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Gas production and activation calculation in MEGAPIE

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Abstract

The MEGAwatt PIlot Experiment (MEGAPIE) project was started in 2000 to design, build and operate a liquid Lead-Bismuth Eutectic (LBE) spallation neutron target at the power level of 1 MW. Gas measurements by γ spectroscopy performed at the beginning of irradiation in August 2006 has led to the determination of main radioactive isotopes released by the LBE. Comparison with calculations performed with several validated codes supplies important volatile elements release fraction estimation in a spallation target. In addition, calculations with MCNPX2.5.0, FLUKA and SNT codes coupled with evolution programs have been performed in order to study the activation of the target. It provides important information on structural materials (such as container, window and bypass tube) and LBE activation just following the end of irradiation and at different cooling times. The induced database is relevant for safety and radioprotection during operation, for the post-irradiation experiments and for target dismantlement.

1. Introduction

The MEGAwatt Pilot Experiment (MEGAPIE) (Bauer et al, 2002) is a major project in the frame of Accelerator Driven System (ADS) studies (European T.W.G., 2001). A continuous beam of 575 MeV protons with a current up to 1.4 mA has irradiated the MEGAPIE target between August and

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December 2006 at the SINQ facility of the Paul Scherrer Institut (PSI, Switzerland). We present in this report studies related to isotope production in the target, related to the issue of gas production, and to the general activation of the LBE and of the structural materials.

2. Gas production

2.1. Gas production study motivation

Radioactive isotopes are produced in the MEGAPIE target structure and LBE during irradiation. Some are directly produced by spallation others are induced by neutron and proton activation. At operated temperature, near 500 K, noble gas and some additional elements (Br, I, Hg) are volatile to some degree, while the behaviour of interest elements such as cadmium or astatine is not well identified (Tall, 2008).

Volatile elements diffuse throughout the LBE and a fraction is released in the liquid metal expansion volume which is highlighted on the schematic view of the MEGAPIE target (see Fig. 1).

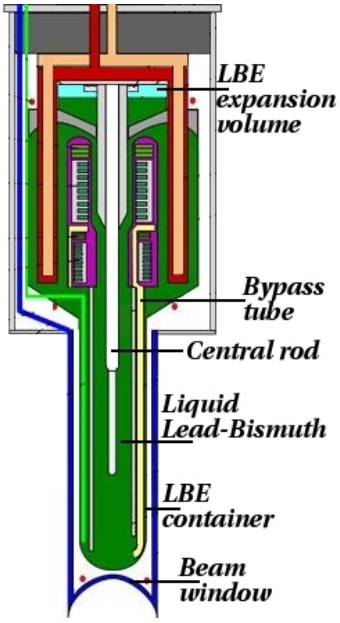

Fig. 1. Schematic view of the MEGAPIE target.

Diffusion and release processes are fundamentally very complex, even more if the circulation of LBE inside the target is considered; theoretical predictions are therefore coarse. The goal of the gas measurement was to determine the gas release by the LBE during irradiation of the target, important data in the framework of ADS safety studies and design.

2.2. Gas measurement

In order to measure short-lived isotopes, fresh gas sampling was carried out about two days after the beginning of irradiation with 1mA·h of accumulated charge. Two gas samples were taken 15 hours after stop of irradiation, and a state close to equilibrium can be assumed. Several γ spectroscopy measurements (see Fig. 2) were performed.

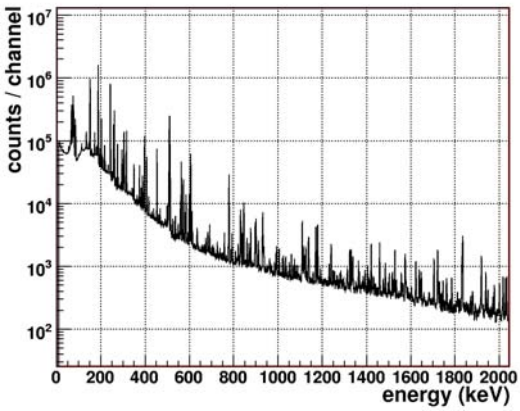

Fig. 2. Gas sample γ spectrum.

