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Comparison on the production of radionuclides in 1.4 GeV proton irradiated LBE targets of different thickness

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Abstract This is the first report on the inventory of radionuclides produced in 1.4 GeV proton induced reaction on Lead–Bismuth Eutectic (LBE) targets. LBE targets of 6 mm diameter and 1 to 8 mm lengths were irradiated with 1.4 GeV protons. The radionuclides ranging from ^{7}Be (53.12 days) to ^{207}Po (5.8 h) were identified in the samples with the help of time resolved γ-ray spectroscopy. However, there is no signature of formation of At radioisotopes, which can be produced by the interaction of secondary particles, typical for thick targets.

Keywords Radionuclide inventory · 1.4 GeV proton · Lead bismuth eutectic target · Gamma spectroscopy

Introduction

To build the next generation high power radioactive ion beam (RIB) facility, EURISOL (European Isotope Separation On-Line), is a part of the Nuclear Physics European Collaboration Committee (NuPECC) long-range plan. It is intended to use liquid Hg or LBE (Lead bismuth eutectic) as proton to neutron converter in the proposed EURISOL facility. Studies on the impact of high-energy protons (~1–1.5 GeV) on liquid Hg have been carried out as a task of EURISOL design study [1]. However, handling of tons of liquid Hg has some practical difficulty, which may be overcome using LBE as target material. Hence, it is important to have a detailed experimental knowledge on the production of radionuclides in the thick LBE targets on impact of high-energy (1.4 GeV) protons. This knowledge is important from multiple angles, to understand fundamental nuclear reactions in the high-energy domain, from the health physics point of view for risk assessment, etc. Moreover, these co-produced radionuclides in the LBE targets can be huge source of isotopes having applications in various fields. For example, accelerator-driven systems (ADS) are designed with LBE target, as well as LBE coolant instead of Na-coolant. Therefore the behavior of these radionuclides in LBE is also of interest [2].

In 1991, Ambe et al. introduced the multitracer technique, which is a breakthrough in tracer studies [3]. A multitracer solution contains a large number of radioactive species, all in their no-carrier-added state and with high specific activity. This multitracer solution allows to mimic natural condition and to study multielemental multidimensional natural dependence. Multitracer solution has been so far prepared by bombarding gold target by high energy ^{12}C/^{14}N/^{16}O projectiles (80–135 MeV/u) followed by radiochemical separation of bulk gold target [3–5]. Therefore radiochemical separation of bulk lead and bismuth targets after bombardment by high-energy protons will also lead to a multitracer solution which could be used in various studies in fundamental sciences.

Earlier several attempts were taken by the scientists to study the impact of high-energy protons on lead or bismuth targets. Rémy et al. [6] in association with CERN and Saclay (Saturne) studied the cross-section for binary and ternary fission induced by high-energy protons (0.6, 2, 3,

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18 and 23 GeV) in thin uranium and lead targets. Gloris et al. [7] determined the cross sections for proton induced reaction on ^{nat}Pb targets of thickness about 120 mg/cm^2 each, by stacked foil technique within the energy range of 80 MeV to 1.6 GeV at the Laboratoire National Saturne/Saclay. They reported the cross sections of around 68 radionuclides starting from ^{46}Sc ($T_{1/2} = 83.8$ days) to ^{207}Bi ($T_{1/2} = 31.55$ years) and concluded that at lower energies the radionuclides are produced by pre-equilibrium dominated reactions, hence the product masses are near to the target nuclei such as $^{nat}Pb(p,xn)^{205}Bi$. But at higher energies, the masses of the product radionuclides are nearly half of the target mass; hence, the mode of production can be related to fission, fragmentation and deep spallation processes, such as, the production of ^{83}Rb ($T_{1/2} = 86.2$ days) and $_{103}Ru$ ($T_{1/2} = 39.26$ days). Titarenko et al. [8] made experimental as well as computer simulation studies using HETC, GNASH, LAHET, INUCL, CEM95, CASCADE and ALICE codes on the yields of residual product nuclei in ^{209}Bi thin targets irradiated by 130 MeV and 1.5 GeV protons at the ITEP U-10 proton synchrotron. Based on the production of radionuclides by 1.5 GeV protons, they demonstrated different production channels, namely, spallation reactions (mass of the product nuclei ranged from ~125–210 amu), fission reactions (~30–140 amu); whereas the production of radionuclides with masses ~125–140 amu are most probably governed by both the channels. The number of radionuclides identified in 1.5 GeV proton bombardment on bismuth targets was around 102. Glories et al. [9] again extended the energy of the protons to 2.6 GeV on Pb targets of thickness 125 mg/cm^2 at the SATURNE II synchrocyclotron of the Laboratoire National Saturne at Saclay and at the cyclotron of the Svedberg Laboratory at Uppsala and reported a wide range of radionuclides starting from ^{7}Be ($T_{1/2} = 53.12$ days) to $_{207}Bi$. Around 129 radionuclides were identified in the proton irradiated Pb targets. They determined 2,000 individual cross sections for 127 reactions and discussed the different modes of production of the radionuclides like multi-fragmentation, three different types of asymmetric and symmetric fission and deep spallation. Using external beam of the ITEP U-10 proton synchrotron, Titarenko et al. [10] also measured the cross section of the produced radionuclides by proton irradiation on $^{206,207,208,nat}Pb$ and ^{209}Bi targets of thickness 127–358 mg/cm^2 for 11 proton energies within the range of 0.04–2.6 GeV and compared their results with the data obtained at other laboratories as well as with theoretical simulations by seven numerical codes. They found that the predictive powers of the codes varied but were satisfactory for most of the radionuclides in the spallation region.

