

# A simulation study on neutronic behavior of non-fissionable and fissionable materials of different geometries as spallation targets in ADS

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## HIGHLIGHTS

- Cone shaped U-238 generates a significant neutron yield, and heat as well.
- Hydrogen is the principal factor in swelling of spallation target and it consists of about 88% of gas production.
- The results show that the shape of the target plays the most important role on angular distribution.
- LBE target provides favorite parameters for both neutronic and physical properties.

## ABSTRACT

Spallation process is the most significant process for neutron generation in industry and medicine. This process has been used in the subcritical reactor core. In this research, we study the neutronic behavior of non-fissionable and fissionable spallation targets consists of U-238, Th-232, Lead Bismuth Eutectic (LBE) and W-184 materials in cylindrical and conic shapes using MCNPX code. Neutronic parameters consist of spallation neutron yield, deposition energy, and angular spectrum of the neutron output. The gas production rate and residual mass spectrum were investigated. The results of this research indicate that the shape of the target must be selected based on target material and operational purposes. The number of neutrons per energy unit is stable at energies higher than 1 GeV, and the rate of change in neutron generation has been reduced after that. Furthermore, hydrogen is the principal factor in swelling of spallation target and consists of about 88% of gas production. It was found that a target of LBE provides the most favorite parameters for both neutronic and physical properties.

## KEYWORDS

MCNPX code  
Spallation process  
Neutronic parameters  
Spallation targets

## 1 Introduction

Accelerator Driven Subcritical Reactors (ADSRs) are a kind of the conceivable fission reactors under attention that have target stability, boost safety and competitive economy (DoE, 1999). An ADSR is formed from the union of a particle accelerator with a proton beam of high energy (from few 100 MeV to few GeV) and subcritical reactor. Particle accelerator uses to initiate spallation reactions in a heavy metal target located inside the reactor core. The core of ADSR is shut down by shutting off the accelerator. This feature along with a subcritical core enhances immunity. The long-term radiotoxicity of nuclear wastes is a problem in the nuclear industry. Transformation of long-lived radionuclides can be used as a solution for this

problem and an Accelerator Driven Systems (ADSs) can therefore reduce these nuclear wastes (Rubbia et al., 2001; Sokolov et al., 2005; Mantha et al., 2007; Bungau et al., 2008).

There are several technologies for the target and accelerator (such as MEGAPIE (Salvatores et al., 1998)), the subcritical core (MYRRHA (Abderrahim et al., 2001), TARC (Rubbia et al., 1995) and MUSE (Mellier et al., 2005)). However, Fixed Field Alternating Gradient (FFAG) (Uesugi et al., 2008) synchrotron seems to be one of the most suitable candidates among the accelerator technologies under consideration for deployment in ADSRs (Pyeon et al., 2009).

In the spallation reaction, a relatively light particle similar to a proton or neutron (obtained from accelerator)

knocks up a heavy nucleus. This reaction is done to occur in two stages. In the first phase (the intra nuclear cascade), a high-energy particle cascade inside the nucleus is created by the retaliation proton. During this intra-nuclear cascade (INC), the nuclei emit high energy ( $>20$  MeV) secondary particles and low energy ( $<20$  MeV) cascade particles. The excited state is normal state of the nucleus after the INC. In the second phase (the evaporation), the excited nucleus reduces mainly by emitting evaporation neutrons with low energies (Demirkol and Tel, 2011).

