3rd International Workshop on ACCELERATOR-DRIVEN
SUB-CRITICAL SYSTEMS & THORIUM UTILIZATION
Virginia Commonwealth University, Richmond, Virginia, USA
October 14-17,2014

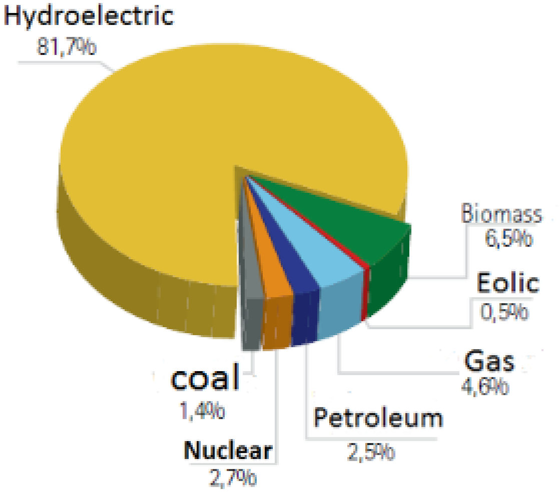
Effects of Fusion and Spallation Spectra on Ganex Reprocessed Fuel Evolution

Carlos E. Velasquez, Graiciany de P. Barros, Maurício Gilberti, Claubia Pereira, Maria Auxiliadora F. Veloso,and Antonella L. Costa

UFMG

Nuclear Energy in Brazil

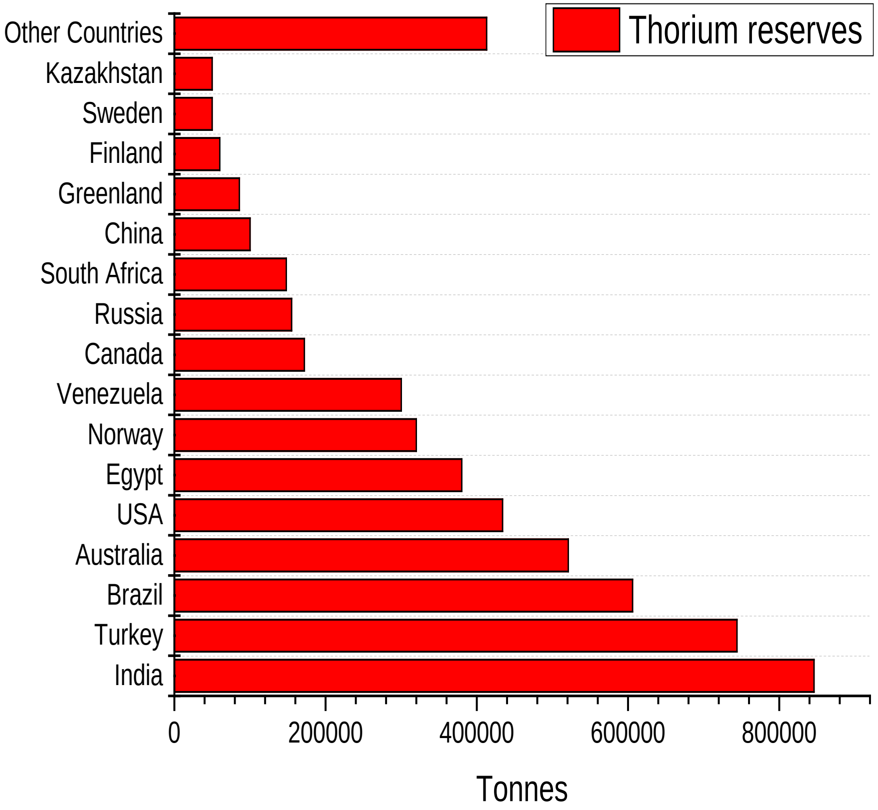
- Currently, there are two reactors operating Angra I and Angra II, in Brazil. The third reactor Angra III is under construction.
- The nuclear contribution to the electricity generation is about 2,7%



Thorium reserves

- Thorium is a valuable energy source since it is about three to four times more abundant in the earth's crust than uranium

- Brazil has the third world thorium reserve.



OECD NEA & IAEA Uranium 2011: Resources, Production and Demand (Red Book) 2011

Nuclear Engineering Department/UFMG

- The nuclear engineering department (DEN) called PCTN at UFMG was founded in 1968.
- The PCTN commitment is to deepen the professional and academic knowledge.
- Train and develop the students research skills and abilities

Fields of Study

- Nuclear Reactor and Nuclear Fuel Cycle Technology
  - Reactor and nuclear fuel cycle physics
  - Sub-critical and minor actinide systems
  - Long-lived fission products recycle
  - High temperature reactor
  - Advanced PWR
- Thermal-hydraulic analysis
- Thorium fuel cycle investigations

The Issue of Nuclear Energy

- Since the beginning of the nuclear fission utilization, nuclear waste has been generating.
- The majority of the spent fuel goes to a final repository long and secures enough to the population contact.
- Nevertheless, the storage capacity eventually wont be enough, not to mention the expenses to take care of it are high.

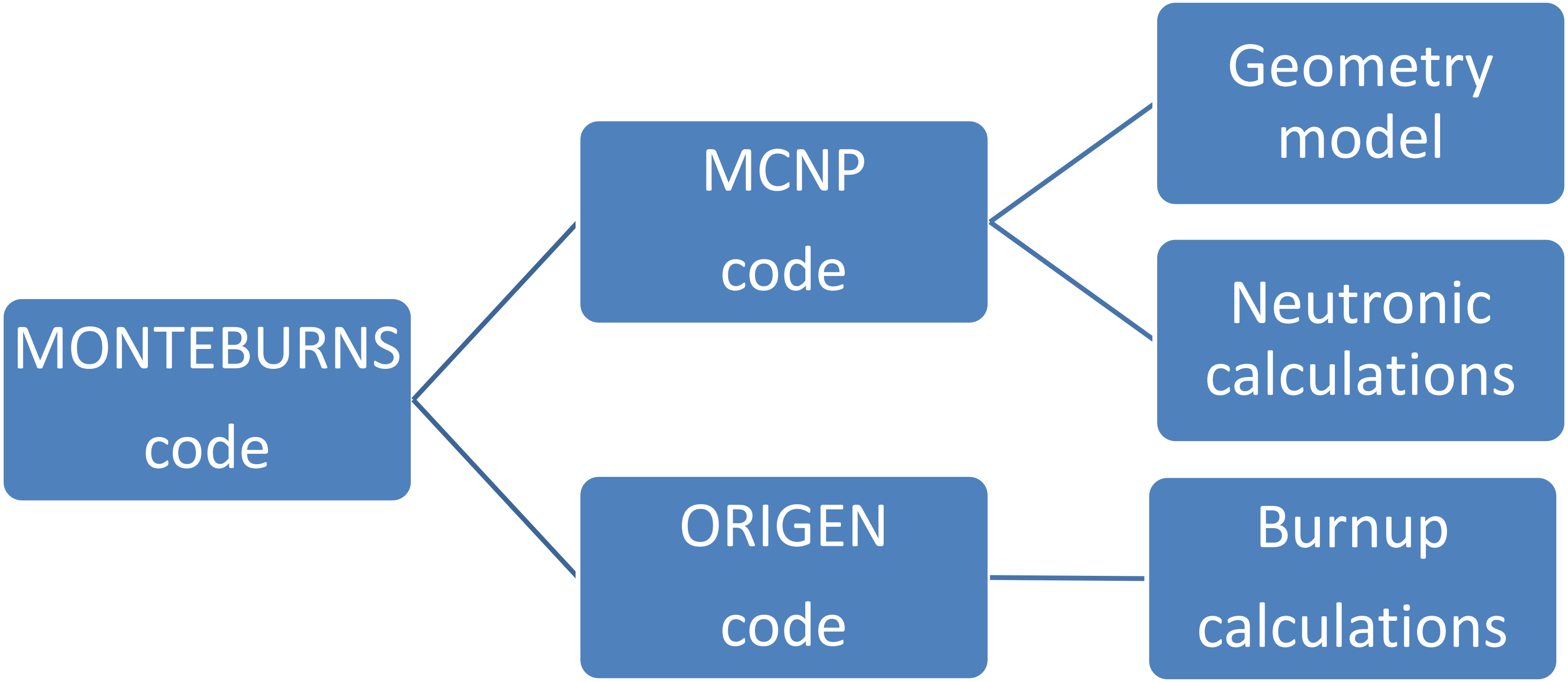
https://www.bartlett.ucl.ac.uk/energy/news/paul-dorfman-nuclear

http://www.glogster.com/jerrodhall/nuclear-waste/g-6maemkvce2okra5ricpvpa0

Goals of this work

1. Study the possibility to induce fuel regeneration and transuranics partioning and transmutation in hybrids systems.
2. Compare the neutronic and depletion behavior under irradiation of the different neutron spectra for fusion and spallation sources.
3. Other studies about ADS involve the influence of the spallation spectrum without the contribution of the fission spectrum

