

Neutronic evaluation of VVER fuel assembly with chemical spectral shift regulation

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Abstract

The performance of the spectral shift control (SSC) method is evaluated and compared to the conventional poison method in the VVER-1000 fuel assembly design. The SSC method can be implemented by gradually adjusting the ratio of heavy water to light water moderator (D_2O/H_2O) during the fuel cycle. In this study, the efficiency of using the SSC design with or without a thermal absorber (gadolinium) is investigated. We apply the SSC with both 12 burnable absorber rods containing 4.0 wt.% Gd_2O_3 (Case 1) and without Gd_2O_3 (Case 2). The neutronic calculations indicate that the discharge burnup is enhanced by 60% and the conversion ratio (CR) is increased by 64.4% at the beginning of the cycle (BOC) compared to the benchmark data. The breeding of Pu^{239} and Pu^{241} is extended to 33.7% and 29.5%, respectively, for the SSC design case (2), and better utilization of U-235 and U-238 has been achieved compared to BM.

Keywords

Chemical spectral shift regulation, Neutronics, fuel cycle, VVER, SERPENT-2

Introduction

During the last decades, there have been numerous efforts to extend the operational cycle of pressurized water reactors (PWRs) from months to years (Burns et al. 2020; Choi et al. 2023; Lindsay et al. 2023). However, these methods typically necessitated enrichment levels greater than 5% and an average discharge burnup level above 62 GWd/MTU. The increase in the enrichment led to an additional excess reactivity produced during the early stages of fuel life, which can be controlled by:

1. Using soluble boron in the cooling system, although effective, can lead to a much lower moderator

temperature coefficient and boron-induced reactivity accidents, especially when used in large quantities.

2. Using control rods, while effective, can lead to an axial peak if inserted too deeply into the core.
3. Using burnable absorbers such as gadolinium, while effective, can lead to diminished neutron economy that can be used in increasing the conversion ratio (CR) of nuclear fuel (Elzayat et al. 2022; Lindsay et al. 2023).
4. Applying the spectral shift control (SSC) method, as demonstrated by the Vulcain experiment in the BR-3 nuclear plant at Mol, Belgium, and successfully operated in this nuclear plant between 1966 and 1968 (Sider and Matzie 1980), resulted in improved resource utilization and a higher CR.

In the concept of the SSC, the neutron spectrum shifts from intermediate at the beginning of the cycle (BOC) to thermal at the end of the cycle (EOC). With a resonance spectrum during the early stage of operation, neutrons can be captured in fertile materials such as U^{238} , increasing the converted fissile plutonium (Kotlyar et al. 2017). This allows for the initial U^{235} enrichment to remain higher to sustain the neutron chain reaction at the EOC, enabling more power to be generated with the same enrichment (Yokornizo et al. 1993). Consequently, better fuel utilization, extending the operation of a single fuel loading to achieve a longer fuel cycle and lower initial U^{235} inventory than conventional light water reactors (LWRs) (Matzie and Sider 1979).

The SSC methods can be categorized into two groups:

1. The chemical method, achieved by varying the (D_2O/H_2O) ratio, initially aimed to enable an ultra-long fuel cycle for the Mixed Moderator PWR (MPWR) (Tochihara et al. 1998), and later extended to studies on VVER 1000 reactors with low-enriched uranium fuel (LEU).
2. The mechanical method, achieved by varying the moderator-to-fuel volume ratio (v_m/v_f) by inserting and withdrawing spectral shift rods.

The chemical method was initially introduced by Mars and Gans in 1961, involving the utilization of a mixture of light and heavy water (Mars and Gans 1961). At the BOC, the D_2O concentration was altered from 79% to 60% mole, gradually decreasing to 2% mole at the EOC. The fluctuation in heavy water concentration regulates the reactivity of the core and enhances the power density, ultimately resulting in an extended cycle life. Several studies and experiments have been conducted to identify the most appropriate changes that could enable the SSC design to achieve its extra benefits. In spectral shift control reactors, introduced in (Alcala 1984), the neutronic and thermal-hydraulic behavior were optimized for low-enriched uranium graphite cells cooled with water to determine the optimal cell design for implementing the SSC system. The D_2O/H_2O ratio concentration utilized in the coolant system flows through tubes arranged within the graphite blocks. In LR-0 reactor model (Nagy et al. 2014), the neutron absorption profile for employing thermal poison in control rods was investigated. It was found that epithermal neutron poisons or using double-layer control rods (with epithermal neutron poison in the outer layer and thermal neutron poison in the inner layer) might be more suitable for a mixed D_2O/H_2O moderator reactor. According to a study on the utilization of the SSC method in single-batch small modular reactors (SMRs), it was found that a combination of a slightly wetter lattice and chemical method can result in a significant enhancement in natural uranium utilization, reaching up to 49%, compared to conventional LWRs (Lindley and Parks 2016).

