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Development of numerical models for Monte Carlo simulations of Th-Pb fuel assembly

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Abstract. The thorium-uranium fuel cycle is a promising alternative against uranium-plutonium fuel cycle, but it demands many advanced research before starting its industrial application in commercial nuclear reactors. The paper presents the development of the thorium-lead (Th-Pb) fuel assembly numerical models for the integral irradiation experiments. The Th-Pb assembly consists of a hexagonal array of ThO_2 fuel rods and metallic Pb rods. The design of the assembly allows different combinations of rods for various types of irradiations and experimental measurements. The numerical model of the Th-Pb assembly was designed for the numerical simulations with the continuous energy Monte Carlo Burnup code (MCB) implemented on the supercomputer Prometheus of the Academic Computer Centre Cyfronet AGH.

1 Introduction

From one side, the development and design of any research or commercial nuclear system need numerous precise neutron transport and burnup calculations for the prediction of its behaviour under radiation field. From the other side, the numerical tools, nuclear data and models applied in these calculations, should be validated experimentally so that the results will be credible. Such validation must be done on a small scale compared to the real nuclear power systems, for instance by means of assemblies designed for dedicated integral experiments.

This paper presents the first step of a detailed calculations being performed before to conduct an integral irradiation experiment on a thorium-lead (Th-Pb) nuclear system, i.e. it shows the procedure applied for the development of numerical models for the High Performance Computing (HPC) using Monte Carlo methods. The presented numerical model reproduces the Th-Pb fuel assembly available at the AGH University, Faculty of Energy and Fuels, Department of Nuclear Energy, Krakow, Poland. The Th-Pb assembly consists of ThO_2 fuel rods manufactured in Bhabha Atomic Research Centre (BARC), India [1] and metallic Pb rods manufactured at AGH University. The design of the assembly allows different combinations of rods for various types of irradiations and experimental measurements. As neutron source the ^{252}Cf isotope has been foreseen since it is easily accessible [2]. Moreover, its energy spectrum, comparing to the other radioisotope sources,

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is the most similar to the fission energy spectrum of main fissionable isotopes and may be approximated by the Watt fission spectrum [3]. The lead was selected as a neutron reflector because of its numerous advantages, such as: low neutron absorption cross section, high atomic mass and thus little thermalisation, neutron multiplication capability in the (n,2n) reaction and perspectives of application as a coolant in the future fast nuclear energy systems [4]. The numerical model of the Th-Pb assembly was designed for the numerical simulations with the Monte Carlo continuous energy Burnup code (MCB) [5] implemented on the supercomputer Prometheus of the ACK Cyfronet AGH [6]. The design of a detailed numerical model of the Th-Pb fuel assembly is necessary to perform reliable simulations in the following scientific areas preceding experimental measurements:

- neutron spectrum and flux density distribution for different configurations of assembly, showing the influence of material composition on those characteristics,
- expected activity of thorium rods and other samples, for the selection of proper irradiation conditions,
- neutron induced transmutations and formation of new isotopes, especially fissile ^{233}U,
- fission and radiative capture reaction rates on various isotopes presented in the initial and irradiated fuel.

Section 2 briefly introduces the background of the thorium fuel cycle. Section 3 presents the MCB code used for numerical simulations. Section 4 shows the engineering geometry of the Th-Pb assembly while Section 5 the designed numerical model. The study is concluded in Section 6.

