

First report on the inventory of radionuclides in 1.4 GeV proton irradiated thick Lead-Bismuth Eutectic (LBE) target

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This work is a part of LIEBE Project



Liquid euctectic Pb/Bi loop for EURISOL **LIEBE project**



Final Report of the EURISOL Design Study 2009, J. Cornell Ed., GANIL

LIEBE project: Instituts and timeline

WP Definition	<u>WP Holder</u>
WP1: Coordination	CERN
WP2: Conceptual Design and Simulation	SCK-CEN
WP3: Construction, Assembly	CERN
WP4: Instrumentation	CERN
WP5: Safety and Licensing	CEA
WP6: Target characterization and analysis	PSI
WP7: Radiochemistry	SINP
WP8: Offline Commissioning	IPUL
WP9: Online Operation	CERN

					LIEBE pro	ject planni	ng - endors	sed 2nd SC	, Lisbon, O	ct 2012			
WP	Q3-12	Q4-12	Q1-13	Q2-13	Q3-13	Q4-13	Q1-14	Q2-14	Q3-14	Q4-14	Q1-15	Q2-15	2018
					CERN-w	ide Long S	Shutdown 1	1					
2 Design			1 st design	CFD, FEM		Final design							
3 Assembl	I					Techn. drawings	componen ts	Procurem ent	Constructi on	Adaptation			
4 Instr.			Definition		Tests@ip ul/cern			software					
5 Safety		CERN authorities	Safety constraint s	Risk analysis + RP			Risk analysis + RP II			Safety file	Beam permit		
6 Target charact					Release (Po)	Inventory							Waste PIE
7 Chemist	:	Activation Pb/Bi			Compar MC code				Activation Pb/Bi-II				
8 Offline		Test pumps IPUL	Test IPUL CERN						commissi oning	commissi oning	Cold check-out		
9 Online		Operation al parameter s		Definition of online tests				Modif target area I			Modif target area II	1 week online – Report	Final report



Aim of this experiment:

Tomaketotalinventoryandquantificationoftheradionuclidesproducedbybombardmentof1.4GeVproton on LBE targets.

 To investigate the production of highly radiotoxic Po and At radionuclides occurring through secondary particle reaction channels.

Experiment:

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- Eight cylindrical LBE samples (Fixed diameter of 6 mm and lenghts from 1 to 8 mm) weighing between 0.43 to 2.83 g.
- LBE samples were encapsulated.
- Irradiation with 1.4 GeV proton beam using RaBIT setup at ISOLDE.
- Irradiation time 1-3 s.
- Intensity of the proton beam was 3.18×10¹² protons/pulse for 1 and 2 mm length target and 3.22×10¹³ protons/pulse for the rest.
- Time resolved gamma measurements.

<u>RaBIT (rapid proton beam irradiation transport) irradiations</u> <u>setup:</u>

- Pneumatic transport system installed in class A lab at ISOLDE
- Samples sent in shuttles into the proton beam line in front of HRS front-end
- Samples transported to class C lab after reduction of dose rate below 10 uSv/h (cooling periods of 7-24 hrs)



Measured activity (Bq) of the long lived (>5 d) radionuclides at EOB in sample #8

