INVESTIGATION OF INCINERATION RATE OF ^{241}AM, ^{243}AM, AND ^{237}NP SPALLATION TARGETS USING A PROTON ACCELERATOR DRIVEN SYSTEM USING MCNPX CODE

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Abstract

Whereas high level nuclear wastes consist of long half-life alpha emitter transuranic elements retain their radio-toxicity over natural level values up 10^6 year at disposal areas, their depleting via transmutation process using critical reactors or accelerator driven systems is widely considered recently. Before high cost trials, computational codes can provide acceptable information and feasibility study on nuclear waste neutronic behavior in critical reactors or accelerator driven systems. Hence, MCNPX 2.6.0 code has been used in this work to evaluate incineration possibility of ^{241}Am, ^{243}Am and ^{237}Np using proton accelerators via spallation process induction through these targets. Neutron yield leaked from the spallation targets, power density, neutron flux leaked from the spallation targets, residual nuclei production inside the investigated targets and some reaction rates has been calculated for the modeled targets. The neutronic calculations of spallation process have been carried out using different proton energies of 0.4, 0.6, 0.8 and 1 GeV protons and 1 mA beam current. According to the computational data, ^{237}Np produces the most leaked neutron yield using all the investigated proton energies.^{237}Np will be transmuted as 6.36kg/y via (n, γ) and fission reactions. It suffers the most heat deposition (36.9 kW/cm^3) and produces the most fission rates (61.2888 #/source particle). The transmutation rate value is 3.32 kg/y for ^{241}Am and 1.73 kg/y for ^{243}Am.

Keywords: Transuranic transmutation, MCNPX 2.6.0simulation, Accelerator driven system

Introduction

Recently depleting minor actinide waste obtained in a nuclear reactor spent fuel has been evaluated by critical reactors under one-cycle or multiple-cycle strategies. Sahin et al. (2008) have been theoretically investigated load of a thorium fuel mixture contained some fractions of transuranic (TRU) elements in CANDU reactor. As their results shows, the modeled core can depletes 100 kg/y of the loaded wastes [1]. Romanello et al. (2008) have been compared TRU burning ability in accelerator driven and critical systems. According to their results, the fast investigated reactors need to a 30-40% power more than accelerator driven systems for burning of TRUs [2]. Perko et al. (2012) have been studied burn up of a TRU-contained fuel in two VVER and PWR modeled reactors. As their theoretical calculations shows, the modeled VVER and PWR cores can deplete TRU fraction of the loaded fuel with a rate of 73 kg/y and 88 kg/y respectively [3]. According to the carried out study by Ikeda et al. (2011), burn up of TRU in an ARR modeled reactor achieves 34 kg/TW$_{th}$h americium depletion. They used an americium nitrate (AmN) blanket in the ARR modeled core [4].

Hence, Investigation of TRU nuclear waste element transmutation via induced-spallation reaction by means of a proton accelerator has been proposed in this work.

Material and methods

MCNPX 2.6.0 code has been used to model spallation process in different cylindrical targets (10×60 cm)of ^{241}Am, ^{243}Am and ^{237}Np irradiated by proton beams of 0.4, 0.6, 0.8 and 1 GeV and beam current of 1 mA (2-mm spatial FWHM), respectively. Leaked neutron yield from the spallation targets, fission and (n, γ) reaction rates inside the spallation targets and power density deposition have been calculated for any modeled spallation target using the different energies separately. Residual mass distribution inside the spallation targets and leaked neutron spectra has been calculated using the computational code for proton energy of 1 GeV and 1 mA current.F1 tally has been used to obtain neutron spectra leaked from the spallation targets. F6 tally has been used to calculate power density inside the modeled targets. Histp card has been used to calculate radionuclide production in any spallation targets [5]. Residual masses have been transferred to gram (g) scale by multiplying the N^{HTAPE} data with A (mass number) × 1.036402E−08 (g) [6]. Large history of particles has been used to reduce the calculation errors to less than 0.5%. INCL4/ABLA model has been used for the spallation neutronic calculations. KCODE mode has been used to calculate effective multiplication, delayed neutron fraction as well as effective delayed neutron fraction of the spallation targets. Burn card has been used to evaluate time behavior of effective multiplication during 1 year in case of any modeled spallation target.

