ON BURNUP MODELLING ISSUES ASSOCIATED WITH VVER–440 FUEL

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Abstract: The paper investigates various computational modelling issues associated with VVER-440 fuel depletion, relevant to burnup credit. The well-known SCALE system and the TRITON sequence are used for the calculations. The effects of variations in depletion parameters and used calculation methods on the isotopic vectors are investigated. The burnup behaviour of Gadolinium, a burnable poison in nuclear fuel, is quite important in actual core analysis, but its behaviour is somewhat complicated, requiring special treatment in numerical modelling and calculations. Therefore, a special part of the paper is devoted to the treatment of Gadolinium-bearing fuels. Moreover, some discussions on power normalization, which plays an inevitable role in burnup calculations, are also included. The analyses in the paper include determination and ranking of the most important actinides and fission products and while emphasis is put on the fuel temperature distribution and its influence on the final isotopic vector of depleted fuel. To assess the acquired modelling experience used to predict the VVER-440 spent fuel nuclide composition, the measured compositions of Novovoronezh NPP irradiated fuel assembly are compared to data calculated by TRITON sequence. The samples of fuel assembly with 3.6 wt. % U-235 enrichment underwent 4-cycle campaign of totally 1109 days in the core and cooling period of 1-13 years. Calculated concentrations are compared to measured values burdened with their experimental uncertainties for totally 47 nuclides. The calculated results show overall a good agreement for all nuclides, differences from measured are pointed out and discussed in the paper.

Keywords: burnup, VVER 440, modelling, SCALE

1. INTRODUCTION

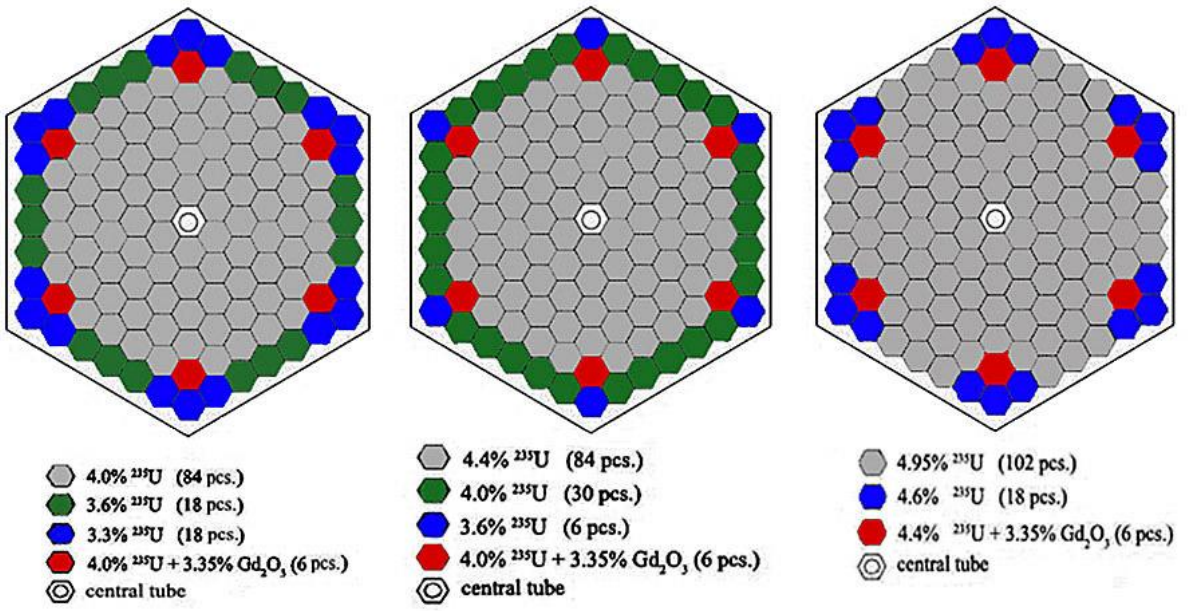
The prediction accuracy of burnup calculations is a critical factor in the reactor analysis sequence. The core properties depend on the actual composition of the fuel; thus, the characteristics of the reactor core undergo changes during burnup. Moreover, the isotopic composition of the spent fuel discharged from the core is a key factor in both the operations and the material control activities of the deep geological repository. An accurate estimate of the time-dependent radionuclide inventory in this material is necessary to evaluate many spent fuel issues, including neutron and gamma-ray source terms for shielding analysis, decay-heat source terms for temperature distribution and radiological and chemical toxicity for environmental impact consideration. Slovakia has four nuclear reactors generating half of its electricity and another two under construction. This paper investigates and summarizes modelling issues associated with VVER-440 fuel depletion performed by the SCALE system [1]. The effects of variations in the depletion parameters and used calculation methods on the isotopic vectors are investigated. The burnup behaviour of Gadolinium, a burnable poison in nuclear fuel, is quite

