Monte Carlo Studies in Accelerator-Driven Systems for Transmutation of High–Level Nuclear Waste

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Abstract

A spallation neutron source was modeled using a high energy proton accelerator. The aim is to maximize the minor actinides and fission products transmutation rates, which is created from the operation of nuclear power reactors for the production of electricity.

The transmutation system is composed of a natural lead target, beam window, subcritical core, reflector, and structural material. The neutrons are produced by the spallation reaction of protons from a high intensity linear accelerator in the spallation target, and the fission reaction in the core. It is used a hexagonal lattice for the waste and fuel assemblies. The system is driven by a 1 GeV, 10 mA proton beam incident on a natural lead cylindrical target. The protons were uniformly distributed across the beam. The core is a cylindrical assembly. The main vessel is surrounded by a reflector made of graphite. The axes of the proton beam and the target are concentric with the main vessel axis. The structural walls and the beam window are made of the same material, stainless steel, HT9. We investigated the following neutronics parameters: spallation neutron and proton yields, spatial and energy distribution of the spallation neutrons, and protons, heat deposition, and the production rates of hydrogen and helium, transmutation rate of minor actinides and fission products.

In the calculations, the Monte Carlo code MCNPX, which is a combination of LAHET and MCNP, was used. To transport a wide variety of particles, The Los Alamos High Energy Transport Code (LAHET) was used.

1. Introduction

For the last fifty 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 comes from transuranic wastes and minor actinides. Every year a light water moderated reactor (LWR), 1GWh (e), discharges about 21 tons radioactive fuel with the following inventory: 20 tons of uranium containing 0.9 % (180 kg) U-235; 200 kg of plutonium (which only 63 % is fissionable), 20 tons of uranium containing 0.9 % (180 kg) U-235; 200 kg of plutonium (which only 63 % is fissionable; 21 kg of minor actinides: 10 kg of neptunium, 10 kg americium, 1 kg curium: 760 kg fission products 18 kg of technetium-99, 16 kg of zirconium-93, 9 kg of cesium-135, 5 kg of palladium107, and 3 kg of iodine 12, which are long-lived elements [1].

Transmutation of minor actinides and long-lived fission products is a promising concept to reduce the radioactive waste and its long-term radiotoxicity. The transmutation technology to incinerate the long-lived radioactive isotopes using an accelerator driven subcritical reactor is one of the best solutions.

2. The system

Neutrons are produced by the spallation reaction of protons from a high intensity linear accelerator in the spallation target and the fission reaction in the core. It is used a hexagonal lattice for the waste and fuel assemblies. The system (Fig.1.) is driven by a 1 GeV, 10 mA proton beam incident on a natural lead cylindrical target, 20 cm radius, 70 cm height entering the target through a 5.3 cm radius hole. The protons were uniformly distributed across the beam of radius 2 cm. The core is cylindrical assembly, 2.3 m radius, 4.6 m high. The wall thickness of the main vessel is 2 cm. The main vessel is surrounded by a reflector made of graphite, 40 cm thick. The axes of proton beam and the target are concentric with the main vessel axis. The structural walls and beam window are made of the same material, stainless steel, HT9.

The vertical proton beam pipe is inserted from the top into the main vessel down to the spallation target region. The hemispherical bottom end of the tube forms the beam window.

The major reasons to choose sodium as coolant are its excellent thermal properties and good compatibility with stainless steel. Most breeder reactors use a hexagonal lattice for the fuel structure, to be consistent with breeder reactor designs, we used a hexagonal lattice for the fuel assemblies.

