BERYLLIUM POISONING IN THE MARIA REACTOR

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ABSTRACT

The poisoning of beryllium blocks initiated by the Be-9(n,α) reaction is discussed in this paper. The present status of the methods used to evaluate Li-6 and He-3 poison concentrations in the MARIA reactor are described. On the basis of conservative assumptions the magnitude of the problem is evaluated. Implications for the reactor operation are formulated, comprising the necessity of keeping long term reactor operation records with special emphasis on beryllium history.

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 Li-6, H-3, He-3 and He-4 isotopes. Buildup of He-3 and Li-6, because of their large thermal neutron absorption cross sections, results in large negative reactivity and flux, spectrum and power distribution modifications.

The negative reactivity has been observed in the beryllium moderated and reflected MARIA reactor after a seven years' break in reactor operation [1]. A systematic study of the problem has been performed with the aim to assist the operator in his decisions [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 operation history. The loss of reactivity caused by shutdown periods, because of tritium decay, has been estimated for the MARIA reactor.

Basic equations

The main reactions leading to beryllium poisoning are:

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

As seen from Eq. (1), beryllium is transformed almost immediately into Li-6. The corresponding equations are:

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

where the dependence of the number densities, N, on time has been omitted to simplify the notation. The subscripts: Be, L, T, and He denote respectively Be-9, Li-6, H-3 and He-3, λ_T is the tritium decay constant equal $1.78 \cdot 10^{-9}$ 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:

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

In Eq. (3) the beryllium loss from the Be(n,2n) reaction is neglected in $\{RR\}_{Be}$. This approximation is justified as the loss can be neglected for the MARIA reactor. It has been estimated that during the MARIA operation in the period 1973-85, the reduction in beryllium density was 0.08% due to (n,2n) reaction and 0.03% due to (n,α) reaction.

The energy interval (E_{min}, E_{max}) covers the entire range of neutron energies and σ_x (E) is the relevant neutron-induced cross section for isotope x. It should also be noted that when reactor is off power, i.e. the neutron flux $\varphi(E,t) = 0$, the last two equations of the system (2) describe the tritium decay and He-3 buildup. The problem (Eqs. 2) is nonlinear because the reaction rates are time-dependent since the flux and spectrum depend on the poison concentrations.

Computational tools and methods

The equations (2) have been linearized by assuming constant reaction rates over a specified time interval. The solution of the simplified system is performed in 2 steps:

1. Calculation of reaction rates, i.e., equation coefficients.
2. Analytic solution of the system (2) with given initial number densities for beryllium, lithium, tritium and helium.

The accuracy of above approach has been checked by additional calculation [2].

The following programs were used in calculations of the reaction rates:

- Lattice spectrum codes WIMSD5 [3] and WIMS-ANL [4] using respectively the WIMS ‘86’ library of Winfrith and the ANL library based on ENDF/B-VI.
- The transport code TRITAC [5,6] in 2 dimensions and the diffusion and fuel management code REBUS in 3 dimensions [7].

Solution of equation (2) is performed using program BERYL [2], based on the analytic formulas. 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 given in input or calculated by the program from the 7-group microscopic 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+7, 8.21E+5, 5.53E+3, 4.0E+0, 6.25E-1, 2.5E-1, 5.8E-2.

The 7-group cross sections for beryllium in the MARIA asymptotic cell were calculated by WIMS, assuming fresh fuel. Fluxes in beryllium were obtained either from calculations of an asymptotic cell or from the whole reactor calculations in a chosen configuration by REBUS.

The character of transmutations taking place during operation of MARIA can be seen in Fig. 1. The solution of Eqs (2) for the asymptotic cell is presented. History of irradiation comprised 700 days at power followed by 700 days of tritium decay and again 700 days at power. The reaction rates were assumed constant over the whole period considered. Initial conditions for the solution of Eqs. (2) were $N_{Be}(0) = 0.11668$ atoms/b-cm, $N_{Li}(0) = N_H(0) = N_{He}(0) = 0$.