Peak detection analysis allowed the identification of the isotopes listed in the Table 1 with respective half-lifes and activities.

Table 1
Isotopes, respective half-life and measured activity in the gas sample.

Isotope	$t_{1/2}$	Activity (Bq)	Isotope	$t_{1/2}$	Activity (Bq)
^{41}Ar	1.8 h	$3.2\ 10^{2}$	^{192}Au	5.0 h	$3.4\ 10^{4}$
^{79}Kr	34.9 h	$4.5\ 10^{4}$	^{193}Au	17.7 h	$1.2\ 10^{4}$
^{85m}Kr	4.5 h	$1.5\ 10^{5}$	^{195}Au	186 d	$1.2\ 10^{2}$
^{88}Kr	2.8 h	$2.7\ 10^{4}$	^{192}Hg	4.9 h	$1.8\ 10^{4}$
^{122}Xe	20.1 h	$1.4\ 10^{4}$	^{193m}Hg	11.1 h	$1.2\ 10^{4}$
^{125}Xe	16.9 h	$9.5\ 10^{4}$	^{195m}Hg	41.6 h	$2.9\ 10^{3}$
^{127}Xe	36.4 d	$5.0\ 10^{3}$	^{197}Hg	64.1 h	$2.1\ 10^{4}$
^{129m}Xe	8.9 d	$7.6\ 10^{3}$	^{197m}Hg	23.8 h	$3.6\ 10^{3}$
^{135}Xe	9.1 h	$5.7\ 10^{2}$	^{203}Hg	46.6 d	$5.0\ 10^{1}$

In a γ spectroscopy from a second sample taken during the same sampling procedure but at a later stage, neither mercury nor gold were observed. While it is not obvious to understand this fact, the most plausible explanation is that mercury and gold are absorbed in the walls of the cover gas system between the two samplings. In order to deduce the activity in the expansion volume, the measured activities must be corrected by a sampling factor that is the fraction of gas ending in the sampling unit; this factor is known from the volumes and temperatures of the various components in the cover gas system and is $3 \cdot 10^{-4}$ for noble gas. Because of the uncertainty on the behaviour of mercury and gold during the sampling procedure, we could only define a sampling factor range for mercury estimated between $3 \cdot 10^{-4}$ and $4 \cdot 10^{-2}$. Gold is excluded from these considerations because it is not released from the LBE but produced by decay from released mercury.

2.3. Comparison with calculation

Gas production calculations were performed with FLUKA/ORIHET3 (Fassò et al., 2005 / Atchison and Schaal, 2001), MCNPX2.5.0/CINDER'90 (Pelowitz, 2005 / Wilson et al., 2006) and SNT (Konobeyev et al, 2002). With MCNPX2.5.0 we carried out calculations with Bertini/Dresner (Bertini, 1969 / Dresner, 1962), INCL4/Abla (Boudard et al., 2002 / Junghans et al. 1998), ISABEL/Abla (Yariv and Fraenkel, 1979 / Junghans et al. 1998) and CEM2k (Mashnik and Sierk, 2001) in order to describe intra-nuclear cascade and evaporation processes. The evolution calculation follows the experimental irradiation time profile. For each isotope excluding gold, evolution takes into account the spallation, the decay and nuclear reaction with low energy neutron flux. Gold is not volatile so it is assumed that it results only from mercury decay and spallation is thus excluded from its evolution calculation.

The ratio between measured and calculated activities gives an estimation of the isotope release fraction in the MEGAPIE target. Fig. 3 shows this ratio as a function of the mass number while Fig. 4 represents the same quantity versus isotope half life. The release fraction represents physically the amount of released atom per produced atom in the LBE. For both figures, mercury release fraction has been calculated using the sampling factor lower limit leading to a maximised value. For easy reading, we don't include all the used codes in the figures. Results with CEM2k and Isabel/Abla are similar.

