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RAPORT IAE – 40/A

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OTWOCK - SWIERK 1999

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Abstract

This report discusses methods used to evaluate ^{6}Li and ^{3}He poison concentrations, initiated by the ^{9}Be(n,α) reaction, in the beryllium blocks of the MARIA reactor. The results are based on ENDF/B-VI neutron cross sections, 3D diffusion neutron fluxes, and solutions to the differential equations which describe the time-dependent poison concentrations as function of reactor operation and shutdown periods. MCNP Monte Carlo calculations were used to verify calculated poison levels for observed critical configurations. Previous evaluations used somewhat less refined methods based on asymptotic solutions for the poison concentrations. It was found that ^{3}He and ^{6}Li in the beryllium blocks limit the available excess reactivities and alter flux and power distributions. Based on analyses of critical cores, it was determined that poison concentrations need be evaluated for only an in-core region and for an ex-core region and not for each beryllium block.

1. INTRODUCTION

Beryllium irradiated by neutrons with energies in the range 0.7 - 20 MeV undergoes (n,α) and (n,2n) reactions resulting in subsequent formation of isotopes lithium (Li-6), tritium (H-3) and helium (He-3 and He-4). Negative implications of this process are met whenever beryllium is used in a thermal reactor as a reflector or moderator. This paper deals with the negative consequences of the (n,α) reaction. The long-term accumulation of helium and tritium gases produces a swelling effect which limits the lifetime of beryllium blocks in a reactor [1]. Because of their large thermal neutron absorption cross sections, the buildup of He-3 and Li-6 concentrations, initiated by the ^{9}Be(n,α) reaction, results in large negative reactivities which alter flux and power distributions.

The effect has been observed in the beryllium moderated MARIA research reactor when, after a long break in reactor operation, a significant decrease in reactor reactivity was observed and which could not be explained by any structural nor material modifications. To explain the effect, a study of Li-6 and He-3 buildup in the reactor and its consequences on reactor characteristics has been undertaken. A summary of the first results has been presented in Ref. 2.

Below, the equations governing the process are recalled and their solution discussed. The effect of He-3 and Li-6 presence in beryllium blocks on reactor reactivity and power density distribution has been analyzed. It has been shown that the effect depends strongly on the reactor shutdown periods. The loss of reactivity caused by shutdown periods, because of tritium decay, has been estimated for the MARIA reactor.

2. FORMULATION OF THE PROBLEM

2.1 Basic equations

The process of beryllium poisoning starts when beryllium gets irradiated by neutrons. The (n,α) reaction with threshold energy of about 0.7 MeV leads to the set of reactions:

$$
\begin{aligned}
&{}^{9}\mathrm{Be}\,(n,a) \rightarrow {}^{6}\mathrm{He} \\
&{}^{6}\mathrm{He} \xrightarrow{\beta^-} {}^{6}\mathrm{Li}, \qquad T_{1/2} = 0.8\,\mathrm{sec} \\
&{}^{6}\mathrm{Li}(n,a) \rightarrow {}^{3}\mathrm{H} \qquad\qquad , \\
&{}^{3}\mathrm{H} \xrightarrow{\beta^-} {}^{3}\mathrm{He}, \qquad T_{1/2} = 12.33\,\mathrm{yr} \\
&{}^{3}\mathrm{He}\,(n,p) \rightarrow {}^{3}\mathrm{H}.
\end{aligned}
\tag{1}
$$

As seen from Eq. (1), some beryllium is transformed almost immediately into ^{6}Li. The number densities of beryllium, helium, lithium and tritium are controlled by the following equations:

$$
\begin{aligned}
\frac{dN_{Be}}{dt} &= -N_{Be} \cdot \{RR\}_{Be} \\
\frac{dN_{L}}{dt} &= N_{Be} \cdot \{RR\}_{Be} - N_{L} \cdot \{RR\}_{L} \\
\frac{dN_{T}}{dt} &= N_{L} \cdot \{RR\}_{L} - \lambda_{T} \cdot N_{T} + N_{He} \cdot \{RR\}_{He} \\
\frac{dN_{He}}{dt} &= \lambda_{T} \cdot N_{T} - N_{He} \cdot \{RR\}_{He}
\end{aligned}
\tag{2}
$$

where the dependence of the number densities, N, on time has been skipped to simplify the notation. The subscripts: Be, L, T, and He denote respectively Be-9, Li-6, H-3 and He-3. The constant λ_T is the tritium decay constant equal $1.78 \cdot 10^{-09}$ sec^{-1} . {RR} stands for the isotope reaction rates: (n,T) for Li-6, (n,p) for He-3, and (n,α) reaction for beryllium:

$$
\{RR\}_{x} = \int_{E_{min}}^{E_{max}} \varphi(E,t)\sigma_{x}(E)dE \,. \tag{3}
$$

The energy interval (E_{min}, E_{max}) covers the entire range of neutron energies in the Be matrix and $\sigma_x(E)$ is the relevant neutron-induced cross section for isotope x.

It should also be noted that when the neutron flux, $\varphi(E,t) = 0$ (operational break) the last two equations of the system (2) are still nontrivial and describe the tritium decay and He-3 build-up.

The coefficient matrix of the system (2) has the form:

$$
\begin{bmatrix}
-\{RR\}_{Be} & 0 & 0 & 0 \\
+\{RR\}_{Be} & -\{RR\}_{L} & 0 & 0 \\
0 & +\{RR\}_{L} & -\lambda_{T} & +\{RR\}_{He} \\
0 & 0 & +\lambda_{T} & -\{RR\}_{He}
\end{bmatrix}
\tag{4}
$$

It is composed of isotopic reaction rates and the tritium decay constant. As the beryllium reaction rate depends only on the neutron flux magnitude above 0.7 MeV and the others on the thermal flux magnitude, the resulting number densities are strongly dependent on the neutron energy spectrum in beryllium. Thus, the presence of neutron flux in equation coefficients {RR} makes the problem nonlinear as the flux level and spectrum depend on number densities of Li-6 and He-3. The effect of nonlinearity on the problem (2-4) will be discussed separately, but for the moment we shall assume that reaction rates are constant over the period of operation.

In the formulation (2-4) the loss from the ^{9}Be(n,2n) reaction is neglected in $\{RR\}_{Be}$. This approximation is justified as the beryllium number density is nearly constant over typical intervals during which reactor is at power. It has been estimated that the whole period of MARIA operation in the years 1973-85 resulted in the 0.11% reduction of ^{9}Be of which 0.03% were caused by the (n,α) reaction.

The character of transmutations taking place during irradiation of beryllium can be seen in Fig. 1, where the solution of Eqs. (2) for $N_{Be}(0) = 0.11668$ atoms/b-cm, $N_{Li}(0) = N_{H}(0) = N_{He}(0) = 0$, constant reaction rates (relevant to MARIA reactor) and irradiation for 700 days followed by 700 days of tritium decay and again 700 days of irradiation is shown.

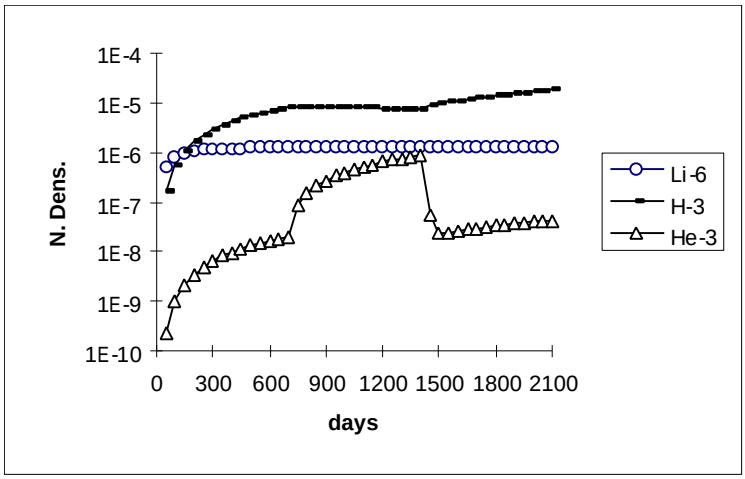

Fig. 1. Solution of Eqs. (2) for $N_{Be}(0) = 0.11668$ atoms/b-cm, $N_{Li}(0) = N_H(0) = N_{He}(0) = 0$, for 700 days of irradiation followed by 700 days of tritium decay and again 700 days of irradiation.

It can be seen that Li-6 density saturates in approximately 200 days, while H-3 and He-3 density is growing with time for the time-scale considered. It is worth noting, that during the 700 days shutdown period He-3 density is increasing about 40 times due to H-3 decay. After return to irradiation a rapid decrease of He-3 density to the pre-shutdown period level is observed. The latter effect is purely theoretical as real reactors are usually operated with shutdown breaks, which will be discussed later.

2.2. Special forms of the equations

In Ref. 3 the requirement for asymptotic solution is discussed. The first of Eqs. (2) can be readily solved giving the decrease of beryllium number density following the exponential function:

$$N_{Be}(t) = N_{Be}(0) \cdot \exp\left\{-\int_0^t \varphi(E)\sigma_{Be}((n,\alpha),E)dE \cdot t\right\} \tag{5}$$

The beryllium reaction rate in all thermal reactors is very low, in the reflector it usually does not exceed 10^{-13}. For the MARIA reactor lattice, it is of the order of 10^{-12} and so for t = 1 yr., $N_{Be}(t)/N_{Be}(0) = 0.99997$. Hence, the number density of beryllium changes negligibly during the time of reactor operation. This fact has been used in Ref. 3 for the analysis of the 30-MW Oak Ridge Research Reactor where the beryllium number density has been assumed constant and only the last 3 equations have been effectively solved:

$$\begin{aligned}
\frac{dN_L}{dt} &= N_{Be} \cdot \{RR\}_{Be} - N_L \cdot \{RR\}_L \\
\frac{dN_T}{dt} &= N_L \cdot \{RR\}_L - \lambda_T \cdot N_T + N_{He} \cdot \{RR\}_{He} \\
\frac{dN_{He}}{dt} &= \lambda_T \cdot N_T - N_{He} \cdot \{RR\}_{He}
\end{aligned} \tag{6}$$

Thus, the set of 4 homogeneous equations (2) can be reduced to the set of three equations (6) with a constant coefficient equal to the product of beryllium number density multiplied by its reaction rate for (n,α) reaction. In both cases the equation coefficients are time dependent as the

neutron flux depends on the amount of strong absorbers which are being formed in the beryllium matrix.

