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Computations of fuel management in MARIA reactor with highly poisoned beryllium matrix

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Abstract. The high-flux research reactor MARIA has been operated in Poland since 1975. Its core consists of loop type fuel channels placed in a beryllium block matrix. Irradiation of beryllium by neutrons results in a build-up of ^{6}Li, and ^{3}He isotopes with large thermal neutron absorption cross-sections. In addition, tritium is formed and decays into ^{3}He which complicates the transmutation chains. Thus, the fuel management of the reactor depends on the beryllium poisoning. The isotopic transmutations in beryllium have to be computed in parallel to the fuel depletion. In this paper a comparison of the measured and computed results is given during: reactor operation in the period up to January 2004, modernization break in the reactor operation from January 2004 to February 2005 and reactor operation from February 2005. The measured and computed effects comprise: reactivity effects due to the fuel burn-up and beryllium poisoning by ^{6}Li, and ^{3}He.

Key words: criticality • excess reactivity • beryllium poisoning • isotope transmutation • nuclear reactor

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Introduction

The core of the MARIA reactor consists of tubular fuel elements placed in a matrix of beryllium blocks, cf. Fig. 1. Beryllium irradiated by neutrons with energies above 0.7 MeV undergoes (n,α) and (n,2n) reactions resulting in a subsequent formation of ^{6}Li, ^{3}H, ^{3}He and ^{4}He isotopes. The ^{3}He and ^{6}Li isotopes have large thermal neutron absorption cross-sections and their presence causes changes in the MARIA core operational characteristics, i.e. reactivity, flux level, its spectrum and fuel element power production.

As the experimental determination of ^{6}Li, ^{3}H and ^{3}He content in the reactor beryllium is unrealistic, a systematic computational determination of its quantities has been undertaken. The REBUS-3 code [6], based on diffusion approximation, is used at the IEA to calculate isotopic transmutations both in fuel elements and in beryllium blocks of the reactor. Microscopic cross-section library of Be has been produced using the WIMS-ANL code [5]. It was shown earlier [3] that in order to get reliable MARIA fuel management predictions it is necessary to follow both the history of fuel element, burn-up and individual beryllium block poisoning. In order to obtain poison levels in individual beryllium blocks, both periods of operation and operational breaks have to be represented in the calculations [1, 2].

The results of fuel management calculations include time evolution of:

