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EVALUATION OF THE TRANSMUTATION OF TRANSURANIC USING NEUTRONS SPECTRUM FROM THE SPALLATION REACTION

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ABSTRACT

The transmutation of transuranic was analyzed by simulating the neutron flux from different spallation sources across arrays of fissile material with isotopic composition PWR reprocessing. A simplified model of Accelerator-Driven Systems (ADS) containing target, moderator graphite, lead-bismuth coolant or sodium coolant, is used. The simulation was made using the particles transport code MCNPX 2.6.0 which allowed to evaluate the rate of transmutation of actinides (Np, Pu, Am, and Cm) at different locations in the system. The objective of the study is to evaluate which the behavior and influences the spectrum of the spallation in the transmutation without the contribution or interference of multiplier, medium subcritical, which would add the contribution of fission neutrons generated, thus interfering in the analysis. The arrangement enable to infer the influence of hardened neutron flux from the spallation reaction in the transmutation, the results show that this is independent of the target material chosen, and the spectrum of spallation has a negligible importance compared to the influence of moderation and scattering generated by the coolant or moderator used.

1. INTRODUCTION

For the last 50 years, the nuclear systems have been producing increasing amounts of highly radioactive waste. The spent fuel consists of a wide variety of elements and isotopes. The majority of the long-lived isotopes come from only a few transuranic elements plutonium, neptunium, americium, curium and some fission products such as technetium and iodine. Fresh light water reactor (LWR) fuel consists of uranium oxide, UO_2. About 95–97% of the uranium composition is ^{238}U and 3–5% fissionable ^{235}U. As the fuel in a reactor is burnt out, its composition changes. Every year a LWR, 1 GWh (e), discharges about 21 tons radioactive

fuel with the following inventory: 20 tons of uranium containing 0.9% (180 kg) ^{235}U, 200 kg of plutonium; 21 kg of minor actinides: 10 kg of neptunium, 10 kg americium, 1 kg curium: 760 kg fission products: 18 kg of ^{99}Tc, 16 kg of ^{93}Zr, 9 kg of ^{135}Cs, 5 kg of ^{107}Pd, and 3 kg ^{129}I, which are long-lived elements [5].

The spent nuclear fuel is moved to a pool of water, where isotopes with short half-lives decay to safer levels and internal heat generation drops. The second step is to isolate the spent nuclear fuel from the biosphere by placing it in a geological repository. However, there are still the issues about the volume of waste and how to minimize release of radioactive material to the environment. Transmutation of Pu and minor actinides and long-lived fission products is a promising concept to reduce the radioactive waste and its long-term radiotoxicity. Partitioning and transmutation are considered as ways of reducing the burden on a geological disposal. Since plutonium and the minor actinides are mainly responsible for the long-term radiotoxicity, when these nuclides are removed from the waste (partitioning) and fissioned (transmutation),the remaining waste loses most of its long-term radiotoxicity [6].

Transmutation is the process of bombarding a material with particles to form new atoms with higher masses and/or to fission the material into atoms with smaller masses. It can reduce the mass, volume, activity, heat load, and/or raditoxicity of waste that must be sent to repository [7]. Many different technologies have been examined for transmutation, including a variety of reactors and ADSs. These systems are primarily distinguished by whether they have fast or thermal neutron spectrums. Transuranics transmutation can be planned in both thermal and fast reactors. However, a critical reactor with solid fuel and liquid coolant , thermal and fast, in which neutron production and neutron losses are in balance, may contain a quite limited amount of particular TRU mixture components like Np, Am or Cm isotopes, due to safety constraints. This is one of the mains reasons why the accelerator-driven systems are currently studied worldwide for nuclear waste burning. Accelerator-driven subcritical reactors have been proposed for many applications such as energy production, fertile-to-fissile transmutation and conversion of long-lived radioisotopes into stable or much shorter-lived isotopes [8–11]. The transmutation technology to incinerate the long lived radioactive isotopes using an accelerator-driven subcritical reactor is one of the best solutions. ADSs can be designed to have a fast neutron energy spectrum, and they are subcritical so safety issues can be considered less significant than in a fast reactor. Another advantage of ADSs is that they can burn mixtures of material that would not maintain criticality in a reactor [12].

This paper is a contribution to the study of transmutation of transuranics in ADS-fission hybrid systems using the code MCNPX 2.6.0 (Monte Carlo N-Particle transport eXtend) [1]. The goal is only to compare the influence of the spectrum of spallation without the contribution of the means subcritical (fission spectrum) in the transmutation. A hybrid system ADS-fission, consists of a source of neutrons generated by spallation reaction, about 0.2% to 10% of the total neutron population, plus the contribution of neutrons generated by fission of fuel in the subcritical means - *spallation* + *fission*. Unlike a critical system to which the simulation uses only a standard fission source, the simulation of an ADS should consider, in principle, a source that corresponds to these two spectra types, and the weighing is not as simple be made. By building a model to assess the influence of only spallation spectrum in the transmutation of TRU (transuranics), the simulations involving evaluate the behavior of the neutron flux emitted by the target (source of neutrons), focusing on the transuranic elements - Pu and minor actinides (Np, Am and Cm) considering graphite as moderator, or

lead-bismuth and sodium as coolant. The analysis allowed evaluating the influences of source alone, or submitted to a moderator and coolant.

2. EVALUATION OF TRANSMUTATION BASED ON RATE CAPTURE AND FISSION

The influence of the spectrum of neutrons generated by different spallation targets was analyzed, which focus on a layer of transmutation. The analysis is based on the simulation of the target spallation and evaluation of the results obtained from the records of reactions capture (n, γ) and fission (n, f) in the layers of transmutation. This are the two main reactions involved, as extensively shown in [1] [20] and [18].

The arrangements is simple, and maintain a wide margin of subcriticality in order not to compute the influence of neutrons generated by fission processes in a medium subcritical, analyzing, so only the influence of the particles generated in the target by the processes of intra-nuclear cascade the spallation reaction.

In the calculations, was used the Monte Carlo code MCNPX 2.6.0, which is a combination of LAHET and MCNP. This code transport a wide variety of particles when include charged particles such as protons and charged pions as well as neutrons with high energies. MCNPX offers options based on four physics packages; the BERTINI [13–16] and ISABEL [17] models taken from the LAHET Code System, the CEM package [14] and INCL. The MCNP transport code is used for radiation transport at low energies (<20 MeV). The transport of high energy particles (>150 MeV) is performed using nuclear models, while for the transport of low energy particles (<150 MeV) cross section libraries are used.

The configuration for the target, beam energy and transport particles, based on the study presented in [20]. The simulation consists of a parabolic proton beam 2.0 GeV, focusing on the surface of a cylindrical target. For the simulation is used Bertini, neutrons microscopic cross sections ENDF/B-VI, for temperature 293,6 °C, and data library proton LANL / T. As shown in [1] temperature has little significance in the results presented here. In spallation reaction was considered a wide range of transport particles, such as muons and pions that participate in the secondary cascade reactions [1] [18].

3. MODELING

The methodology was based on the model used by S.R. Nezhad-Hashemi et al. [2] [3], in which some modifications were made, which allowed the reduction of the error and the adequacy of the analysis to a more realistic evaluation that considers the isotopic composition of TRU reprocessed.

3.1. Reference model

In S. R. Hashemi-Nezhad et al. [2] [3] was analyzed the transmutation through of reaction rate of capture (n, γ) and fission (n, f), in some minor actinide isotopes, ^{237}Np, ^{241}Am, ^{246}Cm, ^{245}Cm, and ^{239}Pu, subjected to a neutrons spectrum generated by a spallation reaction. In the

spallation reaction we used a Pb target inside of the two different systems, one thermal system moderate graphite, and lead-cooled fast system without moderator. This model is considered the interaction of neutrons emitted by the target with the environment moderator / coolant and focusing on fuel. His analysis allows a series of comparative results between the two systems, and its effectiveness in transmuting these isotopes through these rate.

The system: (a) lead target and lead coolant - (Pb, Pb, 0); and (b) the lead target and graphite moderator - (Pb, C, 0). The first letter in brackets "Pb" specifies the target, the second letter "Pb" or "C" the moderator / coolant used, while "0" in the third position indicates that the system does not contain a specific nuclear fuel. The moderator / coolant occupy a bulk of 32 m^3 where the target is incorporated together with the fuel cells. In the case of (Pb, Pb, 0) approximately 370 tons of lead act as coolant and target, while in the system (Pb, C, 0) a target cylinder of 20 cm diameter and 1 meter long of the lead is inserted into the central plane xy. Arranged from the center of the system, 20 spheres of radius 1.5 cm and spaced 15 cm from each other are distributed at the center of the array. The spheres are filled with nuclear isotopes of interest and act as both - layer transmuting and measuring cells (Figure 1]). For the calculations, the number of particles (protons) incidents was nps = 25,000, using the Monte Carlo code MCNP-4B2.

The transmutation results are given in terms of the number of a given reaction in 1 g of the isotope of interest. Basically, to the records (number of events) reactions (n, γ) or (n, f) in the cells with transmutation material divided by the material mass (in grams), i.e. how many events capture or fission occurs for 1 gram of the specified material in the distance to the center of the target.