important in actual core analysis; therefore, a special part of the paper is devoted to this issue. Finally, some discussions on power normalization are also included. Other publications relevant to burnup modelling issues for PWR can be found in [2,3]. To assess the ability of the SCALE system and the associated nuclear data to predict the VVER-440 spent fuel nuclide composition, the measured compositions of Novovoronezh NPP irradiated fuel assembly are compared to data calculated by the TRITON sequence in the last part of the paper. The work done follows the simplified specification of the computational benchmark based on the #2670 ISTC project [4] providing VVER-440 data for 8 samples cut outs of 4 fuel pins of the fuel assembly (FA) No. D26135 with the average reached burnup 38.5 MWd/kgU. The #2670 ISTC project has was carried out between years 2003 and 2005. Measurements of samples cut out of were performed in RIAR Dimitrovgrad, Russia, the final project report is publicly accessible [6]. The FA irradiation was done during the 15 - 18 core loads in the fourth power unit of Novovoronezh NPP.

2. METHODS AND TECHNIQUES

2. 1. VVER-440 geometry model description

All the current VVER-440 fuel assemblies used in Slovakia are hexagonal and the fuel rods are placed in the assembly in a triangular grid pattern. The assembly is enclosed in a hexagonal wrapper with the width across the flat equal to 145 mm (the 2nd generation FA). The FA and emergency reactor control assemblies (ERC) are positioned in a hexagonal grid with a spacing of 147 mm. The fuel rods are in the bundle in a triangular grid pattern with a pitch of 12.3 mm. The fuel rod claddings are made of the E110 zirconium alloy (Zr + 1% Nb), while the wrapper tubes of FA and ERC are made of the E125 zirconium alloy (Zr + 2.5% Nb). The modelled outside diameter of fuel rod cladding is 9.1 mm and the inside diameter is 7.75 mm. The cladding houses a fuel column assembled of uranium dioxide pellet. Generally, several types of profiled fuel assemblies are used to maintain power peaking factors under the design limits. A Gd_2O_3 absorber is integrated with a mass content of 3.35% into number of FAs to aid fuel profiling. The profiling diagrams with various initial enrichments and locations of fuel bundles used in self-developed 2D SCALE models are shown in Fig. 1.



a) ERC fuel part - 3.84 $_{wt}$ % U-235 b) FA - 4.25 $_{wt}$% U-235 c) FA & ERC fuel part - 4.87 $_{wt}$ % U-235

Figure 1. Profiling diagram for fuel rod bundles used in computational model

2. 2. Calculation methodology

An accurate treatment of neutron transport and depletion in VVER-440 fuel assemblies characterized by heterogeneous and complex design requires the use of advanced computational tools. The depletion module TRITON [5], included in SCALE 6.1.3 code system developed by ORNL, was used to perform depletion simulations for two-dimensional FA and ERC models. The TRITON depletion module is coupling the 2D transport code NEWT with the point depletion and decay code ORIGEN-S. The TRITON lattice physics modelling approach for PWR fuel is extensively described in SCALE/TRITON primer [6]. For the sake of brevity, just the most important options used in our best modelling approach (BMA) and based on SCALE/TRITON primer and our experience for burnup calculations are shown here. The developed models are 2D assembly models with reflective boundary conditions on all sides, which represent infinite radial arrays of infinite length fuel assemblies. An unstructured coarse-mesh finite-difference acceleration approach (CMFD) is used with "partial-current" acceleration scheme. All results of sensitivity and parametric evaluations were calculated with the standard SCALE V7-238 multigroup neutron library based on ENDF/B-VII.0 evaluated data [7]. The V7-56 multigroup neutron library based on ENDF/B-VII.1 [8] evaluated data was used in the case of simplified benchmark calculation. Fuel pins with burnable absorbers were depleted by constant flux option instead of constant power approach. The average specific power of each model is derived from the average reactor power of NPP Bohunice unit 4 during cycle 30 and equals to 33.05421 kW_{th}/kgHM. The average concentration of boron acid (H_3BO_3) is obtained using the same approach and reaches cb=2.56 g/kg. The fuel is modelled with temperature of 933 K and the temperature of structural materials and water coolant is 555 K. The fuel pellet density is 10.55 g/cc and the density of zirconium alloys equals 6.55 g/cc. Very fine depletion steps (<0.5 MWd/kgHM) are used before Gadolinium peak reactivity to track the fast poison concertation changes. After peak reactivity, longer steps are used but are kept smaller than 1 MWd/kgHM.