Glasbrenner et al. [11] investigated the production of Po radionuclides by irradiating LBE targets of 2 mm thickness and 16 mm diameter with protons having 71 and 590 MeV energy. They found the formation of ^{206}Po and ^{208}Po radioisotopes in the targets and concluded that the specific activity ratio of $^{206}Po/^{208}Po$ is dependent on the energy and intensity of the proton beam. Using the FLUKA Monte Carlo code, Sunil et al. [12] estimated the neutron yield, neutron ambient dose equivalent, induced activity and the remnant gamma ambient dose equivalent for 0.1–1 GeV protons incident on thick LBE target. Radionuclides such as ^{195}Au ($T_{1/2} = 186.09$ days), ^{201}Tl ($T_{1/2} = 72.912$ h), ^{203}Pb ($T_{1/2} = 51.873$ h), ^{205}Bi ($T_{1/2} = 15.31$ days), ^{206}Bi ($T_{1/2} = 6.243$ days), ^{207}Bi ($T_{1/2} = 31.55$ years), ^{208}Po ($T_{1/2} = 2.89$ years), ^{210}Po ($T_{1/2} = 138.37$ days) and ^{3}H were found to be the important radionuclides in the target handling perspective. Formation of radionuclides in the LBE targets is highly dependent on the energy of the protons employed as it opens several reaction channels, which were studied earlier in either Pb or Bi targets.

This article reports the first experimental study on the production of radionuclides in the 1.4 GeV proton induced reactions on thick LBE targets of different thickness and lengths. It aims to compare the production and activity of various radionuclides in variable lengths of LBE targets. This paper also discusses the production of radiotoxic Po and At radionuclides in the LBE targets.

Experimental

Preparation of the LBE targets

Eight cylindrical thick LBE targets having a fixed diameter of 6 mm with varying lengths from 1 to 8 mm were prepared by melting pure LBE pieces. The chemical composition of LBE targets is Pb:Bi = 45:55 by weight. The weight of the targets was varying from 0.43 to 2.83 g with the increase in length (Table 1). To avoid the risk of contamination from the irradiated samples, each target was sealed by two-component glue (Araldite) before irradiation. The layer of glue was about 0.5 mm covering the total surface of the LBE targets.

Irradiation

The RaBIT (rapid proton beam irradiation transport) irradiation facility at CERN-ISOLDE was used to irradiate the sealed targets by 1–3 pulses of 1.4 GeV protons (Fig. 1). The RaBIT facility consists of a pneumatic transport system installed in a class A laboratory that allows to send samples in containers (referred as shuttle in this article) in front of the beam line and bring these back. Typically the shuttles are made of plastic but during these experiments it was observed that the mechanical integrity of those was

Table 1 Details of 1.4 GeV proton irradiation on the LBE targets

Target history [cyclindrical/6 mm diameter]		Proton irradiation details			
Length (mm)	Weight (g)	Pulses	Duration	No of protons/sample	Remarks
1	0.4273	1	1 s	3.18×10^{12}	Plastic shuttle
2	0.7812	1	1 s	3.18×10^{12}	Plastic shuttle
3	1.0511	1	1 s	3.00×10^{13}	Plastic shuttle
4	1.3742	1	1 s	3.00×10^{13}	Plastic shuttle
5	1.7316	3	1 min 51 s	3.43×10^{13}	Aluminum shuttle
6	2.0882	3	1 min 51 s	3.40×10^{13}	Aluminum shuttle
7	2.4474	3	1 min 51 s	3.26×10^{13}	Aluminum shuttle
8	2.8303	3	1 min 51 s	3.22×10^{13}	Aluminum shuttle

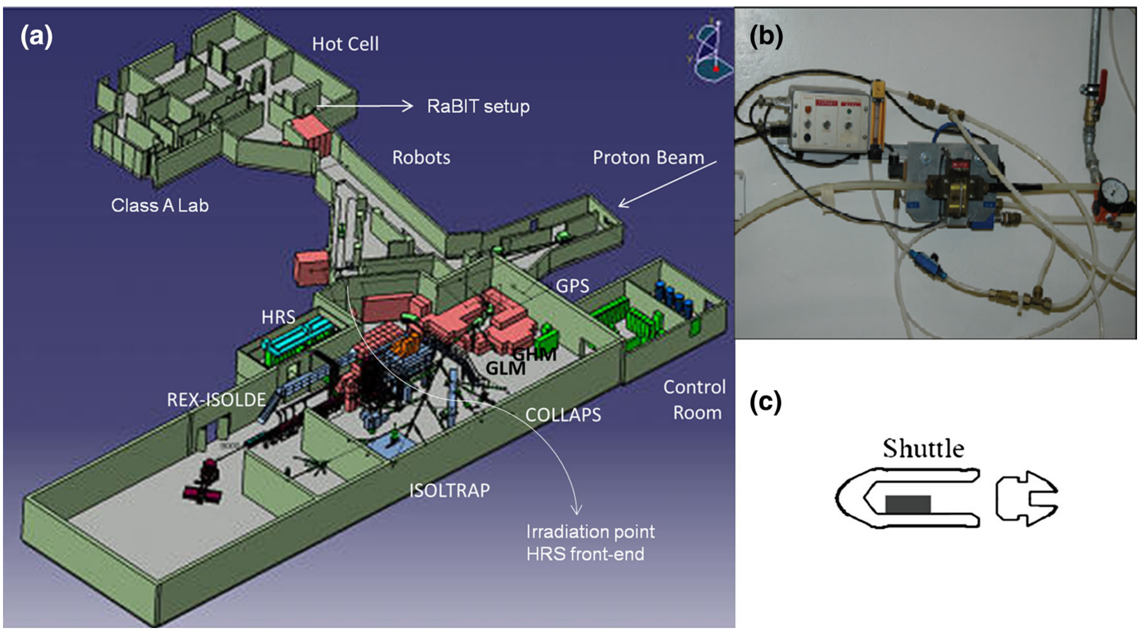

Fig. 1 **a** Location of RaBIT (rapid proton beam irradiation transport) set up at the ISOLDE facility. **b** The RaBIT pneumatic transport system, **c** schematic representation of the plastic/aluminum containers (shuttle) with a loaded sample. The external diameter is 10 mm

