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Polonium Radionuclides that Generate A-Rays from 1h-Irradiated LBE

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Abstract- LBE targets requires precise quantitative and qualitative data. The usage of LBE, which would normally be the best choice for a converter target, is significantly endangered by the presence of α-emitting polonium radionuclides. Research is closely associated with the process of gathering data and understanding about a certain issue. To rephrase, research is the practice of methodically investigating a topic. The 2009 CERN-ISOLDE user's meeting arrived at the consensus that the converter targets should be considered a key source for a number of unusual & potentially therapeutically valuable radionuclides. To detect and measure α-emitting Po radionuclides while there is substantial lead & bismuth around. We achieved this by irradiating LBE targets with medium intensity proton beams, which introduced radiotoxic Po radionuclides into the matrix. We have also investigated the excitation function for the production of different Po radionuclides using LBE (p, xn)$^{207-209}$Po or LBE (p, γ)^{210}Po processes.

Keywords- Polonium, LBE, Radionuclides, LSC, TDCR

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INTRODUCTION

Next-generation RIB facilities will employ large converter targets to create spallation neutrons for fissile material synthesis. A variety of radionuclides are produced when high-energy protons strike the converter target, inducing many nuclear processes, one of which is spallation. Lead-Bismuth eutectic (LBE) has the right combination of characteristics to be a good conversion target.

In addition to several other radionuclides, α-emitting Po radionuclides can be created in the matrix when high energy protons are bombarded into LBE targets. Because it stays in the bloodstream for weeks after inhalation, polonium is a major radioactive worry. ^{210}Po poses a significant risk with a Derived Air Concentration (DAC) value of 10 Bq/m^3 and a half-life of 138.38 days (Buongiorno et al. 2003).

Table 1 contains the nuclear properties of several Po radionuclides that release α-rays.

Table 1 Nuclear properties of the Po radionuclides that emit α-rays (https://www.nndc.bnl.gov/nudat2/)

Radioisotope	Half-life	Decay mode (%)	α–energy (MeV) (I, %)
^{208}Po	2.8 a	α (99.9) ε+β (0.004)	5.115 (100)
^{209}Po	102.0 a	α (99.5) ε+β (0.48)	4.885 (20) 4.883 (80)
^{210}Po	138.3 d	α (100)	4.662 (0.92) 5.300 (100)

The potential for LBE converter targets and the radioprotection of experiment workers are hindered by the radiotoxic Po radionuclides found in the LBE targets. So, the nuclear physics group is concentrating on the most pressing problem: how to reliably measure and forecast, experimentally & conceptually, the production of Po radionuclides from LBE targets. Recognizing & measuring α-emitting Po radionuclides in the LBE matrix is hindered by the presence of stable bulk elements like Pb and Bi, which are one trillion times more abundant than the no-carrier-added (NCA) Po radionuclides produced in the target matrix. The attenuation or complete removal of the α-particles in the LBE matrix would make quantitative

identification of them challenging. Radiochemical methods are the only way to detect α-emitting radionuclides when bulk Pb and Bi are present.

EVALUATION OF THE $^{207–210}$PO RADIONUCLIDE ACCELERATOR

Analysis of the production cross sections of $^{207-210}$Po radionuclides from Bi targets has been documented in many papers. In 1981, Ward et al. investigated the excitation function of ^{209}Bi(p,γ)^{210}Po, ^{209}Bi(p,Γ0) ^{210}Po, and ^{209}Bi(p,2n)^{208}Po reactions from 62-480 MeV projectile energy, as an example. After the target was irradiated, radiochemical methods were devised to extract Po from the bulk Bi. It was lastly counted using a Si surface barrier detector after Po was electroplated onto Ag foil. The overall cross section of the long-lived Po isotopes generated in thin Bi foils (≤10 mg/cm^2) by the ^{209}Bi(p,xn)$^{196-209}$Po reaction was also measured by the same group (Auria et al. 1984) up to 800 MeV projectile energy. Using a gridded-ionization chamber with a pure Bi target in the 30-150 MeV projectile energy range, Daly and Shaw (1964) monitored the ^{209}Bi(p,2n)^{208}Po & ^{209}Bi(p,γ)^{210}Po reactions' cross sections. The ^{209}Bi (p,xn)^{207,208,209}Po reaction was studied by Miyano et al. (1973a, 1974, 1978) using a proton beam with an energy of 10-52 MeV and thin Bi targets ranging from 1.5-1.9 mg/cm^2. The researchers investigated the cross section and recoil ion ranges. After irradiation, the targets were dissolved and then Po ions were electroplated onto thin silver foils.