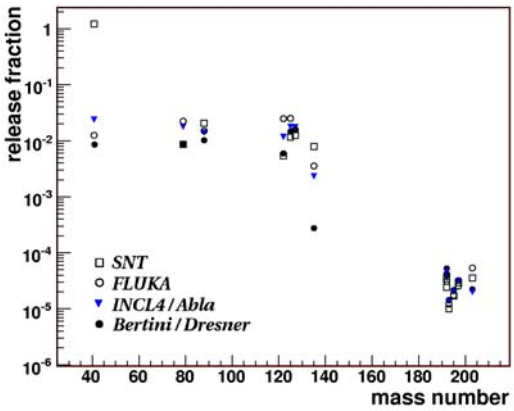

Fig. 3. Ratio between calculated and measured activities as a function of isotope mass number.

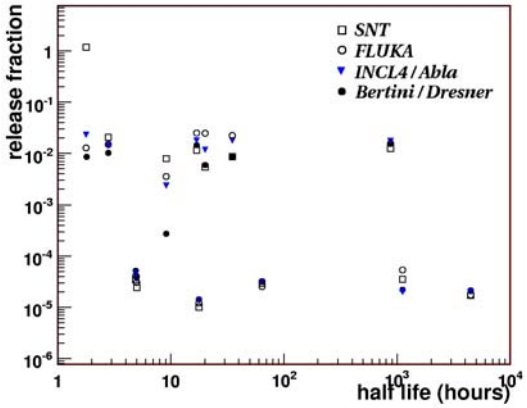

Fig. 4. Ratio between calculated and measured activities as a function of isotope half life.

Results indicate that LBE release is more than three orders of magnitude higher for noble gas compared to mercury. Fig. 4 clearly shows that there is no isotope decay effect in release calculations. The calculated yield of ^{41}Ar with SNT is much lower than the value obtained with the other codes. It results from the use of the improved Fong model (see Fong et al., 1964 and Konobeyev et al., 1999) used in the CASCADE/ASF calculation. The model predicts the sharp decrease of the fission

yields for lead and bismuth isotopes with decreasing of the fragment mass for A < 43.

Average release fraction and associated uncertainty calculation as mean value and standard deviation of data leads to the following value:

$$\tau_{ng} = (1.4 \pm 0.7).10^{-2} \quad (1)$$

$$(3.7 \pm 1.5).10^{-7} \leq \tau_{Hg} \leq (2.8 \pm 1.2).10^{-5} \quad (2)$$

τ_{ng} and τ_{Hg} are the release fraction for noble gas and mercury respectively which has been extracted with the two sampling factor extreme values given above.

2.4. Conclusion for gas production

Experimental data obtained by γ spectroscopy shows that activity in the expansion volume is coming mainly from noble gases (Ar, Kr, Xe) and from heavier elements (Au and Hg). A good agreement among the spallation codes is observed and their supposed reliability allows extracting relevant estimation of volatile element release fraction in the MEGAPIE target. Note that simulated values for ^{135}Xe obtained with Bertini/Dresner and CEM2k deviate from the experimental distribution.

The reader can find in Appendix A the calculated volatile elements activity just after the end of the 123 days irradiation.

3. Activation calculation

3.1. Methodology

Calculations presented in this part are based on the MEGAPIE average irradiation history. It corresponds to a 575 MeV proton beam at 0.947 mA during 123 days, since the target operated from August to December 2006 with a total charge of 2.80 A·h.

