Possibilities of Better Utilization of MOX fuel in VVER type reactors by optimizing neutron spectrum

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Abstract. The introduction of MOX fuel in modern nuclear fuel cycles has posed attention in all leading nuclear countries. However, the introduction of plutonium in the fuel loading in light water reactors has neutron-physical limitations. Improve the consumption of plutonium in MOX fuels is one of the important issues that have to be studied to decrease the risk of proliferation resistance. The impact of varying neutron spectrum on the discharged plutonium was studied through the variation of the fuel pellets diameter in a VVER type MOX fuel assembly without changing the lattice pitch. Neutronic calculations were performed using: MCU, Serpent, and GETERA codes. It was shown that the neutron spectrum plays a dominant role in the consumption and production of nuclides. Also, the usage of assemblies with high moderator to fuel volume ratio burned higher amounts of fissile plutonium in comparison with assemblies with low moderator to fuel volume ratio.

1. Introduction

An average light water reactor produces 20 metric tons of used nuclear fuel (UNF) per year [1]. This UNF is approximately 95% uranium, 1% plutonium, and 4% fission products and other transuranic elements. Plutonium produced in nuclear power reactors is an important component of the future advanced nuclear power [2]. The plutonium in UNF can be recovered through reprocessing. The plutonium can then be used in the manufacture of mixed oxide (MOX) nuclear fuel, to substitute for fresh uranium oxide fuel.

Recycling of reactor-grade plutonium (RGPu) to the nuclear fuel cycle in the form of MOX fuel has long been successfully used in many nuclear power operating reactors. Also, the recycling of weapons-grade plutonium (WGPu) has been considered as an option to decrease the number of nuclear warheads and to bring WGPu to the level of spent fuel standard (SFS). The U.S. Department of Energy (DOE) expressed this standard as follows [3]: “A concept to make the plutonium as unattractive and inaccessible for retrieval and weapons use as the residual plutonium in the spent fuel from commercial reactors”.

In the Russian Federation, the long-term strategy for nuclear power development is based on the use of recycled plutonium mainly in fast reactors, to a minor extent in thermal reactors [4]. However, the introduction of plutonium in the fuel loading in light water reactors has neutron-physical limitations.
The most significant limitations for the introduction of plutonium are the difference in the fraction of delayed neutrons (For $^{239}$Pu it is three times less than that for $^{235}$U), and the reduction in the control rod worth due to the hardening of the neutron spectrum. This difference manifests itself in the efficiency of the control and safety of reactors, hence complicates the share of Pu in light water reactors. This encourages research in studying the physical properties of reactors with mixed loading of MOX and LEU fuels.

2. Neutron spectrum effects

Optimizing the neutron spectrum in a reactor core is considered as an effective way to achieve desired safe and economic performance [5][6]. Changing the neutron spectrum can be done through two main ways: changing the enrichment and/or changing the hydrogen to heavy metal ratio (H/HM). Optimizing the design parameters by selecting H/HM is considered as a main step in reactor design calculations. Changing H/HM for the goal of increasing the consumption of plutonium in MOX cores have been widely studied for the medium and long-term development of nuclear industry [7–9].

2.1. Hydrogen to heavy metal ratio (H/HM)

The hydrogen (H) to heavy metal (HM) atom ratio is usually used to represent to which degree the core is moderated in a light water reactor. H/HM atom ratio is the number of hydrogen atoms per number of fuel atoms in the unit lattice [10], it can be calculated by the relation:

$$\frac{H}{HM} = \frac{N^\text{hydrogen}}{N^\text{fuel}} \times \frac{V_M}{V_F}$$  \hspace{1cm} (1)

Where $N^\text{hydrogen}$, $N^\text{fuel}$ are the hydrogen and fuel atomic number density. $V_M$ and $V_F$ are the moderator and fuel volume. $V_M/V_F$ is the moderator to fuel volume ratio. From the equation and since the density of uranium dioxide does not change in a wide temperature range, it is shown that changing the hydrogen to heavy metal ratio can be accomplished by varying the water density, or by varying the moderator to fuel volume ratio.