1. Volume averaged poison accumulation in each beryllium block.

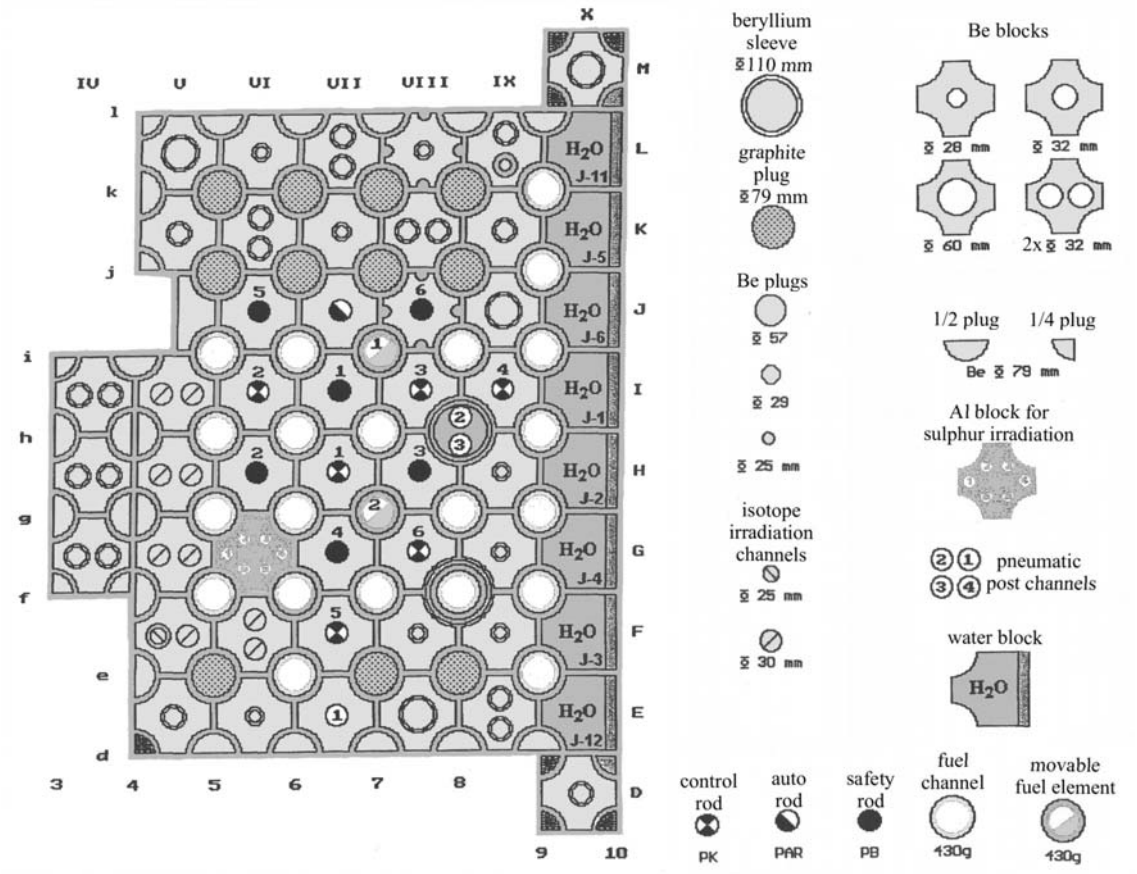

Fig. 1. The horizontal cross-section of MARIA reactor core.

2. Volume averaged fuel element composition.
3. Effective multiplication factor.

MARIA reactor core

A general layout of the MARIA reactor can be seen in Fig. 1. In the figure, the capital letters and Roman numerals are used as coordinates of beryllium blocks, while small letters and Arabic numerals as coordinates of the fuel channels.

The beryllium blocks have an effective length of 1100 mm and are 140 × 140 mm across at their higher and 120 × 120 mm at their lower end. It was shown that the average block transversal dimension 130 × 130 mm can be used in the MARIA core computations. Tubular fuel elements are placed in cooling channels, which, in turn, are placed in the 82 mm diameter cut-outs in the corners of beryllium blocks. The fuel elements consist of 6 concentric tubes with a fuel meat clad with aluminum. Two fuel enrichments have been used in the reactor – the older one with 80% enriched in ^{235}U since 1975 and the newer one with 36% enriched in ^{235}U from 2000. The dimensions of the fuel tubes were the same in both cases. Initially, the newer fuel elements with 540 g of U-235 per fuel element were used which necessitated relatively thin cladding. Since 2005, the fuel with 430 g of U-235 per fuel element has been used and the cladding is thicker.

The important feature of beryllium blocks are vertical cylindrical channels used for reactor control and isotope irradiation. There are five main types of beryllium blocks with different shape, number and radii of the channels. When not in use, they contain beryllium plugs. The beryllium matrix is surrounded by a reflector consisting of graphite blocks, similar in shape to that of beryllium, but without the cut-outs. A beryllium sleeve, 110 mm in diameter, occupies position f8 to make it possible the placement of objects with high diameter. Position h8 has been occupied since 1993 by a hydraulic rabbit system. At position G-VI/I-V, from December 2000 to January 2007 resided an aluminum block for sulphur-35 production.

The MARIA reactor operates in cycles of 100 h on-power and 68 h off-power. If necessary, 261/76 h cycles are also applied. The average fuel element power output during standard operation slightly exceeds 1 MW.

Computational model

Transmutations in beryllium blocks

The diagram of isotopic transmutations taking place during irradiation of beryllium by neutrons are shown in Fig. 2.

In reactor practice the very fast β^- decay of ^{6}He into ^{6}Li can be neglected, and ^{6}Li is considered as being directly produced from ^{6}He. Of strong interest is the formation of the two highly absorbing isotopes: ^{3}He and ^{6}Li. It is worth noting that during breaks in operation ^{3}H decays into ^{3}He, and the process is reversed during reactor operation.

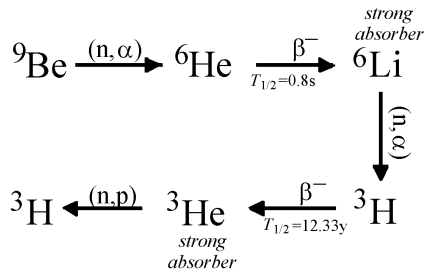

Fig. 2. Isotopic transmutations in beryllium irradiated by neutrons.