There is a strong motivation for improving the accuracy of code predictions used to calculate the creation of neutrons during spallation reactions and the transport of high and low energy neutrons within the target material. It is expected that more codes can ascertain using of ADS in the future. However, now, there is a need for validating the nuclear data and computational tools for the existing ADS applications. In many existing codes, the INC model is used as a basis for first phase calculations. The fundamental assumption of the model is the nucleus-nucleus reaction in terms of binary nucleon-nucleon collisions inside the nucleus. A semiclassical approach is used for explanation intra-nuclear cascade. These calculations follow the history of single nucleons involved in nucleon-nucleon collisions. The Pauli principle is the only quantum mechanical theory inserted into the model. Bertini was the first person who wrote a code of the INC (Bertini, 1963) in 1963. Bertini and ISABEL invoke the Dresner evaporation model with Rutherford Appleton Laboratory (RAL) fission by default. The fission model can be switched to the ORNL model (Yariv and Fraenkel, 1979). This model contains the elemental cross-section data. The MCNPX input cards provide the user control of physics options. The options controlling the Bertini and ISABEL physics modules are taken from the “User Guide”. The user is referred to that document for further information. Nucleons will switch to the Bertini model below 3.0 GeV by default, and pions would switch below 2.0 GeV. Kaons and anti-nucleons would switch to the ISABEL model below 1.0 GeV. Nucleon and pion interactions simulated by Los Alamos high-energy transport code (LAHET) physics use the Bertini intra nuclear cascade model (Hughes et al., 2002; Prael and Lichtenstein, 1989).

The continuous energy of the Monte Carlo code MCNPX can be utilized for modeling neutron transport in critical or subcritical reactors (Hughes et al., 2002). This code is a coupling of two previous calculations codes: LAHET and the Monte Carlo N-Particle transport code (MCNP) (Brown et al., 2000). It authorizes the study of transporting problems in a large range of energies, from 20 MeV to a few GeV. Cross-sections for transport of muons, pions, and antinucleons and single processes nucleons with the energy of  $>20$  MeV are being generated by LAHET, while the MCNP is able to model the transport of electrons, photons, and neutrons within the energy range of 10–11 MeV to 20 MeV. Cross-sections had been taken from libraries of evaluated data.

While thermal-hydraulics under transient conditions and steady state performance were studied in the previous

publications on spallation target (Cheng et al., 2006; Tak et al., 2005, 2004; Taninaka et al., 2011), a little knowledge has been gained about the structural reactions during beam trip transients. Three issues are investigated in the present paper. Evaluating the efficiencies of two geometries including cylinder and cone for four materials of U-238, Th-232, W-184 and LBE as the target; Calculation of heat generation and distribution in the target by analyzing two target geometries. Calculation of the angle and residual mass distributions, and gas production rate as well.

## 2 Materials and Methods

One of the most interesting and urgent components of ADS is the spallation target. In the neutronic point of view, owing that a large value of neutrons is produced by the spallation reaction, the neutron production rate is one of the important parameters for selecting the target material. In the thermal-hydraulic point of view, cooling sufficiently is the basic issue for selecting material of the target.

In this study, the MCNPX code packages have been used to study the spallation neutron yield using the Bertini default physical model for protons of 0.01 to 3.0 GeV irradiated to the target. A cylindrical target with diameter of 15 cm and height of 60 cm (based on the literature (Feghhi et al., 2014; Hassanzadeh and Feghhi, 2015)), and a conical target with diameter of 25.98 cm and height of 60 cm have been simulated (for two equal volumes). In addition, the results corresponding to two non-fissionable materials, such as W-184 and LBE, and two fissionable materials, such as U-238 and Th-232, have been compared. A 1.0 mA proton beam of uniform spatial distribution has been irradiated to the upper surface of the targets.

One of the non-fissionable target materials is tungsten (W-184) which presents chemically inert, less corrosion, resistor to radiation damage, good accessibility, and low price that are close to that of an arbitrary spallation target. It is why tungsten is one of the most important targets that can offer spallation neutron. Nevertheless, absorption of neutrons is an undesired characteristic of tungsten. If tungsten under irradiation contacts with water, it corrodes. Ta cladding is therefore added (Feghhi et al., 2014; Hassanzadeh and Feghhi, 2015; Kawai et al., 2003; Ammerman et al., 2001).

Another non-fissionable target is LBE with good thermal/mechanical properties and low melting temperature ( $123^{\circ}\text{C}$ ). LBE remains liquid when bombards with proton beam, an important amount of heat is deposited inside the target. This character makes LBE more appropriate than other liquid targets. The high boiling temperature of  $1665^{\circ}\text{C}$  is an advantage for this material (Feghhi et al., 2014; Hassanzadeh and Feghhi, 2015; Kawai et al., 2003; Ammerman et al., 2001).