2. 3. Sensitivity and parametric evaluations

The effects of various relevant depletion parameters on the models k_{eff} (due to the reflective boundary conditions identical to k_{inf}) and isotopic changes were investigated, however just the most interesting cases are presented. Variations of a single parameter are studied in each case; the remaining parameters are based on parameters used in BMA model.

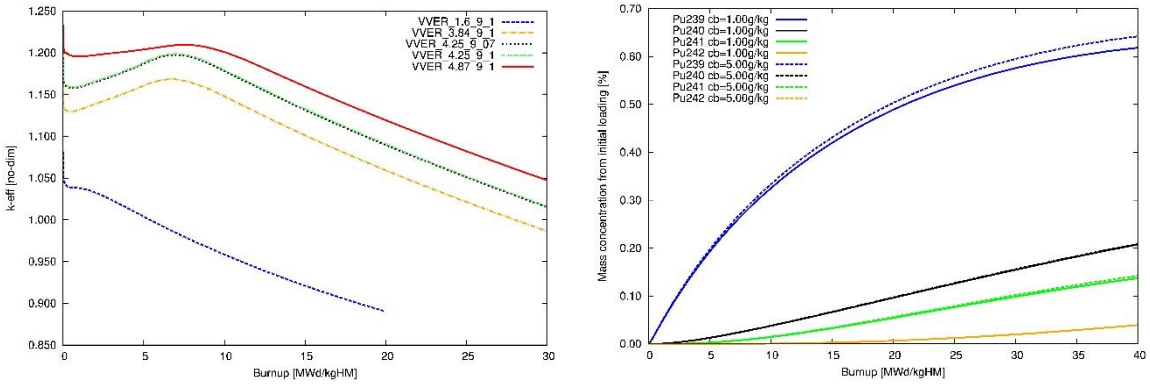
- Several VVER fuel types and enrichments of U-235 are investigated in the paper.
- Since the real-life operating history of each FA or ECS varies significantly, it is necessary to determine an operating history that is bounding in terms of its effect on reactivity, or to define a simple operating history with quantified margin that will bound the effect of operational variations. One of the most important parameters varying is the boron acid concentration. The four concentrations are investigated *cb={1; 2.56; 4; 5} g/kg*.
- The Gadolinium-bearing (Gd) pins should be treated with special approach including constant flux option and consideration of spatial depletion. The influence of both approaches to integral multiplication parameter and to isotopic are studied.
- Calculations were performed to assess the effect of assumed specific power, with varying the specific power from the half of BMA power to the doubled value.
- The moderator (also coolant) temperature varies significantly within the fuel axial profile, therefore it is necessary to assess its influence on the fuel multiplication factor and nuclide concentrations. The three water moderator with boric acid temperatures are investigated $T_M\ vs.\ \rho_{UO_2}$*={541.05 K; 0.7803 g/cc, 555 K, 0.7604 g/cc; 570.45 K, 0.7258 g/cc}*. It should be noted that the moderator/coolant density was calculated with assumption of the design pressure of $p = 12.26\ MPa$.