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Isotopes (T _{1/2})	Activity (Bq)	Isotopes (T _{1/2})	Activity (Bq)	Isotopes (T _{1/2})	Activity (Bq)
⁷⁴ As (17.77 d)	130±6	^{114m} In (49.5 d)	61±7	⁸⁵ Sr (64.84 d)	34±1
¹³¹ Ba (11.5 d)	89±2	¹⁷¹ Lu (8.24 d)	2507±447	¹⁸³ Ta (5.1 d)	1544±125
⁷ Be (53.12 d)	236±50	⁵⁴ Mn (0.85 yr)	3±0.4	¹²¹ Te (16.78 d)	85±9
²⁰⁵ Bi (15.31 d)	2783±99	⁹⁵ Nb (34.975 d)	150±5	^{121m} Te (154 d)	1±0.1
²⁰⁷ Bi (31.55 yr)	7±0.8	¹⁸⁵ Os (93.6 d)	286±5	²⁰² Tl (12.23 d)	965±34
¹³⁹ Ce (137.6d)	5±0.05	¹⁴³ Pm (265 d)	7±0.8	¹⁶⁷ Tm (9.25 d)	517±91
¹⁴⁷ Eu (24.1 d)	308±45	²⁰⁶ Po (8.8 d)	609±18	¹²⁷ Xe (36.4 d)	13±2
¹⁴⁹ Eu (93.1 d)	66±6	¹⁸⁸ Pt (10.2 d)	1753±78	⁸⁸ Y (106.65 d)	65±0.8
⁵⁹ Fe (44.5 d)	23±1	⁸³ Rb (86.2 d)	45±1	¹⁶⁹ Yb (32.02 d)	83±6
¹⁴⁶ Gd (48.27 d)	3±0.2	¹⁰³ Ru (39.26 d)	71±13	⁶⁵ Zn (244.3 d)	4±0.9
¹⁴⁹ Gd (9.28 d)	145±9	⁴⁶ Sc (83.8 d)	7±0.3	⁹⁵ Zr (64.02 d)	31±0.7
¹⁵³ Gd (240.4 d)	I±0.2	⁷⁵ Se (119.8 d)	2±0.1		
¹⁷² Hf (1.87 yr)	3±0.9	^{117m} Sn (13.6 d)	11±4		

Comparison on the presence of Radioisotopes in various samples

Radionuclides (T1/2)	Sample 1	Sample 2	Sample 7	Sample 8
72-As (26.0 h)				
74-As (17.77 d)			_	
110m-Ag (249.79d)				
192-Au (4.94 h)				
194-Au (38.02 h)				
131-Ba (11.5 d)				
135m-Ba (28.7 h)				
7-Be (53.12 d)				
203-Bi (11.76 h)		_		
204-Bi (11.22 h)				
205-Bi (15.31 d)				
206-Bi (6.243 d)				
207-Bi (31.55 yr)				
76-Br (16.2 h)				
77-Br (57.036 h)				
82-Br (35.30 h)				
139-Ce (137.6 d)				



58-Co (70.86 d) 60-Co (5.27 yr) 127-Cs (6.25 h) 129-Cs (32.06 h) 153-Dy (6.4 h) 157-Dy (8.14 h) 171-Er (7.516 h) 146-Eu (4.61 d) 147-Eu (24.1 d) 149-Eu (93.1 d) 59-Fe (44.5 d) 69-Ge (39.05 h) 77-Ge (11.30 h) 146-Gd (48.27 d) 147-Gd (38.06 h) 149-Gd (9.28 d) 153-Gd (240.4 d) 170-Hf (16.01 h) 171-Hf (12.1 h) 172-Hf (1.87 yr) 173-Hf (23.6 h) **193m-Hg** (**11.8** h) 195m-Hg (41.6 h) **197-Hg (64.14 h)** 203-Hg (46.59 d) 111-In (2.8047 d) 114m-In (49.5 d) 185-Ir (14.4 h)



	186-Ir (16.64 h)
/	187-Ir (10.5 h)
	188-Ir (41.5 h)
1	190m2-Ir (3.25 h)
	195m-Ir (3.8 h)
	42-K (12.360 h)
	79-Kr (35.04 h)
	132-La (4.8 h)
	160 L n (34.06 h)
	170 L (2012 d)
	170-Lu (2.012 d)
	171-Lu (8.24 d)
	172-Lu (6.70 d)
	173-Lu (1.37 yr)
	177-Lu (6.64 d)
	54-Mn (0.85 yr)
	95-Nb (34.975 d)
	96-Nb (23.35 h)
	147-Nd (10.98 d)
	185-Os (93.6 d)
	193-Os (30.11 h)
	200-Pb (21.5 h)
	201-Pb (9.33 h)
	203-Pb (51.873 h)
	100-Pd (3.63 d)
	143-Pm (265 d)









