Investigating on Radioactivity of LBE and Pb in ADS Spallation Target

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Abstract Using the Fluka Monte Carlo method, we argue the activity of lead-bismuth eutectic (LBE) and lead (Pb) spallation targets in Accelerator Driven Subcritical System (ADS). Results reveal that the radioactivity of Pb target is lower than that of LBE target when they have the same beam energy and target diameter, the accumulation activity of two target is enhanced as the increase of proton energy and target diameter. In addition, we also discuss the respective contribution of prominent radionuclides of LBE target under various cooling time, it is specified that the used LBE target can be seen as a permanent radioactive waste. For the present works, we expect that it can offer some valuable hints for the future experiment and the assessment of safety hazards of nuclear facilities.

Keywords ADS · Spallation products · Induced activity · FLUKA

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1 Introduction

Accelerator Driven Subcritical Systems (ADS) are a new option of nuclear energy due to their good cost efficiency, environmental benefits, security benefits, etc. [1, 2]. In such a system, an important component is the spallation target, which is the interface between the two principal elements, the high-power accelerator and the subcritical core. Choice of target material and design of target are dependent strongly on several factors like high neutron yield, ease of cooling, low chemical and radio-toxicity production, low risk of fire hazard in operating system, relatively low running cost of the system [3–6]. Presently, lead-bismuth eutectic (LBE) and lead (Pb) are widely used as a spallation target material for neutron production due to its several merits, such as good liquid coolant, high neutron yield, lower thermal neutron capture cross-section, high boiling point (of Pb 1737 °C, LBE 1670 °C) and low vapor pressure (parts per million of mercury in the operating temperature respectively) [7, 8]. These advantages mentioned above will bring great convenience of actual operation in ADS. Furthermore, the higher operating temperature of liquid metal target, which associates with the melting point of target material, is avoided in realistic applications. For the Pb and LBE targets, there is a melting point up to 327 °C for Pb and up to 123.5 °C for LBE, respectively [7], indicating that the Pb target has much higher operation temperature than LBE one. Hence, liquid Pb target requires special structural material that can endure higher temperature than LBE target. From the requirement point of view for target container, LBE has more advantages than Pb. Unfortunately, the LBE has its flaws as target material, the amount of polonium production in the LBE arouses a particular concern because of its high radiotoxicity and the resulting handing problems. Noble gases and the gasous phase of ^{210}Po are also known to occur, and the alpha decay ^{210m}Bi are formed as a result of the reaction ^{209}Bi(n, gamma)^{210m}Bi in LBE spallation target [9, 10].

As we all know, nuclear wastes are transmuted in ADS, meanwhile radioactive spallation residues are produced constantly through the primary interactions and secondary particles [11, 12]. Up to now, there are many experimental studies on the production of radionuclides [13–18], however, because of safety restrictions on the dose rate, targets were cooled for dozens of hours before the first measurement. So, it will lost information on the production of short-lived radionuclides, and experimental measurement could not identify $^{208-210}$Po radioisotopes due to the absence of their suitable characteristic photo peaks. Therefore, it is necessary to study the nuclide production and discuss the respective contribution of prominent radionuclides in spallation target based on nuclear physics calculations, the aim is to give some valuable hints for the future experiment and the assessment of safety hazards of nuclear facilities, including also options for intermediate and final disposal of the target material. In present work, we have not only calculated the induced activity in an ADS employing LBE and Pb targets at beam energies of 0.25, 0.5 and 1.0 GeV, but also surveyed the effects of LBE and Pb targets geometry on the generation of induced activity. Finally, we discuss the residual activity of long-life spallation products in LBE target after one year irradiation with proton beam current of 1.0 mA, beam energy of 1.0 GeV, and the cooling time up to 10^5 years.

2 Method of Calculation

In this manuscript, all calculations were performed with the Fluka Monte Carlo code [19]. The average proton current is 1.0 mA for 1 year of irradiation. Except for the discussion of the effects in target geometry, the rest of calculations always use a 10.2 cm diameter and 60.0 cm length spallation target.