The mechanical method varies depending on the type of reactor. In PWRs, achieving high burnup and CR can be

accomplished by using fertile rods (Okumura et al. 1988). In boiling water reactors (BWRs), the mechanical method of SSC can be achieved by changing the water column in spectral shift rods, causing a change in the void fraction (Yokornizo et al. 1993). In TRU-fueled very high-temperature reactors (VHTRs), the moderator-to-fuel volume ratio changes by varying the number of TRISO particles per unit volume (Tsvetkov et al. 2008). A study on utilizing a movable graphite structure in the Small Advanced High-Temperature Reactor (SmaHTR), where graphite serves as a moderator material, could extend the operational cycle (Kotlyar et al. 2017). In a liquid-fueled molten salt reactor (MSR), zirconium hydride rod assemblies can be utilized as a moderator in the reactor, allowing the moderator-to-fuel ratio to vary throughout the reactor's lifetime (Betzler et al. 2018). The concept of spectral shift rods (SSRs) is analyzed in a small lead-based reactor, where SSRs are divided axially into three sections including moderator, coolant, and control rod absorber from the bottom to the top. During the burnup procedure, the SSRs are gradually withdrawn from the active core to induce a change in the neutron spectrum shift (Zhao et al. 2021).

The main objective of the current paper is to assess the neutronic characteristics of the VVER-1000 LEU fuel assembly (FA) employing chemical spectral shift regulation. This study investigates the impact of varying the ratio between heavy water and light water on the VVER-1000 FA described in the OECD computational benchmark (Kalugin and Gehin 2002). The VVER-1000 FA employs the poison control reactivity method by adding 600 ppm H_3BO_3 in the moderator system and incorporates 12 burnable absorber pins with 4.0 wt.% Gd_2O_3 .

In the previous study (Elzayat et al. 2022), we applied the chemical SSC method through diluting D_2O/H_2O molecular ratio from 65% to 0% in the presence of gadolinium, and from 73% to 0% in the absence of gadolinium, with considering different decrements: 10%, 13%, 15%, and 19% in both cases. These sudden changes in the molecular ratio of D_2O/H_2O leads to a higher reactor response to the reactivity changes and diminish neutronic parameters values. Additionally, ^{135}Xe and ^{149}Sm did not reach equilibrium throughout the fuel cycle. Thus, in this study, the impact of the chemical method in both the presence and absence of gadolinium is examined by diluting the molecular ratio of D_2O/H_2O with equal decrements of 5% in all cases. This dilution process thermalizes the neutron spectrum and burns effectively fissile materials which bred at the BOC. Specifically, the molecular ratio of D_2O/H_2O is increased at the BOC, resulting in a hardened spectrum that facilitates the capture of neutrons in fertile material such as U^{238} rather than in poison control materials, thus enhancing the breeding of Pu fissile material along with suppressing the excess reactivity of the fresh fuel. Therefore, the SSC method has the potential to improve the CR value while achieving high discharge burnups, thereby prolonging the fuel cycle, compared to traditional poison control methods used in typical PWRs.

According to the Monte Carlo simulation, the following parameters were studied: the variation of infinite multiplication factor (k_{∞}), conversion ratio (CR) versus burnup, and the evolution of the isotopic composition of the fuel assembly (^{235}U , ^{236}U , ^{238}U , ^{239}Pu , ^{240}Pu , ^{241}Pu , and ^{242}Pu), the accumulation of fission product poisons (^{135}Xe and ^{149}Sm) and burnout of burnable absorbers (^{155}Gd and ^{157}Gd). The obtained results were compared and analyzed according to the corresponding benchmark mean values (BM) (Kalugin and Gehin 2002).