Elements identified in the Periodic Table

0

	1																	18
1	1																	2
	н																	He
	1.008	2											13	14	15	16	17	4.003
2	3	4											5	6	7	8	9	10
	Li	Be											в	с	N	0	F	Ne
	6.941	9.012											10.811	12.011	14.007	15,999	18,998	20.150
3	11	12											13	14	15	16	17	18
	Na	Mg											Al	Si	P	S	Cl	Ar
	22.990	24.305	3	4	5	6	7	8	9	10	11	12	26.952	28.086	30.974	32.065	35.453	39.948
4	19	20	21	22	23	24	25	26	27	28	29	30	31	32	33	34	35	36
	K	Ca	Sc	Ti	v	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
	39.098	40.078	44.956	47.867	50,941	51.995	54.938	55.845	58.933	58.693	63.546	65.409	69.723	72.64	74.922	78.96	79.904	\$3,798
5	37	38	39	40	41	42	43	44	45	46	47	48	49	50	51	52	53	54
	Rb	Sr	Y	Zr	Nb	Mo	Τc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe
	85.468	\$7.63	88.906	91.224	92.906	95.94	[98]	101.07	102.905	106.42	107.868	112.411	114.818	118.710	121.760	127.60	126,904	151.295
6	55	56	57-71	72	73	74	75	76	77	78	79	80	81	82	83	84	85	86
	Cs	Ba	Lantha- noid:	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn
	132.005	137.527		178.49	180.947	185.84	186.207	190.25	192.217	195.084	196.967	200.59	204.385	207.2	208.980	209	210	222
7	87	88	\$9-103	104	105	106	107	108	109	110	111	112					-	
	Fr	Ra	Actinoids	Rf	Db	Sg	Bh	Hs	Mt	Ds	Rg	Ср						
	223	226		261	262	265	277	264	278	281	272	285						

57	58	59	60	61	62	63	64	65	66	67	68	69	70	71
La	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
138.905	140.116	140.P0S	144.242	[145]	150.36	151.964	157.25	158.Ø25	162.500	164.930	167.259	165.934	173.04	174.967
89	90	91	92	93	94	95	96	97	98	99	100	101	102	103
Ac	Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr
227	232.038	231.036	238.029	237	244	243	247	247	251	252	257	258	259	262





Remarks

> Radioisotopes identified in the samples ranged from 7-Be (53.12 d) to 207-Po (5.8 h) in the Periodic Table.

> The total number of identified radioisotopes in all the four samples are 112. Apart from this, there are 12 more radioisotopes which could not be confirmed due to overlap of photo-peaks and similar half life for different radionuclides.

> Most of the radioisotopes identified in the samples belongs to 4th, 5th and 6th Period.

> The formation of Po radionuclides is confirmed in the samples. But there is no sign of formation of At radioisotopes in the samples.

Remarks

>The modes of production of the radionuclides are mainly fission, electron capture decay of the fission products, fragmentation, spallation, etc,. Also, typical reactions can be found in thick targets like 209Bi(p,pixn)210-xPo or 206Pb(alpha,xn)210-xPo.

> The activity of the long lived radionuclides at the End Of Beam (EOB) in the experimental condition have been calculated.

>It has been difficult to calculate the activity of short lived radionuclides, because of high Compton background in the initial spectra.

<u>lts:</u>

roduction	of ²¹⁰ At	$(T_{1/2} =$	8.3 h)
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ength of BE target	Intensity of proton beam	Time of irradiation	Time elapsed before first measurement	²¹⁰ At
8 mm	3.22 E+13	3 s	26 h	No
				signature
7 mm	3.26 E+13	3 s	27 h	No signature
6 mm	3.40 E +13	3 s	26 h	No signature
5 mm	3.43 E +13	3 s	28 h	No signature
4 mm	3.00 E+13	1 s	30 h	No signature

The complex process of production of ²⁰⁴⁻²¹⁰At was pserved earlier in LBE targets of 20 cm length and 1 cm dius (Tall et al., Int. Conf. on Nuclear Data for Science and echnology, 2007)

In the present experiment maximum length of the ample was 0.8 cm. No ²¹⁰At was observed in any f the samples. Therefore release of At through 19 Bi(p, π -xn)²¹⁰At might be a thick target henomenon, and not observed in thinner targets s used in present experiment.

