ACCUMULATION OF ACTIVATION PRODUCTS IN PB-BI, TANTALUM, AND TUNGSTEN TARGETS OF ADS

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Abstract

Data on new radionuclide production in three types of target, Pb-Bi, tantalum, and tungsten target of ADS are presented in this paper. The irradiation by neutrons produced in blanket and in the target itself do not take into account proton irradiation. The change of isotopic composition, accumulation of new radionuclides, and radiation characteristics (activity, radiotoxicity is water, and radiation dose power) are calculated.

1. Introduction

One of the main parts of the accelerator driven system (ADS) is the neutron-producing target. It can be made of solid heavy metal such as tantalum or tungsten or liquid metal such as lead-bismuth. All these materials are typical for a neutron-producing target. ADS target is irradiated by accelerated protons, by high-energy neutrons from the target itself, and by low energy neutrons from the sub-critical blanket surrounding the target. The average energy of neutrons from the target itself is about several MeV. A flux density of neutrons from the blanket on the target is of the order of 10^{13} $cm^{-2}s^{-1}$ for common-type power blanket with thermal neutrons and can reach several units times 10^{15} $cm^{-2}s^{-1}$ for high flux blanket. Under influence of target irradiation, there are nuclide conversions causing the change of target isotopic composition and radioactive nuclei production.

The change of isotopic composition, accumulation of new radionuclides, and radiation characteristics (activity, radiotoxicity by water, and radiation dose power) caused by external neutrons from blanket and by internal neutrons from the target itself are calculated. The influence of protons on side nuclide production should be calculated separately and is not considered in the paper.

In calculating nuclide conversions by thermal neutrons, reaction rates A_i are taken using values of thermal neutron cross-sections σ_i and resonance integrals I_i of nuclides [1], $A_i = (\sigma_i+\gamma I_i)\Phi$, where Φ is neutron flux density, γ is neutron spectrum hardness showing a ratio of epithermal to thermal neutrons. Value $\gamma = 0.4$ (spectrum typical for light-water thermal-neutron blanket) is considered. For irradiation by internal neutrons from target, monoenergetic neutrons with energies 10 MeV are considered [2,3]. The average high-energy neutron flux density is over the volume of a target about 10^{15} $cm^{-2}s^{-1}$ at energy of protons from accelerator 1 GeV and beam current 10 mA. Corresponding thermal neutron flux from blanket is about 10^{14} $cm^{-2}s^{-1}$. It was accepted that the target has the form of a continuous cylinder with a diameter of 50 cm. Specific activity Q (Ci/g), radiotoxicity RT (litre/g) and radiation dose power $Q\Gamma$ ($R\cdot cm^2/g\cdot hr$) are defined by sums on all radioactive nuclides included in a target:

$$Q = \Sigma\, Q_i$$
$$RT = \Sigma\, RT_i,\ RT_i = Q_i / MPA_i,$$
$$Q\Gamma = \Sigma\, Q\Gamma_i$$

where MPA_i – maximum permissible activity of the given nuclide i in water determined by the modern Russian radiation safety standard [4], Γ_i – gamma-constant of the nuclide i. They are referred to 1 gram of a target.

2. Pb-Bi target irradiation

The initial target is Pb-Bi eutectic containing 44.5% Pb and 55.5% Bi with natural isotopic composition. In Tables 1 and 2, nuclide concentration and radiation characteristics of a target are presented. For low energy neutrons, three Φ values and $\gamma = 0.4$ are considered. Concentrations of nuclides are normalised by 0.445 nuclei of lead and 0.555 nuclei of bismuth in initial target. Only the most important nuclides are submitted in the tables. Radiation characteristics – activity *Q*, radiotoxicity RT and radiation doze power $Q\Gamma$ are referred to 1 gram of a target. Irradiation time T = 1 year.

In irradiation by thermal neutrons, capture cross sections of nuclides in target are very small. So, effects of thermal self-blocking of Pb-Bi target are not essential. Initial nuclide burning is negligible.

An important radionuclide determining the radiation characteristics of a target is ^{210}Po. Because of alpha-decay of this nuclide, radiotoxicity is high. However, radiation doze power is not so great as for other target materials because of low gamma-radiation of ^{210}Po. In high-energy neutron irradiation, ^{210}Po is the main radioactive nuclide, and ^{204}Tl gives also small contribution to radiation characteristics.