Materials and method

VVER-1000 FA design

In the present work, the VVER-1000 FA is presented from the VVER-1000 LEU and MOX Assembly Computational Benchmark (Kalugin and Gehin 2002). There are two variants of FA configurations in this computational benchmark: (1) Low enriched uranium (LEU) + 12 uranium gadolinium rods (UGD) and (2) Mixed oxide (MOX) + 12 uranium gadolinium rods (MOXGD). Here the first type (i.e., UGD variant) is adopted for applying the chemical spectral shift control method. The VVER-1000 FA has a hexagonal shape with 23.6 lattice pitch and the distance between the fuel rods within the lattice is 1.275 cm. The FA is divided into 331 hexagonal unit cells: 18 guide tubes, 1 central instrumentation tube and 312 fuel pins (300 fuel pins with enrichment of 3.7 wt.% U^{235} and 12 UGD with enrichment of 3.6 wt.% U^{235} and 4.0 wt.% Gd_2O_3). Fig. 1 illustrates the LEU assembly configuration obtained by Serpent code. The material composition and density are listed in Table 1. The clad and structural materials are composed of Zr-Nb alloy. The burnup calculations are demonstrated at operating poisoned state conditions where the fuel temperature is 1027 K and non-fuel materials temperature is 575 K. The equilibrium concentrations between Xe^{135} and Sm^{149} are considered in this study. The fuel has been depleted at a constant power

Table 1. The material composition and density (Kalugin and Gehin 2002)

Material type	Isotopic content, (atoms/barn.cm)	material type	Isotopic content, (atoms/barn.cm)
Fuel cell: 3.7 wt.% U^{235}	U^{235} 8.6264E-4 U^{238} 2.2169E-2 O^{16} 4.6063E-2	Fuel gadolinium cell: 3.6 wt.% U^{235} with 4 wt.% Gd_2O_3	U^{235} 7.2875E-4 U^{238} 1.9268E-2 O^{16} 4.1854E-2 Gd^{152} 2.5159E-6 Gd^{154} 2.7303E-5 Gd^{155} 1.8541E-4 Gd^{156} 2.5602E-4 Gd^{157} 1.9480E-4 Gd^{158} 3.0715E-4 Gd^{160} 2.6706E-4
The clad material: Zirconium alloy	Zr 4.259E-2 Nb 4.225E-4 Hf 6.597E-6		
Moderator, 0.6 g/kg of boron	H 4.843E-2 O^{16} 2.422E-2 B^{10} 4.794E-6 B^{11} 1.942E-5		

density of 108 MW/m³ up to a burnup of 40 MWd/kgHM. In this paper, the moderator and coolant consist of a mixture of $\text{D}_2\text{O}/\text{H}_2\text{O}$ instead of H_2O as in the benchmark.

Computational methodology

The Serpent-2 Monte Carlo code version 2.1.31 (Leppänen et al. 2015) is utilized to simulate the VVER-1000 FA. All burnup calculations are conducted with 36 depletion steps over the irradiation time, utilizing the END/B-VII nuclear data library (Chadwick 2006). We adopt 1.25×10^7 neutron histories per burnup step. The simulation process neglects the first 50 cycles before starting active tallying to allow for convergence of the fission source distribution. The reflective boundary condition is applied for all calculations in this study. Each fuel gadolinium pin is divided into 11 equally sized volume annular sub-depletion zones to account for the rim effect resulting from spatial self-shielding. The chemical spectral shift method is applied to two

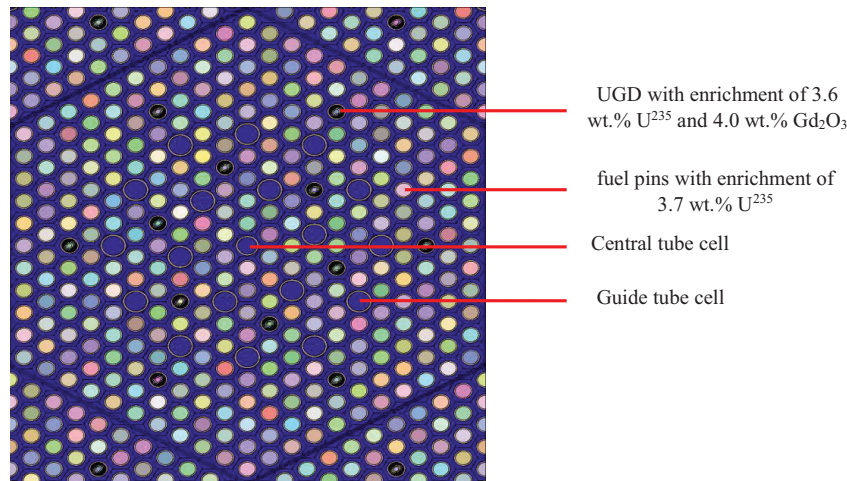


Figure 1. LEU assembly configuration obtained by Serpent code.

different cases (i.e., with and without burnable absorbers) to investigate the influence of the spectral shift on the neutronic characteristics of the assembly.