compromised. Therefore, it was decided to continue the irradiations using shuttles made of a more robust material such as aluminum. The total intensity of the proton beams for the first two targets (1–2 mm length) was 3.18×10^{12} protons/sample and for the other six targets (3 to 8 mm length) was in between (3.0 and 3.43) $\times 10^{13}$ protons/sample. The beam intensity was measured with the help of a current integrator placed before the target. Details of 1.4 GeV proton irradiation on varying lengths of the LBE targets are shown in Table 1. Due to safety restrictions on the dose rate, targets were cooled for 24 h before the first measurement. Therefore, we lost information on the production of short-lived radionuclides; except for the short-lived daughter products of long-lived parents. A regular series of time resolved γ-spectra were collected off-line for each irradiated target for the first 15 days after proton bombardment. A *p*-type HPGe detector (ORTEC) of 2.1 keV resolution at 1.33 MeV in combination with digital spectrum analyzer (DSA 1000, CANBERRA) and Genie 2 k software (CANBERRA) were used for the off-line γ-ray spectrometric measurements. The energy and efficiency calibration of the detector was performed using standard sources of known activity like ^{152}Eu ($T_{1/2}$ = 13.528 years), $_{133}$Ba ($T_{1/2}$ = 10.551 years) and ^{60}Co ($T_{1/2}$ = 5.27 years). The short-lived product radionuclides in the 1.4 GeV proton irradiated LBE targets were identified by the characteristic photo peaks and decay data. The yields of long lived ($T_{1/2} > 5$ days) radionuclides were calculated from the series of γ-spectra taken after 54–60 days and also after 359–368 days of the end of bombardment (EOB) using similar measurement set up. Some of the activities of the long-lived radioisotopes were not determined because of the presence of long-lived parent isotopes, compared to the daughter and improper peak positions in the gamma spectrum. It was difficult to quantify the activity of short-lived radionuclides due to the high Compton background in the initial spectra. Some of the radionuclides could not be confirmed in the sample due to overlapping photo peaks and similar half-lives with other radionuclides.

Results and discussion

The radioisotopes identified ranged from ^{7}Be (53.12 days) to ^{207}Po (5.8 h). The identified short-lived ($T_{1/2} < 5$ days) radionuclides has been listed in Table 2. The list includes ^{206,207}Po, $^{203-207}$Bi, ^{200,201,203}Pb and other successive lower Z elements. Production of ^{7}Be is questionable as it may also be produced by the interaction of 1.4 GeV proton beam with the two-component glues that was used to seal the targets. However, Gloris et al. [9] also reported production of ^{7}Be when bombarded lead targets by 2.6 GeV protons.

The total numbers of identified radionuclides in target lengths 8 to 1 mm are 111, 101, 103, 98, 87, 86, 56 and 45 respectively. Hence, the number of radioisotopes in the LBE targets steadily decreased with the decrease in length of the samples of fixed diameter from 8 mm to 1 mm. This signifies that the production of radioisotopes in the LBE targets is highly dependent on the mass of the LBE target. In other word, as the target length increases more numbers of isotopes are observed. The radioisotopes and their measured activities (long-lived >5 days) at the experimental condition have been tabulated in Table 3. The activities of each radioisotope have been normalized with respect to total proton intensity of 3×10^{13}. The calculated activities at EOB of the long-lived ($T_{1/2} > 5$ days) radioisotopes produced in the LBE targets were higher in sample length 8 mm compared to other samples which also signifies that the production of radioisotopes and their yield is dependent on mass of the LBE targets.

It should be kept in mind that the measured activity is not the absolute activity; even it may differ largely in some cases. This is because the activity produced inside the target is largely attenuated, even some time it can be below the detection limit. For example, from our earlier study it has been found that liquid Hg shows self-shielding of 90 % for 141 keV γ-ray of ^{99m}Tc/^{99}Mo [13]. Therefore the activity reported in this paper is the minimum activity that would be produced in the given experimental condition and can be termed as "visible activity". For example, due to less shielding and less self-attenuation, visible activities in sample 1 and 2 are comparatively higher. Only radiochemical treatment, i.e. dissolving the target, separation of the bulk, etc., followed by quantification should give correct estimate of the 'actual' activity.

The modes of production of the radionuclides include multiple mechanisms, namely, fission, fragmentation, spallation, etc., which was also observed by other scientists while using Pb or Bi as target materials at higher proton energies [7–10]. The produced radionuclides so evidenced in the proton irradiated LBE target undergo various decay processes such as electron capture, alpha decay, β^- decay, isomeric transitions, etc., hence it is difficult to predict accurately the mechanism of formation of each individual radionuclides.

Po formation

Among the various Po radioisotopes, we observed only the formation of ^{206}Po ($T_{1/2} = 8.8$ days) and ^{207}Po ($T_{1/2} = 5.8$ h). Since, proton irradiated LBE targets were cooled for 24 h before the first measurement, short-lived Po radioisotopes (having $T_{1/2}$ between 2–3 h) were not identified. We could not identify $^{208-210}$Po radioisotopes due to absence of their suitable characteristic photo peaks. The ^{206,207}Po isotopes may be produced through secondary particle formation, like ^{209}Bi($p,\pi^- xn$)^{206,207}At (ε)^{206,207}Po or ^{204,206,207,208}Pb($p,\pi^- xn$)^{206,207}Po types of reactions, typical in thick targets, as observed and advocated by Tall et al. [14]. However, it is not possible to comment on the formation of ^{206}At ($T_{1/2} = 30.6$ min) or ^{207}At ($T_{1/2} = 1.81$ h) radionuclides based on our measurements. The ^{206,207}Po radioisotopes may also be produced directly through ^{209}Bi(p,4n)^{206}Po or ^{209}Bi(p,3n)^{207}Po reactions. Table 3 shows that the activity of ^{206}Po is considerably high in thicker targets, which reduces the probability of formation of polonium isotopes through secondary particle reactions, as the yield of the secondary reactions are generally smaller. Tall et al. [14] also predicted for "small" amount of Po radioisotopes that could be formed through secondary particle reactions.