For constant {RR}s and for a sufficiently long time of reactor operation at high flux (i.e. for $\{RR\}_L \cdot t > 5$), the solution of Eqs. 2 or 6 can be approximated by a set of asymptotic formulas given in Ref. 4 with the assumption of $\sigma_{He} \cdot \varphi >> \lambda_T = 1.7814 \cdot 10^{-9}$. In the MARIA reactor the He-3 reaction rates are of the order 10^{-7} - 10^{-6} which means that the asymptotic formulas are valid for $t > 2.8 \cdot 10^4$ hours. The first period of MARIA operation lasted approximately 20000 hours so that the formulas were sufficiently accurate to perform first estimates of the poisoning effects. As the reactor is operated with alternating on-power and off-power periods of order of 10^2 hours the formulas cannot be used for realistic evaluation of beryllium poisoning in operating reactor.

$$N_L^{sat} = \frac{(RR)_{Be}}{(RR)_L} \cdot N_{Be} \tag{7a}$$

$$N_{He}(t) = N_{Be} \cdot \frac{(RR)_{Be}}{(RR)_{He}} \cdot \lambda_T \cdot t + \left(N_L(0) + N_{He}(0) + N_T(0) - N_L^{sat} \cdot \{1 + (RR)_L / (RR)_{He}\} \right) \cdot \frac{\lambda_T}{(RR)_{He}} \tag{7b}$$

$$N_T(t) = N_{Be} \cdot (RR)_{Be} \cdot \left(t + \frac{\lambda_T}{(RR)_{He}^2} \right) + N_L(0) + N_{He}(0) + N_T(0) - N_L^{sat}$$

where the index 'sat' denotes the saturated number density. These equations show that in the asymptotic limit $N_T(t)$ and $N_{He\text{-}3}(t)$ increase linearly with time.

2.3. Mode of operation of the MARIA reactor

The MARIA reactor had been operated since the year 1977 until 1985 with numbers of operating hours per year given in Table 1. For this period the average power output from one assembly has been assumed equal 1.9 MW.

Table 1. Time schedule of MARIA operation in the years 1977 - 85.

Year	Number of hours	Year	Number of hours.	Year	Number of hours.
77	1413	80	723	83	3451
78	2294	81	2215	84	3616
79	1967	82	2437	85	2298
				Total number of hours	20414

Starting from the middle of 1985 the reactor had a break in operation of seven and half years. After this break a shuffling of beryllium blocks had been done and new blocks added. The reactor achieved criticality in the middle of 1993 and up to the end of 1994 was operated for irregular periods specified in Table 2. Starting from 1993 the exact power distributions were available with average power per fuel assembly of about 1.1 MW.

Table 2. Operating and shutdown hours up to January 1995

dates	. hours		dates	. hours	
	operation	shutdown		operation	shutdown
28.06.93			14.06.94	75.1	236.9
29.06.93	24	120	27.06.94	1.4	22.6
05.07.93	12	60	28.06.94	3.3	308.7
08.07.93	12.5	1187.5	11.07.94	100.3	1315.7
27.08.93	23.75	576.25	08.09.94	3.45	332.55
21.09.93	97	95	22.09.94	5.8	282.2
29.09.93	26	22	04.10.94	9.85	62.15
01.10.93	2.25	309.75	07.10.94	8	160
14.10.93	72	240	14.10.94	2.35	429.65
26.10.93	38.25	9.75	02.11.94	5.45	522.55
28.10.93	8.5	135.5	24.11.94	3.55	92.45
03.11.93	13.75	10.25	28.11.94	81.3	86.7
04.11.93	45	1299	05.12.94	266	70
30.12.93	7	3449	19.12.94	272	232
23.05.94	25.25	502.75	09.01.95	84.6	35.4
			Total	1329.7	12206.3

In the last years the reactor operation is based on a weekly schedule of 100 operating hours followed by a 68 hours break. There are additional weekly breaks, caused by economical reasons, every second or third week. The resulting number of hours is 3856 in 1997 [5] and this value has been assumed for subsequent years.

Thus, the estimation of Li-6 and He-3 poisoning had to distinguish the following periods:

I. operation from 1977 until 1985,
II. shutdown for reconstruction,
III.irregular operation in 1993-94,
IV.operation since 1995.

The first estimations of the Li-6 and He-3 build-up had been based on the asymptotic equations (7). The periods III and IV included very short operating times followed by operational breaks of various length. For that reason the general solutions of Eqs. (2) or (6) had to be applied. The analytical solution of those equations has been constructed and a numerical program developed in which an arbitrary number of operating times followed by breaks in operation is taken into account.

3. COMPUTATIONAL TOOLS AND METHODS

The solution of the problem is composed of 2 parts:

1. Calculation of reaction rates, i.e., equation coefficients,
2. Solution of the system (2) or system (6) with known initial values of number densities for beryllium, lithium, tritium and helium.

As already mentioned both systems of equations are nonlinear but the effective solution has been obtained under the assumption of constant reaction rates within the specified periods. The inaccuracy of solution caused by this assumption has been checked in separate calculations.

3.1. Reaction rates calculations

The basic computational tools applied in calculations of reaction rates include:

- Lattice spectrum codes WIMSD5 [6] and WIMS-ANL [7] using respectively the WIMS '86' library of Winfrith and the ANL library based on ENDF/B-VI.
- The transport code TRITAC [8,9] in 2 dimensions and the diffusion-coupled REBUS code in 3 dimensions [10].

Both versions of WIMS have been used for calculations of microscopic cross sections and WIMSD5 was used for calculations of absorption rates normalized to total absorption in the cell equal unity. Those absorption rates had to be then normalized to the flux level in the reactor. The flux level has been obtained from the 2-D TRITAC calculations averaged over 3 types of beryllium blocks: interior, i.e. surrounded by fuel elements, exterior 1 – adjacent to fuel elements, exterior 2 – nonadjacent to fuel elements. The (n,α) rates have been calculated using the WIMSD5 fast fluxes multiplied by $Be(n,\alpha)$ cross sections [11] averaged over respective energy groups.

In WIMS-ANL, 7 group microscopic cross sections have been calculated and combined, following Eq. (3), with the 7 group flux values taken from REBUS calculations to determine the required microscopic reaction rates. The ANL library allowed for explicit determination of 7 group microscopic cross sections for the needed reactions. The average flux values over two types of beryllium blocks have been considered: inner – surrounded by fuel elements and outer – composed of the rest of the beryllium matrix. The flux values have been taken from 3D calculations of a 16 fuel assembly equilibrium core with appropriate values of Li-6 and He-3 number densities modified following the calculated case. The flux level corresponded to the total power output of the reactor core equal to 16.1 MW. This approach will be referred to as 'improved'.

3.2. Solution of the system of equations for number densities

The asymptotic Eqs. (7) were used only to obtain the first estimate of the poisoning for the first period of MARIA operation with lumped total number of operational hours. The obtained results have shown the necessity of more detailed treatment of shutdown periods. Moreover, the time schedule of reactor operation in years 93-98 did not allow for application of asymptotic formulas. A program has been written based on the analytic solution of Eqs. (2) or (6). During the shutdown period, tritium accumulated during reactor operation decayed to He-3 following the last two of Eqs. (2) or (6), with assumed zero neutron flux:

$$\begin{aligned} N_T(t) &= N_T(0)\cdot\exp(-\lambda_T\cdot t) \\ N_{He}(t) &= N_{He}(0) + N_T(0)\cdot\{1-\exp(-\lambda_T\cdot t)\} \end{aligned} \tag{8}$$

For $t << T_{1/2}$, with $T_{1/2}$ = 12.33 yr., the half-life of tritium, the second of Eq. (8) can be approximated by:

$$N_{He}(t) = N_{He}(0) + N_T(0)\cdot\lambda_T\cdot t \tag{8a}$$

The approximation expressed by Eq. (8a) can be applied for the analysis of weekly operation with standard shutdown periods. However, for the first period of MARIA operation where the operating and shutdown times are lumped into 1 year's periods, the use of Eq. (8a) for calculation of He-3 build-up during shutdown leads to an error in He-3 contents of 2% after the first shutdown of 6466 hr. and to 18% after the long shutdown specified as the second period of the reactor lifetime.

The initial conditions applied to calculations for the first period of reactor operation were: zero number densities for lithium, tritium and helium and the number density of beryllium corresponding to the technical specification of beryllium blocks in the MARIA reactor. For the next periods the number densities calculated for the end of the preceding period were used as initial values.

The homogeneous system of 4 Eqs. (2) or 3 Eqs. (6) can be solved analytically leading to rather lengthy formulas given respectively in Appendix A and Appendix B. The 2 solutions are equivalent for the orders of magnitude of reaction rates holding in reactor applications. Namely in the analytical solution of Eqs. (6) the fact that the beryllium (n,α) reaction rates are much smaller than the Li-6 and He-3 reaction rates has been used. Both solutions have been written in the form of independent subroutines and used for calculations of irradiation periods. The results obtained from both of them have been found identical in 4 digits for the whole reactor MARIA lifetime, including a number of very short and long (i.e. lumped) operating times. The subroutine used for calculations of the decay periods is based on Eq. (8). It should be stressed that this subroutine requires application of double precision which has been applied in the whole program. Program BERYL used for calculations of Li-6, H-3 and He-3 number densities is given in Appendix C. It includes two subroutines based on solutions either of Eqs. (2) or of Eqs. (6) which can be used optionally, and is composed of:

1. subroutine solving Eqs. (2),
2. subroutine solving Eqs. (6),
3. subroutine calculating reaction rates, from 7- group neutron flux values and cross sections given in the input,
4. subroutine calculating tritium decay during shutdown following Eqs. (8),
5. error subroutine,
6. the main program.

The program allows for an arbitrary number of steps. At each step we specify number of operating hours followed by a number of shutdown hours.

Reaction rates may be calculated by the program from the 7-group cross sections supplied via DATA statement and 7-group flux values given in the input for the following upper energy group boundaries [eV]:

1.0E+07, 8.21E+5, 5.53E+03, 4.0E+00, 6.25E-01, 2.5E-01, 5.8E-0.

It should be stressed that the reaction rates are in this case calculated with multigroup microscopic cross sections representative of the spectrum in the beryllium matrix of the MARIA reactor. For a different reactor those cross sections should be recalculated and put under the DATA statement in the subroutine RRATES.

4. REACTION RATES

As stated above in the solution of Eqs. (2 or 6) constant reaction rates have been assumed. However, the reaction rates have been recalculated for the basic periods defined in section 2.3 to take into account the influence of absorption modifications on the neutron flux,

4.1 The first period of MARIA operation 1977 - 1985

The calculations have been done on several levels of sophistication.

The first estimates were made to establish the order of magnitude of the effect. They have been based on asymptotic formulas (7). The microscopic reaction rates in beryllium were

calculated by WIMSD5 for the MARIA cell. Reaction rates have been normalized to the flux level corresponding to the power in fuel assembly equal 1.9 MW. They are given in Table 3.

