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# Model analysis of isotopic composition of VVER-440 spent nuclear fuel☆

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## Abstract

This paper presents an analysis of the isotopic composition of spent nuclear fuel under different operating conditions. Calculations were made for fuel assemblies of a VVER-440 reactor with second-generation Gd-2 fuel with enrichment of 4.25% $^{235}U$. The isotopic concentration was analyzed as a function of burnup of nineteen isotopes, selected on the basis of environmental risk, such as main fuel isotopes $^{235}U$, $^{236}U$, $^{238}U$, $^{239}Pu$, $^{240}Pu$, $^{241}Pu$, as well as some actinides, and fission products, such as $^{237}Np$ , $^{238}Pu$, $^{242}Pu$, $^{241}Am$, $^{242}Am$, $^{242}Cm$, $^{244}Cm$, $^{245}Cm$, $^{90}Sr$, $^{93}Zr$, $^{99}Tc$, $^{129}I$, $^{137}Cs$. The SCALE 6 computer code was used for model design and computer analysis.

## 1. Intoduction

Nuclear power entails the production of spent nuclear fuel, which requires skilled management with high safety standards. Successful management of spent fuel is crucial to the stable and sustainable growth of nuclear energy [1]. At the end of 2010, 440 nuclear power reactors (23 VVER-440 units) were operating in 30 countries plus Taiwan, with a combined capacity of over 376 GWe. In 2009 they provided 2,560 billion kWh, about 15% of the world's electricity [2].

In power reactors uranium is mostly used as nuclear fuel, from which other isotopes are produced. Uranium $^{238}U$ produces plutonium $^{239}Pu$ in the reactor through classical irradiation. This then creates other isotopes, usually highly toxic, through neutron capture. This process ultimately leads to the overcharging of the entire fuel cycle. About 70 tonnes of plutonium are accumulated worldwide in spent fuel each year [3]. Over 11,500 tonnes of heavy metals are unloaded from power reactors annually, accounting for the bulk of civilian radioactive material. This material needs to be managed appropriately [1]. Varying the neutron energy spectrum could impact the amount of toxic isotopes in spent nuclear fuel through different operating conditions.

Investigation of the isotopic composition of spent nuclear fuel has a key to play in solving the challenges associated with burnup credit, control and categorization of the amount of nuclear material and estimate the source conditions for radiation analyses. Changes in nuclear fuel composition during burnup can affect its reactivity, material and chemical properties, as well as the radioactive environment [4].

## 2. Relativity of isotopic compostition of spent fuel

The composition of nuclear fuel progresses during burnup and affects reactor reactivity, material and chemical properties. It impacts the radioactivity of the immediate environment, including structural and cladding elements. Additionally, spent fuel isotopic

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composition is the main factor determining the radiation field and major load under which storage and disposal of materials takes place. Therefore, calculation of the nuclear fuel isotopic composition is critical for highly specialized analyses of various fuel cycle options. Isotopic composition of reactor fuel depends on the spatial and spectral distribution of neutrons in the reactor core [5].

Isotopic composition of spent nuclear fuel depends not only on its burnup level, but is also dependent on the operating conditions of the reactor. Spent nuclear fuel with the same burnup value can have different isotopic composition, which is dependent on the neutron energy spectrum in which the burnup occurred. The differences in the spectrum are caused by the different operating conditions, such as the presence or absence of absorber-rods in an assembly, oscillating the concentration of boric acid, dissolved in the moderator, fuel and/or moderator temperature and tolerance in the initial fuel enrichment.

The investigation of spent fuel isotopic composition can be divided into two main phases, the assessment of fuel burnup and the determination of fuel isotopic composition depending on its burnup. The amount of $^{235}U$ in the spent fuel is influenced by the neutron spectrum. The higher the neutron energy spectrum, the more $^{238}U$ is engaged in the burnup process (mainly due to $^{239}Pu$ production) and the more $^{235}U$ is left in the spent fuel at the same burnup level [4].

## 3. Modeling VVER-440 fuel assembly

The SCALE 6 code was used for model design and computer analysis.

The calculations were made for the reactor cell of fuel assemblies and for second-generation VVER-440 fuel Gd-2 with enrichment of 4.25% $^{235}U$ (Fig. 1). Gadolinium, which is contained in fuel assemblies as $Gd_2O_3$, improves average burnup and reduces overall reactivity at the start of the fuel cycle. Calculated assembly is in infinite matrix conditions, as this is the case of the endless set of assemblies.

