Hindawi Publishing Corporation
Science and Technology of Nuclear Installations
Volume 2015, Article ID 104739, 23 pages
http://dx.doi.org/10.1155/2015/104739

Research Article

Transuranics Transmutation Using Neutrons Spectrum from Spallation Reactions

Maurício Gilberti,[1,2] Claubia Pereira,[2,3] Maria Auxiliadora F. Veloso,[2,3] and Antonella Lombardi Costa[2,3]

[1]*Eletrobrás Termonuclear S.A.-ELETRONUCLEAR, Departamento GEI.T, BR 101 Sul-Rodovia Governador Mário Covas Km 517, 23948–000 Angra dos Reis, RJ, Brazil*
[2]*Programa de Pós-graduação em Ciências e Técnicas Nucleares (PCTN), Departamento de Engenharia Nuclear, Escola de Engenharia, Universidade Federal de Minas Gerais, Avenida Antônio Carlos 6627, 31270-901 Belo Horizonte, MG, Brazil*
[3]*Instituto Nacional de Ciência e Tecnologia de Reatores Nucleares Inovadores/CNPq, Brazil*

Correspondence should be addressed to Claubia Pereira; claubia@nuclear.ufmg.br

Received 14 October 2014; Revised 20 March 2015; Accepted 20 March 2015

Academic Editor: Alejandro Clausse

The aim is to analyse the neutron spectrum influence in a hybrid system ADS-fission inducing transuranics (TRUs) transmutation. A simple model consisting of an Accelerator-Driven Subcritical (ADS) system containing spallation target, moderator or coolant, and spheres of actinides, "fuel," at different locations in the system was modelled. The simulation was performed using the MCNPX 2.6.0 particles transport code evaluating capture (n, γ) and fission (n, f) reactions, as well as the burnup of actinides. The goal is to examine the behaviour and influences of the hard neutron spectrum from spallation reactions in the transmutation, without the contribution or interference of multiplier subcritical medium, and compare the results with those obtained from the neutron fission spectrum. The results show that the transmutation efficiency is independent of the spallation target material used, and the neutrons spectrum from spallation does not contribute to increased rates of actinides transmutation even in the vicinity of the target.

1. Introduction

In the last 50 years, the nuclear systems have produced amounts of highly radioactive waste. The spent fuel consists of a wide variety of elements and isotopes. Most of the long-lived isotopes come from few transuranic elements as plutonium, neptunium, americium, and curium and some fission products such as technetium and iodine. Every year, a LWR of 1 GWh(e) discharges about 21 tons of radioactive fuel with the following inventory: 20 tons of uranium containing 0.9% (180 kg) of ^{235}U, 200 kg of plutonium, 21 kg of minor actinides (10 kg of neptunium, 10 kg of americium, and 1 kg of curium), and 760 kg of 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 of ^{129}I) that are radioactive isotopes with a long half-life [1]. Transmutation of Pu, minor actinides, and long half-life fission products is a promising concept to reduce the radioactive waste and its long-term radiotoxicity.

Many different technologies have been investigated for transmutation, including a variety of reactors, fusion-fission systems, and ADSs according to the long-term programme on Partitioning and Transmutation P&T of the Nuclear Energy Agency. The programme focuses on the possible benefits for waste management and used specific fuel cycle strategies, covering plutonium recycling and additional burning of minor actinides in dedicated reactor systems [2]. To understand and quantitatively assess the advantages and drawbacks of different plutonium and minor actinides (MA) burning strategies, it is important to investigate the mechanisms involved in the transmutation. The systems or strategies proposed are primarily distinguished according to their fast or thermal neutron spectra and/or used combined

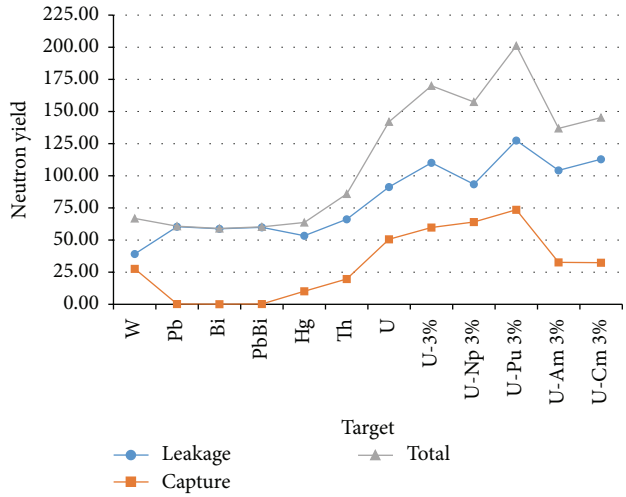

Figure 1: Neutrons produced per incident proton in target, beam energy 2.0 GeV. Image adapted from [13], spallation simulation in MCNPX 2.6.0, Bertini Model.

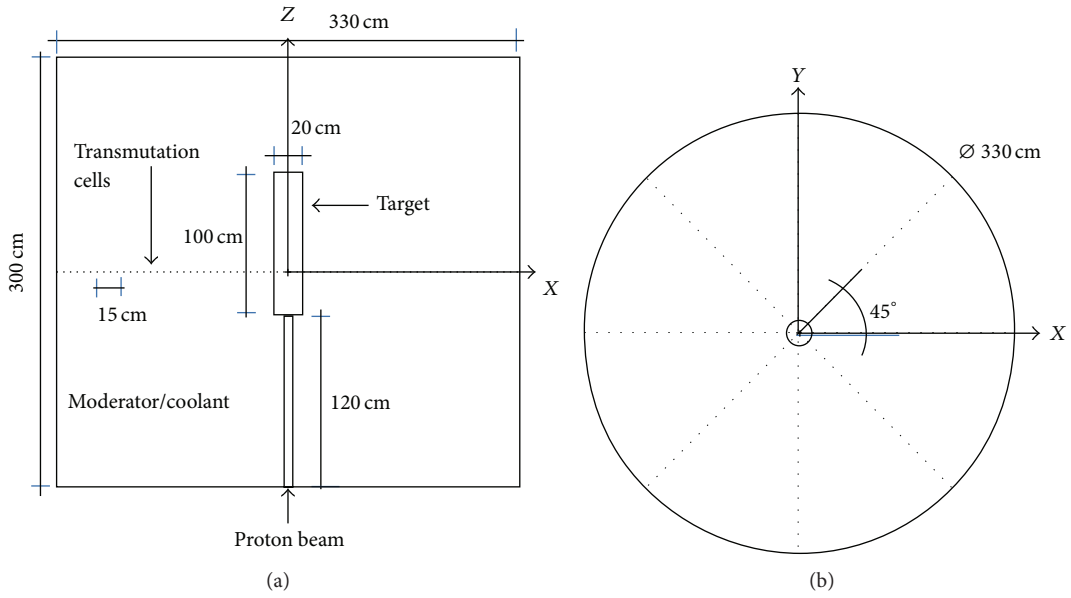

Figure 2: Radial (right) and axial (left) view of the proposed model, according to the Cartesian planes XZ and XY, respectively. The origin of the Cartesian coordinate system is located in the centre of the array (image obtained from the MCNPX Vised, version X 22S).

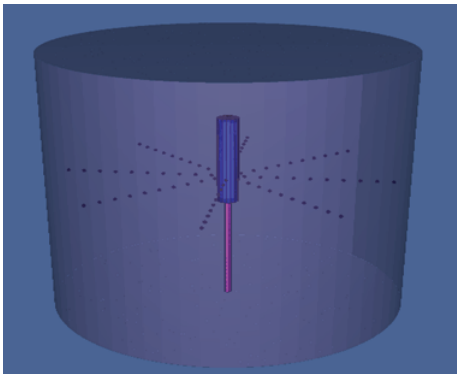

Figure 3: Arrangement view in three dimensions showing the external vessel, the target (blue), transmutation cells (black), and tube of beam (pink) (image obtained from the MCNPX Vised, version X 22S).

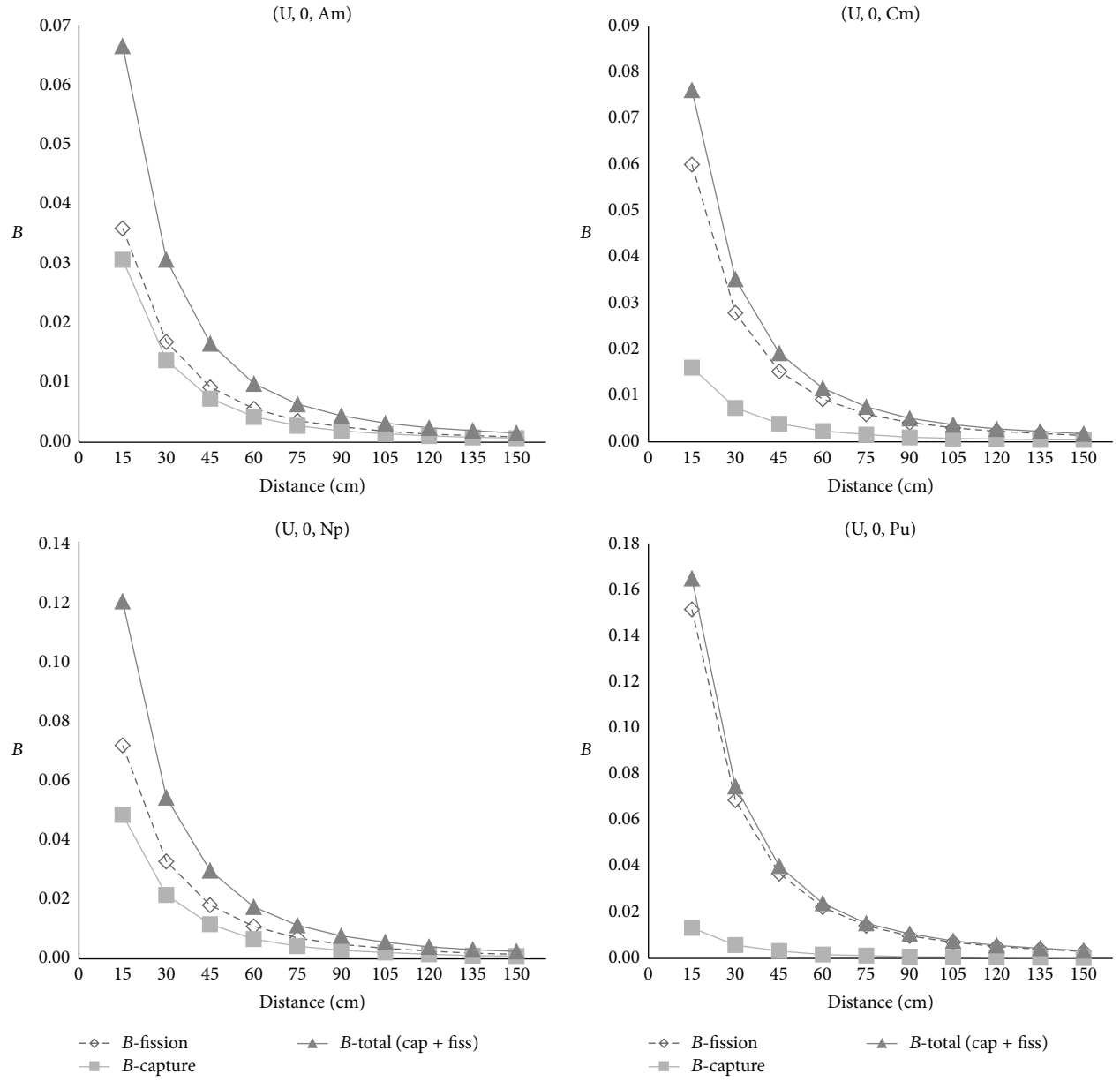

Figure 4: Relative number of reactions B in (U, 0, and TRU) as a function of the distance to the TRU. The proton beam energy of 2.0 GeV and 400,000 particles without moderator/coolant.

burning schemes [3–7]. 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 loss are in balance, may contain a quite limited amount of particular TRU mixture components like Pu, Np, Am, or Cm isotopes, due to safety constraints. This is one of the main reasons why the accelerator-driven systems (ADS) are currently studied worldwide for nuclear waste burning. ADS reactors have been proposed for many applications such as energy production, fertile-to-fissile transmutation, and conversion of long-lived radioisotopes to stable or much shorter-lived isotopes [8–11]. The transmutation technology to burnup the radioactive isotopes with long half-lives using an accelerator-driven subcritical system is one of the best solutions. ADSs can be designed to have a fast neutron energy spectrum, and they are subcritical being safer than 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 best comprehension of mechanisms involved in the transuranic transmutation in ADS-fission hybrid systems. The goal is to compare