Table 3. The first estimation of Li-6 and He-3 absorption and beryllium (n,α) rates in the beryllium moderator

Isotope	Burn-up [MWd/t]	Fast	Thermal	Total
He-3	0.0	2.745E-8	1.01E-6	1.033E-6
	286400	2.443E-8	1.065E-6	1.089E-6
Li-6	0.0	4.946E-9	1.799E-7	1.845E-7
	286400	4.407E-9	1.894E-7	1.938E-7
Be	0.0	1.823E-12	0.0	1.823E-12
	286400	1.615E-12	0.0	1.615E-12

Additional calculations have been carried out with the beryllium block divided into 4 zones of equal cross sectional area. The calculation has given the dependence of reaction rates on the distance from the fuel element shown for Li-6 in Fig. 2. The dependence of Li-6 absorption rate is given for 3 cases:

- For a clean beryllium moderator and fresh fuel,
- For a clean moderator with burned fuel,
- For the moderator poisoned by Li-6 and He-3 to the level estimated for the end of the first period of MARIA operation, i.e., N (Li-6) = $4.1 \cdot 10^{-06}$, N (He-3) = $8.6 \cdot 10^{-08}$ atoms/b-cm

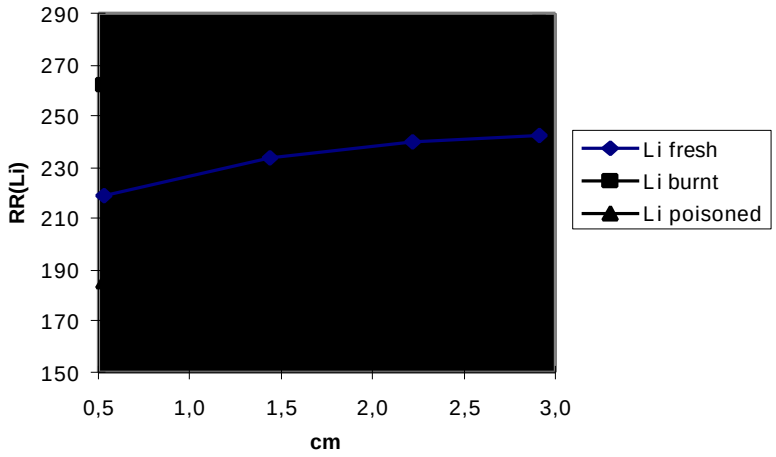

Fig. 2. Dependence of Li absorption rates on the distance in beryllium.

All the above comparisons have been calculated in an infinite lattice. The neutron flux in the reactor MARIA is strongly dependent on the position of the beryllium blocks and hence, the dependence of reaction rates on the position in the reactor had to be taken into account. It has been found impractical to calculate reaction rates for each beryllium block. Therefore 3 types of beryllium blocks were defined:

- interior - blocks surrounded by fuel assemblies
- exterior1 - blocks adjacent to fuel assemblies
- exterior2 - the rest of blocks.

The reaction rates calculated using fluxes taken from 2-D calculations for exterior regions of a small core with 6 fuel elements, used in the first critical experiment, are shown in Table 4.

The reaction rate values have been refined using WIMS-ANL with the library based on ENDF/B-VI. They are quoted in Table 4 under the name 'improved'. Besides, the core layout with 16 fuel elements and realistic fuel burn-up distribution has been used to calculate the flux distribution. Beryllium was poisoned to the level of

$$\text{Li-6 number density} = 7.95 \cdot 10^{-07} \text{ atoms/b-cm},$$
$$\text{He-3 number density} = 8.79 \cdot 10^{-08} \text{ atoms/b-cm},$$

corresponding to a middle value from the first period of reactor operation. In the 'improved' approach the 'exterior1' and 'exterior2' blocks are combined into one zone called 'outer'.

Table 4. Reaction rates in beryllium blocks at various positions in the first core

	Reaction Rate				
Isotope	Interior, first evaluation	Interior, improved	exterior1	exterior2	Outer, improved
Be-9	1.823E-12	1.367E-12	5.861E-13	1.185E-13	1.977E-13
Li-6	1.845E-07	1.297E-07	5.932E-08	1.199E-07	5.746E-08
He-3	1.033E-06	7.384E-07	3.321E-07	6.715E-08	3.271E-07

It is easy to see that absorption rates of Li-6 and He-3 averaged over all outer blocks are close to exterior1, while the beryllium rate is closer to the exterior2. A significant difference can be noted for interior blocks. This can be due to the different number of the fuel assemblies in the cores of 1974 and 1993.

4.2 The second period of MARIA operation

After the reactor shutdown from 1986 to 1993 the He-3 buildup from tritium decay has been so strong that the reaction rates had to be recalculated with actual values of Li-6 and He-3 number densities. Several sets of calculations corresponding to several reloading schemes of the beryllium blocks have been considered. Table 5 gives the reaction rates:

- calculated for the heaviest poisoning, i.e., with Li-6 and He-3 content calculated for the core interior, applied in the whole beryllium matrix,
- calculated for the heavy poisoning in the core interior and the light poisoning, with Li-6 and He-3 content taken for the outer beryllium blocks.

Table 5. Reaction rates in beryllium blocks for different levels of poisoning for the second core at its startup in 1993 (actual power distribution, fuel partially burned)

	Reaction Rate			
	Interior blocks		Outer blocks	
Isotope	Heavy poisoning	Light poisoning	Heavy poisoning	Light poisoning
Be-9	1.698E-12	1.526E-12	2.245E-13	2.575E-13
Li-6	8.831E-08	8.860E-08	2.524E-08	5.558E-08
He-3	5.025E-07	5.042E-07	1.323E-07	3.164E-07

For further calculations the heavy poisoning rates have been applied for interior blocks, and the light poisoning rates for the outer blocks. At the beginning of the fourth period of the reactor life, in January 1995, a significant amount of He-3 has already disappeared due to neutron capture during operating periods (cf. the last of Eqs. (2)). Therefore, the calculation of

reaction rates has been repeated with new poisoning. The new values are given in Table 6 together with values for the beginning of 1996 when the number density of He-3 was further reduced.

Table 6. Reaction rates in beryllium blocks in the beginning of 1995 and 1996 (actual power distribution, fuel partially burned)

	Interior		Outer	
Isotope	January 1995	January 1996	January 1995	January 1996
Be-9	1.398E-12	1.362E-12	2.100E-13	2.022E-13
Li-6	1.213E-07	1.282E-07	5.927E-08	5.192E-08
He-3	6.903E-07	7.300E-07	3.375E-07	3.527E-07

A comparison of Tables 5 and 6 shows an obvious difference between the neutron spectrum in 1993 and 1995 caused by the strong reduction of the He-3 content. The changes between January 1995 and January 1996 are much smaller.

5. NUMBER DENSITIES OF Li-6, H-3 AND He-3 ACCUMULATED IN BERYLLIUM BLOCKS

5.1 The first period of MARIA operation

The first evaluation of beryllium poisoning at the end of the period 1977 - 1985, based on lumped time of reactor operation and the asymptotic approximation (Eqs. 7), has given the number densities shown in Table 7. It is seen that for reaction rates corresponding to the spectrum typical for burned fuel, the number densities are lower by almost 20% for all isotopes. The correction for different positions in the reactor has led to higher contents of the poisonous isotopes in the core center and lower in the outer regions.

Table 7. Number densities [atoms/b-cm] of He-3, Li-6 and H-3 in beryllium blocks with various positions in the reactor.

	Spectrum type				
Isotope	Infinite lattice fresh fuel	Infinite lattice burned fuel	Interior	Exterior 1	Exterior 2
Li-6	1.10E-6	9.31E-7	1.8886E-6	6.0713E-7	1.2256E-7
H-3	1.39E-5	1.23E-5	2.4398E-5	4.6215E-6	6.1766E-7
He-3	2.35E-8	1.99E-8	4.0292E-8	1.2215E-8	2.2993E-9

The number densities of all 3 isotopes differ by more than one order of magnitude depending on the position in the reactor. Table 8 shows number densities of He-3 and Li-6 in four equi-volume zones of the beryllium block, calculated in the infinite lattice approximation.

Table 8. Number densities [atoms/b-cm] of Li-6 and He-3 in beryllium blocks at various distances from the fuel element center

Layer width [cm] Isotope	3.63 – 4.52	4.52 – 5.26	5.26 – 5.91	5.91 – 6.50
Li-6	1.19E-06	1.10E-06	9.80E-07	9.53E-07
He-3	2.52E-08	2.33E-08	2.09E-08	2.03E-08

The first calculations assumed the whole time of reactor operation from Table 1 to be a sum of times of operation in consecutive years 1977 through 1985. Since the experience from the first estimations have shown the strong influence of shutdown periods, the 'improved' calculations, following conditions described in section 3.3, have been carried out assuming for each year the operational period from Table 1 followed by the shutdown period with number of hours summed up to the end of the year. In Table 9 the results for internal blocks are compared with those using one lumped period of operation or a period of operation followed by an extended shutdown.

Table 9. Number densities [atoms/b-cm] of Li-6, H-3 and He-3
in inner and outer beryllium blocks based on 'improved' reaction rates

Position	Inner blocks			Outer blocks
Isotope	Each year separately	Lumped operation	Lumped operation + shutdown	Each year separately
Li-6	1.230E-06	1.230E-06	1.230E-06	3.955E-07
H-3	1.004E-05	1.047E-05	7.541E-06	1.274E-06
He-3	4.503E-07	2.473E-08	2.951E-06	6.528E-09

5.2 The shutdown period

On the basis of number densities obtained at the end of the first period of the reactor operation the buildup of He-3 from H-3 decay during 7 years of reactor shutdown has been calculated using Eq. (8). Table 10 shows the He-3 buildup calculated starting from the values for an infinite lattice given in Table 7.

Table 10. Dependence of He-3 number density [atoms/b-cm] on the length
of the break in reactor operation.

Years	0	1	3	6	9	10
fresh fuel	2.35E-8	7.80E-7	2.17E-6	3.99E-6	5.52E-6	5.98E-6
burnt fuel	1.99E-8	6.93E-7	1.93E-6	3.54E-6	4.91E-6	5.31E-6

It is seen that during a 7 years shutdown of the reactor the amount of He-3 increases by more than two orders of magnitude and its number density can reach the value of about 4E-6 atoms/b-cm. The results for the three types of beryllium blocks are given in Fig. 3. This large increase is because N(H-3)>>NHe-3) at the start of the shutdown (see Table 9).

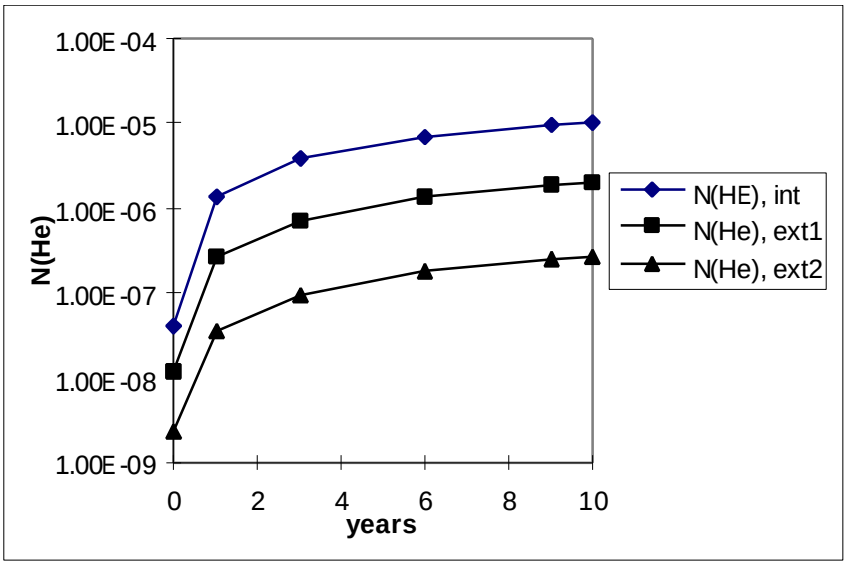

Fig. 3. N(He-3) buildup during the reactor shutdown.

