ESTIMATION OF TRITIUM PRODUCTION IN VVER-440 REACTOR CORE DURING NORMAL OPERATION

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Abstract: Slovakia as one of the world leading countries in the share of nuclear power in electricity production and currently operates 2 nuclear power plants, each with 2 VVER-440 units. In addition to these reactors there are 2 VVER-440 units under construction and 2 units in decommissioning. The VVER-440 technology features thermal neutron spectrum, low enriched uranium dioxide fuel and light-water coolant with diluted boric acid. The reactivity control during normal operation is ensured by the system of emergency reactivity control assemblies (ERC), symmetrically arranged in 37 positions in the reactor core, and by boric acid, homogenously diluted in the moderator. Due to the presence of ^{10}B in the coolant/moderator which has high thermal neutron capture cross-section, the absorption of neutron on these atoms may lead to tritium production. Tritium as the radioactive isotope of hydrogen with half-life of 12.3 years, strongly contributes to the level of radioactivity of the primary coolant, therefore the NPP staff must have appropriate knowledge of its production during operation. The production rate of tritium may be assessed either by experimental measurement or by simulation using state-of-the-art calculation tools. This paper focuses on the estimation of the tritium production for a specific scenario of the operation of the 3rd unit of Mochovce NPP. For simulations the SCALE6 [1] system is used with the detailed calculation model [2] developed at the B&J NUCLEAR ltd. company. The whole-core 3D model of VVER-440 consists of the reactor in-vessel components such as fuel assemblies (including fuel rods, upper spacer grid, intermediate spacer grids, supporting grid, mixing grid, central tube and fuel endings), emergency reactor control assemblies (absorber and fuel part), core basket, barrel and the reactor pressure vessel. The boundaries of the created VVER-440 whole-core model are given by the outer surface of the dry shielding, the level of hot-leg piping and the basement of filtration mechanism. The calculations presented in the paper are performed using self-shielded multi-group cross-section libraries, taking into account the operation conditions of Mochovce unit 3 NPP [3] in the first fuel campaign.

Keyword: *Tritium production, VVER-440, activity, criticality, SCALE system, Mochovce NPP*

1. INTRODUCTION

The light-water coolant as passes through the reactor core, to remove the generated heat from fuel assemblies, gets activated by capturing moderated neutrons. During normal operation numerous neutron induced reactions take place, generating a variety of radioactive nuclides, such as ^{16}N, ^{17}N, ^{14}C or ^{3}H. Due to the relatively long half-life of the majority of these activation products, their concentration in the primary coolant directly influences the radiation exposure of personnel performing maintenance activities. Among all radioactive nuclides produced during reactor operation tritium (^{3}H) is one of the most important. It has high isotopic exchange rate, high residence time in the environment, long half-life (12.3 years) and can easily leak into

the environment in a form of water. In addition, the limits for radioactive releases allow the dilution of tritium (12 GBq per year), in order to decrease its volumetric activity, in waste waters exiting the nuclear power plant. If tritium is not separated effectively from biosphere, it will become a part of the global cycle. Once it is absorbed through food or water, it will steadily exist in human body for long time and induce internal radiation damage [4].

VVER reactors are the most frequently built reactor types in the world. The first units with predecessors of VVER-440 type reactors were erected at the Novovoronesh NPP site in 1972 and 1973 [5]. The second step in the development of VVER-440 type reactors was the V-230 design which was mainly constructed in the period from 1973 to 1982. The third step in VVER-440 development was the V-213 reactor design. Slovakia has four nuclear reactors of this type generating half of its electricity and two more under construction. In 1972, construction of the Jaslovské Bohunice V1 plant commenced, with two VVER-440 V-230 reactors. The first was grid connected in 1978, the second two years later. The V2 units commenced operation in 1984 and 1985. The Slovak NPP Mochovce with VVER-440 V-213 units 1 and 2 were put in operation in the summer of 1998 and the end of year 1999 due to the construction delay caused by political changes in the early 1990s. Two another units 3&4 of VVER-440 V-213 are currently under construction in Mochovce and are planned to be put in operation in 2020 [2]. Since the operation of the Jaslovské Bohunice V2 NP and the first two units of Mochovce NPP started decades ago, estimation of the tritium inventory through calculation would be a very challenging issue. However, the construction of the 3rd unit of Mochovce NPP provides unique opportunity to develop a new methodology to numerically estimate the production rate of tritium in its primary coolant and to compare the result with real experiment, once the reactor is put into operation.

