Neutronic analysis of VVER-1000 fuel assembly with different types of burnable absorbers using Monte-Carlo code Serpent

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Abstract. A neutronic study on the fuel assembly of a Russian type nuclear reactor VVER-1000 fuelled with low enriched Uranium (LEU) plus 12 \( \text{UO}_2 + 4\% \text{Gd}_2 \text{O}_3 \) rods was performed. This type of fuel requires validated computational methods and codes able to provide reliable predictions of the neutronics characteristics. Gadolinium self-shielding effect and isotopes accumulation in Rim region make it necessary to study the geometric modelling effect on the code calculations. The modelling of this fuel type was tested using Monte-Carlo and deterministic codes. In this study, Serpent results are verified using two nuclear data libraries ENDFb.6.8 and ENDFb.7. Also, this study investigates the effect UGd rods division into multiple radial layers on the reactivity, isotopic generation and burnup radial distribution. The same procedure is done on another type of neutron absorber Erbium (UEr) and the results are compared with UGd. The sensitivity of the results determines the validity of Monte-Carlo code in such a computational task comparing two types of neutron absorbers in addition to determining the geometric requirements.

1. Introduction

During reactor operation, the main task is to keep the fission reaction going while preventing any deviations in the fixed parameters for temperature, pressure and reactivity from the safe values. Reactor control is usually carried out by varying the effective multiplication factor (\( K_{\text{eff}} \)). If the value of \( K_{\text{eff}} \) is held at unity, then the power level of the core will remain at a steady level, if \( K_{\text{eff}} \) is increased above unity the power level will rise and if \( K_{\text{eff}} \) is reduced below unity, the power will fall, and the reactor will be shut down.

Reactivity is a measure of reactor control, it is equal to \( (K_{\text{eff}} - 1)/K_{\text{eff}} \), and if this value is greater than zero then the system is said to possess excess reactivity. A negative reactivity value is a measure of the amount by which the reactor is subcritical and it is required to compensate for the excess reactivity specially at the beginning of fuel cycle. At the start-up of a reactor - the first irradiation cycle of the core - a large amount of excess reactivity is available due to the large amount of fuel. Both the fission rate and neutron flux can increase extremely rapidly. Thus, a control system is needed to prevent a large release in reactivity at the first period. The most effective tool for reactivity control at the beginning of reactor operation is the insertion of neutron absorbers either as flat late, cylindrical rods or soluble solution.

Neutron absorbers are set inside fresh fuel assemblies either as a solution in the moderator or Integral Burnable Absorber (IBA). The latest is considered more effective as it contributes in
controlling the neutron generating in the core without relying on control rods or other control mechanisms. It helps in mitigating the increase in reactivity at the beginning of operation especially at the first third of the assembly life [1]. PWR fuel uses several different types of IBAs. The IBAs include gadolinia (Gd₂O₃) or erbium (Er₂O₃) that are mixed with uranium dioxide (UO₂) fuel. Integral neutron absorbers are set in two ways: 1- Burnable poison rods (BPR), placed instead of a guide tube (WABA) or a fuel rod. In one assembly, the number of BPR used alters depending on the neutron absorber thermal absorption cross section. Several general types of BPRs have been applied in PWR fuel. Framatome Cogema Fuels use BPRs composed of B₄C-Al₂O₃ pellets contained in zircaloy tubing (Recently, the borosilicate glass (B₂O₃-SiO₂ with 12.5 wt% B₄O₃) in the form of Pyrex tubing is suggested to be used as BPRs [2]). 2- Outer surface of fuel pellets (IFBA) reducing the pin power peaking throughout the assembly [3].

The selection of neutron absorbers depends on their chemical, physical and -most importantly- nuclear characteristics. Gadolinium has the largest thermal absorption cross section followed by Boron. Hafnium produces larger number of decay isotopes. CdO, Sm₂O₃, Gd₂C₃ and Gd₂O₃ were found to deplete fast, while Er₂O₃, Eu₂O₃, Dy₂O₃ deplete slowly. The residual reactivity of all of them (Except for Gadolinium Oxide and Gadolinium Carbide) is high [4]. Boric acid is usually used as soluble solution in the moderator as it depletes fast with no minimal trace. The isotope B-10 produces helium and lithium [5], which both are stable, non-radioactive isotopes. However, chemical shim can have a major effect on the moderator void coefficient of reactivity [6]. Boron carbide has many unique properties; high neutron worth, chemical stability, high melting temperature, low density and low cost [7]. Since control rod systems are expensive, any reduction in the number of control rods reduces the total cost of the reactor.