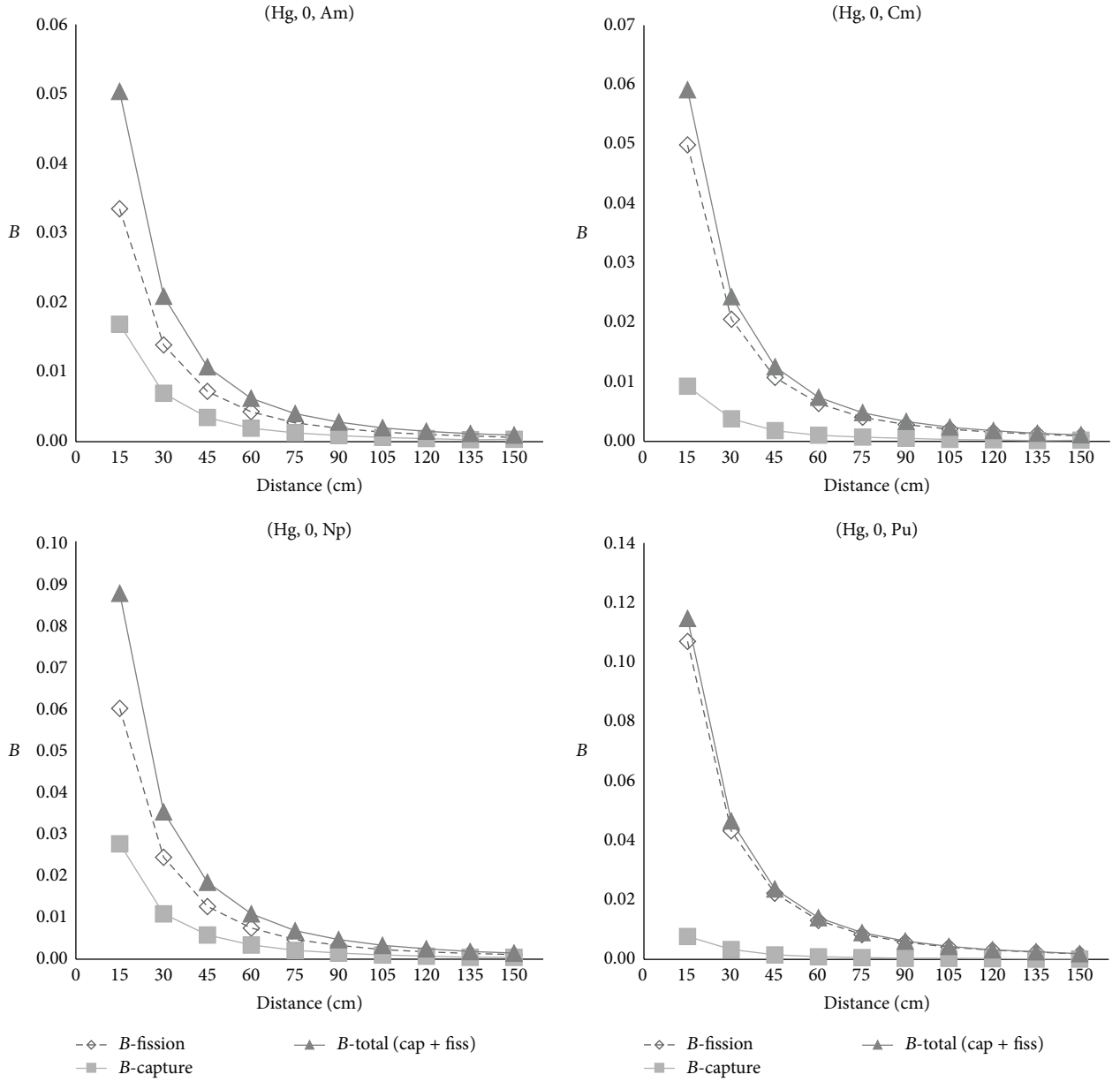

Figure 5: Relative number of reactions B in (Hg, 0, and TRU) as a function of the distance to the TRU. The proton beam energy of 2.0 GeV and 400,000 particles without moderator/coolant.

the influence of the neutrons spectrum from spallation reactions without the contribution of neutron spectrum from fission reactions and to compare the results with the standard fission source. Building a model to assess only the effects of neutrons spectrum from spallation in the TRU transmutation, the simulations evaluate the behaviour of the neutron flux emitted by the target (source of neutrons), focusing on the transuranic elements—Pu and minor actinides (MA)—considering graphite as the moderator and lead-bismuth and sodium, as coolant. The simulation involved the use of the Los Alamos High Energy Transport Code (LAHET) into Monte Carlo code MCNPX 2.6. The LAHET allows simulating the transport of a wide variety of particles and to analyse the production of neutrons through interaction of high energy protons with several targets material, in any form of geometry. The neutrons from spallation reactions are used to drive the subcritical nuclear assembly for calculations of long-lived TRU isotopes transmutation. The targets chosen, energy and geometry, were defined based on optimization developed in [13, 14]. These targets are also commonly reported in the literature.

The analysis will be focused on the capture (n, γ) and fission (n, f) reactions in the layers of transmutation allowing verifying the influences of the spallation source submitted

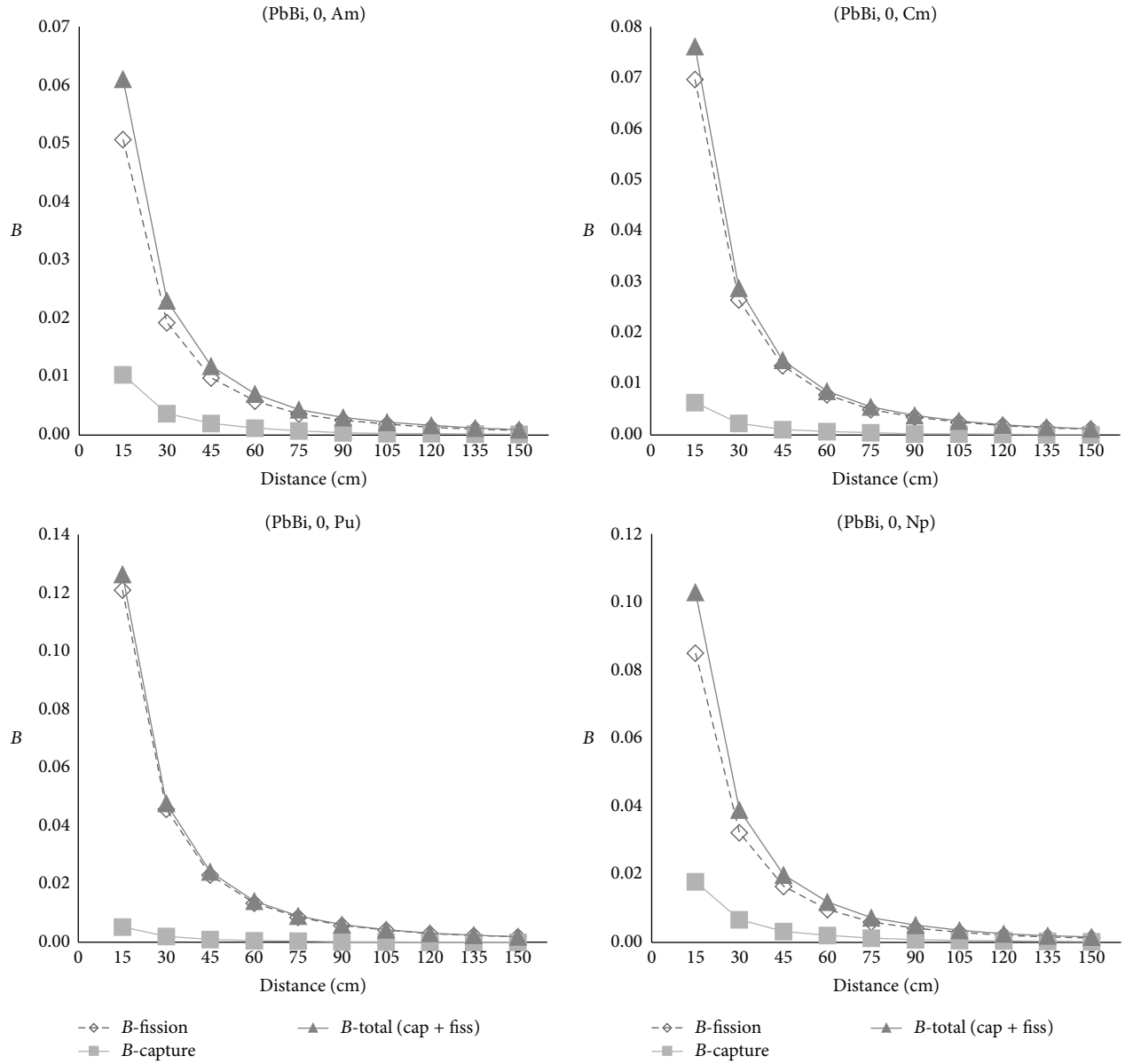

Figure 6: Relative number of reactions B in (PbBi, 0, and TRU) as a function of the distance to the TRU. The proton beam energy of 2.0 GeV and 400,000 particles without moderator/coolant.

in the empty, moderator, or coolant medium. A second extended analysis was generated comparing neutrons spectrum from spallation reactions versus neutrons spectrum from fission reactions. Reactions rates and the burning and depletion of actinides were analysed.

2. Calculation Procedure and Modelling

2.1. Methodology. The methodology was based on the model used by Hashemi-Nezhad and collaborators [15, 16], in which some modifications and new aspects were considered, such as the target and energy beam optimization, according to results obtained in [13]. Modifications in the geometry of the system allowed a significant reduction of the relative error, for values of the ≈1% in all the cells. Also, it opted for more realistic applications of P&T, based on the evaluation of a full isotopic composition of TRU. The isotopic composition of output by mass of fuel burnt 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 used in the transmutation layer.

The arrangement is simple and maintains a wide margin of subcriticality in order to avoid computation of the influence of fission neutrons generated in a subcritical medium.

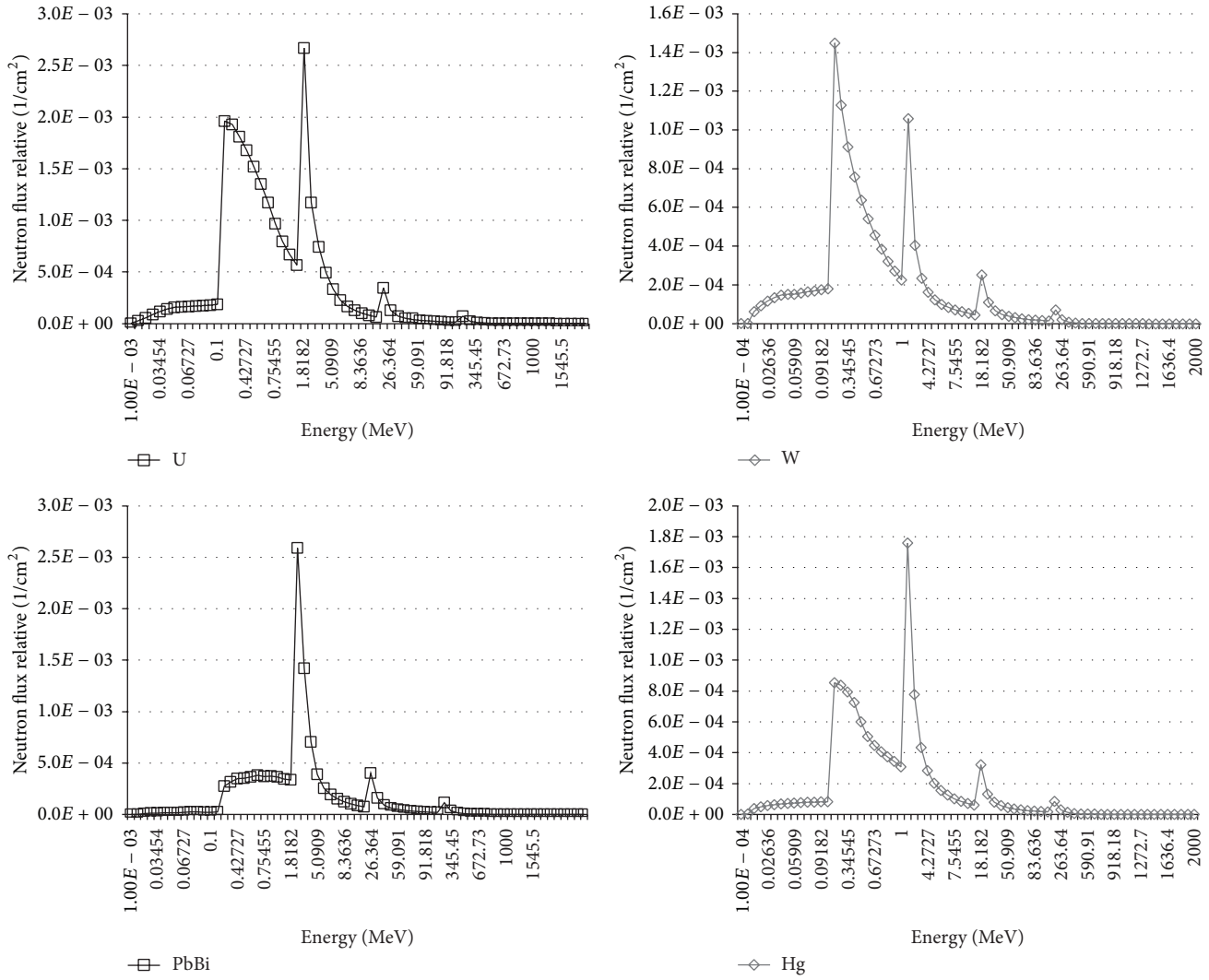

Figure 7: Neutrons emission spectrum for spallation reaction in different targets. 2.0 GeV proton beam, Bertini model.

It is necessary to ensure that only the influence of the hard neutron flux of yield target is computed, by the processes of intra/internuclear cascade, evaporation, and (n, xn) reactions, involved in the spallation reaction.

The configuration for the target, beam energy, and transport particles were based on a study presented in [14]. The simulation consists of a parabolic proton beam of 2.0 GeV, focusing on the top of the surface of a cylindrical target. For the MCNPX 2.6.0 simulations, it used Bertini intranuclear cascade model, continuous ENDF/B-VI neutrons microscopic cross sections at 293.6 K for all material, and LANL/T data library for beam proton. As shown in [13], temperature has little significance in the results presented here. In spallation reaction, it used 2×10^6 incident particles—protons (totalling > 107 neutron histories). It was considered a wide range of transport particles—muons, pions, alpha, deuterons, tritium, and heavy ions that take part in the secondary cascade reactions [13, 17].