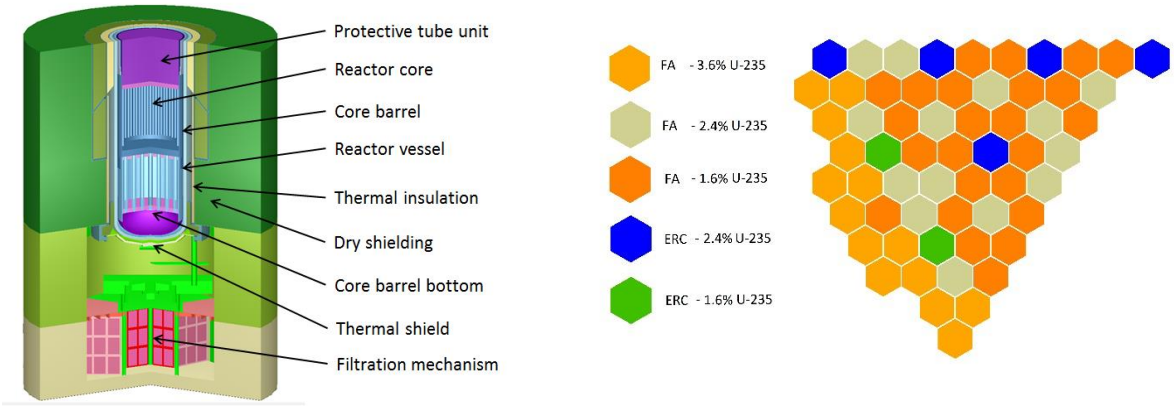
This paper presents the first step of a complex joint long-term research activity between the Slovak University of Technology in Bratislava and the B&J NUCLEAR ltd. research company. This activity is based on the detailed 3D model of the VVER-440 reactor core developed in the state-of-the art SCALE6 calculation system and the experience of the research team with evaluating neutron physical characteristics of VVER-440 units for the Nuclear Regulatory Authority of Slovak Republic for more than 3 years.

2. OVERVIEW OF THE CALCLULATION METHODOLOGY

2. 1. The VVER-440 reactor core

The detailed and precise 3D model of the VVER-440 V-213 reactor has been developed for criticality and shielding calculations in the KENO-VI code of the SCALE6 system. The whole-core 3D model is shown in Figure 1-a. It consists of the reactor in-vessel components, such as fuel assemblies (including fuel rods, upper spacer grid, intermediate spacer grids, supporting grid, mixing grid, central tube and fuel endings), emergency reactor control assemblies (ERC - absorber and fuel part), core basket, barrel and the reactor pressure vessel. The boundaries of the created VVER-440 whole-core model are given by the outer surface of the dry shielding, the level of hot-leg piping and the basement of filtration mechanism [2].

