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Estimation of induced activity in thick lead–bismuth and iron alloy targets by 30 MeV protons

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Abstract

Excitation functions of different radionuclides produced by interaction of protons and neutrons in lead–bismuth eutectic (LBE) and T91, D9 steel alloy targets are calculated using ALICE91 and EMPIRE 2.18 codes and compared with some experimental data. Radioactivity induced by 30 MeV protons is estimated in thick targets of LBE, T91 and D9 that are considered suitable target and beam window materials in accelerator driven subcritical systems (ADSS). The amount of induced radioactivity limits the allowed beam current as well as determines the criteria for hands-on-maintenance and disposal of radioactive wastes in such machines. The present estimation shows that for 500 μA proton beam ^{207}Po is produced with highest activity of around 5×10^6 MBq by proton induced reaction while ^{210}Bi is produced with highest activity (about 10^6 MBq) by interaction of primary neutrons in LBE.

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Keywords: ADSS; LBE; Induced activity; Proton induced reactions; Thick target

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1. Introduction

Significant efforts are being directed towards the development of accelerator driven subcritical systems (ADSS) that employ spallation reactions induced by high current proton beams of energies around 1 GeV. However, studies related to heat generation and target stability besides other logistics are being carried out using much lower energy, high current accelerators [1], which are easily available. In these high current machines one of the factors constraining the beam current and irradiation time is the production of induced radio- and chemical toxicity.

Lead and lead–bismuth alloy are, at present, considered as suitable candidates for the ADSS target-coolant system [2] because of their certain desirable properties like harder neutron spectrum, reduced possibility of boiling and loss of cooling, less fire-hazard (compared to liquid Na), reduced cost. In ADSS thick targets are used, such that the projectile beam is completely stopped inside the targets. The amount of induced activity depends on the target material, the proton flux as well as the transport properties of the incident protons and the neutrons produced. The amount of induced activity need be known for better planning of hands on maintenance and final disposal of the target as radioactive waste. This may be achieved in two ways: experimental measurement and theoretical model based calculation. While the former is the better choice producing more accurate results, it is in a way self-contradictory. This is because some idea about the induced activity is required even to build up an ADSS facility. Secondly, though it is possible at lower energies, it is not always feasible to experimentally measure the induced activity at high energies. On the other hand the reaction models used to calculate the activity are not capable to incorporate the characteristics of all physical processes involved in the reaction. Hence the results need to be validated against experimental data.

In the present work we have estimated the induced radioactivity in thick lead–bismuth eutectic (LBE) target,

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irradiated directly and also by neutrons generated in beam windows of metal steel alloy T91 or D9, in a 30 MeV, 500 μA proton accelerator. For this purpose we have used the cross-sections of different radionuclides produced in the reaction measured experimentally (available in the EXFOR data library) and also calculated using the nuclear reaction model codes ALICE91 [5,7,8] and EMPIRE 2.18 [6]. We have compared the results obtained from the measured and calculated data. We have also compared the measured and calculated cross-sections for proton induced reaction on LBE target above 30 MeV wherever the measured data are available. This is done to test the range of validity of the codes used. Since measured cross-sections are not available for all the radionuclides produced, estimation of cross-section using nuclear reaction model codes will play a dominant role. In the next section we describe the method of calculation. In Section 3 we present the results of our calculation followed by conclusions in Section 4.

2. Induced activity estimation

2.1. Thick target yield

In a thick target the incident projectile gradually slows down inside the target to the threshold of nuclear reactions that take place at different depths at different projectile energies. The total yield of any reaction product in such a thick target is the sum of the yields from several thin targets at gradually decreasing projectile energies. To account for this, we have assumed the thick target to consist of a number of thin slabs. The thickness of each slab is such that the incident beam loses same amount of energy in each slab. Thus if ΔE is the energy lost by the incident beam in each slab, then the thickness Δx_i of the ith slab is [3]

$$\Delta x_i = \frac{\Delta E}{\mathrm{d}E/\mathrm{d}x_{E_i}} \tag{1}$$

where $\mathrm{d}E/\mathrm{d}x_{E_i}$ is the stopping power for the projectile slowed down to energy E_i at the beginning of the ith slab. In the present study ΔE is taken as 1 MeV. Accordingly, attenuation of the incident proton flux at different layers of the thick stopping LBE, T91, D9 is calculated using TRIM [4].

