BERYLLIUM POISONING MODEL FOR RESEARCH REACTORS

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ABSTRACT

Following paper presents current studies on beryllium block poisoning based on beryllium moderated research reactor MARIA. Description of the problem was given along with calculation and measurement methodology. At the moment the experimental campaign is still under development.

1. Introduction

Beryllium is one of the lightest metals having unique nuclear properties. It has relatively low thermal neutron absorption cross section (around 1mbarn at 0,625eV [6]) which makes it a great material to be used as a neutron reflector, allowing to more efficiently utilize the neutrons during reactor operation. It is also very good moderator material, allowing more effectively sustain fission process.
Beryllium undergoes number of reactions during a nuclear reactor operation. Reaction chains of interest due to neutron and gamma environment in a reactor are presented on figure 1. The effect of beryllium's exposure to above reactions is build up of few isotopes.
Helium (^{3}He) build up causes beryllium swelling and therefore changes of its mechanical properties, which is significant limitation in the beryllium operating lifetime. As a result, periodic replacement of blocks is required.

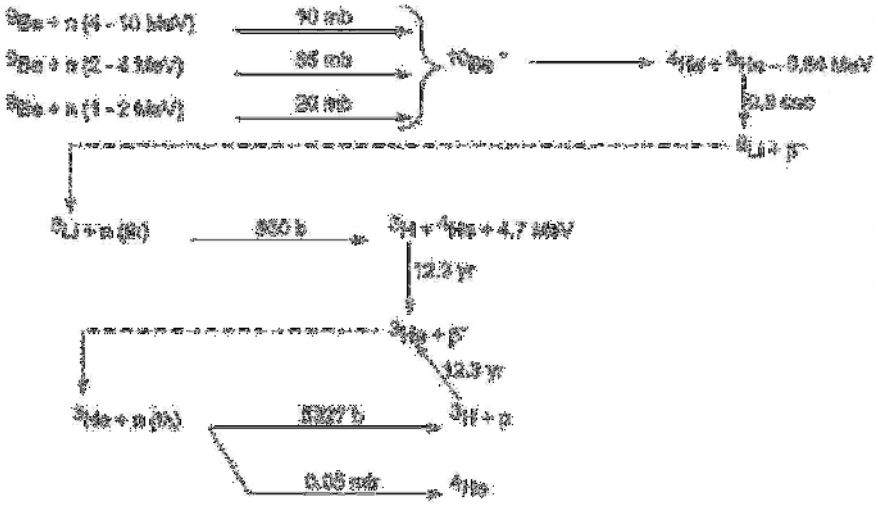

Fig. 1 Main reaction chains from which poisons are generated.

Another aspect directly affecting reactor operation is related to gradual build-up of reaction products of high neutron absorption cross section. The most significant so-called poisons are lithium ($\sigma_{Li\text{-}6\ a}\approx$ 940b) and helium ($\sigma_{He\text{-}3\ a}\approx$ 5300b). These isotopes are generated not only during the reactor operation but also reactor outage. Especially long periods of outage cause significant increases in ^{3}He concentrations, being result of radioactive decay of ^{3}H.

High concentrations of these isotopes result in large negative reactivity, flux and power distribution changes. The damage in material is mainly dependent on the neutron flux, neutron energies and duration of the exposure. In addition, diffusive release of tritium from irradiated beryllium cause particular problem for storage of irradiated beryllium blocks. For all these reasons having more accurate information about the poisoning effect seems to be crucial for better calculation and prediction of reactor operational parameters.

2. Problem description

The poisoning effect has been observed in the MARIA reactor after a seven years' break in its operations.

Located and operated in National Centre for Nuclear Research in Poland, MARIA is a high flux, pool type, water and beryllium moderated material testing reactor (MTR) with graphite reflector. It is using beryllium matrix as a moderator. Fuel channels are situated in a matrix containing beryllium blocks and enclosed by lateral reflector made of graphite blocks in aluminium cans (fig. 2). At the present MARIA is operated using 19.75% U-235 mass enriched (MC-5) fuel.