The 'improved' calculations based on 3D fluxes have given values of Li-6, H-3 and He-3 number densities shown in Table 11.

Table 11. Number densities [atoms/b-cm] of He-3, Li-6 and H-3, obtained by the 'improved' approach after 7 years of reactor MARIA shutdown.

Isotope	Inner blocks	Outer blocks
Li-6	1.230E-06	3.955E-07
H-3	7.059E-06	8.597E-07
He-3	3.426E-06	4.208E-07

5.3. The period of irregular operation

In 1993, new beryllium blocks were introduced into the reactor periphery and the old ones shuffled toward the center. The initial calculation has been carried out for old beryllium blocks and separately for new blocks. The old blocks have been assumed to be loaded into the interior of the reactor core. The new blocks have been placed, respectively, into the interior positions surrounded by fuel channels, exterior adjacent to the fuel channels, and exterior loaded in positions farther from fuel channels. For the period of irregular reactor operation, from the reactor start-up in 1993 up to the end of 1994 the detailed schedule of reactor operation including exact operational breaks has been accounted for. The calculation has been repeated for the 3 types of old beryllium blocks differing in their position during the first operation period and placed now in the inner zone. The results are shown in Table 12. In the same Table 12 the results of the 'improved' approach are quoted for inner and outer beryllium blocks.

Table 12. Number densities [atoms/b-cm] of He-3, Li-6 and H-3 at the beginning of 1995 in old beryllium blocks with various positions in the reactor during the first period of operation.

Isotope	Inner – first approach	Inner – improved approach	Exterior 1	Outer – improved approach	Exterior 2
Li-6	1.794E-06	1.499E-06	9.257E-07	4.294E-07	5.589E-07

H-3	1.389E-05	1.059E-05	2.771E-06	1.281E-06	2.235E-07
He-3	8.755E-07	4.834E-07	9.324E-08	1.285E-07	9.480E-09

For the new beryllium blocks a similar calculation has been carried out with initial values of He-3, Li-6 and H-3 equal to zero. The results are given in Table 13.

Table 13. Number densities [atoms/b-cm] of He-3, Li-6 and H-3 in new beryllium blocks at various positions in the reactor, at the beginning of 1995.

Number density	interior	Exterior 1	Exterior 2
Li-6	5.235E-7	1.846E-7	3.960E-8
H-3	2.086E-7	3.724E-8	1.713E-7
He-3	2.143E-9	9.398E-10	2.403E-9

5.4. The last period of operation

For the last period of the MARIA reactor operation starting from the beginning of 1995 a regular operation has been assumed composed of 100 hours of work and 68 hours of operational break. The predicted number densities of He-3 and Li-6 starting from the reactor start-up in 1993 are given in Figs. 4 and 5. They have been calculated by the 'improved' approach. They are given for the old beryllium blocks present in the reactor from the start of its operation in 1977, and for new blocks loaded in 1993. Time equal to zero in all figures corresponds to the second reactor start-up in 1993. Only the starting values for each year are given in the figures although the detailed scheme has been used in calculations. The curves have been drawn separately for the 'inner' and 'outer' beryllium blocks.

The 'improved' approach has given number densities shown in Tables 14.

Table 14a. Number densities [atoms/b-cm] of He-3, Li-6 and H-3, for inner Be blocks, for the last period of reactor MARIA operation.

Isotope	January 96	January 97	January 98	January 99
Li-6	1.378E-06	1.267E-06	1.245E-06	1.241E-06
H-3	1.316E-05	1.526E-05	1.727E-05	1.926E-05
He-3	8.883E-08	9.800E-08	1.111E-07	1.241E-07

Table 14b. Number densities [atoms/b-cm] of He-3, Li-6 and H-3, for outer Be blocks, for the last period of reactor MARIA operation.

Isotope	January 96	January 97	January 98	January 99
Li-6	4.211E-07	3.998E-07	3.898E-07	3.851E-07
H-3	1.691E-06	1.999E-06	2.294E-06	2.294E-06
He-3	2.645E-08	2.843E-08	3.274E-08	3.700E-08

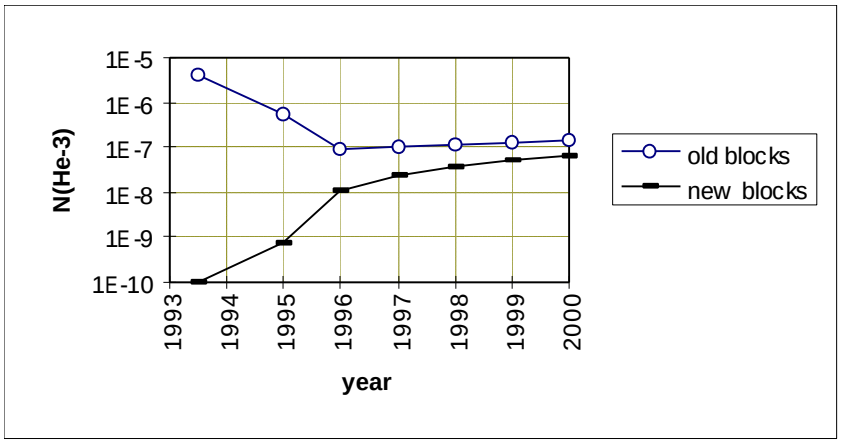

Fig. 4a. Dependence on time of He-3 number densities [atoms/b-cm] in the interior beryllium blocks.

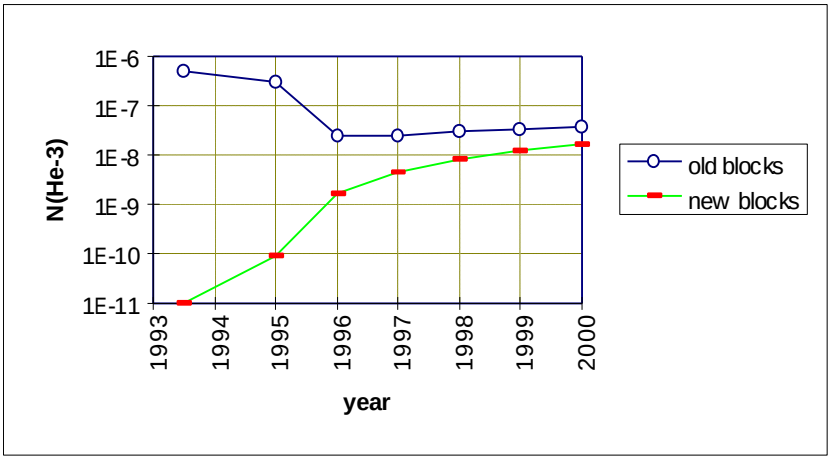

Fig. 4b. Dependence on time of He-3 number densities [atoms/b-cm] in the exterior beryllium blocks.

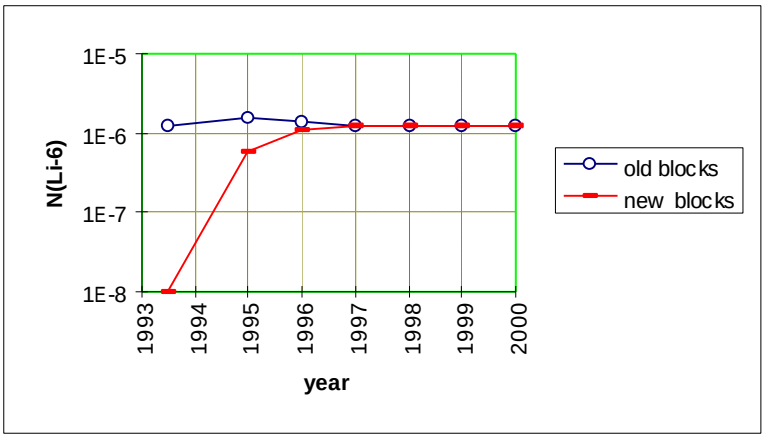

Fig. 5a. Dependence on time of Li-6 number densities [atoms/b-cm] in the interior beryllium blocks.

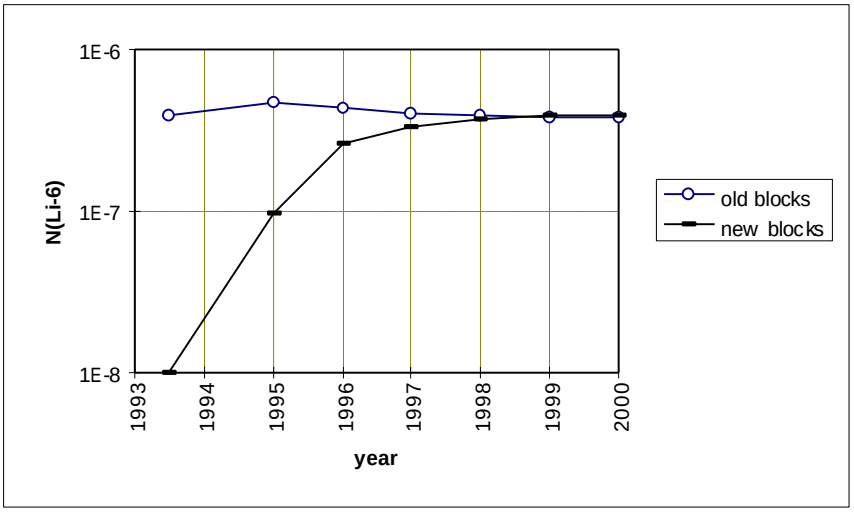

Fig 5b. Dependence on time of Li-6 number densities [atoms/b-cm]in the exterior beryllium blocks.

5.5. The pessimistic approach

In the improved approach two sets of results have been obtained for this period: The first set has been calculated with periods of operation and shutdown following the reactor history of operation as given in Table 2 of section 2.3 for the period of irregular operation 93 -94. The second took into account the fact that the reactor needs additional time to reach its nominal power after each startup. A 5 hours period has been subtracted from the operation after each startup and added to the shutdown period. This approach will be referred to as ‘pessimistic’. For the year 95 the same scheme as for the year 93 has been assumed as no details on the exact operation schedule have been available at the moment. For the year 97 the general scheme described in section 3.1 has been kept but with a shift of 5 hours of the operation time at each reactor startup to the shutdown time. This ‘pessimistic’ approach has given the results shown in Table 15.