Methodology



MONTEBURNS code

MCNP code

ORIGEN code

Geometry model

Neutronic calculations

Burnup calculations

ADS Geometry

- The accelerator tube has a radius of 1.5 cm.
- The core is a cylinder of 6.2 m^3
- The core is filled with hexagonal lattice formed with 156 fuel rods.
- Fuel rod diameter is 1.3 cm
- The pitch is 12 cm
- Rod length is 200 cm

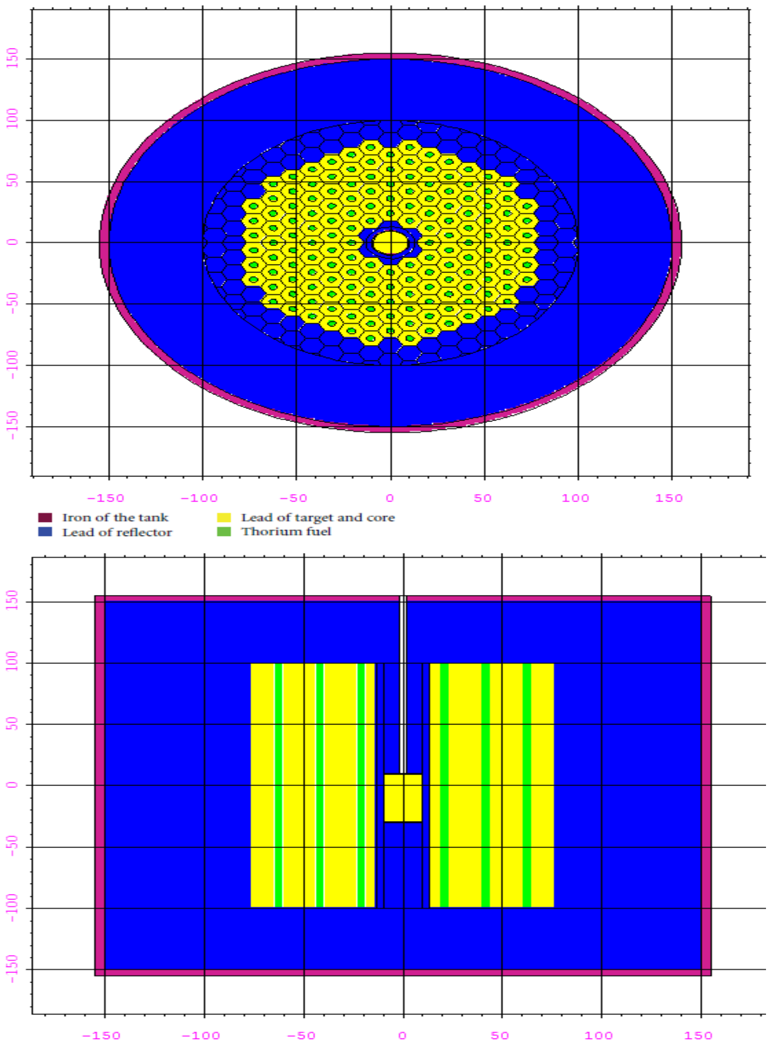


Figure 1. ADS Geometry

The Neutron Spectra

- The evaporation neutron spectrum described where *a* = 1.3 MeV

$$\boldsymbol{p(E) = C\,E\,\mathbf{exp}(-\frac{E}{a})}$$

- The fusion neutron spectrum follows

$$\boldsymbol{p(E) = C\,\mathbf{exp}[-\left(\frac{(E-b)}{\alpha}\right)^{2}]}$$

where “α” is the width in MeV and “b” is the average energy in MeV.

Materials

System structure	Iron
Coolant	Lead
Spallation Target	Lead
Fuel	Ganex diluted with Thorium

Ganex

The reprocessed fuel was obtained from the burned Angra I fuel Type C (33,000MWd/T burned), with 3.1% of initial enrichment left by 5 years in the pool and reprocessed by Ganex process.

Table 1. Recovering percentage from GANEX

Nuclides	Percentagens (%)
Uranium	0.01
Neptunium	99.95
Plutonium	100
Americium	100
Curium	100
Fission products (Nd, Sm, Eu)	5

MIGUIRDITCHIAN, M. et al. Development of the ganex process for the reprocessing of gen iv spent nuclear fuels. In: *Atalante*. Montipellier, França: CEA, 2008.

Ganex Fuel Composition

15.7% Fissile Material & 75.89% Thorium dilution rate

Nuclide	Fraction	Nuclide	Fraction	Nuclide	Fraction
^{232}Th	6.6746E-01	^{239}Np	8.8890E-04	^{242}Cm	4.6855E-04
^{233}U	3.7546E-12	^{238}Pu	3.9895E-03	^{244}Cm	5.3715E-04
^{234}U	3.3322E-07	^{239}Pu	1.0447E-01	^{245}Cm	1.8691E-05
^{235}U	1.7342E-05	^{240}Pu	3.5714E-02	^{143}Nd	2.6515E-03
^{236}U	8.8649E-06	^{241}Pu	3.3580E-02	^{147}Sm	5.3172E-04
^{237}U	1.0634E-08	^{242}Pu	1.2691E-02	^{153}Eu	1.1307E-04
^{238}U	2.1081E-03	^{241}Am	1.5047E-03	O	1.2059E-01
^{237}Np	1.0206E-02	^{242}Am	2.7692E-06		
^{238}Np	1.4078E-05	^{243}Am	2.4269E-03		

Results

- The burnup was performed with
  - Power of 515 MW
  - 10 years burnup
  - Time steps of 45.62days

