Impact of Photo neutrons on Reactivity Worth of ^{3}He In a Reactor with Beryllium Reflector

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Abstract

A study about the contribution of photo neutrons, produced in (γ,n) – reactions of ^{9}Be into the reactivity worth of ^{3}He in a reactor with beryllium reflector is presented. Neutron interactions with beryllium lead to formation of ^{3}T and strong neutron absorbers ^{3}He and ^{6}Li in the reflector (so called beryllium poisoning). After the reactor shut down, the concentration of helium – 3 increases in time due to tritium decay. The negative reactivity effect of neutron absorption by accumulated ^{3}He has been considered in a few papers, concerning reactivity losses in reactors with beryllium reflector. The contributions of the photo neutrons for additional production of tritium during the reactor operation and enhanced build up of helium – 3 after the shut down have not been discussed previously. Calculations of the photonuclear reactions in beryllium are performed for the Belgian Material Testing Reactor BR2 and for a generic reactor model with beryllium reflector. The calculation analysis has shown an increase of about ~ 20% in the reactivity worth of helium – 3 by photo neutrons. The effective fraction of photo neutrons is estimated using MCNP5 with included (γ,n) – reactions.

KEYWORDS: photo neutrons, beryllium, helium – 3, MCNP5

1. Introduction

Studies about the contributions of photo neutrons into the reactivity worth of beryllium have been reported only in a few papers, concerning reactors with beryllium reflector. It was found that (γ,n) – reactions of beryllium insert a small fraction of positive reactivity worth. The effective fraction of the photo neutrons in the reactor BR2 has been determined experimentally more than 30 years ago and it was obtained the value: $\beta_{phot,n}^{measur} = 0.00065$ [1] ($\beta_{eff}^{BR2} = \beta_{del,n+phot,n}^{BR2} = 0.0072$). The contribution of (γ,n) – reactions into the reactivity worth of beryllium, determined with the RADHEAT–V3 code system was equal to: $\beta_{phot,n}^{[2]} \approx 1 \times 10^{-5}$ [2].

Other aspects of the impact of the photo neutrons on the reactivity worth reveal when consider the poisoning effect of beryllium. During the reactor operation beryllium is irradiated by neutrons, which produce ^{3}T and strong neutron absorbers ^{6}Li and ^{3}He in the beryllium reflector. After the reactor shut down, the concentration of ^{3}He increases in time due to the decay of ^{3}T. The negative reactivity effect of the neutron absorption by accumulated ^{3}He, only caused by fission neutrons, has been considered in a few papers concerning reactors with beryllium reflector [3–5]. It was found that the reactivity losses due to the helium–3 poisoning after a long shut down of the reactor can reach large values – up to $\Delta k/k \approx -6\%$ in the 2nd beryllium matrix of the BR2 reactor [3] and about –20% in the Maria reactor after 7.5 years break down of the reactor operation [4].

Investigations about the contribution of the photo neutrons for additional production of ^{3}T during the reactor operation and consequently increased accumulation of ^{3}He after the shut down are not available in the reactor physics literature at the present moment. In this paper we present a calculation analysis for the impact of the photo neutrons, produced through (γ,n)-reactions of ^{9}Be on the reactivity worth of ^{3}He in a beryllium reflected reactor. Detailed calculations for the various contributors into the reactivity worth of helium – 3 are performed for the Belgian Material Testing Reactor BR2 and for a generic reactor model, composed by a fuel assembly in the center and surrounded with beryllium reflector. The effective fraction of the photo neutrons is estimated, using MCNP5 [6] with included (γ,n)–reactions.

2. Beryllium poisoning

The poisoning of beryllium is a complex process, which is caused by neutron absorption of all energies. The poisoning effect starts after the threshold (n,α) – reaction on beryllium–9, irradiated by fast neutrons with energy $E_n > 0.69$ MeV and following transmutations into nuclides of ^{6}Li, ^{3}He and ^{3}T according to the reactions [3–5]:

$$^1_0n+^9_4Be\rightarrow^6_2He+^4_2He;\quad ^6_2He\rightarrow^6_3Li+^0_{-1}e+\nu_{e^-}\qquad (T_{1/2}^{^6He}=0.808s) \tag{1}$$

$$^1_0n+^6_3Li\rightarrow^3_1T+^4_2He; \tag{2}$$

$$^1_0n+^3_2He\rightarrow^3_1T+^1_1p \tag{3}$$

$$^3_1T\rightarrow^3_2He+^0_{-1}e+\nu_{e^-}\qquad (T_{1/2}^{^3T}=12.33\text{years}) \tag{4}$$