For a VVER unit cell (hexagonal lattice), the moderator to fuel volume ratio can be described by the relation:

$$\frac{V_M}{V_F} = \frac{2\sqrt{3}}{\pi} \left( \frac{P}{D_F} \right)^2 - \left( \frac{D_c}{D_F} \right)^2$$  \hspace{1cm} (2)

Where $P$ represents the pitch of the lattice, $D_c$ is the diameter of the pin (outer diameter of clad), $D_F$ is the diameter of the fuel pellet. Note that $V_M/V_F$ for an assembly will be higher than that for a unit cell due to the excess water at the periphery of the assembly.

2.2. Effects of Hydrogen to heavy metal ratio change

Infinite multiplication factor is used to describe the efficiency of the neutron multiplying process. Changes in the H/HM ratio could be described through the parameters of the four-factor formula describing the infinite multiplication factor [11].

$$K_\infty = \eta f p e$$  \hspace{1cm} (3)

Where $\eta$ is the number of fission neutrons produced per absorption in the fuel, $f$ is the thermal utilization, $p$ is the resonance escape probability and $e$ is the fast fission factor. Varying H/HM will lead to change in the infinite multiplication factor because of the dependence of each of the factors on it. Although changing H/HM has no direct effect on $\eta$, it leads to change in the thermal spectrum, and hence slightly on $\eta$. As a result, it is possible to neglect the impact of changing H/HM on $\eta$.

Increasing H/HM increases the share of moderator in the absorption of neutrons. Hence, neglecting the neutron spectrum effects, the increase in H/HM leads to decrease in the thermal utilization. Therefore, $f$ is inversely proportional to H/HM.
With the increase in the H/HM, the resonance escape probability will increase due to the increase of moderation. Therefore, \( p \) is directly proportional to H/HM.

With the increase in H/HM, the probability of scattering on nuclei of hydrogen increases for the above fast fission threshold neutrons. This scattering will lead to a significant reduction of the neutron energy and thus derives neutron energy beyond the fission threshold. As a result, this process will lead to decrease in the value of \( \varepsilon \). Therefore, inverse relation between \( \varepsilon \) and H/HM.

As a summary, it can be noted that the dependence of \( K_{\infty} \) on H/HM is mainly determined by two factors, \( p \) and \( f \). The dependences of these quantities on H/HM have opposite character.

3. Methodology

3.1. VVER-1000 MOX Fuel Assembly

For this study, a profiled VVER-1000 MOX fuel assembly was chosen [12]. It consists of a mix of uranium oxide UO\(_2\) and reprocessed WG (Weapons-grade) plutonium with an average fissile Pu content \( \approx 3.3\% \) in the form of PuO\(_2\). The fuel assembly is shown in figure 1.

![Figure 1. VVER-1000 MOX fuel assembly.](image)

The fuel pellets has a diameter of 7.72 mm, while the clad thickness is 0.722 mm. The hexagonal cell pitch is 12.75 mm. The clad and structural materials are composed of a Zr-Nb alloy. Operating parameters for the assembly is listed in table 1.

| Parameter                                | Description |
|------------------------------------------|-------------|
| State                                    | Operating non-poisoned state |
| Boundary condition                       | Reflective |
| Fuel temperature (K)                     | 1027        |
| Non-fuel temperature (K)                 | 575         |
| Boron concentration in moderator (ppm)   | 600         |
| Power density (MW/m\(^3\))               | 108         |
The study of the effects of changes in the neutron spectrum in the reactor core is carried out by studying the effect of these effects on the fuel assembly. A number of calculations were performed for the assembly with reflective boundary conditions. The diameter of the fuel pellets in the assembly varied from 5.72 to 8.72 mm in 0.2 mm increments (fuel rod diameter from 7.16 to 10.16 mm). This variation was conducted without varying the lattice pitch.