2 Thorium fuel cycle

The existing nuclear reactors operate on the uranium-plutonium fuel cycle (U-Pu), where the fission reaction mostly occurs on the naturally occurring ^{235}U and bred ^{239}Pu. However, the fuel cycle can be also based on the thorium (^{232}Th), which natural abundance is about 3 to 4 times larger than uranium [7]. The thorium itself cannot undergo nuclear fission but serves as a fertile isotope for the production of fissile ^{233}U. Thus, thorium fuel has to be combined with the fissile isotopes like ^{233}U, ^{235}U or ^{239}Pu. The thorium-uranium (Th-U) fuel cycle is a promising alternative against U-Pu fuel cycle, but it demands many advanced research before starting its industrial application in commercial nuclear reactors [8]. The leading country conducting research on the thorium fuel cycle is India due to the large resources of thorium and limited resources of uranium. India postulates implementation of a three stage nuclear power programme linking the closed fuel cycles of Pressurized Heavy Water Reactors (PHWR), Liquid Metal-cooled Fast Breeder Reactors (LMFBR) and Advanced Heavy Water Reactors (AHWR) [7]. In the first stage PHWR will utilize natural uranium fuel and thus produce plutonium and depleted uranium. Next, the depleted uranium and plutonium will fueled LMFBR with thorium blanket. In this way ^{233}U will be bred from ^{232}Th and ^{239}Pu from ^{238}U. The bred ^{239}Pu will continuously fueled LMFBR while ^{233}U will be used in the last step to fuel AHWR with Th component for continuous breeding of ^{233}U. Therefore in equilibrium state AHWR will operate on ^{233}U and will be fuel just with Th. The main advantages and drawbacks of the Th-U fuel cycle comparing to the traditional U-Pu fuel cycle are presented below:

Advantages:

- Good chemical and physical properties,
- Lower inventory of minor actinides and plutonium in spent fuel,
- Proliferation resistance (^{232}U contamination of irradiated fuel),
- Possibility of breeding fuel in thermal reactors and fast reactors,
- Thorium is more abundant, safer and more effective in mining.

Disadvantages:
• Neutron source (other fuel) required to produce fissile ^{233}U from ^{232}Th,
• Difficult and costly fuel fabrication (high γ radiation from ^{232}U),
• Relatively long time of ^{232}Th breeding to ^{233}U,
• Likelihood of using ^{233}U for military purposes,
• R&D and licensing needed before commercial implementation.

3 MCB code

Generally, in Monte Carlo methods a physical system is described by the probability density functions, from which the sequences of random numbers are sampled and used for numerical simulation. The sampling process is repeated many times and events of interest are recorded by scoring functions, called tallies. The final results of the simulation are the averages over events of interest simulated in many trials and associated variations. The Monte Carlo methods in nuclear engineering allow simulation of the advanced nuclear systems with a heterogeneous material composition and complex geometry. The presented paper shows the procedure of design numerical models for the Monte Carlo Continuous Energy Burnup Code (MCB) developed at the AGH University, Faculty of Energy and Fuels, Department of nuclear Energy. The MCB code integrates the commercial Monte Carlo transport code A General Monte Carlo N-Particle Transport Code (MCNP) [3] and the novel Transmutation Trajectory Analysis Code (TTA) [9] at the level of the FORTRAN source code. The MCNP subroutines use Monte Carlo methods for neutron transport simulations while the TTA forms and analyzes transmutation and decay chains for nuclide density evolution in time function. All MCNP features are available in MCB, and some new ones were implemented. In the MCB calculations one could obtain the following parameters: neutron multiplication factor, nuclear reaction rates, potential doses, material activities, neutron flux and power distributions, evolution of material densities and many others. The MCB code was already applied in many national and international projects related to the design of innovative nuclear systems [10].

4 Engineering Geometry

Fig. 1A. presents the Th-Pb fuel assembly located in the radiometric laboratory at the AGH University. The assembly consists of three main components, i.e. support structure, lead reflector and core. The assembly is mounted on a bottom support plate with four wheels, which can move the set-up between chosen position in the laboratory, e.g.: position for preparation of assembly arrangement and insertion of neutron source, position for short-time irradiations and position for long-time irradiations with an additional shielding. The upper support consists of a cylindrical top plate, top bars and grid. The bars and grid determine the rectangular shape of the assembly core. The top and bottom support plates are connected by four side bars. All structural element were fabricated using standard construction steel. The assembly core is surrounded by the reflector mounted using rectangular Pb bricks designed especially for radiation shielding purposes. The bricks are jointed together by means of "V" shape wedges located on each side of them. The standard dimensions of the brick equal 100x100x50 mm (LWH). However, some bricks were cut into smaller pieces to fix them into current assembly arrangement.

