

Monte Carlo Simulation of Damage in Born Neutron Capture Therapy (BNCT) Converter Materials by High-Energy Proton Beam Spallation

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Received: 23 July 2023 / Accepted: 28 April 2024

Abstract

Purpose: High-energy protons are generally used for neutron production by Pb, W, Li, Be, and Ta targets that are used for the Born Neutron Capture Therapy (BNCT) technique. Neutron production targets are destroyed by proton spallation (evaporation of nuclei). The purpose of this study is the investigation of neutron activation and proton spallation damage of converter targets using the MCNPX code, which is based on the Monte Carlo method.

Materials and Methods: The MCNPX code was used to extract the activation and spallation information of secondary particle production in Pb, W, Li, Be, and Ta targets. The neutron activation and proton spallation damage, including radioactive elements production in converter targets, was extracted from data in the MCNPX output file.

Results: Results showed that the highest probability of radioactive elements production by proton with low-level energy in the Ta target are ¹⁸⁰Hf, ¹⁷⁹Hf, and ¹⁷⁸Hf, and in the Li target is ⁷Be, respectively. In addition, the most probable radioactive elements produced by 200, 800, and 1200 MeV proton spallation in lead target are ¹¹⁸Tl and ⁷⁸Pt, and in tungsten target are ⁹⁸Hf, ¹¹⁰Ta, and ¹¹¹Ta, respectively. The calculations showed that the production of radioisotopes in reactions with neutrons is lower than the production in reactions with a proton beam, and with increases in the energy of the proton beam, production of the radioactive elements was increased.

Conclusion: The results illustrated that the radioactive elements are produced in W, Pb, Li, Be, and Te targets in the BNCT method, which should be avoided as radiation hazards.

Keywords: Spallation; Activation; Proton; Neutrons; Radioactive Elements; Born Neutron Capture Therapy.

1. Introduction

In order to treatment of cancerous tumors, various methods were applied such as surgery, laser radiation, brachytherapy, radiation therapy, chemotherapy Methods, etc. [1-3]. In radiation therapy, the neutron, photon, electron, and heavy-ion beam was used for cancer treatment [4-9]. A neutron beam has been used in the treatment of brain tumors using the BNCT technique [10, 11]. Neutron in the BNCT method is produced in different ways [12-14], such as reactor and spallation processes [15-18]. In neutron therapy, the neutron source can be produced by proton irradiation on special targets. These neutrons converted to thermal energy by passing through different materials for the BNCT method [19]. BNCT treatment works by nuclear capture and fission of nonradioactive materials such as ^{10}B with low thermal neutrons. This procedure produces the ^7Li and an alpha particle with high linear energy transfer that deposits all energy in the tumor. By collision of particles with high energy, the spallation process can occur. In this reaction, the light particles were produced with a smaller atomic number. A spallation reaction is comparable to a glass being broken into many pieces. Spallation can be described as a two-step reaction; the target nucleus is heated in the first step, and in the second step, the target is evaporated. The neutron beam can be produced by proton spallation of Li, Be, and Ta targets [20-22]. In the spallation process with high-energy particles, due to the energy transfer to the nucleus in the target, the evaporation process takes place and a wide range of elements with a mass number less than the target nucleus is produced [23]. The neutron beam can be produced by proton spallation of Li, Be, and Ta targets [35,36].

In this research, the proton spallation of Pb, W, Li, Be, and Ta targets is investigated for neutron production. In this paper, the MCNPX code which is based on the Monte Carlo principles, is used for the investigation of spallation processes. In fact, the MCNPX code is a coupling of two previous calculations, the MCNP and LAHET codes. This code can transport the neutron and 32 atomic and nuclear particles. Previously, neutron activation surveys on patients following Boron Neutron Capture Therapy (BNCT) were performed [37], but the radioactive

elements production in converter targets that are built by neutrons and protons has not been investigated by researchers. Also, the spectrum of neutrons produced by Pb, W, Li, Be, and Ta targets can interact activate with these targets. In addition, the production of radioactive elements is determined by using the MCNPX code. The residual radioactivity in BNCT facilities was studied by neutron activation processes [38,39]. But residual radioactivity in targets is not investigated by spallation processes. The aim and innovation of this research is the calculation of the radiation damage of the material that used for neutron production in BNCT method. By using of the production elements as radiation damage, the radiation hazards caused by target for BNCT workers can be investigated.

2. Materials and Methods

The investigation of the neutron activation process and the spallation of protons in Pb, W, Li, Be, and Ta targets has been performed. The radioactive elements in Li, Be, and Ta targets can be produced by proton spallation. The input file of the MCNPX code, written in the first step, includes the geometry card, surface card, and data card, which contain the following components. The geometry of the cells used consists of Pb, W, Li, Be, and Ta in the spallation and activation mode, which is a sphere with a 2 cm radius. In the data card section, information about the elements' percentage of material, information about proton and neutron sources, as well as how to extract spallation data, and the spectrum of neutrons produced, is given. Table 1 shows the information on proton sources used in this project.

The MCNPX code was used to extract the activation and spallation information. The unit of spallation and activation damage (or spallation yield) is the production mass of secondary particle(g) per one gram(g) of target for a single particle of source(sp). Therefore, the unit of spallation damage is $\text{g}/(\text{g.sp})=1/\text{sp}$.

2.1. Validity of the Monte Carlo Simulation Program

In a Monte Carlo simulation project, it is important that the validity of the result is investigated by

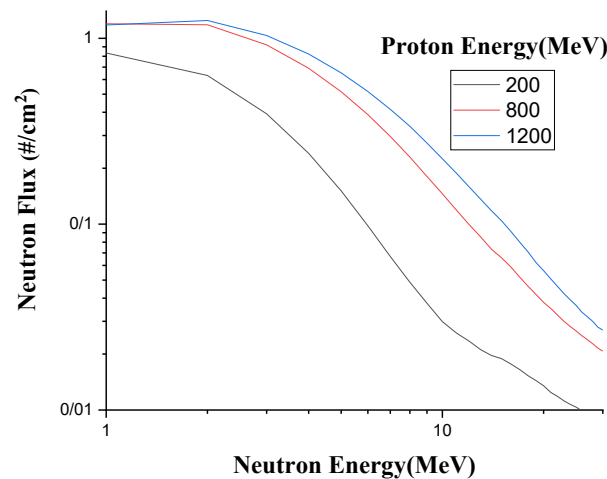
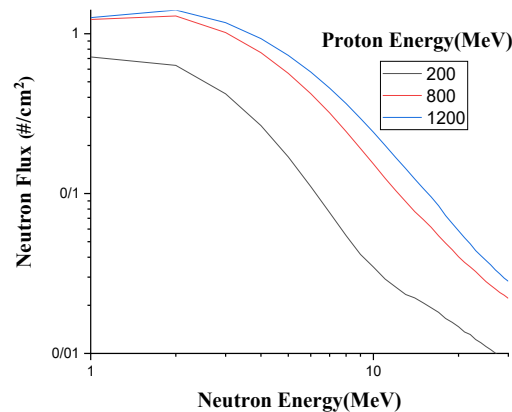
Table 1. Information on proton sources and high-energy ions [17, 28, 24,25]