k_s

- Spallation Source
- Fusion Source

k_{eff}

- Without external source

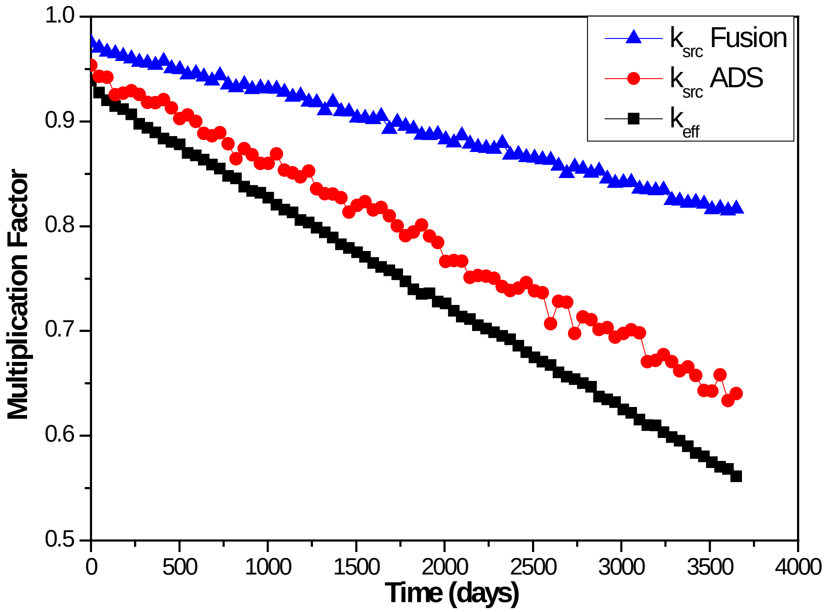


Figure 2.Multiplication Factor

Actinides Mass Transmutation

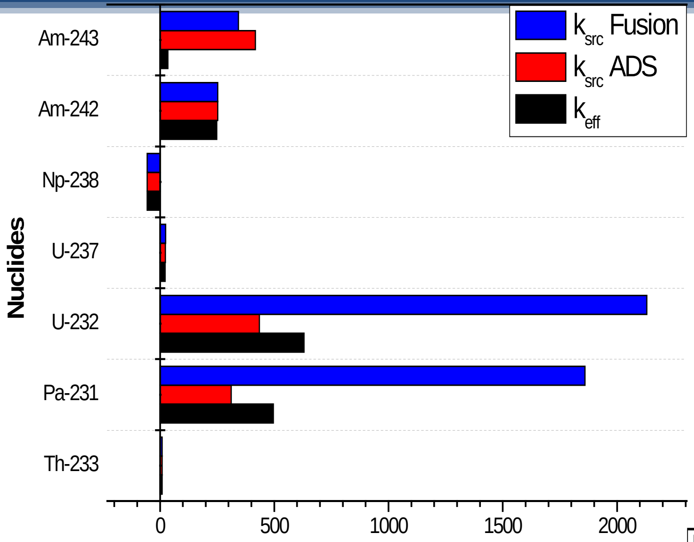


Figure 3. In grams

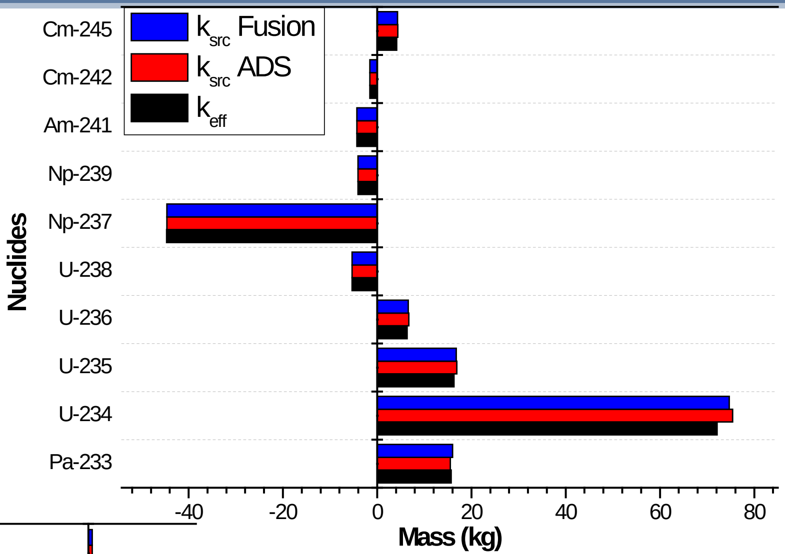


Figure 4. In kilograms

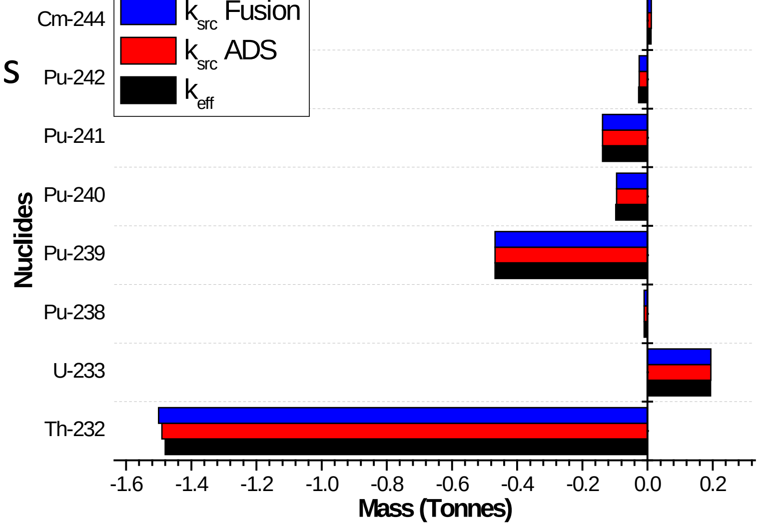


Figure 5. In tonnes

Partial Conclusions

- Subcriticality level was maintained during the 10 year burnup for the different neutron spectra

- It was shown that the use of thorium and reprocessed fuel allows the thorium regeneration ≈ 194 Kg of ^{233}U were produced.

Partial Conclusions

- The radiotoxicity of several actinides were reduced (^{238}U, ^{237}Np, ^{238}Np, ^{239}Np, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am and ^{242}Cm)

- There are two nuclides produced using the fusion with higher amount than using the other neutron source, they are ^{232}U and ^{231}Pa.

Evaluation of Transuranics Transmutation Using Neutrons Spectrum From Spallation Reactions

Objective

- The goal is to examine the behaviour and influences of the hard spallation spectrum in the transmutation without the contribution of the fission spectrum

- To analyze the influence of using different target materials - eutectic mixture of PbBi, Hg and natural U.

- The hard spallation neutron flux is analysed and compared in three conditions;
  (a) Target without moderator or coolant, and with transmutation material – Reference System;
  (b) target immersed in a coolant with transmutation material – Fast System
  (c) target immersed in a moderator with transmutation material – Thermal System.

Methodology

- MCNPX 2.6.0
- Proton beam energy of 2.0 GeV
- 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 model consists of an outer vessel of 330 cm diameter and 300 cm in length.
- The transmutations cells containing the transuranics are placed along the radial axis.