Thus, the equations describing isotopic transformations in beryllium during irradiation by neutrons in the REBUS code look as follows:

$$
\begin{aligned}
\frac{dN_{\mathrm{Be}}}{dt} &= -N_{\mathrm{Be}} \cdot RR_{\mathrm{Be(n,\alpha)}} - N_{\mathrm{Be}} \cdot RR_{\mathrm{Be(n,2n)}} \\
\frac{dN_{\mathrm{L}}}{dt} &= N_{\mathrm{Be}} \cdot RR_{\mathrm{Be(n,\alpha)}} - N_{\mathrm{L}} \cdot RR_{\mathrm{L(n,T)}} \\
\frac{dN_{\mathrm{T}}}{dt} &= N_{\mathrm{L}} \cdot RR_{\mathrm{L(n,T)}} - \lambda_{\mathrm{T}} \cdot N_{\mathrm{T}} + N_{\mathrm{He}} \cdot RR_{\mathrm{He(n,p)}} \\
\frac{dN_{\mathrm{He}}}{dt} &= \lambda_{T} \cdot N_{\mathrm{T}} - N_{\mathrm{He}} \cdot RR_{\mathrm{He(n,p)}}
\end{aligned}
\tag{1}
$$

where: N_x – number density. The dependence on time has been omitted to simplify the notation. The subscripts: Be, L, T, and He denote ^{9}Be, ^{6}Li, ^{3}H and ^{3}He, respectively and λ_{T} is the tritium decay constant equal to 1.78×10^{-9} s^{-1}. $RR_{\mathrm{I(n},x)}$ denotes the isotope I reaction rate of type (n,x):

$$
RR_{\mathrm{I(n},x)} = \int_0^{10\mathrm{MeV}} \varphi(E,t)\sigma_{\mathrm{I(n},x)}(E)dE \tag{2}
$$

where $\varphi(E, t)$ denotes the neutron flux for energy E at time t and $\sigma_{\mathrm{I(n},x)}(E)$ is the relevant neutron-induced reaction cross-section for isotope I and reaction (n,x).

Tritium has a negligible absorption cross-section, but it decays into highly poisoning ^{3}He during breaks in reactor operation.

As the content of poisoning isotopes in the reactor cannot be measured directly, the fuel management calculations included the block average poison build-up calculations for the whole reactor lifetime. The REBUS [6] in-core fuel management code and the 7-group microscopic cross-section library prepared by WIMS-ANL [5] were used as the basic computational tool for beryllium poisoning calculations. To reduce the REBUS running times, the calculations were performed in two dimensions. This, of course, lowered the accuracy of the poison level predictions.

Depletion calculations

The core burn-up calculations were carried out by REBUS for horizontal cross-section of the core in rectangular geometry shown in Fig. 3, with the geometrical buckling equal to 7.46×10^{-4} cm^{-2}, obtained from the 3D calculations. The value of the axial buckling was determined in the following procedure. For a specified time moment, the fuel composition in all elements, and in ^{9}Be ^{6}Li, ^{3}H and ^{3}He atom densities in beryllium blocks,