2.1 Fluka

Fluka is a general purpose tool for calculations of particle transport and interactions with matter, covering an extended range of applications spanning from proton and electron accelerator shielding to target design, calorimetry, activation, dosimetry, detector design, Accelerator Driven Systems, cosmic rays, neutrino physics, radiotherapy etc. It can simulate with high accuracy the interaction and propagation in matter of about 60 different particles, including photons, electrons, neutrinos, muons, hadrons and all the corresponding antiparticles. The program can also transport polarised photons and optical photons. Time evolution and tracking of emitted radiation from unstable residual nuclei can be performed on line. Fluka can handle even very complex geometries, using an improved version of the well-known Combinatorial Geometry (CG) package. Various visualization and debugging tools are also available.

2.2 Induced Activity Calculation

In an ADS system, we always use thick and large mass target to ensure fully stop the projectile beam. In such a thick target, an isotope is produced by the interaction of the incident particle at different degrading energies from incident energy down to reaction threshold. The total yield of isotope is obtained by summing over the yield of the isotope over this entire energy range. The activity of a radioisotope ‘i’ is defined by [3]:

$$A_i = \sum_j N\sigma_i(E_j)\phi_j(E_j)(1 - e^{-\lambda t}) \qquad (1)$$

where A_i represents activity of radioisotope ‘i’, N is the number of target atoms available for reaction, $\sigma_i(E_j)$ is cross-section for production of radioisotope ‘i’ at projectile energy E_j, $\phi_j(E_j)$ is projectile flux at energy E_j, λ is decay constants of radioisotope 'i' and t stands for irradiation time.

3 Results and Discussion

Many types of nuclides are produced in a spallation target due to spallation, fission, (n, gamma), (n, xn) and other nuclear reactions. Which can be identified in three classes, one is represented by sharp peak of the light products dominated by tritium and helium. The second comprises a wide list of intermediate products resulted from either fission and evaporation, and the third is formed by extended range of nuclides sharply peaked at the mass numbers of initial target nuclei [20]. In the following description, we only consider some of the higher radiotoxic isotopes.

3.1 Induced Activity in Spallation Target

The generation of induced activity in an ADS employing LBE and Pb target has been studied. In Table 1, we make a comparison on the radioactivity of the LBE and pure Pb target after one year irradiation at beam energies of 0.25, 0.5 and 1.0 GeV. It can be seen distinctly that activity of the LBE target is higher than that of the Pb target at the same beam energy. Especially, the isotopes of polonium and bismuth are about two orders of magnitude larger than pure Pb target.

The reason results in the formation of ^{210m}Bi and ^{208}Bi are the reaction ^{209}Bi (n, gamma)^{210m}Bi/^{210}Bi and ^{209}Bi(n, 2n)^{208}Bi, respectively. While ^{210}Bi decay through beta emission to form ^{210}Po, ^{210}Po is the most important isotope of polonium that needs to be quantified from the radio-toxicity point of view [11]. ^{205}Pb formed by reaction of ^{204}Pb with ^{204}Pb(n, gamma)^{205}Pb plays a very important contribution to long-lived component of the activity of pure Pb target, also for the LBE target. For pure Pb target, there is no ^{210m}Bi production. We also observed that the accumulation activity of spallation products decreases remarkably with the reduction of proton energy, especially these nuclide with mass number around 150, including ^{150}Gd, ^{148}Gd and ^{154}Dy, among them, the nuclide ^{148}Gd attends widely attention due to its long-lived as well as alpha emitter, and releases significant amount of dose in case of inhalation [20]. The contribution of alpha emitting rare earth element will not be considered when beam energy is 0.25 GeV. This results are agree reasonably well with the Artisyuk's results [20]. Therefore, pure Pb target is better than LBE target in terms of radioactivity and radiotoxicity.