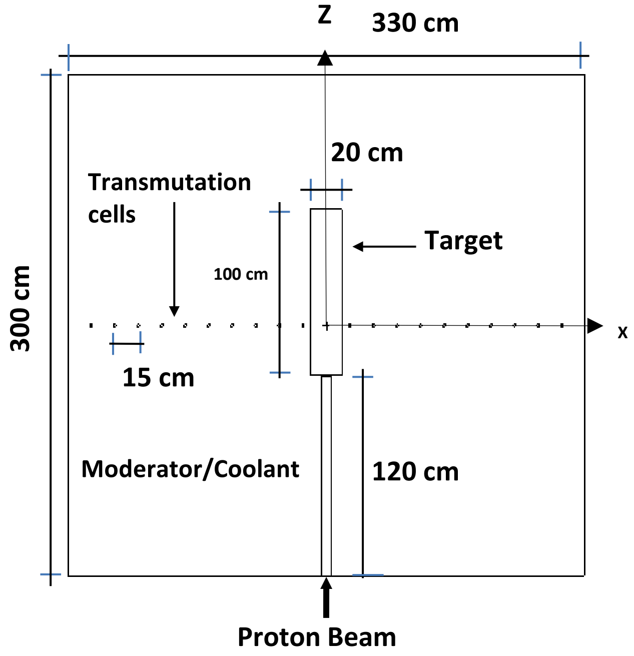


Figure 6 – Axial view of the proposed model

Geometry

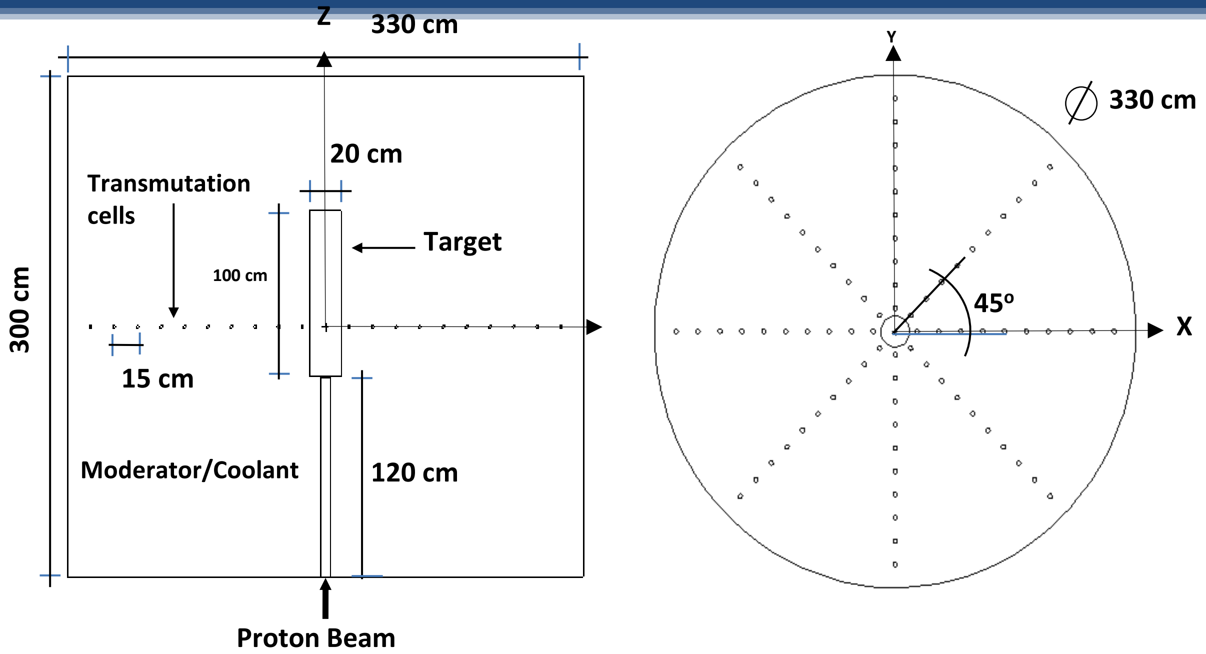


Figure 7 - Radial (right) and axial (left) view of the proposed model, according to the Cartesian plane XZ and XY, respectively.

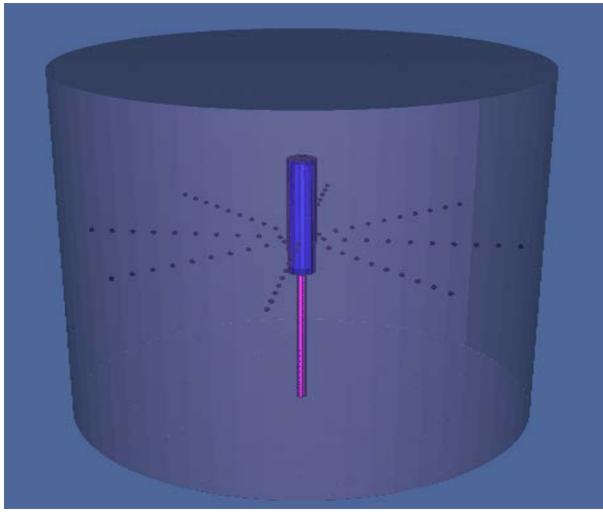


Figure 8 - Arrangement view in three dimensions showing the external vessel, the target (blue), transmutation cells (black) and tube of beam (pink).

Spent Fuel from a PWR

Radionuclide	Mass (kg)	Activity (Ci)
^{234}U	3.14	1.94 X 10^{1}
^{235}U	2.15 X 10^{2}	4.61 X 10^{-1}
^{236}U	1.14 X 10^{2}	7.22
^{237}U	9.15 X 10^{-7}	7.47 X 10^{1}
^{238}U	2.57 X 10^{4}	8.56
Total	2.60 X 10^{4}	α 3.56 X 10^{1}
		β 7.47 X 10^{1}
^{237}Np	2.04 X 10^{1}	1.44 X 10^{1}
^{239}Np	2.05 X 10^{-6}	4.78 X 10^{2}
Total	2.04 X 10^{1}	α 1.44 X 10^{1}
		β 4.78 X 10^{2}
^{236}Pu	2.51 X 10^{-4}	1.34 X 10^{2}
^{238}Pu	5.99	1.01 X 10^{5}
^{239}Pu	1.44 X 10^{2}	8.82 X 10^{3}
^{240}Pu	5.91 X 10^{1}	1.30 X 10^{4}
^{241}Pu	2.77 X 10^{1}	2.81 X 10^{6}
^{242}Pu	9.65	3.76 X 10^{1}
Total	2.46 X 10^{2}	α 1.23 X 10^{5}
		β 2.81 X 10^{6}
^{241}Am	1.32	4.53 X 10^{3}
^{242m}Am	1.19 X 10^{-2}	1.16 X 10^{2}
^{243}Am	2.48	4.77 X 10^{2}
Total	3.81	α 5.01 X 10^{3}
		β 1.16 X 10^{2}
^{242}Cm	1.33 X 10^{-1}	4.40 X 10^{5}
^{243}Cm	1.96 X 10^{-3}	9.03 X 10^{1}
^{244}Cm	9.11 X 10^{-1}	7.38 X 10^{4}
^{245}Cm	5.54 X 10^{-2}	9.79
^{246}Cm	6.23 X 10^{-3}	1.92
Total	1.11	α 5.14 X 10^{5}
Total	2.63 X 10^{4}	α 6.42 X 10^{5}
		β 2.81 X 10^{6}

Evaluation of Transuranics Transmutation Using Neutrons Spectrum From Spallation Reactions

RESULTS

Target without moderator or coolant, and with transmutation material – Reference System