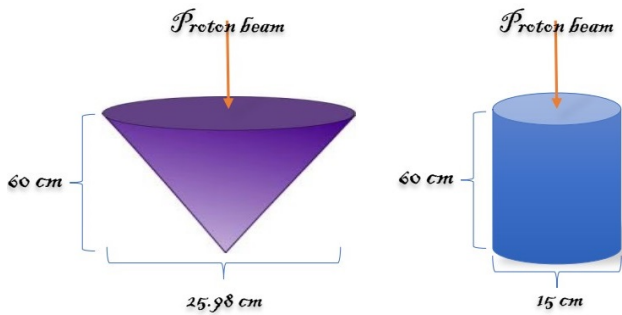
Although the use of fertile material target is more common in the works published so far, Uranium (U-238) could be used as a fissionable solid target with a proton beam of power level about 1.0 MW (*i.e.*, the proton energy,  $E_p$ , is 1.0 GeV and the accelerator current,  $I_p$ , is 1.0 mA. Of

course, it is known that the use of a U-238 target by the proton beam power would be very infeasible. This problem solution is to use a non-actinide target at a higher power level (Feghhi et al., 2014; Hassanzadeh and Feghhi, 2015; Kawai et al., 2003; Ammerman et al., 2001).

Thorium (Th-232) is a fissionable target with a high atomic number, relatively high density, low thermal conductivity and high melting point (1750° C). Th-232 is a desirable target in ADS with high neutron production rate. Moreover, absorption of neutron might cause production of U-233, which is another advantages of this target (Feghhi et al., 2014; Hassanzadeh and Feghhi, 2015; Kawai et al., 2003; Ammerman et al., 2001).

MCNPX simulation of spallation reactions has three stages with a special model used for each of them. The INC is the first phase coincident with the pre-equilibrium stage. Equilibrium evaporation is the second stage that challenges with the fission channel. After evaporation, gamma rays are generated by a de-excitation of the residual nucleus. Thus, this code can use different models for the explanation of individual stages of the spallation reaction (Demirkol and Tel, 2011; Bertini, 1963; Yariv and Fraenkel, 1979).

Spallation neutron yield, deposition energy, angular spectrum of the neutron output, gas production rate and residual mass spectrum by 1.0 GeV protons are calculated for four materials and for cylinder and cone shape geometries. Protons with this energy can be generated in accelerator the FFAG (Uesugi et al., 2008). In addition, in accelerator-driven spallation sources, high-energy particles with general protons' energy of ~ 1.0 GeV have been used (For example, KAERI, XT-ADS and JAEA) (Salvatores et al., 1998; Abderrahim et al., 2001; Rubbia et al., 1995; Mellier et al., 2005; Uesugi et al., 2008; Pyeon et al., 2009). Figure 1 shows a schematic 3-D view of two proposed targets with cylindrical and conical geometries, their dimensions, and the position of the proton source as well.



**Figure 1:** A schematic 3-D view of the cylindrical and conical targets.

### 2.1 Calculation of neutronic parameters

The obtained neutrons per proton is the most important parameter for designing a neutron source in an ADS. In addition to the target material and proton beam energy, geometrical specifications and shape of the target are effective on the spallation neutron yield ( $Y_{n/p}$ ) corresponding to the target. Equation (1) is used to calculate the

spallation neutron yield (Hassanzadeh and Feghhi, 2015):

$$Y_{n/p} = \frac{\langle S_n \rangle}{\langle S_p \rangle} \quad (1)$$

where  $S_p$  is the number of primary protons and  $S_n$  is the number of output neutrons. It is worth mentioning that in these simulations, the ENDF/B-VI neutron libraries are used.