Fig. 1B shows the assembly core composed of Pb and ThO_2 rods arranged in the hexagonal pattern with pitch of 13 mm and length of 1200 mm. The current pattern consist of 246 cylindrical ThO_2 rods and surrounding hexagonal Pb rods. The hexagonal shape of the Pb rods allows their compact package while cylindrical shape of the ThO_2 rods, easy replacement of Pb rods without dismantling the whole assembly. The pins on both ends of the rods allow for their precise placement in the bottom plate and fixing bars. The fixing bars made of Al (see Fig. 1A), serve as a support structure for the pins. The fixing bars freely settle on pins and can be easily removed if necessary. The adjacent rods and outer Pb reflector can be vertically extracted after having removed the Al bars. A gap among rods of about 0.4 mm allows for their easy removal and the modification of the set-up configuration. Seven most inner ThO_2 rods were replaced Six of them were replaced by six Pb rods for the intensification of a neutron flux due to (n,2n) reactions on lead and by a space in the center of the assembly used to insert the Cf neutron source. The use of hexagonal pins surrounded by plane made of Pb bricks will form some gaps at the interface between the bricks and Pb rods (see Fig. 1B). The influence of the gap on the measurements will be checked in the numerical simulations preceding the conduction of the integral experiments.

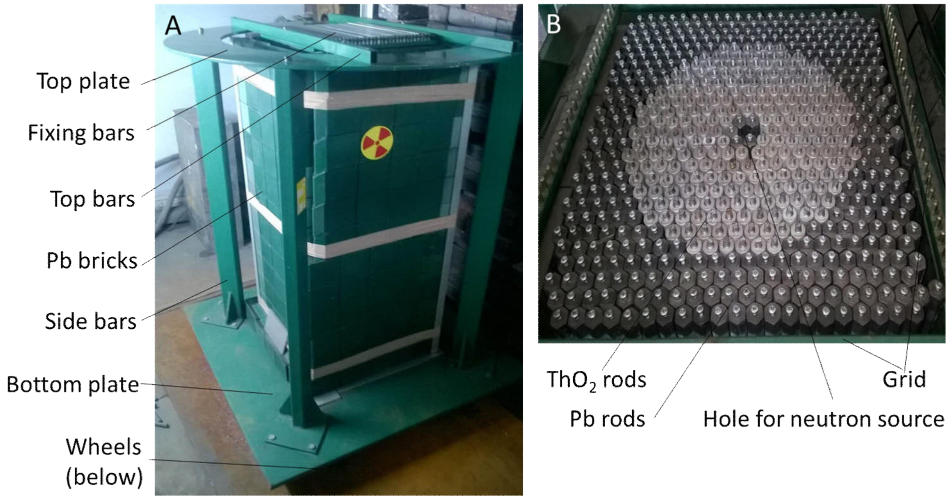

Fig. 1. Th-Pb assembly (A). The core of the assembly without aluminium fixing bars (B).

Fig. 2 shows both types of rods while Fig. 3 schema of the ThO_2 fuel rods manufactured in BARC. Unfortunately, the AGH University does not possess the mock-up of the thorium rod, which would allow the detailed geometrical measurements for the numerical modelling. The dismantling of the rods and extraction of theirs inner components would be troublesome due to the potential risk of alpha contamination as well as legal issues related to radiological protection. Thus, for the numerical modelling the specification of the rods from the available documentation was applied [11]. Table 1 present the main characteristic of the ThO_2 rods.

