Assessment of the Photon and Neutron Source Term for the NPP Krško Spent Fuel

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ABSTRACT

Accurate knowledge of the fuel nuclide inventory is important after reactor shut down, during the fuel storage and subsequent reprocessing or disposal to provide adequate shielding from the photon and neutron radiation. In this paper possibility to calculate the NPP Krško photon and neutron source term with the Serpent code has been analysed. Some deficiencies in the supplied ENDF/B-VII.0 decay library have been observed. In addition, Serpent reports only spontaneous fission rates without (α, n) and (β, n) contributions. To get neutron emission, spontaneous fission rates had to be multiplied with the average number of neutrons born for each particular nuclide manually. Comparison with the Origen code has shown acceptable agreement of the ENDF/B-VII.1 results. Influence of several factors such as fuel burnup, enrichment, temperature, moderator temperature (density), soluble boron concentration, average power, and burnable absorbers has been analysed. In addition, it was demonstrated that, except for the burnup and enrichment, averaging of all other parameters is acceptable approach. IFBA fuel should be accounted for explicitly due to relative high impact on the photon and neutron emissions.

Keywords: PWR fuel, photon source term, neutron source term

1   INTRODUCTION

In a nuclear reactor, the fission of heavy atoms such as isotopes of uranium and plutonium results in the formation of highly radioactive fission products. Due to neutron capture higher actinides are formed, some of them being unstable. All these unstable isotopes radioactively decay and produce decay heat and radiation. Accurate knowledge of the nuclide inventory is important after reactor shut down, during the fuel storage and subsequent reprocessing or disposal. Namely, proper radiation protection is a primary objective in the transportation, storage or processing of irradiated nuclear fuel. The designing of adequate shielding to provide this protection requires appropriate knowledge of the radiation source in spent nuclear fuel.

In this paper possibility to calculate the fuel isotopic composition and determination of the photon and neutron source term with the Serpent code [1] is investigated. Serpent is a well-known

Monte Carlo code used primarily for the calculation of the neutron transport in the reactor. It has been validated for the burn-up calculations [2]. However, in the calculation of the photon and neutron source term different set of isotopes is important than in the neutron transport case. For the purpose of this evaluation only the radioactive decay of fission products and actinides is considered. A typical case of the NPP Krško fuel is selected for comparison with the Origen code [3]. Origen is a well-known computer code system for calculating the buildup, decay, and processing of radioactive materials. Comparison with the Serpent code is performed to verify that the Serpent is taking into account all isotopes important to assess photon and neutron radiation. A similar analysis comparing fuel radioactivity [4] and decay heat [5] has already been performed, showing promising results. After the code validation a sensitivity study is carried out. Fuel isotopic composition is namely pretty dependent on the neutron spectrum and consequently on fuel operating conditions. Influence of several factors such as fuel burnup, enrichment, temperature, moderator temperature (density), soluble boron concentration, average power, and burnable absorbers is analysed.

2 BRIEF CODE DESCRIPTION

2.1 Origen

The Oak Ridge Isotope Generation (ORIGEN) depletion/decay code was developed at ORNL in 1973. Since then, many new versions have been created [3] and are available from RSICC and NEA data bank. It is a well-known point-depletion inventory code and has been used to model nuclide transmutation with capability to generate source terms for accident analyses, characterize used fuel (including activity, decay heat, radiation emission rates, and radiotoxicity), activate structural materials, and perform fuel cycle analysis studies. Origen uses a matrix exponential method to solve a large system of coupled, linear, first-order ordinary differential equations with constant coefficients (Bateman equations). Version 2.1 with "pwrue" library has been used in this paper.

2.2 Serpent

Sensitivity study of the radiation source term is performed with the Serpent code [1, 2]. Serpent is a three-dimensional continuous-energy Monte Carlo reactor physics burnup calculation code, developed at the VTT Technical Research Centre of Finland. It is not a typical Monte Carlo code. While the majority of the other codes use the ray-tracing algorithm as a transport model, Serpent uses the Woodcock delta tracking method. In this way the geometry routines get simplified and calculations are faster compared to conventional Monte Carlo codes. The improved matrix exponential method CRAM (Chebyshev Rational Approximation Method) for solving the Bateman equations [6] has been implemented for the burnup applications. Serpent uses a continuous energy neutron cross section library in an ACE format. Libraries provided with the code distribution are based on the JEF-2.2, JEFF-3.1, JEFF-3.1.1, ENDF/B-VI.8 and ENDF/B-VII.0 evaluated nuclear data files [7]. The code is specialized for two-dimensional lattice physics calculations, but the universe-based geometry description allows the modelling of complicated three-dimensional geometries as well. Detailed geometrical modelling of the NPP Krško fuel assembly in the Serpent code enables accurate determination of fuel isotopics and consequently determination of the photon and neutron source term. In this analyses Serpent 2, version 2.1.28, has been used.

3 COMPARISON OF THE CODES AND LIBRARIES

The NPP Krško is a 2-loop Westinghouse PWR that began electricity production in 1981. The start-up core had a rated thermal capacity of 1,876 MWt, and a 626 MWe gross electric power. Currently, the thermal rating is 1,994 MWt with 727 MWe gross electric power. The core consists of 121 standard 16×16 fuel assemblies with some VANTAGE+ features. A typical fuel assembly with 4.95 % enrichment and no IFBA (Integral Fuel Burnable Absorber) rods was selected as a starting point. A reference case scenario consists of the following reactor operational parameters:

1.   Fuel temperature 900 K,
2.   Moderator temperature 580.46 K with density 0.70871 g/cm^3,
3.   Soluble boron concentration of 1000 ppm.

Parameters are close to the average operational parameters applied in the last NPP Krško cycles. Comparison of several cases is presented in Fig. 1, where photon emission rate per kg of initial uranium fuel is plotted for the burnups up to 60000 MWd/tU. Emissions calculated with the ENDF/B-VII.0 library are almost 40 % lower than emissions predicted with the Origen code. This is somehow in contradiction with the [4] where predicted activities were higher compared to the Origen results. Closer examination has shown that the ENDF/B-VII.0 decay library supplied with the code contains some dubious photon data for some nuclides. For example, there are no photon data for the ^{240}Np, resulting in a zero emission, although ^{240}Np substantially contributes to the total photon emissions. Situation is a little better with the JEFF-3.1.1 library, but the discrepancy with the Origen is still relatively high. Because of that, it was decided to implement ENDF/B-VII.1 library supplied with the MCNP code [8]. Comparison with the Origen shows much better agreement. However, differences are mainly in short lived isotopes as can be seen from Fig. 2, where all 3 libraries give almost identical results after a few days of fuel cooling.
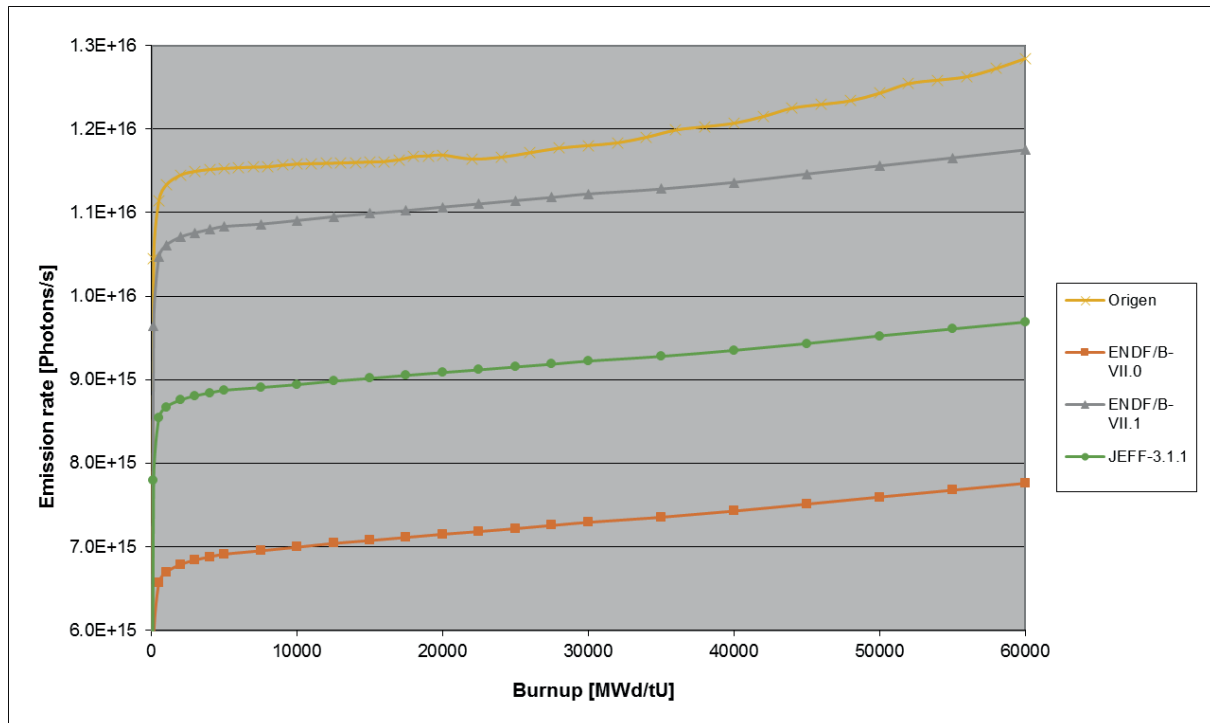