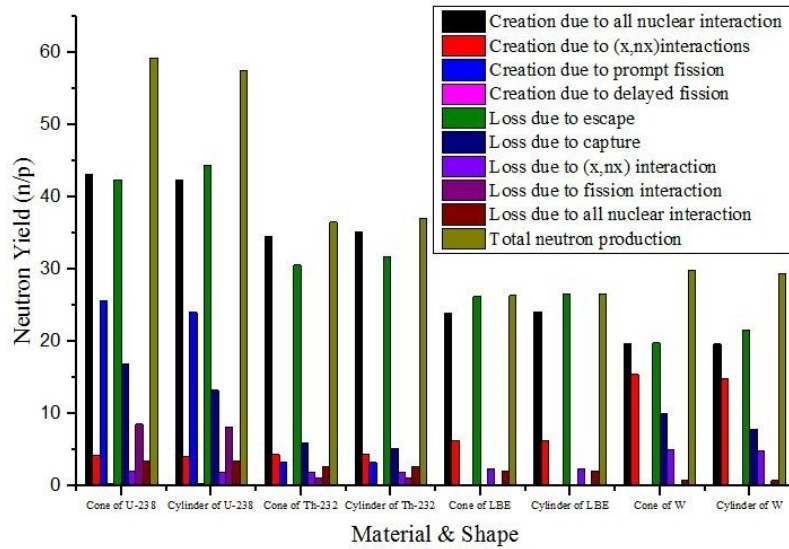
## 3 Results

### 3.1 Spallation neutron yield

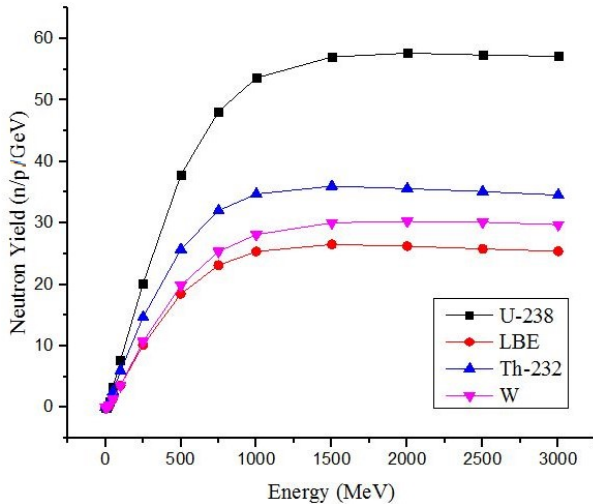
In the first section, the number of spallation neutron yield and leaked is calculated for four targets bombarded with 1.0 GeV protons. The results are compared with the total number of neutrons per unit of proton energy between 10 to 3000 MeV. In order to choose the optimal of neutron production and leakage in spallation target, the size and protons energy should be optimized.

In Fig. 2, the number of spallation neutron yield and leaked from the various process for two shapes and four materials for collision 1.0 GeV proton energy is calculated by MCNPX code. The results show that the number of neutrons in the fissionable targets is more than the non-fissionable targets in all interactions, except loss due to  $(x, nx)$ . It can be explained by cross sections of the process. Also, according to the rate of interactions in this way, the escape of neutrons increases with raising particle energy, but the neutron leakage decreases with increasing of dimensions. The number of the spallation neutron yield of a fission process is zero for LBE and W-184, but is 0.34n/p and 0.32n/p for U-238, and 0.05n/p and 0.03n/p for Th-232 in the cylinder and cone shapes, respectively. All of these parameters are calculated for the volume/surface of the targets.

Figure 3 shows the total number of spallation neutron yield per unit of collision proton energy ( $Y_{n/p}/E_p$ ) in cylindrical shape four non-fissionable and fissionable materials such as W-184, LBE, Th-232, and U-238. In this figure, the incident energy of the proton beam has an approximately linear interrelation with the spallation neutron yield within the range from 0.01 to 1.0 GeV. Maximum value occurs at around 1.0 GeV, and then it begins to decrease at higher energies. Also, the value of the spallation neutron yield for U-238 is nearly two times more than that of other materials. After U-238, high neutron yield is achieved by Th-232, and W-184 and LBE targets have fairly high neutron yields. Due to higher density than LBE which leads to a higher neutron yield, and high melting point as well, W-184 can be used as a spallation target for low thickness (due to neutron capture) However, because of the advantages such as thermo-hydraulic as well as higher mechanical strength, LBE is preferred as a non-fissionable target in ADS. Moreover, as Fig. 3 shows, U-238 target used in this study has the highest neutronic yield and can be effective in the products of the neutron creation reactions which would prevent the transmutation efficiency of the proton ADS (Hassanzadeh and Feghhi, 2015).