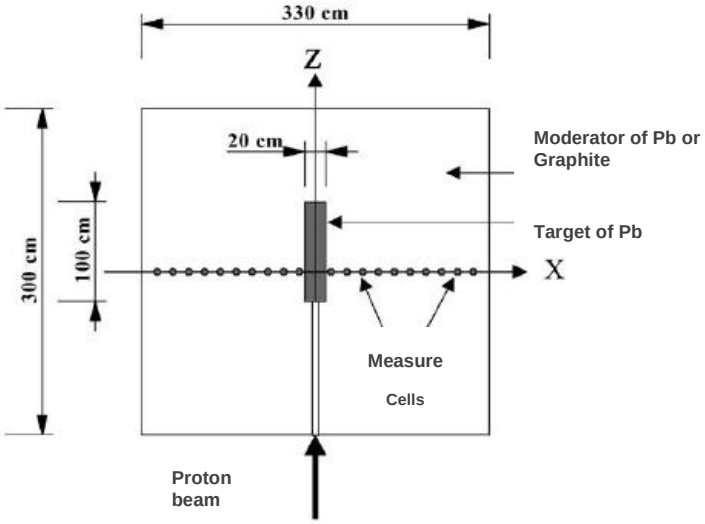


Figure 1: Section XZ arrangement moderator-target used in the simulations of the MCNP-4B2 by S.R. Hashemi-Nezhad et al. The origin of the Cartesian coordinate system corresponds to the center of the arrangement. (adapted from [1]).

3.2. Proposed Model

In this study we used a similar arrangement of reference model, though some aspects were considered as the optimization target and beam energy, defined by the results obtained in [1]. Besides improvements in the geometry of the system allowed a significant reduction of the

error[1]. Also, it opted for a more realistic applications of P&T (Partitioning and Transmutation), based on the evaluation of the isotopic composition full of TRU (Transuranic) - Np, Pu, Am and Cm (considering the use of reprocessing with recovering more than 99% for TRUs, and separately obtaining each element of TRU). The isotopic composition of output by mass of fuel burned in a power plant of 1000 MWe PWR after 5 years cooling is shown in Table 1, and it is used as a reference for defining the percentage of the isotopic composition of the transuranic used in the transmuting layer.

Table 1: Mass of each isotope of actinides in the fuel burned by a 1000 MWe PWR plant after 5 years cooling. (Table extracted from [4]).

Radionuclide	Half-Life	kg	Ci	Elemental Boiling Temperature, °C[b]
^{234}U	2.47×10^{5} yr	3.14	1.94×10^{1}	
^{235}U	7.1×10^{8} yr	2.15×10^{2}	4.61×10^{-1}	
^{236}U	2.39×10^{7} yr	1.14×10^{2}	7.22	
^{237}U	6.75 days	9.15×10^{-7}	7.47×10^{1}	
^{238}U	4.51×10^{9} yr	2.57×10^{4}	8.56	
Total		2.60×10^{4}	α 3.56×10^{1}	4,135
			β 7.47×10^{1}	
^{237}Np	2.14×10^{6} yr	2.04×10^{1}	1.44×10^{1}	
^{239}Np	2.35 days	2.05×10^{-6}	4.78×10^{2}	
Total		2.04×10^{1}	α 1.44×10^{1}	—
			β 4.78×10^{2}	
^{236}Pu	2.85 yr	2.51×10^{-4}	1.34×10^{2}	
^{238}Pu	86 yr	5.99	1.01×10^{5}	
^{239}Pu	24,400 yr	1.44×10^{2}	8.82×10^{3}	
^{240}Pu	6,580 yr	5.91×10^{1}	1.30×10^{4}	
^{241}Pu	13.2 yr	2.77×10^{1}	2.81×10^{6}	
^{242}Pu	3.79×10^{5} yr	9.65	3.76×10^{1}	
Total		2.46×10^{2}	α 1.23×10^{5}	3,508
			β 2.81×10^{6}	
^{241}Am	458 yr	1.32	4.53×10^{3}	
^{242m}Am	141 yr	1.19×10^{-2}	1.16×10^{2}	
^{243}Am	7,950 yr	2.48	4.77×10^{2}	
Total		3.81	α 5.01×10^{3}	2,880
			β 1.16×10^{2}	
^{242}Cm	163 days	1.33×10^{-1}	4.40×10^{5}	
^{243}Cm	32 yr	1.96×10^{-3}	9.03×10^{1}	
^{244}Cm	17.6 yr	9.11×10^{-1}	7.38×10^{4}	
^{245}Cm	9,300 yr	5.54×10^{-2}	9.79	
^{246}Cm	5,500 yr	6.23×10^{-3}	1.92	
Total		1.11	α 5.14×10^{5}	—
TOTAL		2.63×10^{4}	α 6.42×10^{5}	
			β 2.81×10^{6}	

The spheres (transmuting layer) are each filled with transuranium element (Np, Pu, Am, and Cm) according to the percentage of isotope given from Table 1. This analysis allows evaluating the influence of the spectrum of neutrons by the target in each transuranic element.

Preliminary simulations have shown that for obtaining a percentage error below 10% in all cells, it is necessary to transport than $nps > 10^{6}$ protons for simulation, requiring

[1] Initial analysis was performed using the same geometry and number of particles transported by S.R. Hashemi-Nezhad et al., However, the results showed an error very high reaching 40% at distances of 45 cm from the target. This showed the need to transport a high number of protons incident or improvements in the geometry of the model.

considerable computational. Due to this long execution time in some alternative geometry are created, as shown in Figure 2 and 4. These modifications are:

- Increasing the diameter of the balls of 1.5 cm to 3.0 cm.
- Increase the total number of spheres from 20 to 80, forming eight balls each with 10 beams, separated at angles of 45°. The beads were maintained in 15 cm spaced from each other and the center of the array, occupying the initial distance of 15 to 150 cm.
- Consideration of the sum of records (events, traces) of neutrons that focuses on all cells (rings) equidistant from the center of the array.

These changes represent an increase in the number of effective collisions, event logs, neutron cells filled with material transmutation, this due to the increase of the effective layer transmuting.

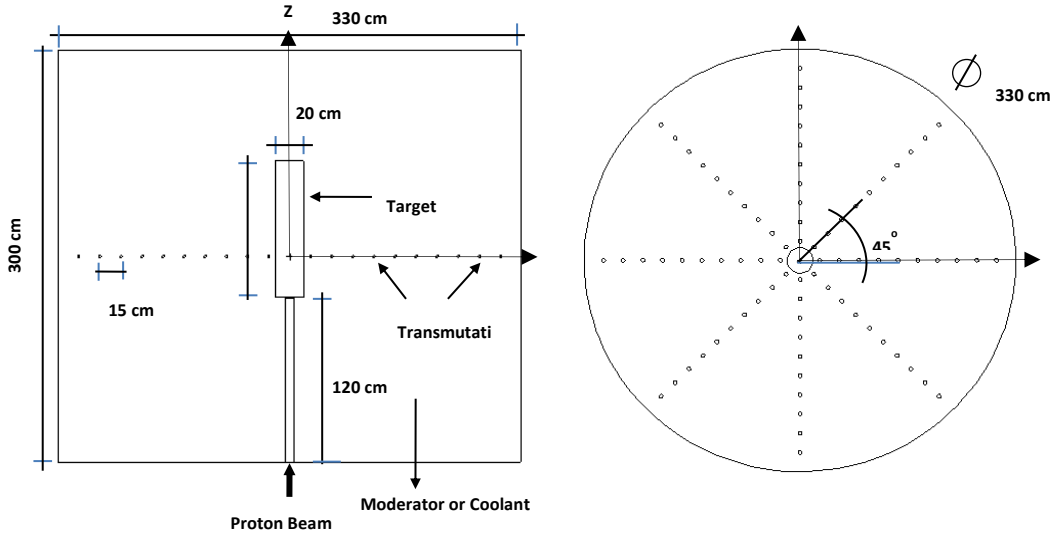


Figure 2: Radial and axial view of the proposed model, according to the Cartesian plane XZ and XY. The origin of the Cartesian coordinate system is located in the center of the array. (image obtained from the MCNPX Vised, version X 22S).

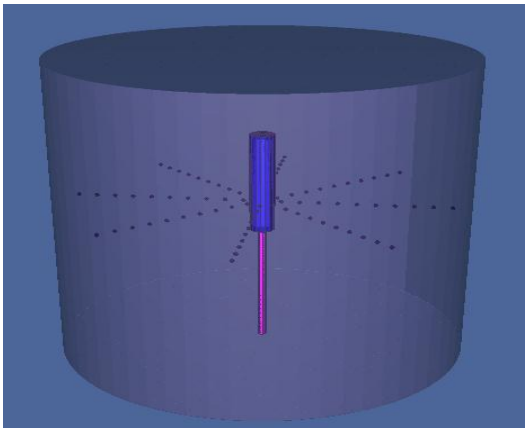


Figure 3: Arrangement view in three dimensions, the image showing the external vassel, the target (blue), transmutation cells (black) and tube of beam (pink). (image obtained from the MCNPX Vised, version X 22S).

The model consists of an outer vessel of 330 cm diameter and 300 cm in length, filled with moderator / coolant homogeneous, occupying a volume (~26 m^3) with the target and spheres embedded. The target cylinder 20 cm in diameter and 1 meter long is centered in the xy plane at position z = -30 cm and z = 70 cm, with the spheres filled with material transmutation radially arranged in the xy plane and z = 0.

A proton beam energy of 2.0 GeV is introduced into the system along the z-axis in a hollow tube of length 120 cm, internal diameter of 6 cm and thickness 1.5 cm. The tube material is composed of alloy steel H9. The proton beam falls directly on the window of the tube toward the target crossing axially. This beam has a parabolic distribution with 1.4 cm in diameter (value assigned according to the results found in [1]).