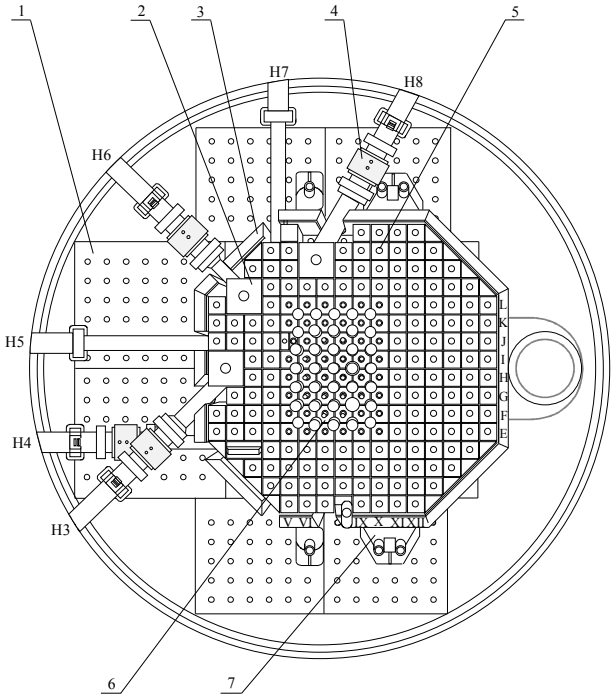

Fig. 2 Horizontal cross section of the reactor

Although actually used computational models satisfy the prediction needs of reactivity coefficients and variations in multiplication factor during irradiation, there is no actual model of poisons spatial distribution development in exploited beryllium. The knowledge of poisons' spatial distribution not only could positively influence core physical characteristics description but also help in a more accurate operation parameters prediction. Moreover detailed information about poisons distribution has a potential use in optimizing reactor configuration.
Typical cycle of MARIA reactor consists of 100 hours of operation, followed by 68 hours shutdown period. There is 30 to 40 operation cycles per year, with a power varying from 30kW to 20MW.

Proper implementation and qualification of the beryllium depletion in a deterministic neutronics calculations as well as improvement of currently used calculation scheme in MARIA core calculations, require a preparation of simple structure calculation and analysis first.
Since nuclear calculations are usually performed in two steps : lattice calculation in a two-

dimensional (2D) infinite arrangement of fuel rods or assemblies and core calculation in a three-dimensional (3D) whole core, following codes were chosen :

a) APOLLO2 transport code for single assembly surrounded with Be blocks in infinite lattice calculation, based on currently used computational scheme
b) MCNP Monte Carlo code used as a reference to validate transport model
c) SERPENT2 Monte Carlo code a second validation code

In order to prepare the experiment, preliminary study and calculations were carried. To increase reliability of results, two codes were used: MCNP associated with JEFF3.1 nuclear data library and SERPENT2 also associated to JEFF3.1 as well as APOLLO2. Few configurations of performing the experimental measurements were considered.

3. Codes and methods

3.1. APOLLO2

APOLLO2 is a 2D deterministic neutron transport code, developed by CEA, AREVA and EdF. It has a modular structure designed to solve Boltzmann transport equation in a multi-energy group scheme for unstructured geometries.

Two calculation methods were considered:

a) collision probability method using interface current approximation and multi-cell geometry, CEA2005 library v10.4 for the X-MASS 172 group structure and Pij solver. In this concept, collision probabilities can be performed using a spatial function distribution that describes a real condition inside the cell. Neutron flux spectrum in each region is different each other because of an existing of the spatial function. CP matrix is performed not only by the optical path length as an exponential parameter, but also the shape of the function that describes the behaviour of neutron flux spectrum in each region.

b) Method of Characteristics with linear surface numerical scheme, SHEM 281-group energy mesh. This method is based on two level approach. First level is based on neutron energy spectrum calculated with 2D geometry using Pij solver in order to provide realistic self-shielded cross sections and local spectra. The second step is performed on the exact 2D geometry using unstructured meshes with MOC solver. The depletion calculations are carried out in this step.

3.2. Monte Carlo –SERPENT2 and MCNP

In Monte Carlo simulations, individual neutrons are tracked one at a time from emission to eventual removal by capture or leakage. How far the neutron moves before interacting with matter, how it interacts and what results from the interaction are all randomized based on known distributions taken from evaluated nuclear data libraries.
Since it is impossible to simulate every neutron in the physical system, an additional normalization condition (e.g., total power or flux density) is also required to determine how many real neutrons one simulated neutron corresponds to.
In Monte-Carlo modelling the geometry is 3D fully heterogeneous and the nuclear data are not tabulated with an energy cutting (multi group description) but have a continuous energy description.