The lithium–6 and helium–3 isotopes have very high absorption cross section of thermal neutrons: $\sigma_a^{^6Li}=942$b and $\sigma_a^{^3He}=5330$b [6]. The equilibrium level of ^{6}Li is determined by formation in a fast neutron spectrum according with (1) and burn up according with (2). After some time of the reactor operation the concentration of ^{6}Li reaches saturation. During the reactor operation ^{3}He is burning out according to the reaction (3) and the tritium concentration increases with the energy produced. After the shut down of the reactor, the concentrations of ^{3}He increases due to the decay of ^{3}T (4), thereby causing reactivity losses of the reactor core. Taking into account the radial and axial distributions of the neutron spectra in the reactor core detailed 3 – D distributions of the reaction rates (1)÷(3) have been calculated in each position of the beryllium matrix in BR2 [7]. After that the reaction rates, caused by fission neutrons: $RR_k^n=\int\varphi(r,E,t)\sigma_{n,l}^k(E)dE$ have been introduced into the following system differential equations for evaluation of the concentrations of ^{6}Li, ^{3}He and ^{3}T versus time [8]:

$$\begin{aligned}
\frac{dN_{Be}(r,t)}{dt} &= -N_{Be}(r,t)\int\varphi(r,E,t)\sigma_{n,\alpha}^{Be}(E)dE\ ,\\
\frac{dN_{Li}(r,t)}{dt} &= N_{Be}(r,t)\int\varphi(r,E,t)\sigma_{n,\alpha}^{Be}(E)dE-N_{Li}(r,t)\int\varphi(r,E,t)\sigma_{n,\alpha}^{Li}(E)dE\ ,\\
\frac{dN_{T}(r,t)}{dt} &= N_{Li}(r,t)\int\varphi(r,E,t)\sigma_{n,\alpha}^{Li}(E)dE+N_{He}(r,t)\int\varphi(r,E,t)\sigma_{n,p}^{He}(E)dE-\lambda_T N_T(r,t)\ ,\\
\frac{dN_{He}(r,t)}{dt} &= -N_{He}(r,t)\int\varphi(r,E,t)\sigma_{n,p}^{He}(r,E)dE+\lambda_T N_T(r,t)\ ,
\end{aligned} \tag{5}$$

Where N_{Li}, N_T, N_{He} are atomic concentrations of ^{6}Li, ^{3}T and ^{3}He in Be matrix, $\sigma^k_{n,l}$ is the microscopic cross section of nuclide k for reaction (n,l), φ(r,E,t) is a neutron flux density, and λ is a decay constant for

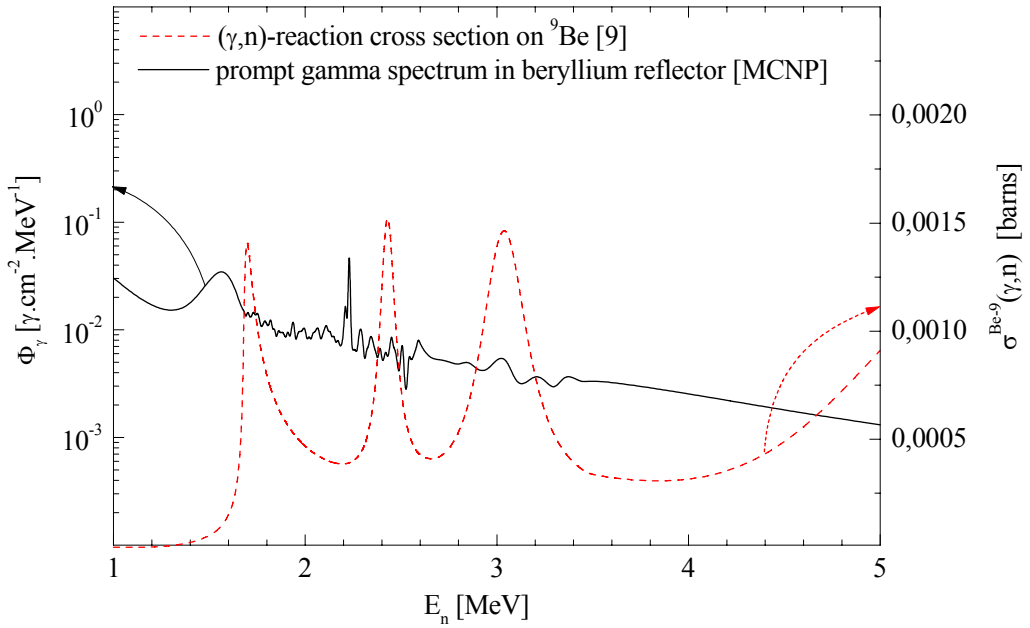
^{3}T. The calculations of the reaction rates (1)÷(3) have been performed with MCNP–4C [7], in which the photonuclear physics is not included. In this paper we present calculations of the reaction rates (1)÷(3) by photo neutrons using MCNP5 with included (γ,n)–reactions.