3.1.1 With MCNPX2.5.0 – CINDER'90 / EASY

Transport codes usually consist of the coupling of a high energy part relying on spallation models and a low energy part utilising nuclear data tables. Within different programmes, and especially the EURISOL-DS (Rapp and David, 2006) project, up to ten physics model combinations available in the transport code MCNPX2.5.0 (Pelowitz, 2005) have been benchmarked. Four model combinations have thus been selected to study the MEGAPIE target: Bertini-Dresner (default option), Isabel-Abla, INCL4-Abla and CEM2k. The evolution code CINDER'90 was used additionally to take into account the subsequent decay of the spallation products and the low energy neutron activation. Calculations were performed with the latest and refined geometry description. The three main regions studied are the lead-bismuth target (LBE), its container and the beam window, part of the container between the target and the proton accelerator (see Fig. 1). Total activities have been computed in each region with the four spallation models. Evolutions in time of these activities have been calculated until 100000 years after the shutdown.

In addition, structure activation calculations were carried out using the European Activation system code (EASY) (Forrest et al., 2005). The code uses the European Activation File (EAF) as an integrated cross-section (neutron energies up to 20 MeV). EAF contains information about more than 1900 nuclides. This calculation uses the MEGAPIE neutron flux calculated with MCNPX.2.5.0 and activation is deduced taking into account only neutron with energy below 20 MeV. The main objective of this assessment is to support the experimental activities that will be carried out during the Post-Irradiation Examination phase. Three structures were examined: the Central Rod made of 316L (CR), the Liquid Metal Container's walls made of T91 (LMC) and the Bypass Tube made of 316L (BPT). Samples from these three structures at 5 different heights will be obtained and measured. For our immediate use, we have arbitrarily focused on the first 20 dominant radioisotopes in each examined volume, and will limit the cooling time to 2160 days after shutdown (~ 6 years).

3.1.2 With SNT

We used the code SNT (Konobeyev et al, 2002) which was specially designed for the modelling of the transmutation and activation of materials irradiated with low, intermediate and high energy particles. Main input data include the irradiation scheme, particles spectra, and nuclear reaction cross-sections. The information about neutron and

proton fluxes in lead-bismuth used in the calculation is presented in Table 2.

Table 2
Average neutron and proton fluxes in lead-bismuth (particles·cm^{-2}·s^{-1})

Energy range	Neutrons	Protons
E < 20 MeV	2.05×10^{13} (96.48 %)	1.10×10^{10} (0.97 %)
20<E<150 MeV	5.91×10^{11} (2.77 %)	1.29×10^{11} (11.44 %)
E > 150 MeV	1.59×10^{11} (0.75 %)	9.88×10^{11} (87.58 %)
Total	2.13×10^{13} (100 %)	1.13×10^{12} (100 %)

3.2. Activation of the LBE

The activation of the target is plotted in Fig 5. All models give more or less the same total activities except at around 10 years decay where tritium is predominant. We have to mention here that predictions of tritium are problematic since some models do not produce it while some others predict a too large amount (Rapp and David, 2006).

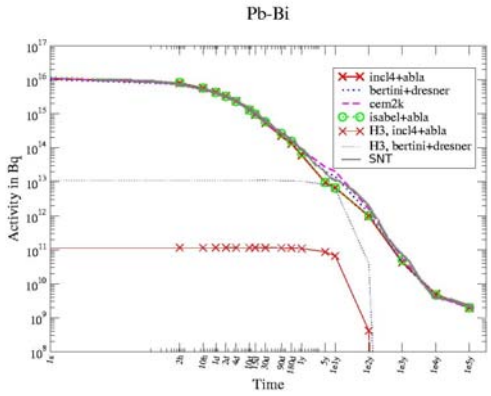

Fig. 5. Total activity in the lead-bismuth target as a function of cooling time after the end of irradiation.

Fig. 6 shows the relative contribution of maximum activity nuclides, extracted with SNT code, in the total activity of irradiated lead-bismuth at the time of cooling up to $5\cdot10^7$ years. Fig. 7 shows contributions of nuclides with different atomic numbers obtained with SNT.