4. Models and results

All calculations were initiated using fresh material compositions, considering 20 operating cycles of the reactor and the same geometrical parameters. Normalized power of 1MW in fuel element was assumed. In all cases reflexive boundary conditions were used. Due to the conical shape of block (fig. 3), an equivalent geometry representative of the middle of the block height was chosen for calculations. Fresh beryllium composition is given in the table 1.

Table 1 Beryllium composition

Isotope	10^{24}*At. Density	Isotope	10^{24}*At. Density
BE9	1.19E-01	MN55	3.99E-06
AL27	1.62E-05	CU63	1.79E-06
FE54	1.15E-06	CU65	7.98E-07
FE56	1.80E-05	NI60	4.90E-07
FE57	4.16E-07	NI61	2.13E-08
FE58	5.54E-08	NI62	6.79E-08
SI28	7.20E-06	NI64	1.73E-08
SI29	3.65E-07	NI58	1.27E-06
SI30	2.42E-07	C	1.37E-04
O16	1.23E-03	CR50	2.75E-07
MG24	1.78E-06	CR52	5.30E-06
MG25	2.25E-07	CR53	6.01E-07
MG26	2.48E-07	CR54	1.50E-07

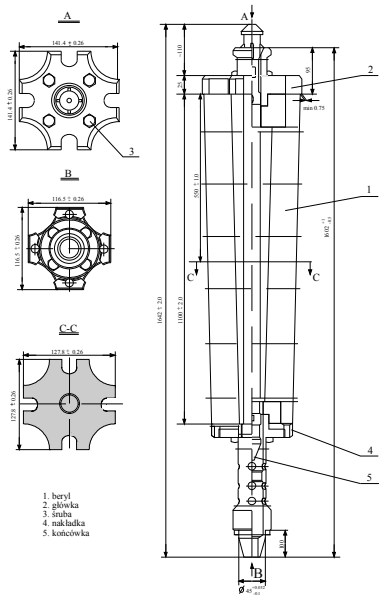

Fig.3. Beryllium block geometry

Typical calculation cell for MARIA consists of one fuel element and This cell is representative of a mesh the MARIA lattice

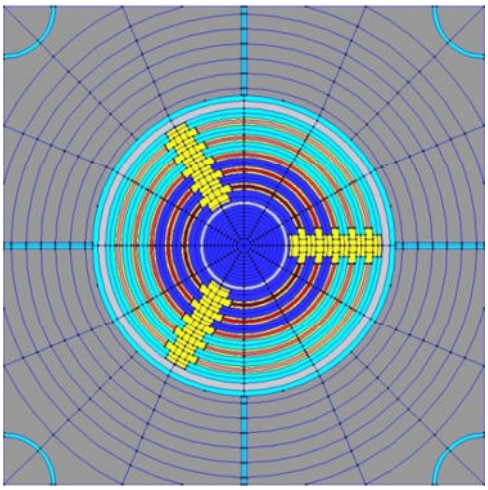
Fig. 4. Calculation cell with beryllium plugs and water gaps.

Calculations with APOLLO2, MCNP or SERPENT were carried out. In APOLLO2 calculations, various cases were investigated. Efficiency and precision tests of two solvers were made for reference (172 groups) and collapsed energy meshes (7 groups). Additionally study of the beryllium depletion chain importance in calculations and its influence on the resulting parameters was made. APOLLO2 data were compared with those of TRIPOLI4 (fresh fuel) and SERPENT (depleted fuel in order to validate calculation scheme. Below, some results are presented. Comparison of multiplication factor (fig. 5) for chosen solver and energy mesh shows that the maximum difference between APOLLO2 and MCNP equals 228 pcm, where comparing SERPENT2 and APOLLO2 the discrepancy varies from 100 pcm to 160 pcm.

The shape of the k_{eff} plot is due to the outage period between cycles, during which poisoning decreases.

The power distribution is given for each of the 5 plate of the assembly.

Example of power distribution in a first fuel layer can be found on fig 6. proving good agreement in results.