3. Photonuclear reactions on ^{9}Be

The photonuclear reaction begins with absorption of a photon by nucleus. Once the photon has been absorbed by the nucleus, one or more secondary particle emissions can occur. For the energy range below 150 MeV [6] the excited nucleus so formed may eject a combination of gamma–rays, neutrons, protons, deuterons, tritons, helium–3 particles, alphas, and fission fragments. The typical gamma radiation in the reactor has continuous energy spectrum with maximum energy below 5 MeV (see Fig. 1) and the secondary particles of interest in this paper, which are emitted after (γ,n) – reactions on beryllium are neutrons and gamma–rays. Beryllium has significant (γ,n)–reaction cross section, with low gamma–rays threshold energy of $Q_{\gamma}^{Be} \approx 1.57$ MeV [9], which is depicted on the same Fig. 1:

$$ {}^{9}_{4}\mathrm{Be} + {}^{0}_{0}\gamma \rightarrow 2\ {}^{4}_{2}\mathrm{He} + {}^{1}_{0}\mathrm{n}, \quad Q_{\gamma} = -1.57 \ \text{MeV} \qquad (6) $$

Figure 1. Typical prompt gamma spectrum, calculated with MCNP in the beryllium matrix (reflector) of the reactor BR2 and cross section of the photonuclear reaction (γ,n) on beryllium [9]. Normalization of the γ–spectrum: to the total number of the prompt photons, produced in 1 fission event.



During the reactor operation a large gamma flux exists inside and around the core. The intensity of the gamma flux is about ~ 15 times higher than the intensity of the fission neutron flux. The main emitters of photo neutrons in the reactor are:

(i) Prompt gamma rays, emitted ~ at the time as the fission has occurred; the average number of the emitted prompt γ – rays per fission event is $N_{\gamma}^{prompt} \approx 7.5$ with average energy about $E_{\gamma}^{prompt} \approx 0.72$ MeV (MCNP calculations);

(ii) Delayed gamma rays, released during the beta decays of fission products; the average number of the emitted delayed γ – rays per fission event is $N_{\gamma}^{delayed} \approx 6.5$ with average energy about $E_{\gamma}^{delayed} \approx 0.45$ MeV (coupled MCNP and SCALE4.4a/ORIGENS [10] calculations);

(iii) Captured gamma rays, resulting from (n,γ) – reactions on various structural materials, which total number is $N_{\gamma}^{captured} \approx 1$ with average energy of about $E_{\gamma}^{captured} \approx 2.0$ MeV (MCNP calculations).

The energies of the emitted photo neutrons are equal to the difference between the energy of the incident gamma ray E_{γ} and the threshold energy of the (γ,n)–reaction Q_{γ}, so that the energies of the photo neutrons in the reactor may vary between thermal and up to about a few MeV. Further, the photo neutrons behave like usual neutrons, i.e. slowing down in multiplication system they can cause fission, (n,xn) – reactions and undergo various reactions of neutron absorption: (n,γ), (n,p) (n, α), etc.

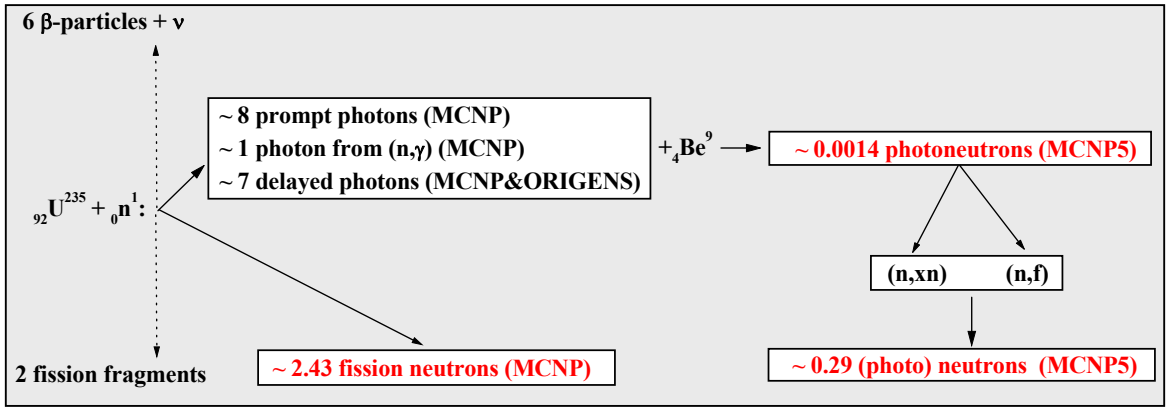
In this paper we adopt the following conception of photo neutron, which includes:

A. Photo neutron born in (γ,n) – reactions of beryllium–9;

B. Secondary neutrons produced by photo neutron in fission, (n,xn) – reactions, etc.

The role of the photo neutrons in the entire fission process is schematically demonstrated on Fig. 2.

Figure 2. Particle balance in fission process, including the emission of photo neutrons by (γ,n) – reactions of beryllium–9 in a generic reactor model with beryllium reflector. The total number of the photo neutrons is evaluated using MCNP5 with included photonuclear reactions.



4. Effective fraction (reactivity worth) of photo neutrons

The effective part of the photo neutrons (or reactivity worth) in this study is defined as the difference between the k_{eff} with included photonuclear reactions (excess reactivity with photo neutrons) and k_{eff} without (γ,n) – reactions (excess reactivity without photo neutrons). Calculations of the neutron – photon balance in a simple cylindrical critical ^{235}U (~ 90%) fueled reactor with one fuel assembly in the center, cooled by light water and surrounded with beryllium reflector as shown at Fig. 3a, are performed using MCNP5. The same evaluations were performed for the detailed 3 – D heterogeneous geometry model of the Belgian Material Testing Reactor BR2 with beryllium matrix, which is given at Fig. 3b. Detailed calculations of the neutron balance in the reactor BR2 – neutron creation and neutron loss – are presented in Table I. The effective multiplication factor for an eigenvalue problem with included or with not included (γ,n) – reactions is calculated and printed automatically by MCNP/MCNP5. For the mode

problem with included (γ,n) – reactions, the estimation of k_{eff} is made only taking into account the contribution from fission neutrons and from photo neutrons, produced in photonuclear reactions of prompt photons with ^{9}Be. In order to estimate the contribution from photo neutrons, produced by the delayed and captured photons, we can use an approximate equation for k_{eff}:

$$k_{eff} = \frac{\nu_f^n * \left(F^n + \sum_i N^{ph_i} * F^{ph_i} \right)}{\left(A^n + \sum_i N^{ph_i} A^{ph_i} \right) + \left(F^n + \sum_i N^{ph_i} F^{ph_i} \right) + \left(L^n + \sum_i N^{ph_i} L^{ph_i} \right) - \left(N_{n,xn}^n + \sum_i N^{ph_i} N_{n,xn}^{ph_i} \right)} \quad (7)$$

