# EXTENDED VALIDATION OF ENGINEERING MODELS FOR EXPRESS-METHOD OF BURNUP EVALUATION OF WWER-1000 FUEL ELEMENTS

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## ABSTRACT

This work continues a series of papers [1-4] on the development of simplified physical models for cesium isotopes buildup in function of fuel burnup and evolution of $^{134}Cs/^{137}Cs$ activity ratio for different fuel assembly (FA) designs. Implementation of these models takes into account operational features for fuel in elongated core cycles at an uprated power level of WWER-1000 power units. This paper presents validation of model calculation results for $^{134}Cs/^{137}Cs$ activity ratios in fuel of four TVSA-PLUS assemblies (with solid $UO_2$-fuel pellets) and one UTVS assembly. Calculated $^{134}Cs/^{137}Cs$ activity ratios are compared against the available NPPs data on spike-events and results of leaking fuel testing for these FAs in failed fuel detection system (FFDS) casks. Validation of the model calculation results against the available experimental data showed that the developed models enable to perform reasonable evaluations of $^{134}Cs/^{137}Cs$ activity ratios in fuel in advanced FA designs.

## 1. Introduction

Radiation safety assurance procedures at operating WWER-1000 power units (PUs) include monitoring of activities of the reference radionuclides in the primary coolant (PC). Preliminary fuel burnup evaluation in case of the presence of a leaking fuel assembly (FA) while reactor operation is of high importance at NPPs. First, this would decrease time expenditures for its identification in the core. Second, it would reduce time for finding a proper substitute in spent fuel storage pools at the course of scheduled maintenance outage at PUs. Nowadays, reliable identification of leaking FAs of advanced designs requires taking into account operational features of enhanced core cycles at WWER-1000 PUs at 104%$N_{nom}$ (thermal output power level), with a perspective power uprating up to 107-110%$N_{nom}$, [1].

In case of fuel operational failures while PU operation with radioactive fission products release into the PC, the most sensitive indicator of failed fuel burnup at WWER-1000 reactors is $^{134}Cs/^{137}Cs$ activity ratio measurements at spike-events that are registered at an intermediate power decrease or at reactor shutdown, [1, 4, 5]. Accumulation of $^{137}Cs$ and $^{133}Cs$ in fuel is practically linear with fuel burnup. $^{134}Cs$ buildup is predominantly governed by the neutron capture reactions in stable $^{133}Cs$ in the resonance energy region.

Evaluation of leaking FAs burnup relied on the calculated dependence of $^{134}Cs/^{137}Cs$ activity ratio in function of fuel burnup for the typical WWER-1000 irradiation history from the Guidance Document for NPPs. However, the results of some recent spike-events analysis using that dependence showed some discrepancy, discussed, for instance, in [1]. Currently, achievement of longer fuel cycles at a higher FAs' linear heat generation rates (LHGRs) leads to an upgraded initial fuel enrichment and the use of burnable neutron absorbers (Gd-doped fuel rods). These factors entail hardening of the neutron flux spectrum that affects the effective neutron capture cross-section in $^{133}Cs$ converting it into $^{134}Cs$. Thus, the series of papers [1-4] focused on the conditions of the fuel operation modern fuel cycles, and the models for a fast evaluation of $^{134}Cs/^{137}Cs$ activity ratio in function of fuel burnup were developed to improve the existent evaluation techniques at NPPs in fuel while PU operation.

## 2. Basic concepts of the models

Developed simplified models in the presented code are implemented for fast cesium inventory evaluations in fuel, and are described in detail $UO_2$ rods [1,2] and Gd-doped fuel rods [3]. The models introduce the approach that provides an opportunity to evaluate cesium isotopes buildup in fuel taking into account the initial hardness of the neutron spectrum ($\gamma$) and its changes during the cycle using the given LHGR evolution (neutronic calculations for the loading pattern) of a Gd-doped fuel rod and the surrounding fuel rods in the FA. In our simplified models, parameter $\gamma$ describes the ratio of epithermal/thermal energy neutrons interactions with fissionable nuclides. Evolution in contributions of epithermal and thermal energy neutrons into total LHGR of an FA during the cycle lead to the changes of $^{134}Cs/^{137}Cs$ activity ratio in fuel, [1].
At the beginning of a fuel cycle, thermal neutrons are considerably captured by present $^{10}B$ and $^{155,157}Gd$ isotopes with high capture cross-sections, so the contribution of thermal neutron flux into the total power of an FA changes in time. Intermediate neutron absorption by $^{235}U$ is less affected, thus the flux of intermediate neutrons $\Phi_{ET}(t)$ changes inconsiderably in time taking into account possible minor changes in LHGRs from a sustained constant value.
Model for the $UO_2$ rods is shortly provided below. Assuming that the relation between FA's LHGR and epithermal neutron flux $\Phi_{ET}(t)$ varies inconsiderably during the cycle defined by (1), and taking into account calculated initial ($t = 0$) thermal neutron flux $\Phi_{T_0}$ and $\gamma_0$, the value of $\gamma(t)$ is estimated at each time step $(j)$ over the cycle time by relation (1):

$$\gamma(t) = \frac{\Phi_{ET}(t)}{\Phi_T(t)}, \quad \frac{\gamma_0 \Phi_{T_0}}{LP_{fr}^{t=0}} = \frac{\gamma_j \Phi_{T_j}}{LP_{fr}^{j}}, \tag{1}$$