2.2. Modelling and Calculations. The system consists of a subcritical arrangement with target immersed in a moderator or coolant with transmutation material. For the analysis of spallation neutron flux, it considers three different target materials—eutectic mixture of PbBi, Hg, and natural U. These materials and others were selected because they are commonly used in nuclear power systems and some of them are proposed as target ADS systems. In [13] their properties as spallation target are widely discussed. Figure 1 and Table 2 show the neutron emissivity results (neutron leakage, total neutrons produced, and neutrons captured, all in the target) for some target materials. The hard neutron flux from spallation is analysed and compared in three conditions: (a) target without moderator or coolant and with transmutation material—named Reference System; (b) target immersed in a coolant with transmutation material—Fast System; and (c) target immersed in a moderator with transmutation material—Thermal System. For compatibility

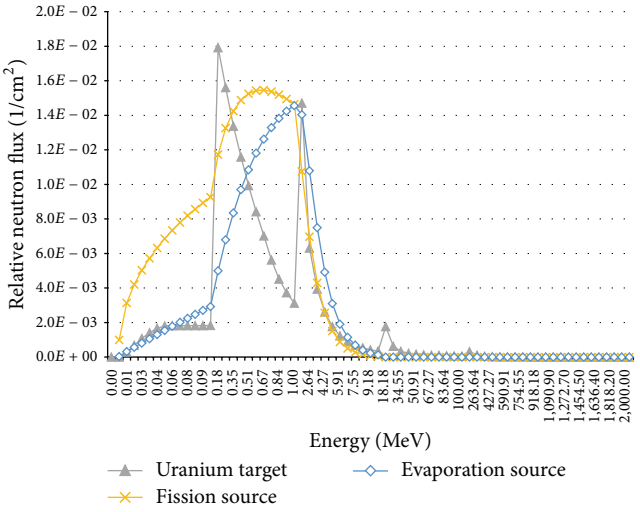

Figure 8: Neutrons emission spectrum for spallation reaction in U target (30 cm diameter, 2.0 GeV proton beam, Bertini Model) compared to MCNPX 2.6.0 standard evaporation and fission source.

the same nomenclature used in Hashemi-Nezhad et al. [15] was maintained, naming the systems as (target, medium, and TRU).

The analysed 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 centred in the XY plane at positions $Z = -30$ cm and $Z = 70$ cm, with the spheres filled with the material transmutation radially arranged in the XY plane and $Z = 0$. The spheres (transmutation layer) are each filled with transuranic elements (Np, Pu, Am, and Cm) according to the percentage of the isotope given in Table 1.

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 of 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 of diameter [13].

Preliminary simulations have shown that, for obtaining a relative error below 10% calculated in all cells, it is necessary to transport more than $nps > 10^6$ protons per simulation, requiring considerable computational time. Due to this long execution time, some alternative geometry is considered such as increasing the number of spheres and diameter, still maintaining a large margin of subcriticality as shown in Figures 2 and 3. The modification was made increasing the diameter of the balls from 1.5 cm to 3.0 cm, increasing the total number of spheres from 20 to 80, constituting 8 rows with 10 spheres spaced in 15 cm from each other and away from the centre, and occupying the initial distance of 15 until 150 cm. It was considered the sum of records (events, traces) of neutrons that reach on all cells (rings) equidistant from the centre of the array. This analysis is more coherent with the practice, where the composition of actinides follows a reprocessing matrix fuel instead of the rates isolated analysis for each isotope. The results obtained are compared with the typical pattern of a conventional critical system.

Table 1: Mass of each isotope of actinides in the fuel burned by a 1000 MWe PWR plant after 5 years cooling (adapted from [22]).

Radionuclide	Mass (kg)	Activity (Ci)
^{234}U	3.14	1.94*E*01
^{235}U	2.15*E*02	4.61*E* − 01
^{236}U	1.14*E*02	7.22
^{237}U	9.15*E* − 07	7.47*E*01
^{238}U	2.57*E*04	8.56
Total	**2.60E04**	**α 3.56E01** **β 7.47E01**
^{237}Np	2.04*E*01	1.44*E*01
^{239}Np	2.05*E* − 06	4.78*E*02
Total	**2.04E01**	**α 1.44E01** **β 4.78E02**
^{236}Pu	2.51*E*04	1.34*E*02
^{238}Pu	5.99	1.01*E*05
^{239}Pu	1.44*E*02	8.82*E*03
^{240}Pu	5.91*E*01	1.30*E*04
^{241}Pu	2.77*E*01	2.81*E*06
^{242}Pu	9.65	3.76*E*01
Total	**2.46E02**	**α 1.23E05** **β 2.81E06**
^{241}Am	1.32	4.53*E*03
^{242m}Am	1.19*E*02	1.16*E*02
^{243}Am	2.48	4.77*E*02
Total	**3.81**	**α 5.01E03** **β 1.16E02**
^{242}Cm	1.33*E* − 01	4.40*E*05
^{243}Cm	1.96*E* − 03	9.03*E*01
^{244}Cm	9.11*E* − 01	7.38*E*04
^{245}Cm	5.54*E* − 02	9.79
^{246}Cm	6.23*E* − 03	1.92
Total	**1.11**	**α 5.14E05**
Total	2.63*E*04	α 6.42*E*05 β 2.81*E*06

The results are shown in terms of the rates of reactions (n, γ) and (n, f) in the cells (spheres) filled with transmutation material. These reactions are the more important involving the TRU transmutations [15, 18, 19]. The results are obtained through a special treatment of MCNPX card event record that allows number counting of captures in a given nuclide or a specific combination of nuclides at the end of each history, converting the pulses created in a power distribution of a detector in neutron capture. The number

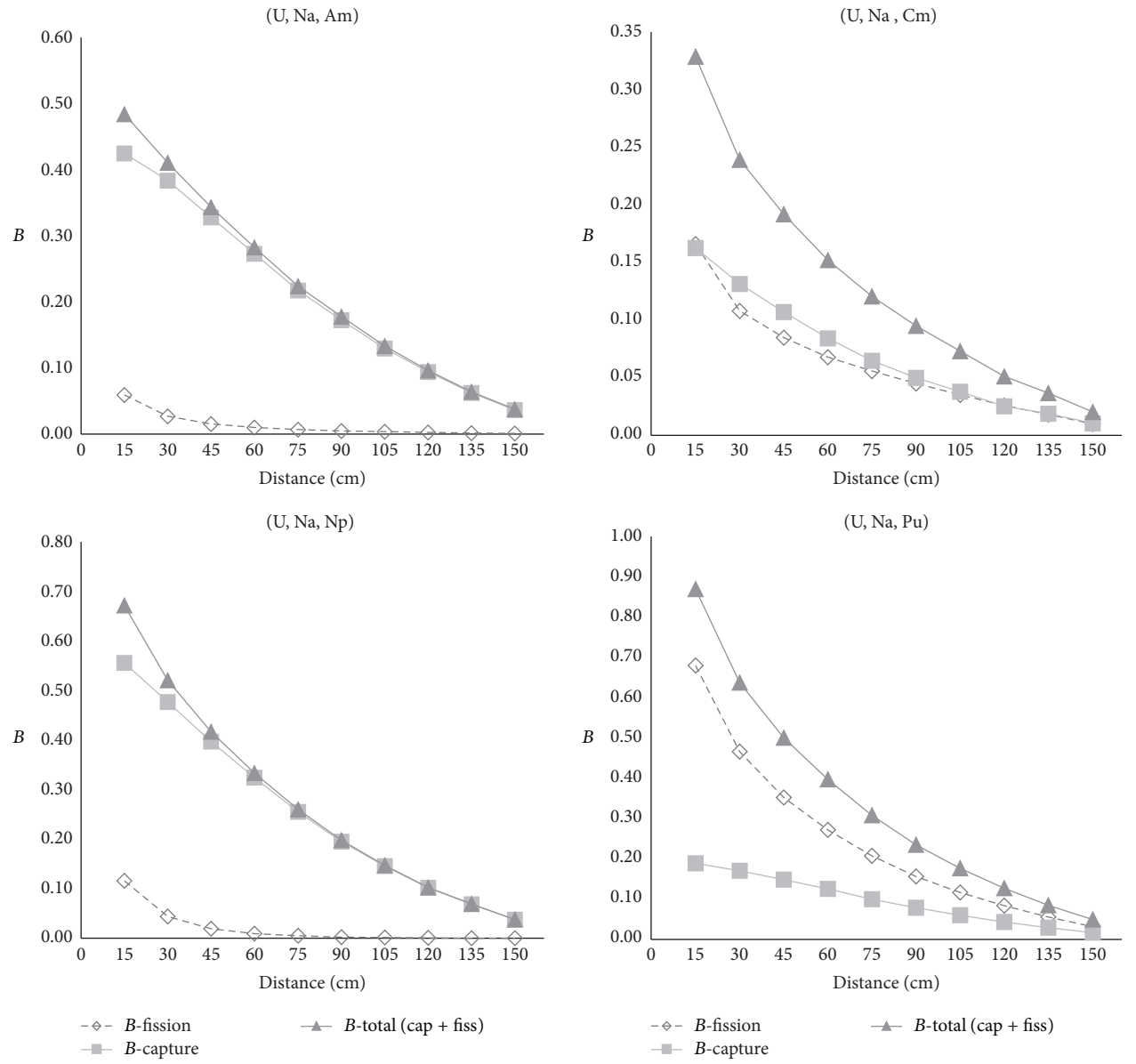

Figure 9: Relative number of reactions B in (U, Na, and TRU) as a function of the distance to the TRU. For proton beam energy of 2.0 GeV and 400,000 particles.

of records of 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 considering the material density, bulk, and Q value (average energy generated by each fission event ~200 MeV).

3. Analysis of Results

3.1. Reference System. The system without moderator is used as a reference model for comparison with the fast sodium or fast lead-bismuth systems and thermal graphite system. Figure 4 shows the rate of TRU transmutation via reactions (n, f) and (n, γ) as a function of the parameter B and the distance x from the centre of the system (U, 0, and TRU), Figure 5 for (Hg, 0, and TRU), and Figure 6 for (PbBi, 0, and TRU). The parameter B is defined as the relative number of interactions, while B' will represent the relative number of B interactions normalized to 1 g of material.

To systems without any moderation and scattering, the transmutation mechanism is more effective since all nuclides have much larger (n, f) fission reaction rates than (n, γ) capture reaction rates.

These behaviours can best be represented by the average value of the ratio between the capture and fission reactions $R = (B_{(n,f)}/B_{(n,\gamma)})$ as shown in Table 3. The ratio R is independent of the number of transport particles (i.e., the number of events) and material mass. This relation quantifies the effect in transmutation rates of the different neutrons spectrum emitted of each target.

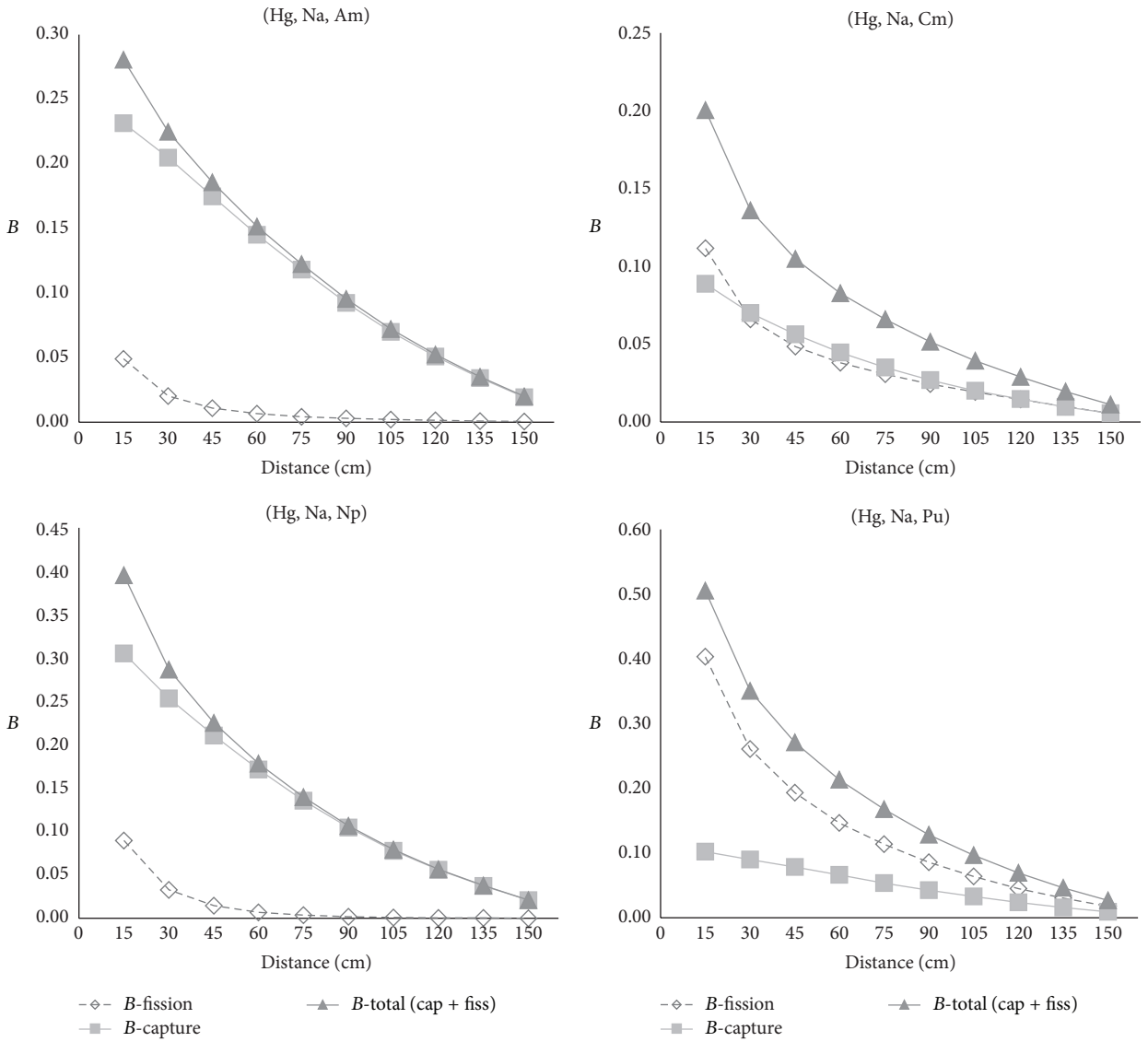

Figure 10: Relative number of reactions B in (Hg, Na, and TRU) as a function of the distance to the TRU. For proton beam energy of 2.0 GeV and 400,000 particles.