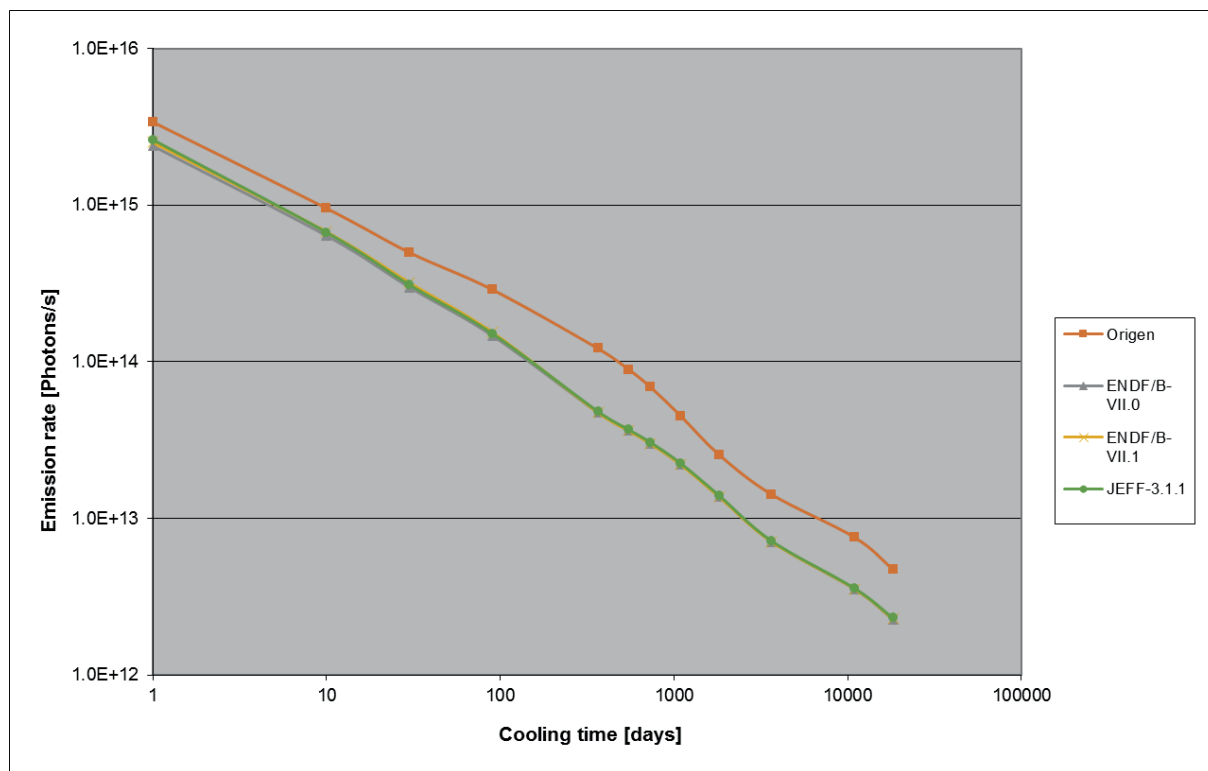Figure 1: Photon emission rate during fuel burnout

Figure 2: Photon emission rate during fuel cooling

When we tried to estimate neutron emissions, we encountered a few obstacles. Serpent currently predicts neutron emission only from spontaneous fission. There are no models to estimate neutrons from (α, n) and (β, n) reactions. Nevertheless, since spontaneous fission contributes a majority of neutron emissions (roughly 90 %), it is judged that the presented analysis is still representative. Additional drawback is that the Serpent reports only spontaneous fission rates. To get number of neutrons, fission rates have to be multiplied with the average number of neutrons born (nu-bar, $\overline{\nu}$). Since this value differs for different fission nuclides, fission rate for each nuclide had to be multiplied with corresponding $\overline{\nu}$. Values from the ENDF/B-VII.1 library have been taken in all Serpent cases. At the end, it should be noted, that since at longer cooling times only insignificant fractions of the neutron source are caused by the photoneutron reactions, this type of neutron source was neglected in the analysis.

Photon emission comparison of a considered cases is presented in Fig. 3, where neutron emission rate per kg of initial uranium fuel is plotted for the burnups up to 60000 MWd/tU. ENDF/B-VII.0 values are again questionable, since they differ noticeable from other cases at low burnups. Closer examination has shown, that the spontaneous fission rate for the ^{238}U is 100 times too large due to wrong data in the decay library. Other libraries are more consistent with the Origen code. During the fuel cooling (Fig. 4) JEFF-3.1.1 library is a little closer to the Origen code. ENDF/B-VII.0 and ENDF/B-VII.1 results are almost identical.