A surface barrier type semiconductor detector was used for measurement. The cross section of the ^{209}Bi(p,γ)^{210}Po reaction was also measured by the same group utilizing the stacked foil approach and a surface barrier type Si detector from 10 to 50 MeV. According to Miyano et al. (1973b), the maximum cross section measured 0.38 mb with a projectile energy of 12 MeV. In a comparison with Kelly E's (1950) findings, Cohen BL (1955) measured the cross section of the identical reaction from 8-22 MeV proton energy. In their study, Vysotskii et al. (1991) documented the ^{209}Bi(p,γ)^{210}Po reaction's cross section in the lower energy range (4.7-6.5 MeV projectile energy), with the maximum 72 μb cross section recorded at 6.2 MeV. In 1956, Andre et al. investigated the ^{209}Bi(p,xn)^{208,209}Po & ^{209}Bi(p,γ)^{210}Po reactions' cross sections using stacked foils of thin Bi targets (100 μg/cm2) up to a proton energy of 10.65 MeV.

Multiple publications have experimentally estimated the production cross sections of ^{206,207}Po radionuclides that produce γ-rays using medium intensity proton beams. The 207Po production cross section from a thin Bi target that was proton irradiated at 130 MeV was reported by Titarenko et al. (1998). In 1971, Birattari and colleagues used a γ-ray and an X-ray detector to study the excitation function of the ^{209}Bi(p,xn)^{206,207}Po reaction from a projectile energy range of 20-45 MeV. Using off-line γ-spectrometry, Chung et al. (2011) estimated the cross-section of the ^{209}Bi(p,3n)^{207}Po reaction from projectile energies ranging from 18 to 551 MeV. The cross section of ^{209}Bi(p,3n)^{207}Po was determined by MokhtariOnarj et al. (2017) using an HPGe detector from 62-100 MeV proton energy.

Reports on the creation of α-emitting Po radionuclides from proton irradiated LBE targets are rare, even if there are few reports on the generation of Po radionuclides from Bi targets. Producing ^{207,208,210}Po radionuclides from a proton-irradiated thin LBE target (7–7.6 mg/cm2) at 8.95–21.95 MeV projectile energy is detailed in this chapter. In order to quantify polonium, previous research mostly used α-spectrometry. We have used the LSC in conjunction with the triple to double coincidence ratio (TDCR) method to quantify the Po radionuclides that generate α-rays. Hence, the fundamentals of LSC & LSC-TDCR method have been stated below before the experiments themselves are described.

LIQUID SCINTILLATION COUNTER (LSC)

This technique is used for measurement of alpha or beta emitting radionuclides. The basic principle of LSC relies upon the conversion of kinetic energy from nuclear decay to light energy. In LSC, sample in the liquid state is mixed thoroughly with a cocktail solution which acts as the detector. The cocktail solution (also known as scintillator) consists of solvents, solutes and surfactants. Functions and examples of each of these components have been given in Table 4.2. The energy from the radioactive decay is first transferred to the surrounding organic solvents which upon de-excitation gives the energy to the solute molecules by non-radiative transfer mechanisms. There are two types of solutes present in most of the cocktails, i.e. primary and secondary. These primary and secondary solutes are fluorophores, release the excess energy in the form of light which then gets converted into electrical signal. The intensity of the signal is directly proportional to the original nuclear decay energy dissipated in the cocktail. A schematic representation of the entire technique is shown in Fig. 2.

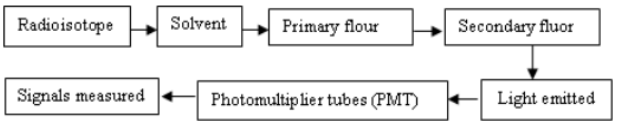

Figure 1: Schematic representation of the LSC technique

Owing to intimate mixing of the sample with the detector and high linear energy transfer (LET) of α–particles, the α-emitting radionuclides can be measured with 100% counting efficiency by this technique. Alongside LSC yields relatively fast results due to high counting sensitivity for both α and β emitting radionuclides and several samples can be measured in a sequence (Eikenberg et al. 2014).

Table 2 Different components in a scintillation cocktail

Component (% of total solution)	Concentration in cocktail	Examples	Function
Solvents (60-99%)	10 M	Toluene, xylene, dioxane	Absorbs kinetic energy from the radioactive decay and transfers energy to solutes.
Primary Solutes or fluors (0.3-1%)	0.01 M	2,5-diphenyloxazole (PPO), 2,5-diphenyl-1,3,4-oxadiazole (PPD)	Absorbs energy from solvent and emits light
Secondary solutes or wavelength shifters	0.0001 M	1,6-diphenylhexa-1,3,5-triene	Absorbs light from primary solutes and emits at longer wavelength
Surfactants	>1 M	Triton X, alkyl phenol ethoxylates	Ensures mixing of organic solvent and aqueous sample solution

The activity of a radionuclide is estimated in LSC either by comparing the counts with a standard or by calculating the detection efficiency with the help of "Free parameter model". The free parameter can be deduced from this model in two ways: from the measurement of a tracer or calculation of coincidence ratio in a specific three Photomultiplier tubes (PMTs) liquid scintillation counter. The latter is known as triple to double coincidence ratio (TDCR) method (Casette and Do, 2008). The concept of TDCR has been discussed in detail in the next section.