Table 2: Neutrons produced per incident proton in target, beam energy 2.0 GeV, and spallation simulation in MCNPX 2.6.0.

	W	Pb	Bi	PbBi	Hg	Th	U	U-3%	U-Np 3%	U-Pu 3%	U-Am 3%	U-Cm 3%
Leakage	39.2	60.5	58.8	60	53.4	66.2	91.3	110.1	93.4	127.5	104.2	112.9
Capture	22.6	0.4	0.2	0.3	10.3	19.8	50.7	59.9	64.1	73.7	32.7	32.4
Total	**66.9**	**60.8**	**59**	**60.3**	**63.7**	**85.9**	**142**	**170**	**157.5**	**201.1**	**136.9**	**145.3**

Table 3: Average value of the ratio of the capture and fission reactions $R = (B_{(n,f)}/B_{(n,\gamma)})$ for TRU fuels analysed in reference systems.

System	Material rate-$R = (B_{(n,f)}/B_{(n,\gamma)})$			
	R_{Am}	R_{Cm}	R_{Np}	R_{Pu}
(Hg, 0, TRU)	2.13	5.82	2.25	14.3
(PbBi, 0, TRU)	4.99	11.06	4.77	22.43
(U, 0, TRU)	1.3	3.88	1.64	11.9

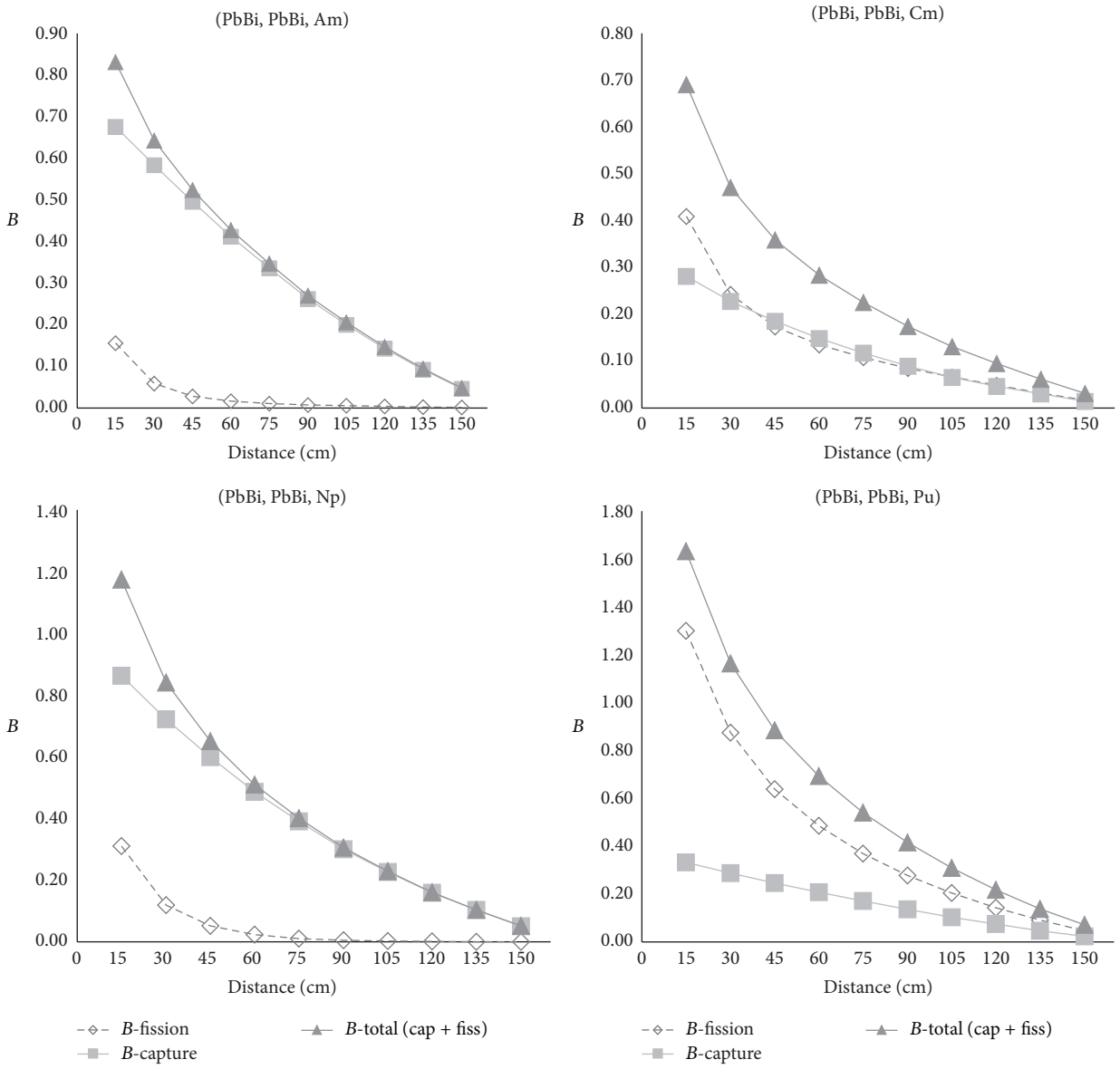

Figure 11: Relative number of reaction B in (PbBi, PbBi, TRU) as a function of the distance to the TRU. For proton beam energy of 2.0 GeV and 400,000 particles.

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

The ratio R also shows that the spectrum of neutrons emitted by the target of PbBi is harder than other targets. It is due to the low PbBi microscopic scattering cross section [13, 14]. Figure 7 shows the neutron emission spectrum to different targets. These results are consistent with the literature [2, 20, 21] corresponding to the typical energies expected for the process of evaporation, characterized by nuclear temperatures of $T = 2 \cdots 8$ MeV that follow Maxwellian distributions [20, 21]. Figure 7 shows also two high peaks: one of them is close to 2 MeV, approximately the value of the fission energy of neutron emission. The second wide peak is due to the process of capture/scattering of the target material. The PbBi target has only one peak centred close to 2 MeV because of

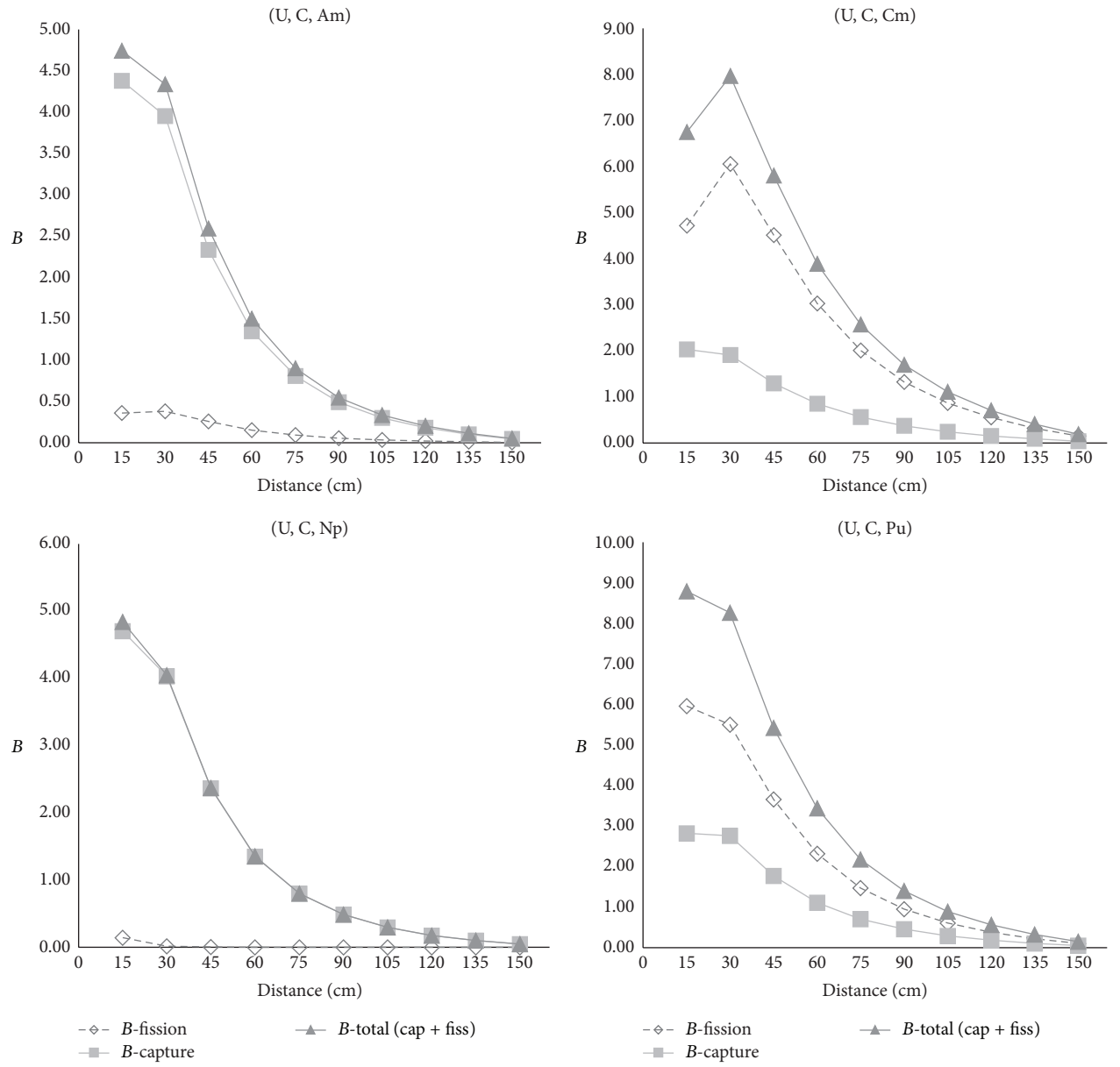

Figure 12: Relative number of reactions B in (U, C, and TRU) as a function of the distance to the TRU. For proton beam energy of 2.0 GeV and 400,000 particles.

its low scattering and capture cross sections. Figure 8 shows the neutron emission spectrum of the MCNPX standard evaporation and fission source, compared with the spallation spectrum of U target simulated using Bertini model. Due to the material properties, the expected theoretical distribution of neutrons emission spectrum in the temperatures involved is slightly modified with peaks resonant emission, although within the energy range expected. Other reactions such as (n, xn) were not considered in this study, due to the low emissivity of the target at energies above 4 MeV as shown in Figures 7 and 8.

3.2. Sodium Cooled Fast System. The sodium cooled system and eutectic lead-bismuth cooled system are shown in Figures 9–11, and Table 4 presents the average value of the ratio R in transmutation materials for all systems.

The analysis of these figures and ratio R show that:

(i) Comparing with the reference system, the Na and PbBi systems show opposite behaviour, increasing (n, γ) reactions to Am, Cm, and Np. Only Pu has the fission reactions as the main mechanism of transmutation.

(ii) The behaviour of the three systems is very similar and weakly dependent on the selected target, being the transmutation a direct function of moderator and coolant. For target distances larger than 30 cm, it is not possible to observe any distinction between

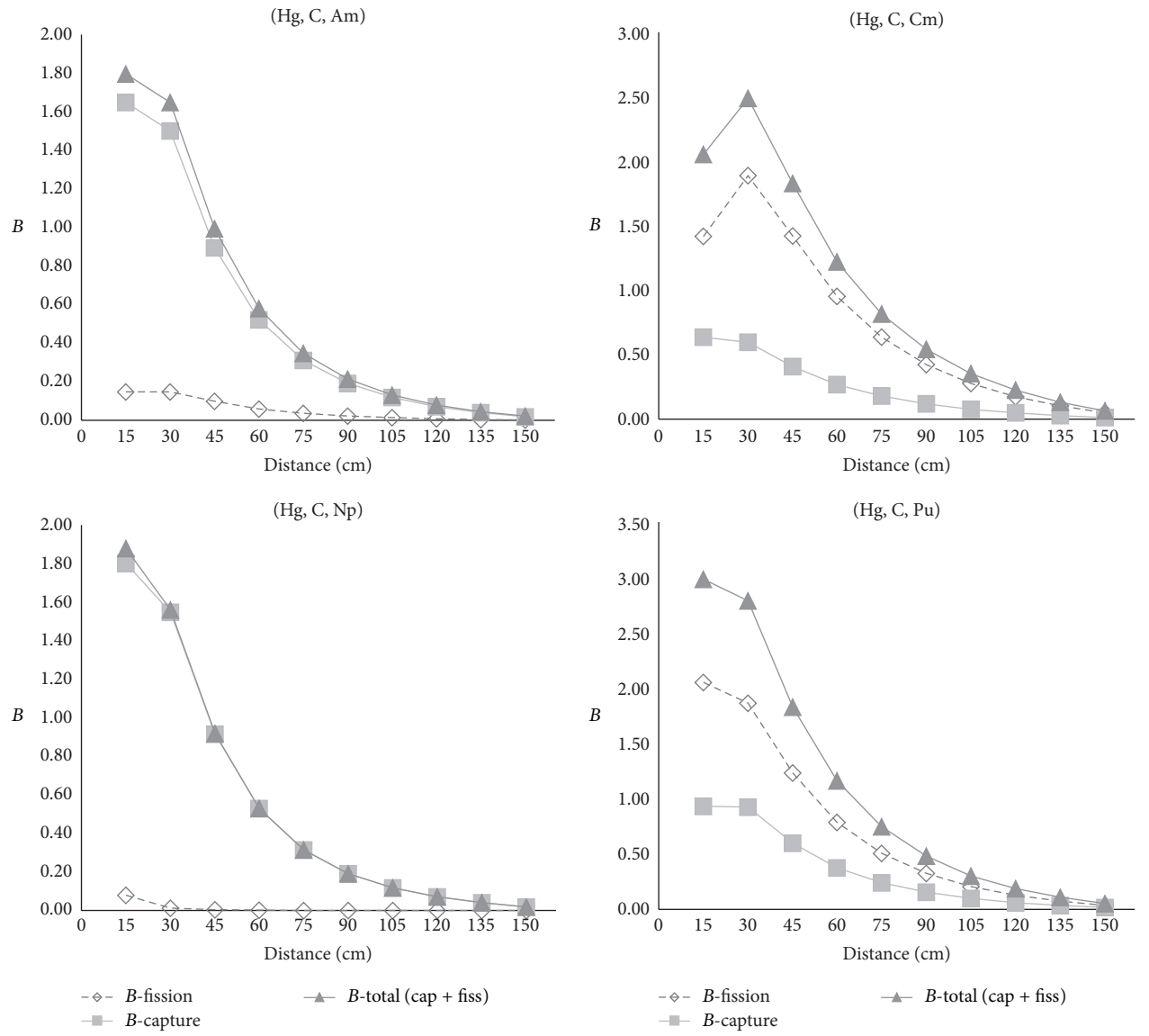

Figure 13: Relative number of reactions B in (Hg, C, and TRU) as a function of the distance to the TRU. For proton beam energy of 2.0 GeV and 400,000 particles.