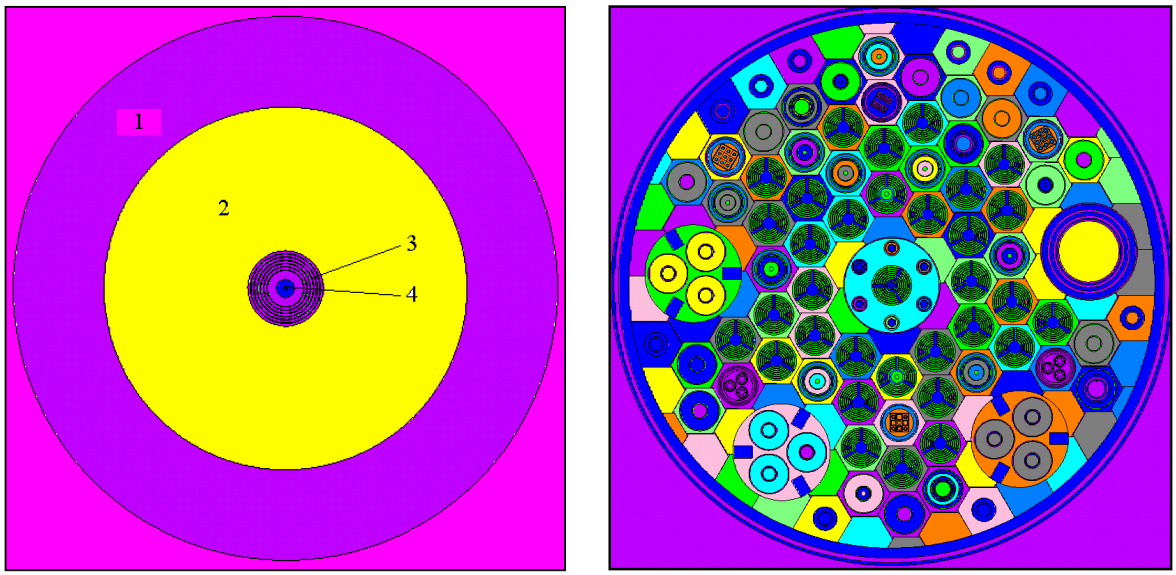
Where: $A = \int_E \int_V \Sigma_a(E)\varphi(E,r)dEdV$ and $F = \int_E \int_V \Sigma_f(E)\varphi(E,r)dEdV$ are the total absorption and the total fission in the reactor per source particle; ν_f^n is the average number of fast neutrons emitted per 1 fission event; L is the leakage term, $N_{n,xn}$ – multiplicity term due to (n,2n) – reaction on ^{9}Be. The superscript "n" is used to denote the contribution into the effective neutron multiplication factor from fission neutron in an eigenvalue problem (column 1 or column 2 in Table I). The superscript "ph_i" is notation for photo neutron, which is produced by the photon of type "i", i=prompt, captured or delayed photon in (γ,n)–reactions of ^{9}Be in a coupled photon – neutron problem with fixed external γ–source (columns 3,4,5 in Table I); N^{ph_i} is the number of photons per 1 fission event.

Table I. Neutron balance in the reactor BR2, calculated with MCNP for different sources of neutrons. Normalization: per 1 source particle in 1 fission event.

Column N°		1	2	3	4	5
Source particle (S.P.)		1 fission n	1 fission n	1 prompt γ	1 captured γ	1 delayed γ
Notation		n	n	ph_{pr}	ph_c	ph_d
N° S.P. per 1 fission event		2.43	2.43	7.32	0.8	6.5
(γ,n) – reaction		off	on	on	on	on
Computational code		MCNP/MCNP5	MCNP5	MCNP5	MCNP5	MCNP5
Source mode		eigenvalue	eigenvalue	external γ	external γ	external γ
Neutron Creation	(γ,n)	0	6.3635E-04	9.6867E-05	5.3512E-04	3.4494E-05
	(n,xn)	0.10753	1.0794E-01	2.4406E-03	1.0329E-02	7.3784E-04
	(n,f)	1	1	2.2645E-02	9.5116E-02	6.7324E-03
Neutron Loss	(n,γ), A	6.4224E-01	6.4228E-01	1.4576E-02	6.1441E-02	4.3306E-03
	(n,f), F	4.1152E-01	4.1180E-01	9.3858E-03	3.9383E-02	2.8094E-03
	(n,xn), $N_{n,xn}$	5.3763E-02	5.3970E-02	1.2203E-03	5.1646E-03	3.6892E-04
	Leakage, L	0	0	0	0	0

Using the data in column 1 & column 2 (or column 1& columns 3,4,5 in Table I) along with Eq. 7 we can determine the effective multiplication factors for the cases without photonuclear reactions and with included photonuclear reactions: $k_{eff}^{n} \approx 1.00001$; $k_{eff}^{n+ph} \approx 1.00068$. Then the effective fraction of the photo neutrons in a ^{235}U (~90%) fueled reactor with beryllium reflector is: $\beta_{eff,BR2}^{ph} \approx 0.00067$ versus the experimental value: $\beta_{phot,n}^{measur} = 0.00065$ [1].

Figure 3. a) MCNP model of a simple cylindrical critical reactor with beryllium reflector and ^{235}U (~90%) in the center, cooled by light water. Legend: 1 – is light water; 2 – beryllium, 3 – uranium plates; 4 – aluminum, Be or H_2O in the center; b) 3–D MCNP model of BR2 reactor.



a b

5. Reactivity worth of ^{3}He by photo neutrons

5.1 Calculation of photon spectra in beryllium reflector

Similarly to the fission neutrons, the photo neutrons produce an additional amount of ^{3}T during the reactor operation, which decays into additional amount of ^{3}He after the reactor shut down and contributes into the total reactivity loss. Any version of MCNP can be used for calculations of the reaction rates (1), (2) and (3), generated by fission neutrons in an eigenvalue problem. The reaction rates generated by photo neutrons, which are produced in photonuclear reactions of prompt, captured and delayed gamma rays with beryllium, are evaluated with MCNP5 in a separated, coupled photon – neutron calculation with an external γ–source, having the corresponding energy distribution. The gamma spectra of different photon types were calculated in the beryllium matrix of the BR2 reactor and they are presented at Fig. 4.