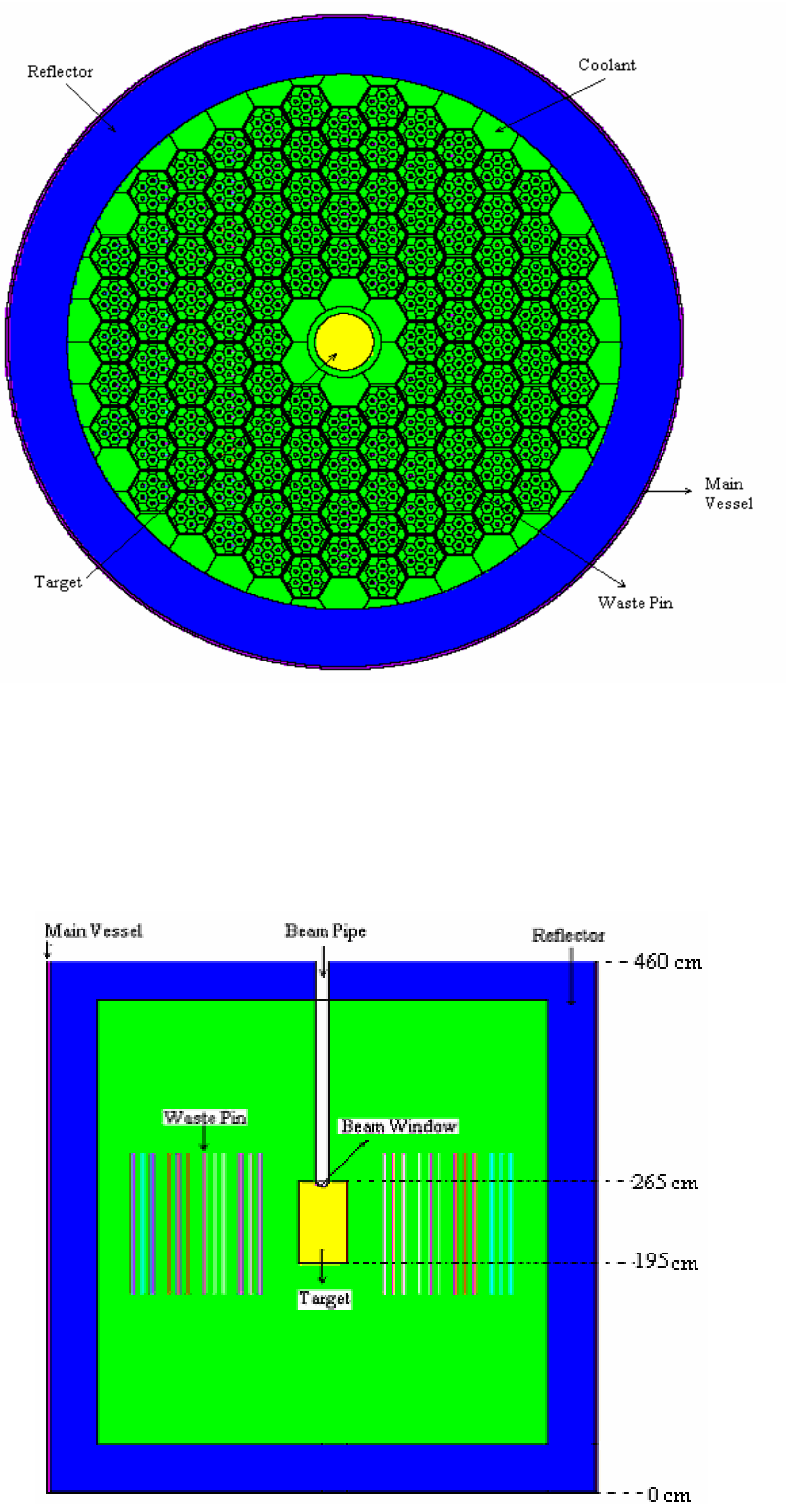


Figure 1. Horizontal and vertical sectional views of the present ADS design

3. Neutronic analysis

In the calculations, Monte Carlo code MCNPX [2], which is a combination of LAHET [3] and MCNP, was used. To transport a wide variety of particles, The Los Alamos High Energy Transport Code, LAHET, was used. These include charged particles such as protons and charged pions as well as neutrons with high energies. MCNPX offers options based on three physics packages; the BERTINI [4] and ISABEL [5] models taken from the LAHET Code System, and the CEM package [6].