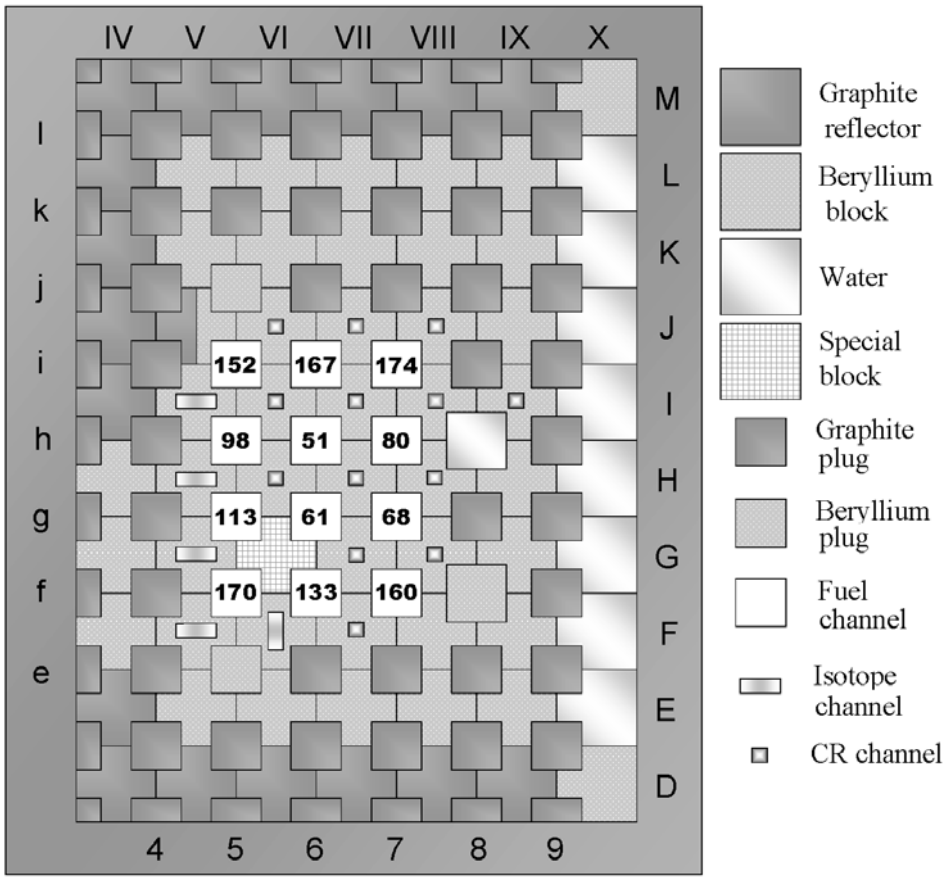

Fig. 3. REBUS computational model of MARIA core.

Table. 1. Beryllium matrix poisoning during one year of MARIA reactor operation

Poisoning characteristics	January 17, 2004	February 3, 2005
Amount of ^{6}Li (g)	2.9	2.9
Amount of ^{3}H (g)	23	20
Amount of ^{3}He (g)	0.29	1.4
Negative reactivity in ^{3}He and ^{6}Li ($)	–4.5	–9.4
Reactivity drop per month ($)	0.4	0.4

obtained in 2D REBUS burn-up calculations, were an input in 3D REBUS axially homogeneous calculations with all control elements withdrawn. Then, the axial buckling was derived so that k_{eff} from 2D is equal to the 3D one. In principle, the k_{eff} calculated in 3D with control rods at critical position should be equal to unity, but this is rarely the case in practice. The relative deviation from $k_{eff} = 1$, i.e. $(k_{eff} - 1)/k_{eff}$ is the bias of the computations. Thus, the bias is a measure of a constant error of the computational model used.

Each beryllium block, fuel element and control rod channel in 2D calculations were represented in the computational mesh as separate material zones with volume preservation. The graphite reflector in the model is thinner than in reality, but this simplification does not practically influence the k_{eff} value. The numbers in fuel element channels are their burn-up values rounded to full MWd.

The depletion calculations are performed in cycles including on-power and off-power periods. First, for an on-power period, the composition of fuel meat in each fuel element and average atom densities of poisoning isotopes in each beryllium block are calculated by REBUS. Five burn-up steps of 15 min are used to simulate the reactor start-up, and 2 h steps are used to simulate the period of stable power operation. Next, for an off-power period, the transmutations in fuel and beryllium during the operational break are calculated. The resulting atom densities are used as input values for the calculations of the next cycle. The fresh fuel composition in all calculations is taken from the manufacturer's certified U-235 content in the fuel element.

Results

Reactivity consequences of a long break in reactor operation

During breaks in reactor operation the accumulated ^{3}H decays into ^{3}He and reactivity of the reactor decreases. The decrease depends on the length of the break. The technical break in MARIA operation took place from January 16, 2004 to February 3, 2005. The decay of tritium during that period caused a 480% increase in ^{3}He content and hence the reactivity drop of ~ 4.9 $, cf. Table 1. The magnitude of the break effect enabled experimental verification of the computational methods.

As the calculations give the k_{eff} values, while the measured quantity is reactivity, the measured values of reactivity were converted to respective effective multiplication k_{exp}, using the formula:

$$k_{exp} = \frac{1}{(1 - \beta \cdot \rho_{exp})} \tag{3}$$

with $\beta = 0.00725$ \$$^{-1}$ used for MARIA core.

The evolution of beryllium matrix poisoning and its effect on the reactivity during one year of operation is shown in Table 1. The effect on the reactivity includes repositioning of four fuel elements, which had to be done to achieve an excess reactivity needed to perform critical experiments in September/October 2004. Thus the real negative effect on the reactivity of additional ^{3}He formation is slightly higher than 4.9 $.

The experiments were performed at different room temperatures while the calculations were performed assuming uniform 20°C. Therefore, necessary temperature correction of the measured reactivity was done. The experimental temperature coefficient equal to -2.4×10^{-2} \$/°C, having approximately a 10% accuracy, was used for that purpose, see Fig. 4.