2. 4. The simplified benchmark

The calculation presented in the paper is based on data published for samples No. 21 and 182 taken from the fuel pin No. 6 of the FA No. D26135. The axial position of sample No. 21 with a length of ~ 10 mm cut out from fuel was approx. 1 m above the lower fuel pin plug. The sample No. 182 was in the lower part of the core just approx. 10 cm above the fuel pin plug. Our results for sample No. 149 from pin No. 54 can be found in our previous work [9]. The data in document [5] are not fully comprehensive, therefore user effect may be present in our approach. The spent fuel assembly was operated from October 1987 up to July 1991 (1369 calendar days). There were 1109 opeation days. A cooling period (since the end of irradiation period to the radiochemical analysis) made up approx. 12 years. Thus, the working assembly is typical of the majority of fuel assemblies, which are stored in NPP storage pools. The nuclide composition of spent nuclear fuel specimens was examined in the analytical and mass-spectrometric analysis laboratories of the RIAR [10]. The examination included dissolution of the cut-out specimen including control over fuel dissolution completeness, radiochemical extraction of U, Pu, Np, Am, Cm, Nd, Cs and Ce from fuel solution, nuclide composition measurements of the extracted elements, measurement of uranium, plutonium, neptunium, americium, neodymium, cerium and caesium mass fractions using isotopic dilution and measurements of Np-237, Cm and Ce-144 contents. All the results are burdened with nuclide specific error range for a confidence probability of P=0.95. These errors are considered in the results section of this paper. The radiochemical examinations of specific groups of nuclides were done in different and are precisely taken into account in the calculation model.

2. 5. Results

The results of different fuel types and the average enrichments of U-235 are shown in Fig. 2-a). As it can be seen, for ERC with the uniform enrichment of 1.6 wt% U-235 and without integrated burnable absorbers, the reactivity decreases monotonically with burnup in a nearly linear fashion. In contrast, for fuels with Gadolinium the reactivity increases as fuel burnup proceeds, reaches a maximum at the burnup where the absorber is nearly depleted (between 6 to 8 MWd/kgHM), and then decreases monotonically. The reactivity peak is more noticeable in the models with average enrichments of 3.84 and 4.25 wt% U-235 than in the 4.87 wt% U-235 case. This effect is caused by the different ratio of fissile to absorbing Gd isotopes and by their opposite influence on the criticality parameters in each assembly type. Interesting finding is that the highest peak value moves to higher burnup for FAs with higher uranium enrichment.

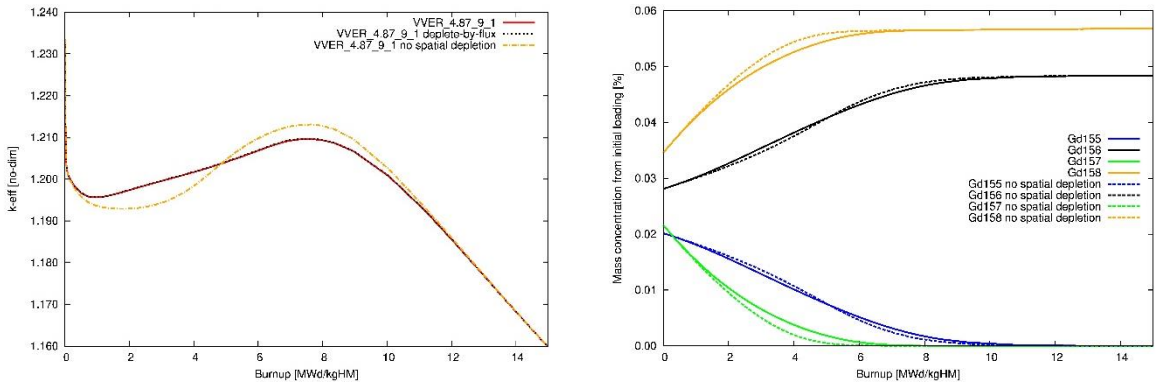


a) Effect of assembly type b) Effect of boron acid concentration

Figure 2. Effects of assembly type and boron acid concentration

It is not shown, but we can conclude that the reactivity trends of various boron acid concentrations are parallel during the whole burnup. The concentrations of important isotopes gathered during depletion with maximal and minimal investigated boron acid concentrations