Results and discussion

According to the obtained data, ^{243}Am provides the least neutron leakage (67.3 n/p at 1 GeV) as a result of spallation process induction using different proton beam energies. ^{237}Np conclude in the most leaked neutron yield (133 n/p at 1 GeV). Using low energies, there is not much discrepancy for the leaked neutron yield from the investigated spallation targets (Fig.1).

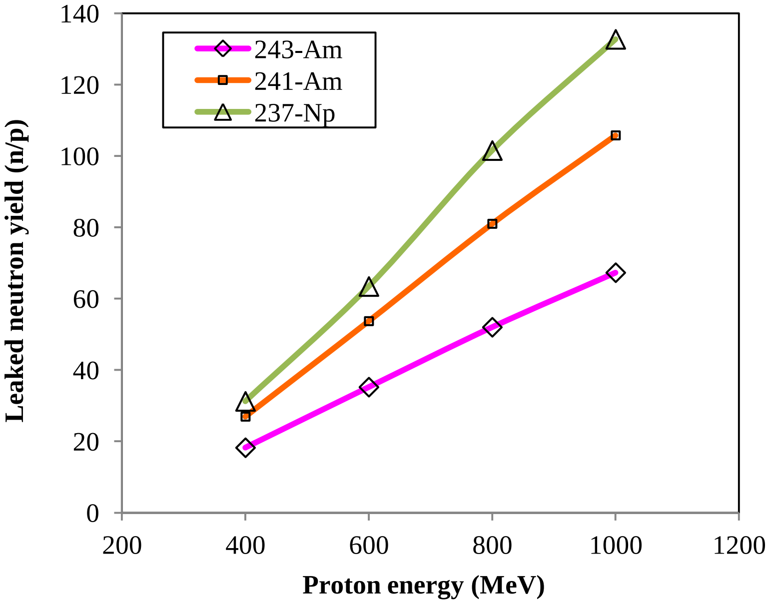

Figure 1. Comparison of the leaked neutron yield on different proton beam energy for the different spallation target.

Computational data showed maximum (n,γ) reaction rate will be occurred for ^{237}Np (19.7 #/Source particle at 1 GeV). The minimum reaction rate belongs to ^{243}Am target in all of energy span (Fig.2).

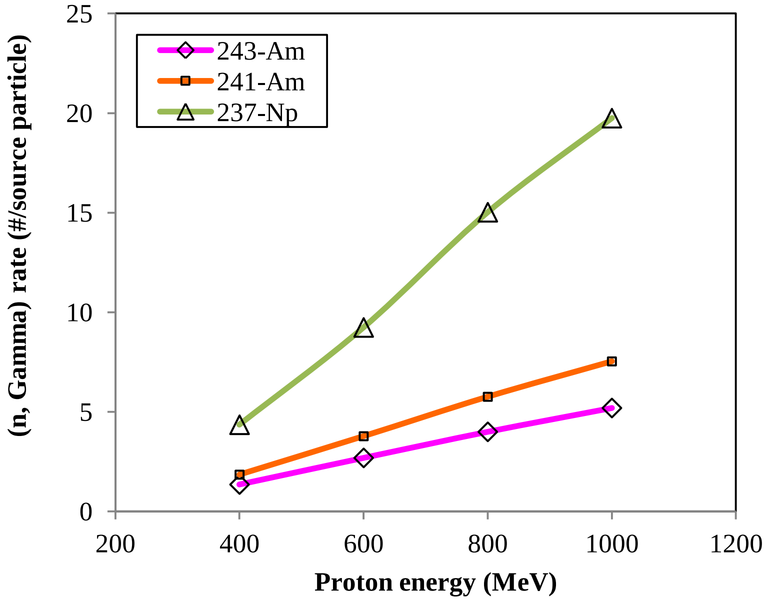

Figure 2. Comparison of (n,γ) reaction rate on different proton beam energy for the different spallation target.