In Table 2, we make a comparison on the radioactivity of the LBE and Pb target with different diameter after one year irradiation at beam energy of 1.0 GeV. It can be seen that the activity of spallation products improves significantly as increasing

Table 1 Comparison on the radioactivity of the LBE and Pb target in different beam energy (Bq)

Nuclide	LBE			Pb		
	0.25 GeV	0.5 GeV	1.0 GeV	0.25 GeV	0.5 GeV	1.0 GeV
^{210}Po	1.91E + 12	6.40E + 12	1.50E + 13	0	4.19E + 09	3.46E + 10
^{209}Po	3.50E + 10	4.58E + 10	4.27E + 10	0	1.69E + 07	1.78E + 08
^{208}Po	5.57E + 12	6.74E + 12	7.84E + 12	2.66E + 08	7.97E + 08	3.72E + 09
^{210m}Bi	5.23E + 05	1.75E + 06	4.09E + 06	0	0	0
^{208}Bi	3.99E + 08	1.31E + 09	3.49E + 09	9.49E + 06	1.16E + 07	1.24E + 07
^{207}Bi	4.03E + 12	9.71E + 12	2.10E + 13	5.93E + 11	7.34E + 11	8.70E + 11
^{206}Bi	1.68E + 14	3.49E + 14	6.62E + 14	4.51E + 13	5.45E + 13	6.29E + 13
^{205}Bi	1.74E + 14	3.30E + 14	5.73E + 14	7.29E + 13	8.73E + 13	9.60E + 13
^{205}Pb	1.26E + 07	2.96E + 07	6.06E + 07	1.40E + 07	3.60E + 07	8.22E + 07
^{202}Pb	2.79E + 09	5.65E + 09	8.82E + 09	3.02E + 09	6.08E + 09	1.05E + 10
^{206}Tl	7.69E + 12	3.05E + 13	6.95E + 13	1.76E + 13	6.91E + 13	1.59E + 14
^{204}Tl	1.73E + 12	6.61E + 12	1.45E + 13	3.80E + 12	1.42E + 13	3.11E + 13
^{202}Tl	1.61E + 13	5.64E + 13	1.16E + 14	3.27E + 13	1.11E + 14	2.26E + 14
^{194}Hg	4.35E + 10	2.12E + 11	3.53E + 11	5.27E + 10	2.47E + 11	4.05E + 11
^{195}Au	2.87E + 13	1.28E + 14	2.12E + 14	3.99E + 13	1.60E + 14	2.60E + 14
^{194}Au	3.77E + 11	3.42E + 12	1.03E + 13	6.67E + 11	5.98E + 12	1.80E + 13
^{193}Pt	4.90E + 11	2.24E + 12	3.93E + 12	7.02E + 11	2.80E + 12	4.67E + 12
^{150}Gd	456.3	1.19E + 04	4.28E + 05	483.4	1.02E + 04	4.65E + 05
^{148}Gd	0	1.62E + 08	1.14E + 10	0	1.04E + 08	1.25E + 10
^{154}Dy	288.4	3751	3.81E + 05	0	1444	4.22E + 05
^{99}Tc	1.60E + 07	5.78E + 07	1.09E + 08	1.10E + 07	4.11E + 07	8.20E + 07
^{3}H	1.32E + 11	1.41E + 12	1.29E + 13	1.30E + 11	1.41E + 12	1.29E + 13

target diameter. In other word, spallation products are highly dependent on the mass of the target material. Only for the activity, lower beam energy and smaller target diameter are expected, but this effect will confront with the overall decrease in neutron production. Whereas the main application of spallation target in ADS is as a source for generation of neutron, thus the balance must be taken into account between the neutron yield and the cumulative activity.