The VVER-440 fuel assemblies (FAs) are of hexagonal shape and consist of 126 UO_2 fuel pins, placed in the assembly in a triangular grid pattern. The bundle of fuel rods in the assembly is enclosed in a hexagonal wrapper with 145 mm width across the flat. The fuel assemblies and emergency reactor control assemblies are positioned in a hexagonal grid with a spacing of 147 mm. The fuel rods are located in the bundle in a triangular grid pattern with 12.3 mm pitch. The fuel rod cladding is made from E110 zirconium alloy, while the wrapper tubes of FA and ERC are made from E125 zirconium alloy [2].



a; 3D reactor model b; Equivalent sixth of the fuel loading pattern

Figure 1. KENO 3D model of the VVER-440 V-213 reactor and fuel loading pattern

As it was mentioned in the introduction part, the analysis presented in this paper was carried out for the first fuel loading of Mochovce unit 3, considering a simplified approach. This approach assumes fresh fuel composition without burnup through the cycle and variable critical boron acid concentration and coolant cycling. The fuel loading pattern used in the present analysis is shown in Figure 1-b. Due to the 60° symmetry of the core, only one sixth of the pattern is presented. The fuel loading pattern is based on our previous work and consists of FA and ERC assemblies with 2.4 % and 3.6% enrichment of ^{235}U [5].

2. 2. Critical boron acid concentration

The critical boric acid concentration is a concentration that ensures critical state of the reactor for the given effective full power day (EFPD) without requiring the positions of the ERC assemblies to be adjusted. Compared to the ERC assemblies the boric acid ensures uniform distribution of absorber in the core, however requires time consuming dilution, if its concentration is to be decreased, and may cause positive reactivity feedback, in case of high concentration. The critical boron acid concentration was calculated using the ANDREA macro code [6]. This code, the product of Nuclear Research Institute (NRI) REZ, has been being developed since 2005. The code is designed to support safety analyses of VVER type reactors [7]. The results of critical boric acid concertation are shown in Figure 2.