TDCR method

Conventionally, the LSC counters were equipped with 2 PMTs while the modern LSC instruments are equipped with 3 PMT systems (Fig 2) which allows measuring the ratio of triple coincidences to the logical sum of double coincidences. This ratio is known as TDCR or triple to double coincidence ratio, a standardization technique of LSC. TDCR is a much simpler approach and it enables the determination of absolute activity without the use of calibration standards as TDCR itself is equivalent to the efficiency of the system (Krapiec M, 2011).

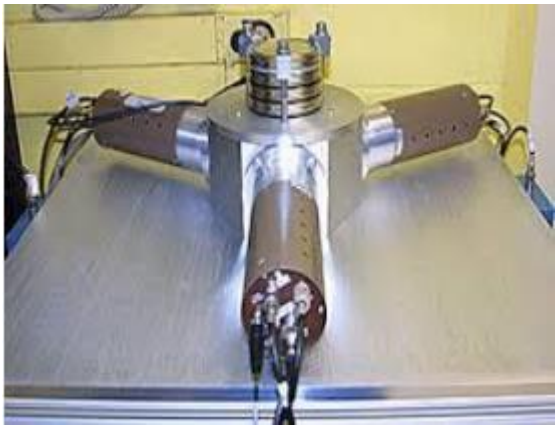

Figure 2: Three PMT in modern LSC-TDCR counters at 120◦ angle

Mathematical expression of TDCR: If an electron of energy E produces mean number of m photons in a scintillator, then the probability of emission of x photons for a mean value m is given by Poisson's law:

$$P(x/m) = m^x e^{-m}/ x!$$

The extreme case would be detection of zero photons; in that case, x tends to 0. Thus non-detection probability in case of one PMT would be

$$Z = P(0/m) = e^{-m}$$

Since detection probability is the complement of non-detection probability, therefore detection probability or efficiency (ε)

$$\varepsilon = 1\text{-}Z = 1\text{-}e^{-m}$$

The value of detection efficiency would vary between 0 and 1 signifying two extreme cases of no interaction and strong interaction between the decaying isotope and cocktail respectively. Therefore efficiency of three PMTs connected in a coincidence circuit can be drawn from equation (ii) as follows:

$$\varepsilon = (1\text{-}e^{-m})\,(1\text{-}e^{-m})\,(1\text{-}e^{-m}) = (1\text{-}e^{-m})^3$$

Thus efficiency (εT) for triple coincidence,

$$\varepsilon_T = (1\text{-}e^{-m})^3$$

In case of double coincidence (εD),

$$\varepsilon_D = (1\text{-}e^{-m})^2\, e^{-m}$$

The above equation shows the coincidence probability for two PMTs with zero detection in the third tube. This is possible for two other combinations (A total of three PMTs, thus for double coincidence, three cases are possible). So,

The sum of double coincidences efficiency = $3(1\text{-}e^{-m})^2 e^{-m}$

Thus, in total we have four combinations i.e. one triple and three double. The logical sum of all these four possible combinations, i.e. sum of all coincidences;

$$All = (1\text{-}e^{-m})^3 + 3(1\text{-}e^{-m})^2\, e^{-m}$$

$$= (1\text{-}e^{-m})^2\,[(1\text{-}e^{-m}) + 3e^{-m}]$$

Multiplying all the terms we can express binomially as;

$$3(1\text{-}e^{-m})^2\text{-}2(1\text{-}e^{-m})^3$$

Thus the ratio of triple coincidences to all coincidences referred as TDCR is,

$$TDCR = (1\text{-}\,e^{-m})^3/[3(1\text{-}\,e^{-m})^2\text{-}2(1\text{-}\,e^{-m})^3]$$

Considering two extreme cases, the number of electrons produced in the PMT tends to a very high number or to zero value, thereby limiting the TDCR value between 0 and 1. TDCR value of 1 signifies that all photons produced by the αparticle scintillation process are registered as triple and double coincidences which confirmed 100% counting efficiency.

Optimisation of α/β separation in LSC

The efficiency of converting an incident radiation into a scintillation pulse is ~10 times higher for the β-particles than for the α-particles. The energy scale of LSC is calibrated with β–sources therefore an α-particle appears at 1/10 of its energy. For example: A 5 MeV α-particle will appear at around 500 keV. Therefore there will be overlap of pulses from α-particles and β-particles (Edler and Passo, Perkin Elmer). But duration of α-induced scintillation is longer than β-induced one which makes it possible to separate the two. Optimisation of α/β separation is an important aspect during measurement of the α-emitters. TDCR offers the advantage of discriminating between α/β spectra.

The basic principle behind α/β discrimination in LSC is based on the fact that alpha particles form more triplet excited states upon interacting with the solvent and the scintillating molecules than the beta particles. Therefore, higher fraction of delayed fluorescence is observed in the case of alpha particles producing long tail alpha pulses (Fig. 3) that can be separated from the predominantly short tail beta pulses. The light flash duration of beta pulses are 5 ns while α pulses are 10 ns. The LSC-TDCR technique therefore discriminates α/β emitters based on their pulse length and enables simultaneous determination of both α and β activity without radiochemical separation.