At the BOC, the D_2O/H_2O ratio starts at 65% in the case of the presence of 12 burnable absorber pins with 4.0 wt.% Gd_2O_3 (case 1), and with 73% in the case without using any burnable absorber (case 2). This ratio decreases with equal decrements of 5% in all cases over the operational cycle until it reaches a concentration of 0% at the EOC. The startup ratio of D_2O/H_2O (i.e., 65% and 73%) has been adopted to ensure acceptable initial excess reactivity. The Serpent-2 code cannot automatically apply the critical density iteration of D_2O during burnup. Therefore, the D_2O concentration is manually adjusted during the simulation through 14 different molecular ratios of D_2O/H_2O distributed over the operating cycle as follows: at the BOC, the molecular ratio of D_2O/H_2O is started with a high value until the model could not further maintain criticality conditions (i.e., $k_{\infty} > 1$). Subsequently, as the fuel cycle proceeds, the subsequent values gradually decrease until reaching 0% D_2O and 100% H_2O at the EOC. The results obtained by Serpent-2 are compared with the benchmark mean values as a reference case, with corresponding statistical errors of $\pm 64\%$ and ± 19 pcm for CR and k_{∞} , respectively.

Result analysis and discussion

The influence of the chemical spectral shift control (SSC) method on the fuel utilization and the cycle length of the VVER-1000 FA has been demonstrated by determining the time evolution of the following parameters: k_{∞} , CR, U^{235} , U^{238} , Pu^{239} , Pu^{240} , Pu^{241} , Pu^{242} , Xe^{135} , Sm^{149} . The simulation was performed by Serpent-2 for two cases: case (1) ($D_2O/H_2O + 4.0$ wt.% Gd_2O_3) and case (2) (D_2O/H_2O).

The variation of the multiplication factor with burnup

Fig. 2 illustrates the variation of the infinite multiplication factor (k_{∞}) during burnup. As depicted in Fig. 2, the reactivity is highly sensitive to the D_2O/H_2O ratio, resulting in small variations in k_{∞} during operation. Fig. 2 shows that in case (2), two important criteria have been achieved. Firstly, the reduction in D_2O quantity is lower than in the second case, which reduces the spectral shift design cost. Secondly, the suppression of k_{∞} has occurred in case (1) more than in case (2) due to the thermal absorption cross section for gadolinium. This effect is more noticeable at the EOC where more thermal neutrons are produced. Additionally, from Fig. 2, there is a small fluctuation in the k_{∞} during burnup due to the variation in D_2O/H_2O ratio that needs to be smoothly lowered and controlled to maintain a constant reactor power and temperature. Compared to the previous work (Elzayat et al. 2022), flattening the k_{∞} curve can be achieved by decreasing the rate at which the ratio of D_2O/H_2O is decreased. Fig. 2 illustrates that the

discharge burnup for both cases (1&2) of the SSC method reached about 40 MWd/KgHM compared to 25 MWd/KgHM in the benchmark model. Through comparing the discharge burnup for both cases with the benchmark mean values (blue line in Fig. 2), the fuel burnup has improved by 60% compared to the benchmark values.

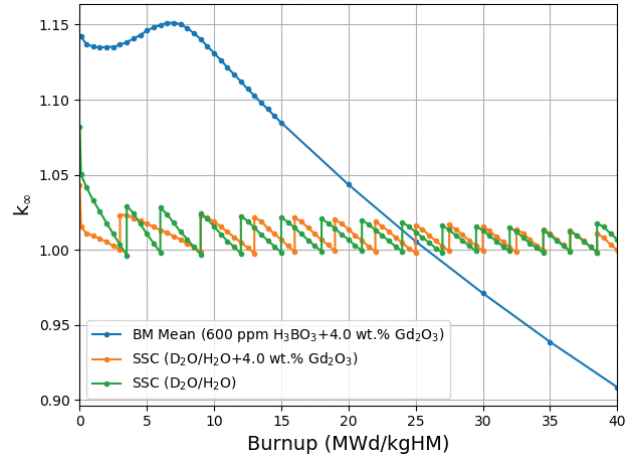


Figure 2. Variation of infinite multiplication factor during burnup.

Time evolution of the average isotopic concentration

Fig. 3 shows the average number density of U^{235} in the fuel assembly as a function of burnup. As depicted in Fig. 3, the U^{235} concentration decreases at a slow rate in both cases of the SSC method, leading to a $\approx 31\%$ increase in its unburned inventory at the EOC compared to the benchmark case. This decrease is due to the limitation of the thermal absorption cross section of U^{235} by the neutron spectrum hardening. Consequently, there is a necessity for increased U^{235} enrichment. However, the high production and effective burning of other fissile nuclides (Pu^{239} and Pu^{241}) counterbalance this drawback of the SSC design.