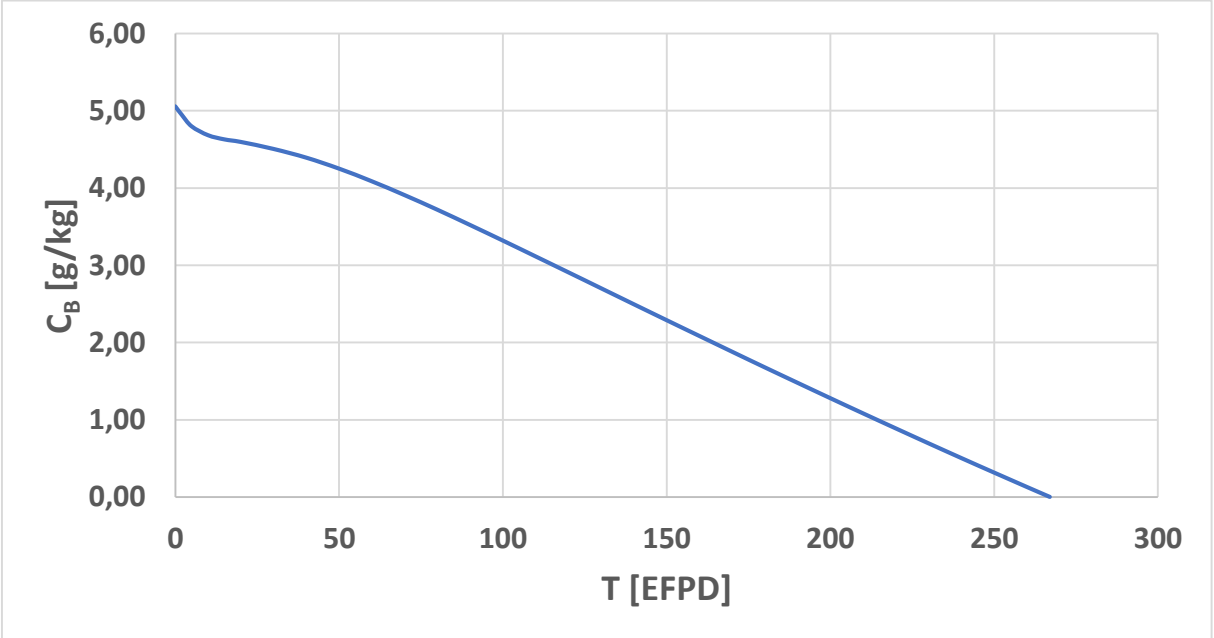


Figure 2. Critical boric acid concentration through the fuel cycle

As it can be seen, the critical boric acid concentration of fresh fuel (0 EFPD) is 5.059 g/kg. As a result of fuel burnup, the reactivity of the system decreases, so does the critical boric acid concentration that reaches 0 g/kg at 266.95 EFPD. Usually, after the boric acid concentration has reached zero, the operation of the reactor continues by withdrawing the ERC assemblies from operation position (in our case 225 cm) to the upper edge of the core (250) and by decreasing the reactor power (1741 MWth in our case), core inlet temperature (268 °C) and the pressure at the main steam collector, however these operational regimes were not considered in our analysis.

2. 3. Dilution of boric acid

By operating the reactor for certain EFPDs the reactivity of the system decreases. To ensure criticality, the reactivity bond by boric acid is released by diluting its concentration. The dilution is performed by pumping out a certain amount of moderator and replacing it with demineralized H_2O. As the boric acid is diluted the concentration of ^{10}B and ^{11}B atoms decreases, however the concentrations of ^{16}O, ^{1}H and ^{2}H proportionally increase. The concentrations of ^{10}B and ^{2}H atoms in coolant are illustrated in Figure 3 and Figure 4.