3.2. Computational tools

An essential step in reactor physics calculation is the availability of computer simulation codes, capable of performing various reactor design calculations tasks. These computer codes can provide sufficiently accurate solutions for a given problem when compared with real operating reactor data. In this study, GETERA code, which is based on first flight collision probability method, MCU and SERPENT codes, which are Monte-Carlo based codes, were selected as the main tools for the calculations.

The GETERA code is intended for calculation of neutron space-energy distribution in nuclear reactor cells and polycells [13]. The first version of the code was created in 1990 in the national research nuclear university (Moscow engineering physics institute). Its library has nuclear data for 135 nuclides in 299 energy groups [14] based on the nuclear data library BNAB-93. The code showed a very good capability to account for fuel depletion calculations [15][16]. Fuel assemblies that contain multiple cells is described in GETERA by polycells model. For VVER type cells, each hexagonal cell is converted to an equivalent circle-shaped cell (Wigner-Seitz cell). The Wigner-Seitz cell model represents the moderator as an annular region that conserves the appropriate ratio of moderator to fuel region. 66 cells of moderator is added to the periphery of the assembly to simulate the outer boundary containing moderator.

MCU (Monte Carlo Universal) is a project on development of a universal computer code for simulation of particle transport such as: neutrons, photons, electrons and positrons in three-dimensional systems by means of the Monte Carlo method [17]. The project was developed at Kurchatov Institute. MCU-FREE is a free version of the version MCU-5 which was developed in 2009 for educational and research purposes [18]. It provides simulation only for neutron transport in three-dimensional systems. The main data bank contains data for 375 isotopes. Main sources for the data bank development are BNAB, LIPAR, ENDF/B, JENDL and JEFF. The MCU-5 package contains the MCU-Office tool [19] responsible for visualization of input and output data.

Serpent is a multi-purpose three-dimensional continuous-energy Monte Carlo particle transport code. It was developed at VTT Technical Research Centre of Finland [20]. Version 2.1.29 was used with libraries based on ENDF/B-VII evaluated data files.

4. Results and discussion

4.1. Initial Reactivity

The H/HM for the fuel assembly has been optimized in order to maximize the increase in initial reactivity and hence the associated increase in fuel burnup. Initial infinite multiplication factor versus \( V_M/V_F \) and H/HM for the assembly is shown in figure 2. The vertical line shows the \( V_M/V_F \) corresponding to the standard fuel assembly.

An assembly with standard dimensions (fuel pellet diameter of 7.72 mm) is in the under moderation region. To achieve the maximum initial infinite multiplication factor, higher H/HM ratios in the assembly is required. The optimal \( V_M/V_F \) corresponds to a fuel pellet diameter of 7.56 mm. The maximum initial infinite multiplication factor has an average value (\( k_\infty = 1.23912 \)) at \( V_M/V_F \) equal to 3.7 and H/HM equal to 7.8.

Optimal value of H/HM divides the entire range of variation of H/HM in two regions. In the first region, where H/HM is less than \((H/HM)_{\text{optimal}}\), the reduction of \( K_\infty \) occurs due to the increase in resonance absorption of neutrons. In the second region, where H/HM is more than \((H/HM)_{\text{optimal}}\), the decrease in \( K_\infty \) occurs due to the increase in absorption of thermal neutrons by the moderator.
Compared to MCU and Serpent, GETERA results were lower in all cases. However, the optimum diameter is the same as in MCU and Serpent.

4.2. Neutron spectrum
To observe the effect of varying the H/HM on the neutron spectrum, the neutron spectrum divided into the 28 neutron energy groups of the BNAB-93 nuclear data library for 3 different $V_M/V_F$ ratios is presented in figure 3.

![Neutron spectrum](image)

**Figure 2.** Initial $K_\infty$ versus $V_M/V_F$ and H/HM.

**Figure 3.** Neutron spectrum presented in 28-energy groups for three different $V_M/V_F$.

It is shown that increasing $V_M/V_F$ shifts the spectrum towards thermal energies while decreasing it increase the population of neutrons with higher energies. This shift in the neutron spectrum, caused by
the change in the level of moderation in the assembly, leads to a change in the rate of the capture and fission reaction of the main nuclides.