Table 1. **Nuclide concentration and radiation characteristics of Pb-Bi target irradiated by external low energy neutrons from blanket**

Nuclide	**Initial concentrations**	**T = 1 yr**		
		$\Phi=10^{13}$ cm^{-2}s^{-1}	$\Phi=10^{14}$ cm^{-2}s^{-1}	$\Phi=10^{15}$ cm^{-2}s^{-1}
^{204}Pb	0.0064	6.4-3	6.4-3	6.1-3
^{205}Pb	0	2.95-6	2.94-5	2.88-4
^{206}Pb	0.107	0.107	0.107	0.107
^{207}Pb	0.0983	0.0983	0.0981	0.0959
^{208}Pb	0.233	0.233	0.233	0.235
^{209}Bi	0.555	0.555	0.555	0.553
^{210}Po	0	6.32-6	6.31-5	6.30-4
Q, Ci/g	–	0.0289	0.289	2.88
RT, litre/g	–	8.90 + 9	8.90 + 10	8.88 + 11
QΓ, (R cm^2/g hr)	–	1.54-3	0.0154	0.153

Table 2. **Nuclide concentrations and radiation characteristics of a target irradiated by 10 MeV neutrons**

Nuclide	**Initial concentrations**	**T = 1 yr**	
		$\varphi = 10^{15}$ cm^{-2}s^{-1}	$\varphi = 10^{16}$ cm^{-2}s^{-1}
^{204}Hg	0	2.38-10	2.38-9
^{204}Pb	0.0064	0.0064	0.0064
^{204}Tl	0	1.11-7	1.11-6
^{206}Pb	0.107	0.107	0.107
^{207}Pb	0.0983	0.0983	0.0983
^{208}Pb	0.233	0.233	0.233
^{209}Bi	0.555	0.555	0.555
^{210}Po	0	1.61-5	1.61-4
Q, Ci/g	–	0.0735	0.753
RT, litre/g	–	2.27 + 10	2.27 + 11
QΓ, (R cm^2/g hr)	–	4.19-3	4.19-2

The comparison of radiation characteristics caused by neutrons from the target itself and neutrons from external blanket is based on the assumption that a high energy neutron flux density of 10^{15} cm^{-2}s^{-1} corresponds to a neutron flux from external blanket of 10^{14} cm^{-2}s^{-1}. The radiotoxicity caused by neutrons from the target itself is about 3 times less, and radiation dose power is 4 times less than the same characteristics caused by neutrons from an external blanket. This result is important as it shows a rather high role of neutrons from the target itself in the process of accumulation of those radionuclides which define the main radiation characteristics of the irradiated target.

3. Tantalum target irradiation

The initial target is made of natural tantalum. In Table 3, nuclide concentration and radiation characteristics are presented for low energy neutron irradiation. For $\Phi = 10^{14}$ cm^{-2}s^{-1}, value $\gamma = 0.1$ and for $\Phi = 10^{15}$ cm^{-2}s^{-1} value $\gamma = 0$ are considered with thin target, so effects of self-blocking are weak. Radiation characteristics are determined by ^{182}Ta and, at some extend, by ^{185}W.

Table 3. **Nuclide concentration and radiation characteristics of tantalum target irradiated by external low energy neutrons from blanket**

Nuclide	Initial concentrations	T = 1 yr	T = 0.5 yr
		$\Phi = 10^{14}$ cm^{-2}s^{-1}	$\Phi = 10^{15}$ cm^{-2}s^{-1}
^{181}Ta	1.0	0.761	0.724
^{182}Ta	0	4.40-3	1.04-3
^{182}W	0	9.56-3	1.13-3
^{183}W	0	0.210	0.252
^{184}W	0	0.015	0.021
^{185}W	0	2.62-5	1.37-4
Q, Ci/g	–	27.8	7.84
RT, litre/g	–	1.1+10	2.7+10
QΓ, (R cm^2/g hr)	–	1.85+5	4.37+4

4. Tungsten target irradiation

In Table 3, partial nuclide introduction to radiation characteristics is shown for $\Phi = 10^{14}$ cm^{-2}s^{-1}. In Table 4, concentration of nuclides and radiation characteristics of a target are presented for low energy neutron irradiation. Concentrations of nuclides are normalised by one nucleus of natural tungsten, radiation characteristics by 1 gram of a target. For neutron flux $\Phi = 10^{14}$ cm^{-2}s^{-1}, value $\gamma = 0.4$ is considered, and for $\Phi = 10^{15}$ cm^{-2}s^{-1}, $\gamma = 0$ and T = 0.5 years is taken. Target diameter 50 cm. Only the most important nuclides are submitted.