The burnout rate of fissile isotopes and poison isotope buildup in the models are based on the proportionality of the local power density, $q_V(r,t)$, to the thermal neutron flux, $\Phi_T(t)$, to the concentrations of the relevant isotopes, $n_j(r,t)$, and to the corresponding two-group fission cross-sections, $\sigma_f^k(t)$, that are averaged over the neutron spectrum in epithermal and thermal groups:

$$q_V(r,t) \propto \sum_k E_k \sigma_f^k(t) n_k(r,t) \Phi_T(t), \tag{2}$$

To derive local concentrations of U and Pu isotopes, we consider the following set of equations:

$$\begin{aligned}
\frac{dn_{235}(t)}{dt} &= -\sigma_a^{235}(t) n_{235}(t) \Phi_T(t) \\
\frac{dn_{238}(r,t)}{dt} &= -\sigma_a^{238} \bar{n}_{238}(r) f(r) \Phi_T(t) - k_{^{238}U(n,f)} \Phi_T(t) \sum_k \sigma_f^k(t) n_k(r,t) \\
\frac{dn_{239}(r,t)}{dt} &= -\sigma_a^{239}(r,t) n_{239}(r,t) \Phi_T(t) + \sigma_c^{238}(t) \bar{n}_{238}(t) f(r) \Phi_T(t) \\
\frac{dn_j(r,t)}{dt} &= -\sigma_a^j(t) n_j(r,t) \Phi_T(t) + \sigma_c^{j-1}(t) n_{i-1}(r,t) \Phi_T(t), \; j = 240 \div 242
\end{aligned} \tag{3}$$

Here $n_j(r,t)$ is the local concentration of the isotope j (j = 235, 238-242) with the respect to the moment of calculation time (t), $\bar{n}_{238}(t)$ is the averaged concentration of $^{238}U$, $\sigma_a^k(t)$ and $\sigma_f^k(t)$ are the two-group effective cross-sections in the used approximation [1] for total neutron absorption and fission, k = 235, 239, 241, and $\sigma_c^{238}(t)$ is for $^{238}U$ neutron capture, respectively. For a given nuclide $j$, total effective neutron cross-section is evaluated describing the neutron spectrum as a combination of a Maxwell-Boltzmann distribution function which is characterized by effective neutron temperature $T_n$, and a component of epithermal energy neutrons, which neutron flux distribution is proportional to the reciprocal of the neutron energy – Westcott $g$-factor, [1]. Therefore, thermal neutron flux $\Phi_T(t)$ is calculated as:

$$\Phi_T(t) = \frac{\left(1 - k_{^{238}U(n,f)}\right) \cdot LP^{fr}(t)}{2\pi \int_{R_{in}}^{R_{out}} \left( \sum_k \sigma_f^k(t) n_k(r,t) E_f^k \right) r dr}, \tag{4}$$

here $R_{in}$ – centerline hole radius, $R_{out}$ – radius of a fuel pellet, $\sigma_f^k(t)$ – fission cross-section of nuclide, $n_k(r,t)$– current concentration of fissionable nuclide, $E_f^k$ = 200 MeV ($k$ = 235, 239, 241) is the energy release per heavy atom fission. This estimation is performed with regard to the evolution of primary fissionable nuclides ($^{235}$U, $^{239,241}$Pu) in fuel.

The radial form function $f(r)$ takes into account the strong resonance neutron absorption in $^{238}$U that leads to the formation of $^{239}$Pu. This distribution function can be interpreted as the combination of a constant production of $^{239}$Pu from thermal neutron capture plus a highly nonlinear term for the production due to resonance absorption, discussed in [1]. Presented engineering models implement the approach for a normalized radial form function $f(r)$ of neutron absorption for rod geometry (7). In this approach, parametrical dependencies for the $f(r)$ are derived from the ratio of $^{238}$U absorption cross-sections: the fracture of full cross-section which is not self-shielded $\sigma_{a,238}^{nss}$, and full cross-section $\sigma_{a,238}$. The value of effective resonance integral $I_{238}^{eff}$ is derived from experimental data and dependencies obtained for $UO_2$-fuel, [1]. Therefore,

$$f(r) = p + (1-p)F(r)\,,\ p = \sigma_{a,238}^{nss} / \sigma_{a,238}\,,\ F(r) = \mathrm{C}_F \frac{\phi(z)}{\sqrt{1-z^2}}\,,\ z = r / R_{out}\,,$$
$$\phi(z) = \frac{1}{\pi}\int_0^{\pi/2}\left[\left(\sqrt{1-z^2\sin^2\varphi} + z\cos\varphi\right)^{1/2} + \left(\sqrt{1-z^2\sin^2\varphi} - z\cos\varphi\right)^{1/2}\right]\mathrm{d}\varphi\,,\ \sin\varphi_0 = z, 0 < \varphi < \varphi_0\,. \quad (5)$$