The MCNP transport code is used for radiation transport at low energies (<20 MeV). The transport of high energy particles (>150 MeV) was performed using nuclear models, while for the transport of low energy particles (<150 MeV) cross section libraries have been used. In an ADS, the energy is produced as a result of the multiplying nuclear cascades initiated by accelerated protons interacting with the spallation target. Spallation reactions in a target material proceed through three stages: In the first stage, an incident particle interacts with the nucleus through successive nucleon-nucleon hard collisions in a potential which describes the density of the nucleus as a function of radius. Intranuclear cascade (INC) model is used this stage, in which high energy particles are emitted. After the INC stage of the reaction the nucleus is left in an excited state. This is the starting point for the second or preequilibrium stage of the reaction. In the third stage, the evaporation phase of the reaction, the remaining nucleus is de-excited either by evaporation or by fission. We used Bertini model for nucleons and pions, and Isabel model for other particle types in the intranuclear cascade stage. Preequilibrium model has been used for the second stage of the reaction. The default physics module options were used with preequilibrium model after intranuclear cascade.

The study was performed in three steps. In the first step, only the spallation target, proton beam pipe and beam window isolated from the core were considered. In the second step, all calculations have been made in absence of fissile materials and wastes. In the third step, the full reactor geometry was taken into account. In the calculations, we did not take time evaluation of the neutron spectrum and other quantities into account.

The target was designed to maximize the neutron yield and to soften the axial power distribution. The target optimization means finding the maximum of neutron production inside the target and the maximum of neutron leakage to the core at given proton energy and target shape and material. Lead is a material highly transparent to neutrons; it has a very low capture cross section, the main isotope in natural lead ^{208}Pb is double magic, and thus exceptionally stable, from the MeV region down to very low energies. But it has a moderate elastic neutron cross section.

One of the fundamental parameters of the spallation target is the number of neutrons produced per beam particle incident on target. Calculation of number of neutrons produced in the target is done by summing up the absorption and leakage. Because of small neutron absorption of lead, the leakage is considered to be equal to the neutron production. Spallation neutron and proton yields per one incident proton are 27.4 and 1.2E-02, respectively. Other particle yields such as, pion and muon per one incident proton have been neglected due to their low particle fluxes.

The neutron leakage on the whole surface of the target for all three steps mentioned above is shown in Table 1. Some of the spallation neutrons produced by protons in the target can escape through the beam pipe, therefore it is desired to minimize the neutron escape through the beam pipe. The neutron leakage through the beam pipe for target- beam pipe system (Fig. 2.) isolated from the core is shown in Table 2. The ratio of the neutron leakage into the proton beam vacuum to the total neutron leakage from the target top face is 1.54%.

Table 1. The neutron leakage on the whole surface of the target and main vessel wall.

Neutron leakage (n/s)	Beam pipe+target	Beam pipe+target +coolant	Full System
Target			
top face	3.25E+17	5.68E+17	1.44E+18
side face	1.34E+18	2.78E+18	9.43E+18
bottom face	4.02E+16	1.63E+17	1.03E+18
Main vessel			
top face	1.12E+14	8.60E+15	8.73E+16
side face	-	4.12E+16	3.42E+17
bottom Face	-	6.92E+15	8.15E+16