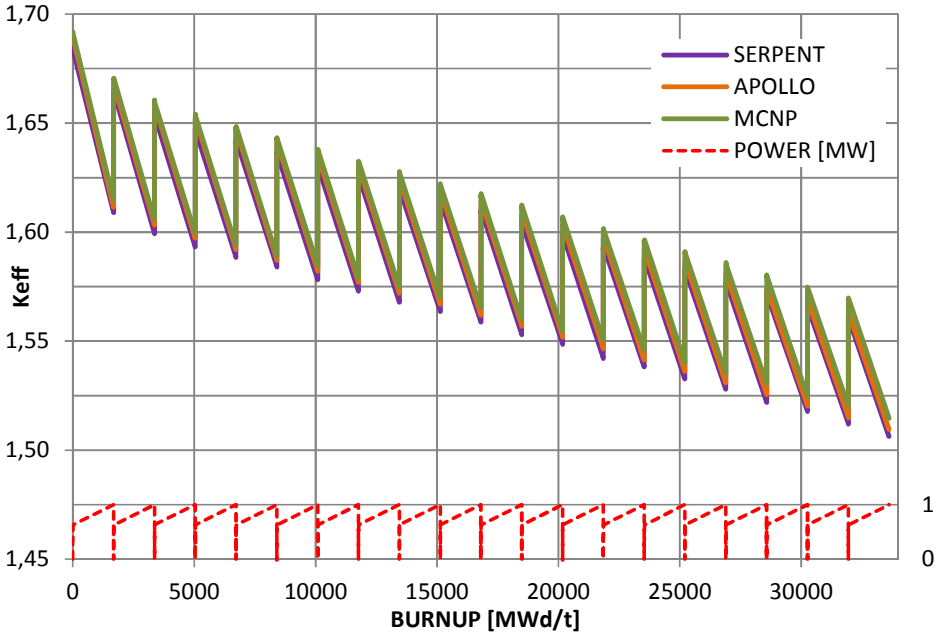

Fig. 5 Effective multiplication factor comparison

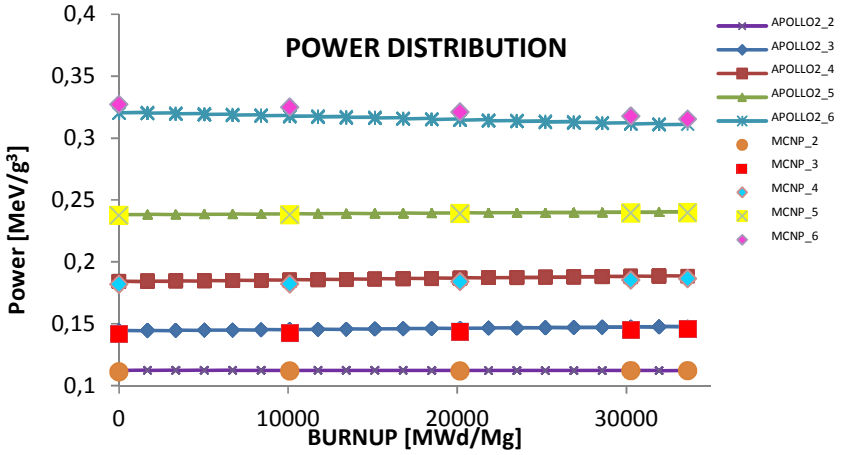

Fig 6. Power distribution comparison

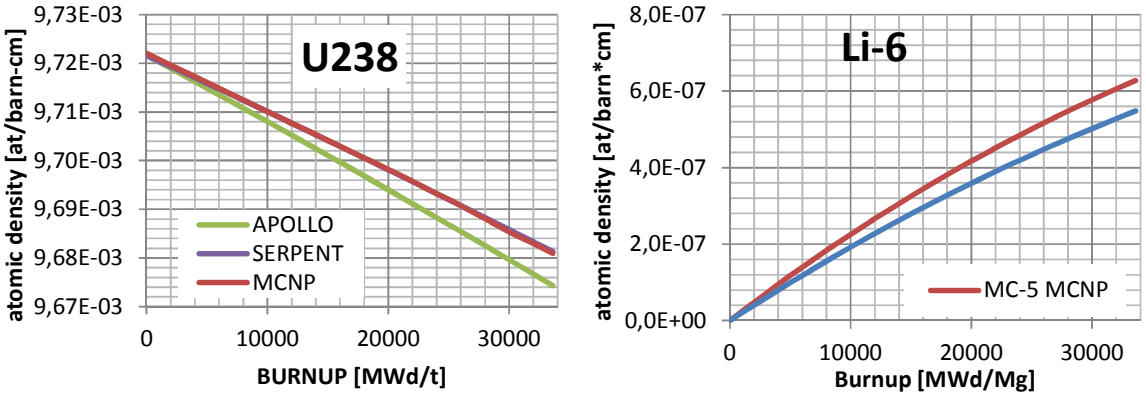

Fig 7. Atomic density change during operation for ^{238}U and ^{6}

Evolution of two isotopes concentration was presented on fig 6. showing some difference in atomic densities.