At formation

We have not observed any signature of At radionuclides in any of the samples. However, it was not possible for us to observe short-lived $^{204-208}$At radionuclides due to 24 h cooling time. The high activity of ^{206}Po does not vouch its formation through the decay of ^{206}At. Tall et al. [14] observed the formation of $^{204-210}$At radioisotopes on irradiating thick LBE targets with 1–1.4 GeV protons at CERN-ISOLDE. The target used by them consisted of lead–bismuth contained in a tantalum cylinder of 20 cm in length and 1 cm in radius, 75 % filled, of mass 547 g. The modes of production of At radionuclides were assisted either by ($p,\pi^- xn$) charge exchange reactions on ^{209}Bi or

Table 2 List of radioisotopes identified in 1.4 GeV proton irradiated LBE targets ($T_{1/2}$ < 5 days)

Radioisotopes ($T_{1/2}$)	Measured γ-energy (keV) (I_γ %)	Type	Present in LBE target length (mm)	Radioisotopes ($T_{1/2}$)	Measured γ- energy (keV) (I_γ %)	Type	Present in LBE target length (mm)
^{42}K (12.36 h)	1,524.7 (18)	I	3, 4, 5, 6, 7, 8	^{165}Tm (30.1 h)	806.6 (8.3)	I	All
^{48}Sc (43.67 h)	1,312.0 (100)	I	3, 4, 5, 6, 7, 8	^{169}Lu (34.06 h)	960.6 (23.5)	I	All
^{69}Ge (39.05 h)	1,106.8 (36)	I	7, 8	^{170}Lu (2.01 days)	1428.2 (3.4)	C	All
^{77}Ge (11.30 h)	1,368.3 (3.2)	I	6, 7, 8	^{170}Hf (16.01 h)	120.2 (19)	I	All
^{72}As (26.0 h)	8,34.1 (79.5)	I	All	^{173}Hf (23.6 h)	297.0 (33.9)	I	All
^{76}Br (16.2 h)	657.1 (15.9)	I	3, 4, 5, 6, 7, 8	^{175}Ta (10.5 h)	857.8 (3)	I	3, 4, 5, 6, 7, 8
^{77}Br (57.03 h)	249.8 (2.9)	I	3, 4, 5, 6, 7, 8	^{176}Ta (8.09 h)	1159.3 (24.6)	I	3, 4, 5, 6, 7, 8
^{82}Br (35.30 h)	554.3 (70.9)	I	3, 4, 5, 6, 7, 8	^{187}W (23.72 h)	479.5 (21.8)	I	8
^{79}Kr (35.04 h)	261.3 (13)	I	2, 6, 7, 8	^{181}Re (19.9 h)	953.4 (3.6)	I	All
^{86}Y (14.74 h)	627.8 (32.6)	C	3, 4, 5, 6, 7, 8	^{182}Re (64.0 h)	1231.0 (15)	I	5, 6, 7, 8
^{87}Y (3.325 days)	484.8 (89.7)	I	1, 2, 3, 4, 5, 6	^{182m}Re (12.7 h)	1,221.4 (24.8)	I	3, 4, 5
^{86}Zr (16.5 h)	242.8 (96)	I	2, 3, 4, 5, 6, 7, 8	^{185}Ir (14.4 h)	1,828.8 (76)	I	3, 4, 5, 6, 7, 8
^{89}Zr (78.41 h)	908.9 (100)	I	1	^{186}Ir (16.64 h)	137.1 (41.3)	I	All
^{97}Zr (16.91 h)	743.4 (93)	I	5, 6, 7, 8	^{187}Ir (10.5 h)	977.4 (61)	I	3, 4, 5, 6, 7, 8
^{96}Nb (23.35 h)	1,200.2 (19.8)	I	2, 3, 4, 5, 6, 7, 8	^{188}Ir (41.5 h)	672.5 (1.4)	C	All
^{96}Tc (4.28 days)	812.6 (82)	I	All	^{190m2}Ir (3.25 h)	186.7 (66.3)	I	3, 4, 5, 6, 7, 8
^{97}Ru (2.9 days)	215.7 (86)	I	6, 7, 8	^{195m}Ir (3.8 h)	684.9 (9.4)	I	7, 8
^{100}Rh (20.8 h)	1,107.2 (13.6)	I	8	^{189}Pt (10.87 h)	721.4 (5.5)	I	3, 4, 5, 6, 7, 8
^{101m}Rh (4.3 days)	306.8 (81)	I	2, 4, 6, 7, 8	^{191}Pt (2.8 days)	359.9 (6)	I	All
^{111}In (2.80 days)	171.3 (90.2)	I	3, 4, 5, 6, 7, 8	^{192}Au (4.94 h)	316.5 (58)	I	3, 4, 5, 6, 7, 8
^{119m}Te (4.70 days)	270.5 (28)	I	6, 7, 8	^{193m}Hg (11.8 h)	258.1 (60)	I	2, 3, 4, 5, 6, 7, 8
^{129}Cs (32.06 h)	548.9 (3.42)	I	3, 4, 5, 6, 7, 8	^{195m}Hg (41.6 h)	560.3 (7.5)	I	2, 8
^{135m}Ba (28.7 h)	268.2 (15.6)	I	All	^{198}Tl (5.3 h)	1,489.6 (2.6)	I	3, 4, 5, 6, 7, 8
^{132}La (4.8 h)	464.5 (76)	I	8	^{199}Tl (7.42 h)	247.2 (9.2)	I	3, 4, 5, 6, 7, 8
^{146}Eu (4.61 days)	633.1 (44)	C	All	^{200}Tl (26.1 h)	367.9 (87)	C	All
^{147}Gd (38.06 h)	929 (19.4)	I	All	^{201}Tl (72.9 h)	167.4 (9.4)	C	All
^{151}Tb (17.60 h)	251.8 (26)	I	2, 3, 4, 5, 6, 7, 8	^{200}Pb (21.5 h)	235.6 (4.3)	I	All
^{153}Tb (2.34 days)	212.0 (31)	C	2, 5, 6, 7, 8	^{201}Pb (9.33 h)	945.9 (7.4)	I	2, 3, 4, 5, 6, 7, 8
^{154}Tb (21.5 h)	1,123.1 (5.7)	I	5, 7, 8	^{203}Pb (51.87 h)	401.3 (3.3)	C	All
^{153}Dy (6.4 h)	274.5 (5.3)	I	3, 4, 5, 6, 7, 8	^{203}Bi (11.76 h)	820.3 (29.6)	I	3, 4, 5, 6, 7, 8
^{157}Dy (8.14 h)	326.2 (90)	I	3, 4, 5, 6, 7, 8	^{204}Bi (11.22 h)	899.2 (98)	I	All
^{171}Er (7.51 h)	308.3 (64.4)	I	8	^{207}Po (5.80 h)	992.2 (60)	I	3, 4, 5, 6, 7, 8

I Independent and *C* Cumulative

by secondary reactions involving ^{3}He and ^{4}He. The masses of LBE targets used in our experiment were between 0.43 and 2.83 g, two orders of magnitude less compared to that used in ref [14]. The production yields of the secondary reactions are generally small. Therefore it may also happen that the amount of At produced through secondary particle reactions in our experiment is too tiny and below the detection limit.