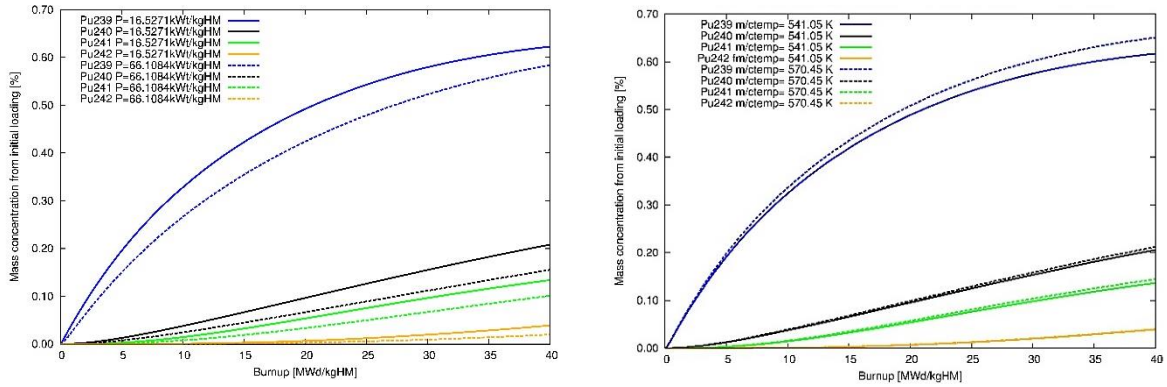
are shown in Fig. 2-b). The agreement diverges mainly for Pu-239 as a function of burnup. As mentioned above, the Gd pins should be treated with special approach including constant flux option and consideration of spatial depletion. The influence of both approaches to multiplication parameter and to isotopic changes is shown in Fig. 3.



a) Effect of deplete-by-flux option vs. spatial depletion b) Effect of spatial depletion for burnable poisons

Figure 3. Effects of Gadolinium-bearing pins modelling

It is evident from Fig. 3-a) that neglecting the depletion by constant flux option does not lead to different results as obtained by using BMA modelling approach. On the other hand, ignoring of fast spatial Gd concentration changes causes significant underestimation of the system reactivity in early burnup stages. Later the reactivity of Gd peak is systematically overestimated. To find the cause, Gd isotopes depletion is shown in Fig. 3-b). The Gd-155 and Gd-157 have much larger thermal cross sections than U-235, thus their concentration changes significantly influence the neutron balance of the system. Gd-157 depletes more rapidly in the case when spatial concentration changes are not taken into account. This faster concentration decrease is consequence of higher effective absorption cross section yielding to lower reactivity of the system. Consequently, this concentration difference causes higher reactivity amplitude of Gd peak in later burnup steps. To assess the effect of assumed specific power, the fuel temperature was kept constant in all calculation steps. Consistent with other studies, the general trend is for reactivity to decrease with increasing specific power. With respect to reactivity the use of minimum specific power seems to be conservative modelling option. As shown in Fig. 3.a), the concentrations of Pu-239 and Pu-241 decrease with increasing specific power, which would also tend to reduce reactivity. We assume that almost all important fission products decrease with decreasing specific power which would tend to increase reactivity.



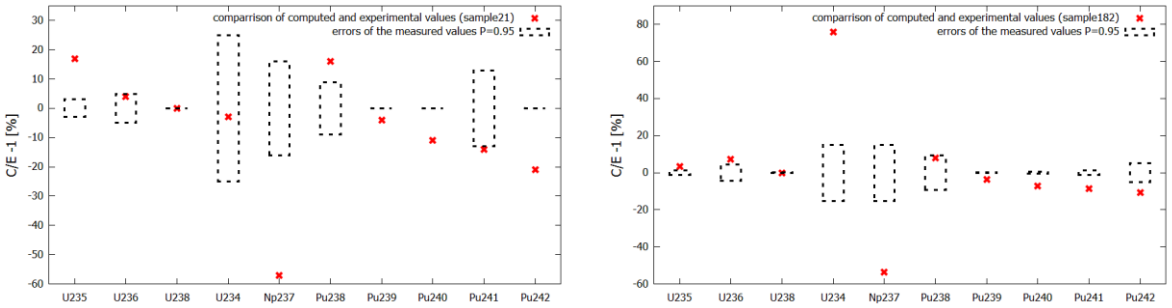
a) Effect of specific power b) Effect of moderator temperature

Figure 4. Effects of specific power and moderator temperature

The VVER-440 FAs are designed as under-moderated, therefore one can expect lower fuel multiplication properties with the higher moderator temperature. The results achieved confirm our expectations and are fully in accordance with theory. The concentrations of fissile

actinide Pu-239, see Fig. 4-b), are shown to rise with increasing moderator temperature. Isotope Pu-239 is formed from fertile U-238 by absorption of a resonance and thermal neutrons and then undergoing double decay through U-239 and Np-239 isotopes. On the other hand, Pu-239 has very high absorption cross-section for thermal neutrons. Consequently, Pu-239 concentration is sensitive to the level of system under-moderation.