Neutron spectrum hardness is defined as the ratio of the flux of fast neutrons ($E_n > 1$ eV) to the flux of thermal neutrons ($E_n < 1$ eV) [21]. The dependence of the hardness of the neutron spectrum on $V_M/V_F$ is shown in figure 4. The hardness of the neutron spectrum decreased with increasing $V_M/V_F$. Higher $V_M/V_F$ (H/HM) means more hydrogen atoms and therefore better neutron moderation (neutrons will have lower energies on average). The results of the hardness of the neutron spectrum obtained by the three programs showed good agreement. Since the optimization was carried out for two-dimensional fuel assembly with reflective boundary conditions, the hardness values of the neutron spectrum are not associated with leakage.

![Figure 4. Hardness of neutron spectrum versus $V_M/V_F$ and H/HM.](image)

It can be observed that the hardness of the neutron spectrum for MOX assemblies is high in comparison with LEU assemblies; this can be explained by the higher absorption cross section in the thermal region in MOX assembly. Also, the number of neutrons produced per fission is higher for $^{239}$Pu than $^{235}$U and the fission yield of $^{135}$Xe from $^{239}$Pu is lower than that for $^{235}$U which means that fewer neutrons will be absorbed by $^{135}$Xe.

4.3. **Burnup depth and cycle length**

Burnup calculations were performed using the GETERA and MCU codes. The fuel has depleted with a constant power density of 108 MW/m$^3$ to an infinite multiplication factor of $k_\infty = 1.03$ at the end of cycle to account for leakage. Burnup depth versus $V_M/V_F$ is shown in figure 5. For burnup calculations using MCU, enough number of neutron histories and burnup steps were used for calculation to obtain low statistical errors.

Typically, an increase in reactivity at the beginning of a cycle ($k_c$ at BOC) will result in higher burnup. For the assembly with $V_M/V_F$ equal to 2.6 (fuel pellet diameter of 6.92 mm), an increase in the burnup depth by 10.8% was observed. Since the burnup depth and the cycle time in effective full power days (EFPDs) compete with each other, higher fuel burnsups with a smaller fuel rod diameter result in shorter cycles. Figure 6 shows the cycle length versus $V_M/V_F$. The longest cycle occurred with $V_M/V_F$ equal to 1.7.
4.4. Plutonium consumption

The discharged plutonium concentrations at the end of cycle (EOC) as a function of $V_M/V_F$ is listed in table 2. It is shown that increasing $V_M/V_F$ (H/HM) ratios decreases the concentration of discharged plutonium averaged over the entire assembly. The maximum plutonium concentration achieved at the lowest studied $V_M/V_F$.

Harder neutron spectrum results in higher percentages of $^{239}$Pu and lower percentages of $^{240}$Pu at the EOC. The percentage of $^{239}$Pu burned at the end of the cycle increases with increasing $V_M/V_F$ as shown in figure 7. However, the accumulation of $^{240}$Pu and $^{241}$Pu increases with increasing $V_M/V_F$. The ratio of the discharged concentrations of $^{240}$Pu and $^{241}$Pu to their initial concentration is shown in figure 8.
Table 2. Discharged plutonium composition for different $V_M/V_F$.