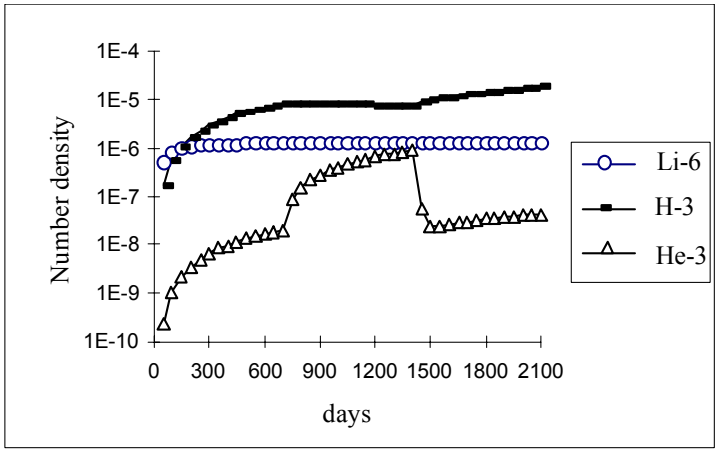

Fig. 1. Solution of Eq. (2) with constant reaction rates.

It can be seen that Li-6 density saturates in approximately 200 days, while H-3 and He-3 density is growing with time. It is worth noting that during the 700 days of shutdown 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.

Spectral effects

The buildup of He-3 and Li-6 in beryllium leads to spectrum hardening. Thus assuming that the reactor is operated at constant power, the ratio of beryllium reaction rate to that of lithium in Eq. (2) increases and so does the density of Li-6 and hence He-3. In the BERYL program the process of spectrum hardening is simulated changing reaction rates at specified time steps.

The operation of a research reactor is performed in cycles including operational breaks. During each break the amount of He-3 increases exponentially with time, causing an additional spectrum hardening after return to power. The result of this process is shown in Figs.2 and 3 in terms of Li-6 and He-3 number densities. The results have been calculated by WIMS and BERYL for the beryllium block of the MARIA reactor asymptotic cell, for average channel power and resulting temperatures. Four cases were considered. The first, denoted 'RRconst_nb', corresponds to constant reaction rates, and operation without breaks. The next case, denoted 'RRmod_nb', has been obtained with reaction rates recalculated after each 8736 hours of reactor operation. It is seen that in this case the Li-6 number density does not reach equilibrium. The He-3 number density grows slightly faster than in the first case. Both cases have

been then recalculated assuming weekly cycle of 100 on and 68 hours off power, plus one additional week off power every fourth week, denoted 'wb'. With the assumption of constant reaction rates, the breaks do not influence the Li-6 buildup and the asymptotic value of its number density is reached. When spectral changes are considered, the increase in Li-6 buildup is observed. As can be seen in Fig.3, spectral changes lead to a rather week increase of He-3 number density, while considering the operation with breaks increases significantly the amount of He-3.

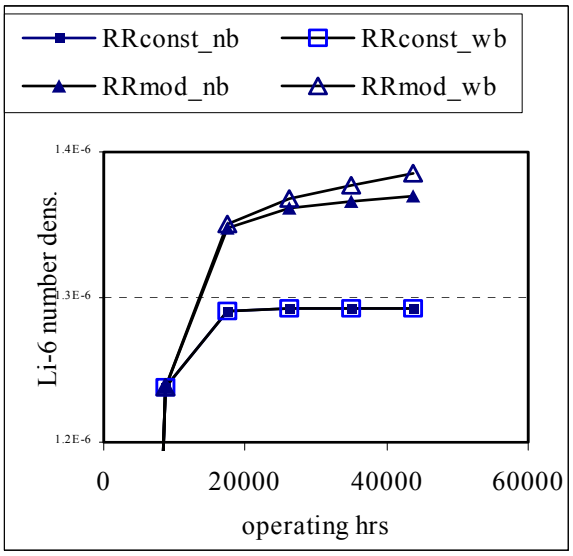

Fig. 2. Buildup of Li-6 in an infinite lattice.

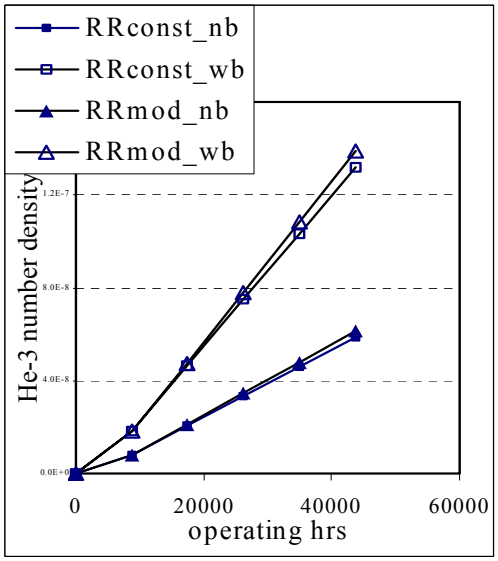

Fig. 3. Buildup of He-3 in an infinite lattice.