**Figure 2:** Comparison of spallation neutron yield and leaked in cylinder and cone shapes of U-238, Th-232, W-184 and LBE targets



**Figure 3:** Comparison of the total number of spallation neutron yield per unite proton energy in the cylinder shape for U-238, Th-232, LBE and W-184 targets.

Using fissile materials as the neutron source in ADS have many difficulties such as swelling and delayed neutron production. These difficulties can be overcome by using non-fissile materials as the spallation target (Hasanzadeh and Feghhi, 2015). Finally, in order to select an optimum target, other parameters such as the gas production rate, residual mass, angular spectrum, and heat deposition should be considered. The mentioned parameters have been investigated in the present paper.

The accuracy of the results are examined through comparison with experimental data for spallation neutron yield in different target materials, dimensions and incident energies (See Table 1). The experimental data were reported by AECL, BNL (Feghhi et al., 2014; Kimura, 1992; Kumar et al., 2003) and the computational data were obtained by MCNPX code for this parameter in U-238 and W-184 targets with different energies. In this

study, the average relative difference between computational and experimental results for spallation neutron yield was about 8.35%. According to the ACEL report (Feghhi et al., 2014), the obtained experimental results by the foil activation method is believed to be accurate to within  $\pm 5.0\%$ . In addition, in this research, the obtained calculations data had an average uncertainty of less than 1.0%.

### 3.2 Energy deposition

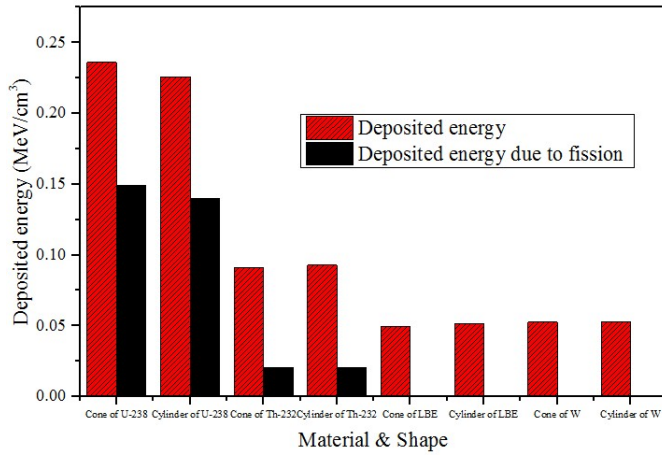
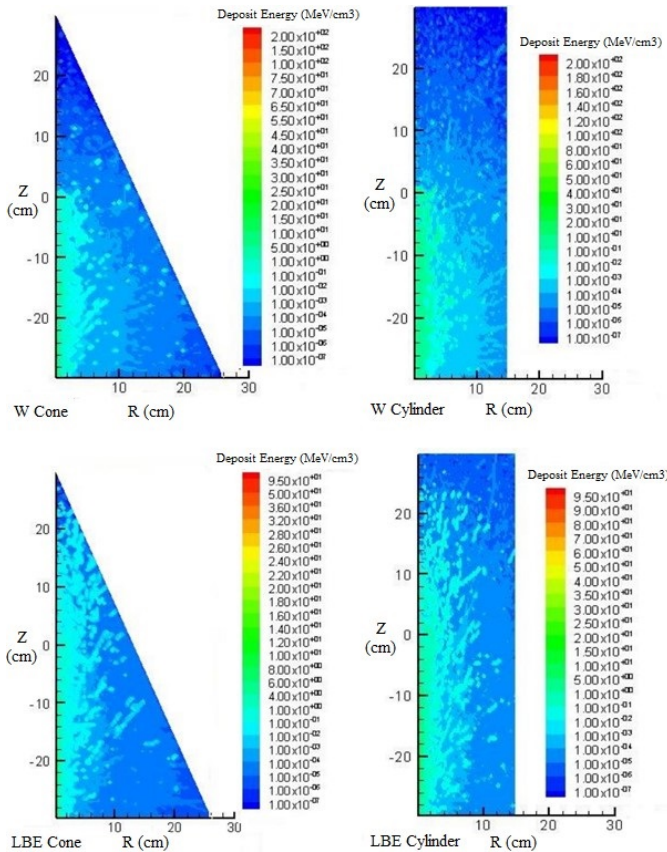
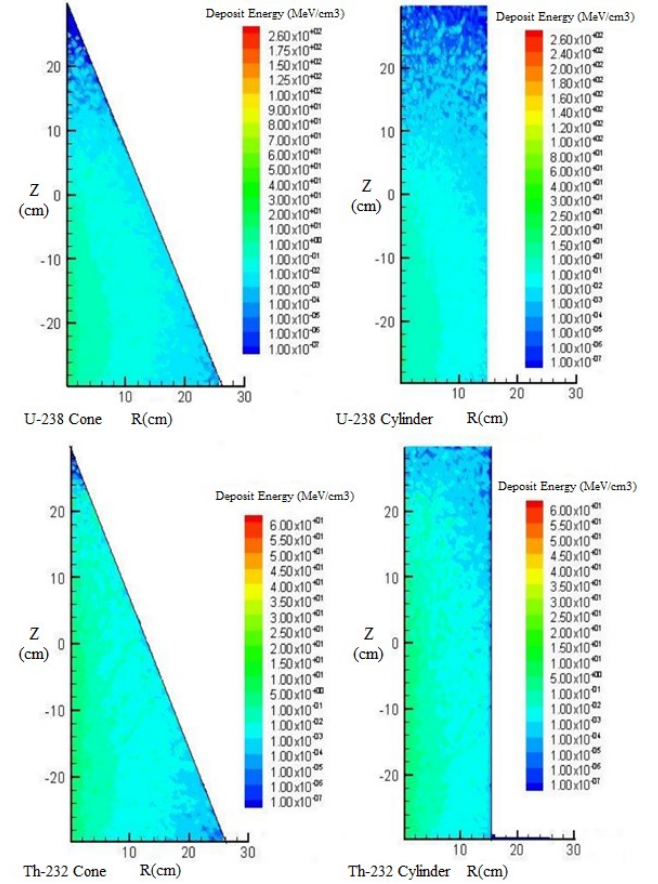
The deposited neutron energy calculated for fissionable and non-fissionable materials due to the collision of 1000 MeV protons for cylinder and cone shapes using MCNPX code are shown in Fig. 4. As can be seen, the differences between fissionable and non-fissionable materials are the amount of energy has been deposited and energy from fission. In the same materials, energy has been deposited are equal almost for cylinder and cone shapes. Therefore, this factor does not depend on the geometry. In other words, the value of the deposited energy is not sensitive to the geometry shape. Also, the highest energy deposition obtains for U-238 material target and the heat is generated bear noticeably less in other targets. Moreover, in the collision of 1 GeV protons with the conical target, the highest energy of 0.24 MeV/cm<sup>3</sup> is deposited in the U-238 target.