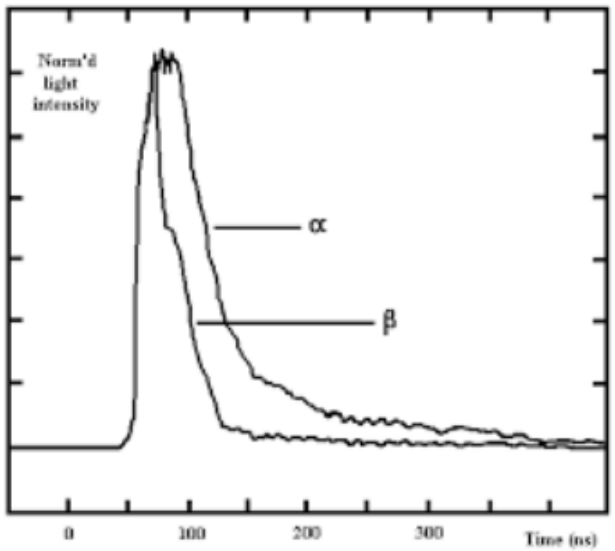

Figure 3: Different pulse length of alpha and beta

In this work, α-activity of polonium radionuclides has been measured by the LSC-TDCR technique after α/β optimisation. Post quantification of polonium radionuclides at different projectile energies, attempt has been made to identify which α-emitter was produced at different projectile energies. We directly measured the Po radionuclides in a complex matrix, i.e. in presence of bulk amount of Pb and Bi. Apart from that, yield and cross section of γ-emitting 207Po was also measured at that energy. To the best of our knowledge, this is the first systematic report on the quantification of α-emitting Po radionuclides produced in LBE targets bombarded by low energy protons of different energies, using the LSC-TDCR technique. However, earlier LSC technique was used to measure Po radionuclides in environmental matrices (Maxwell et al. 2013, Begy et al. 2015) or in some cases to measure vapour pressure of released Po (Neuhausen et al. 2004, Gonzalez Prieto et al. 2014a,b,c, Gonzalez Prieto et al. 2016).

EXPERIMENTAL

Chemicals and reagents

Six LBE targets were prepared by rolling thick LBE foils at the BARC-TIFR target laboratory. The target thicknesses were 7 mg/cm2 (LBE1) and 7.6 mg/cm2 (LBE2 to LBE6). HNO3 (65%) was procured from MERCK and the aqueous Aqualite Ultra-low level cocktail was procured from HIDEX. Deionised water (18.2 MΩcm) was obtained from Thermo Scientific Barnstead Smart2Pure Water Purifying System.

Irradiation details

All the LBE foils were coated with two component glue, 'Araldite', to prevent the loss of volatile Po radionuclides. The targets were irradiated 'one at-a-time' with proton energies from 9-22 MeV at Bhabha Atomic Research Centre-Tata Institute of Fundamental Research (BARC-TIFR) pelletron, Mumbai. Instead of the stacked foil technique, irradiation of one target at a time would minimize the spread of projectile energy. The beam current was measured with the help of a Faraday cup placed at the rear of the target in combination with a current integrator (Danfysik). For each of the targets, the exit energies were calculated by the software SRIM (Zeigler and Bierserk, 2013). The summary of irradiation details is given in Table 3.

Table 3 Irradiation details of the six LBE targets

Target	Thickness (mg/cm^2)	Incident energy (MeV)	Exit Energy (MeV)	Energy at the centre of mass (MeV)	Integrated Charge (μC)
LBE1	7.0	22	21.9	21.95	3000
LBE2	7.6	20	19.9	19.95	3000
LBE3	7.6	18	17.9	17.95	3000
LBE4	7.6	15	14.9	14.95	3000
LBE5	7.6	12	11.9	11.95	3000
LBE6	7.6	9	8.9	8.95	3000

Measurement of ^{207}Po by γ-spectrometry

Before dissolution of the samples, series of gamma spectra were taken in a p-type HPGe detector of resolution 2.35 keV at 1.33 MeV in combination with a digital spectrum analyzer (DSA 1000, CANBERRA) and Genie 2K software (CANBERRA),

which enabled us to measure ^{207}Po quantitatively. The radionuclides were identified from their corresponding photo-peaks and decay data. The energy and efficiency calibrations of the detector were performed using a standard 152Eu (T1/2= 13.53 a) source of known activity.

Measurement of α-emitters by LSC-TDCR technique

For measuring α-activity, LS-spectrometer HIDEX 300SL optimized with triple to double coincidence ratio (TDCR) was used (Fig. 4).