Fig. 4 illustrates the average number density of U^{238} in the fuel assembly vs. burnup. Fig. 4 shows a 0.62% decreasing in the U^{238} inventory at the EOC compared to the benchmark (BM) model and the consumption rate of U^{238} increases with the application of the SSC method, primarily due to the increase of its radiation capture cross-section as a function of the hardened neutron spectrum. This results in a significant accumulation of fissile plutonium isotopes such as Pu^{239} and Pu^{241} breeding in the reactor.

Figs 5, 7 show that, for the SSC design, the buildup rates for Pu^{239} and Pu^{241} isotopes increase by approximately 57% and 45%, respectively, compared to the BM at 25 MWd/HMkg. At the EOC, the maximum values of Pu^{239} and Pu^{241} are 33.7% and 29.5%, respectively, for SSC design compared to the benchmark, as shown in Figs 5, 7. The total amount of Pu^{239} in the assembly increases during the initial burnup. Afterward, more thermal neutrons are produced, leading to the progressive burning of Pu^{239} , as illustrated in Fig. 5.

As shown in Figs 6, 8, the production rate of non-fissile plutonium isotopes (Pu^{240} and Pu^{242}) has achieved a maximum increase of about 25.7% and 3.3%, respectively, at EOC for SSC method compared to the benchmark.

According to Figs 9, 10, the inventory values of Xe^{135} and Sm^{149} have increased by 24.7% and 21.9%, respectively, in the SSC design compared to the benchmark. This increase is attributed to their low burnout rate through the hardened neutron spectrum, which can delay the restarting of the reactor after shutdown. Furthermore, equilibrium cannot be achieved due to the regular variation in the D_2O/H_2O ratio throughout the fuel cycle as shown in Figs 9, 10.

Figs 11, 12 depict the time evolution of the average concentration of Gd^{155} and Gd^{157} in the fuel assembly. As illustrated, gadolinium isotopes exhibit a slower burnout rate compared to the benchmark model. Gd^{155} and Gd^{157} act as thermal neutron absorbers, and their poison effect becomes apparent at the EOC where thermal neutrons are generated.

The variation of the conversion ratio with burnup

Fig. 13 demonstrates the variation of conversion ratio (CR) during burnup for both cases of the SSC method compared to the benchmark model. At the BOC, Fig. 13

shows that the conversion ratio for Case (2) is higher than for Case (1). The conversion ratio for both cases (i.e., 1&2) is higher than for the BM mean values by about 58% and 64.4%, respectively, due to the gadolinium poison effect. As the EOC approaches, CR values decrease, coinciding with the complete depletion of gadolinium isotopes. At this stage, both cases (1&2) exhibit similar moderation conditions, with 100% H_2O content. In the benchmark model, CR starts with a low value since the poison effect of (H_3BO_3 and Gd_2O_3) dominated at BOC. Towards the EOC, the CR increases as the soluble boron and gadolinium isotopes are entirely burnt out. At the BOC, the CR in the case (2) is higher than that of case (1) due to the gadolinium effect. The conversion ratio shows rapid and smaller fluctuations with every change in the D_2O/H_2O molecular ratio, similar to the infinite multiplication factor. These fluctuations can be further reduced by diluting the D_2O/H_2O ratio by 1% throughout the fuel cycle.

We conducted a comparison between two dilution process models relative to the benchmark model (BM). Table 2 shows the variations of neutronic parameters and assembly average isotopic composition with two different dilution processes. Firstly, the molecular ratio of D_2O/H_2O was diluted by 10%, 13%, 15%, and 19%

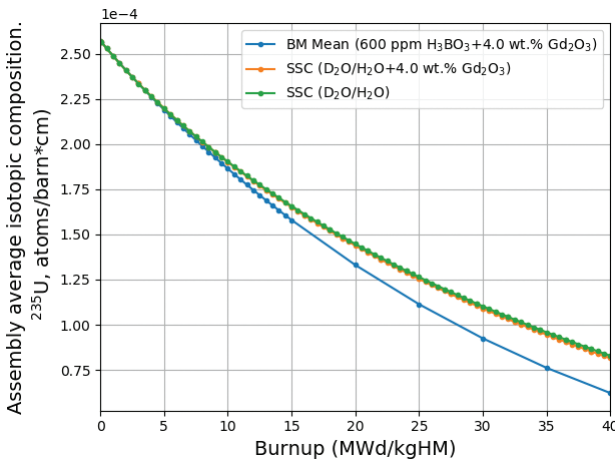


Figure 3. Average number density of U^{235} in the fuel assembly.