In the rest of the paper only values calculated with the ENDF/B-VII.1 library are reported, since the results seem to be the most consistent.
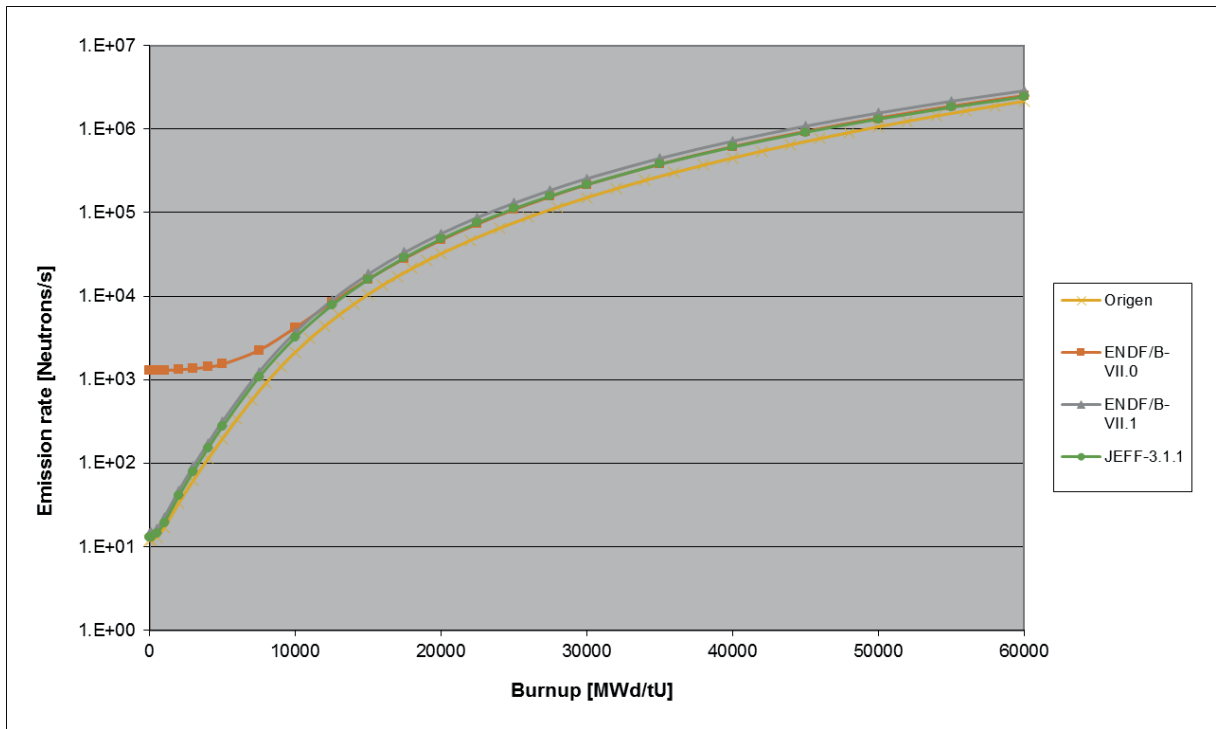
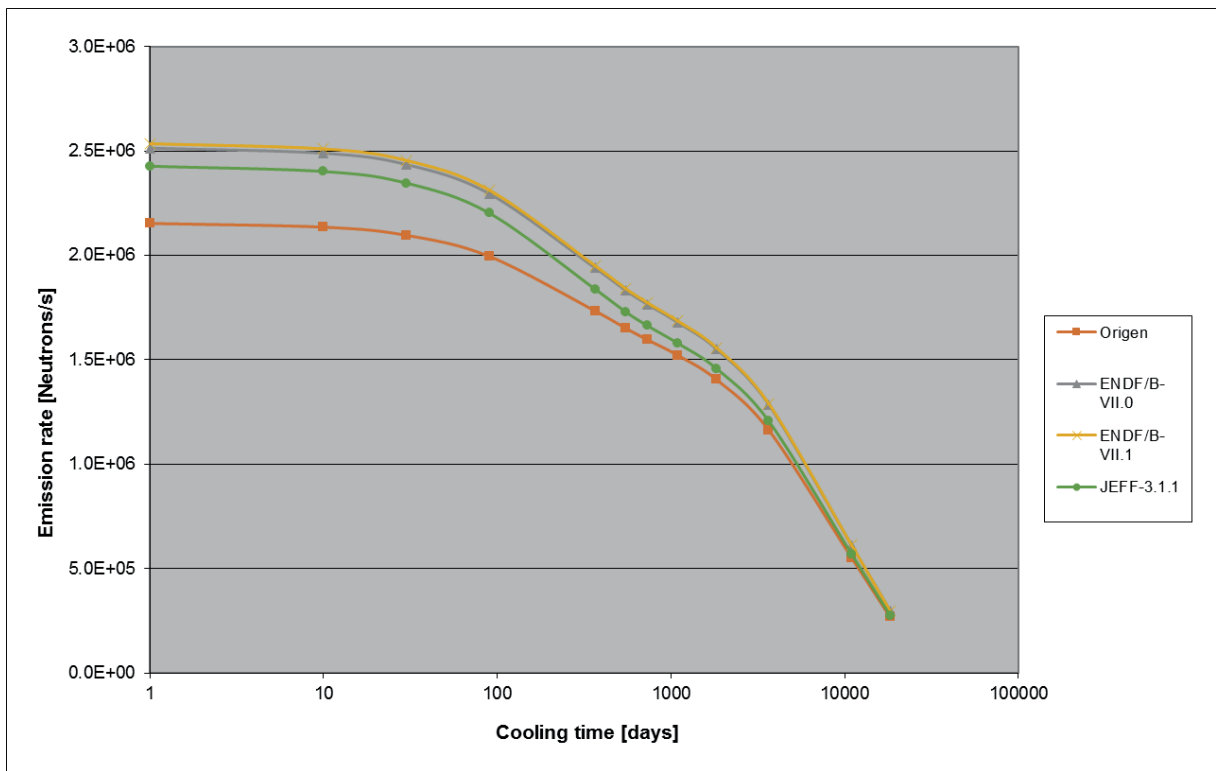Figure 3: Neutron emission rate during fuel burnout

Figure 4: Neutron emission rate during fuel cooling

4 SENSITIVITY STUDY

Photons and neutrons in the spent fuel are produced by the radioactive decay of unstable nuclides. Isotopic inventory of these unstable nuclides in the fuel is constantly changing during the reactor operation and is a function of several parameters such as neutron energy spectrum, present fission nuclides inventory etc. Reactor operating conditions (temperatures of the fuel and moderator, water density, neutron absorbers etc.) directly influence neutron spectrum. It is prudent to evaluate effect of these parameters for the development of optimal calculation strategy to achieve desired accuracy. In addition, in the photon and neutron source term evaluations usually some averaging of material properties or conditions is applied. It is not self-evident that such averaging process would yield also an averaged source term value. To verify such assumptions, a sensitivity analysis is needed to confirm such approach or to identify crucial nonlinear parameters.

4.1 Burnup

Emission rates versus fuel burnup has already been presented in Figs. 1 - 4. As can be seen from Figs. 1 and 3, changes in the isotopic inventory and consequently emission rates are very high during the fuel burnout. Relative differences in the emission rates for the 40000 MWd/tU and 50000 MWd/tU cases are presented in Figs. 5 and 6. They are rather high and are increasing over cooling time. A curve denoted as "Aver" represents the difference between average of the 40000 MWd/tU and 60000 MWd/tU case relative to the 50000 MWd/tU case. Relative difference is less than 0.6 % for the photon case allowing reasonable averaging process. Situation is very different for the neutron emissions. Fig. 3 is showing increased exponential behaviour (be aware of logarithmic scale), which can clearly not be adequately described by linear function. Therefore, averaging process would produce high errors. "Aver" curve on Fig. 6 is showing errors of almost 25 % for the averaging over ±10000 MWd/tU interval. Closer examination has shown that the major neutron contributors are ^{244}Cm (80 % – 97 %) and ^{246}Cm (0.7 % - 3 %) nuclides. Concentration of both nuclides is very sensitive to the fuel irradiation and initial composition.
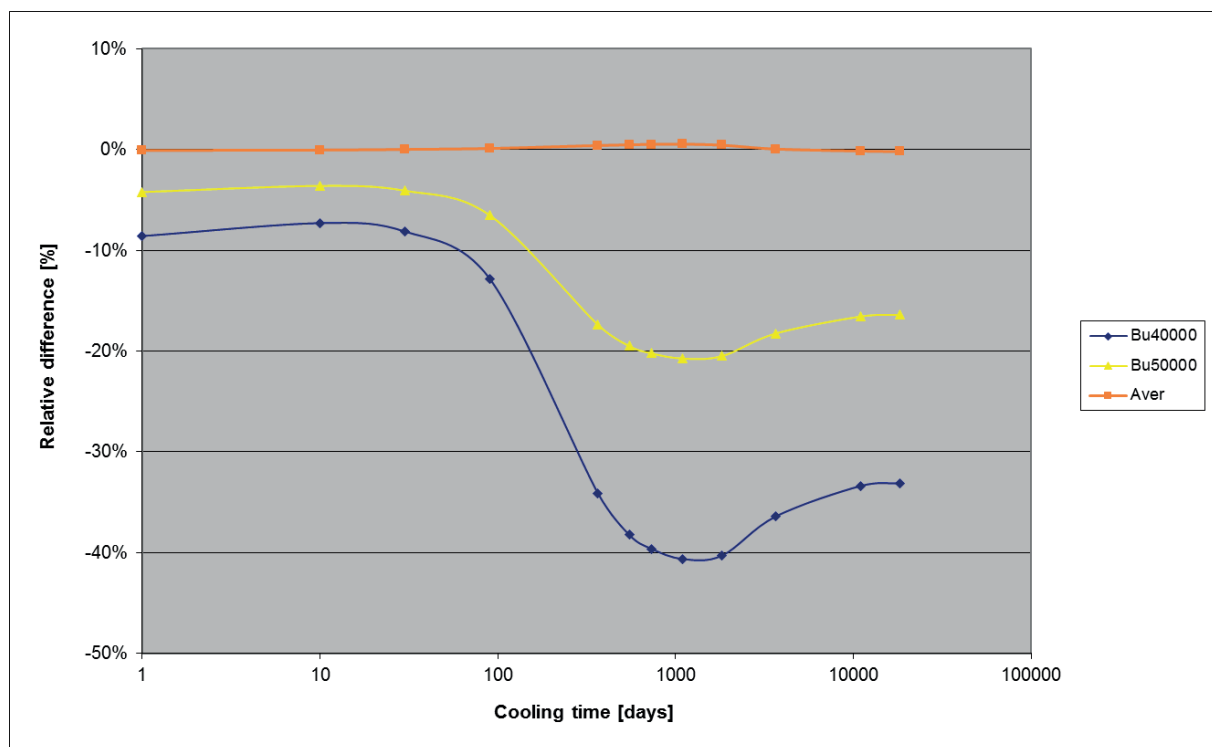