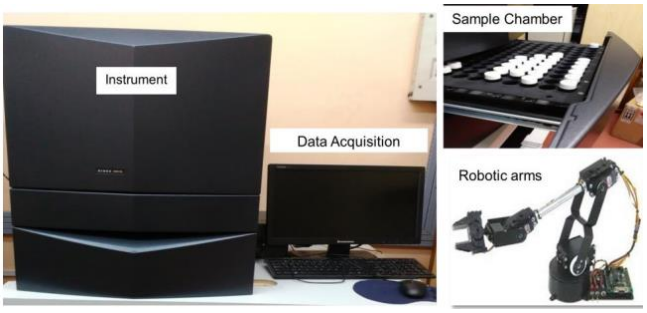

Figure 4: HIDEX 300SL instrument at SINP, Kolkata used for α-analysis

The glue coated LBE foils were dissolved in 4 mL 3 M HNO_3 solution by sonication for 15-20 minutes. High concentration of dissolved solutes (bulk Pb and Bi) and concentrated acids are most likely to quench the light output in liquid scintillation counting leading to an overlapped α/β spectrum. Therefore, efforts were made to optimise the α/β separation. We optimised the α/β separation by adjusting the sample: cocktail ratio and the pulse length index (PLI). PLI is the horizontal line which discriminates the alpha from the beta pulses. Minimum sample: cocktail ratio offered better separation. For better understanding of the optimization process, few intermediate steps of α/β optimization with different combinations of sample: cocktail ratio and PLI has been shown in Fig. 5 whereas Fig..6 depicts a typical 2D plot for the system where the α and β clouds are visually well separated. The individual α-spectrum obtained after the α/β optimization at PLI 6 is shown in Fig. 7

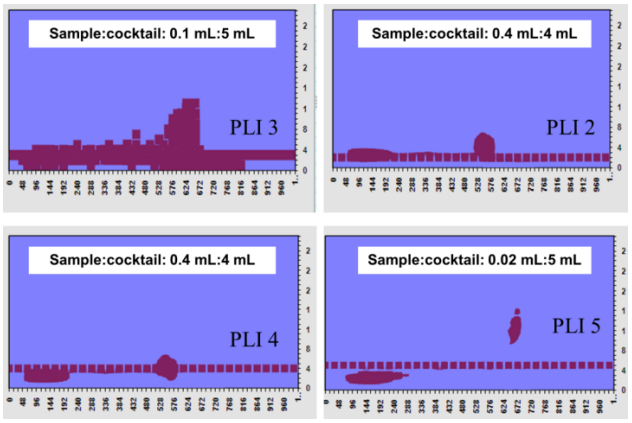

Figure 5: Two-dimensional illustration of few intermediate stages of α/β optimization by varying the sample: cocktail ratio and pulse length index (PLI) (y axis). x-axis represent channel numbers

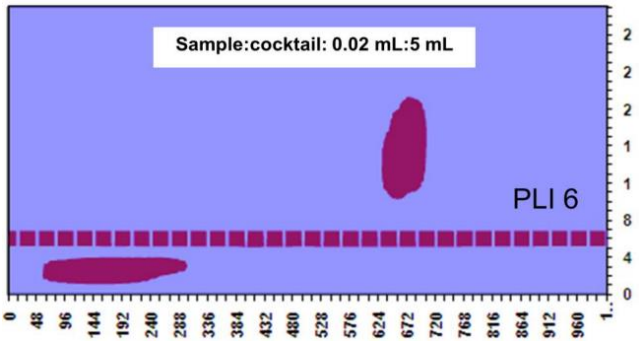

Figure 6: A well separated α/β spectrum of LBE target at 18 MeV

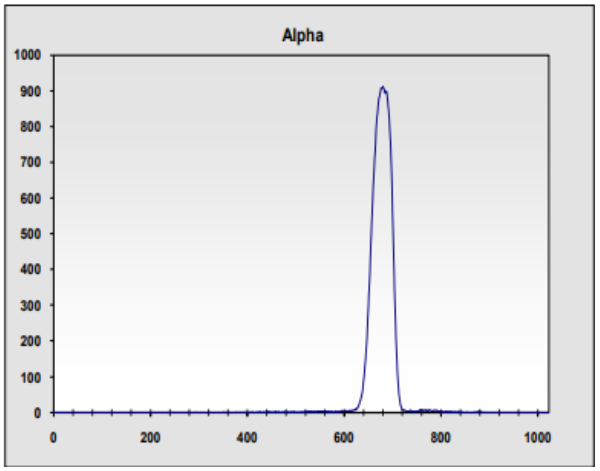

Figure 7: LS α-spectrum of Po radionuclides in LBE after α/β optimization. The x axis represents channel number while the y axis shows counts

Post α/β optimization, 5 mL Aqualite cocktail was added to 0.02 mL of each stock solution and was shaken to obtain a homogeneous, clear solution. Only exception was LBE6, where 5 mL Aqualite cocktail was added to 0.1 mL stock solution. The blank was prepared in similar way by addition of 0.02 mL of deionised water to 5 mL of the same cocktail. Each sample was counted for 5000 s and count of the blank was duly subtracted. The counts thus obtained for 0.02 mL stock solution were corrected for 4 mL (total volume in which each sample was dissolved). TDCR values of α-emitting radionuclides were found to be 1 in majority of the cases.