5. EXPERIMENT calculations

In order to carry out the experiment it is essential to perform very detailed forecasting analysis and calculations. Studies of possible measuring system configurations were made with predicted distribution of lithium, generated during use of beryllium in the reactor. Monte Carlo codes SERPENT and MCNP were used. To simplify primary calculations, following assumptions were made: non-conical case of beryllium block geometry, MR6/485 fuel, initially fresh material compositions. The use of MR6 fuel type instead of MC5 in this model allowed to avoid the problem with fuel elements' orientation. MR6 element is constructed out of fuel tubes (as it can be seen on fig 8.) whereas MC5 element consists out of fuel plates connected by aluminium ribs (as on fig 4.). The amount of ^{235}U of both elements is the same and equals485±5g with nominal length of 1000mm. Two types of meshes to determine poisons concentrations were taken into account.

Horizontal cross section of calculated geometry with four fuel elements (F) located in the corners of the block and aluminium plugs (G) in slots for control rods was presented on fig 8. Layers of beryllium block for which neutron flux and reaction rates were calculated were numbered with letters A-D. The other parts of the block are marked with letter E and numbers 6-9. All calculated values of lithium concentrations can be found in table 2 along with their representation on the diagram on fig 6.

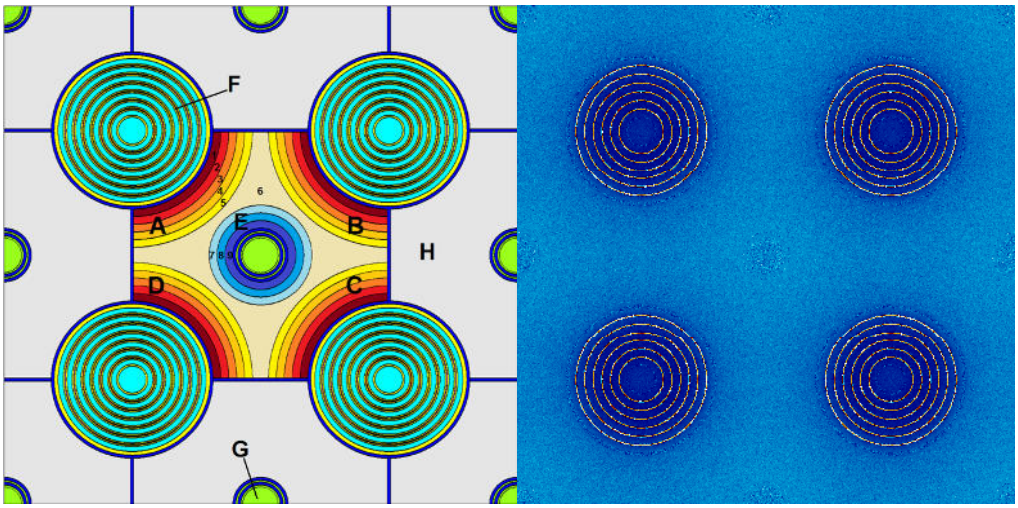

Fig 8. On the left cross section in xy plane of beryllium block with poison calculation zones. On the right representation of neutron flux distribution in energy range of 10^{-11}-10[MeV]

It was assumed that the beryllium block was surrounded by water. Reflection boundary conditions for x and y planes were used, and escape in the z plane. Reactor operations temperatures were used respectively for each material. Using neutron fluxes and reaction rates in equation. (1), lithium concentrations were calculated.

$$\boldsymbol{K}_{^6Li}(t_a) = \frac{m_{at}(^6Li)}{m_{at}(^9Be)} \cdot \frac{\sigma_f}{\sigma_{th}} \cdot \frac{\Phi_f}{\Phi_{th}} \left(1 - e^{-\sigma_{th}\Phi_{th}t_a}\right) [ppm] \quad (1)$$

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Where: m_{at} is atomic mass if the isotope, σ_f is a cross section of $^9_4Be(n,\alpha)^6_2He$ reaction on fast neutrons (Φ_f), σ_{th} neutron cross section of $^6_3Li(n,\alpha)^3_1H$ reaction on thermal neutrons(Φ_{th}) and $\boldsymbol{K}_{^6Li}$ is concentration of ^{6}Li in ppm after time t_α.