Figure 5: Effect of the burnup on the photon emission rate
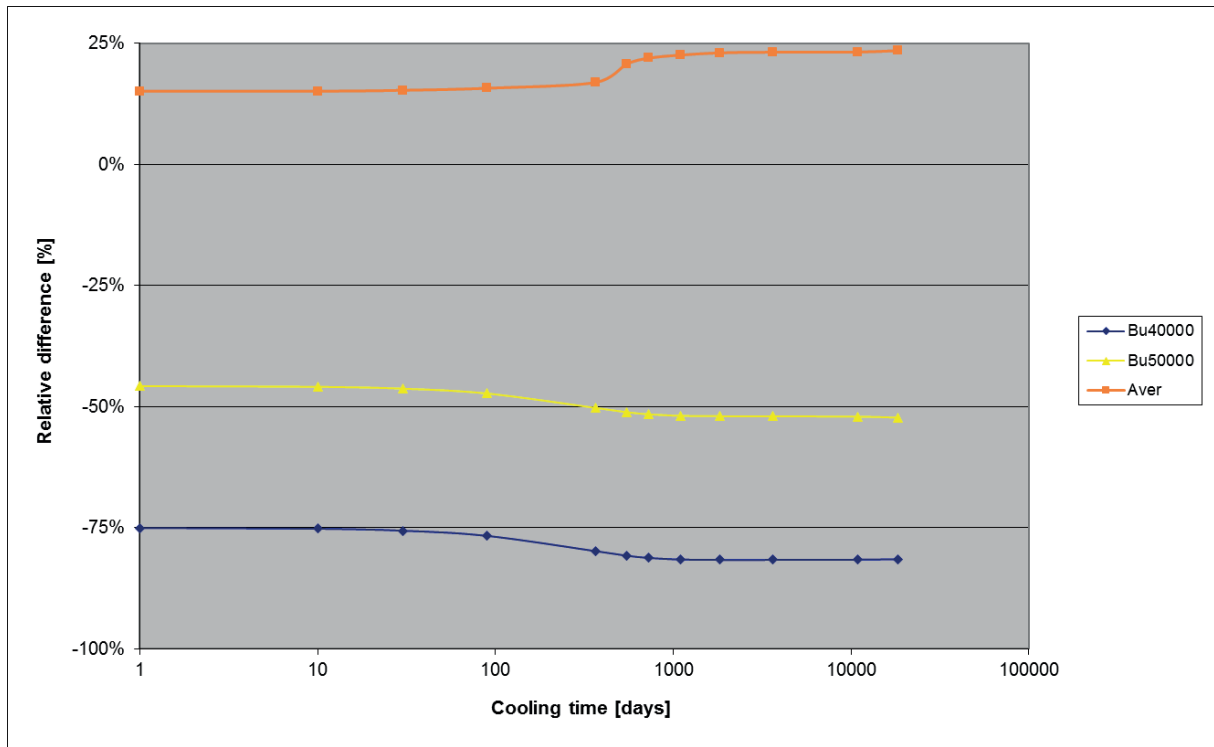
Figure 6: Effect of the burnup on the neutron emission rate

4.2 Enrichment

The effect of the fuel enrichment is shown in Figs. 7-8. Two cases (2.1 % and 3.525 %) are compared to the reference 4.95 % case. It can be estimated that a decrease of 1 % in enrichment can induce increase of more than 5 % in the photon emissions rate and 70 % in the neutron emissions. Moreover, the effect is not linear over the cooling period.

As already mentioned, in practice it is a usual procedure to calculate larger fuel areas with some average properties. But strictly speaking, if the fuel assembly is constituted of 2 geometrically equal regions, the one with 2.1 % enrichment and the other with 4.95 %, the average emission rate would not be equal to the emission rate of the fuel with an average 3.525 % enrichment. Differences between the average emission rate of the 2.1 % and 4.95 % fuel region and the emission rate of the averaged 3.525 % region are plotted as "Aver" in Figs. 7 and 8. Differences in photon emission rate of up to 1 % are visible in the 1-10 days region. Much larger differences of up to 15 % are observed for the neutron emission rate confirming unsuitability of the enrichment averaging process for the neutron source term.
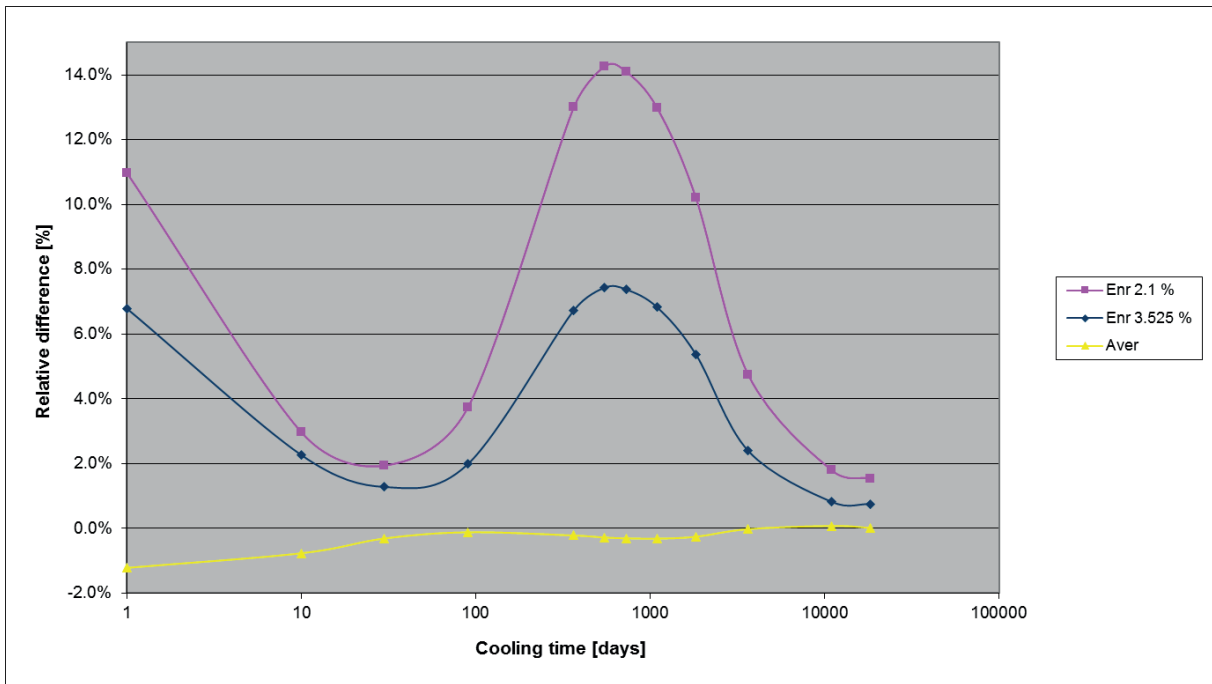
Figure 7: Effect of the fuel enrichment on the photon emission rate
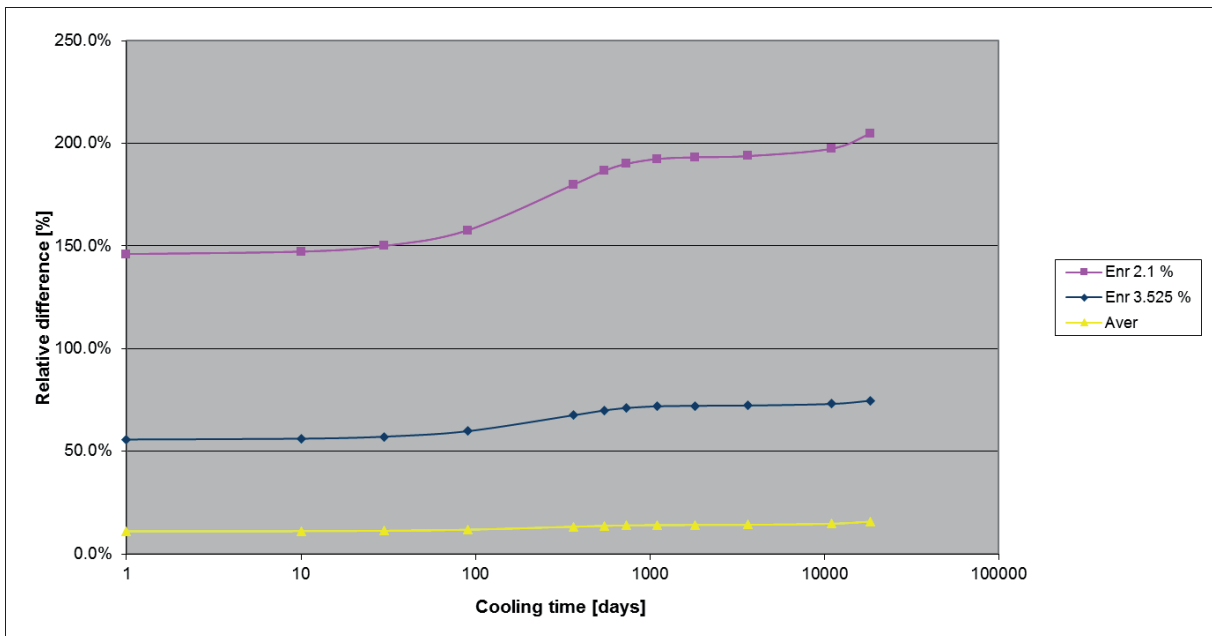