Table 15. Number densities [atoms/b-cm] of He-3, Li-6 and H-3, for outer Be blocks, in the pessimistic approach.

Isotope	Inner		Outer	
	Feb. 97	Feb. 98	Feb. 97	Feb. 98
Li-6	1.424E-06	1.271E-06	4.225E-07	3.985E-07
H-3	1.108E-05	1.396E-05	1.357E-06	1.826E-06
He-3	6.033E-07	8.191E-08	1.389E-07	2.163E-08

5.6. Dependence of number densities of Li-6 and He-3 on reaction rates

The nonlinearity of the problem was assessed by repeating the calculation of the number densities of Li-6 and He-3 with different sets of reaction rates. The results are shown in Table 16.

Table 16. Dependence of number densities [atoms/b-cm]of Li-6 and He-3 calculated for the '95 core, using different reaction rates in inner beryllium blocks.

isotope	reaction rates		number densities	
	actual	first core	based on actual RR	based on first core RR
Be	1.584E-12	1.823E-12	1.167E-01	1.167E-01
Li-6	1.184E-07	1.845E-07	1.329E-06	1.153E-06
H-3	-	-	1.427E-05	1.154E-05
He-3	4.248E-07	1.033E-06	9.209E-07	1.190E-06

The same calculation has been repeated using the 'improved' approach. Results are shown in Table 17. Comparing the last two columns we see the magnitude of influence of the spectrum in beryllium blocks on the buildup of Li-6 and He-3 poisoning.

Table 17. Dependence of number densities of Li-6 and He-3 [atoms/b-cm] calculated for the '95 core, using different reaction rates in inner beryllium blocks, 'improved' approach.

isotope	reaction rates		number densities	
	actual	first core	based on actual RR	based on first core RR
Be	1.698E-12	1.367E-12	1.167E-01	1.167E-01
Li-6	8.831E-08	1.297E-07	1.378E-06	1.153E-06
H-3	-	-	1.316E-05	1.094E-05
He-3	5.025E-07	7.384E-07	8.883E-08	2.066E-07

6. INFLUENCE OF BERYLLIUM POISONING ON REACTOR CRITICALITY AND POWER DISTRIBUTION

6.1. The infinite lattice approximation

The beryllium poisoning is interesting mostly because of its consequences on the reactor reactivity. In other words it was important to know if the presence of He-3 and Li-6 in the above estimated amounts can cause a significant change of reactor reactivity. In the first step the question has been answered in the infinite lattice approximation. A separate set of calculations has been carried out to estimate the effect separately for both of these isotopes. The results are

given in Fig. 6 in the form of k-inf dependence on the concentration of the isotopes for the MARIA lattice with fresh fuel in the estimated range of number densities.

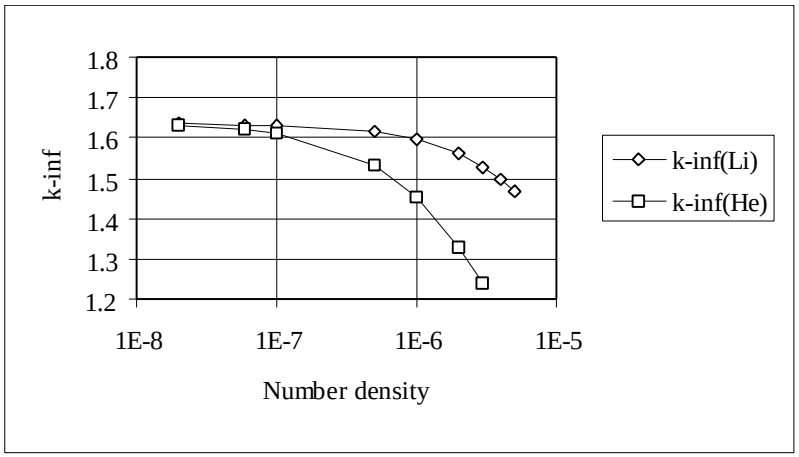

Fig. 6. Effect of He-3 and Li-6 presence in the beryllium moderator on k-inf.

A separate check has shown a negligible effect of beryllium poisoning on the fuel burn-up. The results are shown in Table 18. This conclusion is confirmed in Fig. 7.

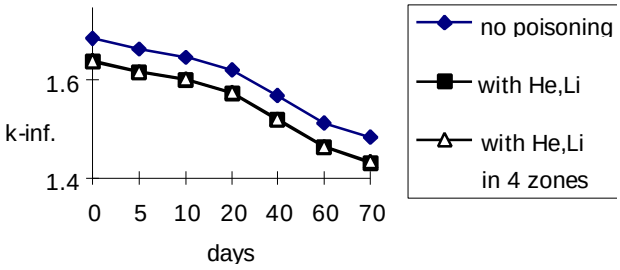

Fig. 7. Dependence of k-inf on burn-up for:

- beryllium blocks without poisoning,
- beryllium blocks with average poisoning,
- beryllium blocks with poisoning calculated for 4 layers.

Table 18. Burn-up characteristics without and with beryllium poisoning.

Be moderator	*k-inf* (0)	*k-inf* (70d)	U-235(70d)	aver.φ_{tot} (0)	aver.φ_{tot} (70d)
no poisoning	1.6253	1.4327	1.6253E-3	5.5414E+14	4.7469 E+14
average poison	1.5887	1.3836	1.6249E-3	4.9530 E+14	4.7995 E+14
poison in 4 zones	1.5905	1.3844	1.6249E-3	4.9525 E+14	4.7990 E+14

6.2. Comparison with critical experiments of 93 and 95

The effect shown in Fig. 6 seems very strong, taking into account the contents of Li-6 and He-3 for beryllium blocks in the MARIA reactor. However, the effect in the reactor with 15 - 17 fuel elements, with the spectrum far from the asymptotic one should be evaluated separately. It has been impossible to reconstruct the history of irradiation of each beryllium block. We had to

accept the fact that the estimation of beryllium block poisoning can be done only with the inevitable error because of different irradiation histories of various beryllium blocks.

The first comparison with critical experiment has been carried out for beryllium moderator with varying number densities of admixed poisoning isotopes. A simplified diagram of reactor configuration is shown in Fig. 8.

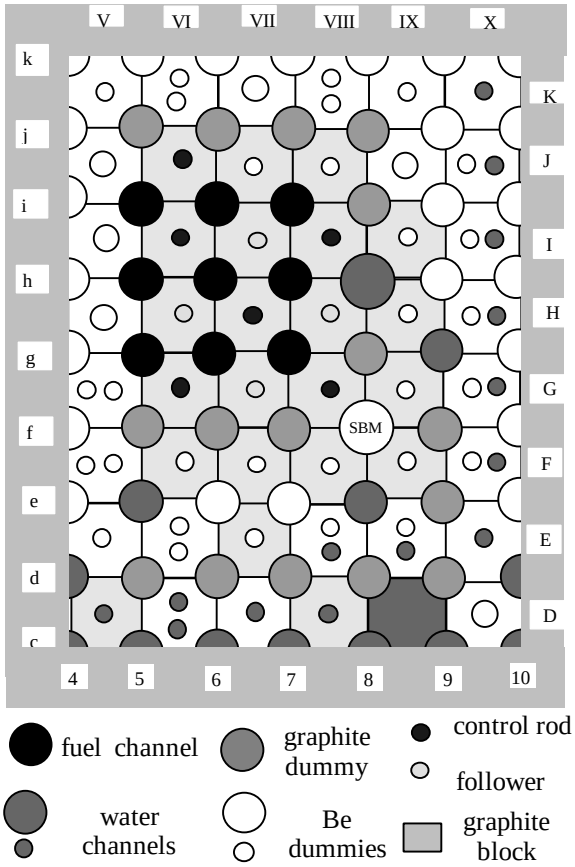

Fig. 8. Horizontal cross section of the MARIA reactor during critical experiments in 1993.

The shaded area shows the positions of the old beryllium blocks. The differences in irradiation channel radii and detailed block shapes are not shown to simplify the diagram. The 'SBM' denotes an experimental stand, later removed. During these experiments the stand consisted of two concentric tubes filled with water.

A set of measurements in different reactor configurations has been available, differing in the filling of particular channels by graphite, beryllium or water. The measured excess reactivity of those configurations, with all control rods withdrawn, could be compared with the sets of calculations performed assuming different level of poisoning in old beryllium blocks. For all the sets the content of Li-6 was kept constant and equal 1.9E-6 atoms/b-cm. The results are given in Tables 19 and 20.

Table 19. Excess reactivity of configurations in \$ (β_{eff}=7.25E-3).

No	Configuration	Calculation : N(He-3)=4E-7	Calculation: N(He-3)=1E-6	Measurement
(a)	Fig. 7	9.03	3.07	2.99
(b)	f-6 ⇒water	8.49	2.58	2.49
(c)	f-6 ⇒water	9.12	3.15	3.03

	e-6 ⇒graphite			
(d)	j-6,j-7,f-5,f-6,f-7 ⇒beryllium	9.42	3.42	3.26

Table 20. Reactivity worth of configuration modifications.

Modification	Δρ [$]		
	N(He-3) 4.0E-7	N(He-3) 1.0E-6	experiment
a⇒b	-0.54	-0.49	-0.50
a⇒c	+0.09	+0.08	+0.04
b⇒c	+0.63	+0.57	+0.52
a⇒d	+0.39	+0.35	+0.27
c⇒d	+0.30	+0.27	+0.23
average discrepancy	+0.08	+0.04	

A systematic comparison of experimental and computational results has shown that the effective average number densities of He-3 equal to $1.0 \cdot 10^{-06}$ and of Li-6 equal to $1.9 \cdot 10^{-06}$ atoms/b-cm, give results close to experiment.

The next set of results concerns the poisoning existing in the reactor after over one year of irregular operation. The core was composed of 16 fuel elements differing in burn-up and power level, which had to be taken into account together with actual control rod positions. The k-eff values obtained from 2D TRITAC calculations, with correction for a constant bias of 30 mk evaluated using the MCNP code, are shown in Table 21 for various poisoning of the old blocks.

Table 21. k-eff for the MARIA reactor configuration of 1995 with various number densities [atoms/b-cm] of He-3 and in old blocks. In new blocks N (He-3) = 9.4E-10, N(Li-6) = 1.8E-07.

No	N(He-3) in old blocks	N(Li-6)in old blocks	k-eff
1	9.3E-08	9.3E-07	1.055
2	8.8E-07	1.8E-06	0.981

It has also been found that the distribution of beryllium poisoning has to be taken into account when calculating the power distribution in the reactor. In Table 22 the power distribution calculated for 3 sets of He-3 and Li-6 concentrations (both cases from Table 21 and the third case, same as 1, but with 3 blocks with higher He-3 and Li-6 concentrations) and for the same reactor configuration is given. The increased number densities have been used in the blocks: K-VI, J-VII and J-VIII (cf. Fig. 8). The change of k-eff for this third case has been negligible, with only 0.4% decrease in comparison with case 1 of Table 21. As seen in Table 22, the power decrease in the fuel channels adjacent to modified beryllium blocks is around 10%. Thus, the application of effective beryllium poisoning in all old blocks instead of a detailed history of each block can lead to the errors in power distributions up to 10%.

