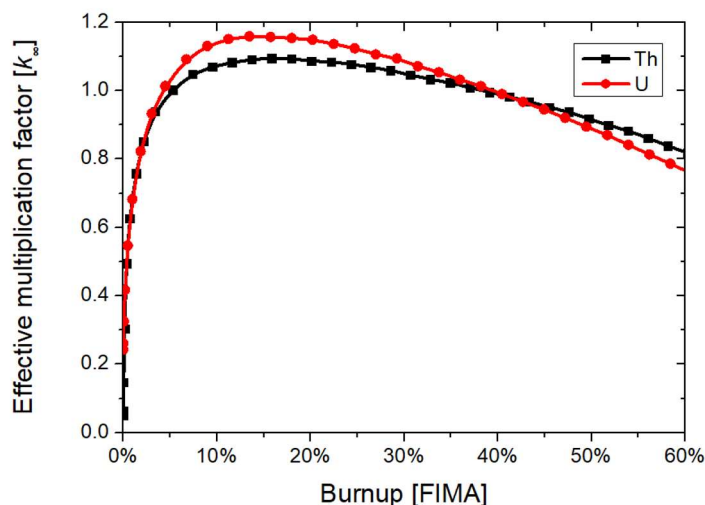


Figure 5. Evolution of k_{∞} for thorium and uranium breeding fuels

As the fissile isotopes (^{239}Pu , ^{233}U) gradually accumulate in the breeding fuel, k_{∞} increases with increasing burnup depth. For the uranium and thorium fuels, k_{∞} reaches 1 at burnup depths of approximately 3.5% FIMA and 5.2% FIMA, respectively. At burnup depths of approximately 13.2% FIMA and 15.5% FIMA, k_{∞} reaches its maximum value. Although the metallic uranium fuel has a higher value of ν compared to the metallic thorium fuel, the presence of Zr isotopes in the cladding material of the metallic uranium fuel contributes to a certain degree of moderation. These combined effects result in a maximum k_{∞} value for metallic uranium that is approximately 6% higher compared to metallic thorium, with values of 1.16 and 1.10, respectively. As the accumulation of fission products increases, the decrease in k_{∞} for metallic thorium is slower compared to metallic uranium, as the total fission product inventory for metallic thorium is only about 50% of that for metallic uranium. This leads to an intersection of k_{∞} values for both fuels at deeper burnups.

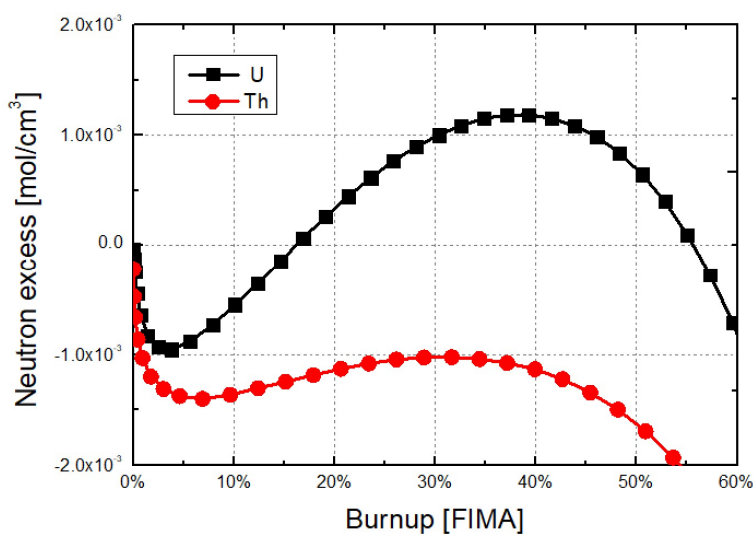


Figure 6. The neutron balance of thorium and uranium breeding fuels

The neutron balance for the thorium-based and uranium-based core models is shown in Figure 6. After the conversion of U-Zr alloy fuel into neutron-producing material (from 3.7% FIMA to 39.3% FIMA), it can accumulate a surplus of 2.12×10^{-3} mol/cm³ of neutrons, which is sufficient to compensate for the neutrons borrowed from the external source during the breeding and conversion process, which requires 0.93×10^{-3} mol/cm³ of neutrons. This nuclear breeding fuel can achieve the B&B burning mode. However, after the conversion of metallic thorium fuel into neutron-producing material (from 6.8% FIMA to 31.6% FIMA), it can only provide a surplus of 0.38×10^{-3} mol/cm³ of neutrons, which is insufficient to compensate for the borrowed neutrons (1.4×10^{-3} mol/cm³) during the breeding and conversion process. Therefore, under the given core design conditions (fuel composition of 37.5%), metallic thorium fuel cannot achieve the B&B burning mode.

The higher neutron surplus in the metallic uranium (U-Zr) core compared to the metallic thorium (Th) core indicates that metallic uranium is more conducive to achieving the B&B burning mode. This is due to the higher density of actinide elements in metallic uranium (14.22 g/cm³) compared to metallic thorium (11.15 g/cm³), resulting in a higher initial heavy metal number density (N_{HM} coefficient) for metallic uranium. Additionally, metallic uranium fuel has higher k_{∞} values for both lifetime and burnup compared to metallic thorium, and metallic uranium can convert to neutron-producing material earlier than metallic thorium (corresponding to the first attainment of k_{∞} in Figure 5).

4. Conclusion

In conclusion, the study demonstrates that under the given fast spectrum conditions, metallic uranium exhibits better breeding performance compared to metallic thorium. The thorium-uranium cycle is more challenging to achieve the B&B mode compared to the uranium-plutonium cycle. To achieve the B&B mode with thorium-based fuel, a more compact design of fuel rods in the core is required to increase the fuel volume fraction and reduce neutron leakage.

Acknowledgments

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