Figure 8: Effect of the fuel enrichment on the neutron emission rate

4.3 Fuel temperature

The effect of fuel temperature is shown in Figs. 9 and 10. Two cases (800 °K and 1000 °K) are compared to the reference 900 °K case. Relative differences of both cases are less than 0.25 % for the photon and 0.5 % for the neutron emission rate. Average emissions are very close to the reference case supporting averaging approach.

It should be noticed that in the reality fuel temperature is not constant over fuel pellet but is highest in the centre and decreases towards the cladding due to heat transfer to the moderator. If we assume constant power across the pellet having constant thermal characteristics, we get a parabolic temperature profile. A 10 region annular case was considered assuming a parabolic temperature profile with a pellet average of 900 °K. Obtained results are plotted in Figs. 9 and 10 as "T profile".

Effect is relatively small and is estimated to be less than 0.2 % in the photon case and 0.3 % for the neutron case.
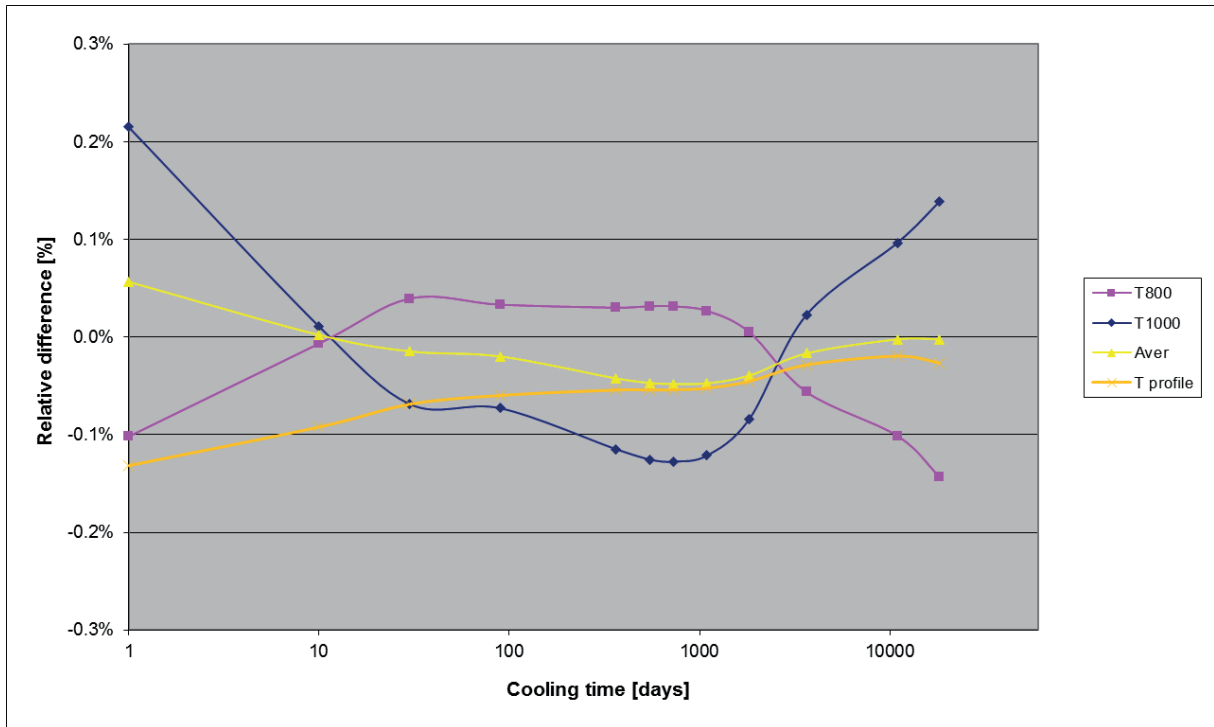

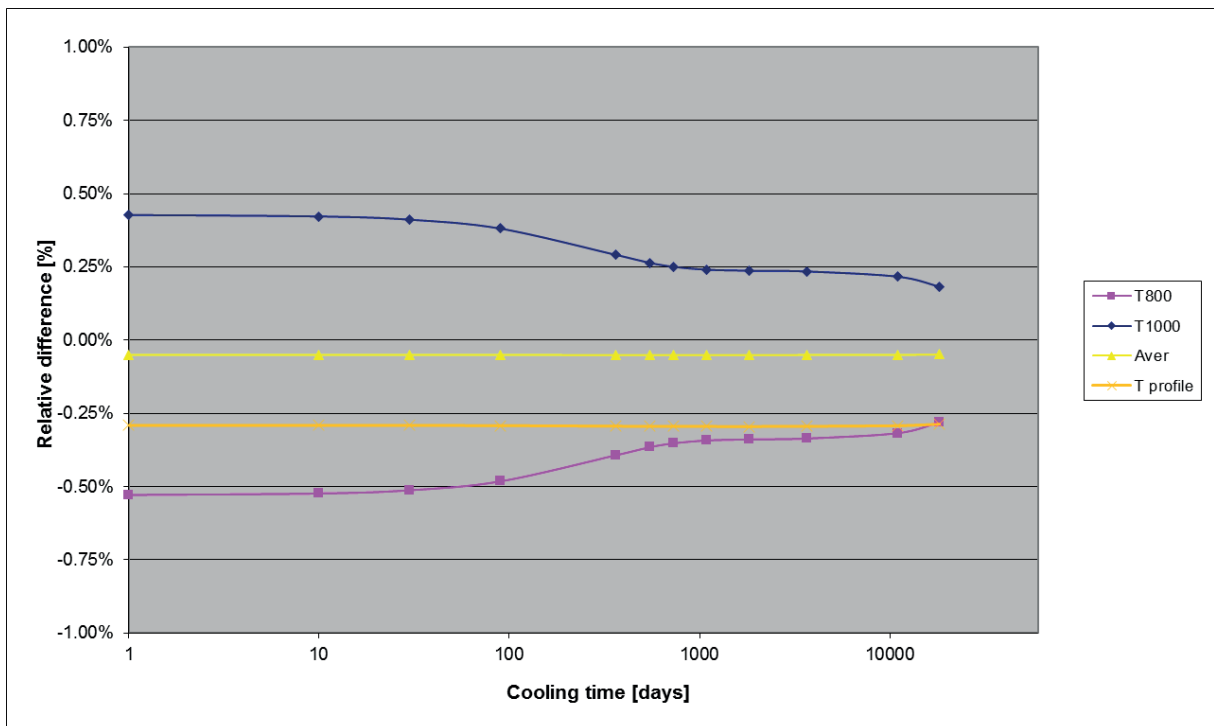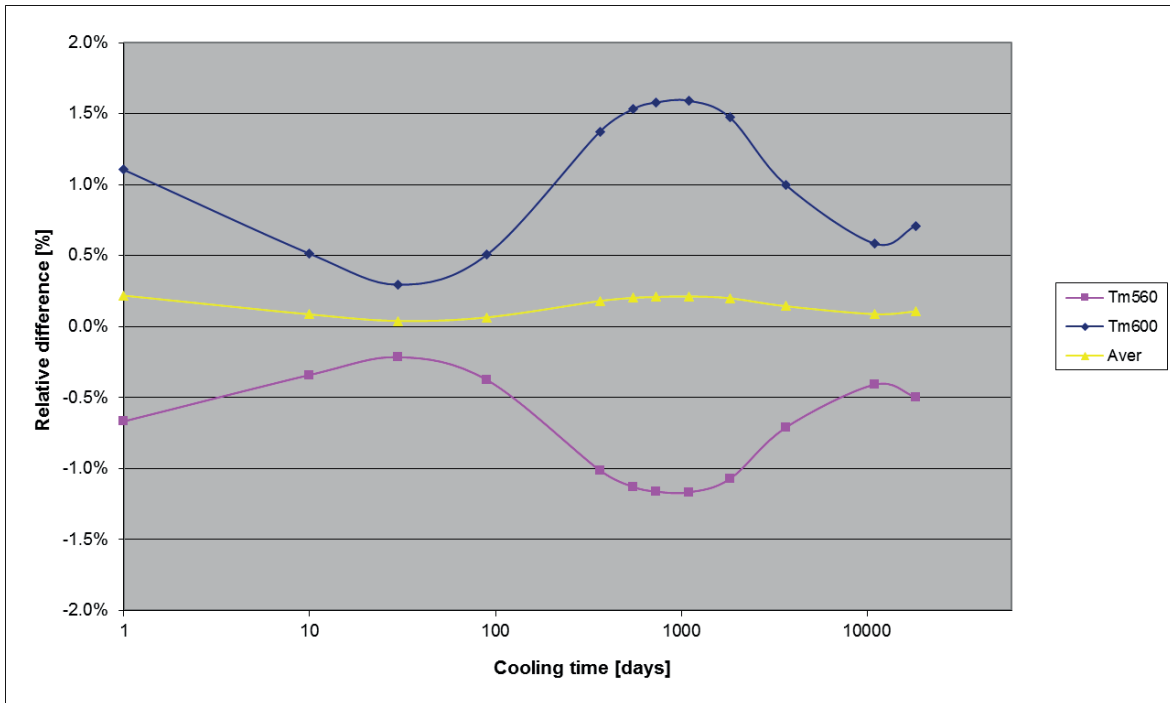Figure 9: Effect of the fuel temperature on the photon emission rate