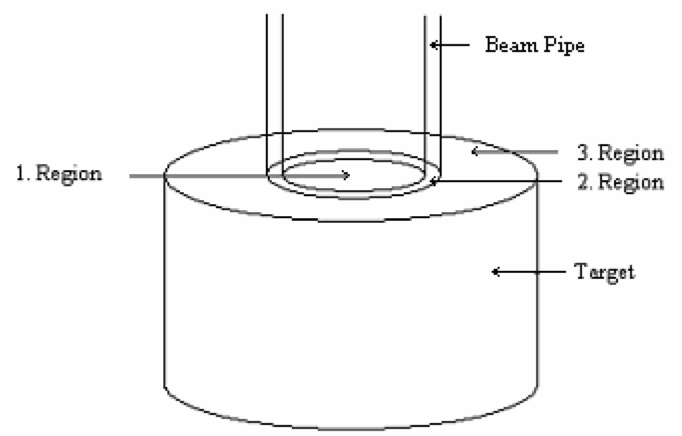


Figure 2. Scheme of the target-beam pipe system

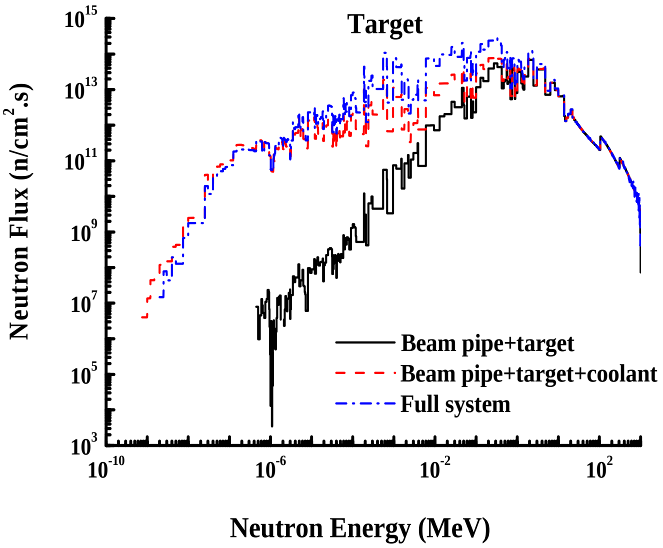
Table 2. The neutron leakage on the target top face for full system

Target Top Face	1. Region (n/s)	2. Region (n/s)	3. Region (n/s)	Total
	1.43E+17	1.54E+16	1.28E+18	1.44E+18

Energy and spatial distribution

The transmutation performance of a nuclear system mainly depends on flux level and neutron spectrum. To investigate the radial dependence for three steps, neutron fluxes and heating were calculated in a set of coaxial cylinders, subdividing the target into parts and also many sections for z-dependence.

The energy distributions of the spallation neutron and proton fluxes, the radial and axial variations of the neutron flux in target and energy distributions of the spallation neutron and proton fluxes in the beam window are shown in Fig. 3-5. The energy distribution of the spallation neutron is rather similar in the target and the beam pipe for all three steps. The energy distribution of the spallation protons for all three steps are the same. The peaks of the neutron distributions appear at energy about 2 MeV for all three steps. For the three systems, the neutron flux is low at energies lower than 1 MeV for target and beam pipe. The neutron flux is maximum at r=0 cm, z=195 cm in the target, Fig. 4.



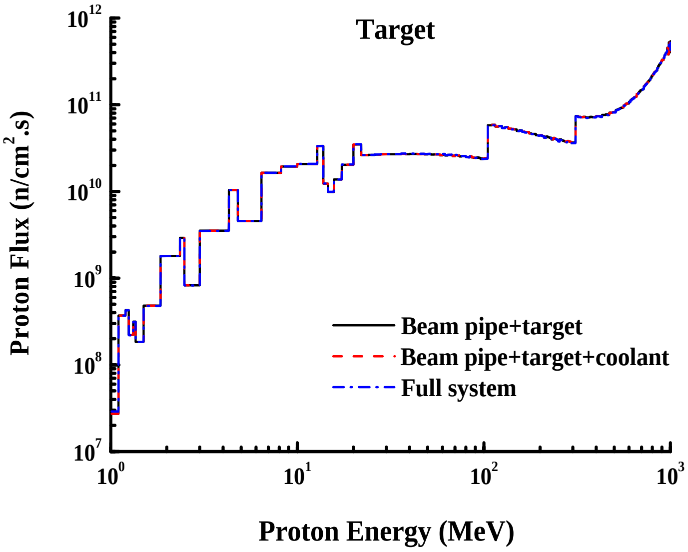


Figure 3. Energy distribution of the spallation neutrons and protons in the energy range 0-1000 MeV in the target.