Ion	E(MeV/u)	amu	E(MeV)	Secondary particle	Target 1	Density (g/cm ³)
H	1.91-2.7	1	1.91-2.7	n	Li	0.51
H	9-28	1	9-28	n	Be	1.85
H	50	1	50	n	Ta	19.6
H	178.5	1	200-1200	n	Li	0.51
H	200-1200	1	200-1200	n	Pb	11.34
H	200-1200	1	200-1200	n	W	19.3

comparing the result with other similar research. For this matter, the produced neutron flux by 200 - 1200 MeV proton irradiation on W and Pb targets was extracted by the MCNPX simulation code and was compared with the practical result. The neutron flux generated in W and Pb by 200,800 and 1200 MeV proton irradiation was extracted and is shown in Figures 1 and 2.

Figures 3 and 4 show the neutron flux of Pb and W target S by 1000 MeV proton beam that measured by experimental method [26].

Comparison of Figures 3 and 4 with Figures 1 and 2 shows a good agreement between the results of this paper and other works. Therefore, the validity of this research is satisfied.

**Figure 1.** The neutron flux of the W target by 200 - 1200 MeV Proton irradiation**Figure 2.** The neutron flux of Pb target by 200 - 1200 MeV proton irradiation

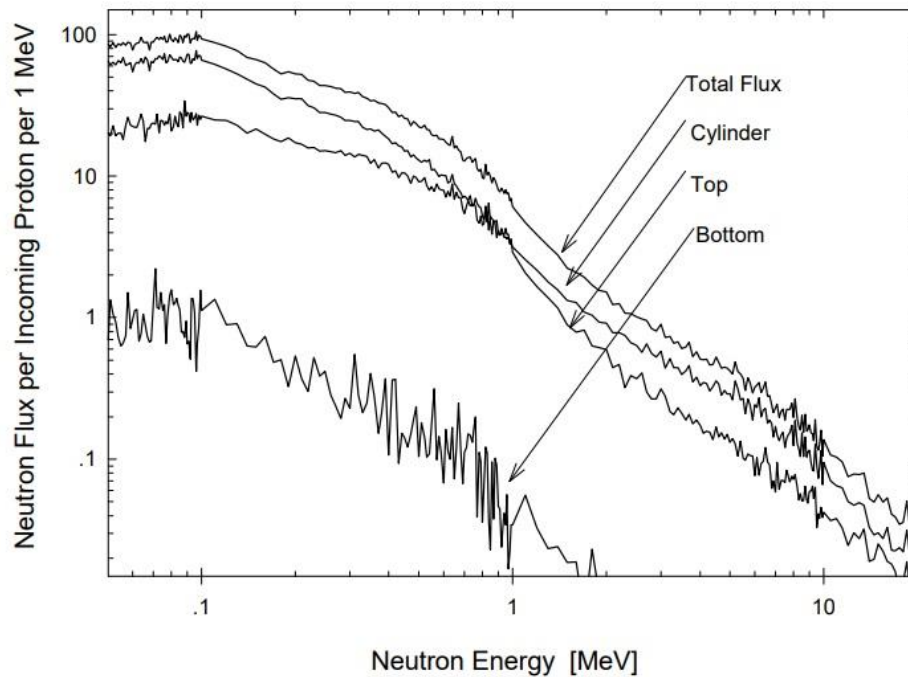


Figure 3. Neutron flux of lead target by 1000 MeV proton beam irradiation [26]

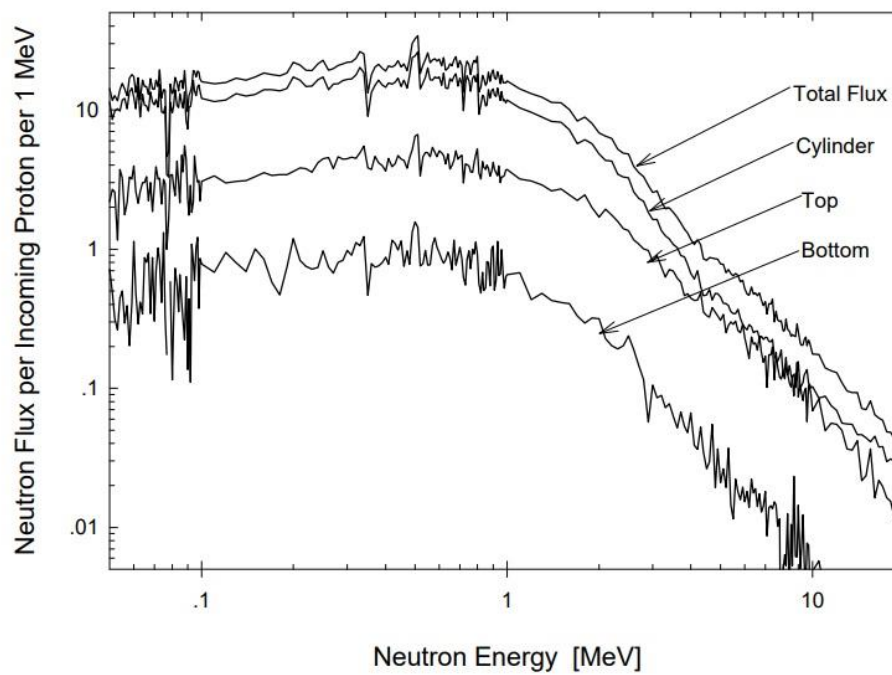


Figure 4. Neutron flux of Tungsten target by 1000 MeV proton beam irradiation [26]

3. Results

The results are presented in 2 steps. In the first step, the proton spallation and activation yield of the Li, Be, and Ta targets were calculated. In the second step, the

proton spallation yield of W and Pb by 200,800 and 1200 MeV Protons for neutron production was investigated.

3.1. Proton Spallation and Activation Yield of the Li, Be, and Ta Targets

The spallation and activation yield of the Li, Be, and Ta targets by proton irradiation was calculated with 1.9-2.5 MeV, 9-28 MeV, and 50 MeV protons that hit the targets, respectively. The radioactive elements produced in the Li, Be, and Ta targets were also calculated, and the major radioactive elements with half-lives longer than a day in every target are listed in Tables 2-5. In Table 2, Z and N are the atomic and neutron numbers.

Table 2. Radioactive elements produced by Proton activation in Li and Be

Element	N	Z	Decay Mode	Half-life
⁷ Be	4	3	ϵ	53.12 d

Table 2 shows that the ⁷Be radioactive element with a half-life of 53.12 d is produced as a result of proton interaction with Li and Be targets.

Table 3 shows the radioactive elements produced in the Ta target as a result of proton radiation with an energy of 50 MeV.