The intensity and the energy spectra of the prompt and captured photons can be computed directly with any version of MCNP. The spectra and the intensity of the delayed photons, escaped from the fission products, accumulated in the fuel elements during irradiation in the reactor, are determined using the code MCNP along with ORIGENS of the SCALE4.4a system. MCNP is used for calculation of the 3 – D power distribution in the core, which is introduced into ORIGENS and used for evaluation of the spectra and the intensity of the delayed photon sources in each fuel element (see Table II).

Figure 4. Photon spectra in the beryllium matrix (reflector) of the BR2 reactor, caused by prompt and delayed photons and by photons, escaped in (n,γ) – reactions. Normalization: to the total number of the photons of type "i", i=prompt, delayed, captured photon, produced in 1 fission event.

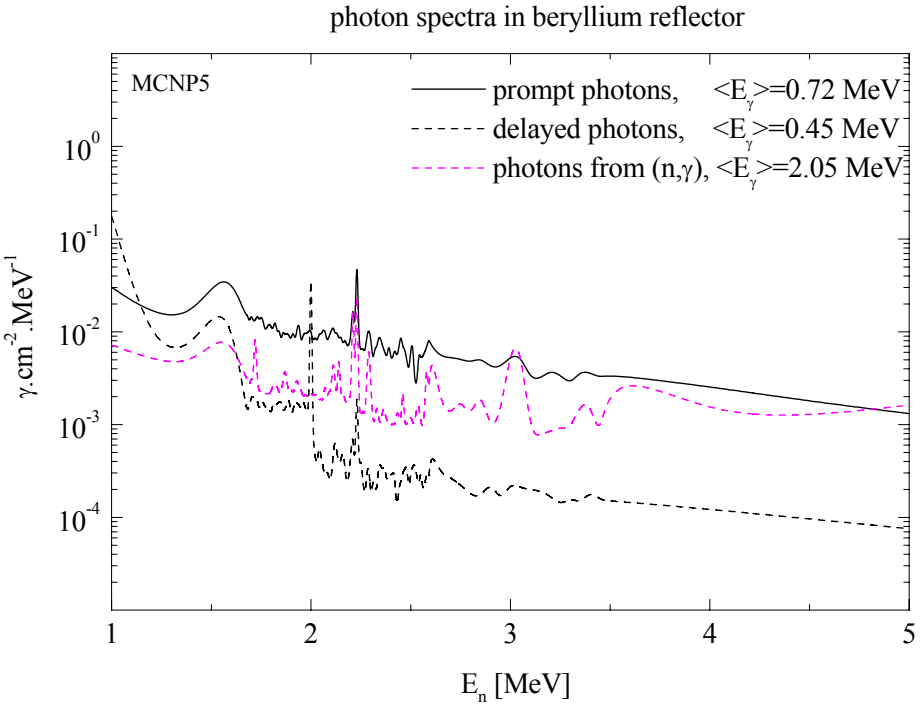


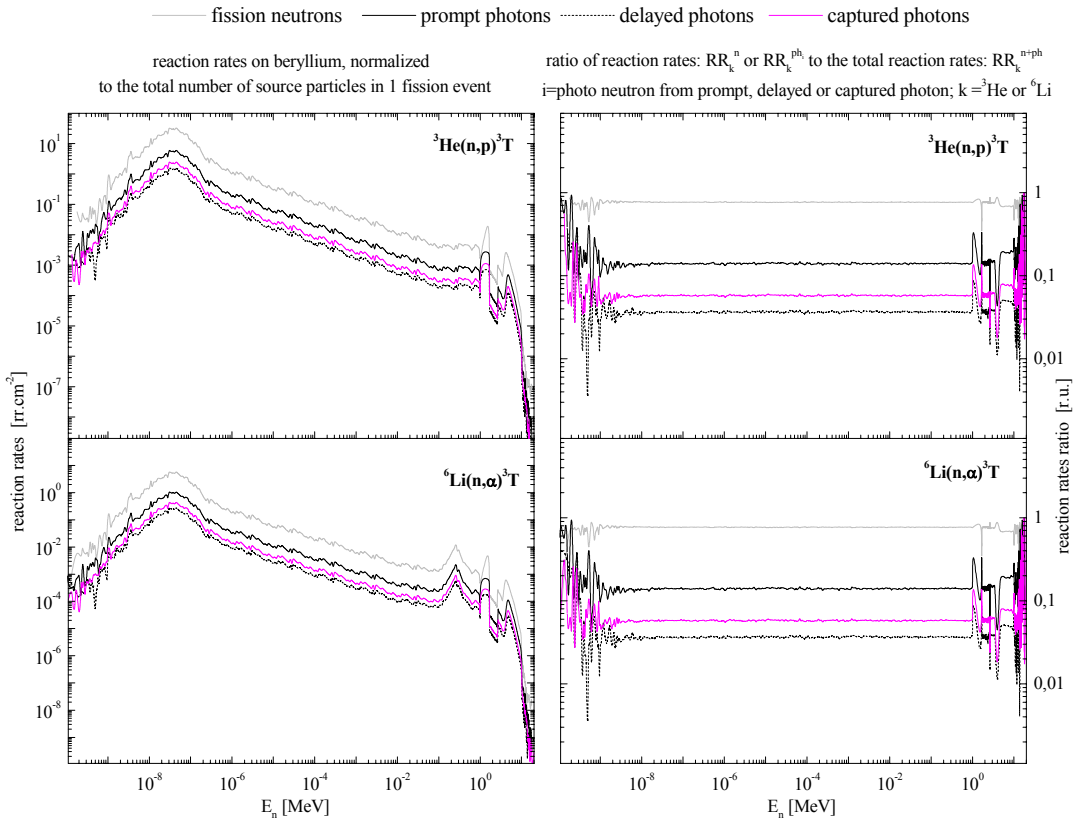
Table II. Intensity and gamma spectrum of delayed gamma-rays, emitted from fission products in the fuel elements. Calculations by MCNP&SCALE4.4a(ORIGENS) at reactor power: P_{BR2}=56 MW.

E_γ, MeV	Photons per second	MeV per second
0 ÷ 0.01	6.0228E+18	3.0114E+16
0.01 ÷ 0.05	5.2973E+18	1.5892E+17
0.05 ÷ 0.1	1.9443E+18	1.4583E+17
0.1 ÷ 0.2	2.3167E+18	3.4751E+17
0.2 ÷ 0.5	3.7409E+18	1.3093E+18
0.5 ÷ 1.0	5.2874E+18	3.9656E+18
1.0 ÷ 1.6	2.2696E+18	3.0640E+18
1.6 ÷ 2.3	5.6440E+17	1.1288E+18
2.3 ÷ 3.0	3.2284E+17	8.5553E+17