Figure 5 shows the contour deposited energy of two non-fissionable including W-184 and LBE in cone and cylinder shapes. The similar figure is shown in Fig. 6 for U-238 and Th-232 fissionable targets in cone and cylinder shapes. Tecplot 10 software is used to draw contours in (Hughes et al., 2002). Deposited energy is focused in the center of the non-fissionable materials, but it is distributed in all areas of the fissionable materials. As mentioned earlier, the value of this parameter is not sensitive to the geometry shape but depends on types of the fissionable and non-fissionable materials.



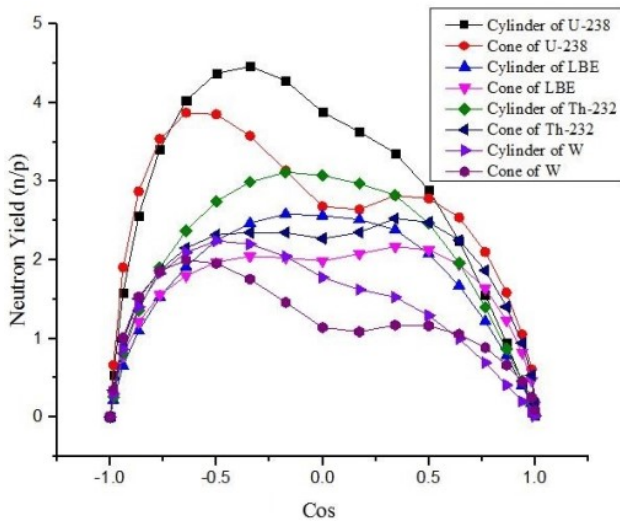
**Table 1:** Comparison of the results corresponding to MCNPX code and experimental data (Kumar et al., 2003; Johnson et al., 1985) for spallation neutron yield with different target materials, dimensions and energies.