Table 3. Radioactive elements produced by proton and neutron activation in Ta

Element	N	Z	Decay mode	Half-Life
¹⁷⁵ Ta	73	102	$\epsilon + \beta^+$	10.5 h
¹⁷⁶ Ta	73	103	$\epsilon + \beta^+$	8.09 h
¹⁷⁷ Ta	73	104	$\epsilon + \beta^+$	56.56 h
¹⁷⁸ Ta	73	105	$\epsilon + \beta^+$	9.31 m
^{178m} Ta	73	105	$\epsilon + \beta^+$	2.36 h
^{178m2} Ta	73	105	IT	60 ms
¹⁷⁹ Ta	73	106	ϵ	1.82 y
¹⁸⁰ Ta	73	107	ϵ, β^-	8.152 h
^{180m} Ta	73	107	β^-	>1.2E+15 y
¹⁷³ Hf	72	101	$\epsilon + \beta^+$	23.6 h
¹⁷⁴ Hf	72	102	α	2.0E15 y
¹⁷⁵ Hf	72	103	ϵ	70 d
¹⁷⁷ Hf	72	105	IT	51.4 m
¹⁷⁸ Hf	72	106	IT	31 y
¹⁷⁹ Hf	72	107	IT	25.05 d
¹⁸⁰ Hf	72	108	IT, β^-	5.5 h

3.2. Proton Spallation Yield of W and Pb Target

Neutron spallation source facility for BNCT and industrial application used the proton beam for neutron generation that hit to the W and Pb targets. In

the final step, the neutron production and spallation yield of W and Pb by 200,800 and 1200 MeV proton irradiation were calculated. The spallation yield of W and Pb by 1200 MeV proton was shown in Figures 5 and 6.

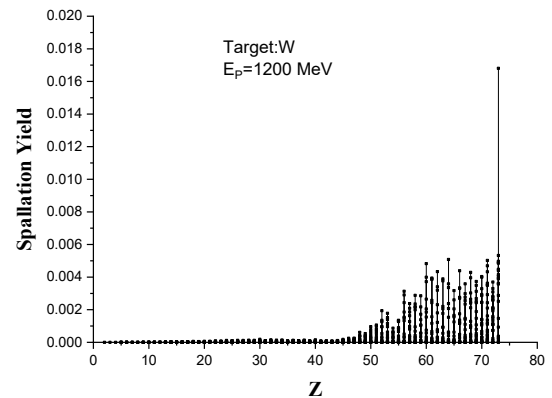


Figure 5. The spallation yield (1/sp) of W by 1200 MeV proton irradiation

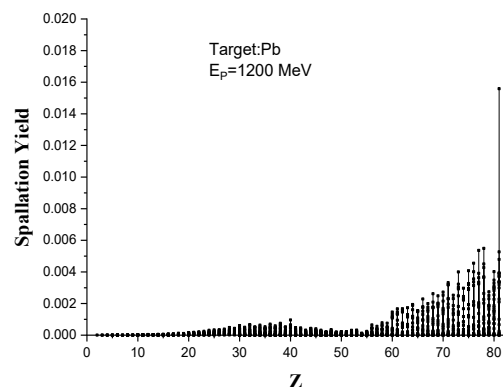


Figure 6. The spallation yield (1/sp) of Pb by 1200 MeV Proton

Figure 5 shows the spallation yield of the elements produced in the tungsten target by proton irradiation with 1200 MeV energy. All produced elements have an atomic number less than 74. The yield of producing elements decreases with decreasing atomic number. The 45 - 74 atomic numbers interval has a higher production yield than the 0-45 atomic numbers range. Also, the spallation yield of elements in each atomic number includes several different isotopes. For example, for 60 atomic number, about 10 isotopes are produced. Also, with the reduction of proton energy, the spallation yield of the produced elements decreases.

Figure 6 shows that the produced elements have an atomic number less than 82. This spectrum of produced elements has two peaks. The first peak is in the atomic number range of 0-50, and the second peak is in the range of 50-82. By decreasing the atomic number from 80 to 55, the production of elements decreases. Also, by decreasing the atomic number from 55 to 40, the yield of element production increases and then decreases for the 0 – 40 interval. With the reduction of proton energy, the yield of element production due to proton spallation decreases. At the same time, several isotopes are produced for each atomic number. The number of isotopes produced for larger atomic numbers is greater than others.

4. Discussion

According to the result, the production yield of ^7Be element is 0.16 that means 16% of the lithium target is used to produce neutrons due to proton irradiation. The ^7Be element produced is a beta-emitter and emits beta radiation after the proton source is turned off. Also, results showed that the ^{179}Ta , $^{180\text{m}}\text{Ta}$ radioactive element and ^{174}Hf with half-lives 1.2×10^{15} y, 1.82 y, and 2×10^{17} year produced in the Ta target by a proton beam that has a high half-life. Other radioactive elements, such as ^{175}Hf , have a shorter half-life of less than 70d. Therefore, cooling of the target after turning off is necessary, and radioactivity of the ^7Be and Ta targets is one of the radiation hazard problems for staff and workers.

According to the Figures 5 and 6, the half-life of radioactive elements produced in the lead and tungsten spallation by 200- 1200 MeV proton irradiation for neutron production was calculated and is shown in Tables 4 and 5.

Table 4 shows the half-life of radioactive elements produced in the proton spallation of a tungsten target. About 150 radioactive elements were produced, of which 37 have a half-life of more than a year, 65 have a half-life of less than a day, and the rest have a half-life of less than an hour. Table 5 shows the half-life of the elements produced as a result of the proton spallation process of lead targets. About 525 radioactive elements with a half-life longer than an hour are listed in this Table. About 250 radioactive elements have a half-life of less than a day, 103

elements have a half-life of more than a year, and 372 radioactive elements have a half-life of less than a year and more than a day. The results of Tables 5 and 6 show that the tungsten and lead targets, for neutron production due to the proton spallation process, will be activated by a wide range of radioactive elements. After turning off the proton source, the radiation emitted in tungsten and lead targets will pose serious risks for the staff. Therefore, when changing targets, this point should be taken into account, and the occurrence of radiation risks should be prevented by installing a suitable shield. If it is necessary to change the target, the protection principles must be fully obeyed. According to the Monte Carlo simulation result, the spallation yield of radioactive production in lead and tungsten by 200-1200 MeV proton was, and the spallation yield of radioactive production in tungsten for 1200 MeV is shown in Figure 7.