The analysis was also expanded considering three different target materials - natural U, Hg and eutectic mixture of PbBi. These targets were chosen because they showed the best results on the neutron emissivity, according to the results obtained in [1]. The neutron flux of the targets are subjected to three different conditions, a reference system (without moderator and coolant), a thermal system graphite-moderated and a fast system with Na or PbBi coolant. This expansion of the analysis enabled us to verify the actual contribution of the spectrum of neutrons emitted by each target in the transmutation.

4. ANALYSIS OF RESULTS

The results are shown in terms of the rates of reactions (n, γ) and (n, f) in the cells (spheres) filled with material transmutation, which can be obtained through a special treatment of MCNPX card event record, which allows to count the number of captures in a given nuclide or a specific combination of nuclides at the end of each story, converting the pulses created in a power distribution of a detector in neutron capture. The number of records fission events occurring in a given cell, is obtained indirectly, by converting the amount of average energy deposited in the cell by fission events. This conversion is made by considering the material density, bulk, and Q value (average energy generated by each fission ~200 MeV).

The parameter B is defined here as the relative number of interactions, while B' will represent the relative number of B interactions normalized to 1 g of material.

4.1. Reference system

The system without moderator is used as a reference model for comparison with the fast systems sodium or lead-bismuth and thermal graphite system. Figure 4 shows the rate of transmutation of TRU via reactions(n, f), and (n, γ) as a function of the parameter B and the distance x from the center of the system $(U, 0, 0)$, Figure 5 for $(Hg, 0, 0)$ and Figure 6 for $(PbBi, 0, 0)$.

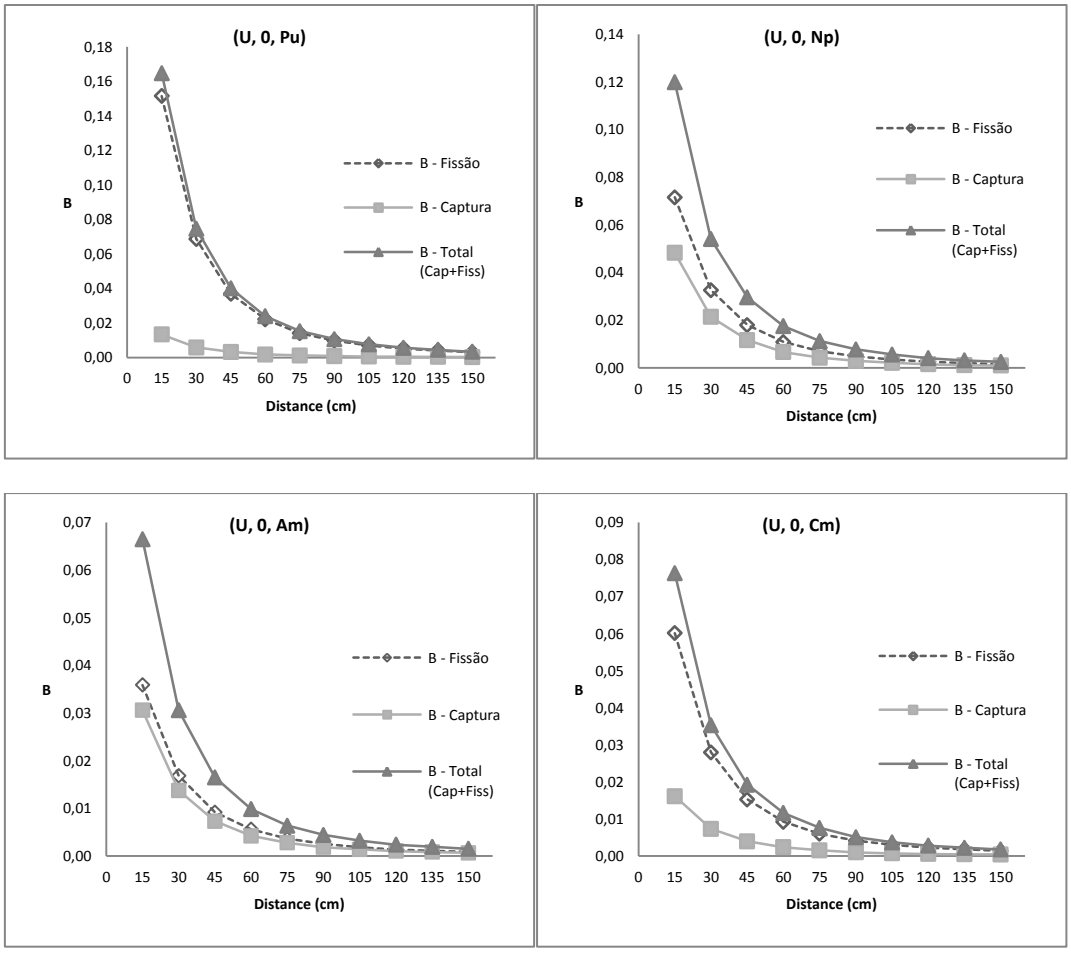


Figure 4: Number relative of reaction B in $(U, 0, 0)$ as a function of the distance to the TRU. The proton beam energy of 2.0 GeV and 400,000 particles with no moderator / coolant.

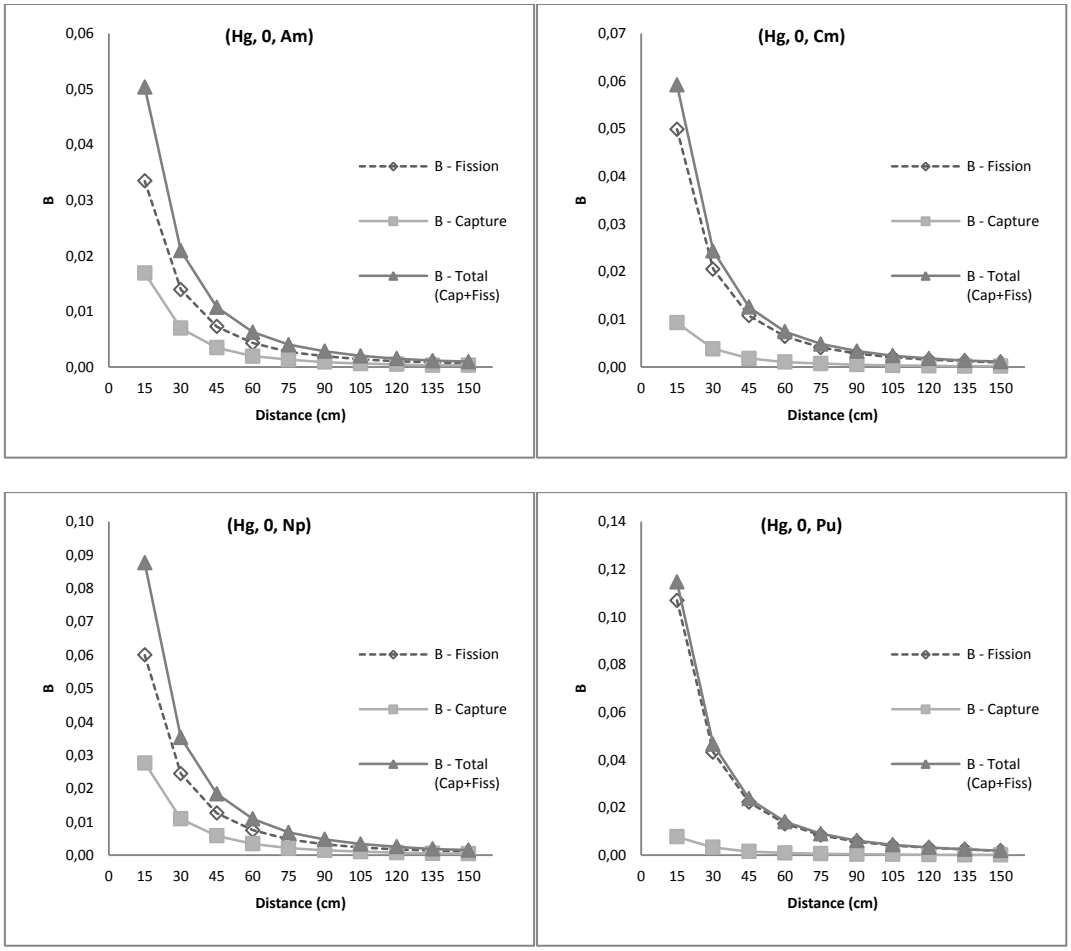
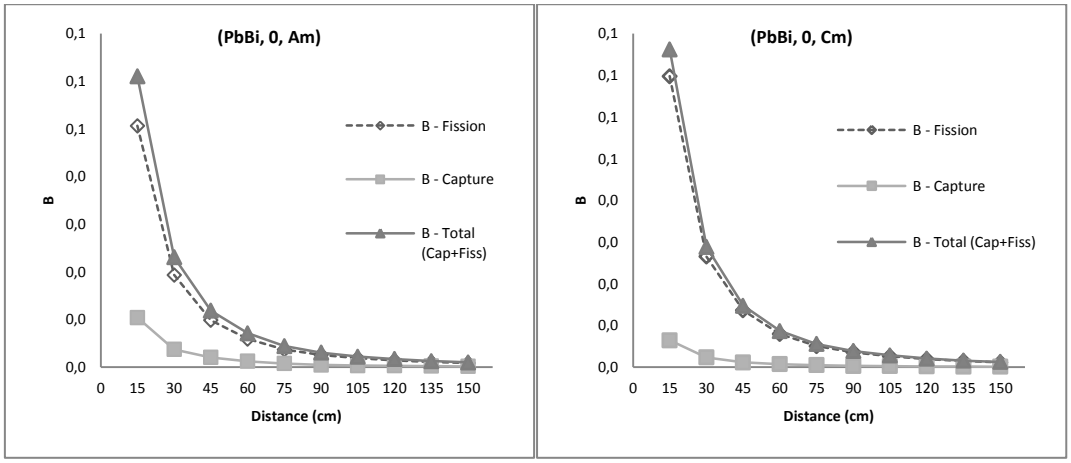


Figure 5: Number relative of reaction B in $(Hg, 0, 0)$ as a function of the distance to the TRU. The proton beam energy of 2.0 GeV and 400,000 particles with no moderator / coolant.