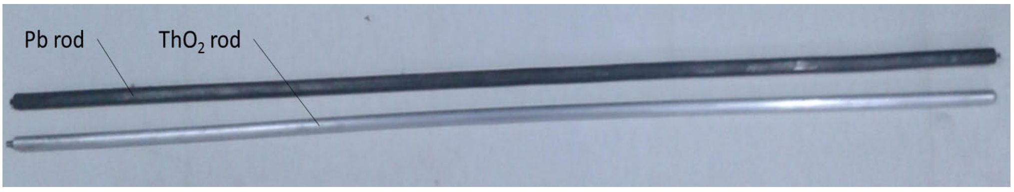

Fig. 2. Pb and ThO_2 rods.

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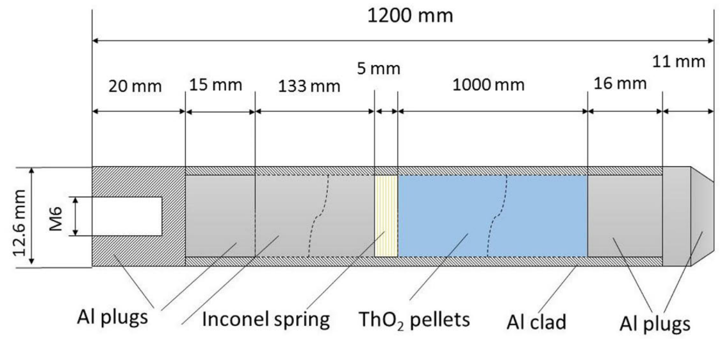

Fig. 3. Schema of ThO_2 rod.

Table 1. Characteristics of ThO_2 rod.

Mass of Th	~736 g
Mass of ThO_2	~836 g
Number of pellets	100
Pellet length	10 mm
Pellet diameter	1.064 mm
Gap thickness	0.13 mm
Clad thickness	0.85 mm
Rod diameter	12.6 mm

5 Numerical model

5. 1 Geometry

The procedure applied for the design of the Th-Pb assembly numerical model is shown in Fig. 4. Firstly, the engineering geometry has to be transferred to the computational geometry. To construct numerical geometry one needs to supply coefficients to the analytical surface equations to form a desired set of surfaces – step 1. Then using the intersection, union and complement operators it is possible to combine the surfaces into cells characterized by theirs volumes – step 2. It is also possible to define, a so called embedded geometry, which allows to build structures containing large arrays of the same elements. To do this, some cells have to be grouped together into entities called *universes*. The universes may be repeated into a square or hexagonal lattice arrangement.

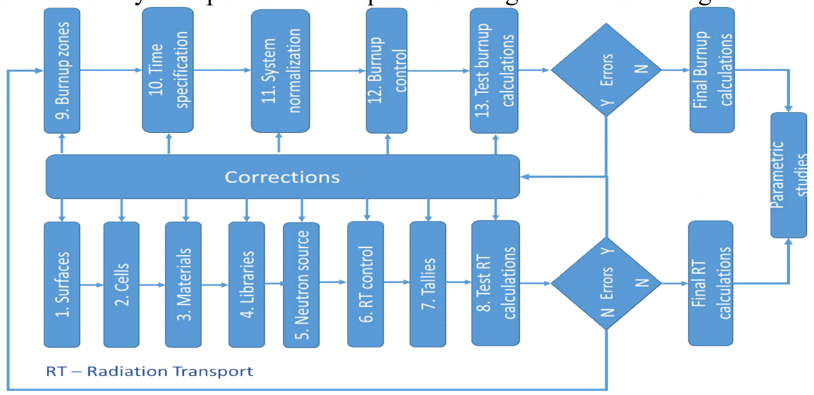

Fig. 4. Procedure applied for design of numerical models.

Fig. 5 shows the computational geometry of main structural elements of the Th-Pb assembly. The presented model is placed in the air cylinder (ϕ=1000 mm), which is surrounded by the void boundary conditions. Two main differences were introduced to the computational geometry comparing with the engineering geometry. Firstly, the wheels below the bottom plate were neglected. Secondly, the orientation of the side bars of the assembly was changed – i.e. the bars have been modeled with their faces parallel to the sides of the assembly. The numerical reconstruction of wheels and side bars rotation would demand transformation of the coordinate system and thus would complicate the numerical model and extend simulation time. Moreover, the changes are of minor importance for neutron transport and burnup simulation because the simplified elements are located outside the assembly core.