Normalization constant $C_F$ is derived from the relation:

$$\frac{2}{R_{out}^2 - R_{in}^2}\int_{R_{in}}^{R_{out}} F(r)rdr = 1 \quad (6)$$

Calculation results obtained with and without the radial distribution models would not show considerable differences as the condition (7) is set for the $f(r)$:

$$\frac{2}{R_{out}^2 - R_{in}^2}\int_{R_{in}}^{R_{out}} f(r)rdr = 1 \quad (7)$$

The model includes the following equations to describe the time rate of change in concentrations of cesium isotopes ($^{133,134,137}$Cs) in fuel:

$$\begin{aligned}
&\dot{n}_{133}(t) = y_{133}^{eff}(t)\dot{F}(t) - \sigma_{133}(t)\Phi_T(t)n_{133}(t)\\
&\dot{n}_{134}(t) = \sigma_{133}(t)\Phi_T(t)n_{133}(t) - \sigma_{134}(t)\Phi_T(t)n_{134}(t) - \lambda_{134}n_{134}(t)\,,\\
&\dot{n}_{137}(t) = y_{137}^{eff}(t)\dot{F}(t) - \lambda_{137}n_{137}(t)
\end{aligned} \quad (8)$$

here $n_{133,134,137}(t)$ – atom density of Cs nuclide [m$^{-3}$], $\lambda_{133,134,137}$ – radioactive disintegration constant of nuclide [s$^{-1}$], $\sigma_{133,134,137}$ – full neutron absorption cross-section of nuclide [10$^{-28}$ m] of the corresponding cesium nuclides, $\Phi_T(t)$ – averaged thermal neutron flux [m$^{-2}$s$^{-1}$], $\dot{F}(t)$ – total fission rate of primary fissionable nuclides ($^{235}$U, $^{239,241}$Pu) in fuel volume [m$^{-3}$s$^{-1}$]. Effective fission yield $y_i^{eff}(t)$ for $^{133,134,137}$Cs isotopes (i = 133, 134,137) per fission of heavy atoms is described by the following expression with the respect to the evolution of their concentrations:

$$y_i^{eff}(t) = \frac{y_i^{235}n_{235}(t)\sigma_f^{235} + y_i^{239}n_{239}(t)\sigma_f^{239} + y_i^{241}n_{239}(t)\sigma_f^{241}}{n_{235}(t)\sigma_f^{235} + n_{239}(t)\sigma_f^{239} + n_{241}(t)\sigma_f^{241}} \quad (9)$$

Therefore, changes of $^{134}$Cs/$^{137}$Cs activity ratio calculations in $UO_2$-fuel using the presented models enable to take into account the features of FAs operation in every particular core cycle.

## 3. Method of the models' validation

Validation of the models for fast evaluation of fuel burnup in failed FAs [1-4] in case of a cladding leakage relied on the data provided by several NPPs with WWER-1000 power units (reactor installation V-320, [1]). The data included information about the reactor core power history, parameters of the coolant purification system, activities of the reference radionuclides in the primary coolant circuit, and average LHGRs of the FAs obtained with neutronic calculations.

At the end of each fuel cycle under consideration (period of 2013-2018), sipping leakage tests in the mast of the refueling machine was performed for all the FAs in the core. Leaking fuel testing in the failed fuel detection system (FFDS) casks was performed for the suspect FAs in the core, and the activities of the reference isotopes in the water samples from the cask circuit were measured. Thus, the leaking assemblies were identified and their fuel burnup was reliably determined. Results of the FAs' testing in the FFDS casks after the reactor shutdown (hereinafter, FFDS results) confirmed that all the leaking assemblies considered in this paper were the only ones in the core in each corresponding fuel cycle (see Table 1).

| # | FA type | Initial FA average fuel enrichment in $^{235}U$, wt.% | Number of $UO_2$/ (U, Gd)$O_2$ fuel rods per FA | Irradiation period, fuel cycles | Calculated FA average fuel burnup (NPP's data), MWd/kgU |
|---|---|---|---|---|---|
| 1 | TVSA -PLUS | 4.34 | 288/24 | 1 | 21.90 |
| 2 | TVSA -PLUS | 4.62 | 288/24 | 2 | 43.03 |
| 3 | TVSA -PLUS | 2.40 | 312/0 | 1 | 13.37 |
| 4 | TVSA -12PLUS | 4.92 | 306/6 | 1 | 22.25 |
| 5 | UTVS | 4.00 | 311/0 | 2 | 25.86 |

Tab 1: Parameters of the fuel assemblies, whose NPP data were used for model calculations validation in the present paper

The data on the registered spike-events (provided by the NPPs) were processed using the Expert System (ES) developed in SRC RF "TRINITI", [1, 5], in particular, the data on the reference isotopes activity measurements in the PC. Values of the specific activities of iodine, noble gases, activated coolant and corrosion products, and cesium in the PC were measured with an allowance of 8÷10 % at the PUs. NPPs data for the considered fuel cycles are shown in Figures 1, 4-5, 10: when the spike-event occurs, activities of cesium isotopes are evaluated subtracting the background activity level (highlighted with orange), and then the values of $^{134}Cs/^{137}Cs$ activity ratios are processed in time (typically, for 1-2 days, highlighted with red). Therefore, $^{134}Cs/^{137}Cs$ activity ratio "intervals" in Figures represent minimum and maximum ratio values from the registered data during the spike-event.