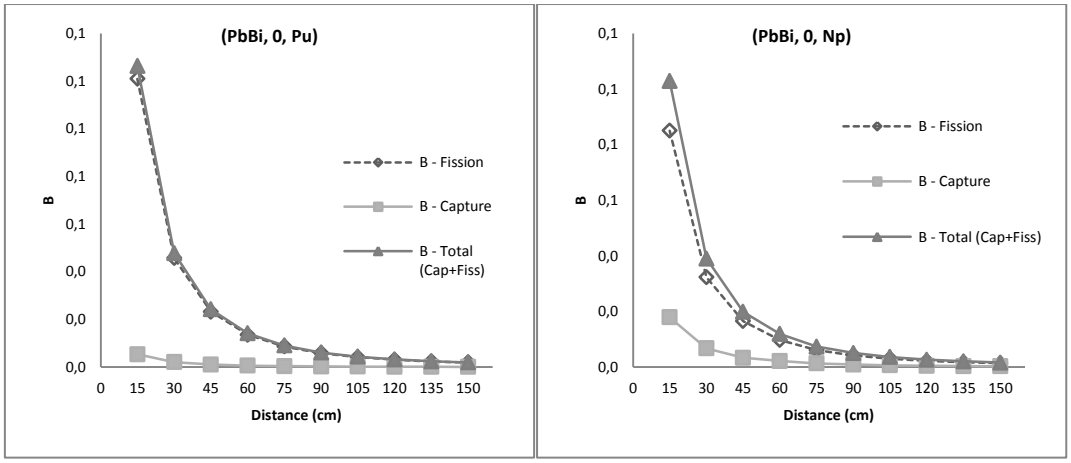


Figure 6: Number relative of reaction B in $(PbBi, 0, 0)$ as a function of the distance to the TRU. The proton beam energy of 2.0 GeV and 400,000 particles with no moderator / coolant.

Analysis of the graphs shows that:

- For transuranic subjected to pure flux emission target, without any moderation, reaction rates for fission (n, f) are higher than the rates of reaction by capture (n, γ) to all elements, and therefore the mechanism more effective for its transmutation.

- The spectrum hardened[2] in a spallation target system without moderator favours therefore the fission reactions in all TRU.

These behaviours can best be represented by the average value of the ratio of the capture and fission reactions $R = \langle B_{(n,f)}/B_{(n,\gamma)} \rangle$ for TRU, as shown in Table 2. The ratio R is independent of the number of transport particles (i.e. the number of event) and the amount of material mass.

The results in Table quantify the effect of the slight difference between the observed emission spectra of targets. As an example for the system $(PbBi\ 0, Pu)$ that each 22.43 fission events plutonium reprocessed at first capture event occurs, and under this condition, the fission reactions occur 22 times more than the capture reactions. The transmutation of plutonium in these conditions follows a direct path via fission reactions. This is in accordance with the cross section of Pu isotopes.

[2] The term "hardened" refers to the fact that the neutrons emitted from the spallation target are mostly in the range of 0.1 MeV to 20 MeV (see [1]).

Table 2: Average value of the ratio of the capture and fission reactions $\boldsymbol{R = \langle B_{(n,f)}/B_{(n,\gamma)} \rangle}$ for TRU fuels analysed in reference systems.

System	Material rate - $\boldsymbol{R = \langle B_{(n,f)}/B_{(n,\gamma)} \rangle}$			
	R_{Am}	R_{Cm}	R_{Np}	R_{Pu}
(Hg, 0, 0)	2,13	5,82	2,25	14,3
(PbBi, 0, 0)	4,99	11,06	4,77	22,43
(U, 0, 0)	1,3	3,88	1,64	11,9

The results show that Pu followed by Cm are the materials that have the highest ratio fission / capture. The data only quantify the expected result, since all major isotopes these actinides analyzed have fission cross sections above capture for range of spectrum above 1 MeV. These information is important when analysing the same conditions, but with the presence half a moderator or coolant.

The ratio R also shows that the spectrum of neutrons emitted by the target of PbBi is more hardened than other targets, is due to the low PbBi scattering cross section (for more information, reference the [1] and [20]). The Figure 7 show the neutron emission spectrum of different targets. These results are consistent with the literature [18], [21] and [22]; and correspond to the typical energies expected for the process of evaporation, characterized by a nuclear temperature T = 2. . . 8 MeV, so that for the spectrum of emitted neutrons follow Maxwellian distributions [18] [20]. The Figures show two peaks, one close to 2 MeV (energy close to the neutron emission per fission reaction) and other lower due to the process of capture / scattering of the target material. The PbBi, has only one peak centered close to 2 MeV. In the Figure 8 show the emission neutron spectrum of the standard fission source and standard evaporation source of MCNPX 2.6.0, compared to the spallation spectrum of U target simulated within Bertini model. Due to the material properties, the expected theoretical distribution of energy of the neutrons emission spectrum in the temperatures involved, it is slightly modified with peaks resonant emission, although within the energy range expected. Other reactions such as the type (x, xn) were not considered, due to the low emissivity of the target at energies above 4 MeV (Figure 7 and Figure 8). As shown below, the moderator and coolant have much importance in the change of the neutron spectrum energy in system population, than target neutrons emission. At distances greater than 30 cm, it is not observed the effect of target in transmutation rates. High energy neutrons that escape from the target, quickly begin to transfer energy to the means.

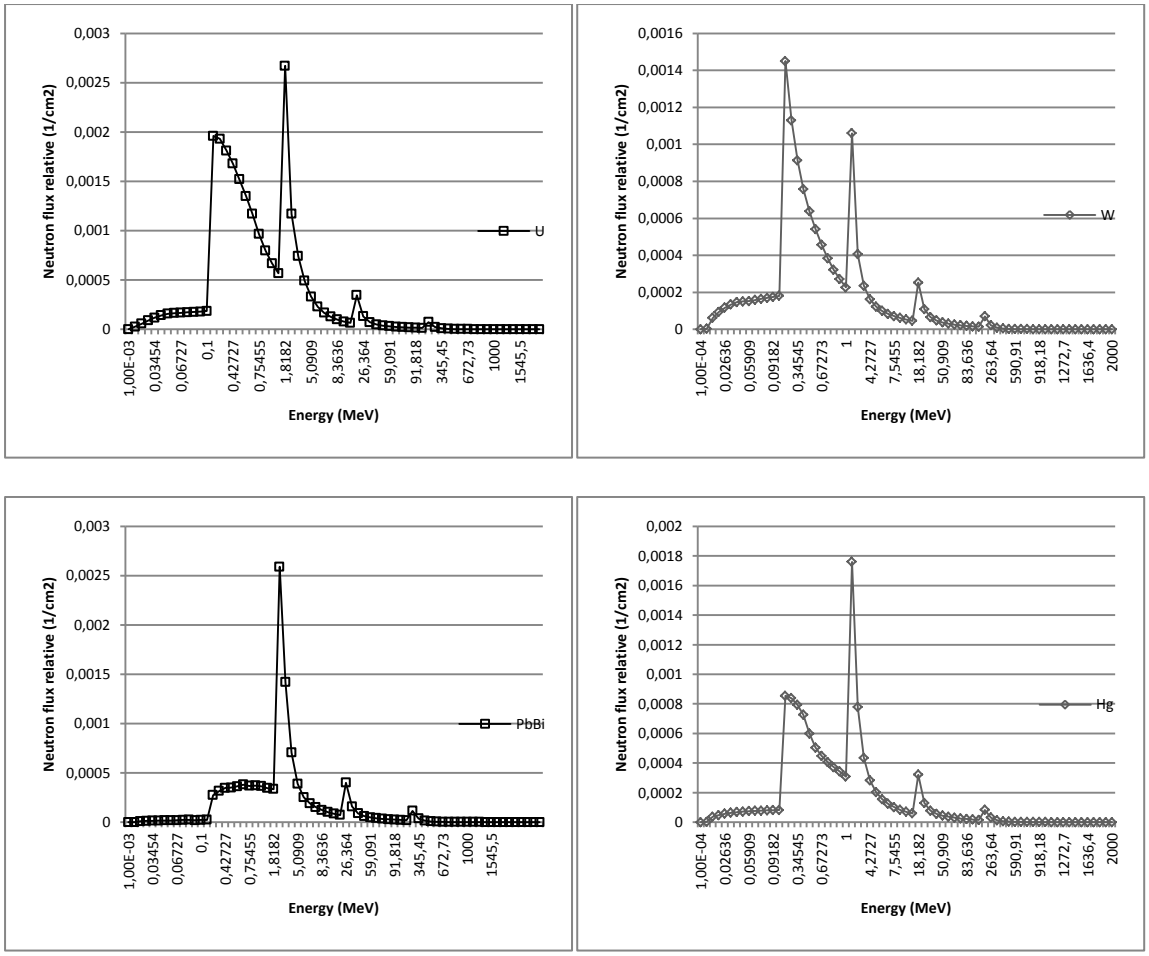


Figure 7: Neutrons emission spectrum of through spallation reaction in different targets. 2.0 GeV proton beam, Bertini model.