Table 4: Average value of the ratio between fission and capture reactions for TRU fuels analysed in Na and PbBi systems.

System	Material rate-$R = (B_{(n,f)}/B_{(n,\gamma)})$			
	R_{Am}	R_{Cm}	R_{Np}	R_{Pu}
(Hg, Na, TRU)	0.034	0.96	0.024	2.06
(PbBi, PbBi, TRU)	0.03	1.026	0.022	2.091
(U, Na, TRU)	0.029	0.92	0.016	2.029

the R values in the three systems, indicating that despite of the low power of moderation, Na and PbBi have more influence than the emission spectrum of the target used. Although these results indicate this direction, this conclusion is not as trivial as shown forward, when performing the same analysis, but using a default neutron fission source.

(iii) The eutectic lead-bismuth has behaviour very similar to sodium. Again, despite of the low power of moderation, sodium and lead-bismuth, when compared

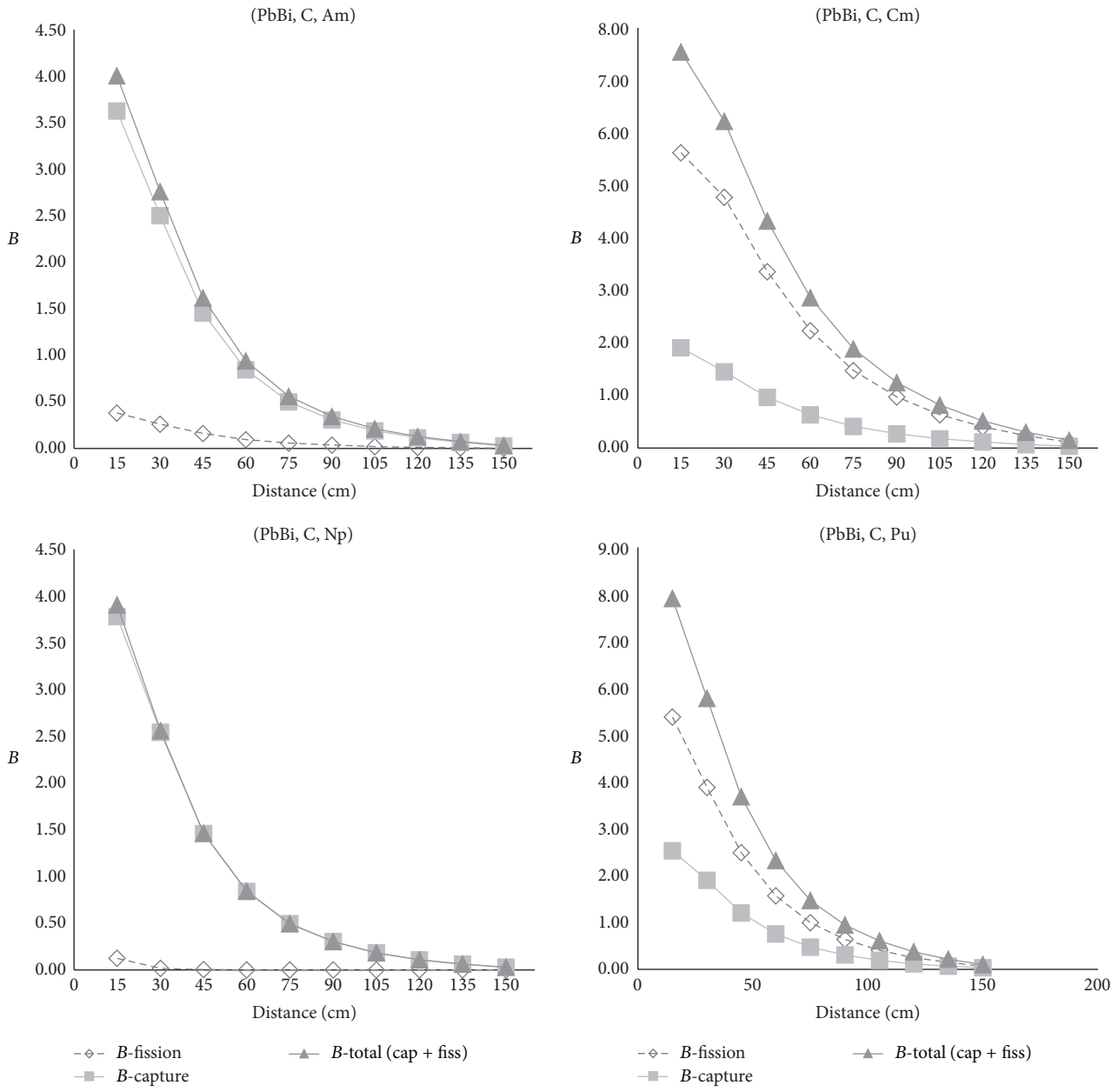

Figure 14: Relative number of reaction B in (PbBi, C, and TRU) as a function of the distance to the TRU. For proton beam energy of 2.0 GeV and 400,000 particles.

to the Reference System, significantly influence the neutrons spectrum and the reactions involved in the transmutation. This behaviour does not indicate significant neutronic differences between the metals in transmutation, although the values of Moderating Power and Moderating Ratio of Na and PbBi are different, as shown in Table 5.

3.3. Graphite Moderated System. The behaviour of the graphite-moderated system (U, C, and TRU) is shown in Figures 12–14. Table 6 shows the ratio R for the graphite moderated systems.

The results show the following.

(i) The ratios R of all targets have values closer than that in the case of Na and PbBi coolants. These data are consistent with expectations due to strong moderation related to the graphite and show that the transmutation of TRU in an ADS thermal system is independent of the chosen target. As it is evidenced in charts, at 30 cm from the target, there is only influence of the moderator on the transmutation.

(ii) For Am and Np, for the three systems, the transmutation occurs mainly via (n, γ) reaction, due to

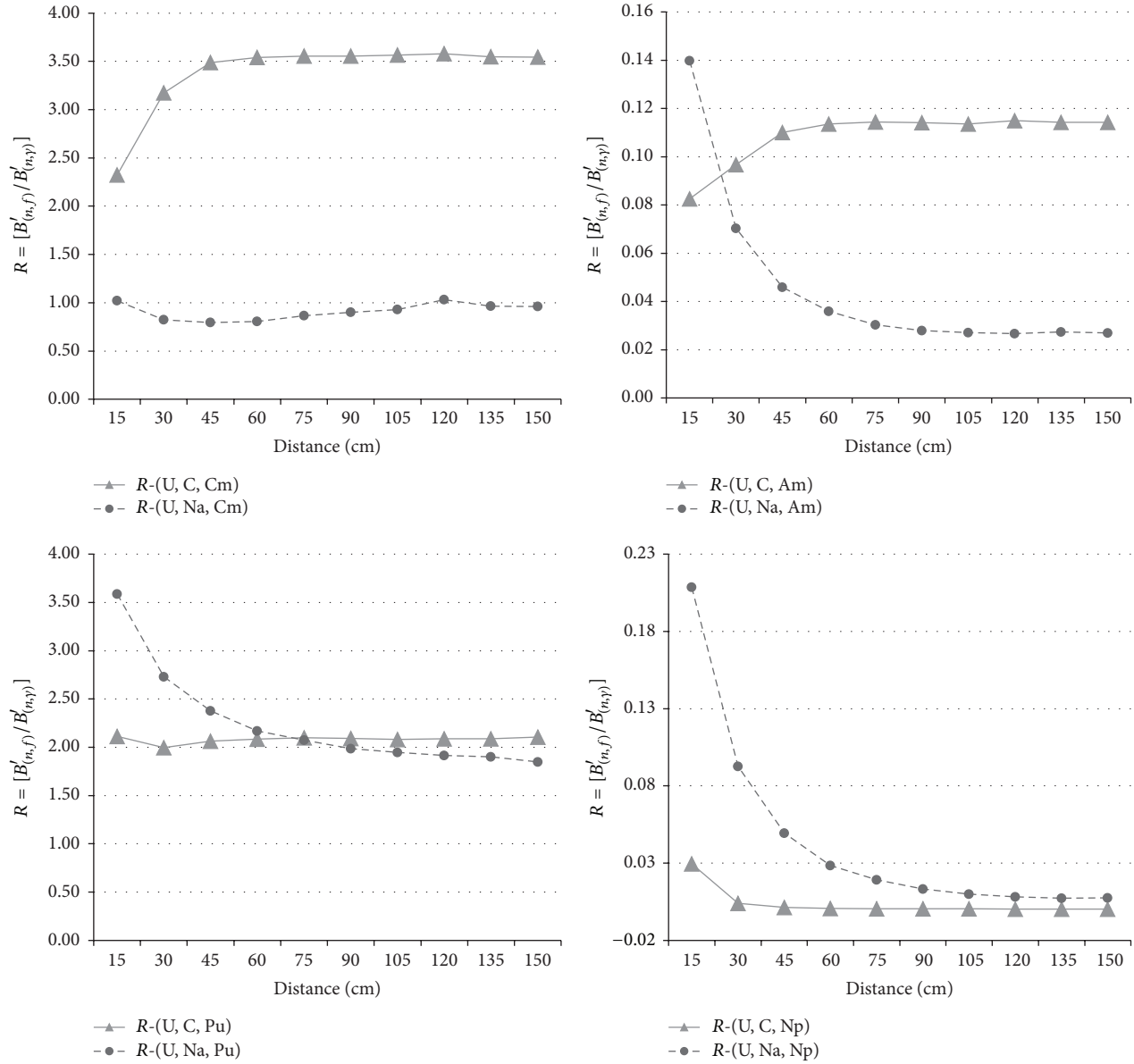

Figure 15: The ratio between the rates of fission and capture to (U, C, and TRU) and (U, Na, and TRU) systems at different locations.

Table 5: Some properties of the moderator/coolant used, with Σ_s and Σ_a being the scattering and absorption macroscopic cross sections, respectively.

Moderator	Average log. energy decrement ξ	Moderating power $\xi\Sigma_s$ (cm^{-1})	Moderating ratio $\xi\Sigma_s/\Sigma_a$	Mean free path $1/\Sigma_t$ (cm)	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 neutrons collisions to cross the energy range 10 keV–1 eV.
[(b)] Number of neutrons collisions to cross the energy range 2 MeV–0.025 eV.

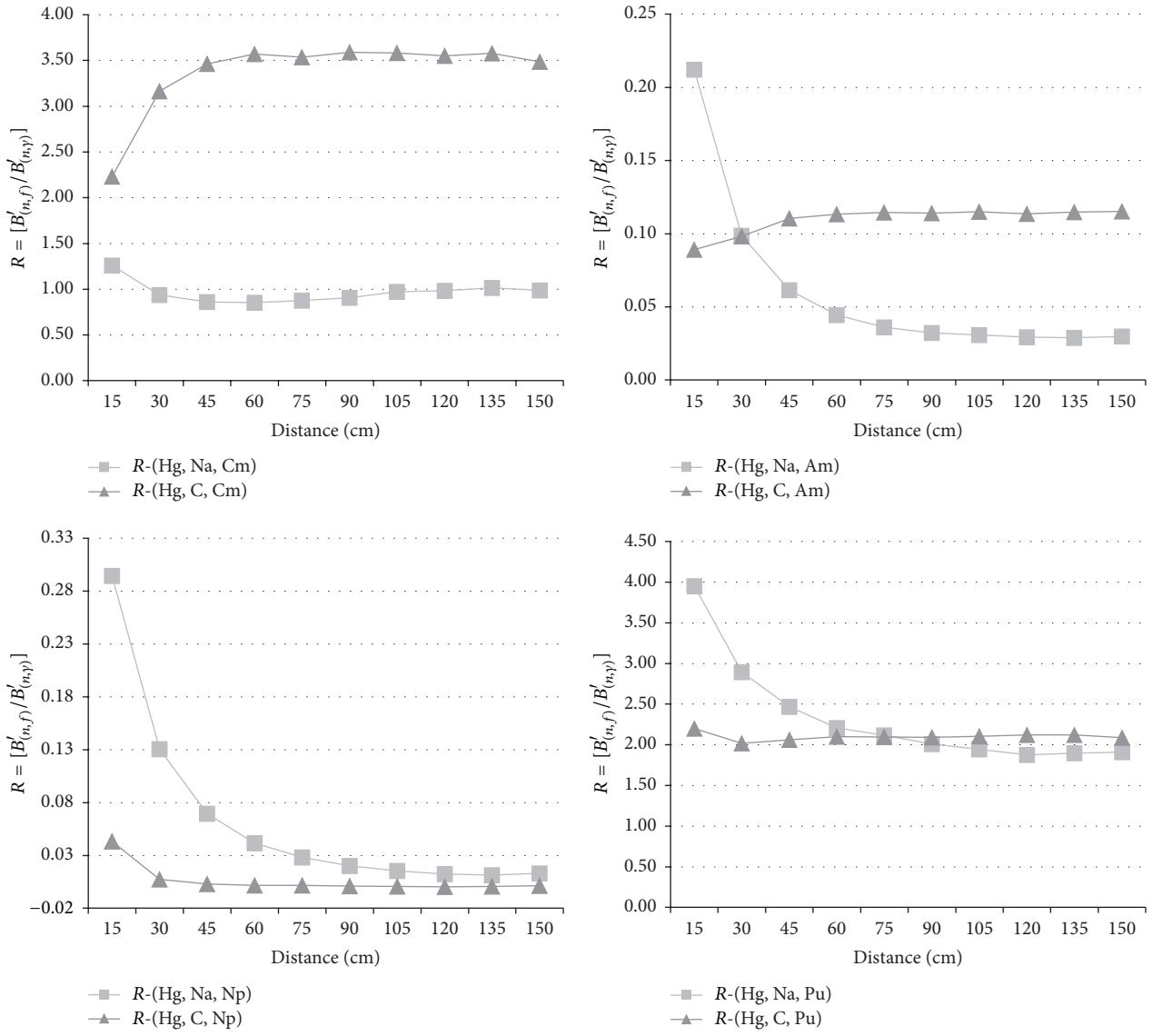

Figure 16: The ratio between the fission and capture rates for systems (Hg, C, and TRU) and (Hg, Na, and TRU) at different locations.