Figure 10: Effect of the fuel temperature on the neutron emission rate

4.4   Moderator temperature

Direct influence of the moderator temperature on the emission rates is negligible. However, reactor is operating in the pressure region, where relatively small moderator temperature changes cause

significant differences in the moderator density producing significant differences in neutron moderation and energy spectrum. The effect is shown in Figs. 11 and 12. Temperature was varied for ±20 °K (water density goes from 0.74972 g/cm^3 to 0.65642 g/cm^3). This is approximately NPP Krško inlet–outlet range at 100 % reactor power. Therefore, upper fuel regions with near core outlet temperatures have up to 3 % higher photon emission and up to 8 % higher neutron emission than bottom regions. Average emissions are up to 0.5 % higher than emissions at average temperature.

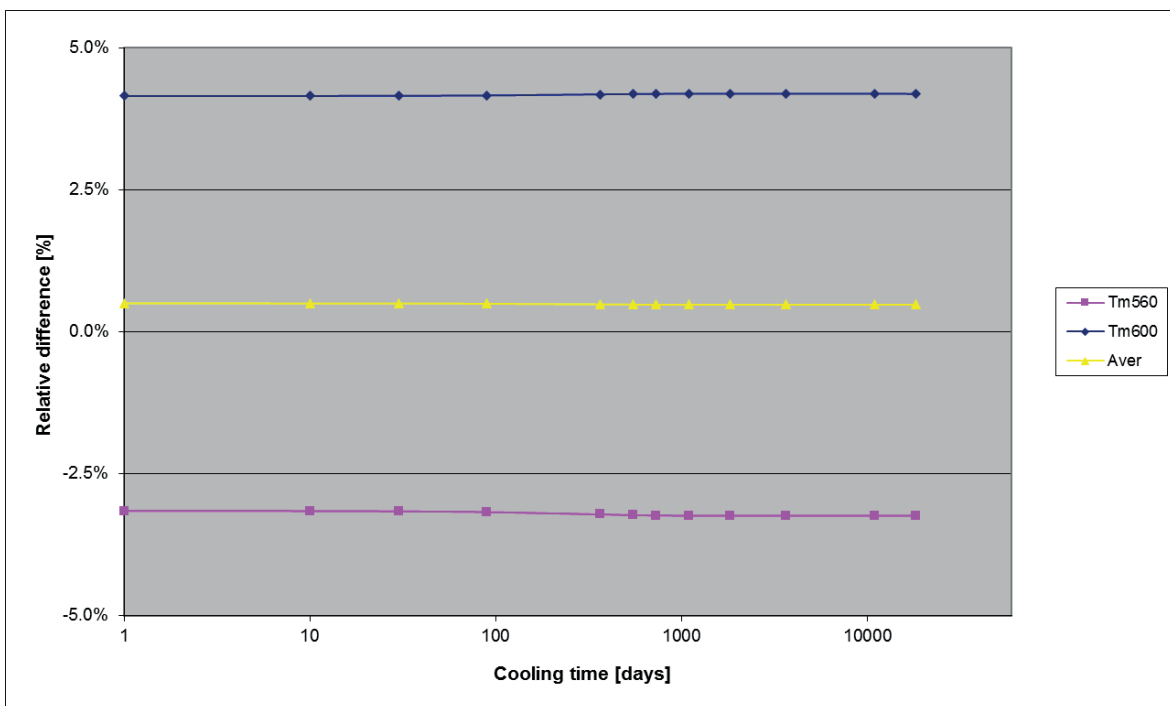Figure 11: Effect of the moderator temperature on the photon emission rate

Figure 12: Effect of the moderator temperature on the neutron emission rate

4.5   Soluble boron

The effect of soluble boron present in the moderator is shown in Figs. 13 and 14, where ±500 ppm variations are analysed. Boron concentration increase of 500 ppm can cause up to 1 % higher photon emission and 2.5 % higher neutron emission. Average emissions are almost the same as the emissions at average boron concentration supporting averaging approach.
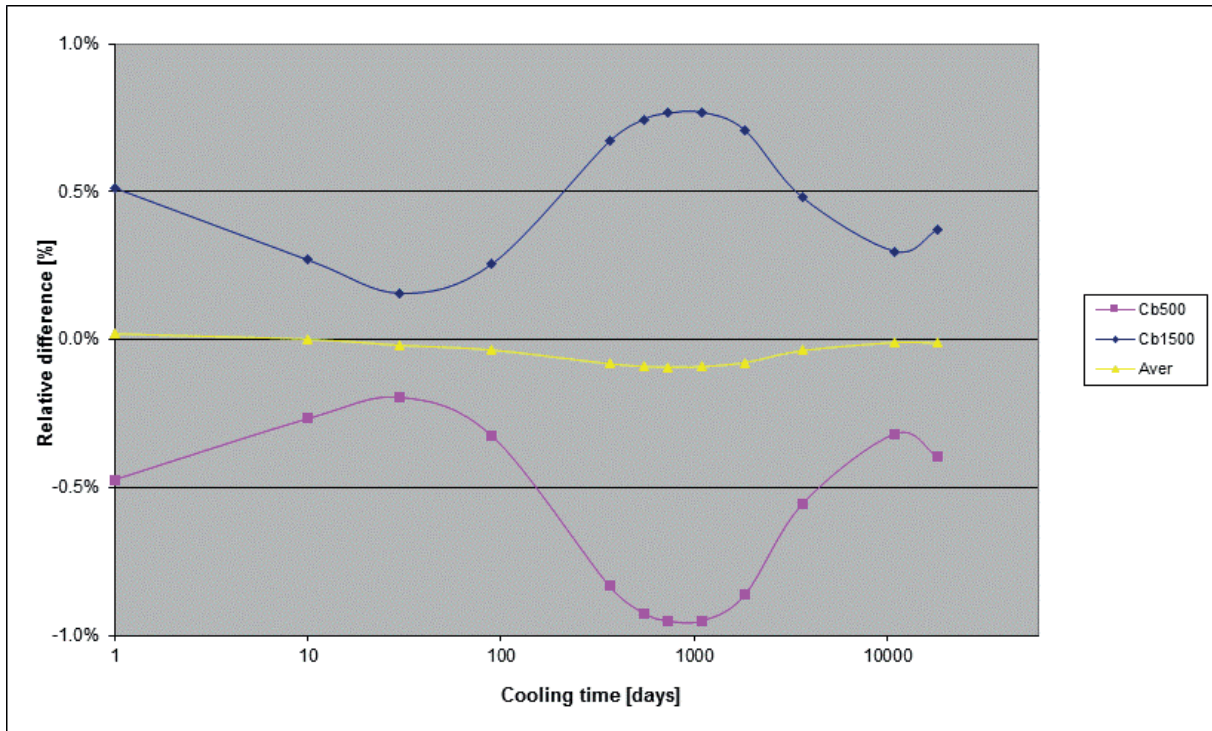