Table 3. **Radiation characteristics of tungsten target irradiated by low energy neutrons from blanket with $\Phi = 10^{14}$ cm^{-2}s^{-1}**

Nuclide	Q, Ci/g	RT, litre/g	QΓ, (R cm^2/g hr)
^{181}W	0.703	1.4 + 7	150
^{185}W	5.68	6.5 + 8	1.6
^{187}W	21.9	–	5.3 + 4
^{188}W	0.297	1.7 + 8	3.3
^{182}Ta	0.703	2.8 + 8	470
^{183}Ta	0.0486	1.6 + 7	80
^{186}Re	1.46	5.8 + 8	140
^{188}Re	2.30	–	730
Total	32.4	1.7 + 9	5.5 + 4

Table 4. **Nuclide concentration and radiation characteristics of tungsten target irradiated by external neutrons from blanket**

Nuclide	Initial concentrations	T = 1 yr	T = 0.5 yr
		$\Phi = 10^{14}$ cm^{-2}s^{-1}	$\Phi = 10^{15}$ cm^{-2}s^{-1}
^{180}W	0.126-2	9.6-4	1.3-3
^{181}W	0	1.2-4	6.6-6
^{182}W	0.263	0.26	0.26
^{183}W	0.143	0.14	0.14
^{184}W	0.306	0.31	0.31
^{185}W	0	6.0-4	7.1-5
^{186}W	0.286	0.28	0.28
^{187}W	0	3.1-5	2.4-5
^{188}W	0	2.9-5	2.0-7
^{182}Ta	0	1.1-5	4.0-9
^{183}Ta	0	3.5-7	3.6-10
^{186}Re	0	7.8-6	6.7-8
^{188}Re	0	2.3-6	3.5-7
Q, Ci/g	–	32.4	18.1
RT, litre/g	–	1.7 + 9	8.5 + 7
QΓ, (R cm^2/g hr)	–	5.5 + 4	4.1 + 4

Table 5. **Nuclide concentrations and radiation characteristics of a tungsten target irradiated by 10 MeV neutrons**

Nuclide	Initial concentrations	T = 1 yr	T = 0.5 yr
		$\varphi = 10^{15}$ cm^{-2}s^{-1}	$\varphi = 10^{16}$ cm^{-2}s^{-1}
^{180}W	1.3-3	1.4-3	2.6-3
^{181}W	0	4.2-3	0.030
^{182}W	0.263	0.26	0.25
^{183}W	0.143	0.15	0.17
^{184}W	0.306	0.29	0.24
^{185}W	0	4.0-3	0.03
^{186}W	0.286	0.27	0.22
^{187}W	0	3.0-8	2.5-7
^{182}Ta	0	1.0-7	1.9-6
^{186}Re	0	3.2-7	1.0-6
^{188}Re	0	4.7-12	2.0-10
Q, Ci/g	–	62.2	486
RT, litre/g	–	5.1 + 9	4.1 + 10
QΓ (R cm^2/g hr)	–	3.9 + 3	9.9 + 4

In low energy irradiation, self-blocking effects are very high. For this reason, production of new nuclides is low in purely thermal spectrum even in high flux 10^{13} cm^{-2}s^{-1}. For neutron spectrum typical for light water blanket, with $\gamma = 0.4$, nuclide production is higher because of epithermal neutrons. Radiotoxicity at $\gamma = 0.4$ is determined by all radioactive nuclides. At $\gamma = 0$, ^{185}W gives a major part to radiotoxicity. Radiation doze power is determined by short-lived ^{188}W.

At 10-MeV neutron irradiation, radiotoxicity is determined by ^{181}W, ^{185}W, ^{184}Re, and radiation dose power by ^{184}Re.

Comparison of these data shows that a radiotoxicity caused by neutrons born in a target is 3 times greater and radiation dose power is 14 times less than the same characteristics caused by low energy neutrons from external blanket. However, it is necessary to mention that radiation characteristics caused by neutrons from an external blanket are defined by short-lived nuclides ^{187}W, ^{188}Re with half-life periods about 1 day, whereas at irradiation by neutrons born in target, main contribution come from nuclides ^{185}W, ^{181}W, and ^{184}Re with half-life periods from 38 up to 121 days. If we consider only radionuclides with half-life periods not less than several tenths of days, then it appears that high-energy neutrons born in a target make radiation dose power 6 times greater than neutrons from an external blanket.

5. Conclusion

Comparison of radiation characteristics produced in irradiation by low energy neutrons from surrounding blanket and high energy neutrons from a target shows a rather high role of neutrons from the target itself in the process of accumulation of those radionuclide which define the main radiation characteristics of the irradiated target.

Absolute values of radiation characteristics allow estimating necessary modes of irradiated target management. For tantalum and tungsten targets, radiation dose power is rather high and decreases slowly at cooling. Radiotoxicity value for all considered targets is close to that of radioactive waste of nuclear reactors. It should be recommended to store irradiated targets after some cooling while taking measures as for middle and high radioactive waste management.

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