1.1. Gadolinium

Only two gadolinium isotopes have the largest thermal neutron absorption cross-section among other isotopes (Gd-155 and Gd-157 have 60,000b and 200,000b, respectively). Inside PWR assembly fuel rods, Gadolinium produces a shift in the thermal neutron spectrum preventing them from penetrating the material block (Self-Shielding) [3]. It exhibits residual reactivity suppression due to the conversion of Gd¹⁵⁵ into Gd¹⁵⁶ and Gd¹⁵⁷ into Gd¹⁵⁸ which both don’t convert into further isotopes. When used as integral neutron absorber, gadolinium needs to be concentrated in small number of fuel rods to avoid too fast depletion. Oxide gadolinium Gd₂O₃ is used in this case because gadolinium reacts quickly with hot water. Gadolinia displaces uranium from the fuel matrix, results in reduced heavy metal loading and shorter cycle. Nevertheless, residual negative reactivity remains due to the presence of other gadolinium isotopes that are not completely destroyed (¹⁵⁵Gd and ¹⁵⁷Gd).

1.2. Erbium

Erbium is also used in mixed form with Uranium oxide (UO₂). Erbium oxide Er₂O₃ (Erbia) has two advantages upon Gadolinium: 1- The low neutron absorption cross section accounts for a smooth redistribution of power after Erbia is consumed. It depletes more slowly than boron and gadolinium, thus providing reactivity control over extended cycle lengths [8]. 2- Erbium leads to a more negative moderator temperature coefficient (MTC) due to its resonance behavior, at which ¹⁶⁷Er has a relatively wide resonance in the epithermal range, at 0.5 eV [9]. This isotope has thermal neutron absorption cross section equal to 649b, it produces Er¹⁶⁸ (2.7b). In the other hand, due to the isotope ¹⁶⁷Er in natural Erbium, Erbia has a non-trivial residual reactivity penalty even at the end of cycle [8].

1.3. Serpent

Monte-Carlo calculation is based on randomly track neutrons, photons or charged particles taking into account the stochastic error. Serpent is a multipurpose three-dimensional continuous-energy Monte Carlo particle transport code developed since 2004 at VTT Technical Research Centre of Finland [10]. One of the characteristic features of Serpent is its tracking routine, it is based both on surface-tracking and the Woodcock delta-tracking method. The latter can reduce the calculation time quite significantly
in typical reactor calculations, in which the geometry dimensions are small compared to the neutron mean-free-path [11]. The code applications include criticality calculations, fuel cycle, research reactors, coupling with thermal hydraulics codes, radiation dose rate calculations, shielding and fusion and medical physics. An extended version was realised for more complicated reactor physics modelling.

2. Model

A joint study was done between the United States and Russian Federation in 2000 to verify the Russian calculation methods on the calculational codes and the design of MOX fuel assemblies and core configurations [12]. The main goal is to work cooperatively to dispose the weapons-grade plutonium. The results of the study are published in the document “A VVER-1000 LEU and MOX Assembly Computational Benchmark”.

The assemblies studied in the benchmark are a uniform LEU fuel assembly with 12 UGd rods and a profiled MOX fuel assembly with 12 UGd rods. In this study the work applied only on the uniform LEU hexagonal fuel assembly which contains 312 fuel pins location, Figure 1, Figure 2 and Table 1. Areas in between the cells and space not belonging to the geometry are filled with water and boron. The fuel is 3.7 wt.% and 3.6 wt.% enriched with Gd2O3 content equal to 4.0 wt.%. The clad and structural material are composed of Zr-Nb-Hf. The burnup calculations are carried at power density equals to 108 MW/m² up to 40 MWd/kgHM burnup with sufficient number of burnup steps to provide accurate results, particularly during the burn out of the Gd absorber. Burnup step − 1 MWd/kgHM in the interval 0-15 MWd/kgHM and 5 MWd/kgHM in the interval 15-40 MWd/kgHM). Both Xe¹³⁵ and Sm¹⁴⁹ are in poisoned state. The temperature of fuel is 1027 K and elsewhere is 575 K. Although, Axial and radial temperature distribution have effect on the calculations, e.g. Rim region [13] [14]. Axially, the boundary condition is infinite while reflected at the sides. The parameters of interest in the benchmark are: $K_{inf}$, isotopic concentrations and burnup radial distribution.