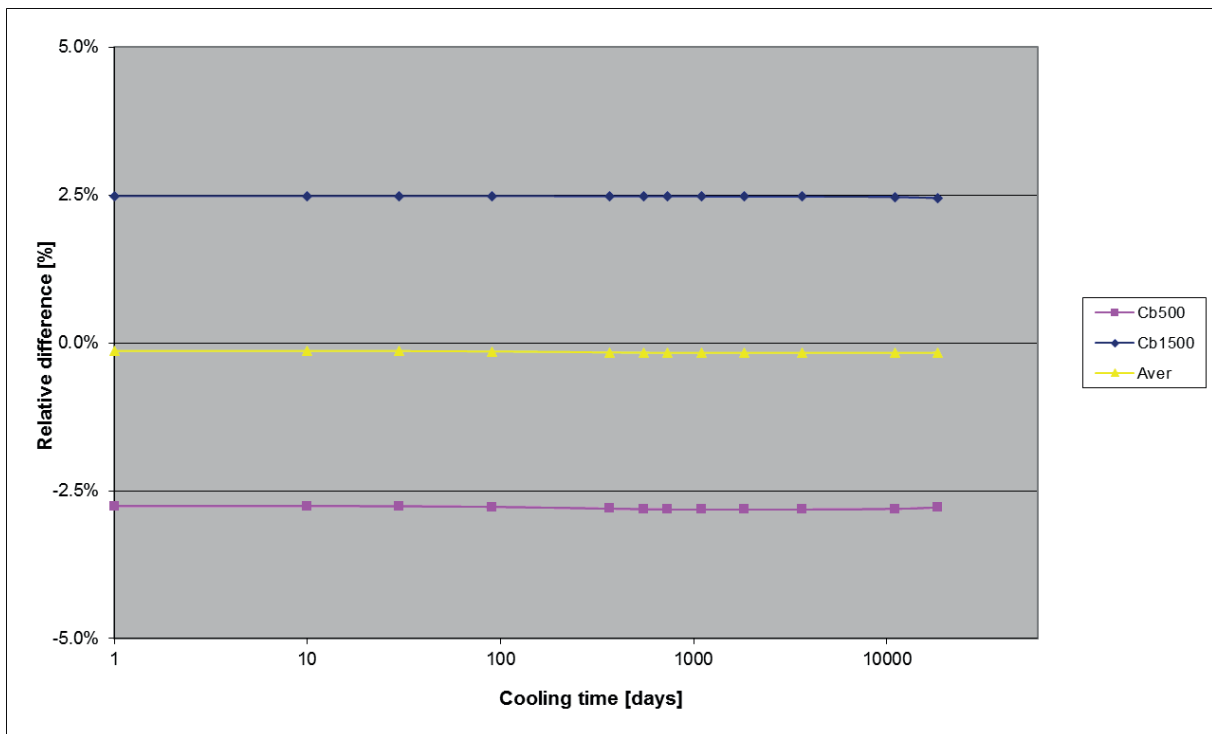Figure 13: Effect of the soluble boron on the photon emission rate

Figure 14: Effect of the soluble boron on the neutron emission rate

4.6   Average power

Cases with 70 % nominal power and 130 % nominal power are presented in Figs. 15 and 16. Photon source term at shutdown is almost proportional to the specific power. 1 % increase in the reactor power results in almost 1 % higher photon emission. That means that the main photon contributors experience some saturation concentration, which is proportional to the specific power. Differences are gradually decreasing over time and are after 10 years cooling approximately 10 times smaller (less than 3 %). Average photon emission is up to 2 % lower than the emission at average specific power. Situation is rather different for the neutron source term (Fig. 16). Emissions are much less dependent on specific power. Differences are less than 2 %. Average neutron emission is up to 0.5 % lower than the emission at average specific power.
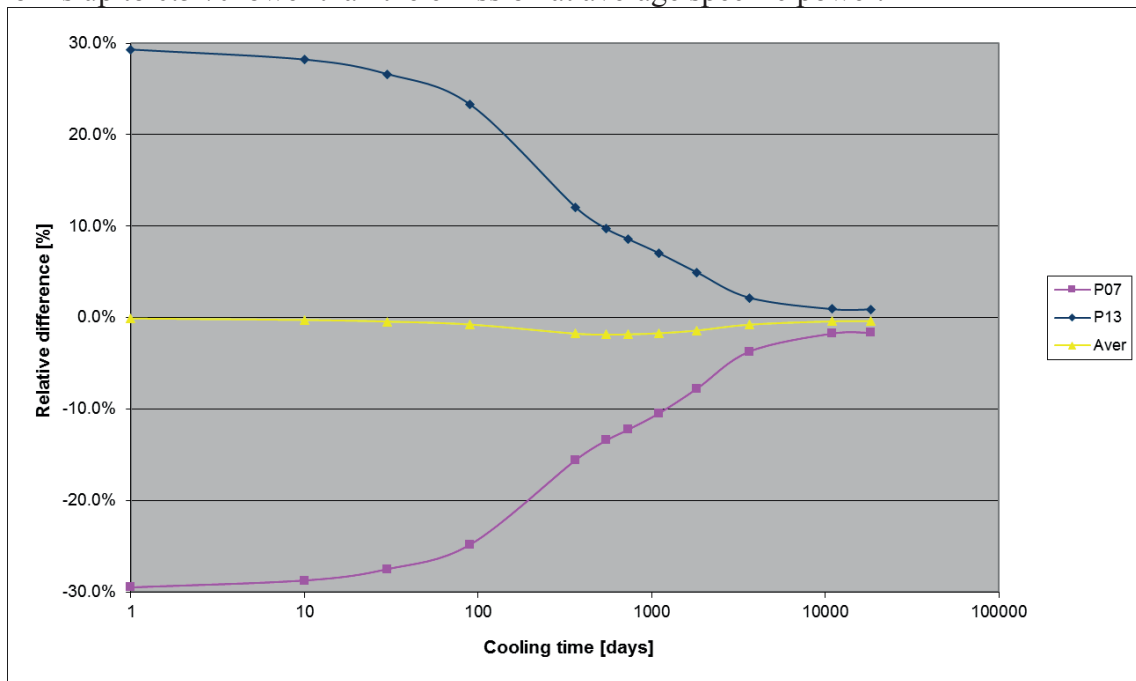

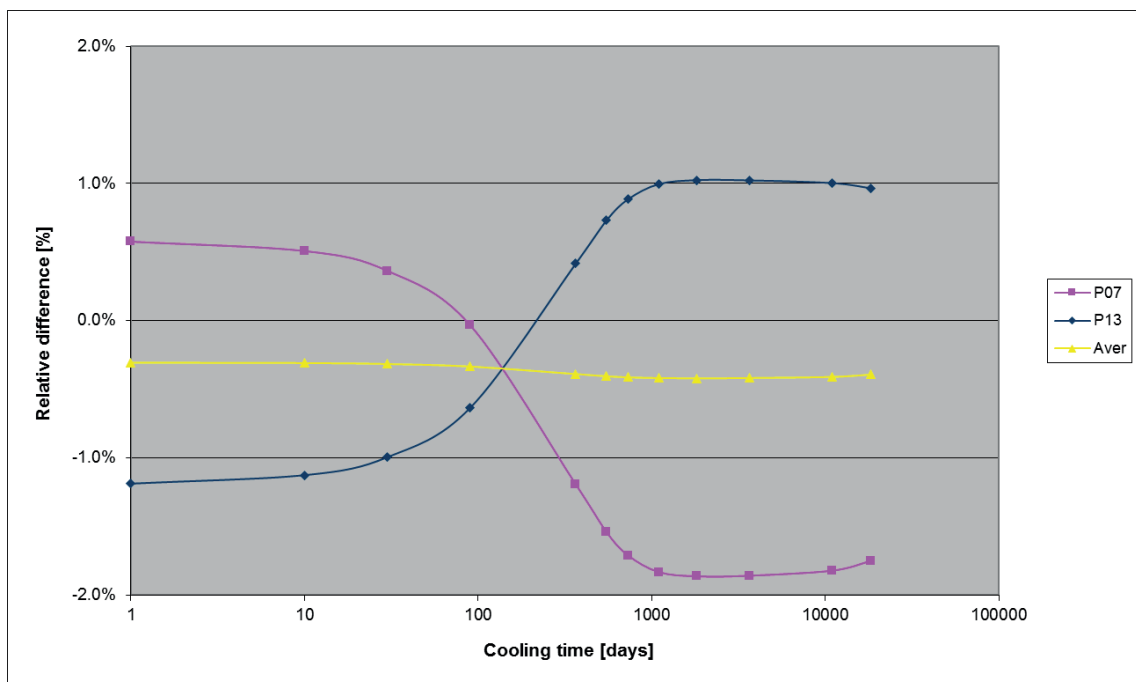Figure 15: Effect of specific power on the photon emission rate