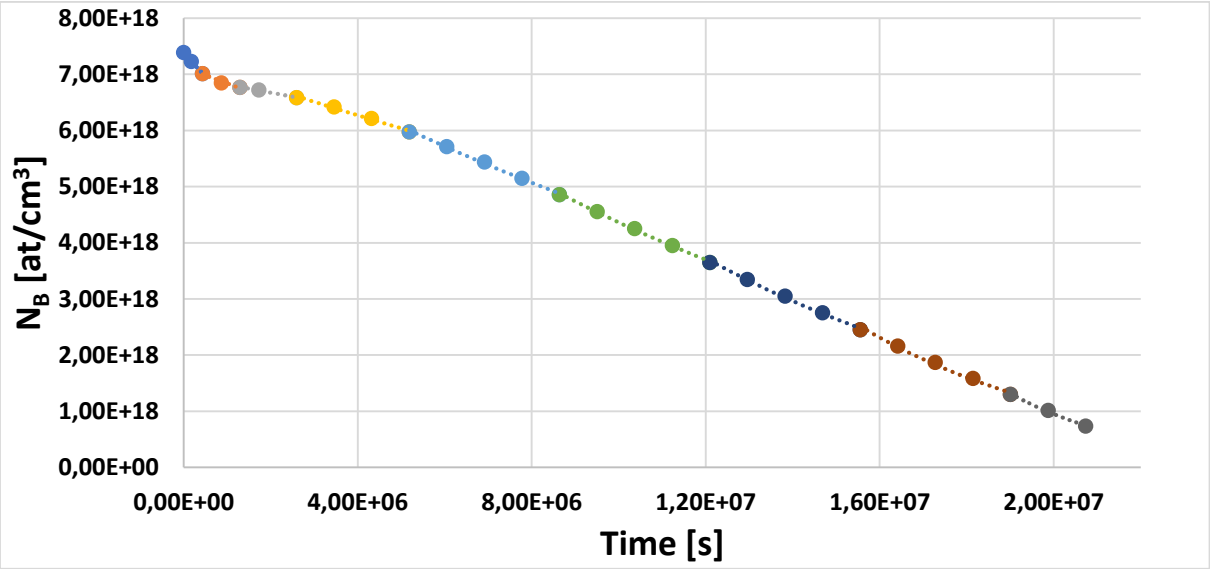


Figure 3. Concentration of ^{10}B in coolant through fuel cycle

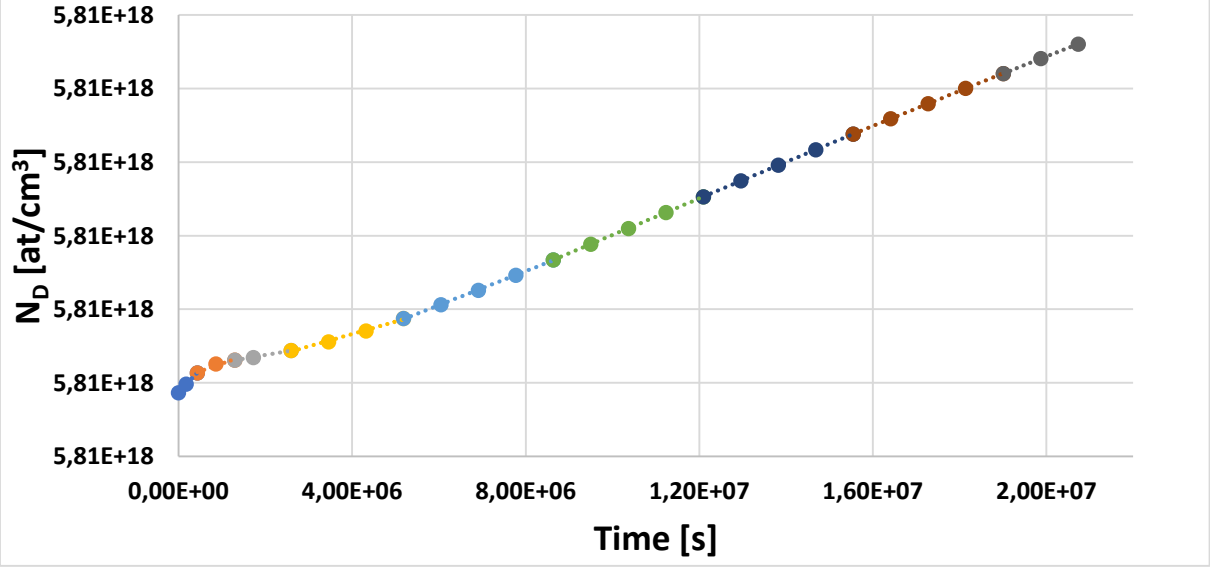


Figure 4. Concentration of ^{2}H in coolant through fuel cycle

In order to use these concentrations in the estimation of ^{3}H inventory, the concentrations should be substituted by simple functions that accurately represent the number density of the sought atoms at the given time step. Since the functions of concentrations of ^{1}H, ^{2}H and ^{10}B, ^{11}B atoms are relatively complex the fuel cycle (0 – 266.95 EFPD) has been divided to 9 intervals while a simple exponential fit was used for each interval.

2. 4. Calculation of tritium inventory in coolant

The calculation of tritium inventory in the first fuel loading of Mochovce NPP unit 3 was performed in the KENO-VI module of the SCALE 6.1.3 system, based on assumption presented in the previous chapters. SCALE6 is a comprehensive modelling and simulation suite developed for nuclear safety analyses by the Reactor and Nuclear Systems Division (RNSD) of the Oak Ridge National Laboratory (ORNL). The KENO-VI is a Monte Carlo criticality solver designed to calculate the k_{eff} and other quantities of three-dimensional systems. The KENO-VI criticality calculations were performed using 238g cross-section libraries based on ENDF/B-VII.0 [8] evaluated data and with cell treatment using the CENTRM/PMC sequence.