Power density deposited in the investigated spallation targets has been calculated using different projectile energies separately. The calculations showed ^{237}Np experience the highest power density than the other investigated targets at whole the studied proton energies.

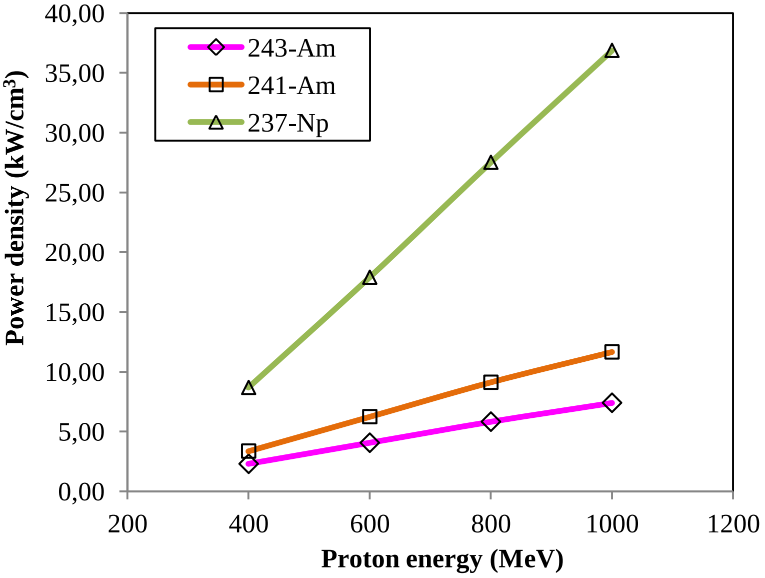

Figure 3. Comparison of power density on different proton beam energy for the different spallation target.

According to the computational data, ^{237}Np concludes in the highest leaked neutron spectra than $^{241/243}$Am targets (Fig.4). A neutron current of in order of 10^{17} n/s is provided using ^{237}Np spallation target using 1 mA current of 1 GeV proton beam.

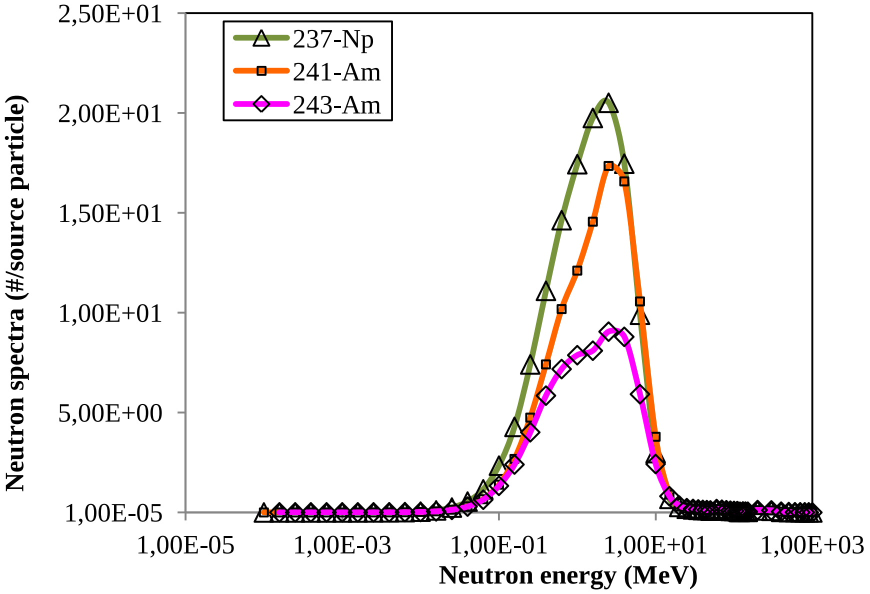

Figure 4. Comparison of neutron spectra leaked the spallation targets, proton energy: 1GeV.