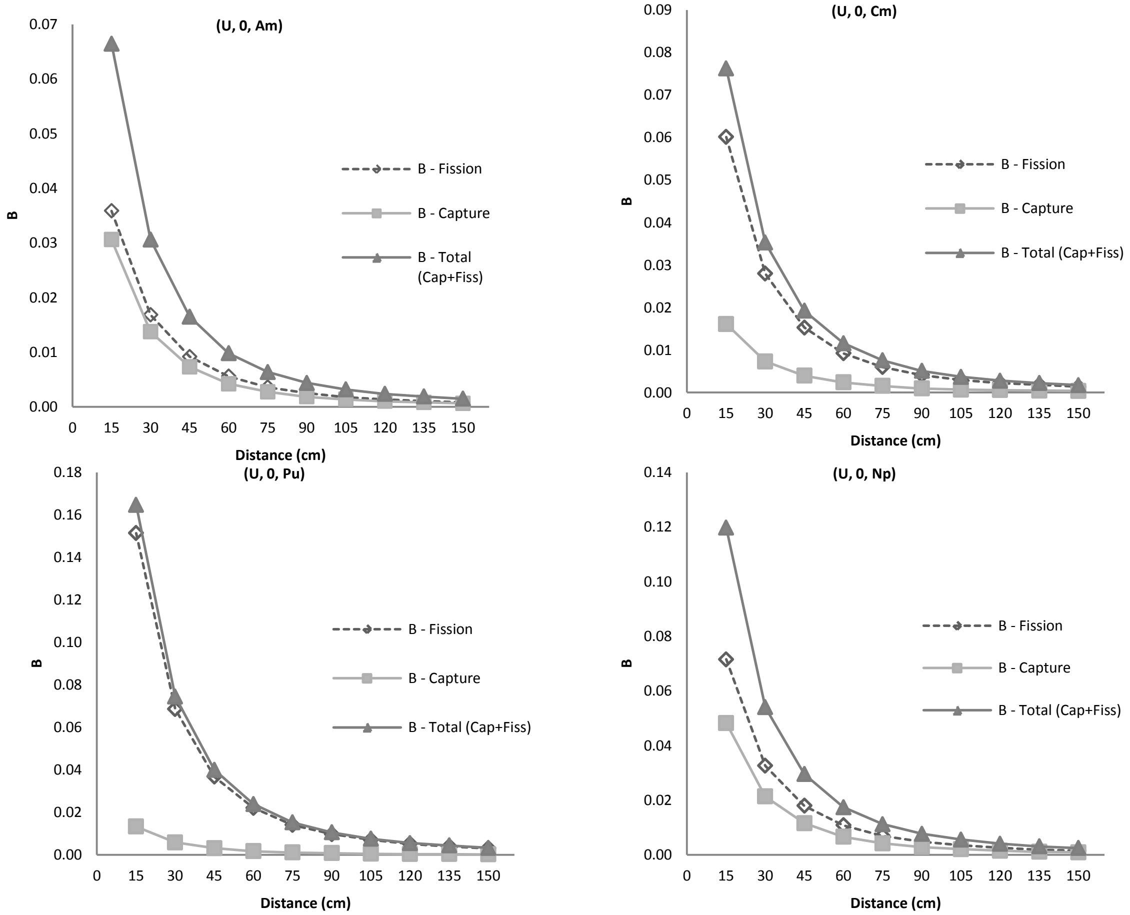


Figure 9 - Relative number of reaction B in $(U, 0, TRU)$ as a function of the distance to the TRU. The proton beam energy of 2.0 GeV and 400,000 particles without moderator / coolant.

Target without moderator or coolant, and with transmutation material – Reference System

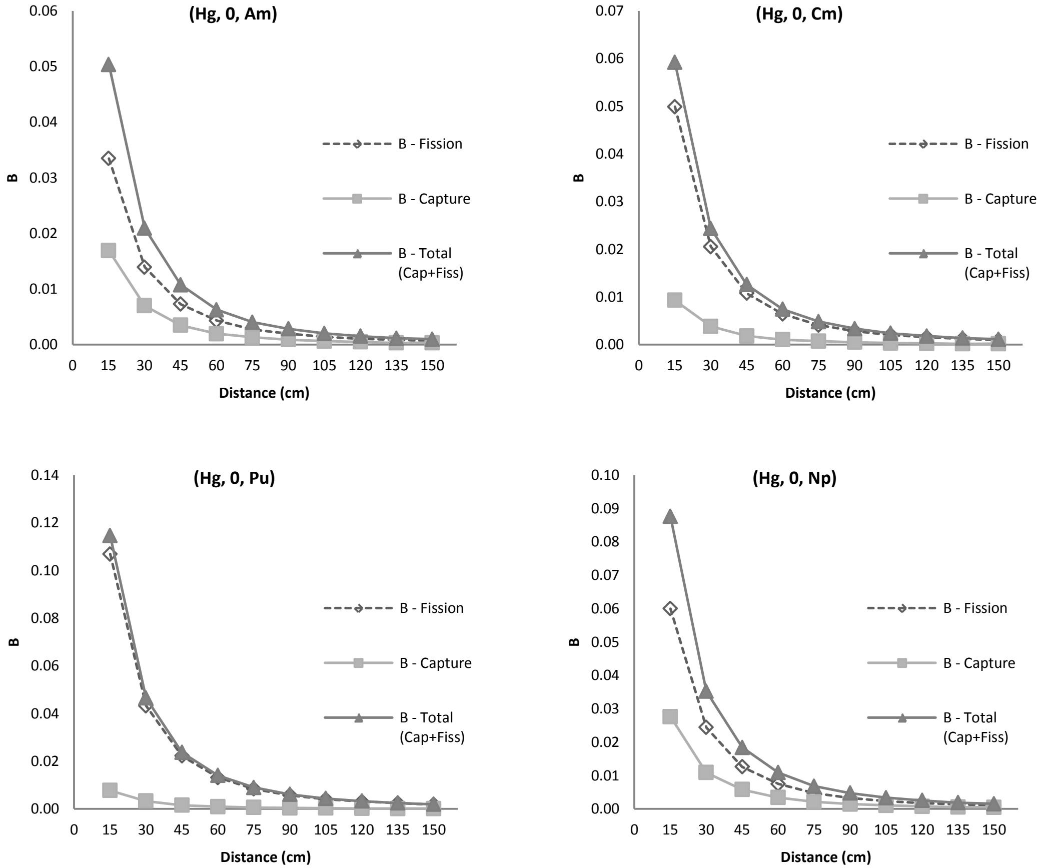


Figure 10 - Relative number of reaction B in $(Hg, 0, TRU)$ as a function of the distance to the TRU. The proton beam energy of 2.0 GeV and 400,000 particles without moderator / coolant.

Target without moderator or coolant, and with transmutation material – Reference System

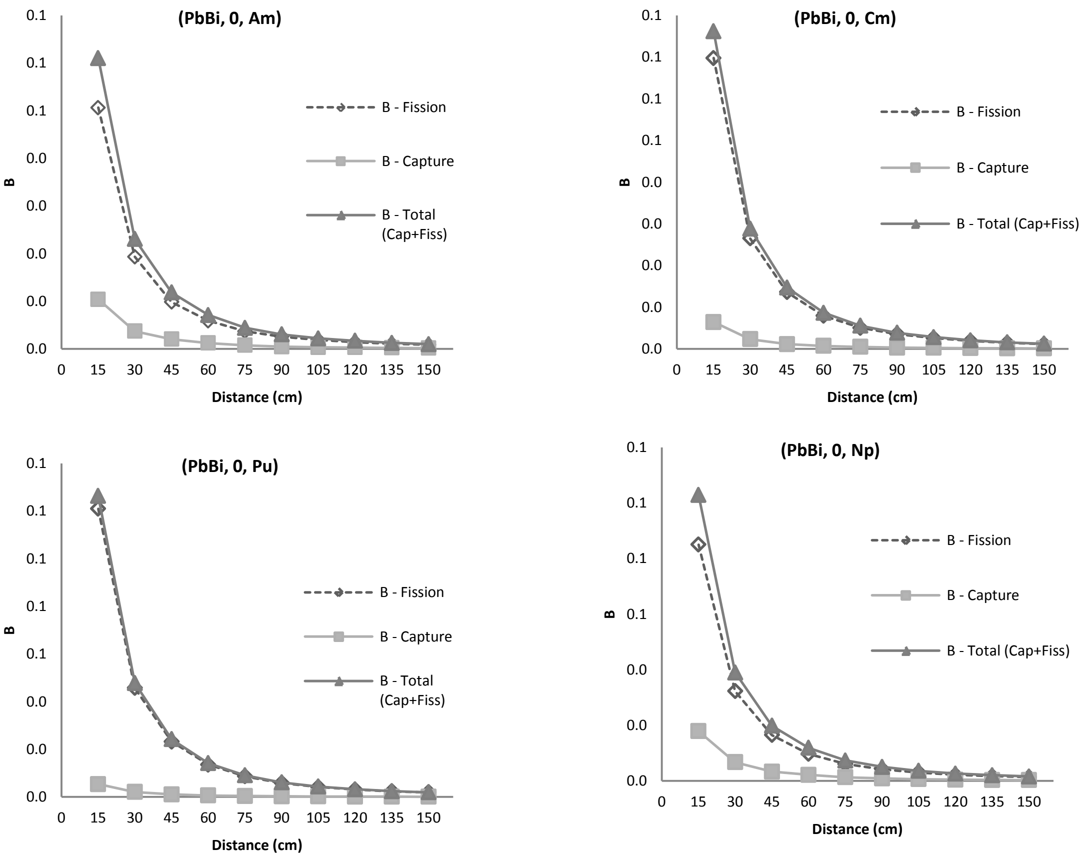


Figure 11 - Relative number of reaction B in $(PbBi, 0, TRU)$ as a function of the distance to the TRU. The proton beam energy of 2.0 GeV and 400,000 particles without moderator / coolant

Analysis of the Previous Results

- The transuranic fission reaction rate (n, f) are higher than the capture reaction rate (n, γ) for all elements.