## 4. Validation of the model calculation results for $^{134}Cs/^{137}Cs$ activity ratio in fuel

Isotope inventory calculations with the developed code in this work were limited to the fast simulation of cesium isotopes buildup and $^{134}Cs/^{137}Cs$ activity ratio in fuel that is used in the standard technique for the evaluation of fuel burnup at NPPs. Obtained calculation results were then compared to the registered $^{134}Cs/^{137}Cs$ activity ratio at spike-events.
Detailed activity analyses in the PC and activity release evaluations in our department are performed using the developed RTOP-CA fuel performance code, [2]. This code is capable of simulations of time-dependent activity release calculations in normal operation modes and the spike-events modeling. Simulation of the radioactive fission products' (RFPs) release into the PC through the rupture opening includes several stages: transport to the fuel grain boundaries, their behavior in the intergrain porosity, and release under the rod cladding. RFPs' behavior in the PC is simulated taking into account nuclear decays, coolant cleanup rates, and penetration of the coolant under the cladding of a failed rod that leads to the change in fuel physical characteristics and conductivity of the pellet-cladding gap. Effect of tramp uranium in the core is also evaluated using this code. Validation of the RTOP-CA models was performed against the results of the carried out reactor experiments (e.g., at MIR research reactor in SSC RIAR), and compared to the activity measurements at NPPs with WWER power units.

### 4.1. Operational parameters of TVSA- PLUS #1 and model calculation results

In TVSA-PLUS #1, the initial enrichment of fuel was 4.4 wt.% in $^{235}U$ in $UO_2$-fuel rods, and 3.6 wt.% in Gd-doped fuel rods (initial content of $Gd_2O_3$ - 8.0 wt.%). TVSA-PLUS #1 operated for one 18-month fuel cycle, its average LHGR ranged between 20-25 kW/m. Calculated FA's average fuel burnup (NPP's data) after the discharge was 21.90 MWd/kgU.
After an intermediate power drop, spiking of cesium activities in the PC was registered; NPP's data are presented in Figure 1.

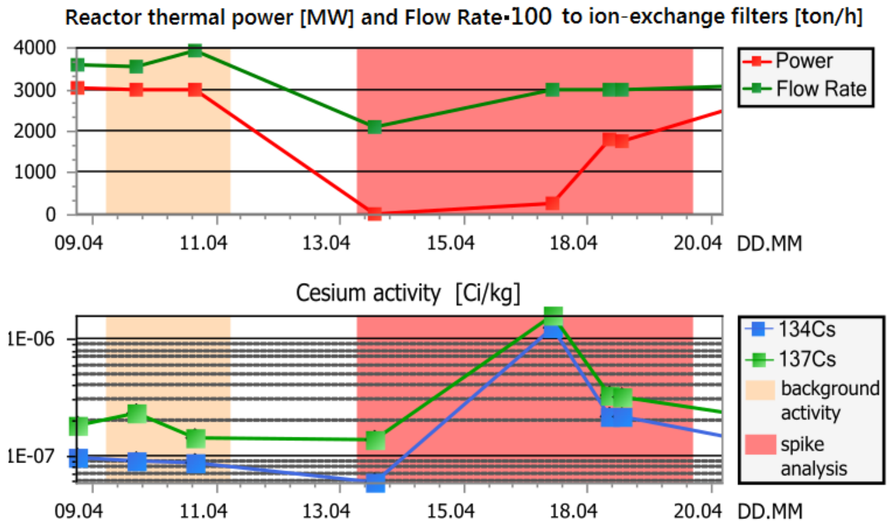


Fig. 1. NPP's data for an intermediate spike-event

Comparison of the model calculation results for the $^{134}Cs/^{137}Cs$ activity ratio in fuel with the handled spike-event data and the FFDS result for TVSA-PLUS #1 is shown in Figure 2.