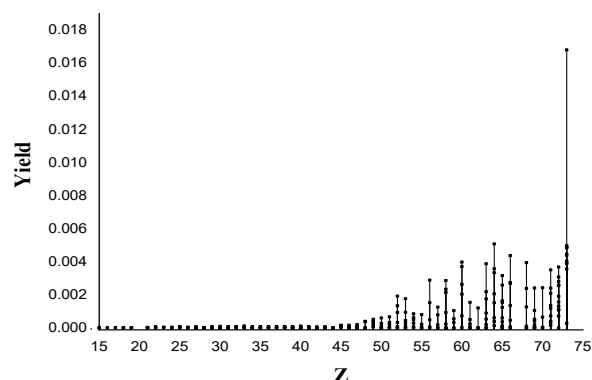


Figure 7. Spallation yield (1/sp) of radioactive production in tungsten by 1200 MeV proton irradiation

Figure 7 shows the benefit of producing radioactive elements as a result of the spallation process with protons with an energy of 1200 MeV in the tungsten target. As it is clear in Figure 7, the radioactive elements with the highest production yield are in the 45-74 atomic number range. It is also clear that the radioactive elements with atomic numbers 15 to 45 have equal spallation yield. The most probable spallation yield of radioactive production in tungsten by 200,800 and 1200 MeV proton was extracted and is shown in Table 6.

The most probable spallation yield of radioactive production in lead by 200,800 and 1200 MeV proton was extracted and is shown in Table 7.

Table 4. Half-life of radioactive elements produced in tungsten spallation by high-energy proton irradiation for neutron production

Radioactive Elements	Half-Life	Radioactive Elements	Half-Life	Radioactive Elements	Half-Life	Radioactive Elements	Half-Life	Radioactive Elements	Half-Life	Radioactive Elements	Half-Life
¹⁸² Os	22.10 h	¹⁶⁹ Yb	32.026 d	⁹⁶ Zr	>3.8E19 y	¹⁴⁵ Sm	340 d	¹⁰⁷ Cd	6.50 h	⁷⁰ Zn	>5E14 y
¹⁸³ Os	13.0 h	¹⁷⁵ Yb	4.185 d	⁹⁷ Zr	16.91 h	¹⁴⁶ Sm	1.03E8 y	¹⁰¹ Pd	8.47 h	^{71m} Zn	3.96 h
^{183m} Os	9.9 h	^{176m} Lu	3.635 h	⁸⁵ Y	2.68 h	¹⁵² Eu	13.537 y	¹⁰³ Pd	16.991 d	⁷² Zn	46.5 h
¹⁷⁴ Ta	1.05 h	¹⁷⁷ Lu	6.734 d	^{85m} Y	4.86 h	^{152m} Eu	9.3116 h	¹⁰⁷ Pd	6.5E6 y	⁶¹ Cu	3.333 h
		^{177m} Lu	160.4 d	⁸⁶ Y	14.74 h	¹⁵⁴ Eu	8.593 y	¹⁰⁹ Pd	13.7012 h	⁶⁴ Cu	12.700 h
¹⁷⁵ Ta	10.5 h	¹⁷⁹ Lu	4.59 h	⁸⁷ Y	79.8 h	¹⁵⁵ Eu	4.7611 y	¹¹² Pd	21.03 h	⁶⁷ Cu	61.83 h
¹⁷⁶ Ta	8.09 h	¹²⁸ Ba	2.43 d	^{87m} Y	13.37 h	¹⁴⁶ Gd	48.27 d	⁹⁹ Rh	16.1 d	⁵⁶ Ni	6.077 d
¹⁷⁷ Ta	56.56 h	¹²³ Xe	2.08 h	⁸⁸ Y	106.65 d	¹⁴⁷ Gd	38.06 h	^{99m} Rh	4.7 h	⁵⁵ Fe	2.73 y
^{178m} Ta	2.36 h	¹²⁴ Xe	>1.6E+14 y	⁹⁰ Y	64.00 h	¹⁴⁸ Gd	74.6 y	¹⁰⁰ Rh	20.8 h	⁵⁹ Fe	44.503 d
^{180m} Hf	5.5 h	¹²⁵ Xe	16.9 h	^{90m} Y	3.19 h	¹⁴⁹ Gd	9.28 d	¹⁰¹ Rh	3.3 y	⁶⁰ Fe	1.5E6 y
¹⁸¹ Hf	42.39 d	¹²⁷ Xe	36.4 d	⁸³ Rb	86.2 d	¹⁵⁰ Gd	1.79E6 y	^{101m} Rh	4.34 d	⁵² Mn	5.591 d
¹⁸² Hf	9E6 y	^{129m} Xe	8.88 d	⁸⁴ Rb	32.77 d	¹⁵¹ Gd	124 d	¹⁰² Rh	207 d	^{44m} Sc	58.6 h
¹⁸³ Hf	1.067 h	¹²¹ I	2.12 h	⁸⁶ Rb	18.631 d	¹⁵² Gd	1.08E14 y	⁹⁹ Tc	2.111E5 y	⁴⁶ Sc	83.79 d
¹³² La	4.8 h 2	^{121m} Te	154 d	⁸⁷ Rb	4.75E10 y	¹⁵³ Gd	240.4 d	^{99m} Tc	6.01 h	⁴⁷ Sc	3.3492 d
¹³³ La	3.912 h	¹²³ Te	>1E13 y	⁷⁶ Kr	14.8 h	¹⁵⁷ Tb	71 y	⁹⁰ Mo	5.56 h	³⁶ Cl	3.01E5 y
¹⁴⁰ Nd	3.37 d	^{123m} Te	119.7 d	⁷⁹ Kr	35.04 h	¹⁵⁸ Tb	180 y	⁹³ Mo	4.0E3 y	³⁵ S	87.32 d
¹⁴¹ Nd	2.49 h	^{125m} Te	57.40 d	⁸¹ Kr	2.29E5 y	¹⁶⁰ Tb	72.3 d	^{93m} Mo	6.85 h	³² P	14.262 d
¹⁴⁴ Nd	2.29E15 y	¹²⁷ Te	9.35 h	⁷⁵ Se	119.779 d	¹⁵² Dy	2.38 h	⁹⁹ Mo	65.94 h	³³ P	25.34 d
¹⁴³ Pm	265 d	¹²⁸ Te	2.2E24 y	⁷⁹ Se	1.13E6 y	¹⁵³ Dy	6.4 h	¹⁰⁰ Mo	1.00E+19 y	³² Si	150 y
¹⁴⁴ Pm	363 d	¹¹⁷ Sb	2.80 h	⁸² Se	1.08E20 y	¹⁶⁷ Tm	9.25 d	⁸⁹ Nb	1.9 h	²⁶ Al	7.17E5 y
¹⁴⁵ Pm	17.7 y	^{118m} Sb	5.00 h	⁷¹ As	65.28 h	¹⁶⁸ Tm	93.1 d	⁸⁶ Zr	16.5 h	²⁸ Mg	20.91 h
¹⁴⁶ Pm	5.53 y	¹¹¹ In	2.8047 d	⁷² As	26.0 h	¹⁷⁰ Tm	128.6 d	⁸⁷ Zr	1.68 h	²² Na	2.6019 y
¹⁴⁷ Pm	2.6234 y	^{113m} In	1.6582 h	⁷³ As	80.30 d	¹⁷¹ Tm	1.92 y	⁸⁸ Zr	83.4 d	²⁴ Na	14.9590 h
¹⁴⁸ Pm	5.370 d	^{114m} In	49.51 d	⁷⁴ As	17.77 d	¹⁷² Tm	63.6 h	⁸⁹ Zr	78.41 h	¹⁴ C	5730 y
^{148m} Pm	41.29 d	¹¹⁵ In	4.41E14 y	⁶⁵ Zn	244.26 d	¹⁷³ Tm	8.24 h	⁹³ Zr	1.53E6 y	¹⁰ Be	1.51E6 y
¹⁴⁹ Pm	53.08 h	^{115m} In	4.486 h	^{69m} Zn	13.76 h	¹⁶⁶ Yb	56.7 h	⁹⁵ Zr	64.02 d		