- The harden spectrum in a spallation target system without moderator favours the fission reactions to all TRU.

target immersed in a coolant with transmutation material – Fast System

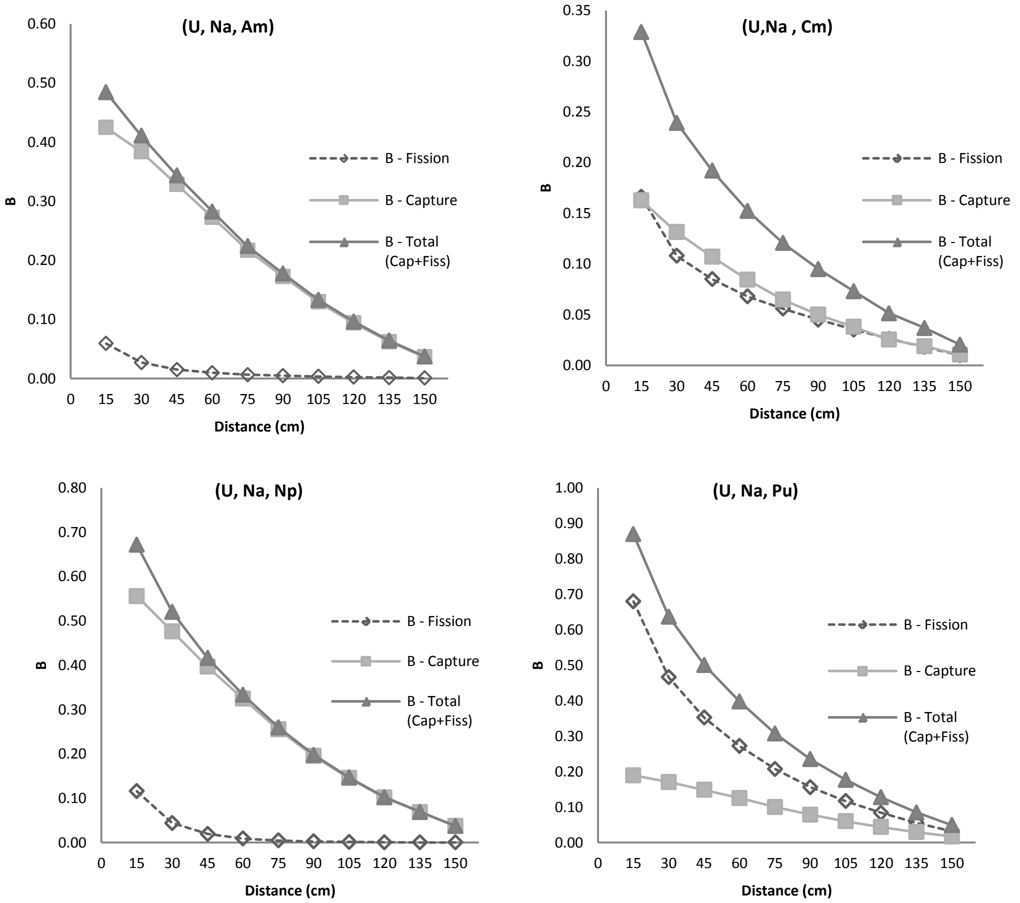


Figure 12 – Relative number of reaction B in (U, Na, TRU) as a function of the distance to the TRU. For proton beam energy of 2.0 GeV and 400,000 particles.

target immersed in a coolant with transmutation material – Fast System

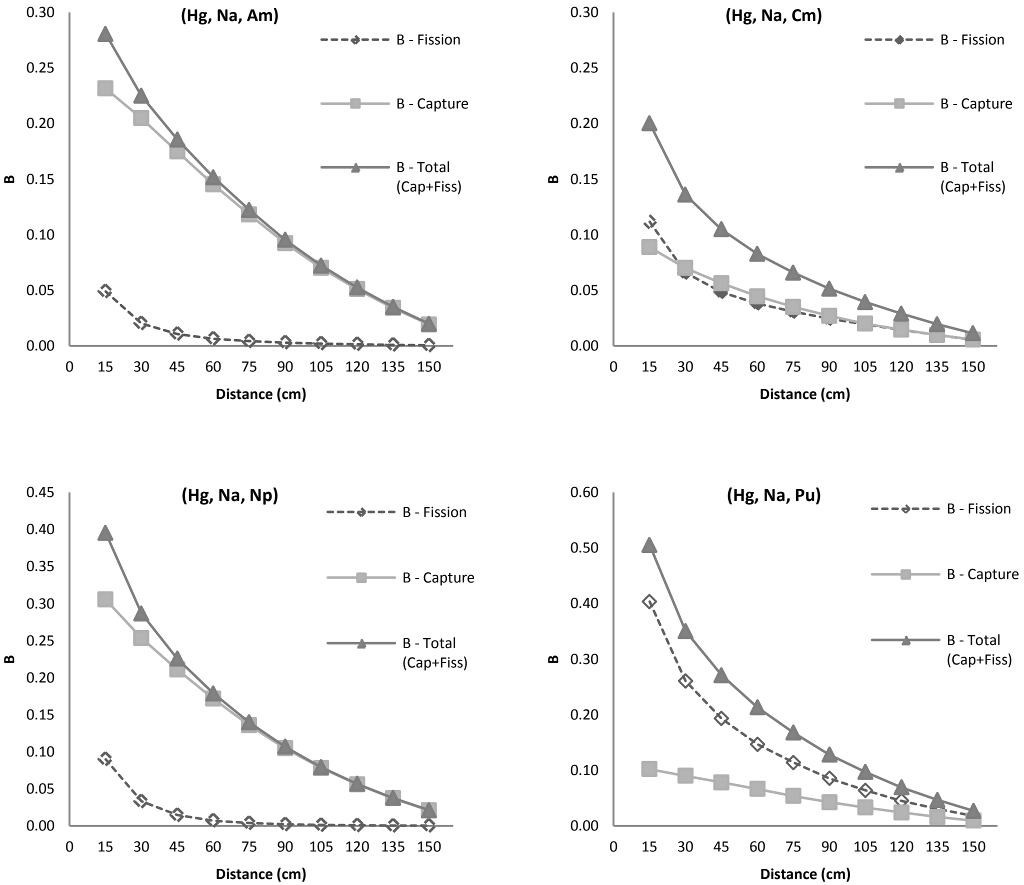


Figure 10 – Relative number of reaction B in (Hg, Na, TRU) as a function of the distance to the TRU. For proton beam energy of 2.0 GeV and 400,000 particles.