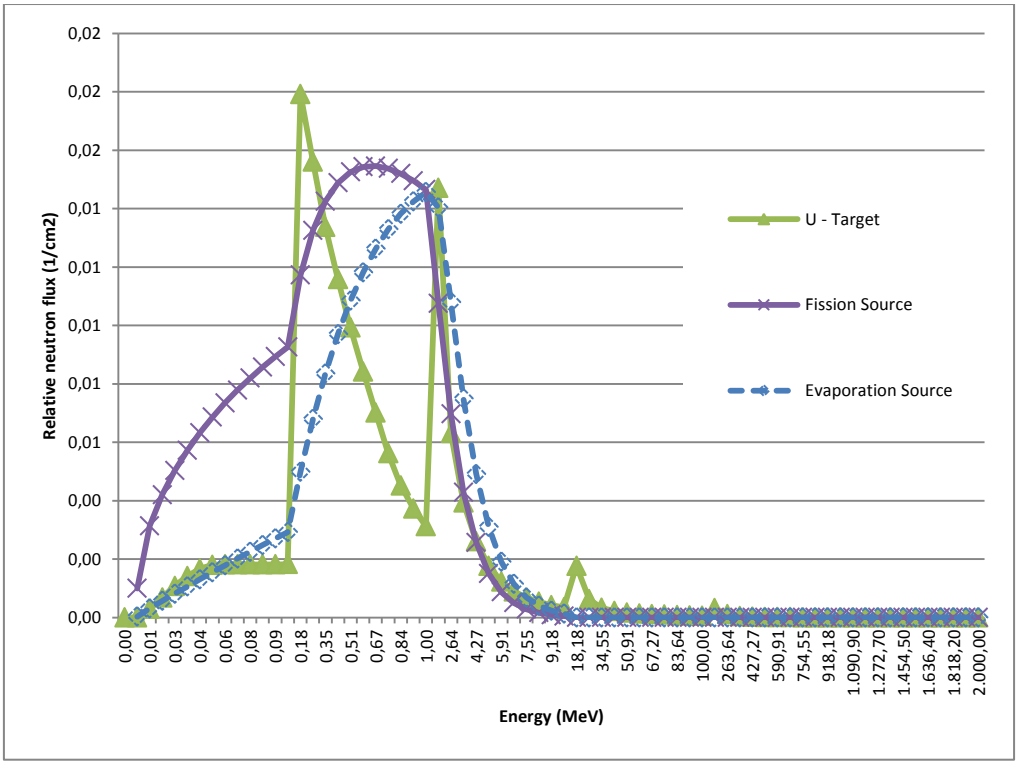


Figure 8: Neutrons emission spectrum of through spallation reaction in U Target (30 cm diameter, 2.0 GeV proton beam, Bertini Model) compared to standard Fission Source and standard Evaporation Source of the MCNPX 2.6.0.

4.2. SodiumColled Fast System

The sodium-cooled system and eutectic lead-bismuth cooled system is shown in Figure 9 to 11, while Table 3 shows the average value of the material transmutation ratio between the capture and fission reactions $R = \langle B_{(n,f)}/B_{(n,\gamma)} \rangle$ for all systems.

Tabela 3: Average value of the ratio of the capture and fission reactions $\boldsymbol{R = \langle B_{(n,f)}/B_{(n,\gamma)} \rangle}$ for TRU fuels analyzed in Na and PbBi systems.

System	Material rate - $\boldsymbol{R = \langle B_{(n,f)}/B_{(n,\gamma)} \rangle}$			
	R_{Am}	R_{Cm}	R_{Np}	R_{Pu}
$(Hg, Na, 0)$	0,034	0,96	0,024	2,06
$(PbBi, PbBi, 0)$	0,03	1,026	0,022	2,091
$(U, Na, 0)$	0,029	0,92	0,016	2,029

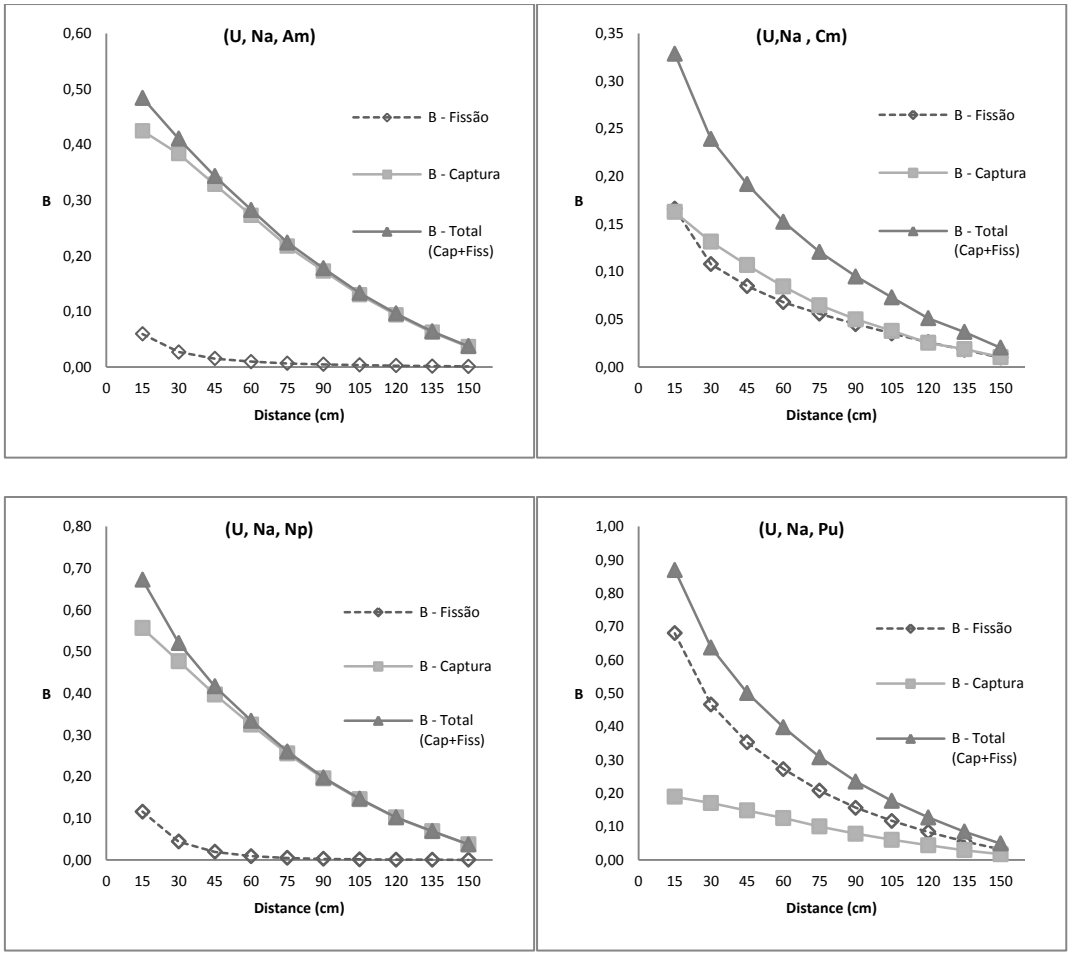
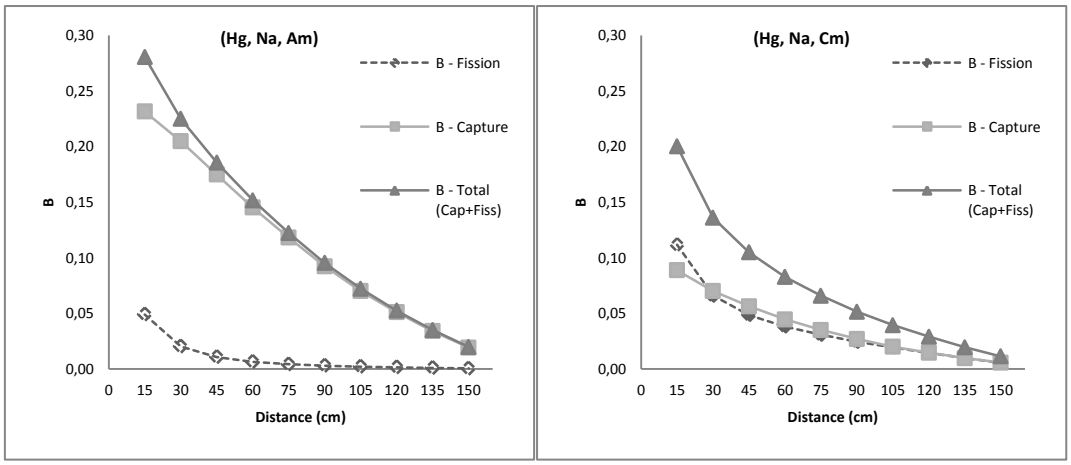


Figura 9: Number relative of reaction B in $(U, Na, 0)$ as a function of the distance to the TRU. The proton beam energy of 2.0 GeV and 400,000 particles.