In Fig. 5, the calculated k_{eff} are compared to the measured k_{exp} values. The k_{eff} straight lines have been

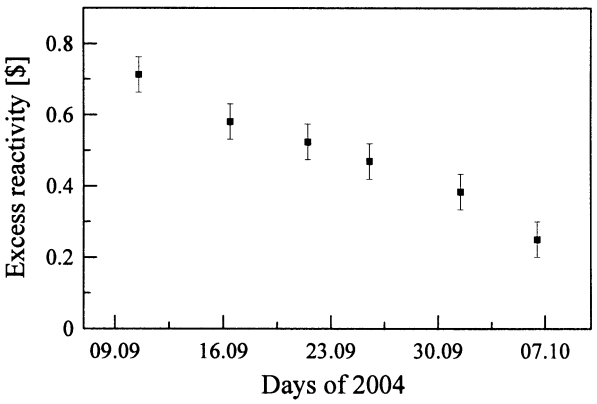

Fig. 4. Measured reactivity corrected to 20°C.

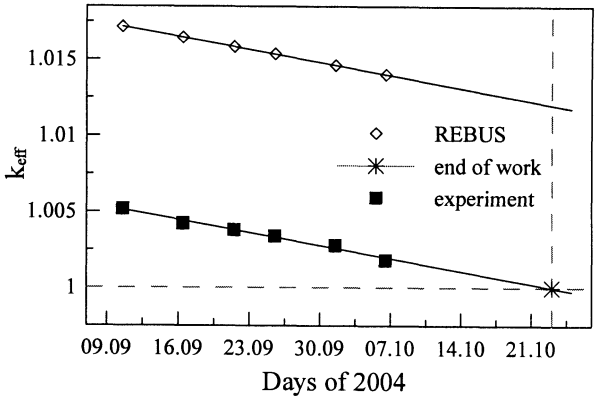

Fig. 5. Experimental and calculated k_{eff} values and interpolated k_{eff} lines.

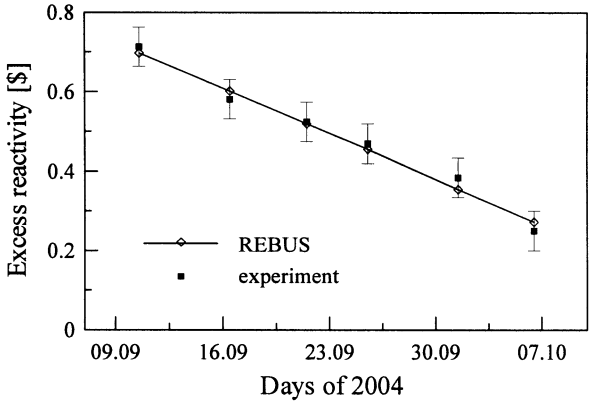

Fig. 6. Comparison of reactivity values with correction for k_{eff} bias.

obtained using the standard least squares method. The coefficients defining the slope of the experimental and computational lines agree to 3 digits. As the statistical error of the experimental slope coefficient is 8×10^{-6}, the computational slope coefficient lies within the statistical error of the experiment.

The intersection of the experimental line with the line $k_{exp} = 1$ determines the moment when the reactivity of the reactor reached zero. The calculated effective multiplication factor is then equal to 1.012, which means that the discrepancy between the computed and measured k_{eff} values is 0.012. This number can be considered as the computational bias. When this bias is subtracted from the results of calculations, a set of computational reactivity values can be compared with the measured values, cf. Fig. 6.

Good agreement between experiment and REBUS calculation can be seen for the reactor core configuration as of September/October 2004. Based on this result, the same bias has been used in the prediction of the reactor start-up experiment in February 2005. The predicted and measured reactivity values are shown in Fig. 7 [4].

Reactor operation after one year shutdown

Although the agreement of predicted and measured stationary reactivity values is acceptable, the work on improvement of the computational model has been continuing. The improvements introduced since 2004 concerned impurities in material specifications, inclusion of experimental channels and preparation of the comprehensive microscopic cross-section library used by REBUS. The improved computer capabilities made possible more extensive application of 3D calculations, so that vertical positions of control rods are routinely accounted for. The computational bias is actually systematically determined as the difference between 3D k_{eff} computed with control rods in critical position and unity. The model of the 3D calculations includes the upper and bottom reflector layers and control rod critical positions, but it does not include the axial dependence of fuel and beryllium isotopic concentrations, as the burn-up calculations are carried on in 2D only. As before, the 2D burn-up calculations use the axial buckling value that ensures the same k_{eff} as the 3D calculations with control rods withdrawn.