2.2. Method of calculation

For LBE target, excitation functions of different radioisotopes produced by proton and neutron induced reaction on ^{204,206,207,208}Pb and ^{209}Bi are calculated using the codes ALICE91 [5] and EMPIRE 2.18 [6]. The calculated excitation functions are compared with experimentally measured data wherever available. In a similar manner excitation functions of various radionuclides produced by proton induced reactions on different stable isotopes of the constituent elements of T91 and D9 have been estimated and compared with available experimental data. From the measured and calculated excitation functions, induced radioactivities in LBE target and T91 and D9 windows due to proton and neutron interaction in a 30 MeV, 500 μA proton accelerator have been estimated for different irradiation periods (Table 1). For this purpose the codes ALICE91 and EMPIRE 2.18 are modified to be used for thick targets. LBE consists of 55% Bi and 45% Pb by weight. The only stable isotope of Bi is ^{209}Bi, while Pb has four stable isotopes, namely, ^{204,206,207,208}Pb with 1.4%, 24.1%, 22.1% and 52.4% abundance, respectively. The elemental composition of T91 and D9 by weight percentages (given in parantheses) are as follows:

T91: Fe (88.82), Cr (9.0), Mo (1.0), Mn (0.45), Ni (0.4), V (0.2), C (0.1), P (0.02), S (0.01).

D9: Fe (63.782), Cr (14.5), Ni (15.5), Mo (2.5), Mn (2.35), Si (0.75), Ti (0.25), V (0.05), Al (0.05), S (0.01), P (0.02), B (10–20 ppm), Nb (0.05), Ta (0.02), N (0.005), Co (0.05), Cu (0.04), As (0.03).

Of these, the isotopic abundances of the major elements are: ^{54,56,57,58}Fe (5.85%, 91.75%, 2.12%, 0.28%), ^{50,52,53,54}Cr (4.34%, 83.79%, 9.50%, 2.37%), ^{58,60,61,62,64}Ni (68.08%, 26.22%, 1.14%, 3.63%, 0.93%), ^{92,94,95,96,97,98,100}Mo 14.84%, 9.25%, 15.92%, 16.68%, 9.55%, 24.13%, 9.63%, ^{25}Mn (100.0%), respectively. The thickness of the window is considered to be 1.5 mm. The projected range of 30 MeV protons in D9 is about 1.7 mm and in T91 it is 1.5 mm. As a result when a beam window is used, radioactivity in the LBE target will be produced solely by the interaction of the neutrons, emitted from the window material, while that in the window material will be due to interaction of protons mostly.

In the case of neutron induced reactions, the energy distribution of neutron yield from thick LBE target is estimated. The excitation functions of the product radionuclides are then folded by this energy differential neutron yield per incident proton to obtain the net excitation

Table 1
Saturation activity of radioisotopes produced from LBE for irradiation upto 400 h by 30 MeV, 500 μA proton beam

Radioisotope (half life)	Decay mode (branching ratio %)	Saturation activity (MBq)
^{207}Po (5.8 h)	ε(99.98) α(0.02)	5.32E + 06
^{202}Bi (1.72 h)	ε, α($<10^{-5}$)	5.04E + 04
^{202}Pb (3.53 h)	IT(90.5), ε(9.5), α	5.23E + 02
^{207}Tl (4.77 in)	β-	2.29E + 03
^{200}Tl (26.1 h)	ε	1.17E + 01
^{204}Bi (11.22 h)	ε	9.70E + 05
^{203}Bi (11.76 h)	ε, α(10^{-5})	6.35E + 04
^{204}Pb(67.2 in)	IT, α	1.69E + 04
^{201}Pb (9.33 h)	ε	1.32E − 02
^{206}Tl (4.2 in)	β-, IT	1.67E + 03
^{199}Tl (7.42 h)	ε	5.56E + 00