Material	Dimension (diameter × length) (cm <sup>2</sup> )	Energy (GeV)	Experimental data	MCNPX code	Relative difference (%)
U-238	10.2 × 61	0.96	40.5	33.025	18.45
	10.2 × 40	0.8	15.0	14.736	1.76
W-184	10.2 × 40	1.0	20.5	19.012	7.25
	10.2 × 40	1.4	28.8	27.093	5.92

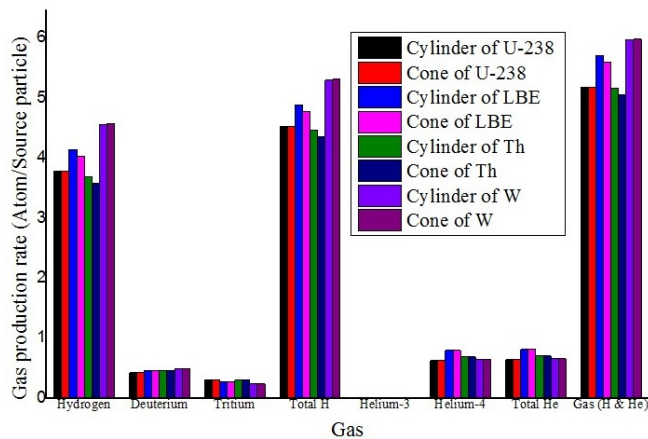
**Figure 4:** Comparison between the results of energy deposited in the cylinder and cone shape U-238, Th-232, LBE, and W-184 targets.**Figure 5:** Comparison between the results of deposited energy contours in the cylinder and cone shape LBE, and W-184 targets.**Figure 6:** Comparison between the results of deposited energy contours in the cylinder and cone shape U-238, and Th-232 targets.

### 3.3 Angular spectrum

Figure 7 shows the angular spectrum of the total leaked neutrons from the surface of the in fissionable and non-fissionable target materials for cylinder and cone shapes. Angular spectra of U-238 and W targets show the same trends of Th-232 and LBE targets. As is shown in this figure, the maximum of neutron yield is between 120 to 140 degrees in the cone shape of U-238 and W targets and 100 to 120 degrees for cylinder shape. For LBE and Th-232 targets in a cylinder shape, the maximum is 90 to 100 and for cone shape is 60 to 70 degrees. Neutron yield corresponding to the cylindrical shape of U-238 is more than other geometries and materials. Therefore, the obtained results show that the angular spectrum not only depends on the geometry shape but also depends on the fissionable and non-fissionable materials.



**Figure 7:** Comparison between the results of the leaked neutron in the cylinder and cone shape U-238, Th-232, LBE, and W-184 targets.



**Figure 8:** Comparison between the results of gas production rate in the cylinder and cone shape U-238, Th-232, LBE, and W-184 targets.

### 3.4 Gas production rate

Target swelling of gas generation due to longtime proton irradiation in the target is one of the most significant parameters for choosing the material in ADS. Figure 8 shows the comparison between the results of gas production average rate (hydrogen and helium) in the cylinder and cone shapes of LBE, W-184, U-238, and Th-232 targets. The average rate of gas production in W-184 target is more than other materials. LBE, U-238, and Th-232 targets are in the next ranks, respectively. Hydrogen is the principal factor in swelling of spallation target and consists of about 88% of gas production. However, the production rate of helium, krypton, and xenon gases is less. Hydrogen atoms are at an interstitial site and helium atoms are at a substitution site. Therefore, the migration behavior is different. At high temperatures, hydrogen atoms escape to sinks and the effect is not so large. Helium atoms play an important role at high temperatures. Even if the production rate of helium atoms is less than 1/5 of that of