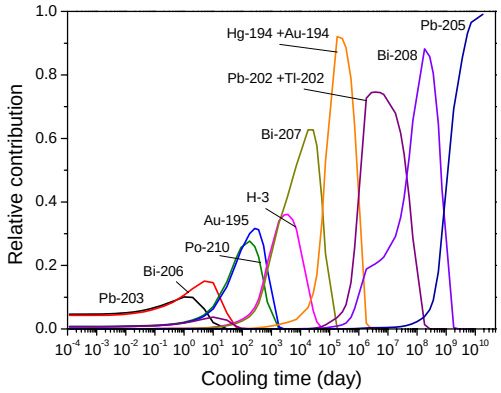

Fig. 6. Relative contribution of nuclides providing the main contribution to the activity of the lead-bismuth after 123 days irradiation.

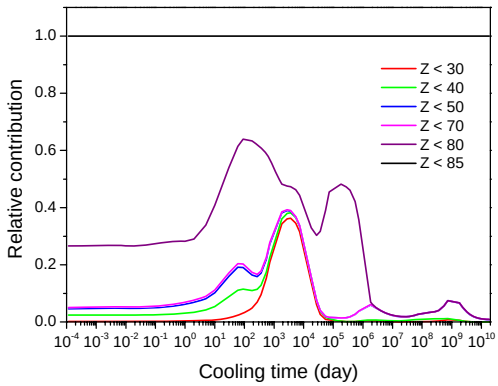

Fig. 7. Relative contribution of nuclides with various atomic numbers in the total activity of irradiated lead-bismuth.

The contribution of various ranges of particle energy distributions in the nuclide production was examined with SNT code. The information obtained is important for the formation and the correction of the priority list for the evaluation of cross-sections for radionuclide yields in the lead-bismuth coolant of ADS under the irradiation. The example of the obtained information about the contribution of different energy ranges of neutrons and protons in the production of radionuclides in lead-bismuth is shown in Table 3.

Table 3
Contribution of different parts of neutron and proton spectra in selected nuclide production in irradiated lead-bismuth (%).

Nuclide	Neutrons <20 MeV	Neutrons 20-150 MeV	Neutrons >150 MeV	Protons <150 MeV	Protons >150 MeV
Po 210	99.96	0.03	0	0.007	<0.001
Po 209	0.004	<0.001	0	11.8	88.2
Po 208	<0.001	<0.001	0	66.6	33.4
Po 207	<0.001	<0.001	0	41.8	58.2
Po 206	<0.001	<0.001	0	56.5	43.5
Po 205	<0.001	<0.001	0	51.9	48.1
Po 204	<0.001	<0.001	0	65.0	35.0
Po 203	0	<0.001	0	45.3	54.7
Po 202	0	<0.001	0	44.5	55.5
Po 201	0	<0.001	0	19.1	80.9
Po 201m	0	<0.001	0	33.2	66.8
Po 200	0	<0.001	0	17.8	82.2
Bi 210	99.97	0.03	0	0	0
Bi 210m	99.92	0.08	0	0	0
Bi 208	73.1	15.2	1.3	1.8	8.6
Bi 207	10.6	66.9	2.0	7.5	13.0
Bi 206	<0.001	62.7	2.6	11.6	23.1
<…>					
Y 85	0	0	3.1	0	96.9
Y 83	0	0	0.04	0	99.96
Sr 93	0	0	<0.001	0	100.
Sr 92	0	0	<0.001	0	100.
Sr 91	0	0	0.1	0	99.9
Sr 90	0	0	0.3	0	99.7
Sr 89	0	0	0.6	0	99.4
Sr 87m	0	0	2.9	0	97.1
<…>					
Cl 39	0	0	<0.001	0	~100.
Cl 38	0	0	<0.001	0	~100.
S 38	0	0	<0.001	0	100.
Mg 28	0	0	7.5	0	92.5
Mg 27	0	0	5.5	0	94.5
Na 24	0	0	5.1	0	94.9
F 18	0	0	4.7	0	95.3
N 13	0	0	4.3	0	95.7
C 14	0	0	6.1	0	93.9
C 11	0	0	5.3	0	94.7
Be 10	0	0	7.2	0	92.8

3.3. Structure material activation results

In Figure 8 we plot the effects of the low energy neutron activation extracted with the evolution code CINDER'90. These neutrons play a significant role in the container surrounded by heavy-water and are negligible for the target where the high energy particles dominate.