The analysis was split into several calculation steps, while the steps represented the reactor operation between 0-5, 5-15, 15-30, 30-60, 60-100, 100-140, 140-180, 180-220 and 220-266.95 EPFD. For each time bin a stand-alone calculation was carried out with the same reactor power, ERC position, temperatures, fresh fuel composition and variable coolant isotopic composition. The isotopic composition of isotope $X_i(t)$ in coolant was calculated using Eq. (1) and the dilution coefficients that were derived from the exponential functions in Figure 3 and Figure 4. The neutron flux was estimated to be constant within a given calculation step.

$$X_i(t) = a_i . e^{b_i t} \tag{1}$$

To estimate the isotopic composition of tritium, 3 production routes were considered:

- Eq. (2) - (n,α) on $^{10}_{5}B$ and subsequently (n,nα) on $^{7}_{3}Li$ - 21 kbarn * 50 mbarn thermal XS
- Eq. (3) - (n,t2α) on $^{10}_{5}B$ – 0.4 barn thermal XS
- Eq. (4) - (n,γ) on $^{1}_{1}H$ and subsequently (n,γ) on $^{2}_{1}H$ – 28 barn * 40 mbarn thermal XS

$$^{10}_{5}B + ^{1}_{0}n \xrightarrow{(n,\alpha)} ^{4}_{2}He + ^{7}_{3}Li + ^{1}_{0}n \xrightarrow{(n,n\alpha)} 2 * ^{4}_{2}He + ^{3}_{1}H + ^{1}_{0}n \tag{2}$$

$$^{10}_{5}B + ^{1}_{0}n \xrightarrow{(n,t2\alpha)} 2 * ^{4}_{2}He + ^{3}_{1}H \tag{3}$$

$$^{1}_{1}H + ^{1}_{0}n \xrightarrow{(n,\gamma)} \gamma + ^{2}_{1}H + ^{1}_{0}n \xrightarrow{(n,\gamma)} ^{3}_{1}H + 2 * \gamma \tag{4}$$

Considering the abovementioned reactions and the radioactive decay of tritium with 12.3 years half-life, the tritium balance equation can be written as follows:

$$\frac{dN_{^{3}_{1}H}(T)}{dT} = \sigma_{n,n\alpha}\varphi N_{^{7}_{3}Li}(T) + \sigma_{n,t2\alpha}\varphi N_{^{10}_{5}B}(T) + \sigma_{n,\gamma}\varphi N_{^{2}_{1}H}(T) - \lambda_{^{3}_{1}H} N_{^{3}_{1}H}(T) \tag{5}$$

where $N_{^{3}_{1}H}(T)$, $N_{^{2}_{1}H}(T)$, $N_{^{1}_{1}H}(T)$ $N_{^{7}_{3}Li}(T)$ and $N_{^{10}_{5}B}(T)$ are the time dependent concentration of ^{3}H, ^{2}H, ^{3}H, ^{7}Li and ^{10}B in the unity of volume, $\lambda_{^{3}_{1}H}$ the ^{3}H decay constant $\sigma_{n,n\alpha}$, $\sigma_{n,t2\alpha}$ and

$\sigma_{n,\gamma}$ are the microscopic cross-sections of (n,α), (n,t2α) and (n,γ) reactions and φ is the corresponding neutron flux. The time dependent concentrations of ^{7}Li and ^{2}H can be defined by Eq. (6) and Eq. (7), where ${N_{^2_1H}}^{DIL}(t)$ represents the concentration of ^{2}H as result of moderation dilution and ${N_{^2_1H}}^{ABS}(t)$ as result of (n,γ) reaction on ^{1}H atoms.

$$\frac{dN_{^7_3Li}(T)}{dT} = \sigma_{n,\alpha}^{^{10}_5B}\varphi N_{^{10}_5B} - \sigma_{n,abs}^{^7_3Li}\varphi N_{^7_3Li} \tag{6}$$

$$N_{^2_1H}(t) = {N_{^2_1H}}^{DIL}(t) + {N_{^2_1H}}^{ABS}(t) \tag{7}$$

The ${N_{^2_1H}}^{ABS}(t)$ ^{2}H component can be calculated by Eq. (8) and the ${N_{^2_1H}}^{DIL}(t)$ component of ^{2}H and the concentrations of ^{10}B and ^{1}H can be calculated by Eq. (1).