Applications

As indicated earlier, the high-energy proton irradiated thick LBE targets are huge source of radionuclides, which are useful in practical applications, e.g., in the field of medical science, industry and technology. In literature, various reports are found for the production of these radionuclides by low energy nuclear reactions (5–7 MeV/u). In this section, we discuss some specific applications of the radionuclides identified in the LBE targets. For example, the long-lived ^{65}Zn ($T_{1/2}$ - = 244.3 days) is used as a tracer to study the bio-kinetics and distribution of zinc in various organs and subcellular compartment of the body [15, 16] whereas $_{72}$As ($T_{1/2}$ = 26.0 h), ^{86}Y ($T_{1/2}$ = 14.74 h) and ^{89}Zr ($T_{1/2}$ = 78.41 h) find their applications in PET imaging [17–19]. Efforts have been made to validate various production routes of different neutron deficient Tc radionuclides like $^{93-96}$Tc. ^{96}Tc ($T_{1/}$

Table 3 Produced long-lived radioisotopes ($T_{1/2} > 5$ days) with their visible activity in 1.4 GeV proton irradiated LBE targets

Radioisotopes ($T_{1/2}$)	Measured γ-energy (keV) with (I_γ %)	Type	Present (P)/measured activity (A_0) at EOB (Bq) per 3 × 10^{13} protons							
			LBE target lengths (mm)							
			1	2	3	4	5	6	7	8
^{7}Be (53.12 days)	477.6 (10.4)	I	66.0 ± 6.6	113.2 ± 8.4	44 ± 1	54 ± 1	170.5 ± 1.7	184.4 ± 2.6	159.2 ± 1.8	219.9 ± 1.9
^{46}Sc (83.8 days)	1,120.5 (99)	I			2 ± 0.1	2 ± 0.1	6.1 ± 0.1	4.4 ± 0.1	4.6 ± 0.1	6.5 ± 0.1
^{54}Mn (0.85 years)	834.8 (99.9)	I			1 ± 0.2	1 ± 0.1	2.6 ± 0.2	2.6 ± 0.1	1.8 ± 0.2	2.8 ± 0.1
^{59}Fe (44.5 days)	1,291.6 (43.2)	I			3 ± 0.4	5 ± 0.5	20.1 ± 0.4	20.3 ± 0.8	19.3 ± 0.5	21.4 ± 0.5
^{58}Co (70.86 days)	810.8 (99)	I	9.4 ± 1.8		14 ± 1	45 ± 1	165.3 ± 2.6	176.5 ± 2.6	165.6 ± 2.8	151.9 ± 2.8
^{60}Co (5.27 years)	1,173.2 (99.9)	I					0.9 ± 0.1	0.9 ± 0.3	0.9 ± 0.2	0.9 ± 0.1
^{65}Zn (244.3 days)	1,115.5 (50.7)	I					2.6 ± 0.1	1.8 ± 0.2	1.8 ± 0.3	3.7 ± 0.2
^{74}As (17.77 days)	595.9 (60.2)	I	56.6 ± 4.7	47.1 ± 5.6	20 ± 1	31 ± 2	105.8 ± 1.7	105.0 ± 1.8	81.0 ± 1.8	121.1 ± 1.9
^{75}Se (119.8 days)	136 (59)	I		9.4 ± 0.9	2 ± 0.1	2 ± 0.1	6.1 ± 0.1	6.2 ± 0.6	5.5 ± 0.1	1.9 ± 0.1
^{83}Rb (86.2 days)	520.5 (46)	I	9.4 ± 1.8	18.8 ± 0.9	7 ± 0.2	11 ± 0.3	36.7 ± 0.3	29.1 ± 0.5	30.4 ± 0.5	41.9 ± 0.4
^{85}Sr (64.84 days)	513.9 (99)	I	9.4 ± 0.9	18.8 ± 0.9	6 ± 0.2	9 ± 0.2	30.6 ± 0.3	28.2 ± 0.3	22.1 ± 0.3	31.7 ± 0.3
^{88}Y (106.65 days)	1,836.1 (99.3)	I	9.4 ± 1.8	18.8 ± 0.9	9 ± 0.3	15 ± 0.4	24.5 ± 0.2	42.3 ± 0.3	35.0 ± 0.4	60.5 ± 0.4
^{95}Zr (64.02 days)	756.7 (54.5)	I	9.4 ± 1.8	9.4 ± 3.7	5 ± 0.2	7 ± 0.3	31.5 ± 0.4	30.9 ± 0.4	27.6 ± 0.5	28.9 ± 0.4
^{95}Nb (34.97 days)	765.8 (99.8)	I/C	28.3 ± 1.8	66.0 ± 1.8	23 ± 0.4	35 ± 0.5	127.7 ± 0.8	118.2 ± 0.6	103.1 ± 0.5	139.7 ± 0.5
^{103}Ru (39.2 days)	610.3 (5.6)	I			28 ± 2		32.4 ± 4.4	56.5 ± 4.4	33.1 ± 3.7	66.1 ± 3.7
^{102}Rh (207 days)	556.4 (96)	C						66.2 ± 6.2		
^{102m}Rh (2.9 years)	697.5 (44)	I					6.1 ± 0.2	5.3 ± 0.2	7.4 ± 0.2	5.6 ± 0.2
^{110m}Ag (249.7 days)	884.7 (72.2)	I			1 ± 0.2	1 ± 0.2	1.7 ± 0.2	0.9 ± 0.1	1.8 ± 0.4	5.6 ± 0.3
^{114m}In (49.5 days)	190.3 (15.4)	I			2 ± 0.3	3 ± 0.8	7.0 ± 0.8	5.3 ± 0.9	4.6 ± 0.7	56.8 ± 5.6
^{113}Sn (115.1 days)	391.7 (64)	I					P	P	P	P
^{117m}Sn (13.6 days)	158.6 (86.4)	I			4 ± 0.8	5 ± 0.5	8.7 ± 9	13.2 ± 0.9	9.2 ± 0.9	10.2 ± 0.9
^{121}Te (16.78 days)	573.1 (80.3)	I	28.3 ± 3.7	47.1 ± 5.6	15 ± 1	23 ± 1	80.5 ± 0.9	74.1 ± 1.8	66.2 ± 0.9	79.2 ± 0.9
^{121m}Te (154 days)	212.2 (81.4)	I				1 ± 0.4	0.9 ± 0.1	0.9 ± 0.3	0.9 ± 0.2	0.9 ± 0.2
^{127}Xe (36.4 days)	172.1 (23.5)	I	9.4 ± 0.9		5 ± 0.7	8 ± 0.7	21.9 ± 0.9	21.2 ± 0.9	17.5 ± 0.9	12.1 ± 0.6
^{129m}Xe (8.88 days)	196.6 (4.6)	I		P					P	P
^{133}Xe (5.24 days)	80.9 (38)	I								P
^{131}Ba (11.5 days)	216.0 (20)	I								82.9 ± 8.4
^{133}Ba (10.5 years)	356.0 (62)	I						0.9 ± 0.3		
^{139}Ce (137.6 days)	165.8 (79.9)	I	9.4 ± 0.9	9.4 ± 1.8	1 ± 0.1	1 ± 0.1	3.5 ± 0.1	3.5 ± 0.1	2.8 ± 0.1	4.6 ± 0.1
^{147}Nd (10.98 days)	439.9 (1.2)	I								P
^{143}Pm (265 days)	741.9 (38.5)	I			1 ± 0.1	2 ± 0.2	7.0 ± 0.3	4.4 ± 0.3	4.6 ± 0.3	6.5 ± 0.2
^{147}Eu (24.1 days)	933.2 (3.6)	I			52 ± 4	51 ± 10	265.9 ± 8.7	262.9 ± 10.6	179.4 ± 9.2	286.9 ± 11.2
^{148}Eu (54.5 days)	629.9 (71.9)	I					1.7 ± 0.2	1.8 ± 0.2		