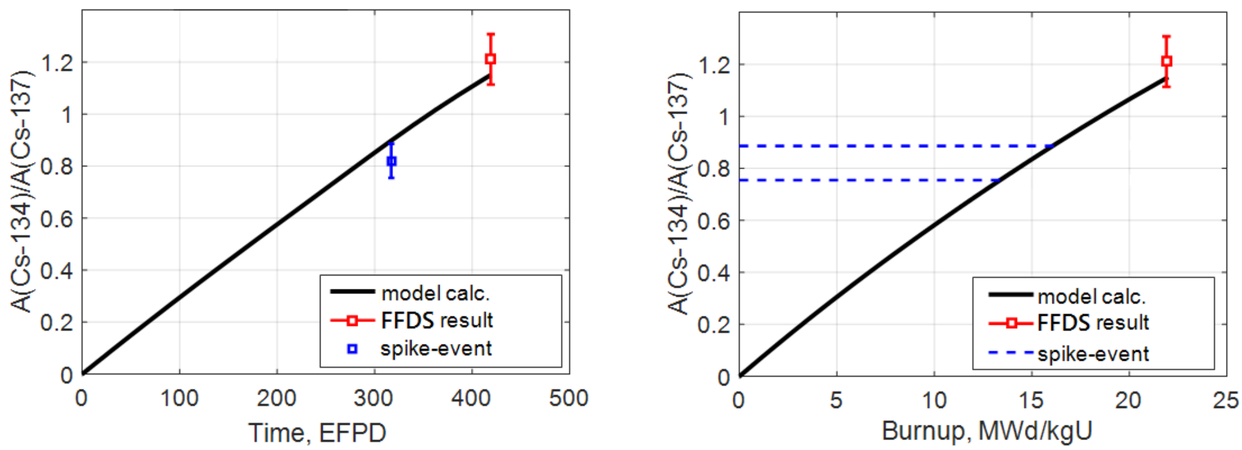


Fig. 2. $^{134}Cs/^{137}Cs$ activity ratio in function of time (left) and fuel burnup (right) for TVSA-PLUS #1

In Figure 2, the registered $^{134}Cs/^{137}Cs$ activity ratio in the PC was 0.82 for the intermediate spike-event that occurred after 317 days after the beginning of the fuel cycle. After the reactor shutdown, TVSA-PLUS #1 was tested in FFDS cask, measured $^{134}Cs/^{137}Cs$ activity ratio in the water sample was 1.21. It can be seen that the model calculation results of cesium activity ratio for TVSA-PLUS #1 are in good agreement with the provided NPP's data.

### 4.2. Operational parameters of TVSA- PLUS #2 and model calculation results

In TVSA-PLUS #2, the initial enrichment of fuel was 4.7 wt.% in $^{235}U$ in $UO_2$-fuel rods, and 3.6 wt.% in Gd-doped fuel rods (initial content of $Gd_2O_3$ - 8.0 wt.%). TVSA-PLUS #2 operated for two 18-month fuel cycles. Average LHGR of TVSA-PLUS #2 is shown in Figure 3. Calculated FA's average fuel burnup (NPP's data) after the discharge was 43.03 MWd/kgU.

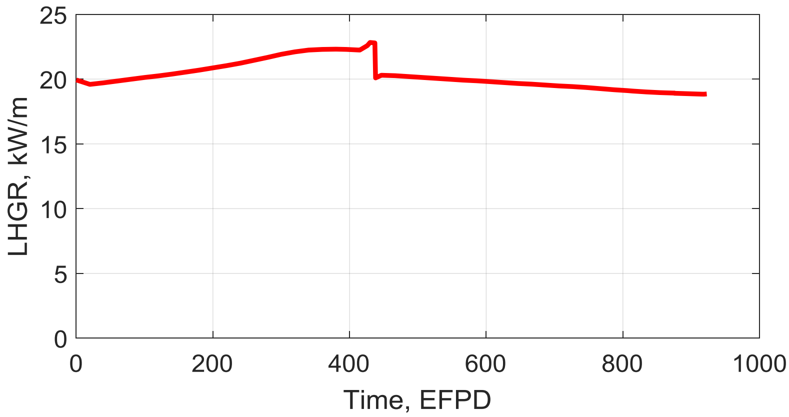


Fig. 3. Average LHGR of TVSA-PLUS #2 for two fuel cycles

At the course of the second cycle of the TVSA-PLUS #2, two spike-events for cesium activities in the PC were registered: after an intermediate power drop at the PU (1) and after the reactor shutdown (2), NPP's data are presented in Figures 4-5.

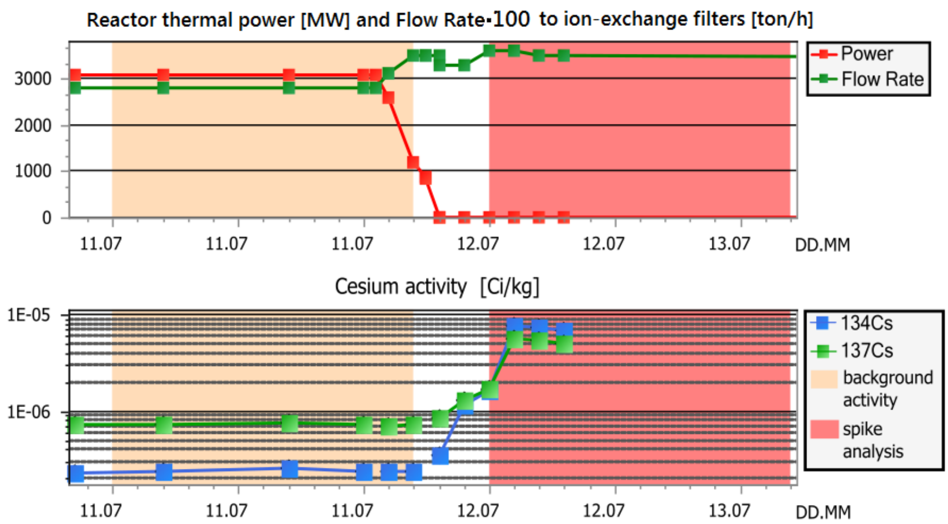


Fig. 4. NPP's data for the intermediate spike-event (1)