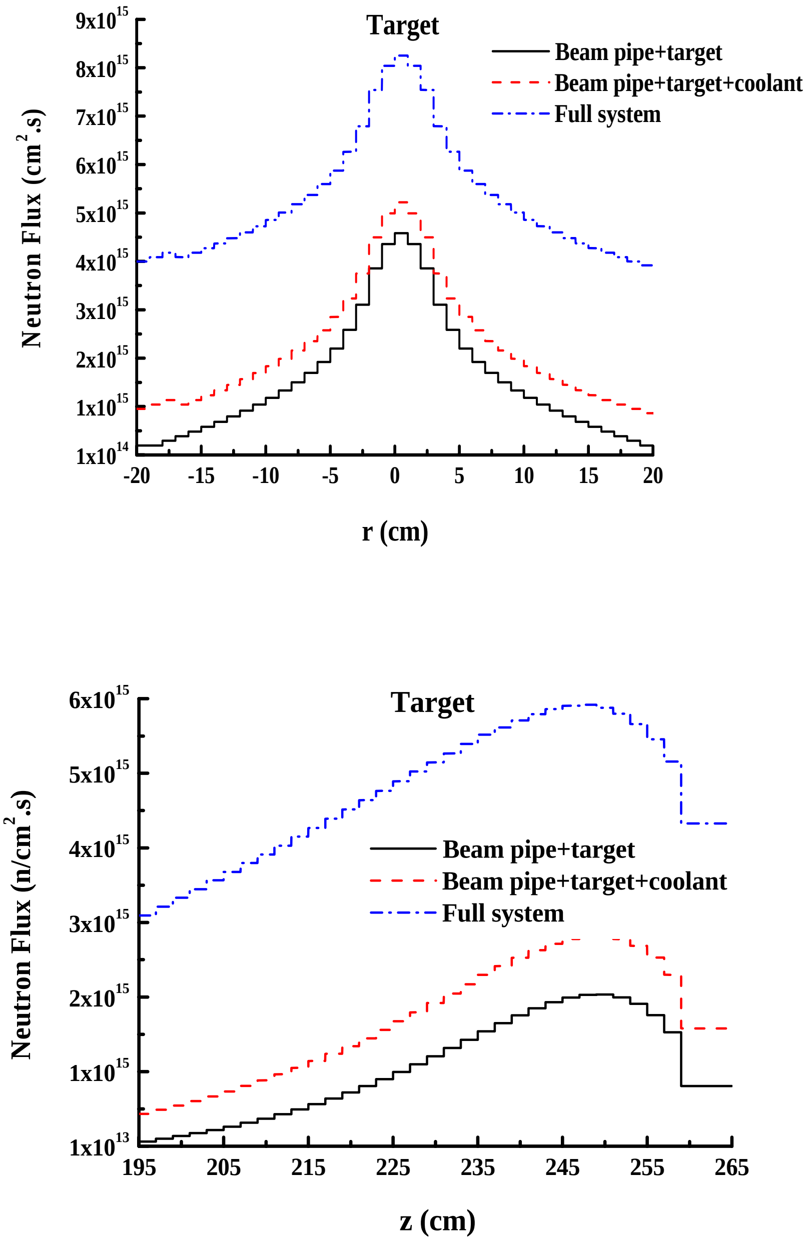
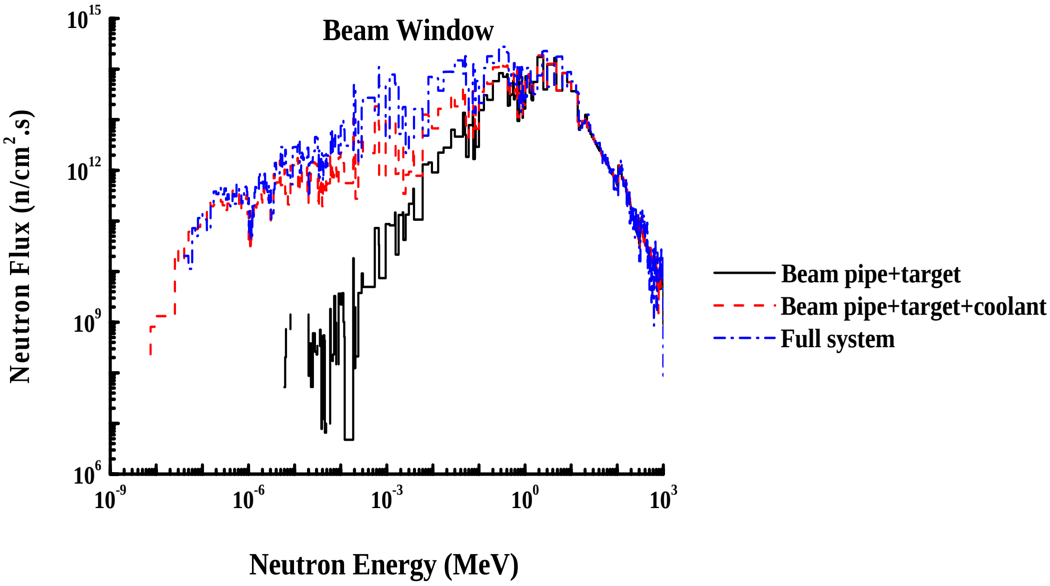


Figure 4. The spatial variation of the neutron fluxes in the target: (a) radial, (b) axial.