![Figure 1. Low Enriched UGd VVER-1000 Assembly; Where; 1. Central tube cell 2. Fuel cell (with UO₂, 3.7 wt.% LEU) 3. Guide tube cell4. Fuel cell (3.6 wt.% LEU with 4.0 wt.% Gd₂O₃).](image)

| Cell Type        | Radius (cm)          |
|------------------|----------------------|
| Fuel cell        | $R_1=0.386$          |
|                  | $R_2=0.4582$         |
| Central tube cell| $R_1=0.48$           |
|                  | $R_2=0.5626$         |
| Guide tube cell  | $R_1=0.545$          |
|                  | $R_2=0.6323$         |

Table 1. Lattice cells dimensions.
3. Results

3.1. Benchmark and Serpent2 Results
Criticality calculation and isotopic generation of assembly are done using two different data libraries: ENDFb.7 and ENDFb.6.8. The Serpent results are compared to other deterministic and Monte-Carlo codes from the benchmark. Results of ENDFb.6.8 show closer results to the benchmark average as it is one of the libraries used there. In the other hand, results of ENDFb.7 show a maximum absolute deviation of about 0.006 pcm specially in the first burnup steps. The values of infinite multiplication factor versus burnup are shown in Figure 3. Atomic densities of U-238 and Pu-239 are measured for one cell containing mixture of oxide uranium and oxide gadolinium. The maximum relative deviation of Serpen2 results for atomic densities of these isotopes is 0.07%. The depletion of U-238 and generating of Pu-239 for benchmark codes and Serpent2 are shown in Figure 4 and Figure 5.

Figure 3. Infinite multiplication factor versus burnup for multiple deterministic and Monte-Carlo codes.
3.2. Radial Measurements of UGd Assembly

The 12 rods composed of oxide uranium mixed with oxide gadolinium are divided into multiple radial layers. The criticality and averaged atomic densities of one UGd rod are measured with respect to burnup steps and rod radius. The optimal neutron population is chosen equal to 50000 as a result of running different values of neutron source as shown in Figure 6. Serpent2 allows setting number of inactive cycles to allow the fission source to converge, they are set to 50. Inactive cycles are followed by 100 active cycles within which the values are calculated.
A shift in the criticality of the assembly is observed at the beginning of the fuel cycle after dividing the UGd rods into 2 layers. The values of $K_{inf}$ are higher at 1-4 MWt.d/kgU and lower at 4-9 MWt.d/kgU. At next burnup steps, no difference is observed as the criticality of radial layers variants (1,2,5,10,15 and 25 layers) are close to each other (Figure 7 and 8). The absolute difference between 1 layers and 2 layers is less than that between 1-5,1-10,1-15 and 1-25. The values after the 5th layer show higher degree of similarity.

Figure 7. $K_{inf}$ values versus burnup for different number of radial layers at 0-40 MWt.d/kgU.
Figure 8. $K_{inf}$ values versus burnup for different number of radial layers at 0-12 MWt.d/kgU.

In the other hand, only the atomic densities of gadolinium isotopes with respect to burnup varies when multiple number of layers is used. A delay in the change of atomic concentrations (decrease in Gd-155, Gd-157 and increase in Gd-156, Gd-158) happens at 4-7 MWt.d/kgU, Figure 9. Isotopes U-238 and Pu-239 overall composition is not affected, Figure 10.

Figure 9. Atomic density of Gd-157 versus burnup for multiple number of radial layers.
Figure 10. Atomic density of Pu-239 versus burnup for multiple number of radial layers.