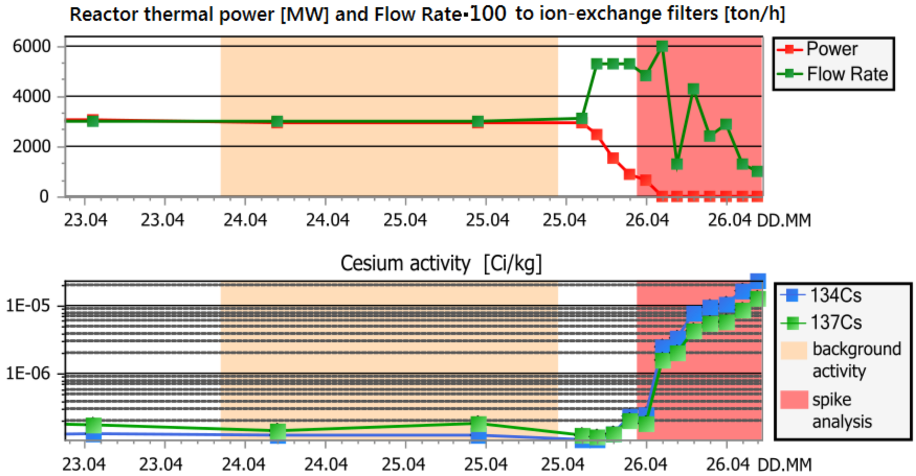


Fig. 5. NPP's data for the spike-event (2) after the reactor shutdown

Registered $^{134}Cs/^{137}Cs$ activity ratio in the PC was in the rage of 1.52÷1.55 for the intermediate spike-event, and 1.69÷1.86 for the spike-event after the reactor shutdown. Comparison of the model calculation results for the $^{134}Cs/^{137}Cs$ activity ratio in fuel with the spike-event data and the FFDS result for TVSA-PLUS #2 is shown in Figure 6. It can be seen that the model calculation results of cesium activity ratio for TVSA PLUS #2 are in good agreement with the provided NPP's data.

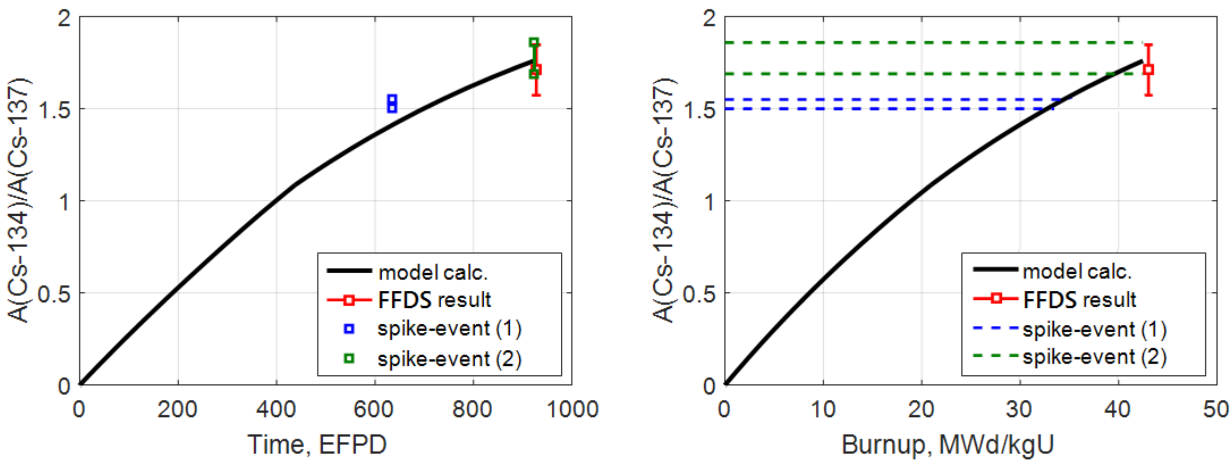


Fig. 6. $^{134}Cs/^{137}Cs$ activity ratio in function of time (left) and fuel burnup (right) for TVSA-PLUS #2

### 4.3. Operational parameters of TVSA- PLUS #3 and model calculation results

In TVSA-PLUS #3, the initial enrichment of fuel was 2.4 wt.% in $^{235}U$. TVSA-PLUS #3 operated for one 12-month fuel cycle, its average LHGR ranged between 18-22 kW/m in the course of the cycle. Calculated FA's average fuel burnup (NPP's data) after the discharge was 13.37 MWd/kgU.
Good agreement of the model calculation results for the $^{134}Cs/^{137}Cs$ activity ratio in fuel with the FFDS result for TVSA-PLUS #3 is observed in Figure 7.