3.2 *Residual Activity in LBE Target*

Many types of nuclides, including alpha-active polonium isotopes ($^{210, 209, 208, 206}$Po) and ^{210m}Bi as well as hard gamma emitting bismuth isotopes ($^{210, 208, 207, 206, 205}$Bi), are produced in a LBE target due to spallation, (n, gamma), (n, xn), (p, xn) and other nuclear reactions. These alpha and hard gamma emitter are the most hazardous due to their relatively long-lived radiowaste. In Figs. 1 and 2, we display the contribution of the isotopes of bismuth and polonium to total activity after

Table 2 Comparison on the radioactivity of the LBE and Pb target with different diameter (Bq)

Nuclide	LBE			Pb		
	10.2 cm	20 cm	30 cm	10.2 cm	20 cm	30 cm
^{210}Po	1.51E + 13	4.75E + 13	9.56E + 13	3.46E + 10	3.72E + 10	3.77E + 10
^{209}Po	4.23E + 10	5.31E + 10	5.63E + 10	1.78E + 08	1.56E + 08	1.48E + 08
^{208}Po	7.90E + 12	9.74E + 12	1.04E + 13	3.72E + 09	4.25E + 09	3.72E + 09
^{210m}Bi	4.11E + 06	1.30E + 07	2.62E + 07	0	0	0
^{208}Bi	3.50E + 09	6.28E + 09	8.28E + 09	1.24E + 07	1.54E + 07	1.62E + 07
^{207}Bi	2.10E + 13	3.60E + 13	4.66E + 13	8.70E + 11	1.05E + 12	1.12E + 12
^{206}Bi	6.63E + 14	1.09E + 15	1.37E + 15	6.29E + 13	7.67E + 13	8.14E + 13
^{205}Bi	5.75E + 14	9.18E + 14	1.15E + 15	9.60E + 13	1.18E + 14	1.25E + 14
^{205}Pb	6.07E + 07	1.01E + 08	1.30E + 08	8.22E + 07	1.40E + 08	1.80E + 08
^{202}Pb	8.85E + 09	1.34E + 10	1.62E + 10	1.05E + 10	1.64E + 10	2.03E + 10
^{206}Tl	6.89E + 13	1.03E + 14	1.27E + 14	1.59E + 14	2.38E + 14	2.88E + 14
^{204}Tl	1.45E + 13	2.13E + 13	2.57E + 13	3.11E + 13	4.62E + 13	5.59E + 13
^{202}Tl	1.17E + 14	1.69E + 14	2.02E + 14	2.26E + 14	3.31E + 14	3.96E + 14
^{194}Hg	3.54E + 11	4.16E + 11	4.44E + 11	4.05E + 11	4.83E + 11	5.18E + 11
^{195}Au	2.12E + 14	2.56E + 14	2.75E + 14	2.60E + 14	3.19E + 14	3.45E + 14
^{194}Au	1.03E + 13	1.19E + 13	1.25E + 13	1.80E + 13	2.06E + 13	2.15E + 13
^{193}Pt	3.94E + 12	4.69E + 12	5.04E + 12	4.67E + 12	5.64E + 12	6.10E + 12
^{150}Gd	4.16E + 05	4.14E + 05	4.27E + 05	4.65E + 05	4.73E + 05	4.62E + 05
^{148}Gd	1.07E + 10	1.12E + 10	1.11E + 10	1.25E + 10	1.23E + 10	1.25E + 10
^{154}Dy	3.81E + 05	3.79E + 05	3.62E + 05	4.22E + 05	4.43E + 05	4.39E + 05
^{99}Tc	1.09E + 08	1.24E + 08	1.30E + 08	8.20E + 07	9.13E + 07	9.51E + 07
^{3}H	1.29E + 13	1.34E + 13	1.35E + 13	1.29E + 13	1.33E + 13	1.35E + 13