Table 3 continued

Radioisotopes ($T_{1/2}$)	Measured γ-energy (keV) with (I_γ %)	Type	Present (P)/measured activity (A_0) at EOB (Bq) per 3 × 10^{13} protons							
			LBE target lengths (mm)							
			1	2	3	4	5	6	7	8
^{149}Eu (93.1 days)	327.6 (3.9)	I/C		66.0 ± 13.2	6 ± 1	8 ± 2	56.0 ± 2.6	51.2 ± 2.6	48.8 ± 2.8	61.5 ± 2.8
^{146}Gd (48.27 days)	154.7 (46.6)	I	9.4 ± 0.9	9.4 ± 0.9	1 ± 0.1	1 ± 0.2	1.7 ± 0.2	1.8 ± 0.3	1.8 ± 0.4	2.8 ± 0.5
^{149}Gd (9.28 days)	149.9 (41.9)	I	P	P	34 ± 7	33 ± 7	91.0 ± 7.0	261.2 ± 21.2	203.4 ± 17.5	135.1 ± 7.4
^{153}Gd (240.4 days)	103.2 (19.6)	I			1 ± 0.2	1 ± 0.2	0.9 ± 0.2	0.9 ± 0.3	0.9 ± 0.3	0.9 ± 0.2
^{167}Tm (9.25 days)	207.8 (41)	I	556.6 ± 47		162 ± 8	192 ± 35	302.6 ± 9.6	318.5 ± 11.5		481.7 ± 10.2
^{169}Yb (32.02 days)	130.5 (11.1)	I	94.3 ± 2.8	150.9 ± 4.7	13 ± 0.7	15 ± 1	23.6 ± 1.7	63.5 ± 1.8	48.8 ± 1.8	77.3 ± 0.9
^{171}Lu (8.24 days)	739.8 (48.1)	I	632.1 ± 85	773.5 ± 132.1	297 ± 20	480 ± 24	2,083 ± 30	2,019 ± 34	1,679 ± 29	2,336 ± 32
^{172}Lu (6.70 days)	1,093.7 (63.5)	I		5,755 ± 434	2,382 ± 76	3,843 ± 127	P	P	P	P
^{173}Lu (1.37 years)	272.1 (21.2)	I	9.4 ± 1.8	9.4 ± 1.8	5 ± 0.1	7 ± 0.2	31.5 ± 0.3	31.8 ± 0.4	23 ± 0.4	24.2 ± 0.4
^{172}Hf (1.87 years)	125.8 (11.3)	I					2.6 ± 0.4	1.8 ± 0.3	1.8 ± 0.5	2.8 ± 0.4
^{175}Hf (70 days)	343.4 (84)	I			49 ± 1	65 ± 1	312.2 ± 2.6	334.4 ± 2.6	230.1 ± 2.8	234.8 ± 2.8
^{183}Ta (5.1 days)	107.9 (10.7)	I		P	2,751 ± 784	3,077 ± 974	3,626 ± 1,402	3,283 ± 866	3,355 ± 924	1,438 ± 432
^{183}Re (70.0 days)	162.3 (23.3)	I	47.1 ± 1.8	283.0 ± 9.4			87.5 ± 2.6	75.9 ± 16.8		P
^{185}Os (93.6 days)	874.8 (6.6)	I	56.6 ± 2.8	132.1 ± 3.7	44 ± 2	62 ± 2	230.9 ± 2.6	225.0 ± 2.6	194.2 ± 2.8	266.5 ± 2.8
^{188}Pt (10.2 days)	381.4 (7.5)	I		547.1 ± 75.4	216 ± 25	367 ± 40	515.2 ± 14.9	514.4 ± 15.0	436.2 ± 14.7	1,633 ± 45
^{203}Hg (46.59 days)	279.2 (81)	I	9.4 ± 0.9							
^{202}Tl (12.23 days)	439.6 (91.4)	I	179.2 ± 9.4	396.2 ± 18.8	137 ± 3	206 ± 24	755.7 ± 3.5	747.3 ± 4.4	639.6 ± 3.7	899.1 ± 23.3
^{205}Bi (15.31 days)	1,764.3 (32.5)	I	424 ± 38	867.9 ± 47.1	328 ± 7	410 ± 10	2,171 ± 12	1,506 ± 19	1,903 ± 14	2,593 ± 13
^{206}Bi (6.243 days)	803.1 (99)	I	2,264 ± 207	P	P	P	P	P	P	P
^{207}Bi (31.55 years)	569.7 (97.8)	I	9.4 ± 0.9	9.4 ± 0.9	1 ± 0.1	1 ± 0.1	3.5 ± 0.1	2.6 ± 0.2	2.8 ± 0.1	6.5 ± 0.1
^{206}Po (8.8 days)	1,032.3 (32.9)	I		P	13 ± 6	73 ± 22	293.0 ± 30.6	497.6 ± 22.9	420.5 ± 22.1	567.4 ± 22.4