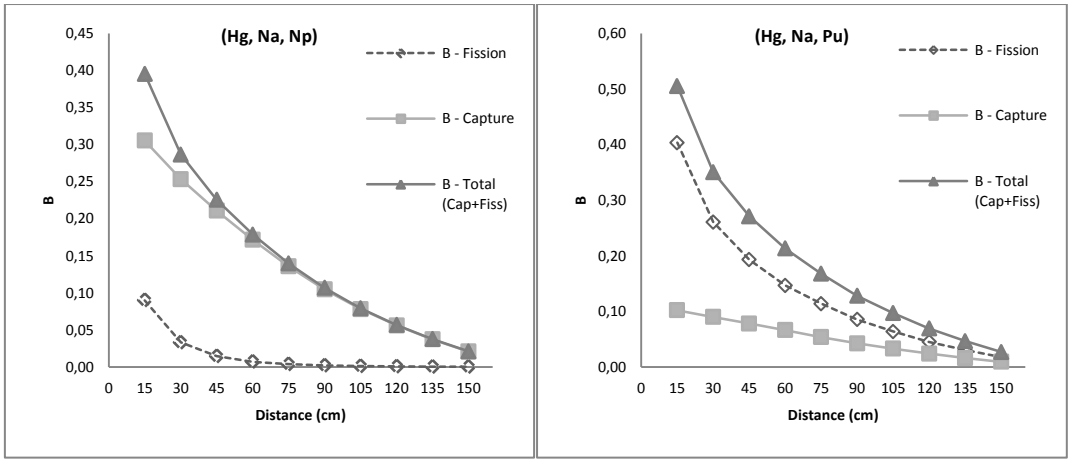


Figura 10: Number relative of reaction B in $(Hg, Na, 0)$ as a function of the distance to the TRU. The proton beam energy of 2.0 GeV and 400,000 particles.

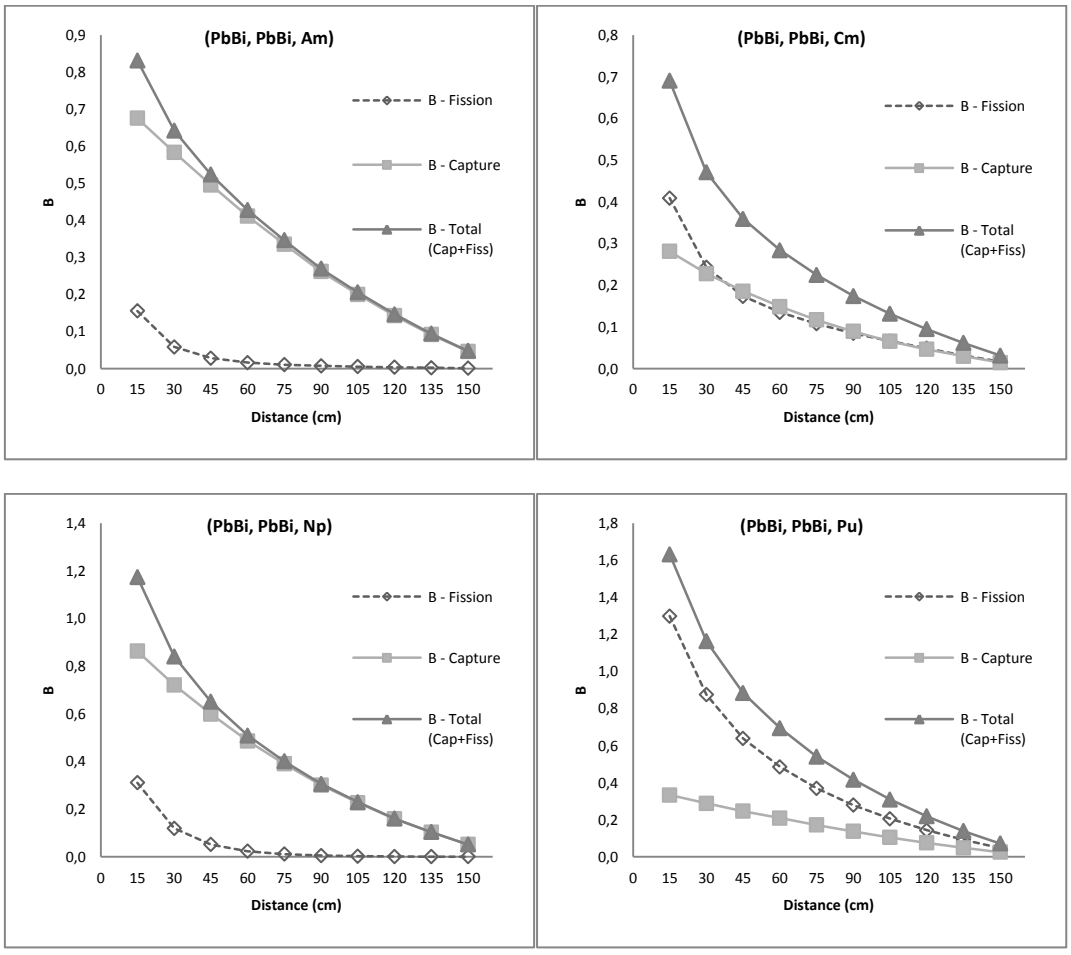


Figura 11: Number relative of reaction B in $(PbBi, PbBi, 0)$ as a function of the distance to the TRU. The proton beam energy of 2.0 GeV and 400,000 particles.

Table 4: Some properties of the moderator / coolant used, being Σ_s and Σ_a the macroscopic cross sections of scattering and absorption of neutrons, respectively.

Moderator	Average log. energy decrement ξ	Power moderation $\xi\Sigma_s$	Reason Moderation $\xi\Sigma_s/\Sigma_a$	Mean free path $1/\Sigma_t$	N_1^a	N_2^b
C	0,158	0,0608	190	2,60	58	115
Pb	0,0096	0,0035	0,58	2,71	959	1894
Bi	0,0095	0,0024	2,4	3,91	969	1914
PbBi	0,0095	0,0029	1,58	3,37	965	1905
Na	0,0845	0,0086	0,66	8,7	109	215
H_2O	0,948	3,27	149	0,29	9,7	19
He	0,425	8,93E-06	45	47619	22	43

[a] Number of collisions of neutrons necessary to cross the energy range 10 KeV - 1 eV.
[b] Number of collisions of neutrons necessary to cross the energy range 2 MeV - 0.025 eV.

The analysis of the graphs and the values for R in Table 3 show that:

- Compared to the reference system, the systems to Na and PbBi present inversion behaviour, with increased capture reactions (n,γ) in lieu of the fission reactions (n,f). Being reactions capture superior to fission in Am, Cm and Np. Only Pu has the main mechanism of transmutation fission.

- The behaviour of the three systems is very similar and suggested that this is weakly dependent on the selected target, being the transmutation direct function of moderator and coolant. For target distances larger than 30 cm, it is not possible to observe any distinction between the R ratio in the three systems, indicating that despite the low power of moderation, Na and PbBi influence the neutron spectrum, i.e. the transmutation reactions (n,γ) and (n,f) much more significant than the target used.

- The eutectic Lead-Bismuth has a behaviour very similar to sodium and both are equivalent. Also show that despite the low power of moderation, sodium and lead-bismuth, when compared to the reference system (unmoderated) significantly influence the spectrum of neutrons and the reactions involved in the transmutation of TRU. This behaviour indicates no difference neutronics representative between the metals. This is due the proximity of the values of Power Moderation $\xi\Sigma_s$ Reason Moderation $\xi\Sigma_s/\Sigma_a$ of Na and PbBi, as shown in Table 4.

4.3. Graphite-Moderated System

The graphite-moderated system $(U, C, 0)$ is shown in Figure 12 to 14, while Table 5 shows the average value of the ratio between the capture and fission reactions $R = \langle B_{(n,f)}/B_{(n,\gamma)} \rangle$ for the graphite-moderated systems.

Table 5: Average value of the ratio of the capture and fission reactions $\boldsymbol{R = \langle B_{(n,f)}/B_{(n,\gamma)} \rangle}$ for TRU fuels analyzed in C systems.

System	Material rate - $\boldsymbol{R = \langle B_{(n,f)}/B_{(n,\gamma)} \rangle}$			
	R_{Am}	R_{Cm}	R_{Np}	R_{Pu}
$(Hg, C, 0)$	0,114	3,544	0,001	2,06
$(PbBi, C, 0)$	0,114	3,554	0,001	2,085
$(U, C, 0)$	0,114	3,544	0,0005	2,087