A comparison of k_{eff} computed using the improved model, and the measured values for the three chosen weeks of reactor operation is shown in Figs. 8, 9, 10.

During reactor operation, the reactivity change is due to fuel depletion and transmutations in beryllium. The single cycle curves show 3 different cases:

1. The first cycle starting on February 7, 2005, after one year break in operation, cf. Fig. 8. It can be seen that after the first day of operation the reactivity of MARIA is increasing. This is caused by the reduc-

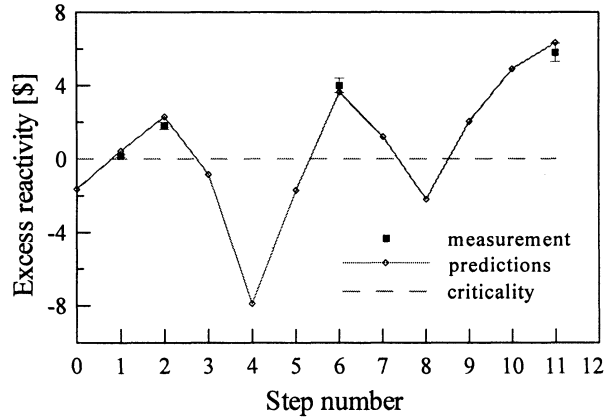

Fig. 7. Predicted reactivity values for consecutive steps of the critical experiment.

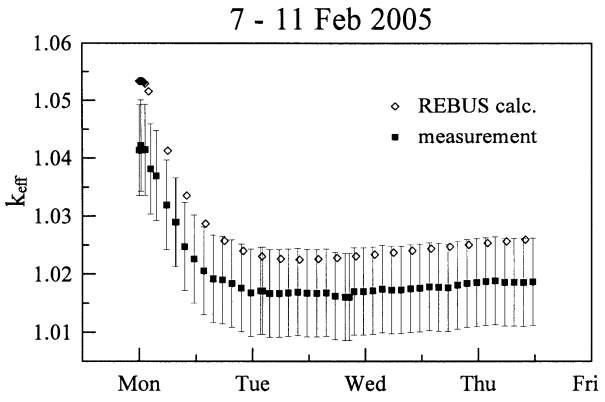

Fig. 8. Effective multiplication factor change for the week starting on February 7, 2005.

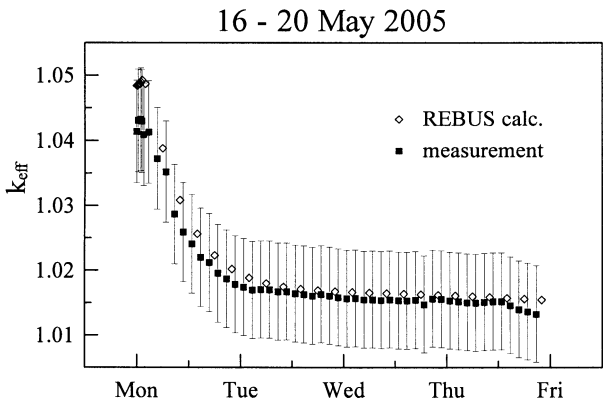

Fig. 9. Effective multiplication factor change for the week starting on May 16, 2005.

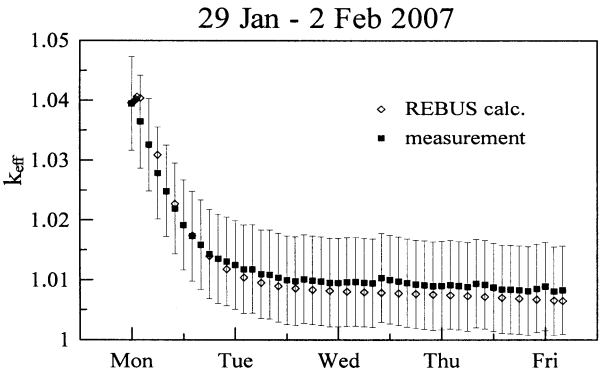

Fig. 10. Effective of multiplication factor change for the week starting on January 29, 2007.