Table 6: Average value of the ratio between fission and capture reactions for TRU fuels analysed in C systems.

System	Material ratio-$R = (B_{(n,f)}/B_{(n,\gamma)})$			
	R_{Am}	R_{Cm}	R_{Np}	R_{Pu}
(Hg, C, TRU)	0.114	3.544	0.001	2.06
(PbBi, C, TRU)	0.114	3.554	0.001	2.085
(U, C, TRU)	0.114	3.544	0.0005	2.087

the low cross sections of the main isotopes. On the other hand, the Cm exhibits a different behaviour having higher rates of reactions than the Pu, inversely as occurred for Na and PbBi. The Pu behaves in all systems reactions as the main mechanism of transmutation, direct function of the high cross sections of its isotopes.

(iii) The results show that the rates of transmutation are independent of target material used. The TRU transmutation depends directly on moderator or coolant used in the system. These results are valid for any target and any energy proton beam incident [13, 14]. Under the conditions analyzed, the PbBi and Na coolants have very similar neutronic behaviours.

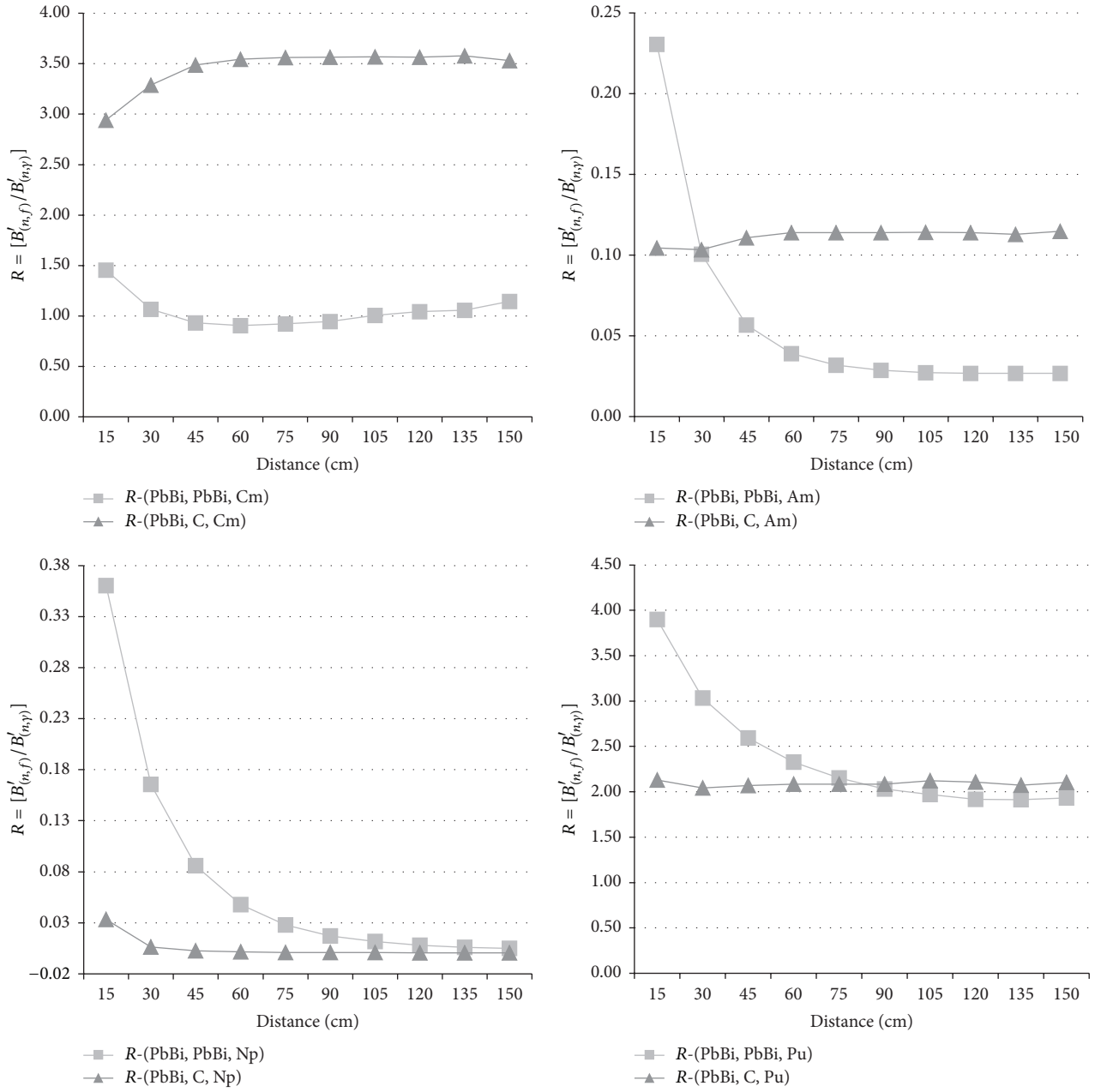

Figure 17: The ratio between the fission and capture rates for systems (PbBi, C, and TRU) and (PbBi, PbBi, and TRU) at different locations.

A comparison between the R for the U, Hg, and PbBi target, in the fast and thermal systems, is shown in Figures 15–17 for all TRU, while Figure 18 shows the values of total reaction rates B_T ($B_T = B_f + B_\gamma$).

The results show that:

(i) Pu—it presents a higher rate of (n, f) than (n, γ) reactions for all systems. It is due to the significant amount of fissile isotopes present, ≈69%, in the composition of Pu reprocessed, mainly the isotopes ^{239}Pu and ^{241}Pu (Table 1).

(ii) Np—its main transmutation mechanism is (n, γ) reactions. The main isotope, ^{237}Np, has a fission cross section for the thermal spectrum very low ~1 b, against a high capture cross section of 10,000 b. Such as quantified in charts, its reaction rates are small in all conditions with low transmutation.

(iii) Am—it has the lowest values for the overall reaction rates B_T. Its main transmutation mechanism is via (n, γ) reactions, which is due to high capture cross section in the thermal spectrum of the main

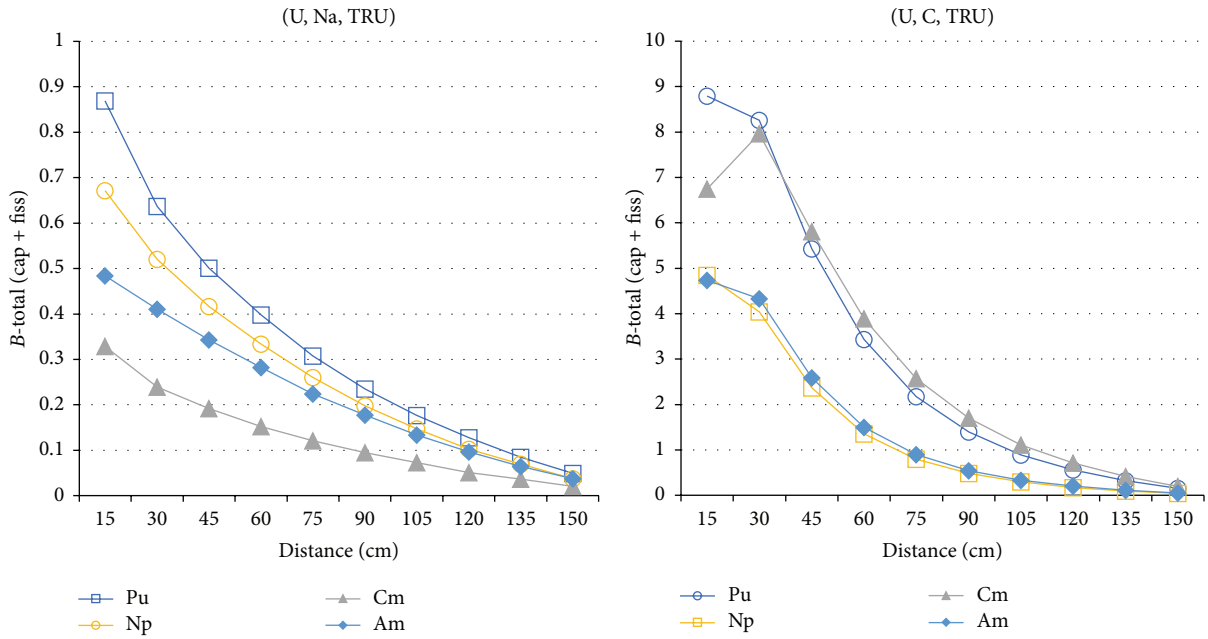

FIGURE 18: B_T TRU for the Na and C systems, as a function of the distance.

the isotopes ^{241}Am (~34%) and ^{243}Am (~65%), whose values are 50,000 b and 5,000 b, respectively.

(iv) Cm—it has features favourable to transmutation in a thermal system; the value of B_T is lesser for all actinides in a system without moderator, and higher than for Pu in a graphite-moderated thermal system. The Cm has three main isotopes with representative mass in the spent fuel, ^{242}Cm (~12%), ^{244}Cm (~82%), and ^{245}Cm (~5%). ^{242}Cm and ^{245}Cm are fissile with high fission and capture cross section for the thermal spectrum, while the ^{244}Cm (with the largest mass percentage) has low fission cross section (~80 b) and high capture cross section (~1,000 b).

3.4. Fission Source. Until now the results were obtained based on simulation of the source of protons focusing on a target, generating neutrons by spallation reaction and analyzing how the neutron spectrum emitted interferes with transmuting layer.

In this section it considers only the neutrons generated by a typical distribution of fission energy (fission source default available in MCNPX 2.6.0 for criticality calculation), allowing us to compare quantitatively the importance and difference between these two coexisting types of sources in a hybrid ADS-fission system. This comparison is performed by means of fission and capture reaction rates, in the uranium target system. The relative error was found to be less than 3% for all cells in all systems examined.

The comparison between sources is better represented by the average value of the ratio between the capture and fission reactions R, which is independent of the number of transport particles (i.e., the number of events) and material mass. This relation R allows us to infer the effect in transmutation rates of fission source in each TRU as shown in Figures 19–22.

The results show the spacing between the curves of the fission source and spallation reaction in the reference system, showing that despite the similarity of the neutron emission spectrum of these sources, the behavior is significantly different when analyzing interactions with actinides. This behavior is observed in Cm and Pu in the Na system, while the Am and Np approach its curves moving away from the center of source origin.

In graphite moderate systems, the curves are very close to each other due to the strong influence of moderator on the neutron spectrum. The results indicate that not considering the spallation source in the simulations involving the depletion of fuel in fast systems can lead to significant errors in the results.

The result shows that the reaction of spallation, with a hardening neutron spectrum, does not favor the increase in transmutation rates of TRU. In all systems examined, and in all TRU matrices, the ratios R are higher for the standard fission source, which indicates that the transmutation mechanism of direct process—fission reaction—is more statistically favorable to occur in a typical critical system, as summarized in Table 7.