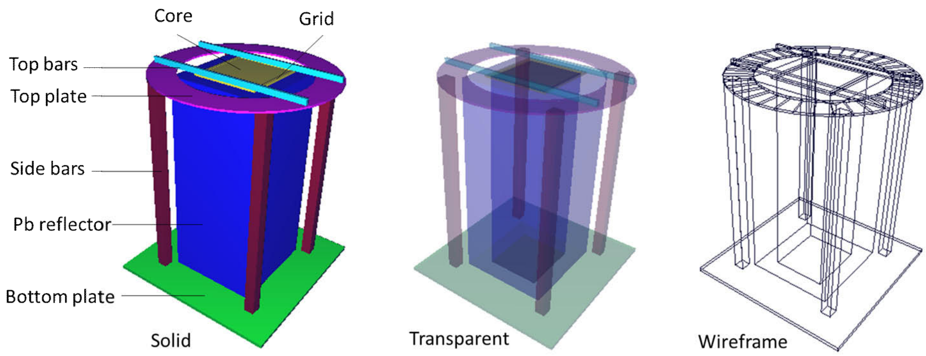

Fig. 5. Numerical model of the Th-Pb assembly.

Fig. 6 presents the cross sections of the assembly core with the Pb and ThO_2 rods and their surroundings. Both types of rods were numerically placed in the core using embedded geometry with the hexagonal lattice. Some simplification were introduced to the numerical geometry of the Th rods. The rods were divided into three axial sections namely, top of the rod, ThO_2 pellets and bottom of the rod. The upper section contains just Al filling, the hole for a M6 thread and homogenized material representing ICONEL spring (see Section 5.2). The ThO_2 in the middle section was not divided into particular fuel pellets but modeled as a one cylinder with a height of 1000 mm. The bottom section contains just compact Al plug without chamfering. The introduced simplifications will not influence the numerical results but significantly reduce the complexity of the model. The Al bars used to fix the rods, were modelled using embedded geometry but with the square lattice. The outer Pb neutron reflector was modelled as a compact cell surrounding the assembly core. The Californium neutron source was placed in the middle of the assembly – see section 5.4. The visual check of the model geometry is possible using dedicated plotting utility called Vised.

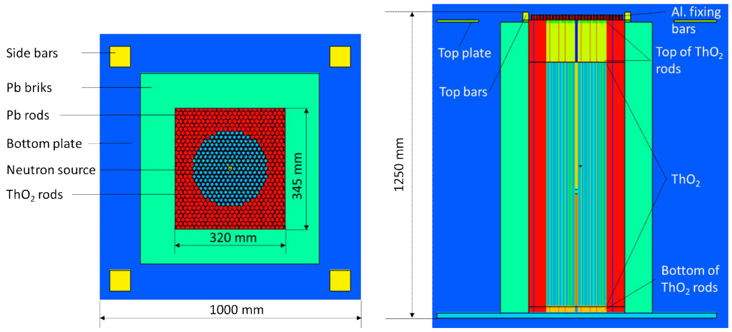

Fig. 6. Radial and axial cross-section of the assembly.

5.2 Materials, Libraries

The materials with specified densities, isotopic composition and nuclear data libraries should be assigned to each cell – step 3,4. They are represented in the input file using ZAID number where: Z means atomic number, A mass number and ID is a cross section library identifier. For instance, 90232.09c defines thorium 232 with 09c cross section library. The identifier of a library indicates the temperature, for which the cross section were evaluated, in this case 900 K. The most common nuclear data libraries available for the MCB numerical simulations are based on JEFF, JENDL and ENDF evaluations.