Fuel assembly parameters [6]:

- outer diameter of the fuel pellet is 7.60 mm,

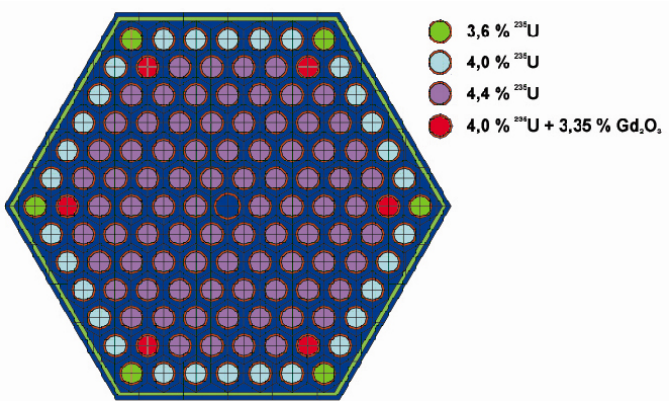


Figure 1: Composition of fuel pins in Gd-2 fuel assembly with 4.25% enrichment of $^{235}U$

- diameter of the central hole in the pellet is 1.2 mm,
- outer diameter of the fuel cladding is 9.07 mm,
- cladding thickness is 0.665 mm,
- grid distance is 250 mm,
- Hafnium mass limit in ZrNb is under 0.01%.

## 4. Results

We obtained the composition of spent nuclear fuel after making the VVER-440 model fuel assembly burnup calculation. We chose the total burnup of 50,000 MWd/tHM, which corresponds to a 5-year fuel assembly irradiation in the reactor core. Differences in isotopic composition are caused by the different operating conditions, which are dependent on the neutron spectrum in which the burnup took place. In the calculation, this represents a change in fuel temperature, moderator temperature, fuel enrichment and concentration of boric acid. For the initial calculation (AVG) or average operational conditions, we included model parameters corresponding to the Gd-2 fuel tabular values. For MIN and MAX cases we used the parameters (Tab. 1) based on available information about the fuel and moderator, where there are minimum (MIN) and maximum (MAX) values of enrichment, fuel temperature ($T_{fuel}$), moderator temperature ($T_{mod}$) and the concentration of boric acid ($H_3BO_3$).

The isotopic concentration was analyzed as a function of burnup of nineteen isotopes, selected on the

Table 1: The 3 operating conditions investigated

| | Enrichment $^{235}U$, % | $T_{fuel}$, K | $T_{mod}$, K | $H_3BO_3$, ppm |
|---|---|---|---|---|
| AVG | 4.25 | 976 | 558 | 450 |
| MIN | 4.20 | 767 | 543 | 0 |
| MAX | 4.30 | 1185 | 573 | 900 |