5. Conclusion

Tables 6 and 7 show that with increasing proton energy, the spallation yield of radioactive elements increases. Also, the atomic and mass numbers of the elements produced with the highest spallation yield increase with increasing proton energy and reach elements with higher mass and atomic numbers. For the tungsten target, ¹⁷⁰Hf, ¹⁸⁴Ta, and ¹⁸⁰Ta elements have the highest spallation yield, and for the lead target, ¹⁹⁹Tl and ¹⁸⁶Pt elements have the highest spallation yield.

A neutron beam was produced by proton irradiation of special targets that were used for the treatment of internal tumors. The proton spallation of W, Li, Be, Ta, Pb targets used in the BNCT therapy process can produce the radioactive elements in these materials. The proton-to-neutron spallation converter materials for the neutron production process can be damaged by this converter. The radioactive elements that exist in proton spallation converters must be considered in the BNCT method. The results showed that the radioactive elements are produced in W, Pb, Li, Be,

Table 5. Half-life of radioactive elements production in lead spallation by high energy proton irradiation for neutron production

Radioactive Elements	Half-Life	Radioactive Elements	Half-Life	Radioactive Elements	Half-Life	Radioactive Elements	Half-Life	Radioactive Elements	Half-Life	Radioactive Elements	Half-Life
²⁰² Bi	1.72 h	¹⁴⁸ Sm	7E+15 y	¹⁰³ Pd	16.991 d	¹⁸⁹ Re	24.3 h	^{129m} Xe	8.88 d	⁷⁴ As	17.77 d
²⁰³ Bi	11.76 h	¹⁴⁹ Sm	>2E+15 y	¹⁰⁷ Pd	6.5E6 y	¹⁷⁶ W	2.5 h	^{131m} Xe	11.84 d	⁷⁶ As	1.0778 d
²⁰⁴ Bi	11.22 h	¹⁵¹ Sm	90 y	¹⁰⁹ Pd	13.7012 h	¹⁷⁸ W	21.6 d	¹³³ Xe	5.243 d	⁷⁷ As	38.83 h
²⁰⁵ Bi	15.31 d	¹⁵³ Sm	46.284 h	^{111m} Pd	5.5 h	¹⁸¹ W	121.2 d	^{133m} Xe	2.19 d	⁶⁶ Ge	2.26 h
²⁰⁶ Bi	6.243 d	¹⁵⁶ Sm	9.4 h	¹¹² Pd	21.03 h	¹⁸³ W	>1.1E17 y	¹³⁵ Xe	9.14 h	⁶⁸ Ge	270.8 d
²⁰⁷ Bi	31.55 y	¹⁴⁵ Eu	5.93 d	⁹⁹ Rh	16.1 d			¹³⁶ Xe	2.36E21 y	⁶⁹ Ge	39.05 h
²⁰⁸ Bi	3.68E5 y	¹⁴⁶ Eu	4.61 d	^{99m} Rh	4.7 h	¹⁸⁴ W	>3E17 y	¹²¹ I	2.12 h	⁷¹ Ge	11.43 d
¹⁹⁸ Pb	2.40 h	¹⁴⁷ Eu	24.1 d	¹⁰⁰ Rh	20.8 h	¹⁸⁵ W	75.1 d	¹²³ I	13.27 h	⁷⁷ Ge	11.30 h
²⁰⁰ Pb	21.5 h	¹⁴⁸ Eu	54.5 d	¹⁰¹ Rh	3.3 y	¹⁸⁷ W	23.72 h	¹²⁴ I	4.1760 d	⁶⁶ Ga	9.49 h
²⁰¹ Pb	9.33 h	¹⁴⁹ Eu	93.1 d	^{101m} Rh	4.34 d	¹⁸⁸ W	69.4 d	¹²⁵ I	59.408 d	⁶⁷ Ga	3.2612 d
²⁰² Pb	5.25E4 y	¹⁵⁰ Eu	36.9 y	¹⁰² Rh	207 d	¹⁷³ Ta	3.14 h	¹²⁶ I	13.11 d	⁷² Ga	14.10 h
^{202m} Pb	3.53 h	^{150m} Eu	12.8 h	^{102m} Rh	~2.9 y	¹⁷⁴ Ta	1.05 h	¹²⁹ I	1.57E7 y	⁷³ Ga	4.86 h
²⁰³ Pb	51.873 h	¹⁵² Eu	13.537 y	¹⁰⁵ Rh	35.36 h			¹³⁰ I	12.36 h	⁶² Zn	9.186 h
²⁰⁴ Pb	>1.4E17 y	^{152m} Eu	9.3116 h	⁹⁵ Ru	1.643 h	¹⁷⁵ Ta	10.5 h	¹³¹ I	8.02070 d	⁶⁵ Zn	244.26 d
²⁰⁵ Pb	1.53E7 y	¹⁵⁴ Eu	8.593 y	⁹⁷ Ru	2.9 d	¹⁷⁶ Ta	8.09 h	¹³² I	2.295 h	^{69m} Zn	13.76 h
¹⁹⁵ Tl	1.16 h	¹⁵⁵ Eu	4.7611 y	¹⁰³ Ru	39.26 d	¹⁷⁷ Ta	56.56 h	^{132m} I	1.387 h	⁷⁰ Zn	>5E14 y
¹⁹⁶ Tl	1.84 h	¹⁵⁶ Eu	15.19 d	¹⁰⁵ Ru	4.44 h	^{178m} Ta	2.36 h	¹³³ I	20.8 h	^{71m} Zn	3.96 h
^{196m} Tl	1.41 h	¹⁵⁷ Eu	15.18 h	¹⁰⁶ Ru	373.59 d	¹⁷⁹ Ta	1.82 y	¹³⁵ I	6.57 h	⁷² Zn	46.5 h
¹⁹⁷ Tl	2.84 h	¹⁴⁶ Gd	48.27 d	⁹³ Tc	2.75 h	¹⁸⁰ Ta	8.152 h	¹¹⁶ Te	2.49 h	⁶¹ Cu	3.333 h
¹⁹⁸ Tl	5.3 h	¹⁴⁷ Gd	38.06 h	⁹⁵ Tc	20.0 h	^{180m} Ta	>1.2E15 y	¹¹⁸ Te	6.00 d	⁶⁴ Cu	12.700 h
^{198m} Tl	1.87 h	¹⁴⁸ Gd	74.6 y	^{95m} Tc	61 d	¹⁸² Ta	114.43 d	¹¹⁹ Te	16.03 h	⁶⁷ Cu	61.83 h
¹⁹⁹ Tl	7.42 h	¹⁴⁹ Gd	9.28 d	⁹⁶ Tc	4.28 d	¹⁷⁰ Hf	16.01 h	^{119m} Te	4.70 d	⁵⁶ Ni	6.077 d
²⁰⁰ Tl	26.1 h	¹⁵⁰ Gd	1.79E6 y	⁹⁷ Tc	2.6E6 y	¹⁷¹ Hf	12.1 h	¹²¹ Te	16.78 d	⁵⁷ Ni	35.60 h
²⁰¹ Tl	72.912 h	¹⁵¹ Gd	124 d	^{97m} Tc	90.1 d	¹⁷² Hf	1.87 y	^{121m} Te	154 d	⁵⁹ Ni	7.6E4 y
²⁰² Tl	12.23 d	¹⁵² Gd	1.08E14 y	⁹⁸ Tc	4.2E6 y	¹⁷³ Hf	23.6 h	¹²³ Te	>1E+13 y	⁶³ Ni	100.1 y
²⁰⁴ Tl	3.78 y	¹⁵³ Gd	240.4 d	⁹⁹ Tc	2.111E5 y	¹⁷⁴ Hf	2.0E15 y	^{123m} Te	119.7 d	⁶⁵ Ni	2.5172 h
¹⁹² Hg	4.85 h	¹⁴⁷ Tb	1.7 h	^{99m} Tc	6.01 h	¹⁷⁵ Hf	70 d	^{125m} Te	57.40 d	⁶⁶ Ni	54.6 h
¹⁹³ Hg	3.80 h	¹⁴⁹ Tb	4.118 h	⁹⁰ Mo	5.56 h	^{178m} Hf	31 y	¹²⁷ Te	9.35 h	⁵⁵ Co	17.53 h
^{193m} Hg	11.8 h	¹⁵⁰ Tb	3.48 h	⁹³ Mo	4.0E3 y	^{179m} Hf	25.05 d	^{127m} Te	109 d	⁵⁶ Co	77.27 d
¹⁹⁴ Hg	444 y	¹⁵¹ Tb	17.609 h	^{93m} Mo	6.85 h	¹⁸² Hf	9E6 y	¹²⁸ Te	2.2E24 y	⁵⁷ Co	271.79 d
¹⁹⁵ Hg	9.9 h	¹⁵² Tb	17.5 h	⁹⁹ Mo	65.94 h	¹⁸³ Hf	1.067 h	^{129m} Te	33.6 d	⁵⁸ Co	70.86 d
^{195m} Hg	41.6 h	¹⁵³ Tb	2.34 d	¹⁰⁰ Mo	1.00E19 y	¹⁸⁴ Hf	4.12 h	¹³⁰ Te	7.9E20 y	^{58m} Co	9.04 h
¹⁹⁷ Hg	64.14 h	¹⁵⁴ Tb	21.5 h	⁸⁹ Nb	1.9 h	¹³² La	4.8 h	^{131m} Te	30 h	⁶⁰ Co	5.2714 y
^{197m} Hg	23.8 h	^{154m} Tb	9.4 h	^{89m} Nb	1.18 h	¹³³ La	3.912 h	¹³² Te	3.204 d	⁶¹ Co	1.650 h
²⁰³ Hg	46.612 d	^{154m2} Tb	22.7 h	⁹⁰ Nb	14.60 h	¹³⁵ La	19.5 h	¹¹⁷ Sb	2.80 h	⁵² Fe	8.275 h
¹⁹¹ Au	3.18 h	¹⁵⁵ Tb	5.32 d	⁹¹ Nb	680 y	¹³⁷ La	6E4 y	^{118m} Sb	5.00 h	⁵⁵ Fe	2.73 y
^{191m} Au	0.92 s	¹⁵⁶ Tb	5.35 d	^{91m} Nb	60.86 d	¹³⁸ La	1.05E11 y	¹¹⁹ Sb	38.19 h	⁵⁹ Fe	44.503 d
¹⁹² Au	4.94 h	^{156m} Tb	24.4 h	⁹² Nb	3.47E7 y	¹⁴⁰ La	1.6781 d	^{120m} Sb	5.76 d	⁶⁰ Fe	1.5E6 y
¹⁹³ Au	17.65 h	^{156m2} Tb	5.3 h	^{92m} Nb	10.15 d	¹⁴¹ La	3.92 h	¹²² Sb	2.7238 d	⁵² Mn	5.591 d