| $V_M/V_F$ | H/HM | $^{238}$Pu (%) | $^{239}$Pu (%) | $^{240}$Pu (%) | $^{241}$Pu (%) | $^{242}$Pu (%) | Concentration Pu ($g/cm^3$) |
|-----------|------|----------------|----------------|----------------|----------------|----------------|---------------------------|
| 2.83      | 5.94 | 0.05           | 61.46          | 26.55          | 10.21          | 1.73           | 0.224                     |
| 2.6       | 5.46 | 0.05           | 62.07          | 25.94          | 10.25          | 1.68           | 0.227                     |
| 2.39      | 5.02 | 0.06           | 62.80          | 25.20          | 10.29          | 1.64           | 0.231                     |
| 2.19      | 4.61 | 0.06           | 64.03          | 24.25          | 10.12          | 1.54           | 0.237                     |
| 2.02      | 4.24 | 0.06           | 65.53          | 23.12          | 9.88           | 1.41           | 0.243                     |
| 1.85      | 3.89 | 0.06           | 67.11          | 21.97          | 9.58           | 1.28           | 0.249                     |
| 1.7       | 3.57 | 0.06           | 69.02          | 20.69          | 9.12           | 1.12           | 0.256                     |
| 1.56      | 3.28 | 0.05           | 71.30          | 19.25          | 8.47           | 0.93           | 0.264                     |
| 1.43      | 3.01 | 0.04           | 74.28          | 17.44          | 7.53           | 0.71           | 0.272                     |
| 1.31      | 2.75 | 0.03           | 77.90          | 15.32          | 6.28           | 0.48           | 0.280                     |
| 1.2       | 2.52 | 0.01           | 81.54          | 13.17          | 4.98           | 0.29           | 0.286                     |

4.5. Accumulation of Actinides

In terms of storage requirements, neptunium is the dominant radiotoxic element with an extremely long half-life. Therefore, the amount of $^{237}$Np must be kept minimum in spent fuel. Several concentrations of actinides at the EOC as a function of $V_M/V_F$ obtained using MCU code is shown in figure 9. In contrast to the concentration of plutonium, the peak of neptunium occurs at $V_M/V_F$ ratios slightly lower than the $V_M/V_F$ ratio for the standard assembly.
It is shown that the maximum concentration of $^{241}\text{Am}$ was obtained at the same $V_M/V_F$ ratio as for $^{237}\text{Np}$, due to the fact that $^{237}\text{Np}$ is a daughter of $^{241}\text{Am}$. Soften the neutron spectrum and the corresponding increase in burning out the $^{241}\text{Am}$ will make it possible to avoid the accumulation of short-lived $^{242}\text{Cm}$, which causes challenging problem dealing with spent fuel due to his high heat release.

5. Conclusion
The main objective of this work was a physical analysis of the possibilities of increasing the efficiency of MOX fuel usage in thermal reactors by optimizing the neutron spectrum. It was shown that the neutron spectrum can be varied by varying the hydrogen to heavy metal ratio through the variation of the moderator to fuel volume ratio. Since the optimization was performed for a 2D fuel assembly with reflective boundary conditions, the optimized H/HM ratio is not related to leakage.

It was shown that the usage of assemblies with higher H/HM ratios will increase the burnup depth, while decreasing the cycle length. An optimal moderator to fuel volume ratio of 2.83 was determined to achieve the maximum increase in burnup. In assemblies with a low H/HM ratios, burnup was limited due to that neutrons were leaked from the system as a result of captures.

The usage of assemblies with high H/HM ratio softens the neutron spectrum and hence increase the discharged $^{240}\text{Pu}$ in the spent fuel. This helps in proliferation resistance point of view. However, the temperature coefficient for the moderator turned out to be less negative than the coefficient for the standard assembly which is an important safety factor. Soften the neutron spectrum leads to more intensive elimination of highly toxic long-lived minor actinides.

The results obtained using MCU and Serpent showed very good agreement. The GETERA results showed deviations from the results obtained with the other two programs. This deviation is mainly due to the fact that GETERA is an engineering program that solves the stationary transport equation using the first collision probability method and the absence of some isotopes in the GETERA data library. Also, GETERA transforms a hexagonal cell into a Wigner-Seitz cell and since the moderation in a hexagonal cell is greater than in a cylindrical cell (Wigner-Seitz cell), plutonium is sensitive to spectrum changes due to 0.3 eV resonances in $^{236}\text{Pu}$ and $^{240}\text{Pu}$ and a 1.1 eV resonance in $^{242}\text{Pu}$.

![Figure 9. Atomic concentration of several actinides versus $V_M/V_F$ and H/HM.](image-url)
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