Usually, in the neutronic and burnup calculations one model only the most important components of the engineering geometry influencing the neutronic parameters, like neutron fluxes and spectra. Therefore, the engineering geometry does not need to be exactly transferred to the geometry of the numerical model. The less significant components are usually modelled as a homogenous mixes of various materials, which sufficiently reflects the physical behavior of the system. According to the visual analysis of the assembly the side bars, top bars and grid are empty inside, thus for the simplification they were modelled as a solid bars with the density and isotopic composition calculated for volumetric mix of inner air and surrounding steel. The numerical model is composed of eleven materials listed in Table 2.

Table 2. Materials composing numerical model [12,13].

Nr	Material	Density [g/cm^3]	Elements
1	ThO_2	9.4	Pellets
2	Cf_2O_3-Pd	12.023	Neutron source
3	Pb	11.35	Reflector bricks and rods
4	SS304L	8.0	Source capsules and holder
5	Aluminium	2.7	Fixing bars, plugs, clad
6	Steel	7.85	Plates
7	Air	0.001205	Free spaces
Nr	Homogenized materials (by volume)	Density [g/cm^3]	Elements
1	Steel 52% + Air 48%	4.08	Grid
2	Steel 31% + Air 69%	2.40	Side bars
3	Steel 13% + Air 87%	1.01	Top bars
4	Iconel 5% + Air 95%	0.4246	Spring

5.3 Neutron source, control, tallies

One of the most important issues in the numerical modeling of the Th-Pb assembly is the reconstruction of a neutron source – step 5. The source is characterized not only by the geometrical parameters but also by its intensity and energy spectrum. Moreover, the radioisotope neutron source degrades due to decay of neutron emitting isotopes and nuclear transmutation due to self-generated neutron field. All these effects should be taken into account in the numerical reconstruction of the neutron source. The Californium neutron source emitting neutrons in spontaneous fission was reconstructed numerically [14]. The source will be placed in the middle of the assembly as it is shown in Fig. 7. The intensity of the fresh source is about 10^8 n/s, which corresponds to the mass of about 43 μg ^{242}Cf. The source was modelled in the form of Cf_2O_3-Pd cermet wire embedded in two stainless steel capsules [15]. The energy spectrum of Cf source is similar to the fission spectrum, thus the Watt fission spectrum with special fitting parameters were used in the numerical modeling.

The numerical modelling with other radioisotope neutron sources like AmBe or PuBe is also planned.

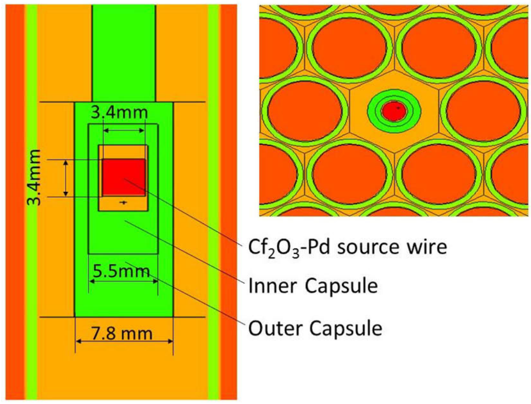

Fig. 7. Numerical model of Cf neutron source.

The main control parameters are related to the statistics of numerical simulations and thus precision of obtained results – step 6. The MCB simulation might be performed in the *kcode* mode, which provides calculation of neutron multiplication factor or in the *source* mode. The second option was chosen for the current calculations because the initial Th-Pb assembly does not contain any fissile material and is unable to induce self-sustaining fission chain reaction. The number of simulated neutron histories depends on the NPS parameter set in the input file. At the moment NPS values are quite small because the current models serves for the testing purposes. After final checkup of the model the NPS will significantly increase to the values allowing the high precision of results together with the acceptable computation time. In the MCNP/MCB modeling the additional large set of control parameters is available on request of the user [3]. These parameters are rather used for the specific types of analyses, which might be performed in additional calculations.