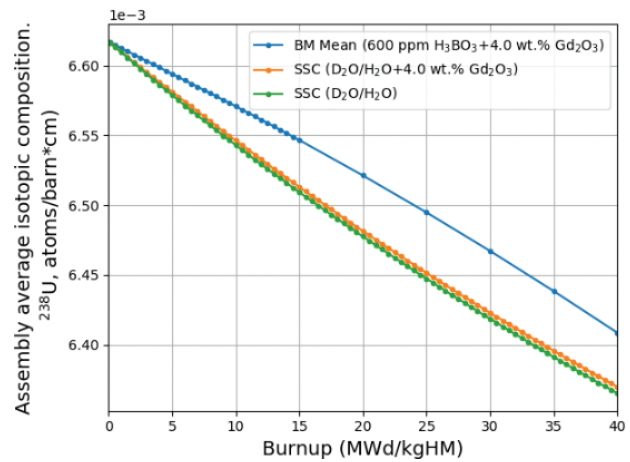


Figure 4. Average number density of U^{238} in the fuel assembly.

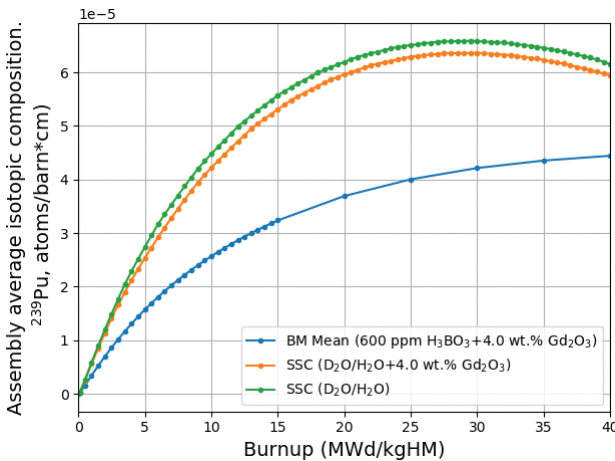


Figure 5. Average number density of Pu^{239} in the fuel assembly.

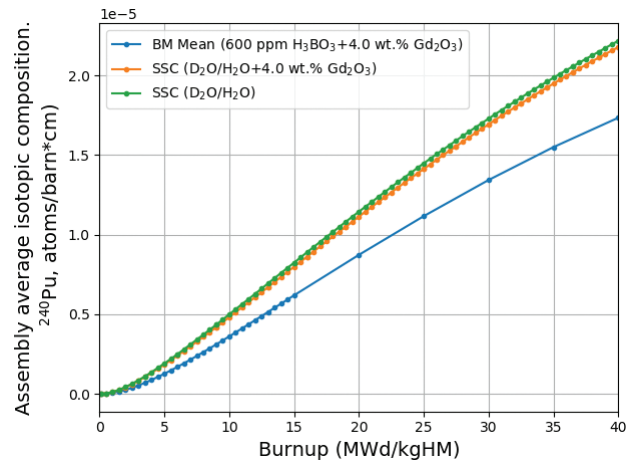


Figure 6. Average number density of Pu^{240} in the fuel assembly.