Table 22. Power distribution for 3 sets of He-3 and Li-6 concentrations. Normalization to the total power equal 16.91 MW.

	5	6	7	8
i		0.87	0.85	
		0.85	0.83	
		0.79	0.74	
h	0.85	1.18	1.19	0.84
	0.90	1.17	1.17	0.82
	0.84	1.16	1.15	0.81
g	1.23	1.40	1.17	
	1.33	1.40	1.13	
	1.26	1.41	1.17	
f	1.17	1.30	1.11	0.67
	1.25	1.29	1.07	0.60
	1.20	1.33	1.14	0.68
e	0.99	1.16	0.95	
	1.04	1.13	0.91	
	1.03	1.20	0.98	

6.3. Comparison with the criticality measurements of February 97

The results of Tables 19 - 22 show the strong influence of beryllium poisoning on basic reactor core characteristics. The most interesting set of results has been obtained in February 1997 where the detailed configuration of fuel element burn-up and control rod positions could be applied. The reactor has been assumed free of Xe-135 as the experiment had been carried out after a shutdown period of at least one week. The diffusion calculations of k-eff by the REBUS code included various levels of poisoning. In the first approach the number densities calculated at the end of the 7 year shutdown period have been used. Then the content of Li-6 and He-3 predicted for the time of experiment has been used. In the first calculations the same content of Li-6 and He-3 has been used for all beryllium blocks in the reactor. Then the beryllium blocks have been divided into ‘inner’ and ‘outer’ types and the contents of poisoning isotopes have been added respectively to each type of blocks. The calculation assumed the ‘inner’ blocks placed in the core region and ‘outer’ blocks placed in the reflector region. No shuffling of the blocks has been assumed. Finally the number densities obtained in the ‘pessimistic’ approach have been applied giving the acceptable agreement with experimental k-eff value, i.e., unity. The results are shown in Table 23 and discussed in Ref. 12.

Table 23. k-eff with various contents of Li-6 and He-3 in beryllium blocks.
Reactor configuration and burn-up from February 97.

Description of the case	k-eff
Li-6 and He-3 from the year 93, ‘inner’ applied in all Be blocks	0.8227
Li-6 and He-3 from the year 93, ‘outer’ applied in all Be blocks	1.0549
Li-6 and He-3 from the year 93, inner and outer Be blocks	0.8736
Li-6 and He-3 from the year 95, inner and outer Be blocks	1.0445
Li-6 and He-3 from the year 96, inner and outer Be blocks	1.0880
Li-6 and He-3 from the year 97, inner and outer Be blocks	1.0699
Li-6 and He-3 from the year 97, inner and outer Be blocks, ‘pessimistic’	1.0275
Li-6 and He-3 from the year 97, inner and outer Be blocks, ‘pessimistic’, corrected for water fractions in Be blocks	1.0242

Li-6 and He-3 from the year 97, inner and outer Be blocks, 'pessimistic', corrected for water fractions in Be blocks, reference MCNP result	1.0015

6.4. Influence of shutdown periods on the beryllium poisoning

The research reactor is operated with shutdown periods. The results presented in Tables 9 and 10 and Fig. 3 have shown the strong influence of the shutdown periods on accumulation of He-3 in beryllium blocks. The Li-6 concentration is not affected directly by the shutdown period. However, accumulation of He-3 modifies the neutron spectrum, and hence the reaction rates. The change of reaction rates, in turn, has the influence on Li-6 content in the following period of reactor operation.

To demonstrate the influence of shutdowns on the He-3 buildup an additional calculation for the period 93-94 has been carried out in 3 approximations:

- The detailed description of reactor operation and shutdown periods (reference case),
- The lumped total hours of operation followed by the lumped shutdown period.
- The lumped shutdown period followed by the lumped operating time.

The comparison of the results for He-3 accumulation is shown in Fig. 9. It can be seen that the lumped operation followed by the lumped shutdown period leads to the strong overestimation of He-3 content while the reverse gives a much smaller underestimation. The exact numbers are here: 5.3E-07 atoms/b-cm for the He-3 number density calculated with the detailed history, 1.2E-06 atoms/b-cm for operation followed by shutdown and 4.3E-07 atoms/b-cm for shutdown followed by operation. In other words, acceptable results can be obtained for lumped shutdown periods followed by lumped operating periods.

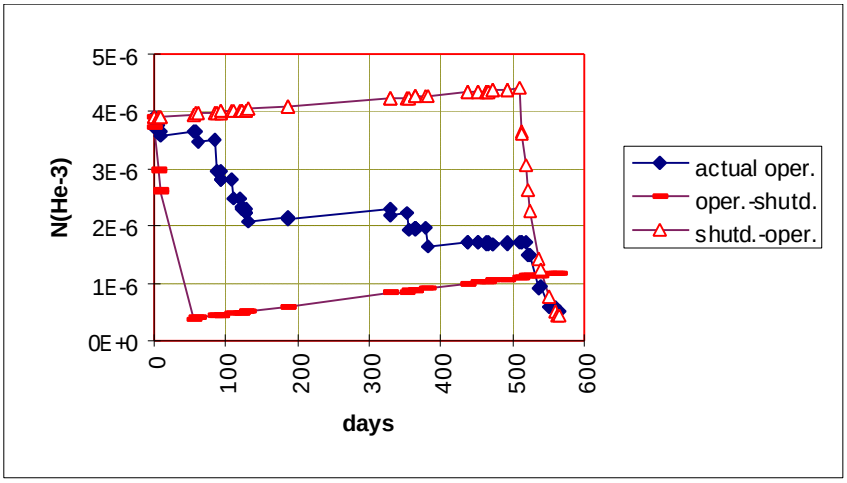

Fig. 9. Changes in He-3 number density [atoms/b-cm] in the period 1993 - 95.

Table 24 shows the effect of reduction of operating hours from 3700 to 2000 per year. The calculations have been carried out with standard cycles of 100 operating hours followed by 68 shutdown hours, with uniform distribution of the cycles over the year. The effect in k-inf will be transferred to k-eff, as it concerns all the beryllium blocks present in the core.

Table 24. Effect of reduction of MARIA operating hours from 3700 to 2000 per year

Operation/Shutdown 3700/4868	Operation/Shutdown 2000/6568

He-3 number density	1.0051E-7	1.4511E-7
Li-6 number density	1.2647E-6	1.2946E-6
k-inf	1.62	1.55

7. CONCLUSIONS

The level of Li-6 and He-3 concentrations in beryllium blocks of the MARIA reactor has been evaluated at different times during the reactor lifetime. It has been shown that the amount of those isotopes accumulated during the reactor history leads to a significant decrease in reactivity.

The distribution of Li-6 and He-3 in the reactor significantly reduces the excess reactivity and has a direct influence on power and neutron flux distributions. Application of effective level of beryllium poisoning is possible but depends on the detailed knowledge of the irradiation history of the beryllium blocks. The rough estimation indicates that a change from blocks internal in the first period of reactor operation to the external - adjacent to fuel channels can lead to the 6% effect in calculated reactivity.

The magnitude and spectrum of the neutron flux have a direct influence on the Li-6 and He-3 build-up through the respective reaction rates. The values of reaction rates depend on several factors of which the most important are shown to be the position in the reactor and the contents of Li-6 and He-3 accumulated already in the beryllium block. Thus, the exact prediction of the reactor poisoning requires the solution of a nonlinear problem. Evaluation of discrepancies caused by application of not updated reaction rates indicates that the 'quasi-static' approach, consisting in modifications of reaction rates only after a major He-3 content modification, is acceptable. An acceptable approximation is therefore to assume that the microscopic reaction rates are constant over the time interval during which the reactor is at power, i.e. few days. However, because of the nonlinearity of the problem, the rates should, in principle, be re-calculated for each subsequent period of operation.

It has been shown that shutdown periods in reactor operation may not be neglected in the analysis as they influence significantly the He-3 buildup from tritium decay. In case of the MARIA reactor the shutdown periods are a very important factor in beryllium matrix poisoning and, hence in the reactivity predictions, because $N(H\text{-}3) >> N(He\text{-}3)$.

It is worth noting that in case of MARIA reactor the time evolution of spatial beryllium matrix poisoning is one of important factors determining conditions of reactor operation and fuel burnup. The irradiation history of beryllium blocks has an essential effect on MARIA reactivity. The seven years' shutdown consequences will be felt during the whole lifetime of presently used beryllium blocks. Reduction of the number of operating hours of the reactor results in an increase of beryllium He-3 poisoning and a subsequent reduction of excess reactivity.

8. REFERENCES

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APPENDIX A

SOLUTION OF EQS. (2)

$$N_{Be}(t) = a_{11} \cdot \exp\{-RR_{Be} \cdot t\}$$
$$N_{L}(t) = a_{21} \cdot \exp\{-RR_{Be} \cdot t\} + a_{22} \cdot \exp\{-RR_{L} \cdot t\}$$
$$N_{T}(t) = a_{31} \cdot \exp\{-RR_{Be} \cdot t\} + a_{32} \cdot \exp\{-RR_{L} \cdot t\} + a_{33} + a_{34} \cdot \exp\{-(\lambda_{T} + RR_{He}) \cdot t\}$$
$$N_{He}(t) = a_{41} \cdot \exp\{-RR_{Be} \cdot t\} + a_{42} \cdot \exp\{-RR_{L} \cdot t\} + a_{43} + a_{44} \cdot \exp\{-(\lambda_{T} + RR_{He}) \cdot t\}$$

where the notation introduced in Eqs. (2-7) has been kept and the coefficients are expressed by initial number densities, reaction rates and tritium decay constant.