It is important to note that despite the B reaction rates being higher than in the moderate system, it is incorrect to infer that thermal systems have higher reaction rates and thus greater efficiency as mentioned in [15]. The behavior displayed in the graphics for the reaction rates only reflects that the cross sections are higher in the thermal spectrum. Fast systems compensate the lower cross section in the spectrum

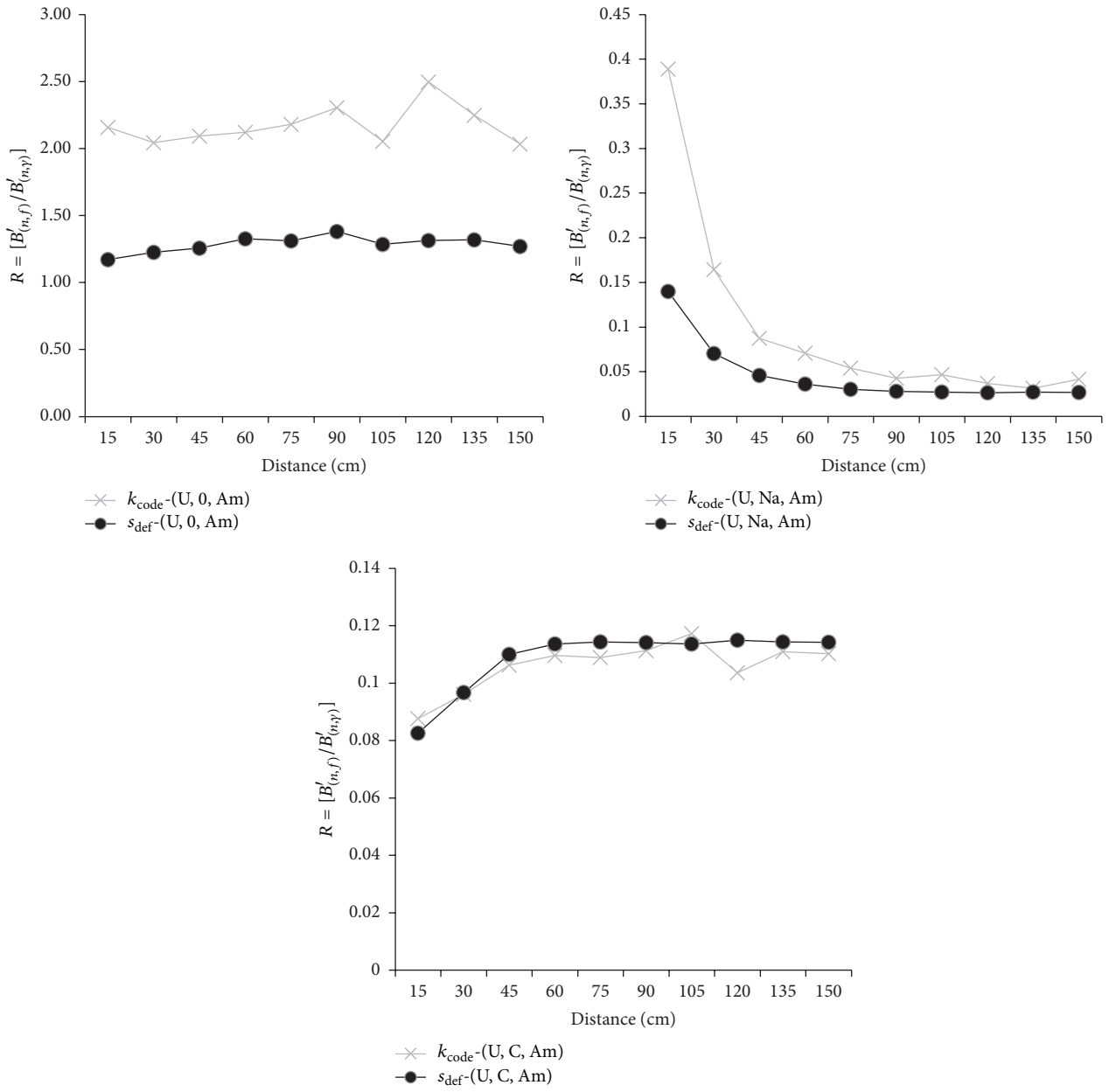

Figure 19: The ratio between fission and capture rates for systems (U, 0, and Am), (U, Na, and Am), and (U, C, and Am), at the different locations in x-axis. Comparison between fission source and spallation reaction models.

Table 7: Comparison of the average value of the ratio of the fission and capture reactions $R = (B_{(n,f)}/B_{(n,\gamma)})$ for TRU fuels analysed in uranium systems.

System	Material rate-$R = (B_{(n,f)}/B_{(n,\gamma)})$			
	R_{Am}	R_{Cm}	R_{Np}	R_{Pu}
$(U, 0, TRU)_{Fission}$	2.14	7.51	3.06	23.12
$(U, 0, TRU)_{Spallation}$	1.3	3.88	1.64	11.9
$(U, Na, TRU)_{Fission}$	0.5	2.85	0.05	6.43
$(U, Na, TRU)_{Spallation}$	0.029	0.92	0.016	2.029
$(U, C, TRU)_{Fission}$	0.11	3.8	0.0003	2.46
$(U, C, TRU)_{Spallation}$	0.114	3.544	0.0005	2.087

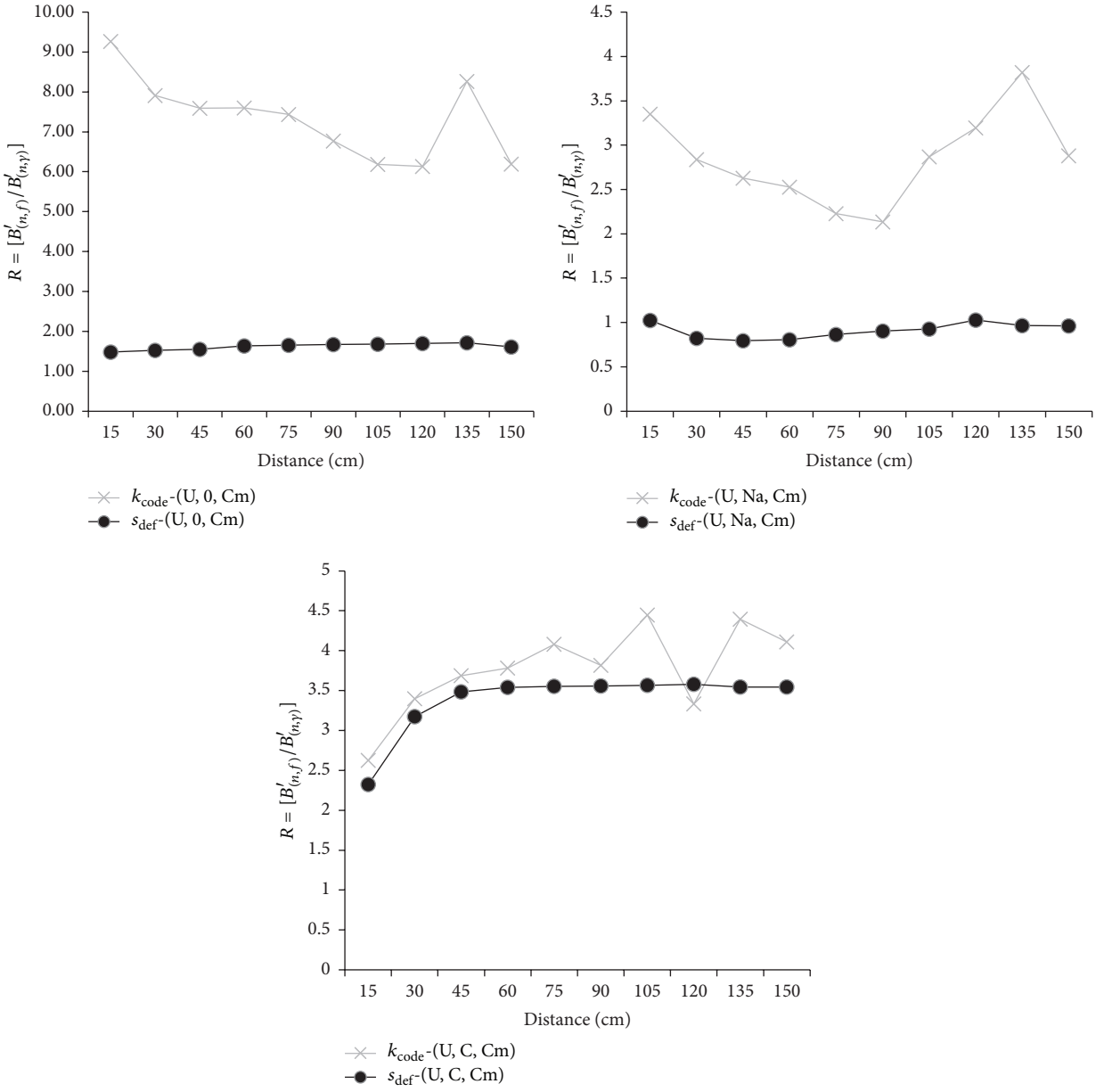

Figure 20: The ratio between fission and capture rates for systems (U, 0, and Cm), (U, Na, and Cm), and (U, C, and Cm), at the different locations in x-axis. Comparison between fission source and spallation reaction models.

and leakage with higher flows of neutrons by increasing the fuel enrichment. A fully closed loop based on thermal spectrum reactors presents difficulties P & T because there is a limit for the number of fuel reprocessing cycles due to the percentage increase of nonfissile isotopes. As an example, Figure 23 shows the burnup of the Pu matrix as fuel for sodium and reference systems. There is a drop in all Pu isotopes; however, the graphite moderated system exhibits a growth of ^{240}Pu and ^{242}Pu, fact related to very low fission cross sections for these nuclides to thermal neutrons, less than 1 b.

4. Conclusion

This study evaluated how the spectrum of neutrons emitted by different spallation targets, Hg, U, and PbBi, interacts with a layer composed of transuranic elements. The goal was to analyse the rates of reactions of fission and capture involved in the purpose of transmutation. The results allowed us to quantify and consider the mechanisms involved when neutron fluxes from spallation target, with or without the presence of a moderation medium, interact with the transmutation layer.

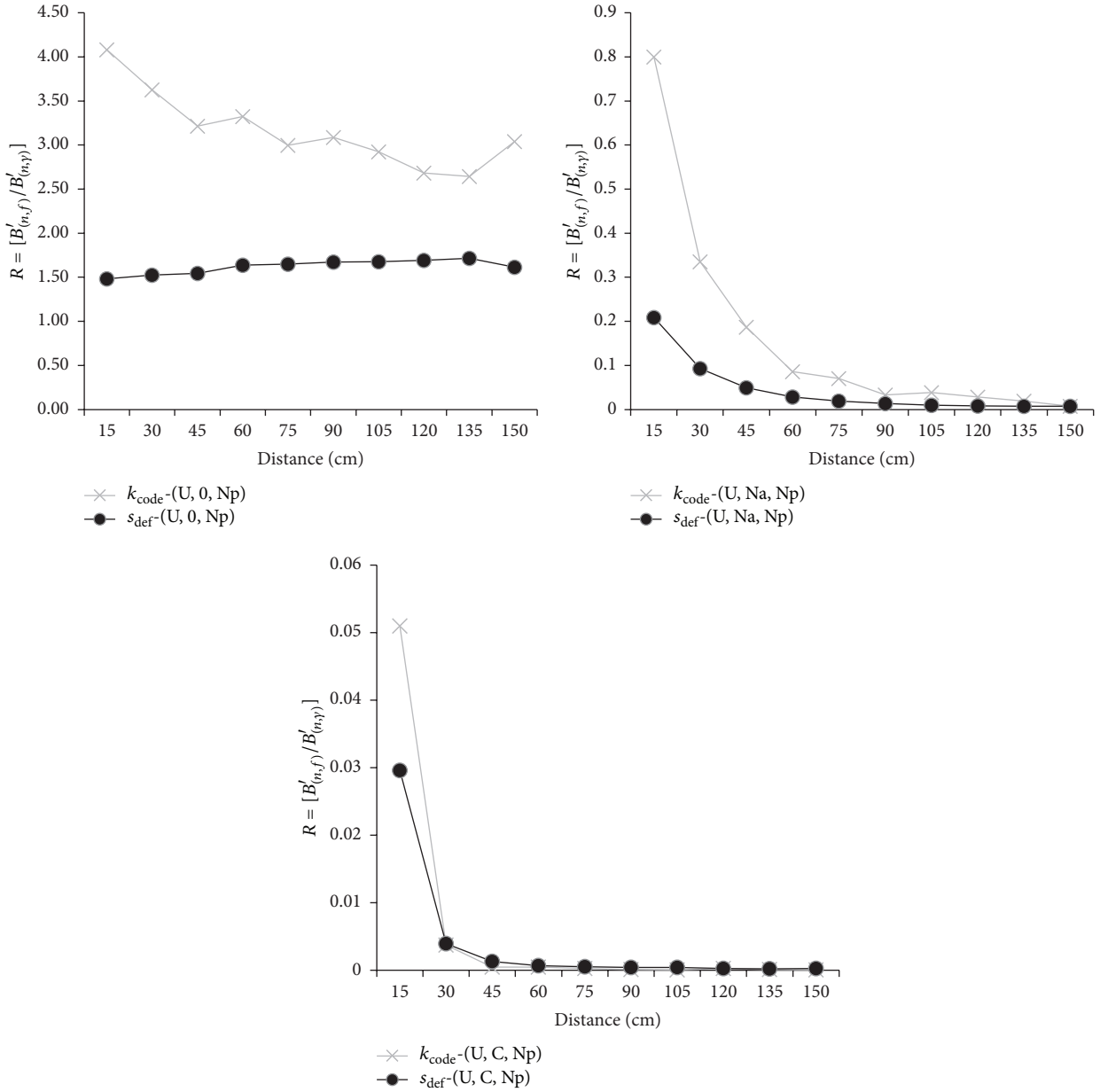

Figure 21: The ratio between fission and capture rates for systems (U, 0, and Np), (U, Na, and Np), and (U, C, and Np), at the different locations in x-axis. Comparison between fission source and spallation reaction models.

In the presence of a medium with low or high power moderation, there is no significant difference in the behaviour of the TRU transmutation using different targets. Transmutation in ADS system is independent of the target being a direct function of the medium, moderator/coolant used. However, in the simulations of burning fuel, the insertion of a spallation spectrum in addition to fission spectrum has relevance especially in fast systems. Spallation simulations involving PbBi and Na suggest that these coolants are neutron equivalent. The results do not indicate that the distribution of PbBi neutron spectrum has advantages front to the Na coolant.