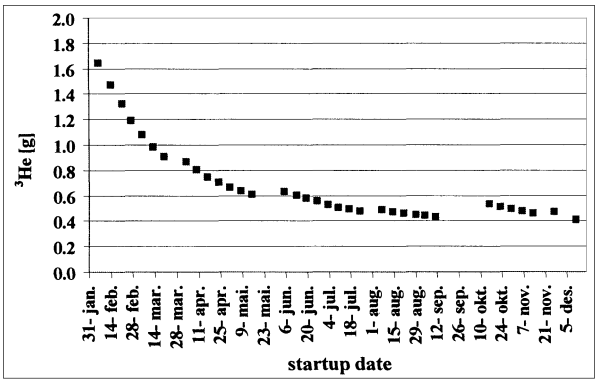

Fig. 11. One year evolution of ^{3}He after reactor startup in February 2005.

tion of ^{3}He content due to its transformation into ^{3}H under irradiation. The reduction in poisoning was strong enough to override the reactivity loss due to fuel depletion. The magnitude of the effect depends on ^{3}H content.

2. A cycle with ^{3}He transformation not sufficient to override the effect of fuel depletion, cf. Fig. 9.
3. The representative cycle with combined effect of fuel depletion and increase in ^{3}He density, cf. Fig. 10.

Figures 8–10 show satisfactory agreement of the measured and calculated detailed reactivity change for a week of the MARIA reactor operation.

The total ^{3}He content in the MARIA beryllium matrix in May 2005 was ~ 0.57 g, and the ^{3}H content was ~ 24 g. In November 2005, the ^{3}He content decreased to ~ 0.44 g and the ^{3}H content increased to 25 g.

Figure 11 shows the ^{3}He content on the second day of each fuel cycle during the year 2005. It can be seen that the ^{3}He content is still decreasing at the end of the year although very slowly. The breaks in reactor operation correspond to the discontinuities in the curve, caused by the increase of ^{3}He content.

Regular reactor operation

The computed evolution of ^{3}He during regular reactor operation in the year 2007 is given in Fig. 12, with computational points showing the ^{3}He content every week.

Figure 13 shows satisfactory agreement of the measured and computed data for the whole year reactivity profile.

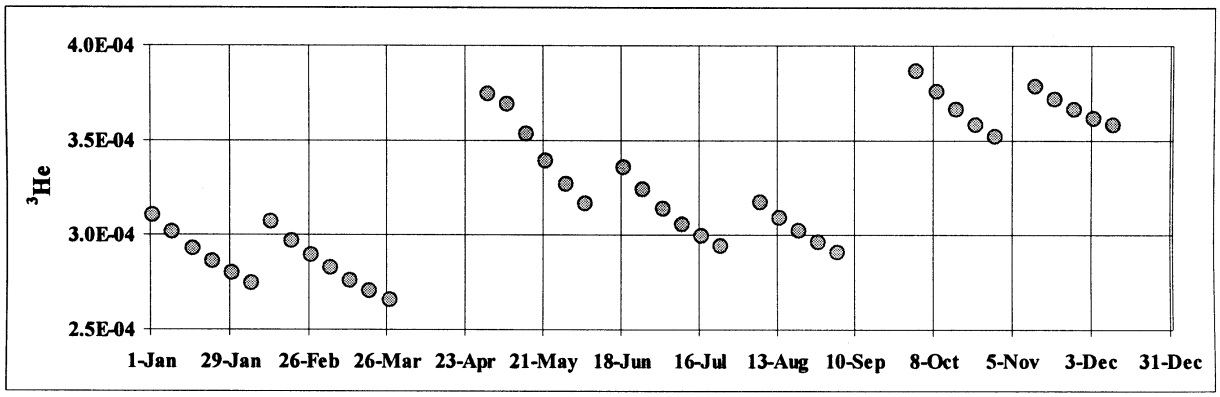

Fig. 12. One year evolution of ^{3}He in 2007 (kg).