target immersed in a coolant with transmutation material – Fast System

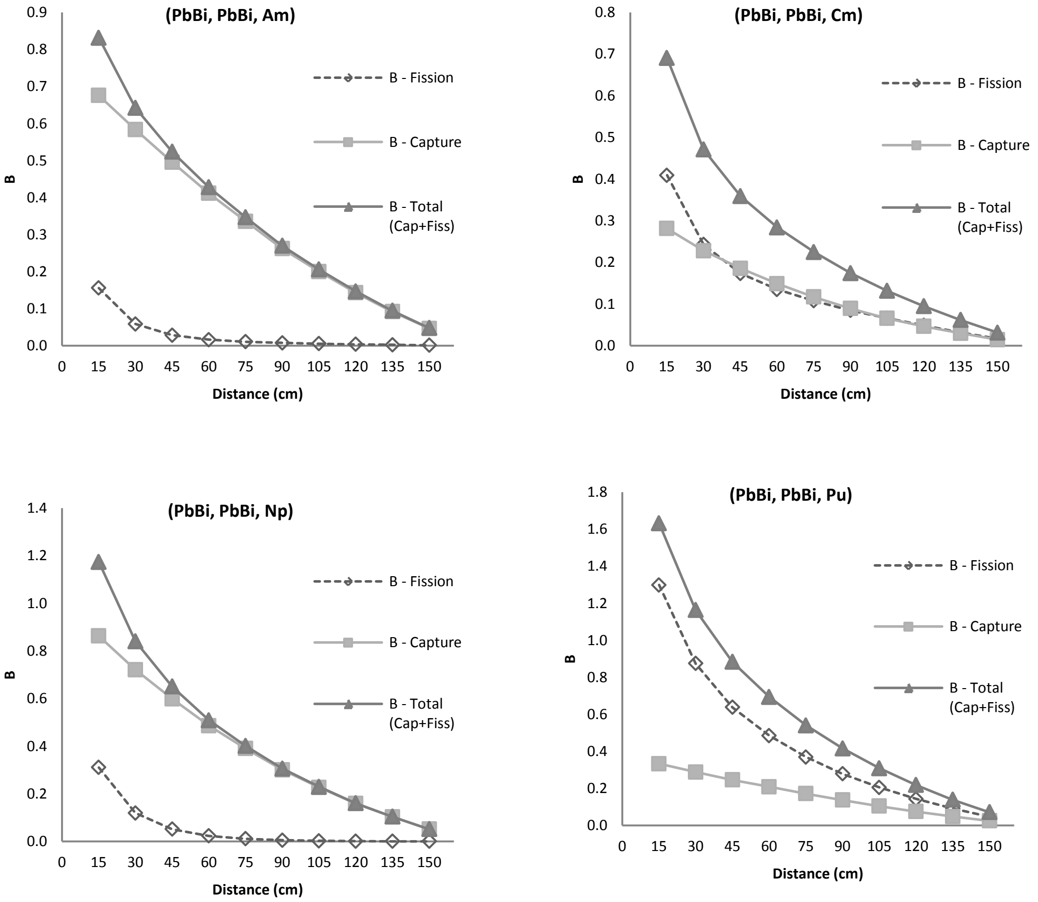


Figure 13 – Relative number of reaction B in $(PbBi, PbBi, TRU)$ as a function of the distance to the TRU. For proton beam energy of 2.0 GeV and 400,000 particles.

Analysis of the Previous Results

- On the systems with Na and PbBi coolant, the capture reaction is superior to fission in Am and Np. Only Pu has the fission as the main mechanism of transmutation.

- For target distances larger than 30 cm, it is not possible to observe any distinction for Cm between the $\boldsymbol{R} = \langle \boldsymbol{B}_{(\boldsymbol{n},\boldsymbol{f})} / \boldsymbol{B}_{(\boldsymbol{n},\boldsymbol{\gamma})} \rangle$

Target immersed in a moderator with transmutation material – Thermal System

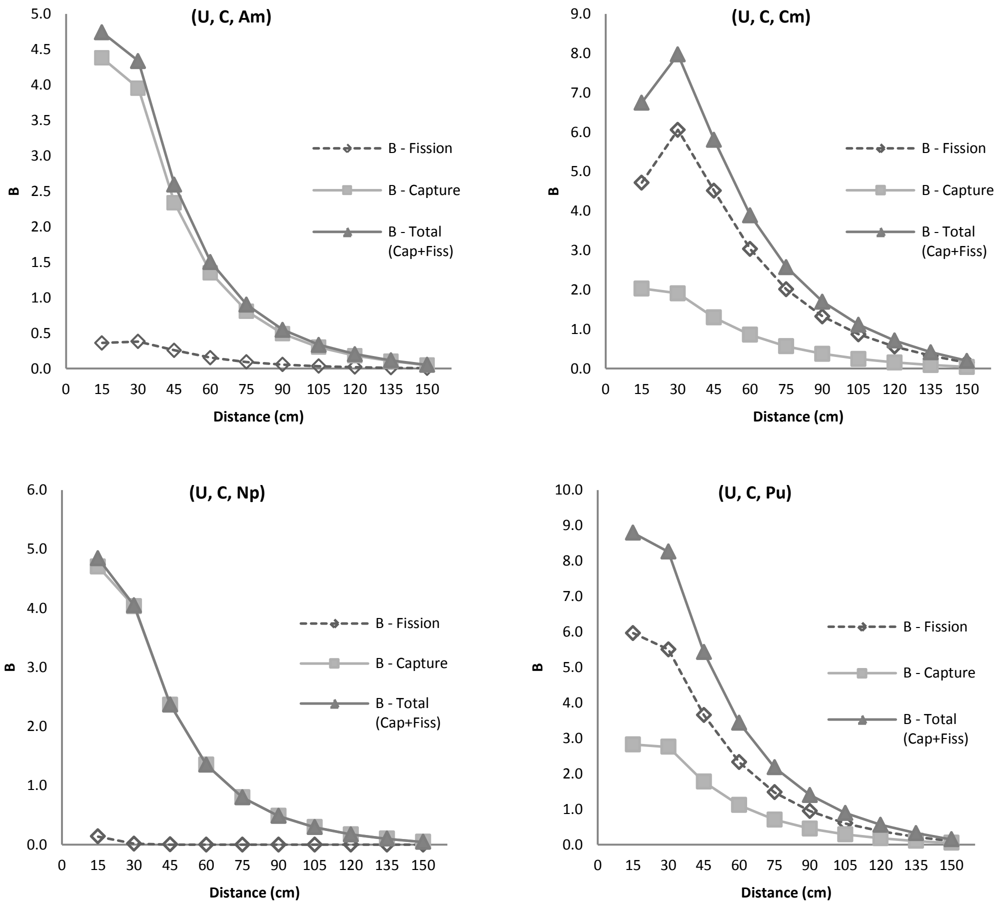


Figure 14 – Relative number of reaction B in (U,C,TRU) as a function of the distance to the TRU. For proton beam energy of 2.0 GeV and 400,000 particles.