The presence of He-3 and Li-6 changes global parameters of a reactor. These changes are shown in terms of absorption in beryllium blocks (Fig. 4) and the infinite multiplication factor of the reactor cell (Fig. 5). Both quantities have been calculated considering spectral changes and the figures show the influence of operational breaks. Absorption is given as a ratio of absorption in the beryllium blocks to that in the fuel element channel.

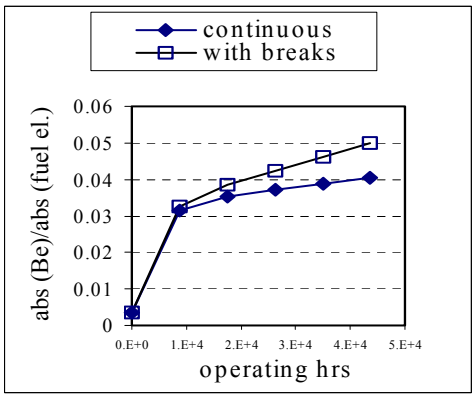

Fig. 4. Dependence of absorption in beryllium on operating time.

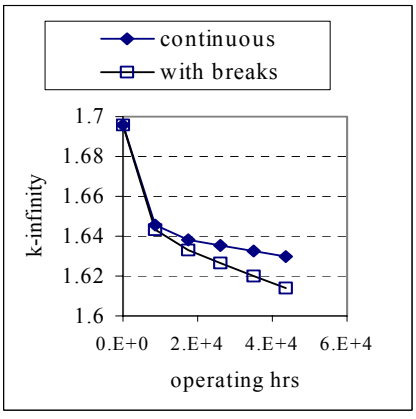

Fig. 5. Dependence of k-infinity on operating time.

Spatial effects

The spatial effects of beryllium poisoning were considered for the realistic history and fuel loading, cf. Fig. 6, of the MARIA reactor.

Total time of operation of the MARIA reactor in the period 1977 - 1985 was 20414 hours. For this period the average power output from one assembly was assumed 1.06 MW. Starting from the middle of 1985 the reactor had a break in operation of seven and half years. During the break the beryllium blocks have been reshuffled. The reactor achieved criticality in the middle of 1993 and up to the end of 1994 was operated for irregular periods with average power per fuel assembly of about 1.1 MW. Thus, in the calculations of Li-6 and He-3 poisoning the following periods had to be considered:

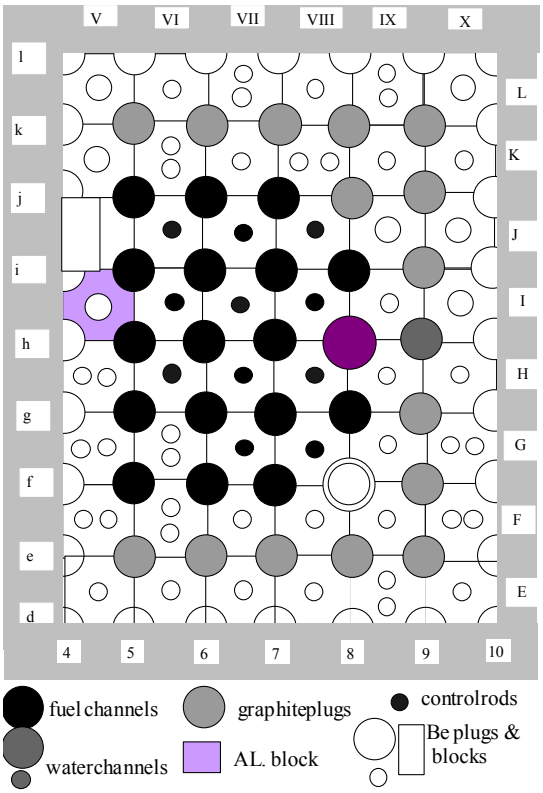

Fig. 6. The horizontal cross section of the MARIA core

1. operation from 1977 to 1985,
2. the shutdown for reconstruction,
3. irregular operation in 1993-94,
4. regular operation since 1995.

The poisoning of the beryllium blocks for the first period of the reactor operation has been calculated taking into account the number of on-power and off-power hours for each year. The neutron flux spectrum and flux levels have been calculated for a 16 fuel element configuration with nearly equilibrium burnup and power distribution obtained from auxiliary REBUS calculations. Beryllium blocks have been grouped in 2 zones: 'inner', comprising beryllium blocks surrounded by fuel elements, and 'outer', comprising the rest of beryllium blocks in the core. The reaction rates have been recalculated for each year using average contents of Li-6 and He-3 for each beryllium zone

Starting from poison number densities at the end of year 1985, the buildup of He-3 from H-3 decay during 7 years of reactor shutdown has been calculated and is shown in Table 1.