$$\frac{dN_{^2_1H}(T)}{dT} = \sigma_{n,\gamma}^{^1_1H}\varphi N_{^1_1H} - \sigma_{n,\gamma}^{^2_1H}\varphi N_{^2_1H} \tag{8}$$

In the above mentioned equations $\sigma_{n,y}^{X}\varphi$ represents the sum of the group-by-group flux weighted collision rate of nuclide X and reaction (n, y), which can be defined by Eq. (9), where $\overline{\varphi(g)}$ is the volume integrated average neutron flux, defined by Eq. (10).

$$\sigma_{n,y}^{X}\varphi \; = \sum_{g=1}^{G} \overline{\varphi(g)}.\,\sigma_{n,y}^{X}(g) \tag{9}$$

$$\overline{\varphi(g)} = \int_V \varphi_{V_i}(g)dV\,;\; \sum_{i=1}^{N} V_i = V \tag{10}$$

In each calculation step the concentration of ^{3}H was estimated using the calculated neutron flux, weighted XS and concentrations of nuclides in the coolant. However, the concentrations of the coolant changes also during a single calculation step, thus this change has to be taken into account, for instance by assuming constant neutron flux and the time integral of number densities through the time step. Since the coolant circulates with 2.5 m/s velocity, it takes approximately 1 s to go through the fuel core, to get activated by thermal neutrons, and additional 17 s to run through the rest of primary circle (PC) and to come back to the core. The activation of ^{3}H can be calculated in two ways, by taking into account the circulation time (t = in-core time) and the total volume of coolant in the primary circuit or by neglecting circulation (t = in-core + out-of-core time) and using only the volume of moderator in the core.

2. 5. Results

The results of the tritium concentration are presented in Figure 5, where CASE 1 represents the conditions where only the in-core time was taken into account and CASE 2 the conditions where the total-time (in + out) was considered. The atom concentrations were achieved by multiplying the result achieved by solving Eq. (5) by the neutron source term (4.523E19 n/s). The results of total and volumetric tritium activities at the end of fuel cycle (266.95 EFPD) are shown in Table 1. The volume of moderator for CASE 1 represents the total volume in PC and for CASE 2 the volume in the core.