Target immersed in a moderator with transmutation material – Thermal System

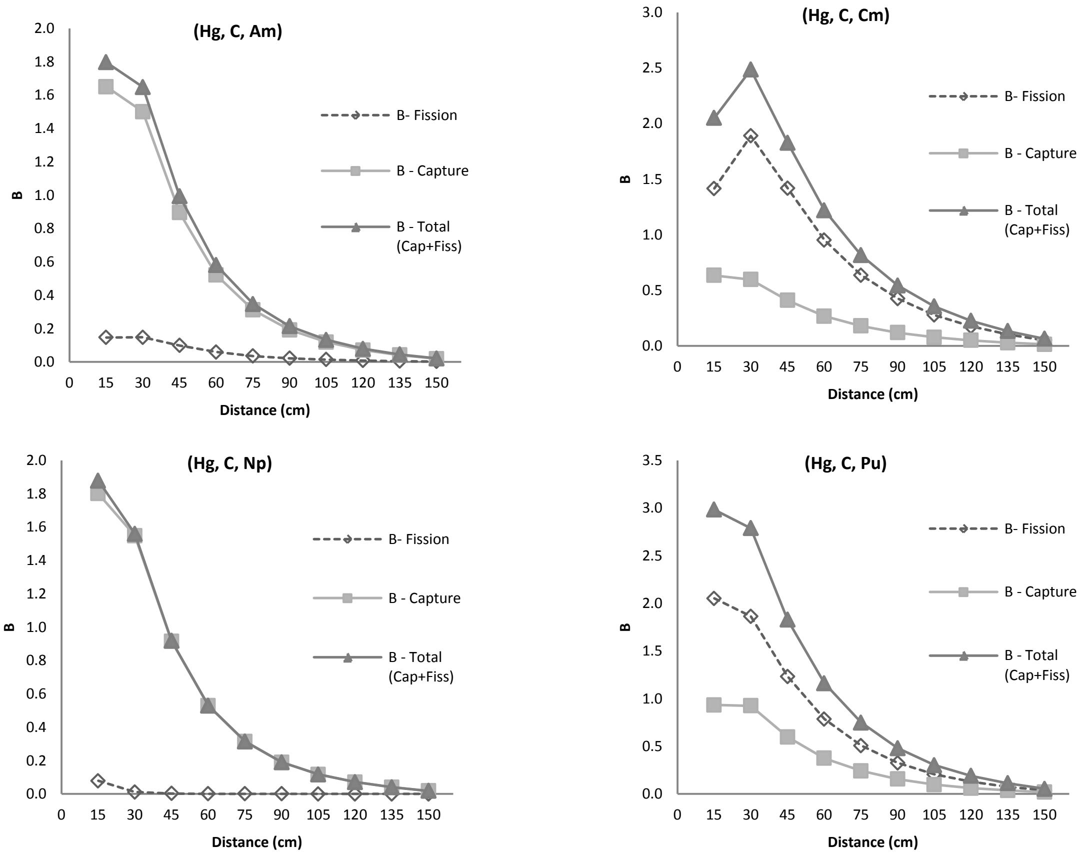


Figure 15 - Relative number of reaction B in (Hg, C, TRU) as a function of the distance to the TRU. For proton beam energy of 2.0 GeV and 400,000 particles.

Target immersed in a moderator with transmutation material – Thermal System

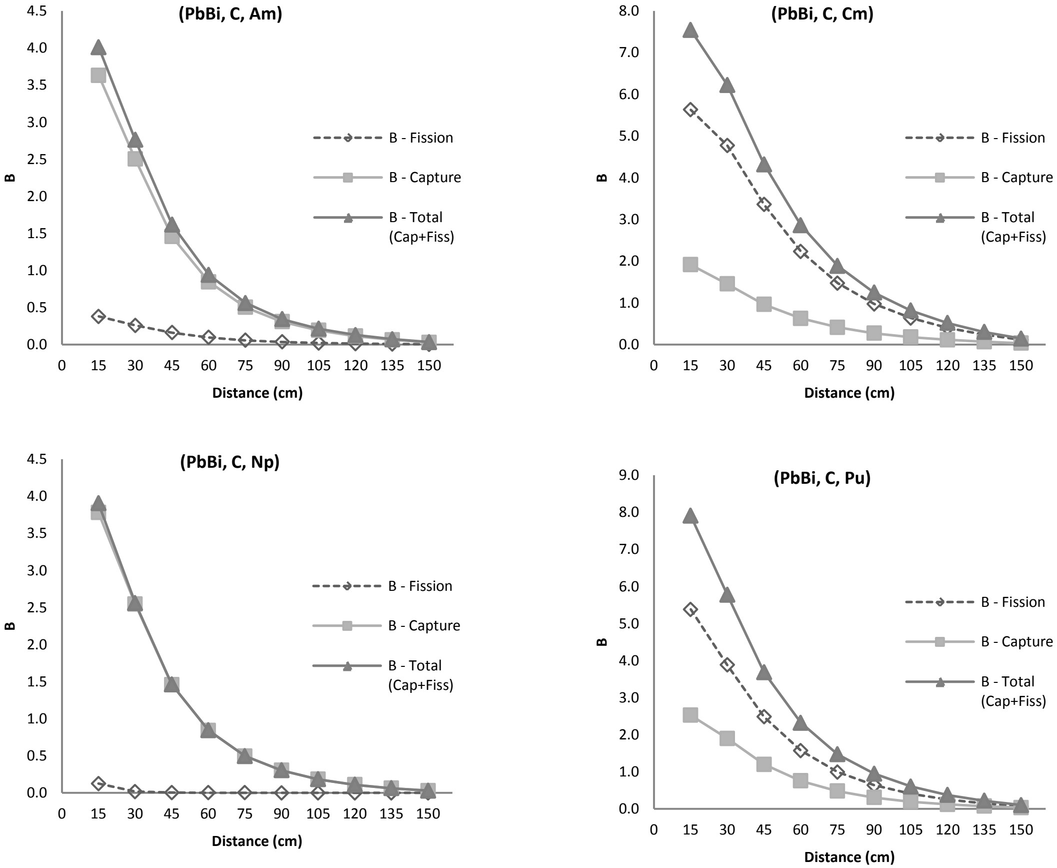


Figure 16 - Relative number of reaction B in $(PbBi, C, TRU)$ as a function of the distance to the TRU. For proton beam energy of 2.0 GeV and 400,000 particles.

Partial Conclusions

- the main mechanism of transmutation in environments with moderator/coolant, for Am and Np are capture reactions.

- In turn, the Cm transmutation is favourable using graphite-moderated on the system. Being the fission the main mechanism for transmutation.

Acknowledgments

cnen
Comissão Nacional
de Energia Nuclear

CAPES

CNPq
Conselho Nacional de Desenvolvimento
Científico e Tecnológico

FAPEMIG

References

M. Gilberti, C. Pereira, and M.A. F. Veloso, "Evaluation of the Transmutation of Transuranic using Neutrons Spectrum from the Spallation Reaction," in *International Nuclear Atlantic Conference*, Recife, 2013, pp. 1-5.

G. P.Barros, C. Pereira, F.Veloso, M.A., and A. L.Costa, "Neutron Production Evaluation from a ADS target utilizing the MCNPX 2.6.0," *Brazilian Journal of Physics*, pp. 414-418, 2010.

G. P.Barros, C. Pereira, M.A. F.Veloso, and A. L.Costa, "Study of an ADS Loaded with Thorium and Reprocessed Fuel," *Science and Technology of Nuclear Installations*, vol. 2012, pp. 1-12, 2012.

C.E. Velasquez, C. Pereira, M.A. F. Veloso, and A. L.Costa, "Thickness Evaluation Layer for Transmutation of Minor Actinides and Pu in a Fusion-Fission System," in *International Nuclear Atlantic Conference*, Recife, 2013.

S. Cota and C. Pereira, "Neutronic Evaluation of the Non-proliferating Reprocessing Nuclear Fues in Pressurized Water Reactors," *Annals of Nuclear Energy*, vol. 24, no. 10, pp. 829-834, 1997.

D. Warin, "Future Nuclear Fuel Cycles: Prospect and Challenges for Actinide Recycling," *Materials Science and Engineering*, vol. 9, pp. 1-6, 2010.

American Nuclear Society. (2006, November) The Use of Thorium as Nuclear Fuel. [Online]. http://www.ans.org/pi/ps/docs/ps78.pdf

C. Sanzo, M. Abdou, and M. Youssef, "Transuranic Transmutation Efficiency of a Small Fusion-Fission Facility for Spent Uranium-oxide and Inert Matrix Fuels," *Fusion Engineering and Design*, vol. 85, pp. 1488-1491, 2010.

D. Poston and H. Trellue, "User´s Manual, Version 2.0 for Monteburns Version 1.0," Los Alamos National Laboratory, LA-UR-99-4999, 1999.

A. Croff, "A User´s manual for the Origen 2 computer code," Oak Ridge National Laboratory, Oak Ridge, ORNL/TM-7175, 1980.

X-5 Monte Carlo Team, "MCNP - A General Monte Carlo N-Particle Transport Code," Los Alamos National Laboratory, California, 2003.

OpenCourseWare MIT. (2009) ORIGEN. [Online]. http://ocw.mit.edu/courses/nuclear-engineering/22-251-systems-analysis-of-the-nuclear-fuel-cycle-fall-2009/labs/MIT22_251F09_ORIGEN.pdf

S. Degweker, B. Ghosh, A. Bajpai, and S. Paranjape, "The physics of Accelerator Driven Sub-Critical Reactors," *PRAMANA-Journal of Physics*, vol. 68, pp. 161-171, 2007.