Capture reactions in Am and Np, in environments with moderator/coolant, are 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 of the Cm is favoured in the moderated system having the fission as the main transmutation mechanism. About the Pu, it is easily converted in all systems having high reaction rates, with

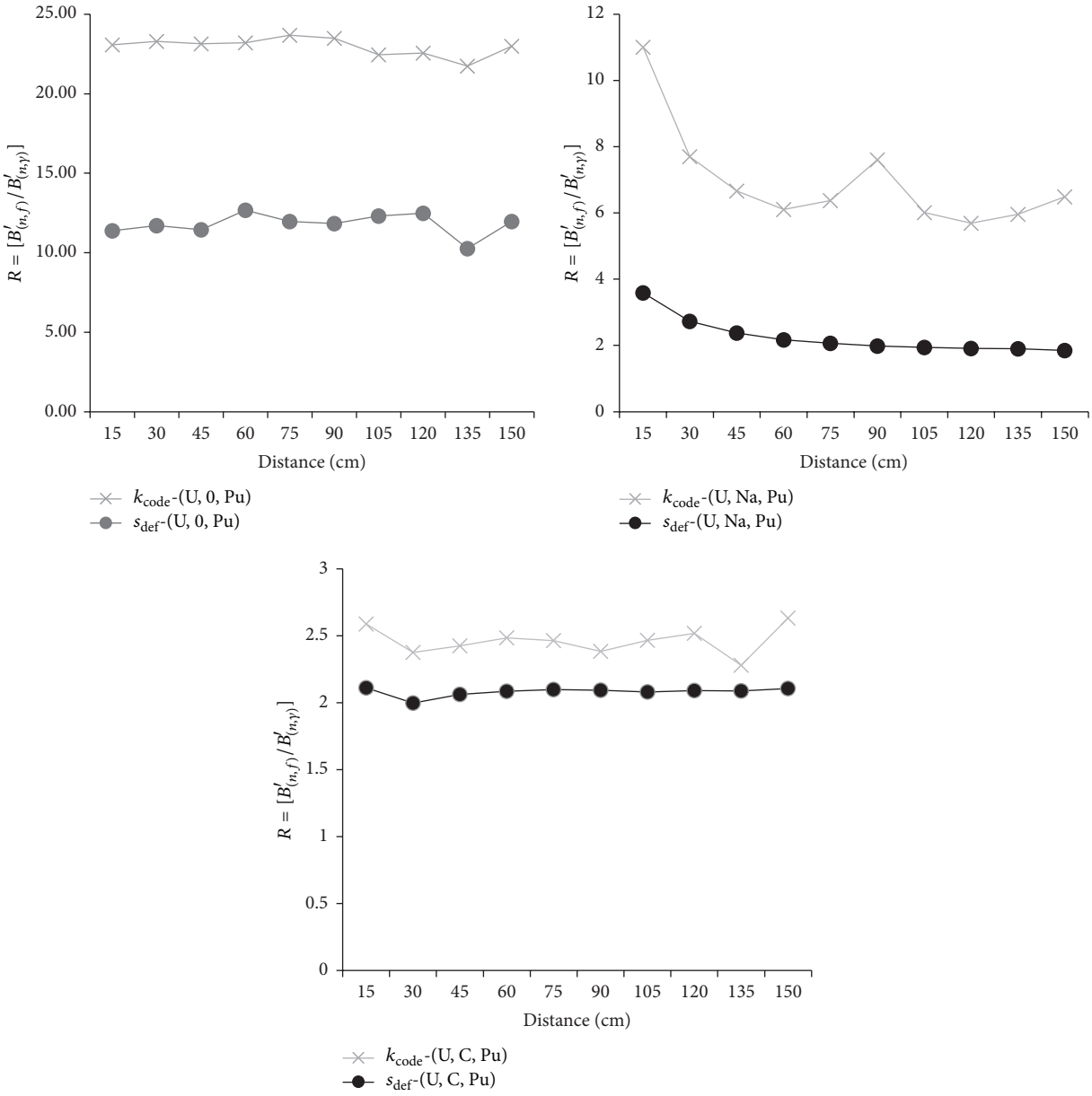

FIGURE 22: The ratio between fission and capture rates for systems (U, 0, and Pu), (U, Na, and Pu), and (U, C, and Pu), at the different locations in x-axis. Comparison between fission source and spallation reaction models.

fission reactions being higher than the capture reactions. However, mechanisms of transmutation via capture reaction may not be the best solution for reducing the isotopic inventory. The conversion of these isotopes with lower fission cross section may make the implementation of closed fuel cycles based on thermal systems difficult.

The efficiency of the reactor is most closely connected to the fuel burnup, the amount of energy generated per mass of fuel. The higher the reactor burnup, higher rates of reaction involved, thus increasing the chances of interaction between the neutron flux and low cross sections TRU isotopes along the burnup cycle. This is due to the greater probability of direct reactions (n, f) or by indirect reactions by (n, γ) successive absorptions and decay chains. The most hardening spallation spectrum in a ADS-fission hybrid system does not seem to have advantages transmutation in relation to a standard critical system. The advantages of recycling in ADS systems appear to be limited security issues, the flexibility of fuel combinatons, and the application dedicated to inventory reduction. As shown in [2], systems that combine closed fuel cycle and fast reactors have greater reduction in the inventory of spent fuel as well as economic advantages. From the point of view analyzed in this work, the results are corroborated herewith.

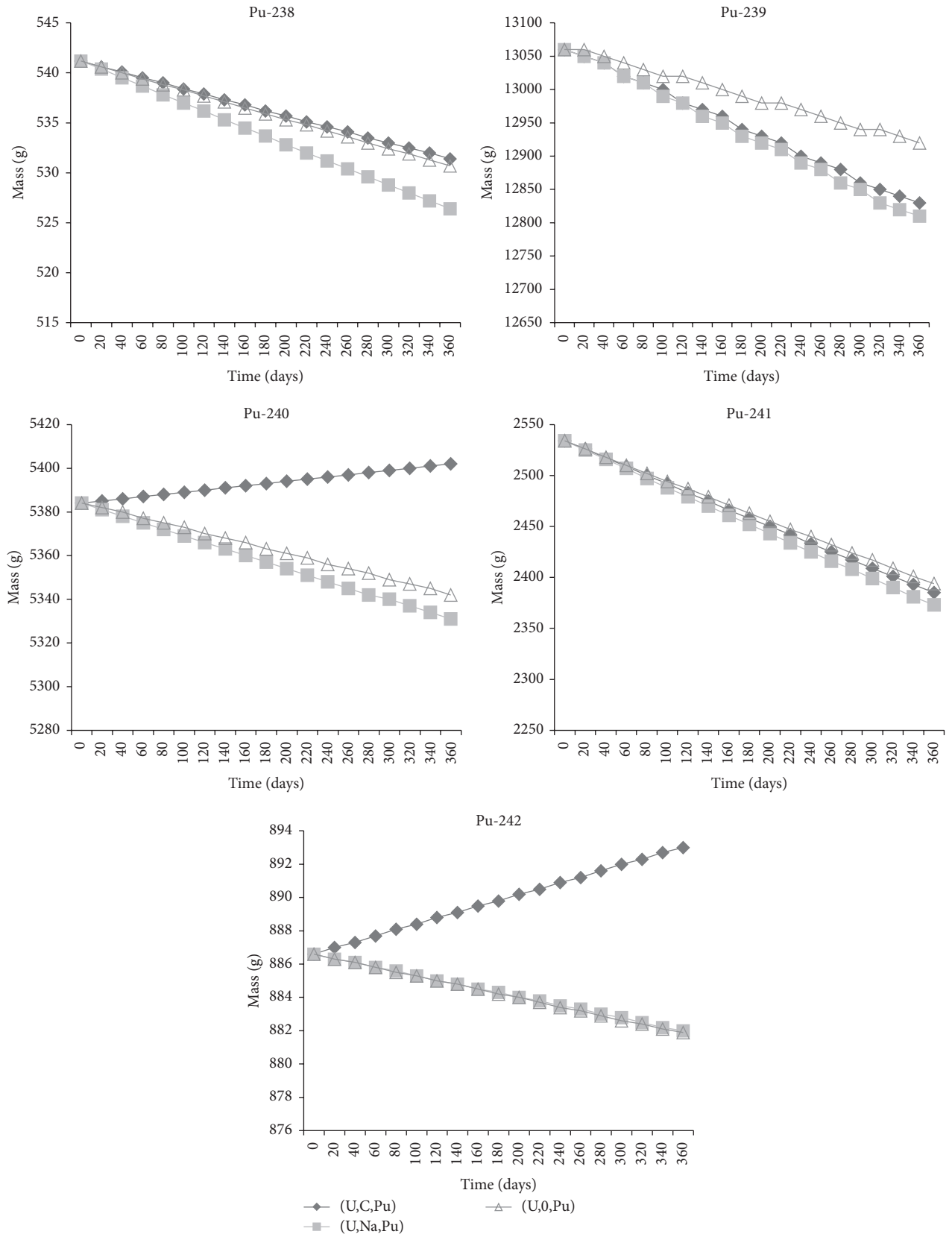

Figure 23: Variation of the mass to Pu isotopes, for 360 days of burnup, in the uranium target systems analyzed. MCNPX 2.6.0 standard fission source.

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

Acknowledgments

The authors are grateful to the Brazilian Research Funding Agencies, CNEN (Comissão Nacional de Energia Nuclear), CNPq (Conselho Nacional de Desenvolvimento e Pesquisa), CAPES (Coordenação de Aperfeiçoamento de Pessoal de Nível Superior), and FAPEMIG (Fundação de Amparo à Pesquisa do Estado de Minas Gerais), for the support.

References

[1] R. L. Garwin and G. Charpak, *Megawatts and Megatons a Turning Point in the Nuclear Ages?* Alfred A. Knopf, New York, NY, USA, 2001.

[2] Nuclear Energy Agency Organisation for Economic Co-operation and Development, *Accelerator-Driven Systems (ADS) and Fast Reactors (FR) in Advanced Nuclear Fuel Cycles: A Comparative Study*, Nuclear Energy Agency Organisation for Economic Co-operation and Development, Paris, France, 2002.

[3] G. P. Barros, C. Pereira, M. A. F. Veloso, and A. L. Costa, "Thorium and reprocessed fuel utilization in an accelerator-driven system," *Annals of Nuclear Energy*, vol. 1, pp. 1–7, 2015.

[4] R. V. Sousa, L. P. A. R. Tanure, D. F. Costa, C. Pereira, M. A. F. Veloso, and A. H. De Oliveira, "A preliminary neutronic evaluation of the high temperature nuclear reactor (HTTR) using reprocessed fuel," *Annals of Nuclear Energy*, vol. 65, pp. 232–238, 2014.

[5] A. Fortini, F. B. A. Monteiro, M. E. Scari et al., "Recent advances on the use of reprocessed fuels and combined thorium fuel cycles in HTR systems," *Progress in Nuclear Energy*, 2014.

[6] C. E. Velasquez, G. P. de Barros, C. Pereira, M. A. F. Veloso, and A. L. Costa, "Axial neutron flux evaluation in a Tokamak system: a possible transmutation blanket position for a fusion-fission transmutation system," *Brazilian Journal of Physics*, vol. 42, no. 3-4, pp. 237–247, 2012.

[7] F. C. Silva, C. Pereira, M. A. F. Veloso, and A. L. Costa, "Shifting study of a VHTR using reprocessed fuel with various TRISO packing fractions," *Nuclear Engineering and Design*, vol. 248, pp. 42–47, 2012.

[8] C. Rubbia, J. A. Rubio, S. Buono et al., "Conceptual design of a fast neutron operated high power energy amplifier," Tech. Rep. CERN/AT/95-44 (ET), European Organization for Nuclear Research (CERN), Meyrin, Switzerland, 1995.

[9] C. M. Van Atta, J. D. Lee, and W. Heckrotte, "The electronuclear conversion of fertile to fissile material," Tech. Rep. UCRL-52144, Lawrance Livermore Laboratory, 1970.

[10] C. D. Bowman, E. D. Arthur, P. W. Lisowski et al., "Nuclear energy generation and waste transmutation using an accelerator-driven intense thermal neutron source," *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, vol. 320, no. 1-2, pp. 336–367, 1992.

[11] M. Salvatores, I. Slessarev, A. Tchistiakov, and G. Ritter, "The potential of accelerator-driven systems for transmutation or power production using thorium or uranium fuel cycles," *Nuclear Science and Engineering*, vol. 126, no. 3, pp. 333–340, 1997.

[12] A. Rineiski and W. Maschek, *Dynamics and Reactivity Control in Accelerator Driven Systems for Nuclear Waste Burning*, Nuclear Energy, Forschungszentrum (FZK), Institute for Nuclear and Energy Technologies, Karlsruhe, Germany, 2006.

[13] M. Gilberti, *Contribuições ao Desenvolvimento de Sistemas Sub-Críticos Dirigido por Aceleradores Aplicados à Transmutação de Rejeitos Altamente Radioativos [Master thesis]*, Departamento de Engenharia Nuclear, UFMG, Belo Horizonte, Brazil, 2011, (Portuguese).

[14] M. Gilberti, C. Pereira, M. A. F. Veloso, A. L. Costa, and C. A. M. Silva, "Evaluation of spallation source for neutrons production by Monte Carlo methods," in *Proceedings of the 8th International Conference on Mathematical Problems in Engineering, Aerospace and Sciences*, pp. 1–8, São José dos Campos, Brazil, 2010.

[15] S. R. Hashemi-Nezhad, R. Brandt, W. Westmeier et al., "Monte Carlo calculations on transmutation of trans-uranic nuclear waste isotopes using spallation neutrons: difference of lead and graphite moderators," *Nuclear Instruments and Methods in Physics Research A*, vol. 481, no. 1–3, pp. 547–557, 2002.

[16] S. R. Hashemi-Nezhad, R. Brandt, W. Westmeier et al., "Monte Carlo studies of accelerator driven systems: energy and spatial distribution of neutrons in multiplying and non-multiplying media," *Nuclear Instruments and Methods in Physics Research A*, vol. 482, no. 1–3, pp. 537–546, 2002.

[17] F. Goldenbaum, *The Physics of Spallation Processes: Theory, Experiments and Applications*, Institut Für Kernphysik: Forschungszentrum Jülich Gmbh, 2004.

[18] J. J. Duderstadt and L. J. Hamilton, *Nuclear Reactor Analysis*, John Wiley & Sons, New York, NY, USA, 1976.

[19] H. Nifenecker, "Hybrid nuclear reactors," *Progress in Particle and Nuclear Physics*, no. 43, pp. 683–827, 1999.

[20] F. Goldenbaum, *The Physics of Spallation Processes: Theory, Experiments and Applications*, Institut Für Kernphysik, Forschungszentrum Jülich Gmbh, 2004.

[21] K. Tucek, *Neutronic and Burnup Studies of Accelerator-driven Systems Dedicated to Nuclear Waste Transmutation*, Department of Physics, Royal Institute of Technology, Stockholm, Sweden, 2004.

[22] Committee on Separations Technology and Transmutation Systems Board on Radioactive Waste Management Commission on Geosciences and Environment and Resources National Research Council, *Nuclear Wastes: Technologies for Separations and Transmutation*, National Academy Press, Washington, DC, USA, 1996.