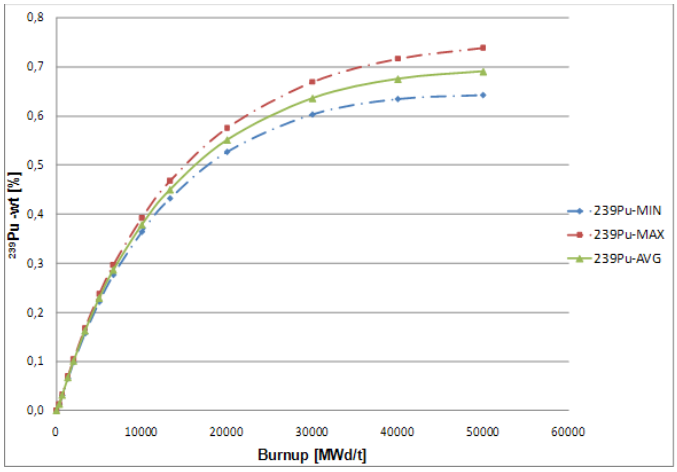


Figure 2: $^{239}Pu$ dependencies of burnup for all three operational conditions

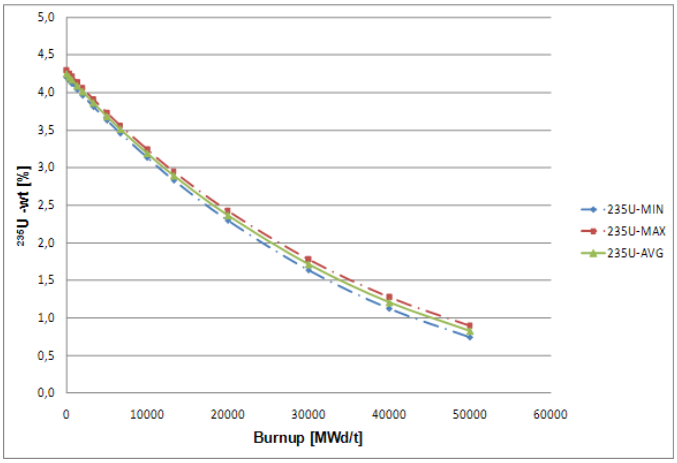


Figure 3: $^{235}U$ dependencies of burnup for all three operating conditions

basis of environmental risk, such as main fuel isotopes $^{235}U$, $^{236}U$, $^{238}U$, $^{239}Pu$, $^{240}Pu$, $^{241}Pu$, as well as some actinides, and fission products, such as $^{237}Np$ , $^{238}Pu$, $^{242}Pu$, $^{241}Am$, $^{242}Am$, $^{242}Cm$, $^{244}Cm$, $^{245}Cm$, $^{90}Sr$, $^{93}Zr$, $^{99}Tc$, $^{129}I$, $^{137}Cs$.

The neutron energy spectrum in the fuel assembly influences the change in the concentration, by way of example plutonium $^{239}Pu$ isotope is shown in Fig. 2. During burnup in 126 fuel pins of standard VVER-440 the concentration gradually increases to around 0.7 wt of $^{239}Pu$.

The results of different isotopic composition of spent fuel for modified operational parameters towards base state are processed in Tab. 2. Differences between the maximum and minimum operational conditions are very small for most of the isotopes. Higher differences than the average are observed only for $^{235}U$, $^{239}Pu$ and $^{241}Am$ isotopes.

## 5. Conclusion

Operating conditions can affect changes in the neutron energy spectrum during burnup, along with changes in the isotopic composition of the spent nuclear fuel. For second-generation VVER 440 nuclear fuel with burnable absorbers and 4.25% $^{235}U$ enrichment there are differences in fuel temperature, moderator temperature, fuel enrichment and concentration of boric acid.

The results from the SCALE 6 code show differences in the isotopic composition of spent nuclear fuel for different operating conditions. It is possible to give differences in percentage terms for the concentration of investigated isotopes in spent nuclear fuel in respect of the AVG, MAX or MIN case. The results show that the isotopes most affected by operating conditions are $^{235}U$ ± 9%, 7% ± $^{239}Pu$ and $^{241}Am$ ± 9.5%. On the other side, operational conditions have a minimal or negligible impact on fission products and $^{236}U$, $^{242}Pu$ and $^{244}Cm$ isotopes.

## Acknowledgement

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Table 2: Percentage differences for MIN and MAX cases against the average isotopic concentration (AVG)

| | $\delta_{MAX}$, % | $\delta_{MIN}$, % | | $\delta_{MAX}$, % | $\delta_{MIN}$, % |
|---|---|---|---|---|---|
| $^{235}U$ | 8.90 | -9.44 | $^{241}Am$ | 9.41 | -8.62 |
| $^{236}U$ | 0.46 | -0.37 | $^{242}Am$ | 1.16 | -1.83 |
| $^{238}U$ | -0.15 | 0.15 | $^{242}Cm$ | 2.66 | -3.84 |
| $^{237}Np$ | 3.63 | -3.97 | $^{244}Cm$ | 1.35 | -2.12 |
| $^{238}Pu$ | 3.38 | -4.27 | $^{245}Cm$ | 5.78 | -7.10 |
| $^{239}Pu$ | 7.04 | -7.05 | $^{90}Sr$ | -0.50 | 0.58 |
| $^{240}Pu$ | 1.33 | -1.44 | $^{93}Zr$ | -0.39 | 0.39 |
| $^{241}Pu$ | 6.14 | -6.55 | $^{99}Tc$ | -0.09 | 0.18 |
| $^{242}Pu$ | -0.83 | 0.67 | $^{129}I$ | 0.43 | -0.48 |
| | | | $^{137}Cs$ | -0.11 | 0.17 |

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