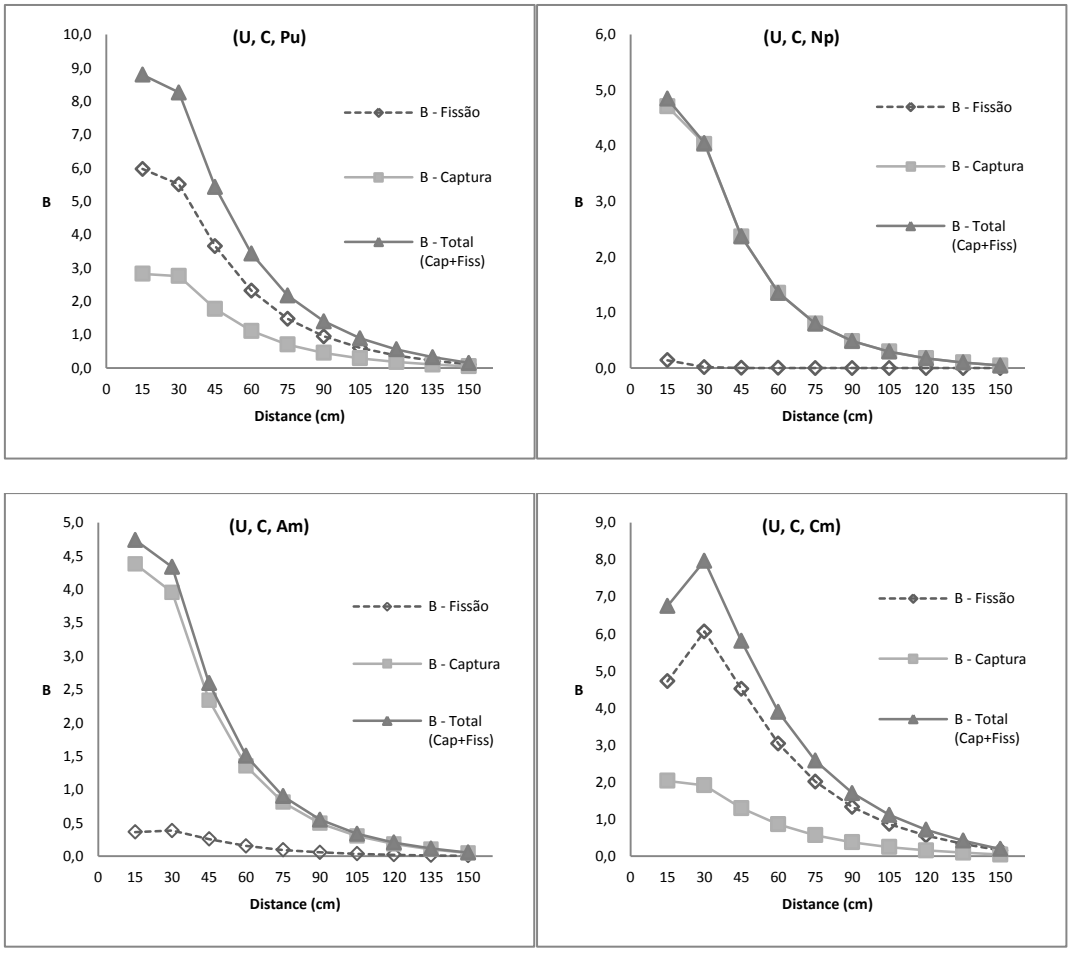


Figure 12: Number relative of reaction $\boldsymbol{B}$ in $(\boldsymbol{U, C, 0})$ as a function of the distance to the TRU. The proton beam energy of 2.0 GeV and 400,000 particles.

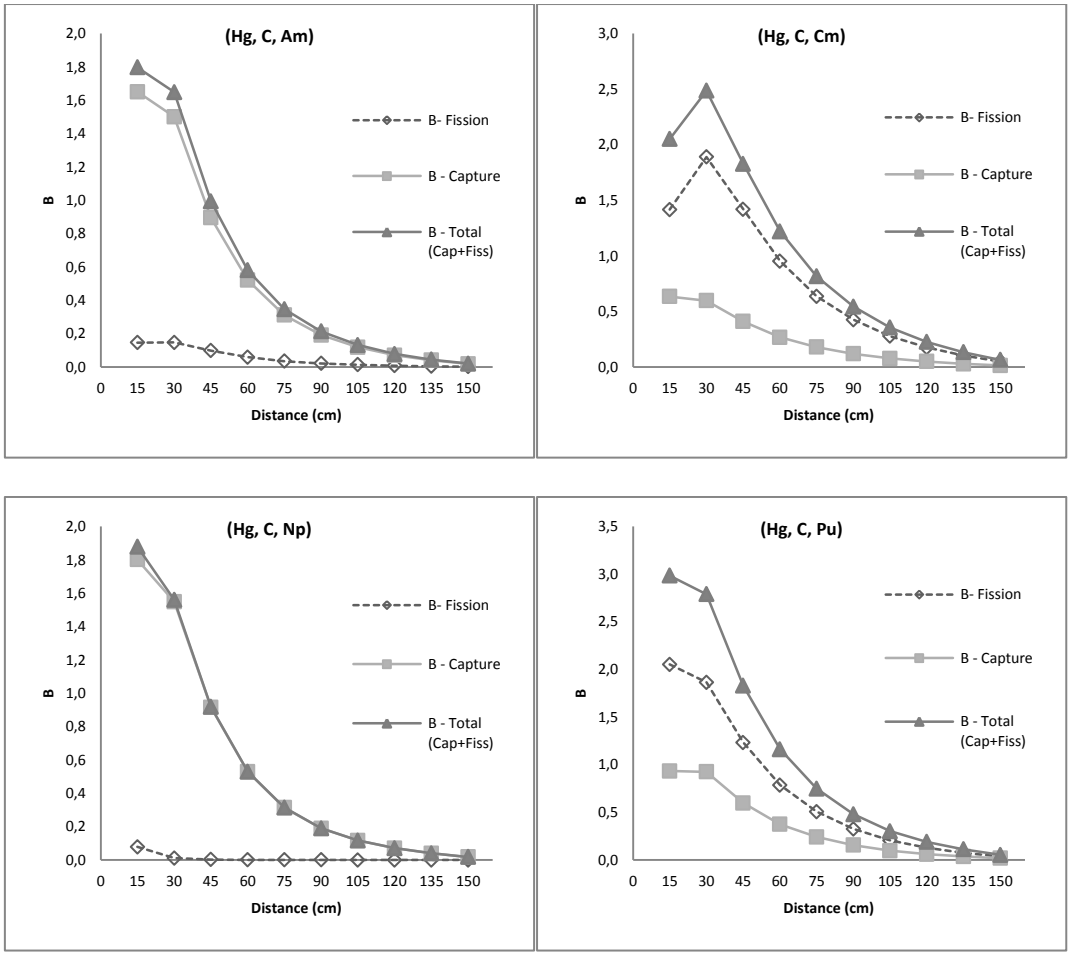
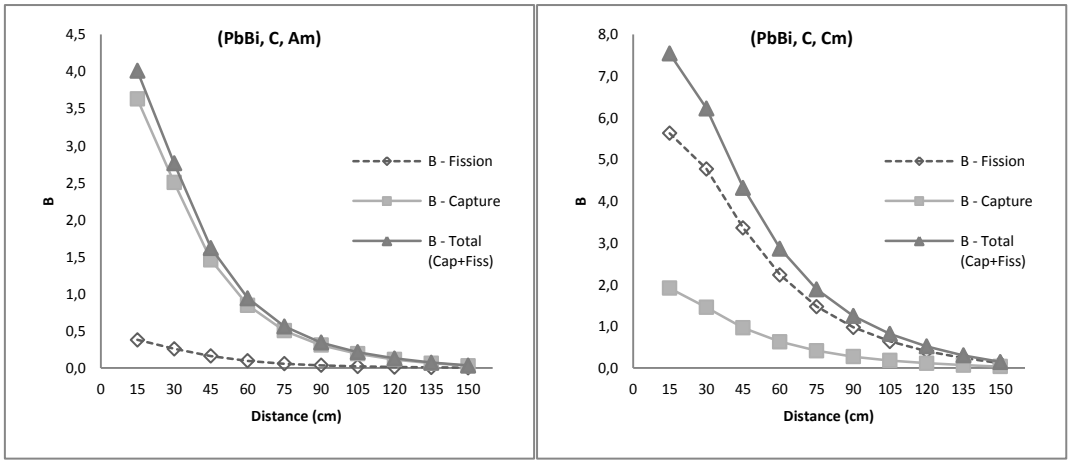


Figure 13: Number relative of reaction B in $(Hg, C, 0)$ as a function of the distance to the TRU. The proton beam energy of 2.0 GeV and 400,000 particles.



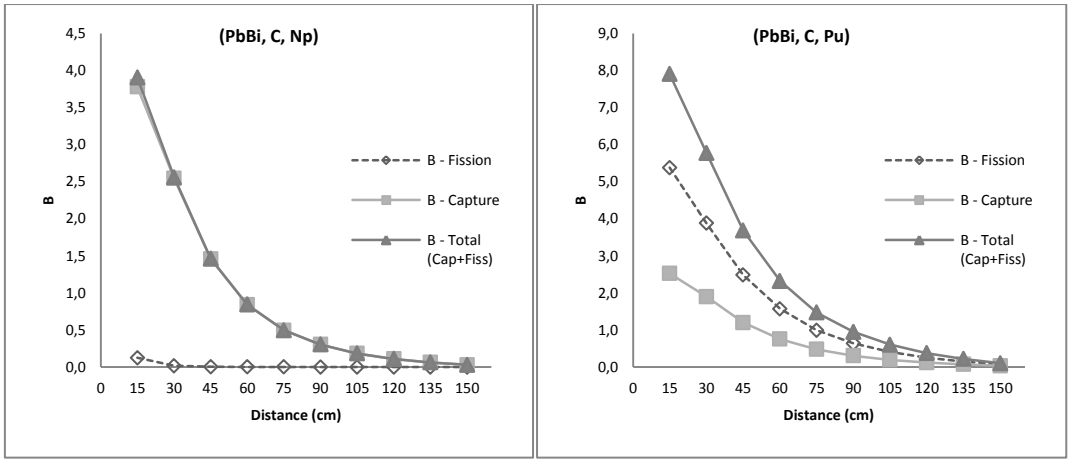


Figure 14: Number relative of reaction B in $(PbBi, C, 0)$ as a function of the distance to the TRU. The proton beam energy of 2.0 GeV and 400,000 particles.