1 year irradiation at a beam current of 1 mA, beam energy of 1 GeV, and a cooling time up to 10^5 years. It is observed in Fig. 1 that during the first month of cooling ^{205}Bi ($T_{1/2}$ = 15.31 days), ^{206}Bi ($T_{1/2}$ = 6.24 days) and ^{210}Bi ($T_{1/2}$ = 5.012 days) give the prominent contribution to total bismuth activity, and then ^{207}Bi ($T_{1/2}$ = 32.9 years) plays a major part in the activity. After 300 years of cooling, the activity of ^{207}Bi decreases to 3.16E + 10 Bq, and then ^{208}Bi ($T_{1/2}$ = 3.68E + 05 years) and ^{210m}Bi ($T_{1/2}$ = 3.04E + 06 years) dominate the total inventory. Therefore, we conclude that for bismuth isotopes long-lived radionuclides ^{207}Bi, ^{208}Bi, and ^{210m}Bi are the most hazardous. Particularly, the ^{210m}Bi decays by alpha particle emission that needs to be quantified from the radio-toxicity point of view.

In Fig. 2, we show the activity of polonium isotopes with a half live greater than one day, such as ^{206}Po (lifetime of 8.8 days), ^{208}Po (lifetime of 2.9 years), ^{209}Po (lifetime of 102 years) and ^{210}Po (lifetime of 138 days). All of these nuclides decay almost by alpha emission except that ^{206}Po has only about 5.45 % probability of alpha emission. In addition, we can see in Fig. 2 that in the initial days of decay, ^{206}Po dominates an essential activity of polonium isotopes. During the period of

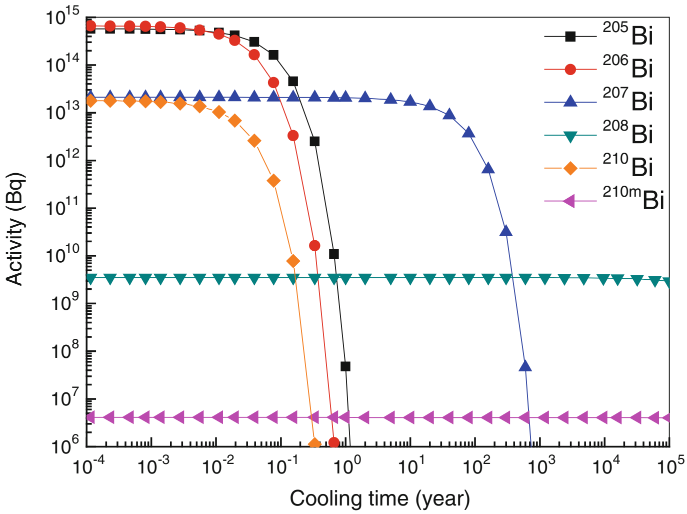

Fig. 1 Residual activity of the isotopes of bismuth produced in LBE target

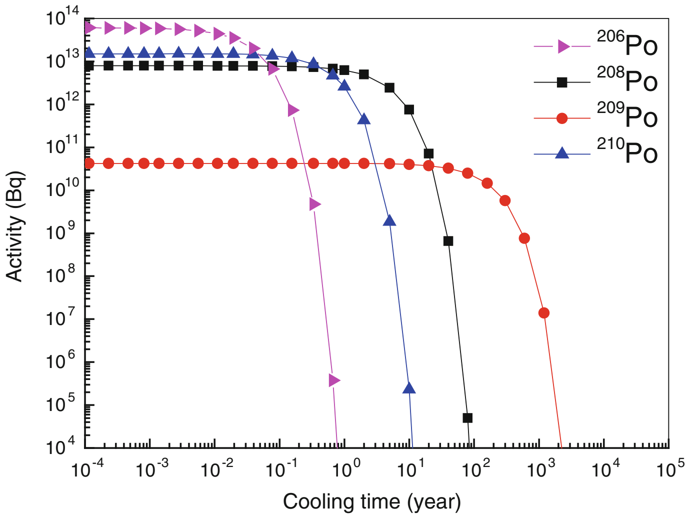

Fig. 2 Residual activity of the isotopes of polonium produced in LBE target

cooling from a month to about five years, ^{210}Po as the major radionuclide, which needs to be quantified from the radiotoxicity point of view because of the gaseous phase. After that ^{208}Po and ^{209}Po become the predominant radionuclide in the total polonium activity.