¹⁹⁴ Au	38.02 h	¹⁵⁷ Tb	71 y	^{93m} Nb	16.13 y	¹³² Ce	3.51 h	¹²⁴ Sb	60.20 d	⁵³ Mn	3.74E6 y
¹⁹⁵ Au	186.09 d	¹⁵⁸ Tb	180 y	⁹⁴ Nb	2.03E4 y	^{133m} Ce	4.9 h	¹²⁵ Sb	2.7582 y	⁵⁴ Mn	312.3 d
¹⁹⁶ Au	6.183 d	¹⁶⁰ Tb	72.3 d	⁹⁵ Nb	34.975 d	¹³⁴ Ce	3.16 d	¹²⁶ Sb	12.46 d	⁵⁶ Mn	2.5785 h
^{196m} Au	9.6 h	¹⁶¹ Tb	6.88 d	^{95m} Nb	86.6 h	¹³⁵ Ce	17.7 h	¹²⁷ Sb	3.85 d	⁴⁸ Cr	21.56 h
¹⁹⁸ Au	2.69517 d	¹⁵² Dy	2.38 h	⁹⁶ Nb	23.35 h	¹³⁷ Ce	9.0 h	¹²⁸ Sb	9.01 h	⁵⁰ Cr	>1.8E17 y
^{198m} Au	2.27 d	¹⁵³ Dy	6.4 h	⁸⁶ Zr	16.5 h	^{137m} Ce	34.4 h	¹²⁹ Sb	4.40 h	⁵¹ Cr	27.7025 d
¹⁹⁹ Au	3.139 d	¹⁵⁴ Dy	3.0E+6 y	⁸⁷ Zr	1.68 h	¹³⁹ Ce	137.640 d	¹¹⁰ Sn	4.11 h	⁴⁸ V	15.9735 d
^{200m} Au	18.7 h	¹⁵⁵ Dy	9.9 h	⁸⁸ Zr	83.4 d	¹⁴¹ Ce	32.501 d	¹¹³ Sn	115.09 d	⁴⁹ V	330 d
¹⁸⁶ Pt	2.2 h	¹⁵⁷ Dy	8.14 h	⁸⁹ Zr	78.41 h	¹⁴² Ce	>5E16 y	^{117m} Sn	13.60 d	⁵⁰ V	1.4E17 y
¹⁸⁷ Pt	2.35 h	¹⁵⁹ Dy	144.4 d	⁹³ Zr	1.53E6 y	¹⁴³ Ce	33.039 h	^{119m} Sn	293.1 d	⁴⁴ Ti	63 y
¹⁸⁸ Pt	10.2 d	¹⁶⁵ Dy	2.334 h	⁹⁵ Zr	64.02 d	¹⁴⁴ Ce	284.893 d	¹²¹ Sn	27.06 h	⁴³ Sc	3.891 h
¹⁸⁹ Pt	10.87 h	¹⁶⁶ Dy	81.6 h	⁹⁶ Zr	>3.8E19 y	¹³⁷ Pr	1.28 h	^{121m} Sn	55 y	⁴⁴ Sc	3.927 h
¹⁹⁰ Pt	6.5E11 y	¹⁵⁸ Er	2.29 h	⁹⁷ Zr	16.91 h	^{138m} Pr	2.12 h	¹²³ Sn	129.2 d	^{44m} Sc	58.6 h
¹⁹¹ Pt	2.802 d	¹⁶⁰ Er	28.58 h	⁸⁵ Y	2.68 h	¹³⁹ Pr	4.41 h	¹²⁵ Sn	9.64 d	⁴⁶ Sc	83.79 d
¹⁹³ Pt	50 y	¹⁶¹ Er	3.21 h	^{85m} Y	4.86 h	¹⁴² Pr	19.12 h	¹²⁶ Sn	~1E+5 y	⁴⁷ Sc	3.3492 d
^{193m} Pt	4.33 d	¹⁶⁵ Er	10.36 h	⁸⁶ Y	14.74 h	¹⁴³ Pr	13.57 d	¹²⁷ Sn	2.10 h	⁴⁸ Sc	43.67 h
^{195m} Pt	4.02 d	¹⁶³ Tm	1.810 h	⁸⁷ Y	79.8 h	¹⁴⁵ Pr	5.984 h	¹⁰⁹ In	4.2 h	⁴¹ Ca	1.03E5 y
¹⁹⁷ Pt	19.8915 h	¹⁶⁵ Tm	30.06 h	^{87m} Y	13.37 h			¹¹⁰ In	4.9 h	⁴⁵ Ca	162.61 d
²⁰⁰ Pt	12.5 h	¹⁶⁶ Tm	7.70 h	⁸⁸ Y	106.65 d	¹³⁸ Nd	5.04 h	¹¹¹ In	2.8047 d	⁴⁷ Ca	4.536 d
¹⁸⁴ Ir	3.09 h	¹⁶⁷ Tm	9.25 d	⁹⁰ Y	64.00 h	^{139m} Nd	5.50 h	^{113m} In	1.6582 h	⁴⁸ Ca	>6E18 y
¹⁸⁵ Ir	14.4 h	¹⁶⁸ Tm	93.1 d	^{90m} Y	3.19 h	¹⁴⁰ Nd	3.37 d	^{114m} In	49.51 d	⁴⁰ K	1.277E9 y
¹⁸⁶ Ir	16.64 h	¹⁷⁰ Tm	128.6 d	⁹¹ Y	58.51 d	¹⁴¹ Nd	2.49 h	¹¹⁵ In	4.41E14 y	⁴² K	12.360 h
^{186m} Ir	1.90 h	¹⁷³ Tm	8.24 h	⁹² Y	3.54 h	¹⁴⁴ Nd	2.29E15 y	^{115m} In	4.486 h	⁴³ K	22.3 h