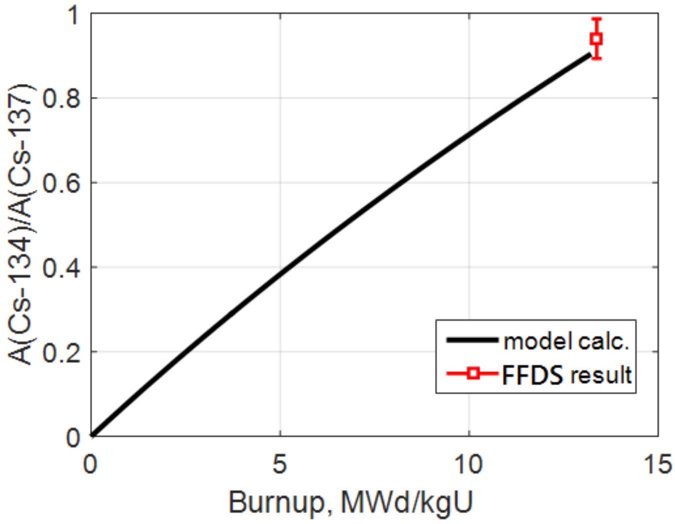


Fig. 7. $^{134}Cs/^{137}Cs$ activity ratio in function of fuel burnup for TVSA-PLUS #3

### 4.4. Operational parameters of TVSA- 12PLUS #4 and model calculation results

In TVSA-12PLUS #4, the initial enrichment of fuel was 4.95 wt.% in $^{235}U$ in $UO_2$-fuel rods, and 3.6 wt.% in Gd-doped fuel rods (initial content of $Gd_2O_3$ - 5.0 wt.%). TVSA-12PLUS #4 operated for one 18-month fuel cycle, its average LHGR ranged between 20-22 kW/m in the course of the cycle. Calculated FA's average fuel burnup (NPP's data) after the discharge was 22.25 MWd/kgU.
Good agreement of the model calculation results for the $^{134}Cs/^{137}Cs$ activity ratio in fuel with the FFDS result for TVSA-PLUS #3 is observed in Figure 8.

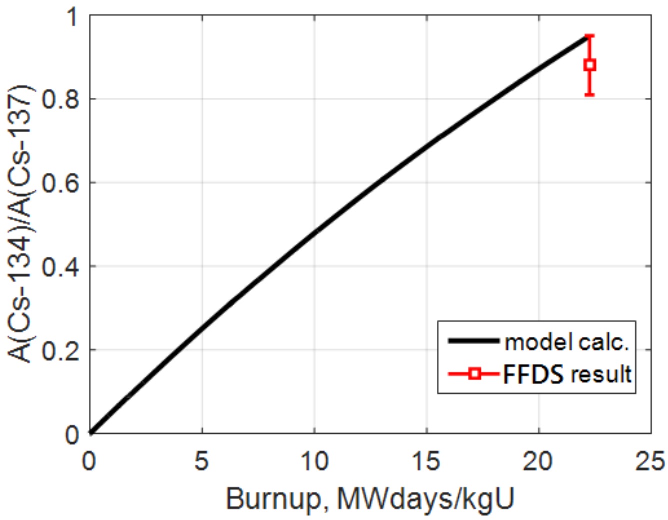


Fig. 8. $^{134}Cs/^{137}Cs$ activity ratio in function of fuel burnup for TVSA-12PLUS #4

### 4.5. Operational parameters of UTVS #5 and model calculation results

In UTVS #5, the initial enrichment of fuel was 4.1 wt.% in $^{235}U$ in 245 $UO_2$-fuel rods, and 3.7 wt.% in 66 peripheral $UO_2$-fuel rods. UTVS #5 operated for two 12-month fuel cycles. Average LHGR of UTVS #5 is shown in Figure 9. Calculated FA's average fuel burnup (NPP's data) after the discharge was 25.86 MWd/kgU.

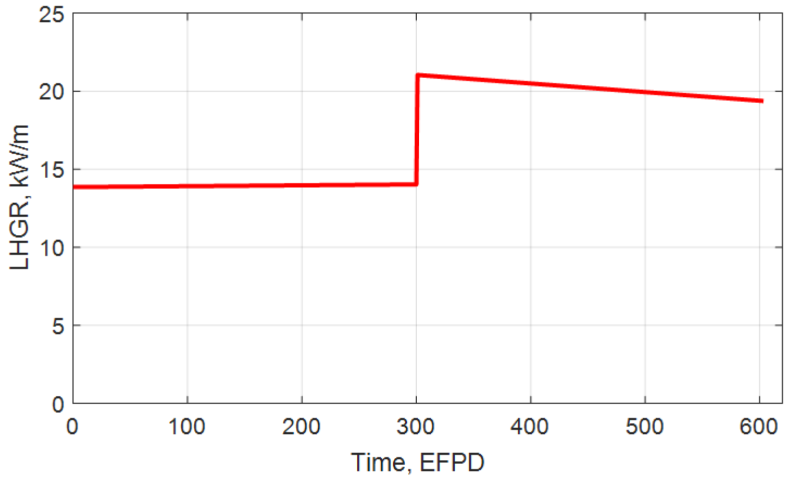


Fig. 9. Average LHGR of UTVS #5

After the reactor shutdown (second fuel cycle), a spike-event for cesium activities in the PC was registered, NPP's data are presented in Figure 10.