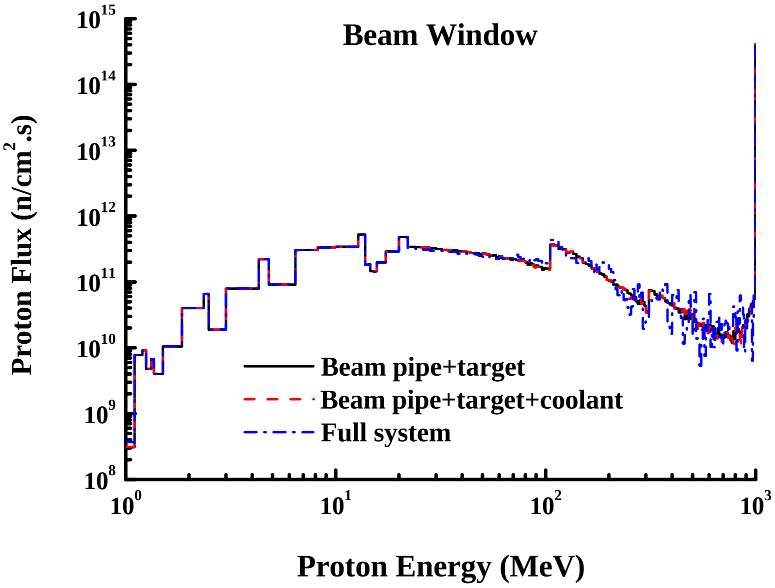


Figure 5. Energy distribution of the spallation neutrons and protons in energy range 0-1000 MeV in the beam window.

Heat deposition

The heat deposition by neutrons and protons in the some parts of the system for the three steps is given in Table 3., and radial variation of the heating in the target is shown in Fig. 6. It is observed that the amount of energy deposited in the target by neutrons is the mostly same for three steps. There is a negligible energy deposition by neutrons in the other structural materials except beam window.

Table 3. Energy deposited by neutrons in the some parts of the system for three calculation steps

Heating (kW/cm^3)	Beam pipe+ target	Beam pipe + target +coolant	Full System
Target	28.66	29.12	31.93
Beam window	84.83	107.74	96.09
Beam pipe	0.0	3.00	4.86
Main vessel wall	0.0	2.65E-03	2.14E-03