The results show that:

- The ratio R of all targets analyzed are even closer than the coolant Na and PbBi. These data are consistent with expectations due to strong moderation exercised by graphite, and show that the transmutation of TRU in an ADS system is independent of the chosen target. Is evidenced in charts that 30 cm from the target, there is only the moderator influence on the transmutation.

- For Am and Np, the three systems, the transmutation occurs mainly via reaction (n, γ),in contrast to Cm exhibits a different behaviour with the main mechanism of transmutation the reactions (n, f) with a R ratio higher than Pu, inversely as occurred for Na and PbBi. The Pu behaves in all systems with fission reactions (n, f) the main mechanism of transmutation.

- The results show that the rate of transmutation are independent of target material used and the emission spectrum of spallation reaction. Transmutation or reactions in TRU depend directly of moderator or coolant used in the system. This results are valid for any target and any energy proton beam incident [1], [20]. Under the conditions studied, the coolant PbBi and Na, have a very similar behavior, and expected evolution of burning near in these systems.

A comparison between the ratio R for the target U in the fast and thermal systems is shown in Figure 15 for all TRU, while Figure 16 shows the values of reaction rates total B_T ($B_T = B_f + B_\gamma$).

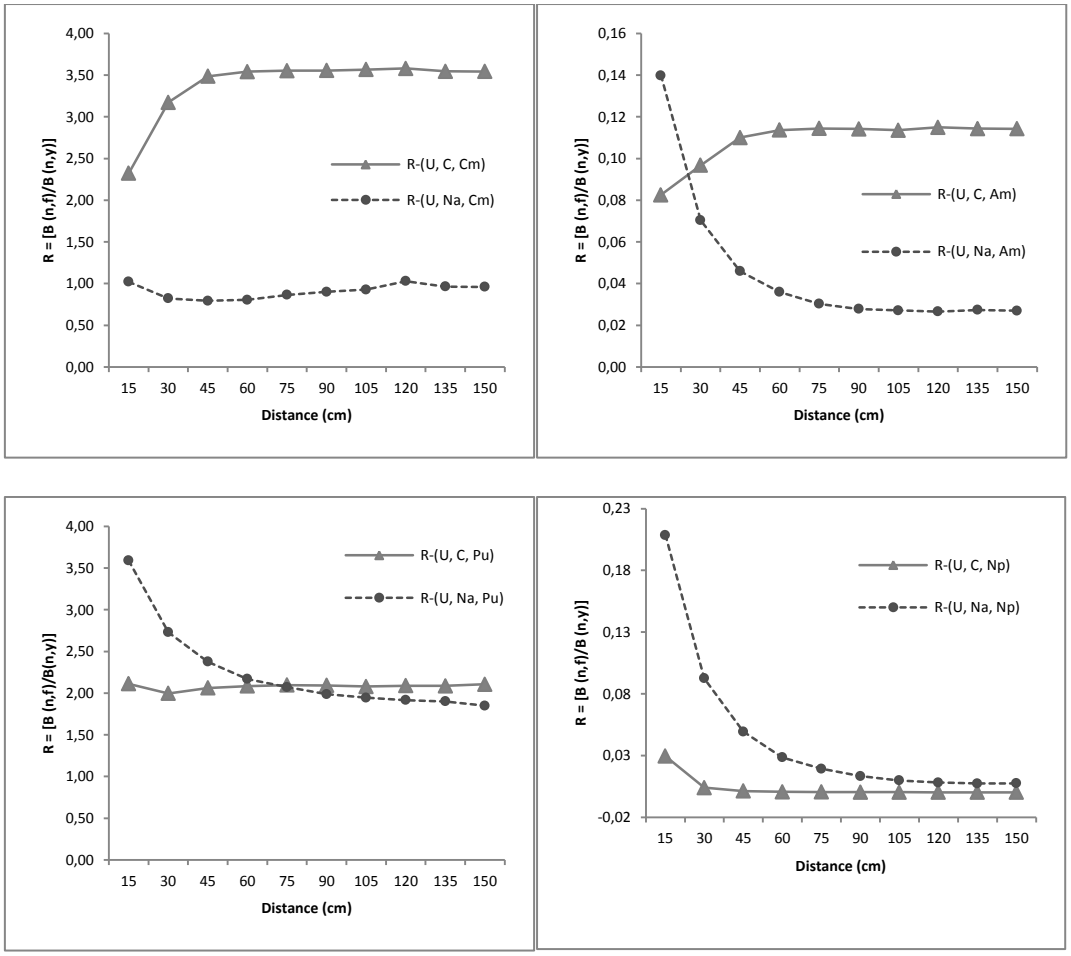


Figure 15: The ratio of the rates of fission and to capture systems $(U, C, 0)$ and $(U, Na, 0)$, to different locations of the axis x.

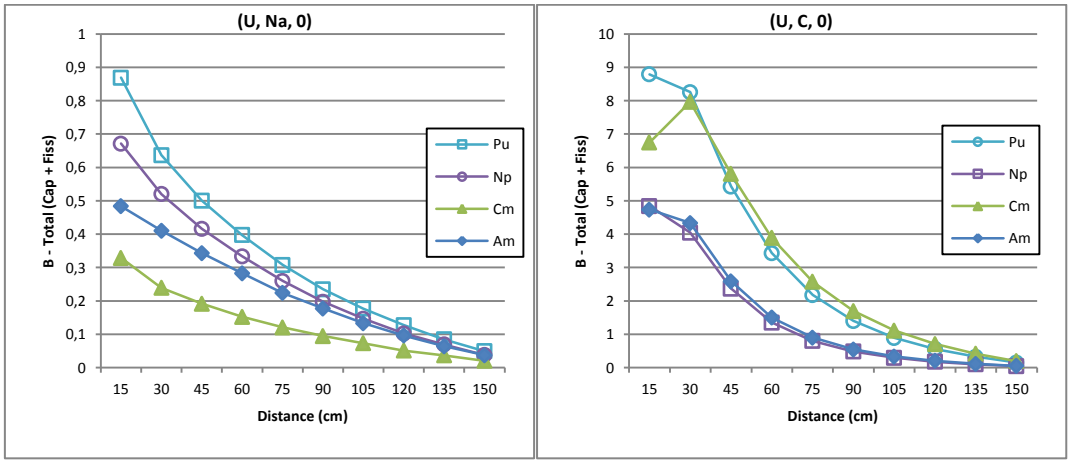


Figure 16: **Value B_T TRU for the Na and C systems, depending on the distance x.**

The analysis results show that:

- **Pu** - shows a rate of reactions (n, f) reactions up to (n, γ) for all systems. The high values for reaction rates in Pu are due to the significant amount of fissile isotopes present ≈ 69% of the composition of Pu reprocessed, mainly due to the isotopes ^{239}Pu and ^{241}Pu (Table 1).

- **Np** - have primary mechanism transmutation reactions (n, γ). The main isotope ^{237}Np has a cross section of the thermal spectrum fission very low ~1 b, against a high capture cross section of 10,000 b. Such as quantified in Figure 4 to 6 for all systems, their reaction rates are small in all conditions and present a difficulty transmutation. In fast environment with the sodium coolant, fission rates are far below.

- **Am** - is the transuranic which presents the lowest values for the overall reaction rate B_T. Their main mechanism is via transmutation reactions (n, γ), which is due to high capture cross-section in the thermal spectrum of the main the isotope ^{241}Am (~ 34%) and ^{243}Am (~ 65%), which represents 50,000 b and 5000 b, respectively.

- **Cm** - is more favourable transmutation in a thermal system, its value B_T is less than the Pu and Np to a system without moderator, less than all elements in a fast sodium-cooled, and superior to Pu in a graphite-moderated thermal system. The Cm has three isotopes most representative mass in the spent fuel, ^{242}Cm (~ 12%), ^{244}Cm (~ 82%) and ^{245}Cm (~ 5%), the ^{242}Cm and ^{245}Cm fissile with fission cross section and capture cross section high for the thermal spectrum, while the ^{244}Cm (with the largest mass percent) have fission cross section too low 80 b for reaction (n, f) and 1,000 b for reaction (n, γ).

5. CONCLUSION

This study evaluated how the spectrum of neutrons emitted by different spallation targets - Hg, U and PbBI, which interact in a layer composed of transuranic elements, with the purpose of transmutation, from the analysis of the rates of reactions of fission and capture involved. The results allowed us to quantify and consider what are the mechanisms (reactions) involved when subjected to neutron fluxes of spallation target, with or without the presence of a medium that exerts some moderation to neutrons.

In the presence of a medium with low or high power moderation, no significant difference in the behaviour of transmutation of TRU, using different targets. Transmutation is independent of the spectrum hardened neutron flux of the spallation reaction, even at different targets, being a direct function of the medium, moderator / coolant used. This suggests that in the simulations of burning the fuel, the insertion a spectrum of spallation, in addition to fission spectrum have low importance.

Reactions capture in Am and Np, in environments with moderator / coolant, is the main mechanism of transmutation of these elements, and they presented, on average, lower overall reaction rate, making it difficult to reduce their inventory. In turn, the transmutation Cm presents a more favourable graphite-moderated system, and the main mechanism fission

transmutation in this environment. Already Pu is easily converted into all systems having high reaction rates, and being fission reactions over reactions capture. However, mechanisms transmutation via capture reaction may not be the best solution for reducing the isotopic inventory. The conversion of these elements isotopes with lower fission cross section may it difficult to implement of closed fuel cycles based on thermal systems.

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