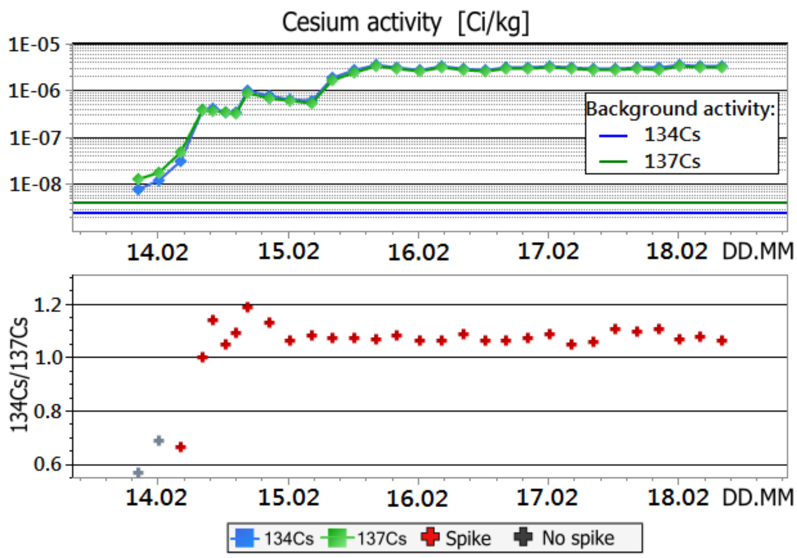


Fig. 10. NPP's data on cesium activities in PC for the spike-event after the reactor shutdown

Registered $^{134}Cs/^{137}Cs$ activity ratio in the PC was in the rage of 1.00÷1.19 for the spike-event after the reactor shutdown. Comparison of the model calculation results for the $^{134}Cs/^{137}Cs$ activity ratio in fuel with the spike-event data and the FFDS result for UTVS #5 is shown in Figure 11. The calculated dependence of $^{134}Cs/^{137}Cs$ activity ratio in function of fuel burnup for the typical WWER-1000 irradiation history from the Guidance Document for NPPs (RD 2004) is also shown alongside the results. It can be inferred that the RD dependence shows a good agreement for the older FA type without Gd-doped fuel rods.

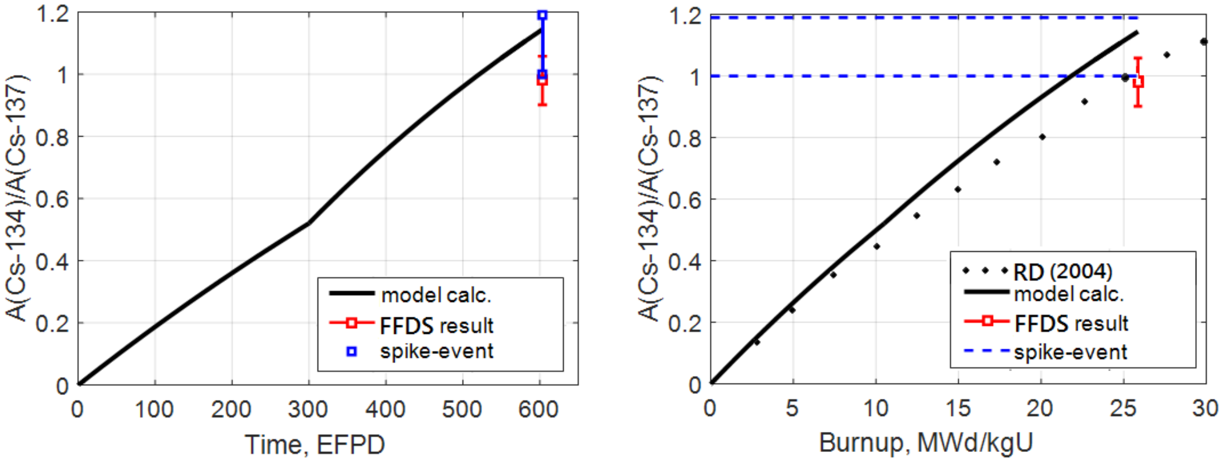


Fig. 11. $^{134}Cs/^{137}Cs$ activity ratio in function of time (left) and fuel burnup (right) for UTVS #5

## 5. Conclusions

1. Comparison of the NPPs data on the $^{134}Cs/^{137}Cs$ activity ratio in PC of WWER-1000 PUs and the FFDS water samples for the leaky FAs with the results of the models' calculations leads to the conclusion that the developed approach allows determining the burnup in a failed FA with a satisfactory accuracy.
2. In case of registered spike-events of cesium activities in the course of reactor operation, implementation of the developed models allows for an increased reliability of the technique for the fuel burnup evaluation in a leaky FA while performing cladding integrity monitoring.

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