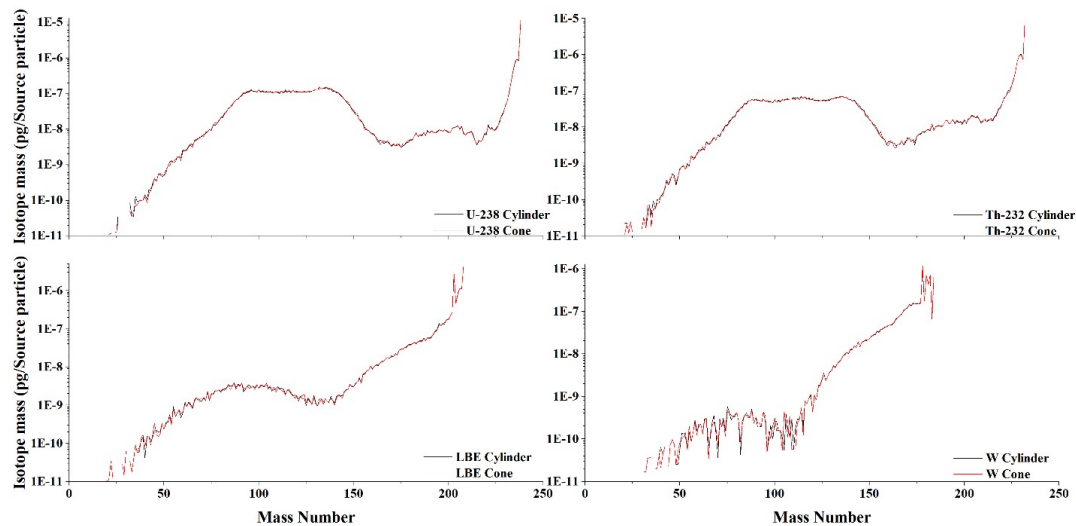
hydrogen atoms, the effect of helium atoms is not ignored for void swelling. Thus, the cone shape of Th-232 provides the minimum average rate of gas production. In addition, the obtained results show that this parameter weakly depends on the geometry shape, but it highly depends on types of fissionable and non-fissionable materials.

### 3.5 Residual mass spectrum

Radiotoxicity after irradiation is an important parameter for choosing the optimal spallation target. Therefore, radionuclide distribution has been computed for the chosen spallation targets. Figure 9 shows the residual mass distribution in the cylinder and cone shapes of U-238, Th-232, LBE and W-184 targets. The results show that while the residual mass is not dependent to the shape of the target, it depends to its material. In addition, the highest radionuclide production occurs for the U-238 material target. Th-232 is the second target between the other studied targets for this case. Thus, the obtained results show that the residual mass distribution does not depend on the geometry shape but it depends on types of fissionable and non-fissionable materials.

## 4 Conclusions

One of the most important elements in the ADS reactor is selection of the spallation target. Neutron production rate is the most essential character for target material selection because large amounts of neutron are produced by spallation reaction in the target. Therefore, in this article, we calculated the number of spallation neutron yield and leaked from the cylinder and cone shapes of U-238, Th-232, LBE, and W-184 targets using MCNPX code. The results were compared with the experimental data reported in the literature. The average relative difference between computational and experimental results for spallation neutron yield was about 8.35%. The value of energy deposition in the target from colliding proton with 1 GeV energy was also calculated. The angular spectrum, gas production rate, and residual mass distribution of collision of 1 GeV protons were calculated. According to results, cone shape of U-238 provides more neutron yield, but generates more heat too and therefore is not suitable for spallation target. However, LBE and W-184 materials are appropriate candidates to be used as spallation target. In addition, hydrogen is the principal factor in swelling of spallation target and consists of about 88% of gas production. Furthermore, the results show that the shape of the target plays the most important role in the angular spectrum. In general, the results of this research shows that fissionable materials can not be used for real ADS. In addition, specifications of solid targets such as thermal hydraulics are not beneficial, especially in high beam powers. Finally, LBE target gives more favorite parameters for both neutronic and physical properties.



**Figure 9:** Comparison between the results of radionuclide production in the cylinder and cone shape U-238, Th-232, LBE and W-184 targets.

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