When taking the atomic densities with respect to UGd rod radius a deep change is observed at the outer layers of the rod. Both U-238 and Pu-239 exhibit decrease and increase, respectively. This change happens in Rim region where the rod experience material defect due to long and heavy irradiation at high burnup values. This change is related to the high epithermal neutron absorption cross section of U-238 which decays producing Pu-239. For accurately detect the exact radius where the change starts, U-238 and Pu-239 at different burnup steps are measured. When the rod is divided into 5 radial layers the change starts at 0.34 cm, in the other hand, it starts at 0.36 cm with 10 radial layers (rod radius equal 0.386 cm, Figure 11 and Figure 12. It can be said that the concentrations are doubled at this region.

The results of burnup distribution through the rod radius is also affected by the radial division. At high burnup values (e.g. 40 MWT.d/kgU), as seen in Figure 13, the burnup is more than 50% higher at the outer rod layer when 10 radial layers are used. This is due to the high thermal neutron flux at the outer layers. The same measurements are done to the three-dimensional model of this assembly where the axial boundary condition is set to black (Total neutron absorption). The same effect on criticality and on atomic densities is measured, however, the values are lower, Figure 14.

Figure 11. Atomic density of U-238 versus radius at different burnup steps for UGd rods divided into; (A):5 layers, (B): 10 layers.
Figure 12. Atomic density of $u-238$ versus radius at different burnup steps for UGd rods divided into: (a): 5 layers, (b): 10 layers.

Figure 13. Burnup distribution through UGd rod radius at 40 MWt.d/kgU.
Figure 14. Comparison between the results of 2D and 3D burnup distribution with 5 radial layers.

3.3. Radial Measurements of UEr Assembly

Erbium has lower thermal neutron absorption cross section than that of gadolinium, thus it can’t build up the same criticality (hence, negative reactivity) as that of UGd assembly with the same number of rods. Therefore, more UEr are needed to get the same worth at the beginning of the fuel cycle. In order to do that, different rod numbers and different fuel loading patterns are examined, Figure 15. The pattern shown in Figure 15. c. is chosen, it has 34 UEr rods. Less number of UEr rods causes the reactivity to linearly drops, while when more UEr rods are added the reactivity increases slightly at the beginning and then drops, Figure 16. Nevertheless, after 5 MWd/kgU, more than needed negative reactivity is built in the assembly causing the overall reactivity to be less than that of gadolinium. This implies a decrease in the fuel cycle life. It is may be a result of using natural erbium in the fuel composition, as it consists big amount of isotope Er-166. Thus, fuel with 0%wt. Er-166 is used and the results are shown in Figure 17 among a comparison with UGd assembly’s reactivity.

Figure 15. Different fuel loading patterns for UEr assembly.
The values of multiplication factor are not affected by rod division as the self-shielding effect for erbium is low comparing to gadolinium. However, the change in atomic densities at the outer layers of UEr rods is similar to that of UEr. It is important to mention that Xe-135 exhibits more fluctuation in UGd rods that in UEr rods. In general, Erbium depletes slowly making the concentrations of other isotopes different than in UGd assembly, specially at the first burnup steps, such as the case shown in Figure 18 for Pu-239 generation.
Figure 18. Atomic concentration of Pu-239 in UEr rods using different number of radial layers.

4. Conclusion
Serpent 2 is used in the calculations of VVER-1000 assembly complex fuel composition and the results are compared with several Monte-Carlo and deterministic codes results using two data libraries ENDFb.6.8 and ENDFb.7. Serpent2 show good agreement with the benchmark average values, specially when ENDFb.6.8 is used as it is one of the libraries used in the benchmark.

The radial division of UGd rods help in getting more accurate results, accounting for the strong self-shielding of gadolinium and isotopes accumulation in the Rim-region. For criticality calculations, 5 radial layers are sufficient. However, 10 layers show different results than that when 5 layers are used when considering the isotopic radial generation. Using more number of layers accompanied with more time and memory consumption.

Erbium has lower thermal neutron absorption cross section, this indicates much lower self-shielding. Therefore, bigger number of UEr rods are required to reach the same effect on assembly’s reactivity. The radial division of UEr has no effect on the criticality calculations, however it helps in getting more accurate results for atomic densities as self-shielding effect has no effect in the development of Rim region.