¹⁸⁷ Ir	10.5 h	¹⁶⁹ Yb	32.026 d	⁹³ Y	10.18 h	¹⁴⁷ Nd	10.98 d	¹⁰⁷ Cd	6.50 h	³⁷ Ar	35.04 d
¹⁸⁸ Ir	41.5 h	¹⁶⁶ Yb	56.7 h	⁸² Sr	25.55 d	¹⁴⁹ Nd	1.728 h	¹⁰⁹ Cd	462.6 d	³⁹ Ar	269 y
¹⁸⁹ Ir	13.2 d	¹⁶⁹ Lu	34.06 h	⁸³ Sr	32.41 h	¹⁵⁰ Nd	>1.1E19 y	¹¹³ Cd	7.7E+15 y	⁴² Ar	32.9 y
¹⁹⁰ Ir	11.78 d	¹⁷⁰ Lu	2.012 d	⁸⁵ Sr	64.84 d	¹⁴³ Pm	265 d	^{113m} Cd	14.1 y	³⁶ Cl	3.01E5 y
^{190m} Ir	1.2 h	¹⁷¹ Lu	8.24 d	^{87m} Sr	2.803 h	¹⁴⁴ Pm	363 d	¹¹⁵ Cd	53.46 h	³⁵ S	87.32 d
^{190m} Ir	3.25 h	¹⁷² Lu	6.70 d	⁸⁹ Sr	50.53 d	¹⁴⁵ Pm	17.7 y	^{115m} Cd	44.6 d	³² P	14.262 d
¹⁹² Ir	73.831 d	¹⁷³ Lu	1.37 y	⁹⁰ Sr	28.79 y	¹⁴⁶ Pm	5.53 y	¹¹⁷ Cd	2.49 h	³³ P	25.34 d
^{192m} Ir	241 y	¹⁷⁴ Lu	3.31 y	⁹¹ Sr	9.63 h	¹⁴⁷ Pm	2.6234 y	^{117m} Cd	3.36 h	³² Si	150 y
^{193m} Ir	10.53 d	^{174m} Lu	142 d	⁹² Sr	2.71 h	¹⁴⁸ Pm	5.370 d	^{106m} Ag	8.28 d	²⁶ Al	7.17E5 y
¹⁹⁴ Ir	19.28 h	¹⁷⁶ Lu	3.78E10 y	⁸¹ Rb	4.576 h	^{148m} Pm	41.29 d	^{108m} Ag	418 y	²⁸ Mg	20.91 h
^{194m} Ir	171 d	^{176m} Lu	3.635 h	^{82m} Rb		¹⁴⁹ Pm	53.08 h	^{110m} Ag	249.79 d	²² Na	2.6019 y
¹⁹⁵ Ir	2.5 h	¹⁷⁷ Lu	6.734 d	⁸³ Rb	86.2 d	¹⁵⁰ Pm	2.68 h	¹¹¹ Ag	7.45 d	²⁴ Na	14.9590 h
^{195m} Ir	3.8 h	^{177m} Lu	160.4 d	⁸⁴ Rb	32.77 d	¹⁵¹ Pm	28.40 h	¹¹² Ag	3.130 h	¹⁴ C	5730 y
^{196m} Ir	1.40 h	¹²⁸ Ba	2.43 d	⁸⁶ Rb	18.631 d	¹⁴⁵ Sm	340 d	¹¹³ Ag	5.37 h	⁷ Be	53.12 d
¹⁸² Os	22.10 h	¹²⁹ Ba	2.23 h	⁸⁷ Rb	4.75E10 y	¹⁴⁶ Sm	1.03E8 y	¹⁰⁰ Pd	3.63 d		
¹⁸³ Os	13.0 h	^{129m} Ba	2.16 h	⁷⁶ Kr	14.8 h	¹⁴⁷ Sm	1.06E11 y	¹⁰¹ Pd	8.47 h	¹⁰ Be	1.51E6 y
^{183m} Os	9.9 h	¹³¹ Ba	11.50 d	⁷⁹ Kr	35.04 h	¹⁸² Re	64.0 h	^{134m} Cs	2.903 h	⁸³ Br	2.40 h
¹⁸⁴ Os	>5.6E13 y	¹³³ Ba	10.51 y	⁸¹ Kr	2.29E5 y	^{182m} Re	12.7 h	¹³⁵ Cs	2.3E+6 y	⁷² Se	8.40 d
¹⁸⁵ Os	93.6 d	^{133m} Ba	38.9 h	⁸⁵ Kr	10.756 y	¹⁸³ Re	70.0 d	¹³⁶ Cs	13.16 d	⁷³ Se	7.15 h
¹⁸⁶ Os	2.0E15 y	^{135m} Ba	28.7 h	^{85m} Kr	4.480 h	¹⁸⁴ Re	38.0 d	¹³⁷ Cs	30.07 y	⁷⁵ Se	119.779 d
^{189m} Os	5.8 h	¹⁴⁰ Ba	12.752 d	⁸⁸ Kr	2.84 h	^{184m} Re	169 d	¹²² Xe	20.1 h	⁷⁹ Se	1.13E6 y
¹⁹¹ Os	15.4 d	¹²⁷ Cs	6.25 h	⁷⁶ Br	16.2 h	¹⁸⁶ Re	3.7183 d	¹²³ Xe	2.08 h	⁸² Se	1.08E20 y