Recording some aspect of the average particle behavior is called tallying and tallies are the scoring functions used for tallying. Tallies provide summary information about the simulation of the physics problem. The standard type tallies can be in the input file or by the tally modification at the level of the source code. The MCB code needs particle fluxes for the estimation of the reaction rates, which in turn are necessary for calculation of the nuclide density time evolution. The numerical procedures lying behind, the so called F4 tally used to estimate the neutron flux, were implemented in the MCB code for the heating and reaction rates calculations. Thus, in the basic numerical model of Th-Pb assembly the specification of tallies in the input file is superfluous, because the quantities of interest will be automatically calculated by the MCB subroutines. However, in the future parametric analysis some tallies will be used for additional studies.

5.4 Burnup

The second stage in the design of numerical model, after test radiation transport calculations (step 8) is the implementation of the inherent MCB function for burnup calculations – step 9. The change in the fuel composition caused by the series of particle/neutron interactions and decays is described as fuel burnup. The detailed burnup analysis should predict the change in the isotopic fuel composition as the function of time and space. The control card BURN specified in the input file launches subroutines calculating the time evolution of nuclide density. The user is obliged to specify physical constraints over a transmutation system to start burnup calculations.

The geometry of the numerical model for neutron transport calculation is modified in order to divided fissile/fertile material into radial and axial burnup zones – step 9. In case of the Th-Pb assembly the thorium rods were divided into eight rings of ThO_2 rods around neutron source. Each ring in addition was divided into 20 axial zones of 50 mm length each, which gives in total 160 burnup zones. The number of burnup zones depends on the purpose of the modelling and may vary during the design of the experiments. For example, the number of rings probably will be decreased to seven, because of the use of six Pb rods around the Cf source for the intensification of neutron flux. Each burnup zone containing ThO_2 is distinguished assigning a different material number, doing so the burnup distribution of the whole assembly is obtained.

The time intervals for the reaction rates recalculations and thus the changes of material densities might be defined as a series of individual irradiation periods, or as cumulative values – step 10. For the current numerical design the irradiation time will not exceed 30 days divided into individual periods of a few days. Some additional actions related to the material processing may be activated at specified time. The system will be normalized to the source intensity of 10^8 n/s including the degradation of the neutron source due to the its decay and transmutation in every time step – step 11. The last step before burnup test calculations is related to the implementation of burnup control parameters, which determine efficiency and numerical solution of Bateman equations – step 12. For the initial calculations the standard set of burnup control parameters was used. The algorithmics used for burnup control is quite complex and it is not considered in this study [9].

6. Conclusions

The study presented the procedure developed for the design of the numerical model for the simulations of the Th-Pb fuel assembly. The design of the numerical model is the first step in order to start the integral irradiation experiments and initiation of R&D in the thorium fuel cycle at the AGH University. In the frame of this study:

- the detailed numerical model of Th-Pb assembly was designed,
- the advanced features of MCNP/MCB code were used to reconstruct real irradiation conditions,
- the model was tested in series of initial short-time and low-precision numerical simulations, to verify its credibility for modelling the irradiation experiments,
- it was shown that, designed model may serves as a basic model for a series of advanced numerical calculations and can be easily modified by the user,
- it was presented that, the development of the numerical models for the Monte Carlo simulations is a long-term task and demands detailed knowledge about engineering geometry, irradiation conditions, neutron source and other parameters influencing experimental measurements.

The future activities related to the modelling of Th-Pb assembly will focus on the modification of the numerical model for the detailed, high-precision calculations using HPC computers of ACK Cyfronet AGH. Afterwards, the numerical results of this unique will be used as a basic proof of competences of the scientific team and to boost the AGH team, towards participation in scientific projects related to thorium fuel cycle. The scientific collaboration is necessary for the knowledge exchange and useful to promote he possibility to receive funds for radiometric laboratory following to the establishment of the advanced radiometric measurement facility.

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