Table 1: Number densities after the first and second period.

	End of 85		Middle of 93	
	Be zone		Be zone	
Isotope	'inner'	'outer'	'inner'	'outer'
Li-6	1.230E-6	3.955E-7	1.230E-6	3.955E-7
H-3	1.004E-5	1.274E-6	7.059E-6	8.597E-7
He-3	4.503E-7	6.528E-9	3.426E-6	4.208E-7

For the last two periods of reactor operation, the detailed history of on-power and off-power periods has been taken into account. The calculations were performed in cycles, starting with the amount of Li-6, H-3 and He-3 from the middle of 93. Again, a nearly equilibrium configuration of the MARIA reactor has been chosen. Fluxes and k-eff have been calculated using REBUS. Next, the BERYL program has been used to calculate reaction rates from the average REBUS flux values for inner and outer beryllium blocks and the values of Li-6 and He-3 at the beginning of 94. The cycle has been repeated for years 1994-1999. The results are shown in Figs. 7-11.

It is worth noting that during the long shutdown of the MARIA reactor a considerable amount of He-3 accumulated (from tritium decay) which started to burn out under irradiation in 1993. It is seen in Figs.7 and 8 that independently of the position in the reactor He-3 number density was decreasing for 2.5 years before it started to increase.

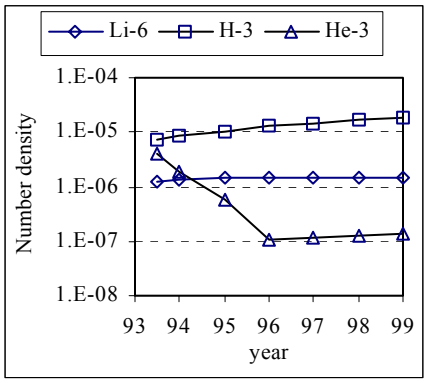

Fig.7. Number densities in inner beryllium blocks.

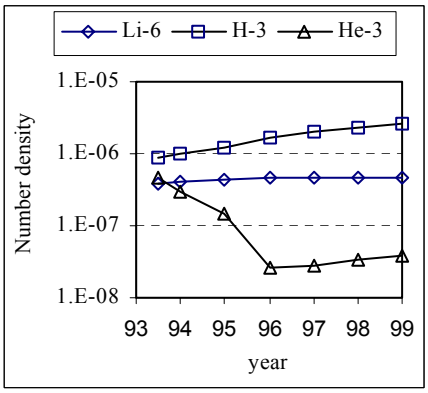

Fig.8. Number densities in outer beryllium blocks.

The differences in the level of parasitic absorption and flux spectrum between the inner and outer blocks are shown in Figs.9 and 10. As before, absorption, per unit volume, is given in terms of a ratio of absorption in beryllium to that in the fuel channel. Flux spectrum index is expressed as the ratio of fast to thermal flux with the cutoff at 0.625 eV. Finally the effective multiplication factor is given in Fig.11.

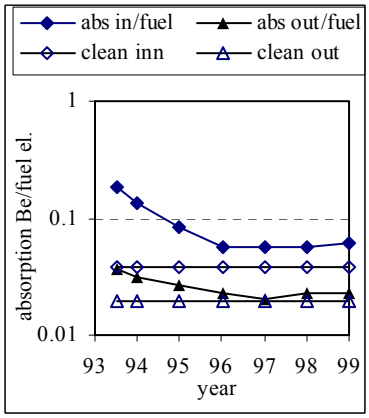

Fig.9. Ratio of Be to fuel channel absorption

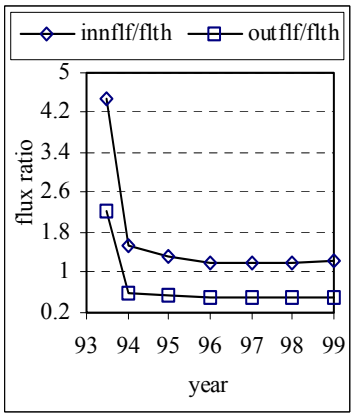

Fig.10. Ratio of fast to thermal flux

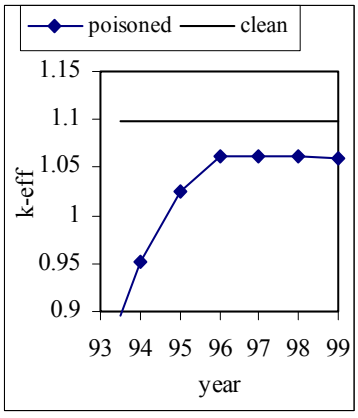

Fig.11. K-effective

Inaccuracies in of Li-6 and He-3 contents prediction

The above results include errors coming from two main sources: the computational methods and the deficiencies in operational records. In the following the factors influencing poison accumulation, not accounted for in the present study are listed.