For the first equation:

$$a_{11} = N_{Be}(0)$$

$$a_{12} = a_{13} = a_{14} = 0$$

For the second equation:

$$a_{21} = \frac{RR_{Be}}{(RR_{L} - RR_{Be})} \cdot a_{11} = N_{Be}(0) \cdot \frac{RR_{Be}}{(RR_{L} - RR_{Be})}$$

$$a_{22} = N_{L}(0) - N_{Be}(0) \cdot \frac{RR_{Be}}{(RR_{L} - RR_{Be})}$$

$$a_{23} = a_{24} = 0$$

For the third equation:

$$a_{31} = N_{Be}(0) \cdot \frac{(RR_{He} - RR_{Be}) \cdot RR_{L}}{(RR_{L} - RR_{Be}) \cdot (RR_{Be} - \lambda_{T} - RR_{He})}$$

$$a_{32} = \frac{(RR_{He} - RR_{L})}{(RR_{L} - \lambda_{T} - RR_{He})} \cdot \left[N_{L}(0) - N_{Be}(0) \cdot \frac{RR_{Be}}{(RR_{L} - RR_{Be})} \right]$$

$$a_{33} = \frac{RR_{He}}{(\lambda_{T} + RR_{He})} \cdot \left[N_{Be}(0) + N_{L}(0) + N_{T}(0) + N_{He}(0) \right]$$

$$a_{34} = \frac{RR_{L} \cdot \lambda_{T}}{(\lambda_{T} + RR_{He}) \cdot (RR_{L} - \lambda_{T} - RR_{He})} \cdot \left[N_{L}(0) + N_{Be}(0) \cdot \frac{RR_{Be}}{(RR_{Be} - \lambda_{T} - RR_{He})} \right] +$$
$$N_{T}(0) \cdot \frac{\lambda_{T}}{(\lambda_{T} + RR_{He})} - N_{He}(0) \cdot \frac{RR_{He}}{(\lambda_{T} + RR_{He})}$$

For the fourth equation:

$$a_{41} = N_{Be}(0) \cdot \frac{RR_L \cdot \lambda_T}{(RR_L - RR_{Be}) \cdot (RR_{Be} - \lambda_T - RR_{He})} = \frac{\lambda_T}{(RR_{He} - RR_{Be})} \cdot a_{31}$$

$$a_{42} = \frac{\lambda_T}{(RR_L - \lambda_T - RR_{He})} \cdot \left[N_L(0) - N_{Be}(0) \cdot \frac{RR_{Be}}{(RR_L - RR_{Be})} \right] = \frac{\lambda_T}{(RR_{HE} - RR_L)} \cdot a_{32}$$

$$a_{43} = \frac{\lambda_T}{(\lambda_T + RR_{He})} \cdot \left[N_{Be}(0) + N_L(0) + N_T(0) + N_{He}(0) \right] = \frac{\lambda_T}{RR_{He}} \cdot a_{33}$$

$$a_{44} = -\frac{RR_L \cdot \lambda_T}{(\lambda_T + RR_{He}) \cdot (RR_L - \lambda_T - RR_{He})} \cdot \left[N_{Be}(0) \cdot \frac{RR_{Be}}{(RR_{Be} - \lambda_T - RR_{He})} + N_L(0) \right] +$$
$$- N_T(0) \cdot \frac{\lambda_T}{(\lambda_T + RR_{He})} + N_{He}(0) \cdot \frac{RR_{He}}{(\lambda_T + RR_{He})} = -a_{34}$$

APPENDIX B

SOLUTION OF EQS. (6) [3]

$N_{Be} = \text{const; auxiliary quantity introduced } A = N_{Be} \cdot RR_{Be}$

$$N_L(t) = N_L(0) \cdot \exp\{-RR_L \cdot t\} + \frac{A}{RR_L} \cdot (1 - \exp\{-RR_L \cdot t\})$$

where the notation used in Eqs. (2-7) has been kept.

Two auxiliary coefficients, a1 and a2, are introduced:

$$a_1 = \frac{1}{(1+\lambda_T / RR_{He})} \cdot \left[N_L(0) + N_T(0) + N_{He}(0) - \frac{A}{RR_{Li}} + \frac{(\lambda_T \cdot A / RR_{He})}{(\lambda_T + RR_{He})} \right]$$

$$a_2 = \frac{1}{(1 + RR_{He} / \lambda_T)} \cdot \left[\frac{RR_L}{(RR_L - \lambda_T - RR_{He})} \cdot (N_L(0) - A / RR_L) + N_T(0) - \frac{RR_{He}}{\lambda_T} \cdot N_{He}(0) - \frac{A}{(\lambda_T + RR_{He})} \right]$$

With the above notation the solution for tritium and helium number densities can be written:

$$N_T(t) = a_1 + a_2 \cdot \exp\{-(\lambda_T + RR_{He}) \cdot t\} + \frac{A \cdot RR_{He}}{(\lambda_T + RR_{He})} \cdot t + \frac{(RR_{He} - RR_L) \cdot (N_L(0) - A / RR_L)}{(RR_L - \lambda_T - RR_{He})} \cdot \exp\{-RR_L \cdot t\},$$

$$N_{He}(t) = \frac{\lambda_T}{RR_{He}} \cdot a_1 - a_2 \cdot \exp\{-(\lambda_T + RR_{He}) \cdot t\} + \frac{A \cdot \lambda_T}{(\lambda_T + RR_{He})} \cdot (t - 1/RR_{He}) + \frac{\lambda_T \cdot (N_L(0) - A / RR_L)}{(RR_L - \lambda_T - RR_{He})} \cdot \exp\{-RR_L \cdot t\}.$$

APPENDIX C

PROGRAM BERYL

The input data should be stored on file ‘beryl.inp’. The output is stored on file ‘beryl.out’.
Input consists of the following cards:
card 1: initial number densities for Be-9, Li-6, H-3, He-3. (4 numbers)
card 2: reaction rates for Be-9, Li-6, He-3 (3 numbers).
If the first entry on card 2 is zero than the next 2 entries are neglected and the program reads card 3.
card 3: 7 values of the neutron flux i groups defined in section 3.2.
cards 4 - NN: pairs of numbers denoting the time of operation and the time of shutdown in hours. NN has to be less or equal 110.
Note! Program BERYL reads input data in the free format. It implies that all the entries are obligatory, i.e. if the reaction rates are to be calculated still three values on the card 2 have to be present, although the values for Li-6 and He-3 reaction rates are redundant. The same applies to all the input data. For instance: The time of operation or the time of shutdown with zero time length must be specified.

Listing of the program:

```
      PROGRAM BERYL
CALCULATION OF HELIUM AND LITHIUM CONTENTS IN BERYLLIUM MODERATOR
C INPUT DATA ON FILE BERYL.INP, OUTPUT ON FILE BERYL.OUT
C CARD1:4 INITIAL NUMBER DENSITIES IN FREE FORMAT FOR: Be,Li-6,H-3,He-3
C CARD2:3 REACTION RATES IN FREE FORMAT FOR: Be,Li-6,He-3
C IF CARD2 HAS THE FIRST NUMBER <E-19 ADDITIONAL CARD IS NEEDED:
C CARD3:7 FLUX VALUES TO CALCULATE REACTION RATES
C BELOW UP TO 110 PAIRS (EACH PAIR ON A SEPARATE CARD):
C TIME OF IRRADIATION, TIME OF SHUTDOWN IN HOURS
      IMPLICIT REAL*8 (A-H,O-Z)
      DIMENSION TI(110),TD(110),ZERO(4),FLUX(7)
      DIMENSION RNUM(110,6),Y(4)
      DIMENSION HTI(110),HTD(110)
      DATA LAT/1.7814D-09/
      REAL*8 LAT
      OPEN(UNIT=5,FILE='beryl.inp')
      OPEN(UNIT=6,FILE='beryl.out',FORM='FORMATTED')
      READ(5,*)(ZERO(I),I=1,4)
      WRITE(6,100)
      WRITE(6,101)
      WRITE(6,20)
      WRITE(6,21)(ZERO(I),I=1,4)
   20 FORMAT(17X,'INITIAL NUMBER DENSITIES:')
   21 FORMAT(2X,'BE-9=',1P,E11.4,' LI-6=',1P,E11.4,' H-3=',1P,
     &E11.4,' HE-3=',1P,E11.4/)
      READ(5,*)RRBE,RRLI,RRHE
      IF(RRBE.GE.1.0E-19) GO TO 2
      READ(5,*,END=1,ERR=1,IOSTAT=ISTOP)(FLUX(I),I=1,7)
      WRITE(6,22)
   22 FORMAT(8X,'REACTION RATES WILL BE CALCULATED FROM FLUX VALUES:')
      WRITE(6,23)(FLUX(K),K=1,7)
   23 FORMAT(1P,7E10.3/)
      CALL RRATES(FLUX,RRBE,RRLI,RRHE)
      GO TO 2
    1 CALL ERROR(1)
    2 WRITE(6,30)
      WRITE(6,31) RRBE,RRLI,RRHE
   30 FORMAT(18X,'BE RR',7X,'LI-6 RR',5X,'HE-3 RR')
   31 FORMAT(14X,1P,3E12.4)
      WRITE(6,*)' '
      I=0
   10 I=I+1
```

```
      IF(I.GT.111) CALL ERROR(2)
      READ(5,*,END=12,ERR=11,IOSTAT=ISTOP) HTI(I),HTD(I)
      GO TO 10
   11 CALL ERROR(3)
   12 I=I-1
      HI=0.0
      HD=0.0
      NI=I
      DO 13 I=1,NI
      TI(I)=3600.0D0*HTI(I)
      TD(I)=3600.0D0*HTD(I)
      HI=HI+HTI(I)
      HD=HD+HTD(I)
   13 CONTINUE
      WRITE(6,40) NI
   40 FORMAT(13X,'BUILD-UP WILL BE CALCULATED FOR ',I3,' STEPS'/)
      DO 18 I=1,NI
C     CALL TERESA(ZERO,RRBE,RRLI,LAT,RRHE,TI(I),Y)
      CALL MANUEL(ZERO,RRBE,RRLI,LAT,RRHE,TI(I),Y)
      DO 14 N=1,4
   14 RNUM(I,N)=Y(N)
      CALL DECAY(lat,TD(I),Y(4),Y(3),Y(4),Y(3))
      DO 15 N=3,4
   15 RNUM(I,N+2)=Y(N)
      DO 16 M=1,4
   16 ZERO(M)=Y(M)
   18 CONTINUE
      WRITE(6,102)
      WRITE(6,103)
      DO 19 I=1,NI
      WRITE(6,104)HTI(I),HTD(I),(RNUM(I,ITYP),ITYP=2,6)
   19 CONTINUE
      WRITE(6,*)' '
      WRITE(6,105)
      WRITE(6,103)
      WRITE(6,104)HI,HD,(RNUM(NI,ITYP),ITYP=2,6)
      STOP
  100 FORMAT(20X,'PROGRAM BERYL')
  101 FORMAT(3X,'CALCULATES LI-6, H-3, HE-3 BUILD UP IN IRRADIATED BERYL
     &LIUM MATRIX'/)
  102 FORMAT(7X,'HOURS',15X,'NUMBER DENSITIES')
  103 FORMAT(1X,'OPERATION SHUTDOWN  LITHIUM    TRITIUM    HELIUM    H3
     && HE AFTER SHUTD.')
  104 FORMAT(1X,2F9.1,1P,6E11.4)
  105 FORMAT(5X,'TOTAL HOURS',15X,'FINAL NUMBER DENSITIES')
      END
      SUBROUTINE ERROR(N)
      IMPLICIT REAL*8 (A-H,O-Z)
      IF (N.NE.1) GO TO 2
      WRITE(6,100) N
  100 FORMAT(1X,'ERROR ',I2,' , <7 FLUX VALUES')
      GO TO 99
    2 IF (N.NE.2) GO TO 3
      WRITE(6,101) N
  101 FORMAT(1X,'ERROR ',I2,' , TOO LONG IRRADIATION HISTORY')
      GO TO 99
    3 WRITE(6,102) N
  102 FORMAT(1X,'ERROR ',I2,'IN IRRADIATION AND DECAY TIMES')
   99 STOP
      END
```

```
      SUBROUTINE RRATES(fl,rrat)
      IMPLICIT REAL*8 (A-H,O-Z)
      DIMENSION fl(7),rrat(3),sigma(7,3)
      DATA sigma
     &/3.64835E-26,2.79085E-30,0.0,0.0,0.0,0.0,0.0,
C RR(Be) na reakcje (n,alpha)+(n,2n)
C    &/1.30105E-25,2.79085E-30,0.0,0.0,0.0,0.0,0.0,
     &1.96931E-25,1.03530E-24,1.95482E-23,1.24381E-22,
     &2.41717E-22,4.86679E-22,9.70933E-22,
     &7.81605E-25,2.93625E-24,1.11137E-22,7.08028E-22,
     &1.37551E-21,2.76993E-21,5.52947E-21/
      do 20 i=1,3
      rrat(i)=0.0
      do 10 k=1,7
      rrat(i)=rrat(i)+fl(k)*sigma(k,i)
   10 continue
   20 continue
      RETURN
      END

      SUBROUTINE MANUEL(y0,c1,B,xlam,C,t,y)
C solution of 3 equations with Be term as RHS of the first eq
C assumes Be reaction rate much smaller than those of Li-6 and He-3
      IMPLICIT REAL*8 (A-H,O-Z)
      DIMENSION y0(4),y(4)
      y(1)=y0(1)
      A=c1*y0(1)
      e2=dexp(-B*t)
      e34=dexp(-(xlam+C)*t)
      bc=B-(xlam+C)
      ac=A/(xlam+C)
      y(2)=y0(2)*e2+A/B*(1-e2)
      a1=(y0(2)+y0(3)+y0(4)-A/B+xlam*ac/C)/(1+xlam/C)
      a2=(B/bc*(y0(2)-A/B)+y0(3)-C/xlam*y0(4)-ac)
     &/(1+C/xlam)
      a3=(C-B)*(y0(2)-A/B)*e2/bc
      a4=xlam*(y0(2)-A/B)*e2/bc
      y(3)=a1+a2*e34+ac*C*t+a3
      y(4)=xlam/C*a1-a2*e34+xlam*ac*(t-1/C)+a4
      RETURN
      END

      SUBROUTINE TERESA(y0,c1,c2,c3,c4,t,y)
C solution of 4 homogeneous equations (including Be (n,alpha))
      IMPLICIT REAL*8 (A-H,O-Z)
      DIMENSION y0(4),y(4)
      c21=c2-c1
      c41=c4-c1
      c24=c2-c4
      c34=c3+c4
      c134=c1-c3-c4
      c234=c2-c3-c4
      e1=dexp(-c1*t)
      y(1)=y0(1)*e1
      e2=dexp(-c2*t)
      e34=dexp(-c34*t)
      a22=y0(2)-c1/c21*y0(1)
c     y(2)=y0(2)*e2+c1/c21*y0(1)*(e1-e2)
      y(2)=y0(1)*c1/c21*e1+a22*e2
      a31=c41*c2/c134/c21*y0(1)
      a32=-c24/c234*a22
```

```
      a33=c4/c34*(y0(1)+y0(2)+y0(3)+y0(4))
      a34=c2*c3/c34/c234*(c1/c134*y0(1)+y0(2))+1/c34*(c3*y0(3)-c4*y0(4))
      a41=c3*c2/c134/c21*y0(1)
      a42=c3/c234*(y0(2)-c1/c21*y0(1))
      a43=c3/c4*a33
      a44=-a34
      y(3)=a31*e1 + a32*e2 + a33 + a34*e34
      y(4)=a41*e1 + a42*e2 + a43 + a44*e34
      RETURN
      END

      SUBROUTINE DECAY(lat,t,he0,tr0,he,tr)
      IMPLICIT REAL*8 (A-H,O-Z)
      real*8 lat
      exlat=dexp(-lat*t)
      te=tr0*exlat
      he=he0+tr0*(1-exlat)
      tr=te
      RETURN
      END
```