Activation calculations with the code EASY (see Fig. 9) show relevant information for the MEGAPIE post-irradiation analysis. All studied volumes reach a plateau closed to $3 \cdot 10^{12}$ Bq/kg in the period between 4 to 6 years after shutdown.

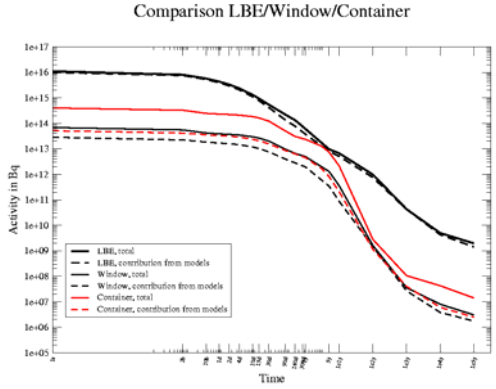

Fig. 8. Total activity in the lead-bismuth target, in the window and in the container taking into account the low energy neutrons (E < 20 MeV) or not. The spallation model used is INCL4-Abla.

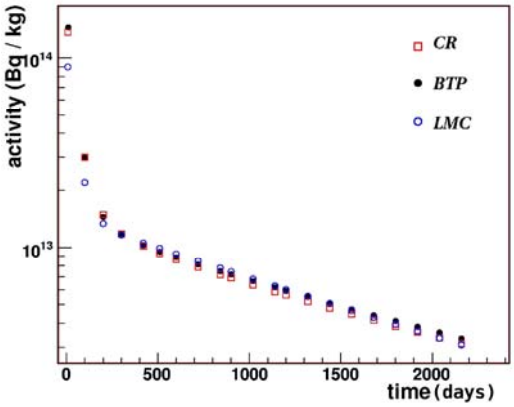

Fig. 9. Activity calculated with EASY in the central rod, the bypass tube and the liquid metal container.

For the cooling period considered by this report, from 21/12/2006 until 20/12/2014 (2160 days), the evolution of the specific activities of both materials 316L and T91 is mainly governed by two radioisotopes: ^{51}Cr (τ=27.7 days) and ^{55}Fe (τ=2.7 years), both are beta emitters. The most important γ contribution comes from the ^{60}Co. While it is not the most important radio-element in terms of activity (at least for the first few years ~ 5y-10y), it is certainly the most important radioisotope in terms of dose-rate.

3.4. *Effects of impurities*

We investigated (Fig. 10 and 11) the effect of impurities and minor alloy elements (see Table 4 for main nuclei) in the lead-bismuth and in the container/window with MCNPX.2.5.0 and CINDER'90.

Table 4
Main impurity and minor alloy elements concentrations in the LBE and the window (T91).

LBE		T91	
nucleus	ppm	nucleus	ppm
Sn	198	Si	3197
In	51	Mn	4180
B	33	V	2480
Ag	13	Ni	1260

Looking at the mass distribution for the window under irradiation (Fig. 10) we see that the minor alloy elements influence the regular distribution (from Fe and Cr), especially for A≈30 and A > 60.

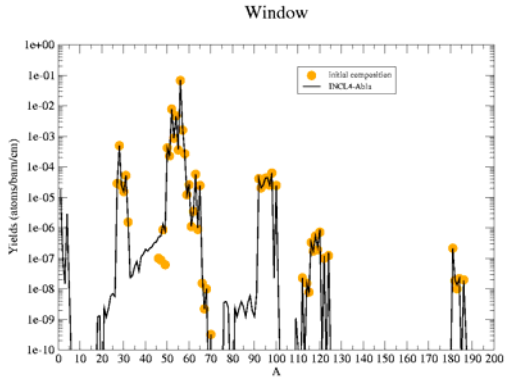

Fig. 10. Mass distribution in the window for an irradiation with 575 MeV protons . The spallation model used is INCL4-Abla.