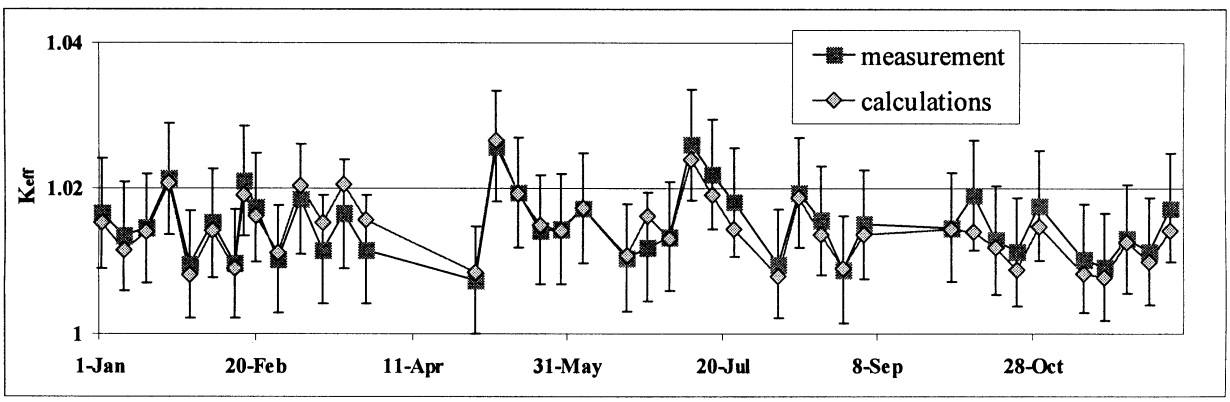

Fig. 13. One year evolution of reactivity.

Table. 2. Beryllium matrix poisoning at the beginning of the years 2004–2008

Poisoning characteristics	2004	2005	2006	2007	2008
Average power in preceding year (MW)	15.20	0.0	16.40	16.70	16.80
Operating time in preceding year (%)	0.46	0.0	0.44	0.46	0.45
Amount of ^{6}Li (g)	3.27	3.27	3.80	3.91	4.33
Amount of ^{3}H (g)	25.12	23.69	25.14	27.51	30.81
Amount of ^{3}He (g)	0.31	1.75	0.38	0.31	0.42
Negative reactivity in ^{3}He and ^{6}Li ($)	4.23	13.25	4.58	4.85	4.21

Summary of beryllium poisoning in the years 2004–2008

Table 2 gives the summary of beryllium poisoning during the last 4 years.

The number densities of January 2004 are the result of MARIA operation since 1974. Then, the break in operation in the period January 16, 2004 – February 2, 2005 took place, during which the decay of tritium into highly absorbing ^{3}He caused a strong reactivity loss equal to 9 $. The figure was obtained from 3D calculations. After reactor start-up in February 2005, the accumulated ^{3}He was transmuted back into tritium and in January 2006 the dynamic equilibrium between ^{3}H and ^{3}He was established. It can be observed that during the whole year 2006 the decay of accumulated ^{3}He was greater than its production. The last line of Table 2 gives the reactivity difference between the core with actual beryllium poisoning and the same core assuming clean beryllium in all blocks and plugs. It should be stressed that in 2007 the reloading of two beryllium blocks took place which slightly decreased the beryllium poisoning effect.

Conclusions

The positive reactivity effects of application of beryllium as a moderator in the MARIA reactor are substantially reduced by the neutron-induced isotopic transmutations in beryllium leading to beryllium poisoning.

The fuel management analysis of the MARIA reactor cannot be carried out without a simultaneous analysis of beryllium blocks poisoning because of a strong influence of that poisoning on core reactivity.

The computational tool for such an analysis has been prepared and has proved to give satisfactory agreement with experimental values.

References

1. Andrzejewski K, Kulikowska T (2000) Beryllium blocks poisoning during the first period of reactor MARIA operation (1975–1985). Report IAE-69/A. Institute of Atomic Energy, Otwock/Świerk
2. Andrzejewski K, Kulikowska T (2002) Poisoning of reactor MARIA beryllium blocks in the period 1993–2000. Report IAE-81/A. Institute of Atomic Energy, Otwock/Świerk
3. Andrzejewski KJ, Kulikowska TA (2004) Isotopic transmutations in irradiated beryllium and their implications on reactor MARIA operation. Nucl Technol 146;1:72–82
4. Andrzejewski KJ, Kulikowska TA, Marcinkowska ZE, Pytel KM (2005) Reactivity of MARIA reactor with strongly poisoned beryllium matrix; experiment and computations. Report IAE-120/A. Institute of Atomic Energy, Otwock/Świerk
5. Deen JR, Woodruff WL, Costescu CI, Leopando LS (2004) WIMS-ANL user manual. Rev. 6. Report ANL/TD/TM-99-07. Argonne National Laboratory, USA
6. Olson AP (2001) A user's guide for the REBUS-PC code, version 1.4. Report ANL/RERTR/TM-32. Argonne National Laboratory, USA