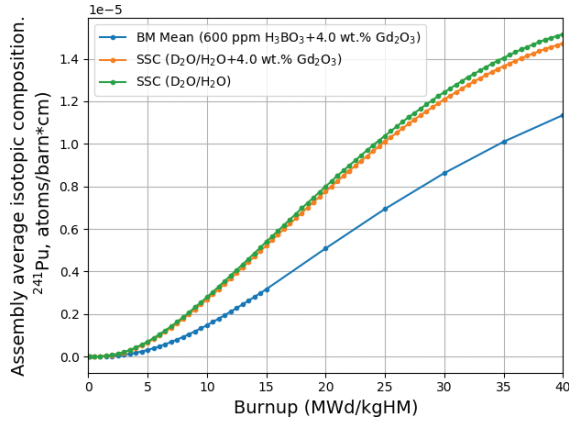
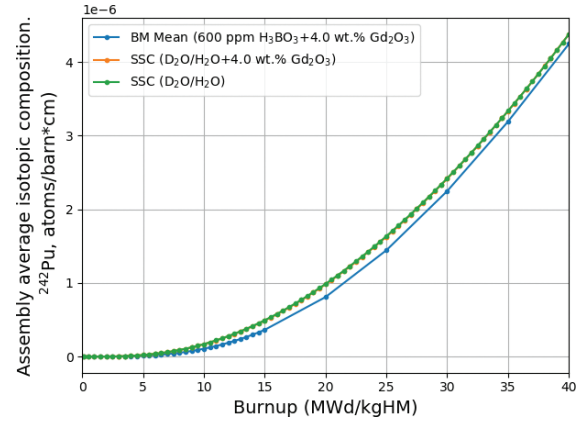
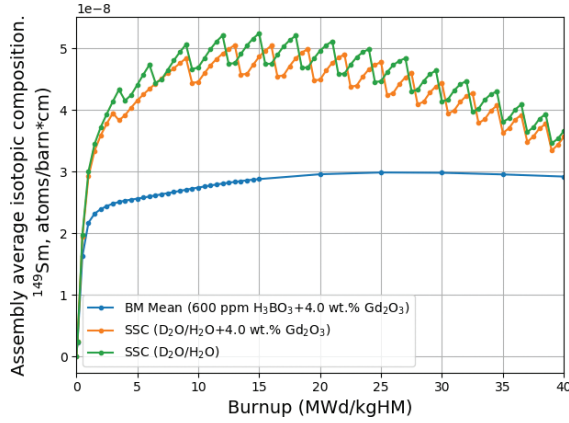
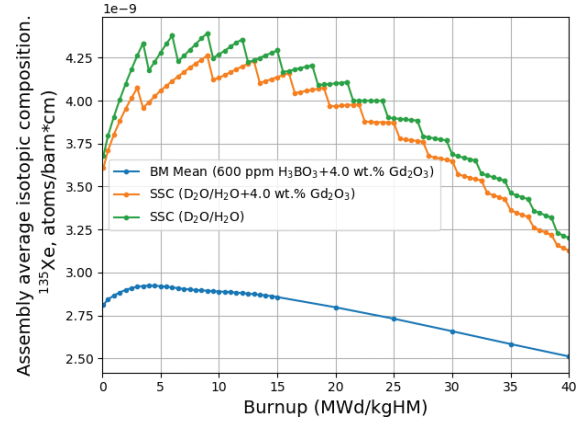
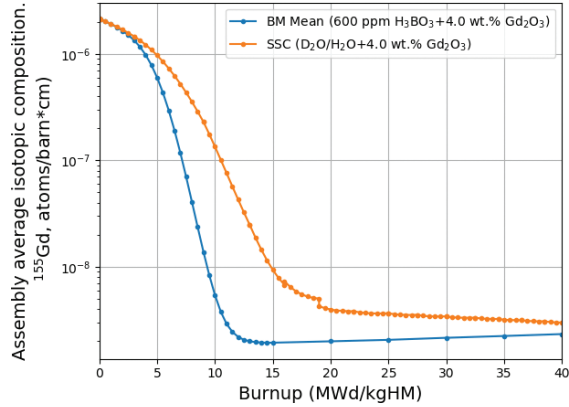
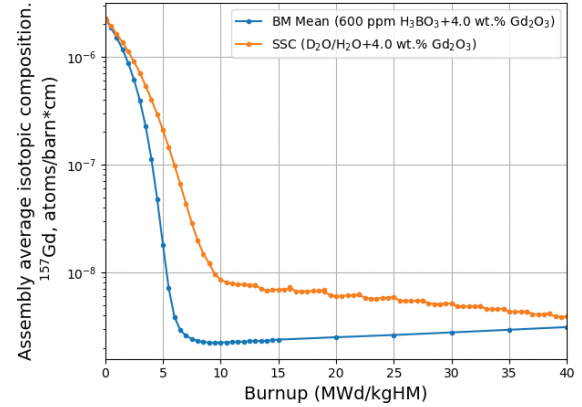
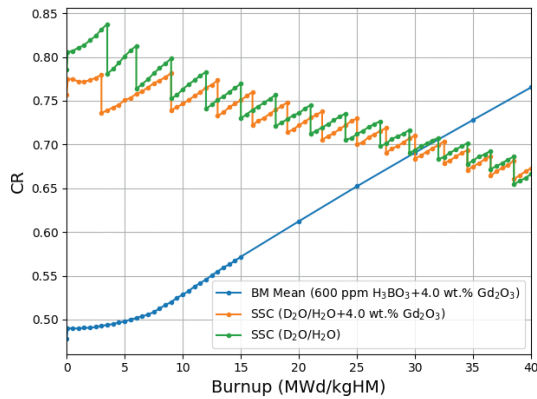
Figure 7. Average number density of Pu²⁴¹ in the fuel assembly.Figure 8. Average number density of Pu²⁴² in the fuel assembly.Figure 9. Average number density of Sm¹⁴⁹ in the fuel assembly.Figure 10. Average number density of Xe¹³⁵ in the fuel assembly.Figure 11. Average number density of Gd¹⁵⁵ in the fuel assembly.Figure 12. Average number density of Gd¹⁵⁷ in the fuel assembly.