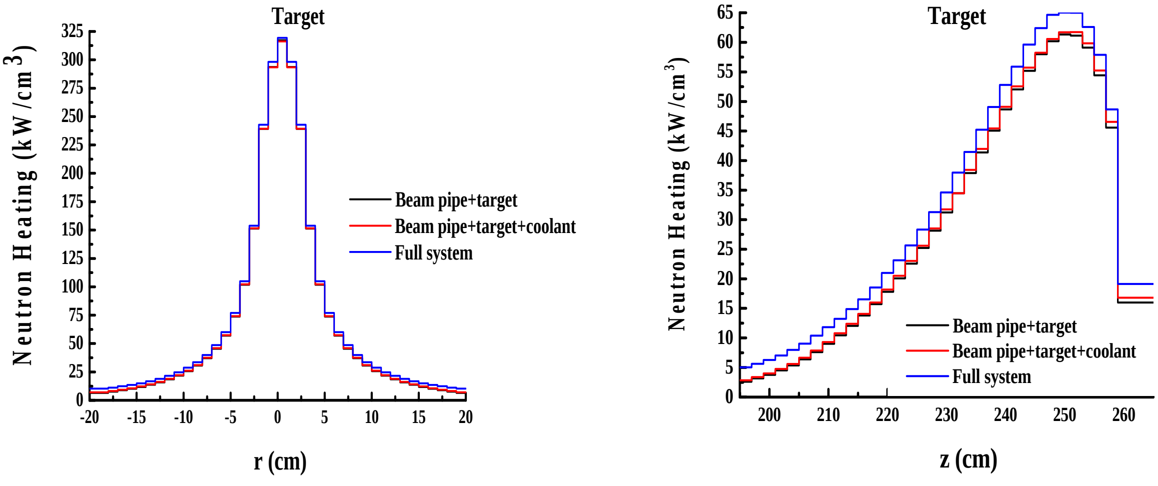


Figure 6. Radial and axial variation of the heating in the target

H and He production rates

The gas production rates in the target and beam window for full system are shown in Table 3. The results show that the gas production in the beam window is more intense than the target, as expected.

Table 4. Gas production in the target and beam window

Gas Production (apmm/fpy)	Target	Beam Window
H	2798.21	33000
H-2	304.1	3770
H-3	181.67	810
Total H	3283.45	37200
He-3	11.12	565
He-4	514.69	3320
Total He	525.81	3880
Total H + He	3809.20	41100

Transmutation

The core of the system mainly consists of the spallation target region and fuel (and waste) assembly region. The fuel (and waste) region consists of the pin-bundle type fuel assemblies and is cooled by liquid sodium. Figure 1 shows the schematic view of the target and the fuel assembly. The fuel elements consist of a stainless steel cladding, 0.35 cm thick, characterized by an external radius of 1.35 cm and a total height of 120 cm, end cap included. The fuel is a cylinder, 119.30 cm height. As mentioned above, we used a hexagonal lattice for fuel assemblies. The fuel subassemblies, bundles, are arranged in an annular array of five rings, each fuel subassembly contains 7 fuel rods. There are 114 bundles in the system. The core configuration for the calculations is characterized by:

10 bundles in ring 1 (^{239}Pu)
20 bundles in ring 2 (^{237}Np)
24 bundles in ring 3 (^{129}I)
30 bundles in ring 4 (^{99}Tc)
30 bundles in ring 5 (^{241}Am)

All calculations are based on pure ^{99}Tc, ^{129}I, ^{237}Np, ^{239}Pu and ^{241}Am isotopes.

The neutron multiplication factor, $k_{eff,}$ with its standard deviation was calculated by "kcode" card of the MCNP code and was found as, after running a large number of histories, 0.96646 ± 0.00119 for the core without the proton beam.

Transuranic elements (TRU) and fission fragments (FF) are two main components of high-level nuclear waste, representing 1.1% and 4% of spent nuclear fuel, respectively. TRU, produced by neutron capture in the fuel can only be successfully incinerated by fission, while the radioactivitiy of the FF can be modified by neutron capture.

Isotopes to be transmuted are minor actinides, such as ^{239}Pu, ^{237}Np, ^{241}Am, and are long-lived fission products, such as ^{99}Tc and ^{129}I . Minor actinides should be transmuted mainly through fission reactions has the possibility increasing higher actinides, while the thermal capture is main transmutation reaction for long –lived fission productions. The main nuclides of minor actinides from power reactor, such as Np, Am and Cm, have threshold fission reaction, so, hard neutron spectrum is desirable.

Table 5. Transmutation calculation results

	^{99}Tc	**^{129}I**	**^{237}Np**	**^{239}Pu**	**^{241}Am**
Initial mass (kg)	910.434	182.509	1068.769	522.932	1082.229
Total transmutation (kg/fpy)	1.455	0.806	36.434	47.254	4.212

The capture reactions rates of LLFP increases in the thermal energy region of ADS is hard, so it is necessary to establish. The region with softer spectrum for transmutation of LLFP. However, the effect on the MA transmutation properties of ADS is significant. Results of transmutation calculation are given in Table 5.

Results

Accelerator driven transmutation system was investigated. The system consists of 1 GeV, 10 mA proton accelerator and A subcritical core with an effective neutron multiplication factor of 0.96. For the neutronic analysis, we used the MCNPX code system. In the neutronic calculations, time evaluations have not been taken into account. The transmutation rates of minor actinides and long-lived fission products vary in the range 0.806-47.254 kg/fpy. The

ADS proposed in this study produces energy from nuclear waste material, while a considerable amount of this spent fuel is incinerated.

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