As the presented data at Table 1 shows there are only less than 3% neutrons with an energy of E_n>20 MeV. About 50% of the leaked neutrons are fast and about 45% are epithermal.

Table 1-Energy range of neutron spectra leaked from the spallation targets.

Neutronenergy	^{237}Np (%)	^{241}Am (%)	^{243}Am (%)
1e-04 MeV<En<1 MeV	.47E+01	.76E+01	.43E+01
1 MeV<En<20 MeV	.39E+01	.06E+01	.27E+01
20 MeV<En<1000 MeV	.33E+00	.88E+00	.98E+00

Residual nuclei calculations showed ^{237}Np encounter with the highest values than $^{241/243}$Am targets after 1 GeV proton beam injection inside the targets. The proton beam current was assumed to be 1 mA for all the calculations. However the long half-life alpha emitter isotopes are producing during the spallation target irradiation, but their depletion rate via (n,γ) and fission reaction is faster (Fig.5).

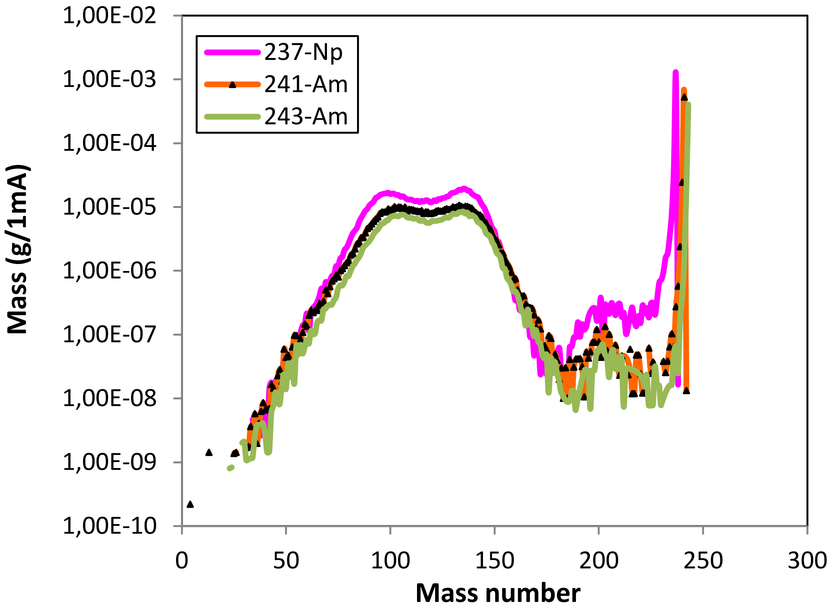

Figure 5. Comparison of residual nuclei production inside the spallation targets, proton energy: 1 GeV.

For instance, about 0.31kg/y of ^{239}Pu is produced inside the ^{243}Am spallation target using 1 GeV proton beam of 1 mA current. Also about 0.01 kg/y ^{237}Np will be produced in the investigated target. Hence, the long half-life alpha emitter isotope production rate is ignorable inside the spallation targets in comparison with their incineration rates.
According to the obtained data, the most neutron delayed fraction belongs to ^{241}Am target (Table 2).

Table 2- Comparison of different physical and neutronic properties of spallation targets.

Target specifications	^{237}Np	^{241}Am	^{243}Am
Density$(\frac{g}{cm^3})$	20.25	13.67	13.67
Meltingpoint(°C)	644	1176	1176
Boilingpoint(°C)	4000	2011	2011
Termal conductivity$(\frac{W}{mK})$	6	10	10
β (pcm)	194	214	190
β_{eff}(pcm)	75	231	182
Neutron per fission(ν)	3.00	3.67	3.87
k_{eff}	0.86213	0.76451	0.64974

Comparison of absorption (absorption via all the reactions moreover (n,γ)) reaction and (n,γ) reaction rates showed total absorption reaction belongs to (n,γ) reaction and the other absorption reactions such as (n, p), (n,d), (n,t) … have not noticeable weight.
Using 1 mA current of 1 GeV, ^{237}Np will be transmitted as 6.36 kg/y via this reaction. The value is 3.32 kg/y for ^{241}Am and 1.73 kg/y for ^{243}Am. The highest (n,γ) and fission rates belong to ^{237}Np while gas production rate inside the spallation targets are approximately close to each other (Table 3).