RESULTS AND DISCUSSIONS

Measurement of ^{207}Po by γ-spectrometry

The γ-spectrum of LBE1 target irradiated at 22 MeV proton energy reveals the formation of ^{203}Pb, ^{203,204,205,206}Bi and ^{207}Po in the matrix (Fig. 8). The

nuclear characteristics of the produced isotopes have been mentioned in Table 4

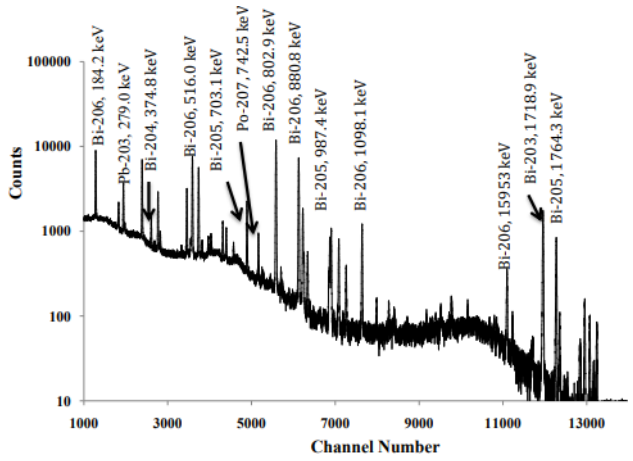

Figure 8: Gamma spectrum of LBE1 target at 22 MeV taken 30.5 h from the EOB

Table 4 Nuclear characteristics of the produced γ-emitting radionuclides (https://www.nndc.bnl.gov/chart/)

Radio-nuclides	Half-life	Decay mode (%)	E_γ(keV) [I_γ(%)]	Contributing reactions	E_{Th} (MeV)
^{206}Bi	6.2 d	ε+β$^+$ (100)	183.9 (15.8) 343.5 (23.4) 398.3 (10.7) 516.2 (40.7) 803.1 (99.0) 881.0 (66.2)	^{206}Pb(p,n) ^{207}Pb(p,2n)	4.56 11.33
^{205}Bi	15.3 d	ε+β$^+$ (100)	703.4 (31.0) 1764.0 (32.5)	^{206}Pb(p,2n)	11.63
^{204}Bi	11.2 h	ε+β$^+$ (100)	374.7 (82.0)	^{204}Pb(p,n)	5.27
^{203}Bi	11.7 h	ε+β$^+$ (100)	820.3 (29.6) 825.2 (14.6)	^{204}Pb(p,2n)	12.50
^{203}Pb	51.8 h	ε (100)	279.2 (81.0)	^{204}Pb(p,pn)	8.40
^{207}Po	5.8 h	ε+β$^+$ (99.9) α (0.021)	742.6 (28.2) 911.7 (16.9) 992.3 (59.3)	^{209}Bi(p,3n)	18.10

There is only one production route for ^{207}Po i.e. ^{209}Bi(p,3n) reaction which has a threshold energy of 18.1 MeV. Therefore, ^{207}Po activity was only observed at 19.95 MeV and 21.95 MeV. After quantifying γ–emitting ^{207}Po at those two energies, production cross sections of LBE(p,3n)^{207}Po [produced through ^{209}Bi(p,3n)^{207}Po] reaction at 19.95 MeV and 21.95 MeV were also estimated using the following formula:

$$A = n\sigma(E)\varphi(1 - e^{-\lambda T})$$

where, A= Activity (Bq) of a particular radionuclide at the EOB

σ (E)= Cross section of formation of the radionuclide at an incident energy E

φ= Beam density in particle/s

n= No. of atoms/cm2

λ= Disintegration constant

T= Irradiation time

Various sources of errors were considered. Error due to counting varied from 1.8% to 10.2%, error in target thickness (~5%), errors in beam current and beam energy (~10%), etc., were also taken into consideration. The total error related to the cross-sectional measurement of ^{207}Po was found to be 11-15%. The yields of ^{207}Po at the EOB and the cross sections have been shown in Table 5.

Table 5 Yield and cross section of ^{207}Po at the EOB at different energies

Proton Energy (MeV)	Yield at EOB, kBq	Cross section (mb)	Cross sections (mb) of other references (normalized from Bi target to LBE target)
21.95	1383±23.9	277±32	167±8[Birattari et al. 1971]
19.95	151±11.0	23±2	25±1[Biratarri et al. 1974], 60[Miyano et al. 1974]

The measured cross sections were compared with other experimental data of Miyano et al. (1974), Birattari et al. (1971). Both the authors used pure Bi target, therefore, while comparing the cross section, it was scaled down to the composition of LBE wherein Bi is only 55.5%. The experimental cross section obtained at 22 MeV is higher than that of the reported value by Birattari et al, probably because we have not allowed Po to escape from the matrix. At 20 MeV, our cross section is comparable with Birattari et al. However, Miyano et al reported higher cross section.