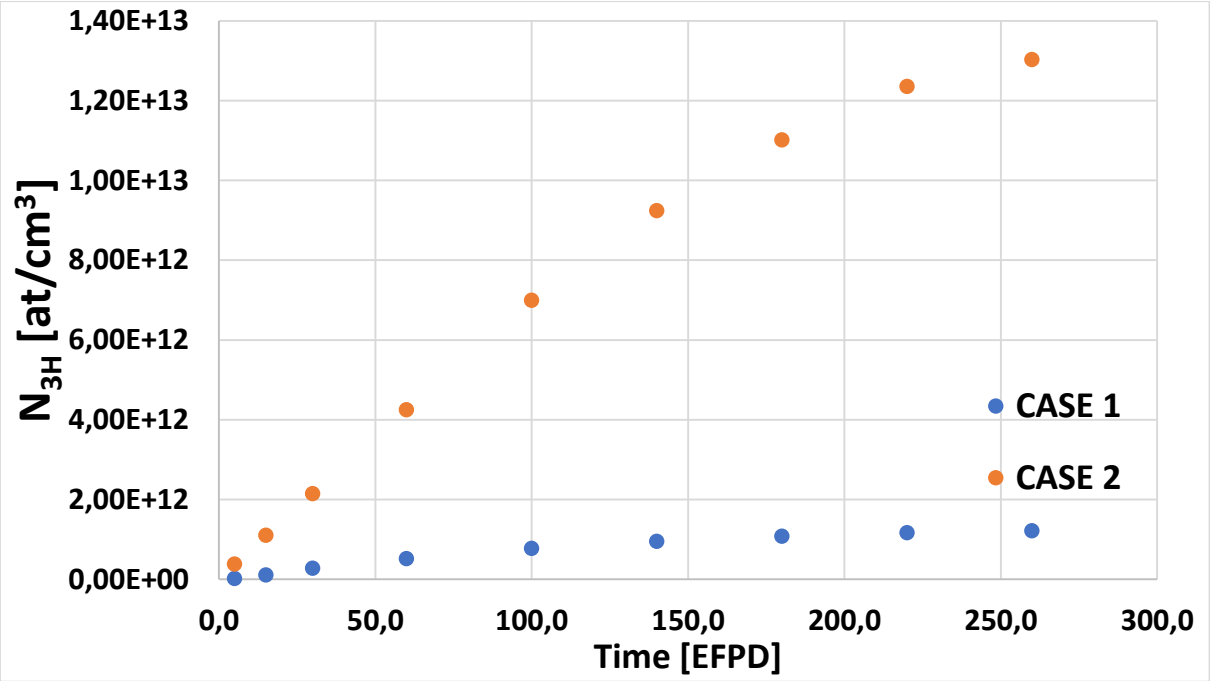


Figure 5. ^{3}H number density through fuel cycle

Table 1. Tritium activity at the end of fuel cycle

Coolant assumption	Circulation	No circulation
Decay constant [s^{-1}]	2.5638E-09	
Moderator volume [m^3]	242.00	12.77
Total activity [Bq]	7.6107E+11	4.4917E+11
Volumetric activity [Bq/l]	9.6403E+06	5.6895E+06

2. 6. Discussion

The number density and the activity of ^{3}H is strongly influenced by coolant circulation. The difference in the total ^{3}H activity is more than 40 %. Due to decreasing boron concentration saturation of the curve of ^{3}H number density can be seen. Among all reactions 87.95 % occurred on ^{10}B, 11.57 % on ^{2}H and 0.48 % on ^{7}Li. In CASE 1 the total volumetric activity of ^{3}H reached 9.64 MBq/l and in CASE 2 5.69 MBq/l. Just for better imagination, this activity is more than 28 000 or 56 000 times higher than the international limit for ^{3}H in drinking water [9]. According to document [10] published in 2018, the total released activity of tritium per one reactor unit is 2441.5 GBq. Compared to our results this value is approximately 3 times higher. However, considering the difference in the critical boric acid concentrations, enrichment, fuel cycle length and the rough estimation of coolant circulations, the results are acceptable.

3. CONCLUSION

In this paper the results of the first approach to estimate the tritium production rate in the Slovak NPPs was presented using SCALE6, the ANDREA diffusion solver and several in-house c++ utilities. The estimation of the tritium inventory was performed assuming fresh fuel isotopic composition, reference core parameters, and various critical boric acid concentration, calculation by the ANDREA code. Three nuclear reactions were taken into account, ^{10}B (n,α) followed by ^{7}Li (n,nα), ^{10}B (n,t2α) and ^{1}H (n,γ) followed by ^{2}H (n,γ). The change of the moderator isotopic composition was assumed by using an exponential fit through a calculation step. The total tritium activity was assumed either by considering the coolant circulation time with the total coolant volume (reference case) or by neglecting circulation and using only the

volume of coolant in the model. In the reference scenario the volumetric activity of tritium at the end of fuel cycle reached 9.64 MBq/l. Among this activity 87.95 % was produced due to the ^{10}B (n,t2α), and 11.57 % by ^{2}H (n,γ) reactions, thus in further steps it will be justified to neglect the minor tritium producing reaction on ^{7}Li. In the following steps of this research activity several improvements will have to be done. First of all, it will be necessary to use a more complex exponential fit of number densities for coolant dilution. It will be required to better assume the circulation time through the core and the neutron flux that contributes to tritium activation. In addition, fuel burnup will also have to be taken into account. After having our methodology improved, it will be implemented in the complex calculation system to optimize the fuel loading pattern of VVER-440 reactors in Slovakia.

4. ACKNOWLEDGEMENT

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