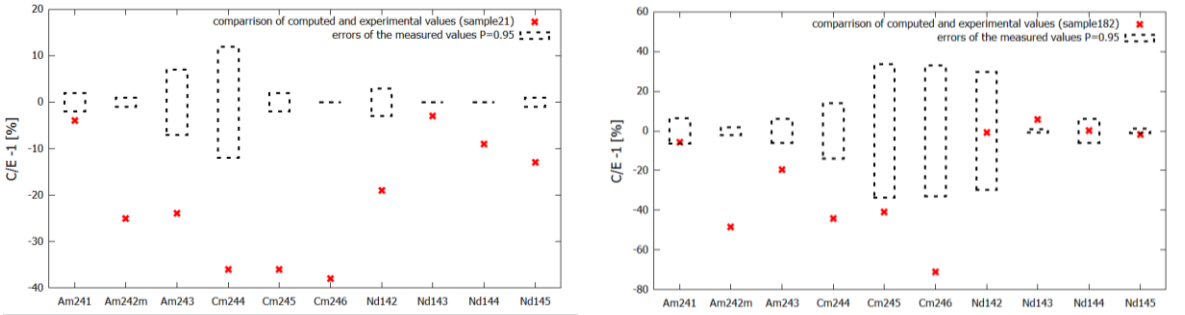
The results of the simplified benchmark calculation for samples No. 21 and 182 of main and minor actinides are mainly shown in Fig. 5 and Fig. 6. The calculated (denoted as C) and experimentally determined (E) nuclide concentrations are presented in the form of C/E-1 ratio. As can be seen, the highest relative deviation is in the case of U-235 and sample No. 21, where the calculation overestimates the average measurement value approximately by 20 %. The resonance absorption by U-238 is enhanced by Doppler broadening with increased fuel temperature. According to almost no deviation of calculated and measured values for U-238, we conclude that fuel temperature was modelled in appropriate way.



a) Comparison of results – sample 21 b) Comparison of results – sample 182

Figure 5. Graphical comparison of results part 1

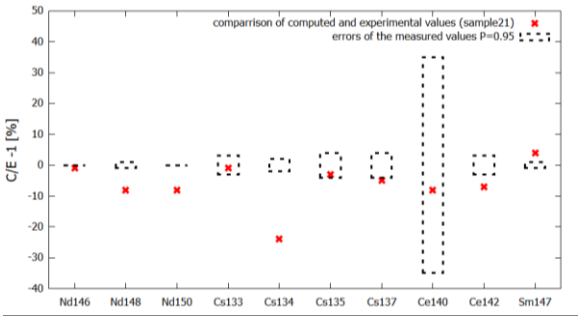
As shown above, the concentration of Pu-239 is sensitive to the level of system under-moderation due to its very high absorption cross-section for thermal neutrons. The comparison of results shows the good agreement for Pu-239 nuclide and both samples; therefore, we assume that fuel density and system under-moderation was modelled correctly. It is worth mentioning, that overall results of sample No. 182 are systematically closer to the measured values. The reason of this behaviour is not clear to us and should be investigated further.



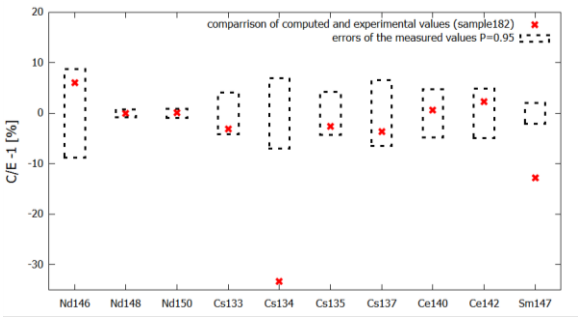
a) Comparison of results – sample 21 b) Comparison of results – sample 182

Figure 6. Graphical comparison of results part 2

Contrary, the final concentration of Pu-241 is slightly underestimated by approx. 5 to 10 %. This underestimation can influence the multiplication properties of final spent fuel configuration. In case of the neutron absorbing materials the concentration of Pu-238 is significantly overestimated, what is compensated by underestimating of Np-237. The precursor of Np-237, which precedes formation of U-232 and thus limits fuel reprocessing by Tl-208 gamma radiation, is the U-236, of which the calculated concentration lies near to the measurement error boundary.