Measurement of α–emitters by LSC-TDCR

Before α-counting by the LSC-TDCR technique the samples were kept aside for two months, so that all the γ-emitting isotopes listed in Table 4 decayed out. The total activity of the α-emitting Po radionuclides have been measured by the LSCTDCR counter and shown in Table 6.

Table 6 Total activity of the α-emitting Po radionuclides at 58th day from the EOB

Energy (MeV)	CPM in 0.02 mL	DPM in 0.02 mL	TDCR	Activity in total 4 mL solution (Bq)
21.95	295.1	295	1.0	983.3
19.95	256.5	257	0.999	846.7
17.95	530.4	531	1.0	1770
14.95	274.0	274	1.0	913.3
11.95	21.6	22	1.0	73.3
8.95*	1.4*	2*	0.985	1.3

*In this sample, CPM & DPM values in 0.1 mL solution are given

Identification of α-emitting Po radioisotope

Proton energy up to 22 MeV could produce either of α-emitting $^{208\text{-}210}$Po or a mixture of them. LSC is a quantitative tool, not a qualitative one. However, attempt has been made to get an indication of which isotope of Po has been produced in the matrix. For the identification of α-emitting Po radionuclides, time resolved alpha counts were taken for 2-13 months from the EOB. Fig. 9 shows the plot from 58th day to 13 months, resulting in a half-life of 2.64 a, which

is comparable but slightly less than the half-life of ^{208}Po (2.898 a).

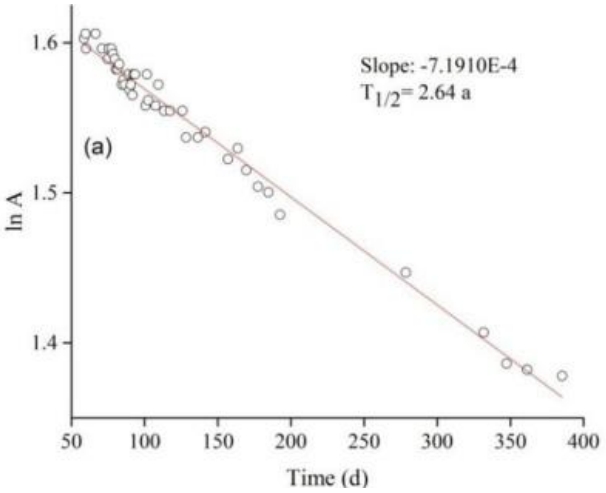

Figure 9: Time resolved plot of α-counts from 58th day to 13 months after the EOB (LBE foil irradiated at 21.95 MeV)

The later part of the above figure, i.e. data from 136 d to 385 d, i.e. after elapsing one half-life of ^{210}Po (138.3 d), if produced, was plotted again (Fig. 10). The half-life of the α-emitting isotope obtained from Fig. 10 is 2.81 a, closer to 208Po.

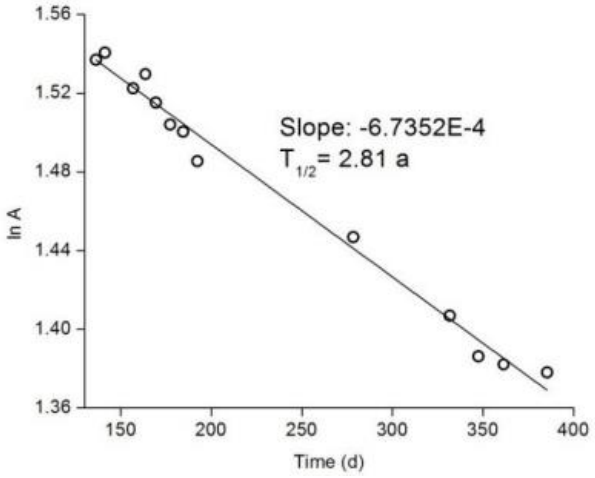

Figure 10: Time resolved plot of α-counts from 136 d to 13 months after the EOB (LBE foil irradiated at 21.95 MeV)

Therefore, it is concluded that in the initial stages there was slight mixture of ^{210}Po along with ^{208}Po. ^{210}Po has a half-life of only 138 d and therefore decayed out with time, leaving only ^{208}Po in the solution.

Calculation of production cross section of 208,210Po

The cross section of ^{209}Bi(p,2n) reaction was calculated by the activation formula mentioned. The cross sections were plotted against different projectile energies and compared with the experimental work by Miyano et al. 1973a (Fig. 11). The total error related to the cross-sectional measurement of ^{208}Po i.e. error in counting, target thickness, beam current, etc. was estimated to be 11-14%. The cross-sectional values were higher than that of Miyano et al. This may again be ascribed to the fact that we have used glue to prevent volatilization of Po during irradiation and the samples were measured directly without any radiochemical separation.