Examples of an input file to be prepared by the user on the file beryl.inp:

Input 1: Reaction rates given in input

```
1.1668E-1 0.0 0.0 0.0
1.367-12 1.297-7 7.384E-7
   3707      6466
   1967      6793
    723      8061
   2215      6545
   2437      6323
   3451      5309
   3616      5168
   2298      6462
    0.0     65688
      24    120
      12    60
      12.5  1187.5
      23.75 576.25
      97    95
      26    22
      2.25  309.75
      72    240
      38.25 9.75
      8.5   135.5
      13.75 10.25
      45    1299
      7     3449
      25.25 502.75
      75.1  236.9
      1.4   22.6
      3.3   308.7
      100.3 1315.7
      3.45  332.55
      5.8   282.2
      9.85  62.15
      8     160
      2.35  429.65
      5.45  522.55
      3.55  92.45
```

```
      81.3  86.7
      266   70
      272   232
      84.6  35.4
```

Input 2: Reaction Rates calculated

```
1.1668E-1 0.0 0.0 0.0
0.0 0.0 0.0
 5.542E+12 1.287E+13 1.382E+13 3.664E+12 2.417E+12 3.092E+13 4.694E+13
   3707      6466
   1967      6793
```

Example of an output file created on beryl.out (for input 1):

```
                    PROGRAM BERYL
   CALCULATES LI-6, H-3, HE-3 BUILD UP IN IRRADIATED BERYLLIUM MATRIX

                 INITIAL NUMBER DENSITIES:
  BE-9= 1.1668E-01 LI-6= 0.0000E+00 H-3= 0.0000E+00 HE-3= 0.0000E+00

                  BE RR       LI-6 RR     HE-3 RR
                1.3670E-12  1.2970E-07  7.3840E-07

       BUILD-UP WILL BE CALCULATED FOR  38 STEPS

       HOURS                NUMBER DENSITIES
 OPERATION SHUTDOWN  LITHIUM    TRITIUM    HELIUM    H3 & HE AFTER SHUTD.
    3707.0   6466.0 1.0119E-06 1.1144E-06 2.2802E-09 1.0691E-06 4.7544E-08
    1967.0   6793.0 1.1428E-06 2.1104E-06 4.8561E-09 2.0204E-06 9.4818E-08
     723.0   8061.0 1.1677E-06 2.4868E-06 1.8681E-08 2.3615E-06 1.4397E-07
    2215.0   6545.0 1.2077E-06 3.7285E-06 8.8657E-09 3.5752E-06 1.6212E-07
    2437.0   6323.0 1.2227E-06 5.1096E-06 1.2044E-08 4.9066E-06 2.1509E-07
    3451.0   5309.0 1.2284E-06 7.0810E-06 1.6585E-08 6.8440E-06 2.5361E-07
    3616.0   5168.0 1.2295E-06 9.1512E-06 2.1573E-08 8.8529E-06 3.1990E-07
    2298.0   6462.0 1.2297E-06 1.0467E-05 2.5387E-08 1.0042E-05 4.5028E-07
       0.0  65688.0 1.2297E-06 1.0042E-05 4.5028E-07 6.5897E-06 3.9025E-06
      24.0    120.0 1.2297E-06 6.8436E-06 3.6623E-06 6.8384E-06 3.6675E-06
      12.0     60.0 1.2297E-06 6.9599E-06 3.5529E-06 6.9572E-06 3.5556E-06
      12.5   1187.5 1.2297E-06 7.0800E-06 3.4400E-06 7.0263E-06 3.4937E-06
      23.8    576.3 1.2297E-06 7.2527E-06 3.2810E-06 7.2259E-06 3.3077E-06
      97.0     95.0 1.2297E-06 8.0292E-06 2.5601E-06 8.0243E-06 2.5650E-06
      26.0     22.0 1.2297E-06 8.2092E-06 2.3950E-06 8.2081E-06 2.3962E-06
       2.3    309.8 1.2297E-06 8.2235E-06 2.3820E-06 8.2072E-06 2.3983E-06
      72.0    240.0 1.2297E-06 8.6628E-06 1.9841E-06 8.6495E-06 1.9974E-06
      38.3      9.8 1.2297E-06 8.8625E-06 1.8064E-06 8.8619E-06 1.8069E-06
       8.5    135.5 1.2297E-06 8.9067E-06 1.7670E-06 8.8990E-06 1.7748E-06
      13.8     10.3 1.2297E-06 8.9698E-06 1.7118E-06 8.9692E-06 1.7124E-06
      45.0   1299.0 1.2297E-06 9.1856E-06 1.5218E-06 9.1094E-06 1.5980E-06
       7.0   3449.0 1.2297E-06 9.1425E-06 1.5690E-06 8.9425E-06 1.7690E-06
      25.3    502.8 1.2297E-06 9.0704E-06 1.6556E-06 9.0412E-06 1.6848E-06
      75.1    236.9 1.2297E-06 9.3852E-06 1.3839E-06 9.3710E-06 1.3981E-06
       1.4     22.6 1.2297E-06 9.3769E-06 1.3930E-06 9.3755E-06 1.3944E-06
       3.3    308.7 1.2297E-06 9.3894E-06 1.3824E-06 9.3708E-06 1.4010E-06
     100.3   1315.7 1.2297E-06 9.7509E-06 1.0785E-06 9.6690E-06 1.1604E-06
       3.5    332.6 1.2297E-06 9.6813E-06 1.1501E-06 9.6607E-06 1.1707E-06
       5.8    282.2 1.2297E-06 9.6816E-06 1.1531E-06 9.6641E-06 1.1706E-06
       9.8     62.1 1.2297E-06 9.6994E-06 1.1410E-06 9.6955E-06 1.1448E-06
       8.0    160.0 1.2297E-06 9.7237E-06 1.1212E-06 9.7137E-06 1.1312E-06
       2.4    429.6 1.2297E-06 9.7220E-06 1.1243E-06 9.6952E-06 1.1511E-06
       5.5    522.5 1.2297E-06 9.7146E-06 1.1349E-06 9.6821E-06 1.1674E-06
```

3.5	92.5	1.2297E-06	9.6949E-06	1.1566E-06	9.6891E-06	1.1624E-06
81.3	86.7	1.2297E-06	9.9571E-06	9.4105E-07	9.9516E-06	9.4659E-07
266.0	70.0	1.2297E-06	1.0572E-05	4.7937E-07	1.0567E-05	4.8411E-07
272.0	232.0	1.2297E-06	1.0959E-05	2.4834E-07	1.0942E-05	2.6463E-07
84.6	35.4	1.2297E-06	1.1039E-05	2.1668E-07	1.1036E-05	2.1918E-07

TOTAL HOURS		FINAL NUMBER DENSITIES				
OPERATION	SHUTDOWN	LITHIUM	TRITIUM	HELIUM	H3 & HE AFTER SHUTD.	
21743.7	129021.3	1.2297E-06	1.1039E-05	2.1668E-07	1.1036E-05	2.1918E-07