Figure 16: Effect of specific power on the neutron emission rate

4.7   Burnable absorbers

Introduction of burnable absorbers causes spectrum hardening and therefore changes in fuel isotopic composition and consequently increases photon end neutron emissions. The effect is presented in Figs. 17 and 18, where cases with 64 IFBA rods and 116 IFBA rods per assembly are plotted. The fuel with 116 IFBA rods is producing more than 4 % more photons and almost 11 % more neutrons than the fuel with no IFBA rods.
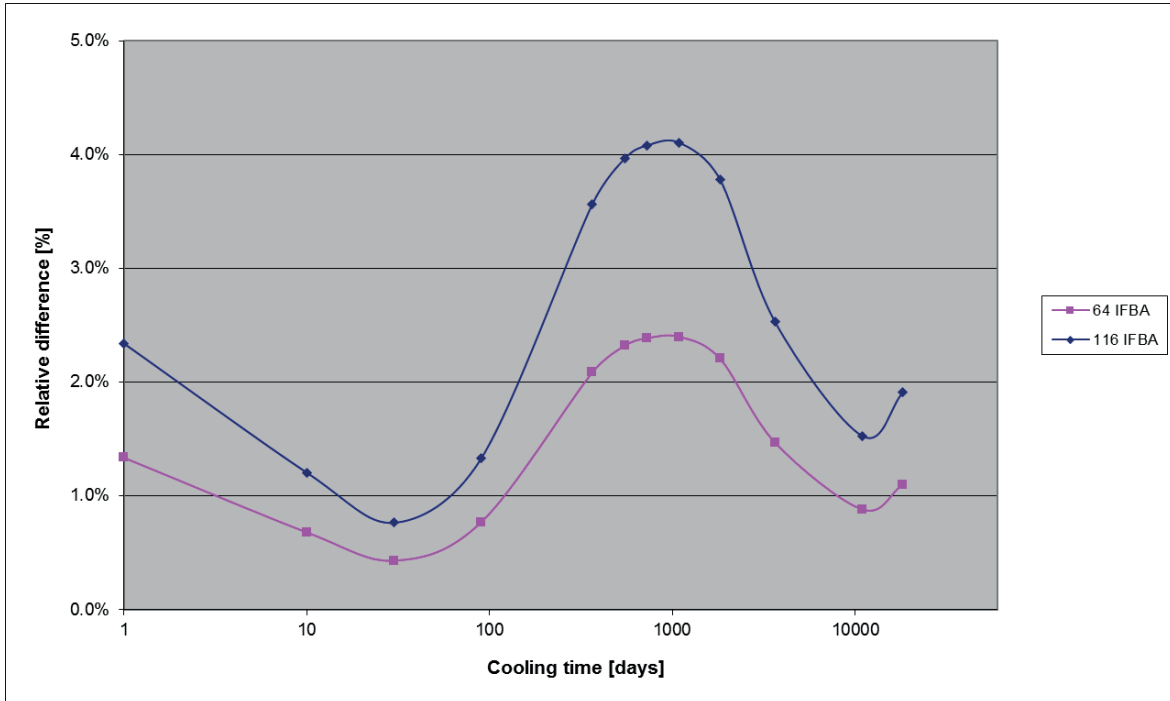
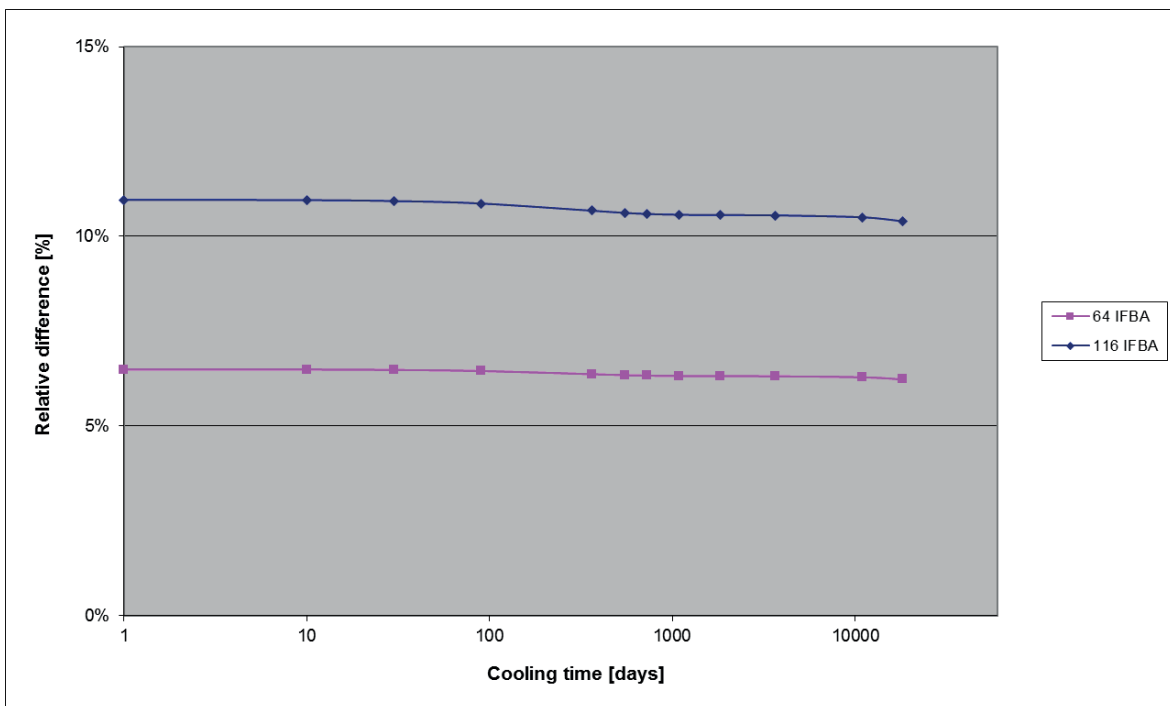Figure 17: Effect of IFBA rods on the photon emission rate

Figure 18: Effect of IFBA rods on the neutron emission rate

5   CONCLUSION

Analysis of the NPP Krško photon and neutron nuclear fuel source term with the Serpent code has been performed. Some deficiencies in the supplied ENDF/B-VII.0 decay library have been detected. In addition, it was noticed that the Serpent reports only spontaneous fission rates without (α, n) and (β, n) contributions. To get neutron emission, spontaneous fission rates had to be multiplied with the average number of neutrons born for each particular nuclide manually. Comparison with the Origen code has shown acceptable agreement of the ENDF/B-VII.1 results.

Since the photon and neutron source term are dependent on the fuel isotopic composition and consequently also on neutron flux spectrum, the effect of several operational parameters was examined. Influence of the following parameters on the emissions have been analysed:

1. burnup,
2. fuel enrichment,
3. fuel temperature, profile,
4. moderator temperature,
5. soluble boron concentration,
6. average power,
7. burnable absorbers.

Realistic operating ranges were considered. In addition, it was demonstrated that, except for the burnup and enrichment, averaging of all other parameters is acceptable approach. IFBA fuel should be accounted for explicitly due to relative high impact on the photon and neutron emissions.

ACKNOWLEDGMENT

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