The concentration of impurities in LBE is low; as far as radio-activity is concerned, impurities in MEGAPIE are not important at any time after the shutdown. This is well illustrated for the LBE in Fig.11.

3.5. *Conclusion on activity calculations*

Activities and quantities of nuclei, volatile elements included, have been predicted in the three main regions of the MEGAPIE target: the lead-bismuth target itself, its container and the window. The central rod, the liquid metal container walls and the bypass tube activity have been investigated too. The codes used are the transport code MCNPX2.5.0 and the evolution code CINDER'90 and EASY. Four spallation models within MCNPX have been compared and three of them are known to give generally very good results for the spallation reactions.

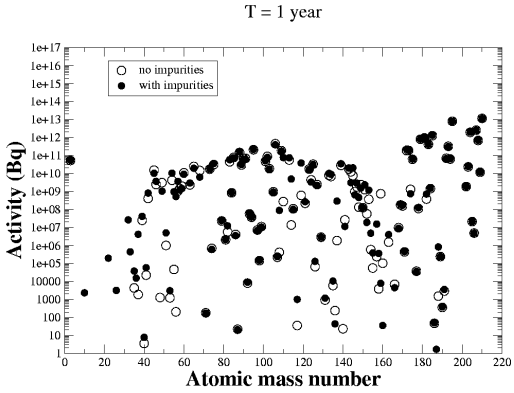

Fig. 11. Activity of radionuclides contained in the LBE after 123 day-irradiation with 575 MeV protons at 0.947 mA using INCL4-Abla and the cooling during 1 year depending on the mass number of nuclides.

Models are generally in good agreement, however, some improvements are needed, and new releases of INCL4, Abla and CEM (known as CEM03 in a beta version of MCNPX) should be available in the future.

Calculation with the code SNT gives relevant results concerning most radioactive isotope after irradiation. Atomic number atoms contribution in the LBE total activity has been extracted. Finally, the contribution of different energetic parts of neutron and proton in the nuclide production in irradiated lead-bismuth has been investigated and highlights important cross section for ADS study.

4. Acknowledgements

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personnel for the great support given during installation, irradiation and post-test analysis.

Appendix A. Cumulated volatile isotope activities after the end of irradiation.

Table 1 : Calculated activity after 123 days of irradiation of isotopes. Only activities upper than one Becquerel after one month cooling are specified.

	FLUKA	MCNPX2.5.0	SNT
^{37}Ar	1.46E+10	2.31e+06	2.07E+09
^{39}Ar	1.53E+08	7.50e+07	4.84E+05
^{42}Ar	1.02E+09	3.48e+08	7.32E+06
^{79}Kr	4.47E+11	6.01e+11	1.18E+12
^{81}Kr	2.77E+06	3.28e+06	6.62E+06
^{83m}Kr	7.93E+12	5.30e+12	1.12E+13
^{85}Kr	1.25E+11	1.27e+11	1.70E+11
^{122}Xe	9.63E+10	1.87e+11	4.42E+11
^{127}Xe	1.20E+12	1.03e+12	1.53E+12
^{129m}Xe	1.04E+12	3.49e+08	9.53E+07
^{131m}Xe	1.77E+09	2.37e+09	2.27E+09
^{133}Xe	1.22E+11	1.18e+11	1.21E+11
^{133m}Xe	8.01E+08	8.56e+08	8.16E+08
^{194}Hg	9.35E+10	7.12e+10	1.04E+11
^{197}Hg	2.70E+14	2.74e+14	2.83E+14
^{203}Hg	3.96E+12	1.24e+13	6.16E+12

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