3.0 ÷ 5.0	1.4233E+17	5.6932E+17
5.0 ÷ 10.0	6.6645E+15	4.9984E+16
10.0 ÷ 20.0	2.0888E-10	3.1333E-09
Total	2.7915E+19	1.1625E+19

5.2 Contributors into the total reaction rates of neutron absorption in beryllium

The calculations of the reaction rates $^{9}Be(n,\alpha)^{6}Li$, $^{6}Li\ (n,\alpha)^{3}T$, $^{3}He\ (n,p)^{3}T$ were performed for a generic reactor model and for the full–scale 3 – D heterogeneous geometry model of BR2 (Fig. 3b) with detailed 3–D fuel burn up profile and detailed 3–D distribution of ^{6}Li, ^{3}T, ^{3}He concentrations in the beryllium matrix. The model was developed using the combined MCNP&SCALE4.4a(ORIGENS) method [11]. The reaction rates (1)÷(3) caused by fission neutrons are computed using any version of MCNP. The calculated spectra from prompt, delayed and captured photons are used in separated coupled photon – neutron problems by MCNP5 with included photonuclear reactions for evaluation of the reaction rates by all types of photons. The energy distributions of the reaction rates due to absorption of fission neutrons and photo neutrons, which are emitted after interactions of photons with beryllium, are given at Fig. 5.

Figure 5. MCNP5 calculations of the reaction rates of neutron absorption in beryllium, caused by fission neutrons and by photo neutrons, emitted in (γ,n) – reactions of prompt, delayed and captured photons with the beryllium reflector. Normalization: to the total number of neutrons or photons in 1 fission event.