Temperature radial distribution has effect on Rim region and isotopic generation, thus, this work can be extended into thermal-hydraulic study of rod radial division. Also, the concentration of Erbium isotopes has affect on the reactivity of the assembly, specially at the end of the fuel cycle. Therefore, this affect can be studied to minimize the reactivity penalty.

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References
[1] Galahom, A. Abdelghafar 2016 Investigation of Different Burnable Absorbers Effects on the Neutronic Characteristics of PWR Assembly Annals of Nuclear Energy 94 22–31 (Elsevier Ltd) (doi:10.1016/j.anucene.2016.02.025)
[2] Shaw, J 2013 Reactor Operation: Principles of Control and Operation 1st Editio (doi:10.1016/B978-0-08-013325-6.50005-5)
[3] Yahya, Mohd-syukri, Hwayneal Yu, and Yonghee Kim 2016 Burnable Absorber-Integrated Guide Thimble ( BigT ) – I: Design Concepts and Neutronic Characterization on the Fuel
Assembly Benchmarks Journal of Nuclear Science and Technology 53 (7) (Taylor & Francis: 1048–60. doi:10.1080/00223131.2015.1090937)

[4] Jo, Chang Keun, Yonghee Kim, and Jae Man Noh 2009 Burnable Poison for Reactivity Management in a Very High Temperature Reactor Annals of Nuclear Energy 36 (3) 298–304 (Elsevier Ltd) (doi:10.1016/j.anucene.2008.12.012)

[5] Subramanian, C Suri, A and Murthy T. 2010 Development of Boron-Based Materials for Nuclear Applications Technology Development Article 313 14–22.

[6] Fadaei, Amir Hosein 2011 Investigation of Burnable Poisons Effects in Reactor Core Design Annals of Nuclear Energy 38 (10) 2238–45 (Elsevier Ltd) (doi:10.1016/j.anucene.2011.06.005)

[7] Galahom, A 2017 Study of the Possibility of Using Europium and Pyrex Alloy as Burnable Absorber in PWR Annals of Nuclear Energy 110 1127–33 (Elsevier Ltd) (doi:10.1016/j.anucene.2017.08.052)

[8] Franceschini, Fausto 2004 Use of isotopically modified erbium to improve fuel cycle economics in iris 5th International Conference on Nuclear Option in Countries with Small and Medium Electricity Grids INAC 2017 pp. 1–12

[9] Franceschini, Fausto, and Bojan Petrović 2009 Fuel with Advanced Burnable Absorbers Design for the IRIS Reactor Core: Combined Erbia and IFBA Annals of Nuclear Energy 36 (8) 1201–7 (doi:10.1016/j.anucene.2009.04.005)

[10] Leppänen, Jaakko, Maria Pusa, Tuomas Vaittinen, Ville Valtavirta, Toni Kaltiaisenaho, and Monte Carlo 2015 The Serpent Monte Carlo Code: Status, Development and Applications in 2013 Annals of Nuclear Energy 82 142–50 (doi:10.1016/j.anucene.2014.08.024)

[11] Leppänen, Jaakko 2017 On the Use of Delta-Tracking and the Collision Flux Estimator in the Serpent 2 Monte Carlo Particle Transport Code Annals of Nuclear Energy 105 161–67 (doi:10.1016/j.anucene.2017.03.006)

[12] Science, Nuclear. 1964. A VVER-1000 LEU and MOX Assembly Computational Benchmark. Mexico Hungary Korea Nuclear Energy Agency Organisation For Economic Co-Operation And Development

[13] Saldikov I, Tikhomirov G, Ternovykh M, and Gerasimov A 2017 Computation Methods and Techniques for Solution of Coupled Multiphysics Problems in Precision Calculations of VVER VII International Conference on Coupled Problems in Science and Engineering

[14] Demin M, Abu Sondos M, and Smirnov A, 2018 The Comparative Analysis of Neutrons Properties of the Nuclear Fuel Produced by the Westinghouse and the TVEL for the Reactors VVER-1000 by Code SERPENT, KnE Eng., vol 3, no 3, p 21.