Values of lithium concentrations calculated with MCNP and SERPENT codes are presented on fig 9. In both codes ENDFB-VII nuclear data library was used. The relative statistical error on average equals 0.01. As it can be seen, SERPENT concentration values are slightly lower than MCNP results and the difference equals approximately 0.6%.
It is important to notice that lithium concentrations will change not only regarding the distance from fuel elements which are 10cm shorter than beryllium block, but also with the block height due to neutron escape at each end of the beryllium block.

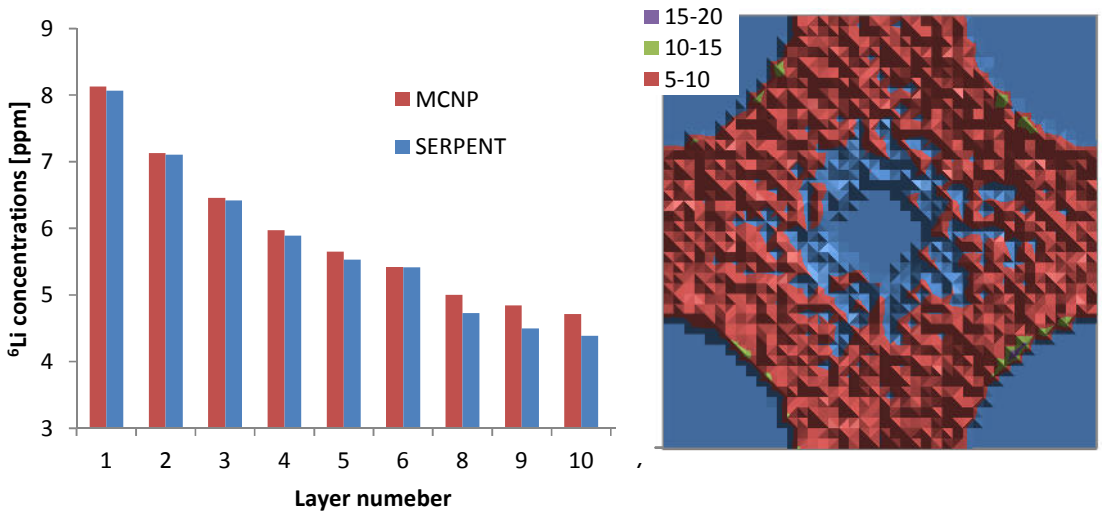

Fig 9. Comparison of calculated lithium concentrations. On the right average distribution in block calculated with SERPENT for a 0.3x0.3x10cm mesh.

Having estimated amounts of lithium in each zone of beryllium block, it is possible to calculate approximate time needed to conduct measurements with use of fission chambers.
For this purpose it is assumed that beryllium will be irradiated by a source of known properties. For this purpose a PuBe neutron source is used. It has a cylindrical shape, strength of $2 \cdot 10^7$ n·cm^{-2}s^{-1}. Detectors, fission chambers were used.
Few measuring systems were proposed with different detectors and neutron source position. Primary calculations were conducted for a fresh beryllium composition. As a second step, the amounts of lithium calculated previously were added respectively to each beryllium layer.

Currently proposed measuring system was presented on fig 10.

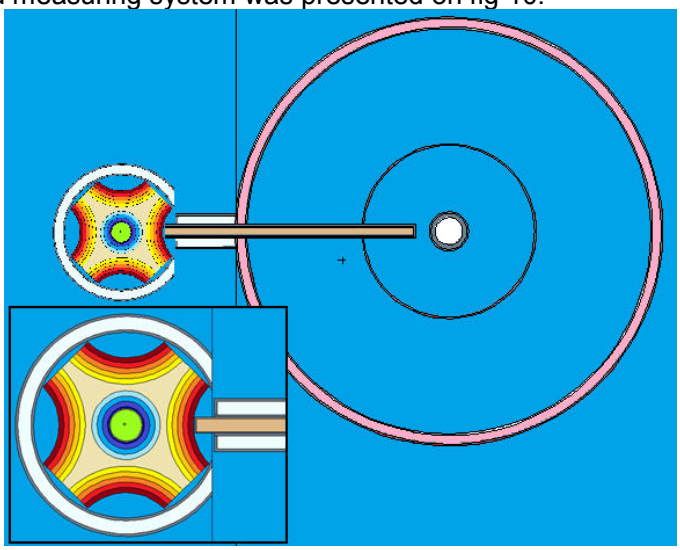

Fig 10. Measuring system

Detector is located inside the block and is marked with a green colour. Neutron source is placed inside a cylinder filled with water and surrounded with layer of B_4C. Collimator is inserted in the cylinder, and faces the block. Whole system is immersed in water. This construction allows for maximum thermalisation of fast neutrons in water and directing as many as possible to beryllium.