1. Fuel burnup: The beryllium poisoning has a rather weak influence on the fuel burnup process. The absorption in fuel channel (fuel + aluminum + water), calculated with clean beryllium is 4% lower than with strongest poisoning observed in 1993, while the absorption in beryllium increases 4.5 times in inner blocks and 1.8 times in outer blocks. Thus, no wonder that, e.g. the U235 depletion after 70 days of nominal power operation is affected by the maximum calculated poisoning only at the fourth significant digit. The reverse, i.e. the influence of the fuel burnup level on Li-6, H-3 and He-3 buildup takes place through the spectral changes which in beryllium are again much weaker than those caused by the Li-6 and He-3 buildup. Thus the lack of detailed fuel burnup history in the present analysis has been found to be of a minor importance when compared to other sources of errors.
2. Power distribution: The actual power distribution has a strong influence on Li-6, H-3 and He-3 content, through the difference in flux levels in beryllium blocks. The lowest operational channel power level is usually about 0.6 - 0.7 MW while the peak power is about 1.4 - 1.6 MW. It means that the flux level in beryllium blocks adjacent to particular fuel channels differs strongly and so does the content of Li-6 and He-3. In our study only one, 'inner' zone with average neutron flux has been considered. For blocks in beryllium reflector analogous differences in flux levels can occur.
3. Poison distribution within the beryllium block: The beryllium block has horizontal cross section of 13*13 cm. A separate analysis has shown differences in Li-6 and He-3 content within the beryllium block from the fuel channel edge to the middle of the block estimated for at least 20% in Li-6 and 19% in He-3 number densities [2]. The similar effect has to be expected in the vertical direction due to the flux vertical distribution. Thus the poisoning should be in fact much higher in the central zone of the core. Besides, the vertical poison distribution will influence the burnup profile of the fuel.
4. Step length for reaction rates update: The calculations may include errors caused by too long time steps between recalculation of reaction rates. The problem includes both spectral and power level effects. Separate numerical tests have been carried out to estimate those errors [2] and to choose a sufficiently long time step. The remedy would be to follow the spectral changes exactly, as it is done in the fuel depletion calculation.
5. Update of core configuration: The presented analysis has been carried out for chosen, quasiequilibrium reactor core configurations. It has been found that a change of configuration applied in the present analysis to another one, also taken from the MARIA operation history has changed the resulting k-effective values by 0.004. This can be considered as the order of error introduced by not following the exact fuel loading scheme.
6. History of reactor operation: Large error can be introduced through unreliable operational records. The extension of operational breaks in the period 95-97 by 5 hours each has lead to the decrease of k-effective by 4%. Thus the exact figures of on- and off-power records have to be applied.
7. Beryllium blocks history: It is felt that the distribution of the poison may influence the power production. For MARIA reactor, the effect of the replacement of 3 beryllium blocks, at positions L-VI, K-VII, K-VIII of Fig.6 in 95 reactor core, with low Li-6 and He-3 content by blocks with high content has been checked. The effect in reactivity was only 0.4%, but modification in the power distribution was up to 10 % in the vicinity of the replaced blocks and several percent in the distant fuel channels [2]. The effect in k-eff is slightly misleading because the blocks are placed at the core periphery.

Conclusions

Numerical prediction of beryllium poisoning is necessary because of lack of experimental methods of its determination.

The influence of beryllium poisoning on reactivity, absorption in beryllium blocks and neutron flux spectrum has been shown. A strong dependence of the amount of created He-3 and Li-6 on the operational breaks has been observed.

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 are felt during the whole lifetime of presently used beryllium blocks.

To predict reliable values of beryllium poisoning in each block, the inclusion of beryllium transmutation chains into a fuel management code is necessary. To be able to simulate the transformations of Be9, Li-6, H-3 and He-3 in each block, the records of reactor operation have to include not only the history of fuel elements but also of each beryllium block. The exact power operation history should be used in the analysis.

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