^{191m} Os	13.10 h	¹²⁹ Cs	32.06 h			^{186m} Re	2.0E+5 y	¹²⁴ Xe	1.6E+14 y	⁷¹ As	65.28 h
¹⁹³ Os	30.11 h	¹³¹ Cs	9.689 d	⁷⁷ Br	57.036 h	¹⁸⁷ Re	4.35E10 y	¹²⁵ Xe	16.9 h	⁷² As	26.0 h
¹⁹⁴ Os	6.0 y	¹³² Cs	6.479 d	^{80m} Br	4.4205 h	¹⁸⁸ Re	17.005 h	¹²⁷ Xe	36.4 d	⁷³ As	80.30 d
¹⁸¹ Re	19.9 h	¹³⁴ Cs	2.0648 y	⁸² Br	35.30 h						

Table 6. The most probable spallation yield of radioactive production (SPRP) in tungsten by 200,800 and 1200 MeV proton (E_p) irradiation

Z	N	SPRP for $E_p=200$ MeV	Z	N	SPRP for $E_p=800$ MeV	Z	N	SPRP for $E_p=1200$ MeV
72	98	0.00409	73	111	0.018218	73	110	0.016805
72	100	0.003931	73	101	0.007474	64	82	0.005084
72	101	0.003341	73	103	0.007048	73	109	0.004973
72	102	0.003286	73	109	0.00589	73	102	0.00489
72	99	0.003263	73	105	0.005878	73	100	0.004855
72	103	0.002551	72	99	0.005794	73	104	0.00448
71	98	0.002075	73	100	0.005722	73	106	0.004398
72	106	0.001758	73	102	0.005659	66	86	0.004387
71	99	0.001485	71	98	0.005576	73	107	0.004053
71	100	0.001347	73	107	0.005507	60	78	0.003994
72	107	0.00112	71	99	0.005458	68	90	0.003958
72	108	0.000922	66	87	0.005139	73	101	0.003928
71	101	0.000906	73	104	0.005093	73	105	0.003896
71	102	0.000696	73	106	0.004884	63	82	0.003887
72	109	0.00049	73	110	0.004849	60	77	0.003718
70	96	0.000444	72	101	0.004471	72	98	0.003694
71	103	0.000409	64	83	0.004404	64	84	0.003586
72	110	0.000291	72	98	0.004395	73	103	0.003574
71	105	0.00016	68	90	0.004232	71	98	0.003517
70	99	0.000152	72	100	0.004107	64	83	0.003354
71	106	0.000145	69	94	0.003921	65	84	0.003175
69	94	0.000102	66	86	0.003789	72	100	0.00309

and Ta targets and must be avoided during radiation hazards calculation. The simulation results show that proton spallation of W and Pb targets produced the (¹⁷⁰Hf, ¹⁸⁴Ta, ¹⁸⁰Ta) and (¹⁹⁹Tl, ¹⁸⁶Pt) radioactive elements with high spallation yield, respectively, which should be considered as radioactive hazards for these targets. With increased energy of the proton beam, the amount of production of the radioactive elements was increased. Also, the Monte Carlo result shows that the production of radioactive elements in reactions with secondary neutrons is lower than in reactions with the primary proton beam.

Table 7. The most probable spallation yield of radioactive production in Lead by 200,800 and 1200 MeV protons

Z	N	SPRP for E _p =200 MeV	Z	N	SPRP for E _p =800 MeV	Z	N	SPRP for E _p =1200 MeV
81	118	0.012081	78	108	0.008673	78	108	0.005488
81	120	0.010675	78	109	0.006797	78	109	0.004507
81	116	0.010262	78	110	0.006741	78	110	0.004274
81	119	0.009813	80	114	0.00612	80	114	0.004019
81	117	0.00969	80	112	0.005897	81	118	0.003936
81	121	0.008471	81	118	0.005782	81	120	0.003922
81	115	0.007583	77	108	0.00567	77	108	0.003914
81	114	0.007011	81	116	0.005622	81	116	0.003731
81	123	0.00642	81	114	0.005378	81	123	0.00371
80	114	0.003784	81	120	0.005118	80	112	0.003481
80	115	0.002871	80	113	0.004956	81	121	0.00338
80	117	0.00266	81	117	0.004851	81	119	0.003366
80	113	0.002614	81	119	0.004749	81	114	0.003343
80	112	0.002609	81	115	0.004678	73	100	0.003225
80	123	0.001019	80	115	0.004343	81	117	0.003193
79	112	0.000697	81	121	0.004325	80	113	0.003098
79	114	0.000602	81	123	0.004239	81	115	0.002986
79	113	0.000555	78	111	0.004023	80	115	0.002823
79	115	0.000399	79	112	0.003958	78	111	0.002636
79	116	0.000368	73	100	0.003512	68	90	0.002618
78	110	0.000367	80	117	0.003351	79	112	0.00251

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