If we normalize the reaction rates by photo neutrons $RR_k^{ph_i}$ for the nuclide k to the total number of photons of type "i", generated in fission as demonstrated at Fig. 2, then we obtain the following ratios:

$$\frac{\sum_i RR_k^{ph_i}}{RR_k^n + \sum_i RR_k^{ph_i}} \approx 0.2 \div 0.25\,; \qquad \frac{RR_k^n}{RR_k^n + \sum_i RR_k^{ph_i}} \approx 0.8 \div 0.75 \tag{8}$$

Where: RR_k^n – reaction rate due to absorption of fission neutrons by the nuclide k. The contributions into the reaction rates (1)÷(3) from fission neutrons and from photo neutrons, produced by prompt, delayed and captured photons are summarized in Table III. The substitution of $RR_k^n + \sum_i RR_k^{ph_i}$ in (5) instead of RR_k^n gives about 20%÷25% increase of the tritium production and 20%÷25% increase of the build-up of helium – 3 in the shut down. These results are valid for an arbitrary reactor with beryllium reflector without regard of the age of the irradiated beryllium.

Table III. Main contributors to the reaction rates of neutron absorption in beryllium (1)÷(3).

Reaction Type	fission neutrons	prompt photons	delayed photons	captured photons
$^9Be(n,\alpha)^6Li$	76.5%	14.1%	3.6%	5.8%
$^6Li(n,\alpha)^3T$	76.7%	13.9%	3.6%	5.8%
$^3He(n,p)^3T$	76.7%	13.9%	3.6%	5.8%

5.3 Reactivity contributors into the total worth of helium – 3

The calculated final reaction rates (1), (2) and (3), generated by fission neutrons and by photo neutrons, which are emitted after interactions of different types of photons with beryllium, are introduced into a system differential equations (5) for the evolution of the concentrations of ^6Li, ^3He and ^3T with time. The evaluated atomic concentrations of ^6Li, ^3He and ^3T are introduced into the input MCNP file for the final eigenvalue calculation of the multiplication factor k_{eff} and for estimation of the reactivity worth of helium – 3. The reactor BR2 operates about 120 full power days per year, which is equivalent to 5÷6 cycles, ~ 20 days each long with big shutdown between cycles. The reactivity effects due to the beryllium poisoning by accumulated during the shut down ^3He have been investigated for different operation cycles of the BR2 reactor, started after long shut down. The various contributors to the total reactivity worth of helium–3 are summarized in Table IV. The deviations of the estimated total reactivity effects by accumulated ^3He are within 10% from the measured reactivity values.

Table IV. MCNP calculations of the contributors to reactivity worth of helium–3 in dollars for different operation cycles of the BR2 reactor (1 BR2 dollar: $\beta_{eff}^{BR2} = \beta_{del,n+phot,n}^{BR2} = 0.0072$ [1]).

BR2 operation cycle	fission neutrons	prompt photons	delayed photons	captured photons
cycle 01/2004A.6	3.01 $	0.70 $	0.13 $	0.31 $
cycle 01/2005A.3	3.33 $	0.79 $	0.14 $	0.34 $
cycle 03/2005A.4	2.91 $	0.68 $	0.12 $	0.30 $

6. Conclusion

The reactivity worth of helium–3, which is accumulated in the beryllium reflector due to decay of tritium after the reactor shut down, has been discussed in a few papers, concerning reactivity loss in reactors with beryllium reflector. Publications about contributions from photonuclear interactions of photons with beryllium for additional production of tritium during the reactor operation and therefore enhanced build up of the strong neutron absorber ^3He after the shut down are not available at the present moment. This paper presents a detailed calculation analysis of four main contributors into the total

reaction rates of neutron absorption in beryllium and into the total reactivity worth of helium–3. The calculations are performed using the Monte Carlo code MCNP5 with included photonuclear reactions:

i. contributions from fission neutrons are about 70%÷75% from the total reactivity worth of helium – 3
ii. the contributions into the total worth of helium – 3 by photo neutrons, emitted in photonuclear interactions of different types of photons with beryllium are as follows:
    a. 13% to 15% from prompt photons
    b. 3% to 5% from delayed photons
    c. 5% to 7% from photons, escaped in (n,γ) – reactions on structural materials

The effective fraction of photo neutrons was estimated for the Belgian Material Testing Reactor BR2: $\beta_{phot,n}^{MCNP5} = 0.00067$ which is in a good agreement with the experimental value.

7. Future work

Additional investigations will be made regarding the impact of the main reactor materials like light water, aluminum, stainless steel and uranium enrichment of the fuel on the poisoning of beryllium by photo neutrons. Calculations for eventual contributions into the reactivity worth of helium – 3 from neutrons, emitted in (α,n) – reactions in beryllium will be performed using MCNPX.

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