***I* Independent and *C* cumulative

$_2$ = 4.28 days) has been proposed for use in prevention of coronary restenosis [20, 21]. The ruthenium radionuclides such as ^{97}Ru ($T_{1/2}$ = 2.9 days) are used in cisternography. The ^{97}Ru-bleomycin labeled compound is used in diagnosis of tumors and also as a chemotherapeutic agent [22] while ^{103}Ru ($T_{1/2}$ = 39.2 days) is used to study the migration of ^{103}Ru radionuclide in tumor cells and liver [23, 24]. One of the mostly used radionuclides ^{111}In ($T_{1/2}$ = 2.80 days) finds its applications in the labeling of cellular blood components, monoclonal antibodies, myocardial damage detections, localization of abscess in polycystic kidneys, radiolabeled immunoglobulin therapies, imaging for cancer, etc. Apart from this, ^{111}In is also used as a probe in γ–γ time differential perturbed angular correlations (TDPAC) studies [25, 26]. Similarly, rhenium radionuclide such as ^{181}Re ($T_{1/2}$ = 19.9 h) owing to its suitable nuclear properties can be useful in radioimmunotherapy, radionuclide synovectomy, bone pain palliation, etc., [27] whereas, ^{101m}Rh ($T_{1/2}$ = 4.3 days) can be used as anti-tumor agent for radiotherapeutic use [28]. The lanthanides ^{153}Gd ($T_{1/2}$ = 240.4 days) finds application in single photon emission computed tomography (SPECT) used for diagnosing neurological disorders, cardiac blockages or abnormalities, and primary and metastasized cancer. ^{153}Gd is also used in industrial radiography, for ascertaining the strength and integrity of structural materials and piping systems [29–31]. Recently terbium radionuclides, especially ^{149}Tb ($T_{1/2}$ = 4.11 h) is under focus as an excellent radionuclide for targeted therapy. However, Maiti [32] decisively showed in her recent paper that no low energy nuclear reaction, i.e. reaction using light or heavy ion projectiles are suitable for production of terbium radionuclides in large quantity required for clinical purpose [32, 33]. We found several terbium radionuclides (e.g., ^{151,153,154}Tb, listed on Table 2), therefore it is expected that α-emitting ^{149}Tb will also be produced in large quantities in proton irradiated LBE targets, which we could not observe because the radionuclide has no suitable γ-energies. ^{157}Dy ($T_{1/2}$ = 8.14 h) is used as a scanning agent for bone and bone marrow [34, 35], while ^{169}Lu ($T_{1/2}$ = 34.06 h) is the parent isotope of ^{169}Lu/^{169}Yb generator system. ^{169}Yb ($T_{1/2}$ = 32.02 days) is gaining interest in brachytherapy [36, 37]. ^{191}Pt ($T_{1/2}$ = 2.802 days) is used to investigate the effect of labeled ^{191}Pt-cisplatin in terms of the antitumor effect and general toxicity on tumor in the body [38]. ^{195m}Hg ($T_{1/2}$ = 41.6 h) is the parent radioisotope of ^{195m}Hg/^{195m}Au generator system and has potential use in angiocardiography [39, 40]. Similarly, ^{205}Bi ($T_{1/2}$ = 15.31 days) is used as a tracer in biodistribution studies and in the design and testing of α-emitting radiotherapeutic agents [41]. Apart from the medical uses, the other radioisotopes such as $_{75}$Se ($T_{1/2}$ = 119.8 days) are also used as tracers to study the selenium adsorption properties in bentonites (clay), migration in plants and fruits [42, 43], whereas ^{95}Nb ($T_{1/2}$ = 34.97 days) is used as a $_{95}$Zr/^{95}Nb chronometer for dating nuclear events [44, 45]. ^{133}Ba ($T_{1/2}$ = 10.5 years) is used as a tracer to study perturbed angular correlation parameters in bovine serum albumin, sorption of radionuclide in estuarine sediments [46].]

Conclusion

This is the first experimental study on the production of radioisotopes by the bombardment of 1.4 GeV protons on thick LBE targets and assessment of the produced radioisotopes by γ-spectroscopy. The production of radionuclides in the LBE targets depends on its mass and intensity of the 1.4 GeV proton beam. Production of ^{206,207}Po is evident in all the samples (1–8 mm). The high activity of polonium radionuclides favors their direct production mechanism rather than production through secondary particle reactions. In the present experimental condition we have not observed any "visible" At activity. The production of various At radionuclides were observed by Tall et al. [14] in very thick LBE targets and was interpreted as secondary particle reactions. David et al. also presented model for production of At by double charge exchange reactions [47]. This experiment therefore leaves ample opportunity to define the limit of "thick" targets from where secondary particle reactions are initiated. The production of various radionuclides via direct nuclear reaction, fission, spallation, fragmentation also opens a window for future prospective regarding the complex nuclear process occurring at the high-energy proton irradiated thick LBE target.

Apart from the valuable information in fundamental science, the product radioisotopes have various applications in the field of medical sciences and technology. The 1.4 GeV proton irradiation on LBE targets for 1 s to 1 min 51 s, produced activities at the end of bombardment (EOB) is in the order of 10^2–10^3Bq for few radioisotopes such as ^{7}Be ($T_{1/2}$ = 53.12 days), ^{74}As ($T_{1/2}$ = 17.77 days), ^{171}Lu ($T_{1/2}$ = 8.24 days), ^{172}Lu ($T_{1/2}$ = 6.70 days), ^{183}Ta ($T_{1/2}$ = 5.1 days) etc. Therefore, it is possible to achieve large-scale production of these radioisotopes, alternative to reactor with high specific activity.