Figure 13. Variation of conversion ratio during burnup.

along the fuel cycle, as shown in Elzayat et al. (2022). Secondly, the molecular ratio of D_2O/H_2O was diluted by 5% throughout the burnup process. Through adjusting the D_2O/H_2O molecular ratio smoothly, an improvement occurs in the neutronic parameter as in k_∞ and CR which varies with small fluctuations with varying the D_2O/H_2O ratio, as listed in Table 2. Additionally, a better utilization occurs for burning U^{235} and U^{238} to bred more fissile material Pu isotopes. Comparing Figs 5, 7 with those in previous study for the same isotopes (Elzayat et al. 2022) shows that Pu²³⁹ and Pu²⁴¹ exhibit a higher buildup value for a 5% decrement along the fuel cycle, increasing by 9.5% and 15.5%, respectively,

Table 2. Comparison of neutronic parameters and assembly average isotopic composition for different dilution processes

Neutronic parameters		Previous work (Elzayat et al. 2022)	Current work
		The dilution process	
		10%, 13%, 15%, and 19%	5%
k_{∞}		Shows rapid and large fluctuations (the maximum fluctuation in $k_{\infty} < 7\%$)	Shows rapid and small fluctuations (the maximum fluctuation in $k_{\infty} < 2.9\%$)
CR		Shows rapid and large fluctuations (the maximum fluctuation in CR < 14%)	Shows rapid and small fluctuations (the maximum fluctuation in CR < 6%)
Assembly average isotopic composition	U^{235}	The inventory increases by 32% compared with BM at EOC.	The inventory increases by 31% compared with BM at EOC.
	U^{238}	The inventory decreases by 0.45% compared with BM at EOC.	The inventory decreases by 0.62% compared with BM at EOC.
	Pu^{239} and Pu^{241}	The inventory increases by 39% and 28%, respectively, compared with BM at EOC.	The inventory increases by 33.7% and 29.5%, respectively, compared with BM at EOC.
	Pu^{240} and Pu^{242}	The inventory increases by 18% and 3%, respectively, compared with BM at EOC.	The inventory increases by 25.7% and 3.3%, respectively, compared with BM at EOC.
	Xe^{135} and Sm^{149}	The inventory increases by 51% and 66%, respectively, compared with BM at EOC.	The inventory increases by 24.7% and 21.9%, respectively, compared with BM at EOC.

compared to larger decrements at 25 MWd/HMkg. This indicates that a smaller decrement produces harder neutrons, which breed more plutonium fissile isotopes that are effectively burned at the EOC. The buildup rate of Xe^{135} and Sm^{149} decreases when diluting by 5% compared to using a small number of large decrements as listed in Table 2. Table 2 demonstrates better utilization of neutrons throughout the dilution process by 5% along the fuel cycle, which can change the reactivity in a smoother manner.

Conclusion

Based on the preceding analysis, it is evident that the SSC design offers several advantages, including enhanced conversion ratio throughout the operational cycle, improved fuel utilization, and a prolonged reactor life cycle.

It was observed that for both cases of the SSC method, there is a 60% improvement in the fuel discharge burnup compared to the benchmark reference model. Moreover, the existence of gadolinium in the first case of the SSC method would result in a reduction of D_2O quantity by

12% at the BOC and by 100% at the EOC relative to the second case, without affecting the achieved discharge burnup. This, in turn, could potentially reduce the capital cost of implementing the SSC method.

Smoothly reducing the (D_2O/H_2O) ratio by 5% results in an enhancement in the variation of k_{∞} during burnup and in the conversion ratio values, besides better utilization of the fuel source to breed more fissile material. Furthermore, significant improvements can be achieved by decreasing it by 1% or lower throughout the process.

Comparing case (2) to case (1), several improvements are notable, including enhanced conversion ratio, increased breeding of plutonium isotopes, better utilization of U^{238} , and reduced use of enriched U.

However, there are also drawbacks to consider. The equilibrium condition of Xe^{135} and Sm^{149} has not been achieved. It is important to highlight that in the SSC design, the significant contribution of Pu^{239} and Pu^{241} to power generation will reduce the values of the main kinetic parameters. These continuous physical changes in the reactor parameters during the operating cycle emphasize the necessity of conducting a precise safety analysis for this type of reactivity control.

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