Table 3- Comparison of different neutronic properties of spallation targets, Proton energy: 1 GeV, Beam current: 1 mA

Quantity	Energy (MeV)			
	400	600	800	1000
^{237}Np				
(n,G) (#/s)	2.72E+16	5.77E+16	9.39E+16	1.23E+17
(n+p,f) (#/s)	8.74E+16	1.82E+17	2.97E+17	3.89E+17
Transmutation rate (kg/y)	1.423651	2.980534	4.850051	6.362937
Gas production (atom/y)	2.40E+08	3.47E+08	4.92E+08	6.48E+08
^{241}Am				
(n,G) (#/s)	1.15E+16	2.35E+16	3.59E+16	4.71E+16
(n+p,f) (#/s)	5.35E+16	1.08E+17	1.65E+17	2.16E+17
Transmutation rate (kg/y)	0.820617	1.661986	2.536969	3.326655
Gas production (atom/y)	2.39E+08	3.35E+08	4.59E+08	6.01E+08
^{243}Am				
(n,G) (#/s)	8.41E+15	1.68E+16	2.49E+16	3.25E+16
(n+p,f) (#/s)	2.73E+16	5.41E+16	8.09E+16	1.05E+17
Transmutation rate (kg/y)	0.451024	0.894867	1.335081	1.736473
Gas production (atom/y)	2.31E+08	3.24E+08	4.43E+08	5.77E+08

As it is seen in Fig.6, ^{237}Np, $^{241/243}$Am effective multiplication factors are approximately constant during 1-year burn up.

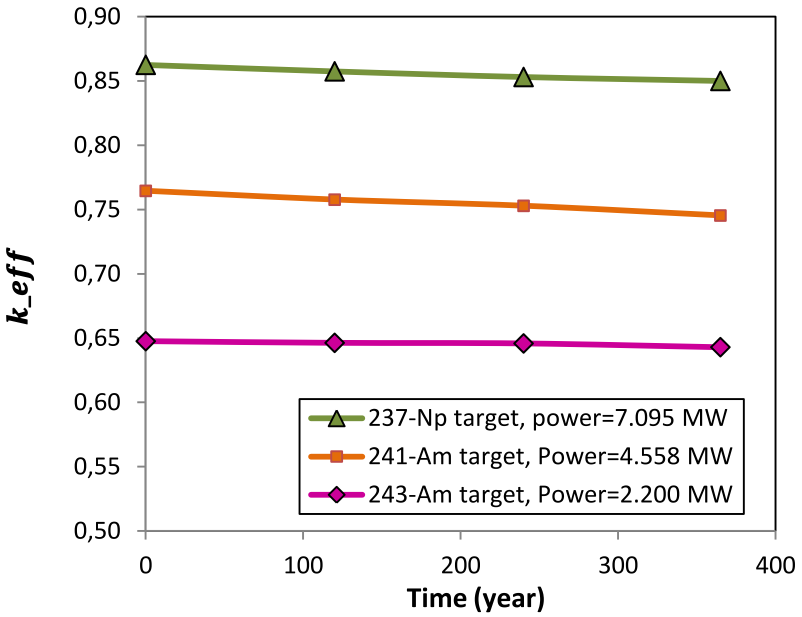

Figure 6. Comparison of effective multiplication factor of the spallation targets during transmutation time.

Conclusion

Many neutrons can be produced in spallation nuclear reactions induced in heavy targets which creates an intense neutrons flux for research, medical and industrial domains, nuclear waste transmutation as well as a pulsed neutron source for subcritical reactors. The carried out study showed utilization of ^{237}Np, ^{241}Am and ^{243}Am as the heavy spallation target not only produces high neutron currents in order of 10^{17} n/s but (n,γ) and fission reaction rates inside them provides depletion rate of these nuclear wastes during the spallation process.

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