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References

1. Final report of EURISOL design study, J.C. Cornell (ed.), GANIL, France (2009)
2. Tsujimoto K, Sasa T, Nishihara K, Oigawa H, Takano H (2004) J Nucl Sci Technol 41:21–36

3. Ambe S, Chen SY, Ohkubo Y, Kobayashi Y, Iwamoto M, Yanokura M, Ambe F (1991) Chem Lett 20:149–152
4. Lahiri S, Zhi Q, Wen YH, Wu XL (2002) J Radioanal Nucl Chem 252:589–590
5. Ambe S, Chen SY, Ohkubo Y, Kobayashi Y, Maeda H, Iwamoto M, Yanokura M, Takematsu N, Ambe F (1995) J Radioanal Nucl Chem 195:297–303
6. Rémy G, Ralarosy J, Stein R, Debeauvais M, Tripier J (1971) Nucl Phys A 163:583–591
7. Gloris M, Michel R, Herpers U, Sudbrock F, Filges D (1996) Nucl Instrum Meth B 113:429–433
8. Titarenko YE, Shvedov OV, Igumnova MM, Michel R, Mashnik SG, Karpikhin EI, Kazaritskya EI, Batyaev VF, Koldobsky AB, Zhivun VM, Sosnin AN, Prael RE, Chadwick MB, Gabriel TA, Blann M (1998) Nucl Instrum Meth A 414:73–99
9. Gloris M, Michel R, Sudbrock F, Herpers U, Malmborg P, Holmqvist B (2001) Nucl Instrum Meth A 463:593–633
10. Titarenko YE, Batyaev VF, Mulambetov RD, Zhivun VM, Barashenkov VS, Mashnik SG, Shubin YN, Ignatyuk AV (2006) Nucl Instrum Meth A 562:801–805
11. Glasbrenner H, Eikenberg J, Gröschel F, Zanini L (2004) J Nucl Mater 335:270–274
12. Sunil C, Biju K, Sarkar PK (2013) Nucl Instrum Meth A 719:29–38
13. Lahiri S, Maiti M, Dutta B (2010) In: Lahiri S, Maiti M, Das SK (eds) Application of radiotracers in chemical, environmental and biological sciences, vol 3. Saha Institute of Nuclear Physics, Kolkata, India
14. Tall Y, Cormon S, Fallot M, Foucher Y, Guertin A, Kirchner T, Zanini L, Andersson M, Berg K, Frånberg H, Gröschel F, Manfrin E, Wagner W, Wohlmuther M, Everaerts P, Köster U, Ravn H, Noah Messomo E, Jost C, Kojima Y (2007) Int Conf Nucl Data for Sci and Technol doi: 10.1051/ndata:07762
15. Chadha VD, Bhalla P, Dhawan D (2010) Digest Liver Dis 42:446–450
16. Canion B, Landsberger S (2013) J Radioanal Nucl Chem 296:379–382
17. Lahiri S, Nayak D (2002) J Radioanal Nucl Chem 254:289–292
18. Herzog H, Rosch F, Stocklin G, Lueders C, Qaim SM, Feinendegen LE (1993) J Nucl Med 34:2222–2226
19. Deri MA, Zeglis BM, Francesconi LC, Lewis JS (2013) Nucl Med Biol 40:03–14
20. Lahiri S, Maiti M (2012) Radiochim Acta 100:85–94
21. Maiti M, Lahiri S (2010) Phys Rev C 81:024603
22. Maiti M, Lahiri S (2011) Radiochim Acta 99:359–364
23. Choi YH, Lim KM, Yu D, Park HG, Choi YG, Lee CM (2002) Ann Nucl Energy 29:429–446
24. Qadeer R (2013) J Radioanal Nucl Chem 295:1649–1653
25. Lahiri S, Maiti M, Ghosh K (2013) J Radioanal Nucl Chem 297:309–318
26. Yang Z, Zhu H, Lin X (2013) J Radioanal Nucl Chem 295:1371–1375
27. Dutta B, Lahiri S, Tomar BS (2013) Radiochim Acta 101:19–26
28. Skakun Y, Qaim SM (2008) Appl Radiat Isotopes 66:653–667
29. Adak S, Bhalla R, Raj KKV, Mandal S, Pickett R, Luthra SK (2012) Radiochim Acta 100:95–107
30. Nayak D, Lahiri S (1999) J Radioanal Nucl Chem 242:423–432
31. Maiti M, Lahiri S, Tomar BS (2012) J Radioanal Nucl Chem 291:427–432
32. Maiti M (2011) Phys Rev C 84:044615
33. Maiti M, Lahiri S, Tomar BS (2011) Radiochim Acta 99:527–533
34. Lebowitz E, Greene MW (1971) Int J Appl Radiat Isotopes 22:789–790
35. Lahiri S, Nayak D, Das SK, Ramaswami A, Manohor SB, Das NR (1999) Appl Radiat Isotopes 51:27–32
36. Nadi H, Sadeghi M, Enferadi M, Sarabadani P (2011) J Radioanal Nucl Chem 289:361–365
37. Lahiri S, Nayak D, Ramaswami A, Manohor SB (2000) J Radioanal Nucl Chem 243:701–705
38. Soares MA, Mattos JL, Pujatti PB, Leal AS, dos Santos WG, dos Santos RG (2012) J Radioanal Nucl Chem 292:61–65
39. Wackers FJ, Giles RW, Hoffer PB, Lange RC, Berger HJ, Zaret BL, Pytlik L, Plankey M (1982) Am J Cardiol 50:89–94
40. Zaichick V, Zaichick S (2013) J Radioanal Nucl Chem 298:1559–1566
41. Nayak D, Lahiri S, Roy K, Basu S, Ramaswami A (2003) Appl Radiat Isotopes 58:447–450
42. Mukhopadhyay K, Nayak D, Lahiri S (2002) J Radioanal Nucl Chem 251:159–162
43. Suzuki KN, Machado EC, Machado W, Bellido AVB, Bellido LF, Osso JA Jr, Lopes RT (2014) J Radioanal Nucl Chem 299:357–361
44. Douysset G, Petit GL, Gross P, Jutier C (2014) Appl Radiat Isotopes 87:152–156
45. Lahiri S, Mukhopadhyay B, Das NR (1997) J Radioanal Nucl Chem 221:167–171
46. Barros H, Laissaoui A, Abril JM (2004) Sci Total Environ 319:253–267
47. David JC, Boudard A, Cugnon J, Ghali S, Leray S, Mancusi D, Zanini L (2013) Eur Phys J A 49:29