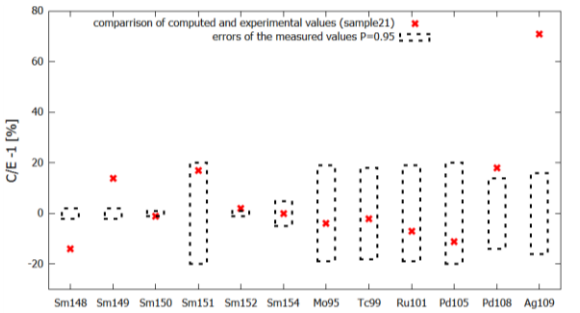
a) Comparison of results – sample 21



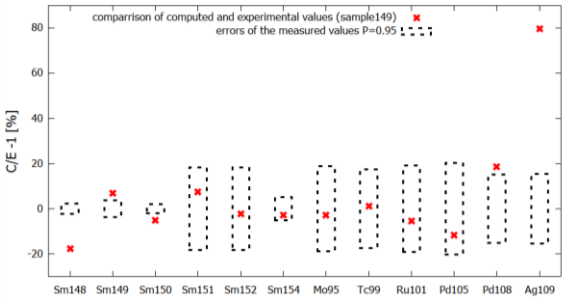
b) Comparison of results – sample 182

Figure 7. Graphical comparison of results part 3

The minor actinides concentrations are underestimated varying on the nuclide to nuclide basics. The Am-241 concentration is slightly underestimated (approx. -4 to -10%). This deviation should be taken into account in follow-on criticality and decay heat calculations. The Fig. 6 to Fig. 8 mainly show results for fission products, for which most calculation results lies slightly under or in the error range of measurements. The higher discrepancies can be found for Cs-134 (-30 %), Sm-148 (-19 %) and Ag-109 (80 %) nuclides. The results of Cs-137 as the primary source of penetrating gamma radiation from spent fuel until 300 years of discharge are satisfactory and lie under the measurement errors.



a) Comparison of results – sample 21



b) Comparison of results – sample 182

Figure 8. Graphical comparison of results part 4

3. CONCLUSION

The analysis of spent nuclear fuel properties and nuclide compositions is a very important stage for the nuclear fuel cycle as well as for neutron-physical calculations of VVER-type reactor cores and their future improvement. This paper described the main results of analyses performed to achieve a better understanding of the modelling issues associated with VVER-440 reactors. The influence of calculation approaches and parameters have been summarized, however much more effort is needed, and further calculations are planned. It was demonstrated that special care should be given to Gadolinium bearing pins. Surprisingly the constant by flux depletion option does not play a significant role in the concentration calculations. The influence of operational history to final reactivity was proven and briefly analysed. The concentration of boron acid significantly influences Pu-239 production as a function of burnup. It was found out that with respect to reactivity the use of minimum specific power seems to be conservative modelling option. The relevant effect was found in the case of variations of moderator/coolant temperature. This observation just supports the fact that axial depletion should be carefully considered where mainly the moderator temperature plays a vital role. In fact, all investigated parameters have little effect on fission-product worth. The most sensitive isotope in all investigated cases was Pu-239 isotope. To assess the currently developed Best Modelling Approach the comparison between the calculation results and experimental data from Novovoronezh NPP was provided. Results obtained indicate that the U-235 calculation

overestimates the average measurement value approximately by 20 %. The minor actinides concentrations were in general underestimated, where in case of Am-241 the underestimation reached approximately -10%. For the investigated fission products, the good agreement was achieved, where most calculation results fell into the error range of measurements. The higher discrepancies were identified in the case of Cs-134 (-30 %) nuclide. This work was based on simplified benchmark definition and is affected by user experience and chosen modelling approach. The influence of the used cross-section library is questionable and will be further investigated. In the case of simplified benchmark, the higher number of energy groups can bring better results mainly for the main and minor actinides, however, used 52 group library is already optimized for pressurized water reactors. Therefore, other approaches and nuclear data libraries should be studied to support the "best practices" used for VVER-440 burnup credit calculations.

4. ACKNOWLEDGEMENT

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