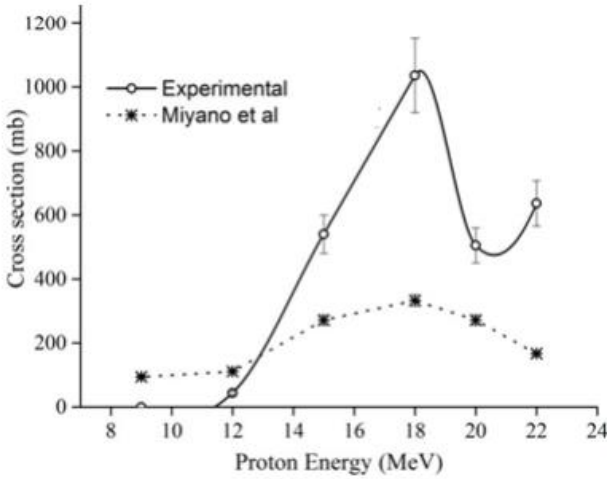

Figure 11: Excitation function of LBE(p,2n)208Po reaction

We have also tried to evaluate the cross section of ^{210}Po. For this we considered that at a later stage of counting, e.g., at 361 d, the total yield (A) is only due to ^{208}Po, when 2.6 half-lives of ^{210}Po have already passed and a negligible amount of ^{210}Po might be present at that time. From this data, we calculated the yield of ^{208}Po at the 58th day (B), which is subtracted from the measured yield (C) on that day. The difference (C−B) is attributed to the contribution of ^{210}Po on the 58th day, this yield for ^{210}Po was again calculated to the EOB. The cross section of ^{210}Po was found in μb range. The cross sections of both ^{208,210}Po are given in Table 7.

Table 7 Cross section of α-emitting 208,210Po at the EOB by LSC-TDCR technique

Proton Energy (MeV)	^{208}Po Cross section, mb	^{210}Po Cross section, μb
21.95	636±71	7.5
19.95	505±55	19
17.95	1036±116	22
14.95	540±60	9
11.95	44±5	1
8.95	0±0	2

The cross section of the ^{209}Bi(p,γ) ^{210}Po reaction has been compared with the TENDL-2017 (TALYS Evaluated Nuclear Data Library) by Koning and Rochman (2012). Our experimental results were also compared with the works of Miyano et al. (1973b), Kelly E (1950) (Fig. 12). Both our data and the evaluated data by Koning and Rochman showed

an extremely low cross section for the $^{209}Bi(p,\gamma)$ ^{210}Po reaction. Our data was found to be the closest data set when compared to the evaluated data. The cross section data in this energy range given by Miyano et al. (1973b) and Kelly E (1950) are higher by more than two orders of magnitude with the evaluated data. This shows that LSC-TDCR is an efficient technique to measure α-emitting isotopes even at very low concentration. The slight deviation between our data and the TALYS evaluated data might be due to low statistics, and there may also be some spurious light output in the LSC system, which influences such low statistics but is otherwise insignificant.

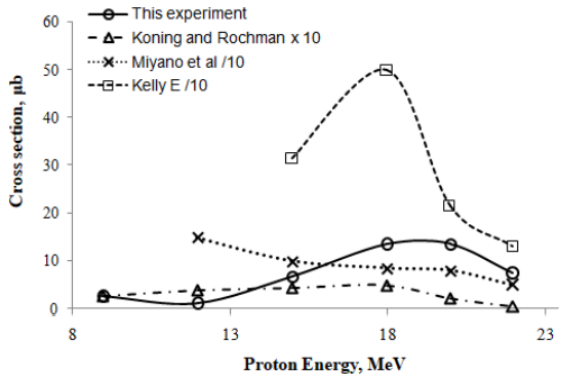

Figure 12: Cross section plot of the $^{209}Bi(p,\gamma)$ ^{210}Po reaction at different energies and comparison with the available experimental and evaluated data

CONCLUSION

There are substantial risks associated with using LBE as a spallation target and coolant in ADS due to the α-emitting polonium radionuclides. The importance of accurately estimating the amount of polonium created within the LBE targets during proton bombardment cannot be overstated. Traditionally, alpha spectroscopy has been used to detect Po, although a liquid scintillation counter provides more precise results. Even with bulk Pb and Bi present, Po in a complicated matrix might be estimated using modern LS measurement techniques that are equipped with TDCR & α/β separation. No radiochemical separation was employed in this initial report on the systematic measurement of $^{208,210}Po$ generated from LBE at varying proton energy. It was feasible to measure ^{210}Po since the LSC-TDCR approach is sensitive even when identifying extremely low abundance αemitting radionuclides. The amount of ^{210}Po produced is so little in the experimental energy range that it is completely unwarranted. Different proton energies have also been used to quantify short-lived γ-emitting ^{207}Po.

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