To perform measurements, it is necessary to know the time during which measurable amount of interactions in detector will occur. Assuming that the efficiency of the detector ε=0.3, it is possible to determine approximated number $\boldsymbol{I}$ of counts in the detector per time unit.

$$\boldsymbol{I} = \frac{\mathbf{N}}{\mathbf{t}} = \frac{\boldsymbol{\alpha \cdot \varepsilon \cdot s}}{\boldsymbol{\sigma_{U5}}} \quad [11] \tag{2}$$

Where:

$\boldsymbol{\alpha} = \int_{\boldsymbol{E_0}}^{\boldsymbol{E}} \boldsymbol{\sigma(E)\varphi(E)dE}$ is a reaction rate in the detector, s – neutron flux of a PuBe source $2 \cdot 10^7$ n/s, σ_{U5} – fission cross section for thermal neutrons in ^{235}U = 582.2 barns

Time t_p needed for measurements depends on the difference between number of counts in fresh (I_0) and poisoned (I_1)block, therefore the difference in counts is equal to:

$$\boldsymbol{x = I_0 - I_1 = \frac{N_0}{t_0} - \frac{N_1}{t_1}} \tag{3}$$

Assuming that measurement time will be the same in both cases, $t_0 = t_1 = t_p$, and expecting the uncertainty $(\Delta \boldsymbol{x})^2$ = 0.1, the measurement time equals:

$$\boldsymbol{t_p = \frac{I_o + I_1}{(I_o - I_1)^2 \cdot \left(\frac{\Delta x}{x}\right)^2}} \tag{4}$$

Using equation (4), values of neutron fluxes and reaction rates that were calculated in SERPENT and MCNP codes, the estimated number of counts in each detector was determined for both fresh and poisoned beryllium. Regarding those values, minimum measurement time was set. Depending on the size of the B_4C cylinder, and therefore volume of the water inside, length of the collimator and side of beryllium bloc maximum of the neutron flux was found in different distances. As for the moment maximum flux was calculated for 10 cm of collimator length and that length of collimator was chosen for the experiment. Comparison of thermal neutron fluxes for considered collimator lengths was given on fig 11. Based on all calculations, time for proposed measuring sets offers very promising values, varying from around 140 to 3000 s.

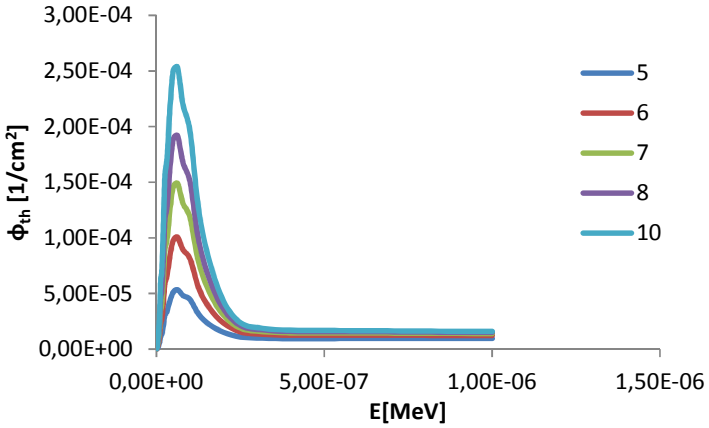

Fig 11. Neutron flux regarding different collimator length

6. Summary

As it was presented in above studies, various calculation and experimental systems were considered. Calculations and measurement method is still under development. Further analysis assume performing detailed calculation of optimal measurement system, performing the experiment with fresh beryllium and poisoned beryllium, that was used during the reactor operation. Measurements will be evaluation source for the calculations method in deterministic code. Some preliminary studies showed that even very low amounts of impurities in beryllium samples, e.g. Fe isotopes, activate the sample strongly enough not to allow for direct measurements. High purity beryllium needs to be used in order to avoid such problems. Measurements of the beryllium currently used in the reactor's core seems to be the best solution concerning not only its isotopic composition, but also due to the geometry.

7. References:

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