Figure 3 presents the total activity generated in the LBE target and a few prominent radionuclides, except for the isotopes of bismuth and polonium, the

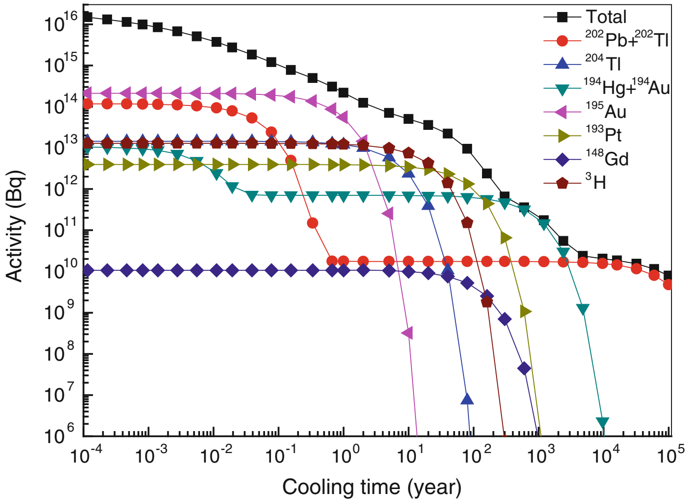

Fig. 3 Residual activity of the other nuclide produced in LBE target

contribution of these isotopes to the total activity is larger than 5 % at various cooling times, the activity of long-lived radionuclide ^{148}Gd which decay via alpha particle emission is also given to make a comparison with other isotopes. In addition, as the major radionuclides ^{195}Au, there is a cooling time up to 2 years and then the activity decreases quickly as increasing time, the activity of ^{195}Au devotes to the total activity up to 29 % after one year of cooling, tritium and ^{204}Tl can be retained during 20 years of cooling. After about 300 years of cooling, the nuclides of ^{194}Hg, ^{194}Au, ^{202}Pb and ^{202}Tl dominate in the total inventory, the contribution of ^{194}Hg and ^{194}Au to total activity approaches 90 % when cooling time is up to 600 years. After that, ^{202}Pb together with ^{202}Tl make the main contribution to the total activity. These results are consistent with research by Lemaire [21]. Therefore, we can conclude that LBE as spallation target material, the necessary measures must be taken to deal with used target as radioactive waste.

4 Summary and Conclusions

The Fluka Monte Carlo method is applied to explore the activity of LBE and Pb spallation target, it is found that the radioactivity of Pb target is significantly lower than that of LBE target under the same beam energy and target diameter, and there is no ^{210m}Bi production in pure Pb target. The accumulation activity of spallation products decreases remarkably with the reduction of proton energy, especially these nuclides with mass number around 150, when beam energy is 0.25 GeV, the contribution of alpha emitting rare earth element will not be considered. In addition, the respective contribution of prominent radionuclides of LBE target are surveyed at various cooling time, the results reveal that the alpha-active polonium isotopes ($^{210,\ 209,\ 208,\ 206}$Po), ^{210m}Bi and hard gamma emitting bismuth isotopes ($^{210,\ 208,\ 207,\ 206,\ 205}$Bi) are the most important radionuclides from the radiotoxicity and long term storage point of view. Moreover, ^{195}Au, ^{204}Tl, ^{3}H, ^{193}Pt, ^{194}Hg, ^{194}Au, ^{202}Pb and ^{202}Tl also have the important contribution to total activity in various cooling time, and then LBE as a target material must be classified as a radioactive waste permanently, for the Pb target, the situation is not so highlighted. Therefore, we hope that our results can give some valuable hints for the choice of target material and the future experiment.

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